A BRIEF HISTORY OF THE CHEMICAL TECHNOLOGY DIVISION

COVER PHOTOGRAPHS

Center: Floyd L. Culler, Director of the Chemical Technology Division, 1953–1965.
Upper left: Don E. Ferguson, Director of the Chemical Technology Division, 1965-1983.
Upper right: Dave 0. Campbell examining equipment at the Three Mile Island Nuclear Reactor.
Middle right: Ray G. Wymer, Director of the Chemical Technology Division, 1983-1988.
Bottom: George W. Parker in his research laboratory.
Lower left: Charles D. Scott, Senior Corporate Fellow, adjusting an experimental ion-exchange system.

ORNL/M-2733

A BRIEF HISTORY OF THE CHEMICAL TECHNOLOGY DIVISION

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Chemical Technology Division

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PREFACE

A new atomic era arose from the wartime activities in the 1940s. During this era, much public attention was focused on the "big science" programs, physics, and engineering technologies associated with the production of the actual hardware (e.g., weapons and nuclear power reactors). Chemical and engineering separations technologies were some of the main forces behind the success enjoyed by the atomic energy programs. Historians view the technologically successful achievements as a colossal team effort embracing not only physicists and mechanical engineers, chemists and chemical engineers, military personnel and civilians, but also a whole host of workers involved in supply, logistics, and construction.

This document attempts to reconstruct a portion of the role played in that team history by the Chemical Technology Division (Chem Tech) of the Oak Ridge National Laboratory (ORNL). Chem Tech had its early foundations in 1944 in the chemical separations studies conducted at the Clinton Laboratories under the auspices of the Manhattan Project. It actually came into existence as an ORNL division on February 1, 1950.

Chem Tech's early contributions were landmark pioneering studies. Unknown and dimly perceived problems-chemical hazards, radioactivity, and criticality-had to be dealt with. New chemical concepts and processes had to be developed to test the new theories being developed by physicists. New engineering concepts had to be developed and demonstrated in order to build facilities and equipment that had never before been attempted. These achievements were great scientific accomplishments and compare in importance to other major technological landmarks of historical significance. Since their beginnings, ORNL and Chem Tech have been at the cutting edge in research and development associated not only with chemical separations of new radioactive elements, but also with the evolution of new engineering technologies involving peaceful applications of nuclear energy.

In response to changing national objectives, ORNL and Chem Tech have diversified their objectives and missions over time. Today, Chem Tech remains a premier part of ORNL, one of the most respected research laboratories in the world.

This document is a history of the first 42 years of an ORNL division that provided pioneering national leadership in nuclear fuel reprocessing and subsequently continued its prominence as an international center of chemical engineering R&D relevant to nuclear energy, alternative energy sources, isotope production and distribution, and advanced waste management science and technology.

Richard K. Genung
Director
Chemical Technology Division
Oak Ridge National Laboratory

ACKNOWLEDGMENTS

Special acknowledgment must be made to the U.S. Department of Energy (DOE) and previous federal sponsors: the “Manhattan Project,” the Atomic Energy Commission (AEC), and the Energy Research and Development Administration (ERDA). Many current and past managers, scientists, and engineers in these organizations have made significant contributions to the research and development (R&D) and work tasks assigned to Oak Ridge National Laboratory (ORNL) and to the Chemical Technology Division (Chem Tech). Their sponsorship has permitted Chem Tech to receive international acclaim through successful accomplishment of the work reported in this brief history.

Many individuals helped to compile this history of the Chem Tech. Special appreciation is given to Alvin Weinberg, Herman Postma, Miles Leverett, Floyd Culler, Frank Bruce, Don Trauger, Clair Scherston, Dave Campbell, Warren Eister, Ray Wymer, Frank Harrington, Rex Leuze, Walt Bond, Paul Haas, Bill Arnold, Charles Coleman, Dave Crouse, Fred Hurst, Bill Unger, Scott Ludwig, Hal Goeller, Ed Nicholson, Ray Blanco, Gene Newman, John Bigelow, Tom Lomenick, Emory Collins, Karl Notz, Herschel Godbee, Kaye Johnson, Arlene Kibbey, Larry Shappert, Martha Stewart, Elias Greenbaum, Phil McGinnis, Dick Lorenz, Hank Cochran, Carl Burtis, Charles Scott, Lloyd Youngblood, Bob Hightower, Zane Egan, Jerry Strandberg, Wilson Pitt, and Bob Brooksbank for both written and oral contributions that present a more complete history of this large and complex division. Because of the diversity of programs and the lengthy tune involved (over 40 years), the compilers have presented a selective history. We would appreciate comments from the readers concerning the completeness of this volume. Please let us know if a more in-depth volume is desired, and, hopefully, we can add historically important information to an additional compilation concerning this dynamic ORNL division in the future.

We appreciate the editorial and publications assistance of Debbie Stevens, Becky Wilker, Larry Davis, Sandi Lyttle, Vicky Rolfe, Nancy Smith, and other members of the Publications Division. We also appreciate the assistance of Chem Tech staff members Joy Williams and Brenda Breeden for their help in preparing the draft manuscript and Vic Pardue for designing and preparing the cover.

EXECUTIVE SUMMARY

A significant part of the story of atomic energy occurred at the Oak Ridge National Laboratory. In 1943, the Clinton Engineering Works and Clinton Laboratories (code named X- 10 site) were established in an unknown southern Appalachian community now known as Oak Ridge, Tennessee. This facility was later renamed the Oak Ridge National Laboratory (ORNL). The principal mission of the newly created laboratory complex was to develop separations processes for plutonium and uranium on an engineering scale and, thus, to assist in the development of the atomic bomb and, later, peaceful application of atomic energy. The Chemical Technology Division (Chem Tech) and its predecessor, the Technical Division, played major roles in this undertaking.

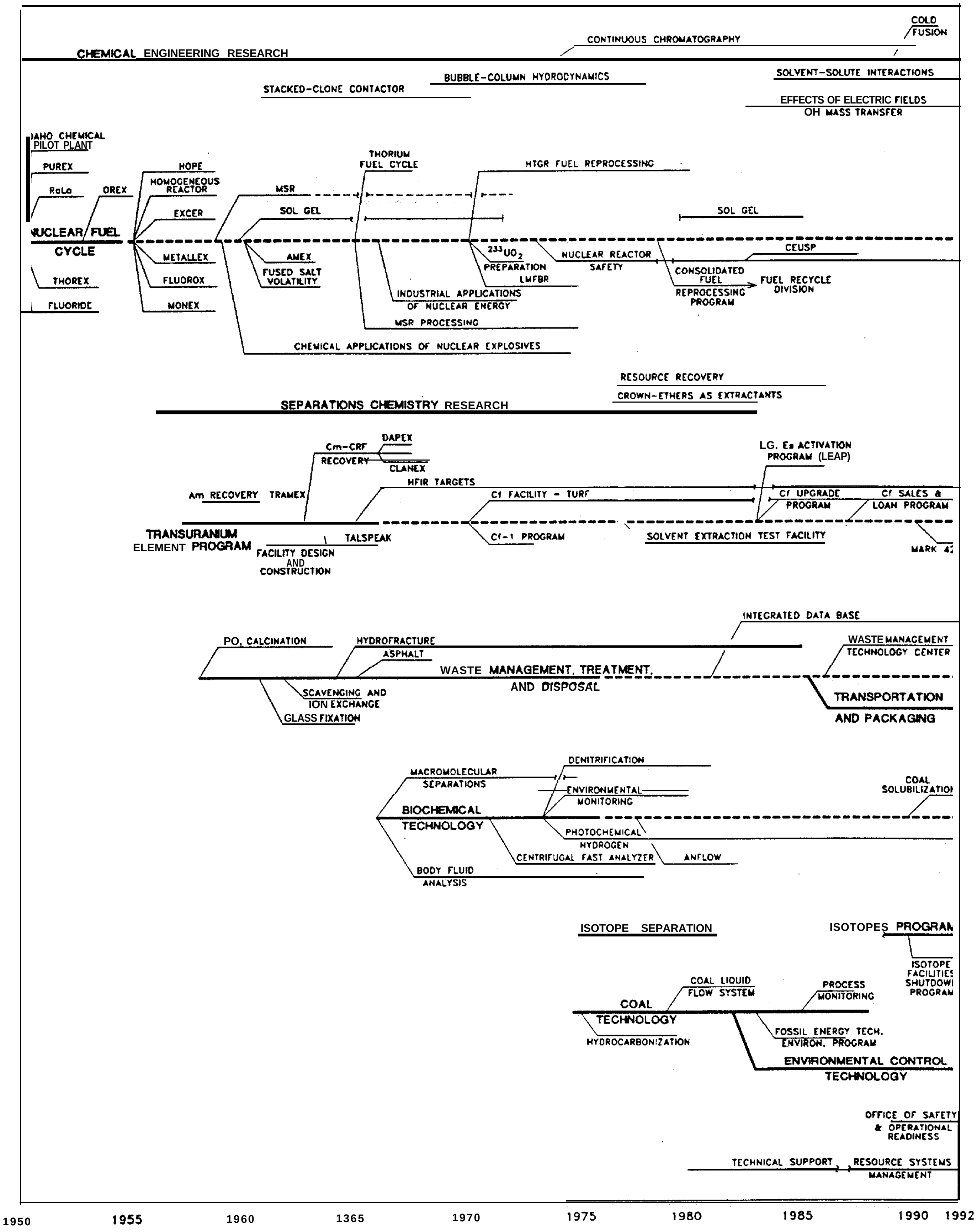
Throughout the ensuing years, the federal government, under the auspices of the Manhattan Project, the Atomic Energy Commission (AEC), the Energy Research and Development Administration (ERDA), and the U.S. Department of Energy (DOE), has operated ORNL using several contractors such as the University of Chicago, Monsanto Chemical Company, Carbide and Carbon Chemicals Company (a division of the Union Carbide and Carbon Corporation), and the current contractor, Martin Marietta Energy Systems, Inc.

This volume presents a selective history of Chem Tech activities from 1950 to 1992. The formation of Chem Tech is deeply intertwined with chemical separations, especially uranium and plutonium, and nuclear fuel reprocessing. The initial major activities were design of the Idaho Chemical Processing Plant, development of the tributyl phosphate process for the recovery of uranium from Hanford metal waste, development of the Purex process for the recovery of uranium and plutonium from Hanford irradiated metal, development of the RaLa process for separation of barium from fuel units, homogeneous reactor fuel studies, and design/construction of the ORNL Metal Recovery Plant. An important initial and ongoing task was the training of operators for Hanford and Savannah River projects and each of the later commercial endeavors. With diversification of national and ORNL missions, Chem Tech undertook R&D studies in many areas, including biotechnology; clinical and environmental chemistry: nuclear reactors, safety, and regulations; effective and safe waste management and disposal; computer modeling and informational data bases; isotope production; and environmental control.

Kudos and anecdotes from ORNL leaders and notable scientists provide personal perspectives of the division and its impacts. These include contributions from Miles C. Leverett, Director, ORNL Technical Division, 1943-1948; Alvin Weinberg, Director, ORNL, 1954-1974; Herman Postma, Director, ORNL, 1974-1989; Floyd Culler, Director, Chem Tech, 1953-1965, Assistant Director of ORNL 1965-1970, and Deputy Director of ORNL 1970–1977; Donald Trauger, Associate Director, ORNL, 1970–1986; Frank Bruce, Associate Director, ORNL. 1970-1978; Ray Wymer, Director, Chem Tech, 1983-1988; and several notable Chem Tech scientist and engineers, including Warren Eister, Ray Blanco, Rex Leuze, Frank Harrington, Ed Nicholson, Clair Scherston, and Dave Campbell.

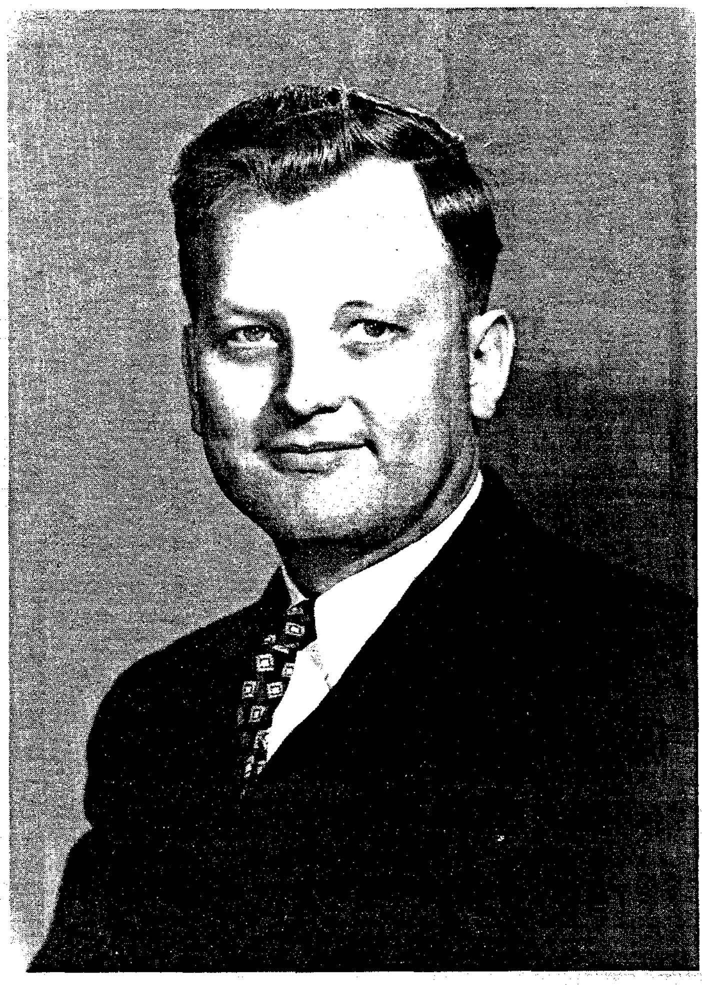
Listings of Chem Tech leaders, its ever-changing operational organization, some of its numerous and diverse activities, and several facilities are provided in this document. Many of these Chem Tech activities were of considerable national importance to DOE programs. The changing missions of Chem Tech are encapsulated in the evolving activities. The following time line summarizes the activities and provides an interesting historical perspective.

ORHL DWG WA-2051



DIRECTORS OF THE CHEMICAL TECHNOLOGY DIVISION

Frank L. Steahly



Dr. Frank Steahly, a native of Portsmouth, Ohio, was born April 16, 1916. In 1937, he received a BS degree in chemistry from Ohio State University. He received MS and PhD degrees in organic chemistry from the University of Cincinnati in 1939 and 1941, respectively. Frank joined the Clinton Laboratories in September 20, 1943, and became the first director of the ORNL Chemical Technology Division on February 1, 1950. He served in that capacity until April 30.1953, when he went to the Idaho Test Site with Phillips Petroleum. In 1955 Dr. Steahly joined the Research Center at the Union Carbide Charleston, West Virginia, site working in coal hydrogenation. He became associate director in charge of research and development of the Olefins Division of Union Carbide, Dr., Steahly died in 1966 at the age of 50. [Sources: Carolyn Ladd, Personnel Records, ORNL, October 26, 1992; Clair W. Scherston, St. Albans, West Virginia, personal communication, October 26, 1992; ***Oak Ridge National Laboratory News*** 2(3), 1 (February 24, 1950)].

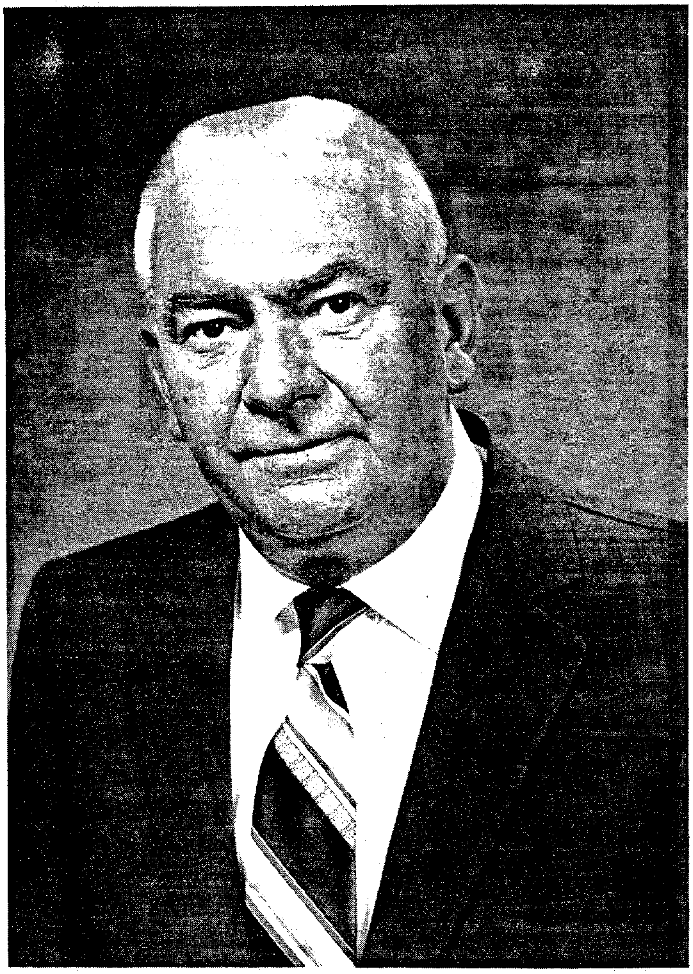
Floyd Leroy Culler

Mr. Floyd Culler was born January 5, 1923. In 1943, he received a BS degree. in chemical engineering from Johns Hopkins University. After working at Eastman Kodak, Floyd was assigned to work with Tennessee Eastman on the Manhattan Project. He began working at Y-12, Clinton Engineering Works in 1945. Floyd transferred to ORNL on May 6, 1947, when he joined the Technical Division. He became the Chemical Technology Division Design Section Chief in 1950. In 1953 Floyd was selected to be the second division director of the Chemical Technology Division, serving in that capacity until 1965. Subsequently, Floyd served as Assistant Director of ORNL from 1965-1970 and as Deputy Director of ORNL from 1970-1977. He retired from ORNL on December 31, 1977. In 1978 he became President of the Electric Power Research Institute; Palo Alto, California, and is currently President Emeritus of EPRI.

Mr. Culler served on the Board of Directors of the American Nuclear Society and is also active in the American Institute of Chemical Engineers and the National Academy of Engineering. He also served on the National Research Council's Committee on Science and Public Policy and the Committee on Nuclear and Alternative Energy Systems.

(Sources: Ann Pemet, ed., Who's ***Who in Atoms,*** 6th ed., Francis Hodgson Books Limited, Guernsey, British Isles, 1977, p. 111; ***International Who's Who in Energy and Nuclear Sciences,*** Longman Group Limited, Harlow, Essex, UK, 1982, pp. 92-93)

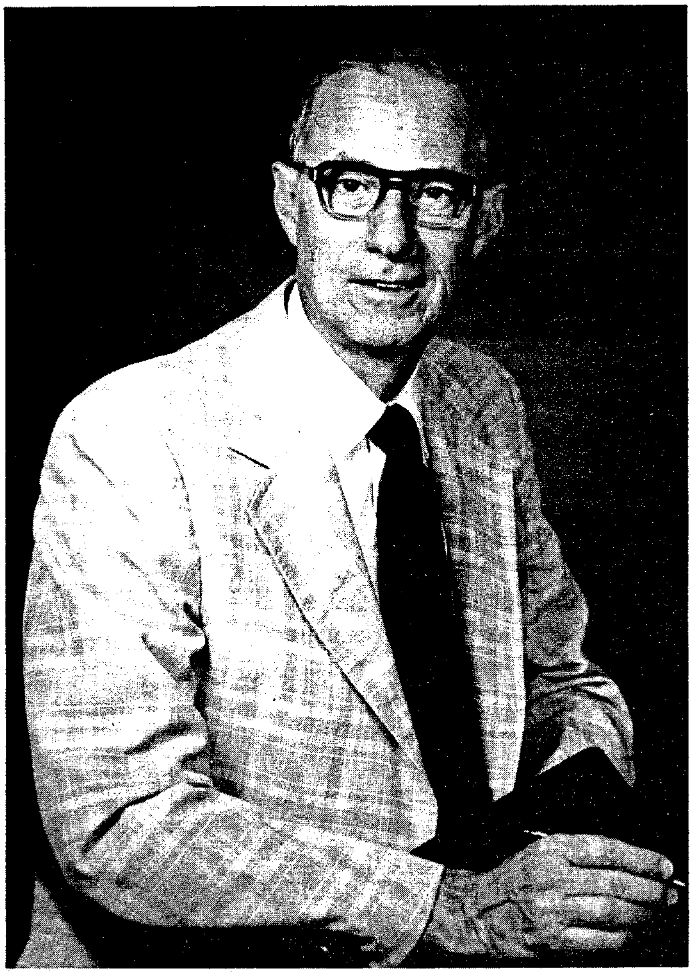
Don Ernest Ferguson



Mr. Don Ferguson was born December 10, 1923. In 1944 he received a BS degree in chemical engineering from the Tennessee Technological University in Cookeville. He received a MS degree in mathematics and chemistry from The University of Tennessee in 1950. On July 23.1946, Don started working at the Clinton Laboratories. He was assigned to the Process Development Section of the Technical Division. Several sections of the Technical Division later became major groups in the Chemical Technology Division. Don became section chief of the Chemical Technology Division Chemical Development Section. A in 1955. He was selected to be the third director of the Chemical Technology Division in 1965 and served in that capacity until 1983. In 1983 became a technical assistant to Don Trauger, Associate Director of ORNL. Don retired from ORNL on January 31, 1987, and died June 6, 1988.

Mr. Ferguson was active in the American Nuclear Society and served as an advisor to the Third International Conference on the Peaceful Uses of Atomic Energy in Geneva in 1964. (Sources: Carolyn Ladd, Personnel Records, ORNL, October 26, 1992; ***Who's Who in Atoms, Volume I***, 5th ed., Harrap Research Publications, London, 1969, p. 462)

Raymond George Wymer



Dr. Ray Wymer was born October 1, 1927, in Colton, Ohio. In 1950 he received a BS degree in chemistry from Memphis State College, and in 1953 he received both MS and PhD degrees in chemistry from Vanderbilt University in Nashville, Tennessee. He came to work for the Chemical Technology Division in 1953. In 1956 he left ORNL to become an Associate Professor of Chemical Engineering at the Georgia Institute of Technology in Atlanta, Georgia, and served concurrently in several areas as Directors of the Radioisotopes Laboratory, Radiation Effects, and Radiochemical Separations projects. In 1958 he became Chief of Nuclear Chemistry for Industrial Reactor Laboratories; Inc., in Plainsboro, New Jersey. Dr. Wymer returned to the Chemical Technology Division in 1959. In 1964 he became section chief of the Chemical Development Section A. He was selected as the fourth division director of the Chemical Technology Division in 1983 and served in that capacity until 1988.

Dr. Wymer is active in the American Chemical Society, American Institute of Chemists, American Institute of Chemical Engineers, American Nuclear Society, and Sigma Xi. He was a U.S. representative to three International Atomic Energy Agency Panels in Vienna, Austria, and served on the National Research Council's Subcommittee on Nuclear and Radiochemistry and on several National Academy of Science panels. He has served as consultant to the U.S. Department of State on nuclear nonproliferation. He also coauthored several books, including the following:

R. G. Wymer and S. Peterson, ***Chemistry** in **Nuclear Technology,*** Addison-Wesley, 1963.

R. G. Wymer and B. L. Vondra, ***Light Water Reactor Nuclear Fuel Cycle,*** CRC Press, Boca Raton, Fla., 1981.

(Sources: Carolyn Ladd, Personnel Records, ORNL, October 26, 1992; ***Who's Who in Atoms, Volume*** II, 5th ed., Harrap Research Publications, London, 1969, p. 1712; R. G. Wymer and B. L. Vondra, ***Light Water Reactor Nuclear Fuel Cycle,*** CRC Press, Boca Raton, Fla., 1981)

Richard K. Genung

Dr. Richard Genung was born September 9, 1947, in Urbana, Illinois. In 1969 he received a BE degree in chemical engineering from Vanderbilt University and MS and PhD degrees from The University of Tennessee in 1972 and 1975, respectively. He did his research as a graduate student investigator at ORNL and, subsequently, came to work in the Chemical Technology Division on July 1, 1975. Programs he worked on at ORNL include Basic Energy Sciences, Conservation and Renewable Energy, Fossil Energy, and Waste Management R&D. He was part of the ORNL R&D team that received an IR-100 Award for development of the portable centrifugal fast analyzer. Richard led another engineering team that demonstrated commercial application of the ANFLOW wastewater treatment process in Knoxville and was one of the leaders who helped establish waste management and environmental technology as a major mission for the Chemical Technology Division. He was named Director of the Chemical Technology Division in 1988.

Dr. Genung has held an adjunct faculty position in the Chemical Engineering Department at The University of Tennessee. He has been active in the Knoxville and Oak Ridge Chapters of Alpha Chi Sigma, Sigma Xi, the American Institute of Chemical Engineers, and the Energy Systems Chapter of the National Management Association.

LIST OF ACRONYMS

AACC	American Association for Clinical Chemistry
A B R	Advanced Breeder Reactors
AE	Architech Engineer
AEC	Atomic Energy Commission
AERE	Atomic Energy Research Establishment
AGNS	Allied General Nuclear Services
ALARA	As Low As Reasonably Achievable
ANFLOW	Anaerobic Upflow Wastewater Treatment System
ANL	Argonne National Laboratory
ANP	Aircraft Nuclear Propulsion
ANS	Advanced Neutron Source
APD	Anhydrous Propylene Diamine
ARE	Aircraft Reactor Experiment
ASFI	Ashland Synthetic Fuels, Inc.
ASTP	Army Specialized Training Program
AVLIS	Advanced Vapor Laser Isotope Separation
BES	Office of Basic Energy Sciences
BFA	Body Fluids Analysis
BP	Bismuth Phosphate Process
BWR	Boiling-Water Reactor
CANE	Chemical Application of Nuclear Explosives
CDB	Characteristics Data Base
CEUSP	Consolidated Edison Uranium Solidification
CFR	Consolidated Fuel Reprocessing
CFRP	Consolidated Fuel Reprocessing Program
CMF	Containment Mockup Facility
CRADA	Cooperative Research and Development Agreement
CRBR	Clinch River Breeder Reactor
CRF	Curium Recovery Facility
CRI	Containment Research Installation
CTD	Chemical Technology Division
CWMO	Central Waste Management Organization
DAPEX	Dialkylphosphoric Acid Extraction
DF	Decontamination Factor
DOE	Department of Energy
DOT	Department of Transportation
DSM	Division of Security Materials
EBR	Experimental Breeder Reactor
ECT	Environmental Control Technology
EDS	Exxon Donor Solvent
EDTA	Ethylenediamine Tetraacetic Acid
EGCR	Experimental Gas-Cooler Reactor
EPA	Environmental Protection Agency

EPC	Emulsion Phase Contactor
EPRI	Electric Power Research Institute
ERDA	Energy Research and Development Administration
ESA	Environmental and Safety Organization
ESD	Environmental Sciences Division
FFTF	Fast Flux Test Facility
FP	Fission Products
FUETAP	Formed Under Elevated Temperature and Pressure
FVP	Fluoride Volatility Process
GE	General Electric Co.
GRI	Gas Research Institute
HAZWDDD	Hazardous Waste Disposal, Development, and Demonstration
HAZWRAP	Hazardous Waste Remedii Action Program
HBCU	Historically Black Colleges and Universities
HBU	High Burnup
HFIR	High-Flux Isotope Reactor
HI	Horizontal Induction Heated
HLW	High-Level Radioactive Waste
HPLC	High-Pressure Liquid Chromatography
HR	Homogeneous Reactor
HRE	Homogeneous Reactor Experiment
HRI	Hydrocarbon Research, Inc.
HRT	Homogeneous Reactor Test
HT	High Temperature
HTGR	High-Temperature Gas-Cooled Reactor
IAEA	International Atomic Energy Agency
ICPP	Idaho Chemical Processing Plant
IDB	Integrated Data Base
IEA	Institute of Atomic Energy
ILW	Intermediate-Level Radioactive Waste
INEL	Idaho National Engineering Laboratory
KAPL	Knolls Atomic Power Laboratory
LANL	Los Alamos National Laboratory
LBU	Low Burnup
LEAP	Large Einsteinium Activation Program
LITR	Low-Intensity Test Reactor
LLNL	Lawrence Livermore National Laboratory
LLLW	Low-Level Liquid Waste
LLW	Low-Level Radioactive Wastes
LLWDDD	Low-Level Waste Disposal, Development, and Demonstration Program
LMFBR	Liquid-Metal Fast Breeder Reactor
LOFT	Loss of Flow Test
LWR	Light-Water Reactor
M&C	Metals and Ceramics Division
MAN	Molecular Anatomy Program

MEI	Minority Educational Institutes
MFO	Mixed Function Oxidase
MFRP	Midwest Fuel Recovery Plant
MPPF	Multipurpose Processing Plant
MRS	Monitored Retrievable Storage Facility
MSBR	Molten-Salt Breeder Reactor
MSR	Molten Salt Reactor
MSRE	Molten Salt Reactor Experiment
MTHM	Metric Ton of Heavy Metal
MTR	Materials Test Reactor
MVST	Melton Valley Storage Tank
NASA	National Aeronautics and Space Administration
NBS	National Bureau of Standards
NCI	National Cancer Institute
NFS	Nuclear Fuel Services
NIGMS	National Institute of Health Geneml Medical Sciences
NIH	National Institute of Health
NRC	Nuclear Regulatory Commission
NRWTP	Nonradiological Wastewater Treatment Plant
OCR	Office of Coal Research
OCRWM	Office of Civilian Radioactive Waste Management
OECD	Organization for Economic Cooperation and Development
OEEC	Organization for European Economic Cooperation
OHER	Office of Health and Environmental Research
OPEC	Organization of Petroleum Exporting Countries
ORAU	Oak Ridge Associated Universities
ORNL	Oak Ridge National Laboratory
ORR	Oak Ridge Reactor
ORSRT	Oak Ridge School of Reactor Technology
OSRD	Office of Scientific Research and Development
OWI	Office of Waste Isolation
P/T	**Partitioning/Transmutation**
PAH	Polycyclic Aromatic Hydrocarbons
PCB	Polychlorinated Biphenyls
PDA	Propylene Diamine
PETC/METC	Pittsburgh Energy Technology **Center/Morgantown** Energy Technology Center
PNL	Pacific Northwest Laboratory
PP&A	Program Planning and Analysis
PWR	Pressurized-Water Reactor
PWTP	Process Waste Treatment Plant
REDC	Radiochemical Engineering Development Center
RSIC	Radiation Shielding Information Center
RSSF	Retrievable Surface Storage Facility
SARPS	Safety Analyses Report on Packagings
SASA	Severe Accident Sequence Analysis

SERI	Solar Energy Research Institute
SETF	Solvent Extraction Test Facility
SP-Ix	Scavenging Precipitation Ion Exchange Process
SRC	Solvent-Refined Coal
SRS	Savannah River Site
STF	Small Test Facility
SWSA	Solid Waster Storage Area
TAAG	Technical Assistant and Advisory Group
TAG	Technical Advisory Group
TBP	Tributyl Phosphate
TCE	Trichloroethylene
TFBR	Tapered Fluidized-Bed Bioreactor
TMI	Three Mile Island
TNA	Thermal Neutron Analyses
TRU	Transuranium Element
TSF	Tower Shielding Facility
UNOP	Unit Operations
UT	The University of Tennessee
WEC	World Energy Conference
WIPP	Waste Isolation Pilot Plant
WMTC	Waste Management Technology Center
WMTSG	Waste Management Technology Support Group
WPS	Westinghouse Plasma Systems

1. THE CURTAIN RISES: PROLOGUE

Socrates: Why have you come at this hour, Crito?
It must be quite early.
crito: Yes, certainly.
Socrates: What is the exact time?
clito: The dawn is breaking.

Plato, 428-348 B.C.
Crito

Oak Ridge National Laboratory represents an experiment in scientific and governmental administration which is unique. It is a national institution operated by a private corporation for the purpose of furthering nuclear chemical technology on the one hand and basic research, in conjunction with southern universities, on the other.

Alvin *M. Weinberg*
History of Oak Ridge National Laboratory
April 8, 1949

Great history often results when people with unequalled talents and experience come together under extraordinary circumstances and are challenged to accomplish seemingly herculean tasks in an impossibly short time period. Such was the birth of atomic energy.

Few events in history can match the collective success story of the development of atomic energy. The management skills and enormous industrial capacity, coupled with an extraordinary pool of scientific and engineering talent, of the United States and the free world, motivated by wartime intensity to protect free society's values, accomplished the production of atomic weapons within a brief three-year period. The dawning of the atomic energy era occurred with the creation of an ultimate weapon of destruction. The creation of the atomic bomb soon evolved into a completely new source of energy production with the peacetime application of atomic energy for electric power production. The complete story involves previously unheard of international scientific and industrial collaboration. A very significant part of that story took place in an unknown southern Appalachian community now known as Oak Ridge, Tennessee. There the Clinton Engineer Works (Clinton Laboratories) was established in 1943. This facility was later to become the Oak Ridge National Laboratory (ORNL). In addition to serving as a pilot plant for plutonium production, a principal mission of the newly created laboratory complex was to develop separations processes and to apply the processes on an engineering scale to separate plutonium and other radioisotopes in sufficient quantities to permit successful development of the atomic bomb and, later, peaceful application of atomic energy. New chemistries had to be explored. New engineering techniques had to be developed. New hazards had to be understood and mitigated. The Chemical Technology Division and its predecessor the Technical Division were a central part of this vast undertaking.

This document presents a historical perspective of the Chemical Technology Division from inception to maturity.

1.1 HISTORICAL BACKGROUND

1.1.1 Early Background and the Manhattan Project

Known as the Manhattan Project, a massive scientific effort was mobilized in the United States in an effort to develop atomic weapons before our then wartime enemies Germany and Japan could do so.1 The project began somewhat tenuously as a response to a thoughtful letter from Albert Einstein to President Franklin Delano Roosevelt. Einstein's letter expressed **concern** regarding potential development of "extremely powerful bombs" through atomic energy. Thus, in October 1939, President Franklin Delano Roosevelt approved undertaking **research** on uranium. The President appointed Lyman J. Briggs, Director of **the** National Bureau of Standards, as head of the Advisory Committee on Uranium, and in early 1940 this committee recommended that the government fund limited research on isotope separation, including research being conducted by Leo Szilard and Enrico Fermi at Columbia University on chain reactions.*

In June 1940 Roosevelt created the National Defense Research Committee, chaired by Vannevar Bush, president of the Carnegie Foundation, which reorganized the Uranium Committee to increase scientific representation. Bush appointed E. 0. Lawrence, Director of the Radiation Laboratory at the University of California at Berkeley, **who also** researched electromagnetic production of ^{235}U, as advisor to Briggs. Soon thereafter, the uranium committee funded plutonium research at Berkeley (Glenn T. **Seaborg** and co-workers) and mass spectogmph work at the University of Minnesota (Alfred 0. Nier, pioneer in electromagnetic separations).*

By executive order on June 28.1941, Roosevelt created **the** Office of Scientific Research and Development (OSRD) under the leadership of Bush, who by that time reported directly to **the** president. The National Defense Research Committee under James **Conant,** president of Harvard University, became the OSRD Development Section on Uranium (code name S-l). Optimistic British reports catalyzed Bush and **Conant,** who strengthened S-l by the addition of Fermi as head of theoretical studies and Harold C. Urey as head of isotope separation and heavy-water research. In late 194 1, Bush appointed Eger V. Murphee, a chemical engineer with Standard **Oil** Company, as head of a group to oversee and supervise engineering and pilot plant studies; Urey as program chief for diffusion and centrifuge separations, including heavy-water studies; Lawrence as program chief for electromagnetic separations and plutonium studies: and Arthur Compton, from the University of Chicago, as program chief for chain reaction and weapons studies. Bush took responsibility for coordination of **engineering** and scientific work as well as approval of construction **contracts.** Roosevelt assigned all uranium policy decisions to the Top Policy Group, which consisted of Bush, **Conant,** Vice-President Henry A. Wallace, Secretary of War Henry L. Stimson, and Army Chief of Staff George C. Marshall .[2]

During the first half of 1942, Urey directed work at Columbia on **gaseous** diffusion and centrifuge systems, Lawrence directed work **on** electromagnetic separations at Berkeley, Compton directed pile experiments and plutonium production efforts at the University of Chicago Metallurgical Laboratory, and Murphree directed studies on ways to move **from** laboratory experiments to production facilities. Murphree coordinated efforts with E. I. du Pont de Nemours and Company. (**Du** Pont) and the Harshaw Chemical Company for large-scale production of uranium hexafluoride. Compton funded Fermi's pile research at Columbia, Samuel K. Allison's pile experiments at Stagg Field, theoretical work of Eugene Wigner at Princeton, and J. Robert Oppenheimer at Berkeley and moved Seaborg's plutonium work from Berkeley to Chicago?

In early 1942, Bush decided that production planning could wait **no** longer and arranged for Army participation in the S-l meetings. Roosevelt had approved **the** Army's involvement **on** October 9, 1941. Bush orchestrated transfer of responsibilities for process development, materials procurement, engineering design, and site selections to the Corps of Engineers. By general order on August 13, 1942, the Manhattan Engineer District of the U.S. Army Corps of Engineers was established, and on September 17 **the Army** appointed Colonel Leslie R. Groves to head the atomic project. Groves was a person of decisive administrative and management skills. Within two days, Groves had secured higher priority ratings for project materials and obtained an excellent production site in Tennessee.* Six days later, he was promoted to the rank of general. This major industrial, academic, and military effort became known as the Manhattan Project.[2]

1.1.2 The University of Chicago and Early Plutonium Studies

Essential to the wartime endeavor was the production and isolation of the fissionable heavy elements, uranium and the newly discovered plutonium. Using lanthanum fluoride as a carrier, Seaborg, discoverer of plutonium while at Berkeley, and his co-workers isolated a weighable amount of plutonium in August 1942 at the Metallurgical Laboratory, University of Chicago. Other separation processes under investigation at that time by several groups included use of adsorption, solvent-extraction, volatility, and peroxide oxidation. The awesome challenge assigned to Seaborg and his co-workers was to develop plutonium separations processes that could be scaled up more than a billion-fold from laboratory experiments to processes capable of being used in production plants.[1]

Du Pont agreed to accept responsibility for the design and development of a plutonium production plant and, consequently, accepted responsibility for the design and construction of a plutonium separations pilot plant at the Clinton Engineering Works in Tennessee. Although the pilot plant was to be operated by the University of Chicago Metallurgical Laboratory, Du Pont was expected to train personnel at the pilot plant for later assignment to the production facilities scheduled for construction at Hanford.* The industrial management capability of Du Pont contributed very significantly to the success of the atomic projects.[3]

Seaborg of the Metallurgical Laboratory and Charles M. Cooper of Du Pont, and their respective staffs, collaborated on establishing processes for use in plutonium separation facilities. Although Seaborg had initially favored the lanthanum fluoride carrier process, he had more recently studied phosphate processes. Research by Stanley G. Thompson indicated that bismuth phosphate as a carrier retained greater than 98% plutonium. Thus, at least two effective separations processes at the microchemistry level were available for consideration for use in the production facilities.*

1.1.3 Clinton Laboratories, Du Pont, and a Pilot Plant for Production and Separation of Plutonium

By late 1942 the decision was made to use the bismuth phosphate method for plutonium separations.* The bench-scale (actually microchemistry) process was developed principally by Seaborg and his co-workers at the Metallurgical Laboratory, University of Chicago.1 In early 1943, Du Pont began construction of the chemical separation plant in Oak Ridge, while Seaborg continued refining the bismuth phosphate method. Du Pont broke ground for the X-10 site in February 1943. The X-10 facilities would include the air-cooled graphite pile or reactor for producing plutonium, a pilot plant for isolating plutonium, and some support facilities. The chemical separations pilot plant construction started in March, and the reactor went critical and began operation November 4, 1943.[2] Several photographs of the early construction progress on the major facilities are depicted in Figs. 1.1 to 1.4.

The Metallurgical Laboratory, University of Chicago, was chosen by the Manhattan District of the U.S. Army Corps of Engineers as the contractor to manage the Clinton Laboratories.4 Although in essence the entire facility was considered a pilot plant in plutonium production and separations, among the work tasks assigned to the University of Chicago were the following:'

- Conduct the necessary studies and develop a workable and dependable method for chemically separating and isolating plutonium from uranium metal and from fission products.
- Develop a process for recovering the partially depleted uranium metal that had been irradiated and used in the development work at the pilot plant.
- Develop methods for producing certain other radioisotopes such as barium and lanthanum for use at other Manhattan project sites.

These work tasks became the. foundation from which the Chemical Technology Division later arose.

The first major facility constructed at Clinton Laboratories, X-10 site, was the graphite reactor for irradiating uranium and producing plutonium. The second major facility constructed was the pilot plant (now Building 3019 but formerly Building 205). where the process for separating

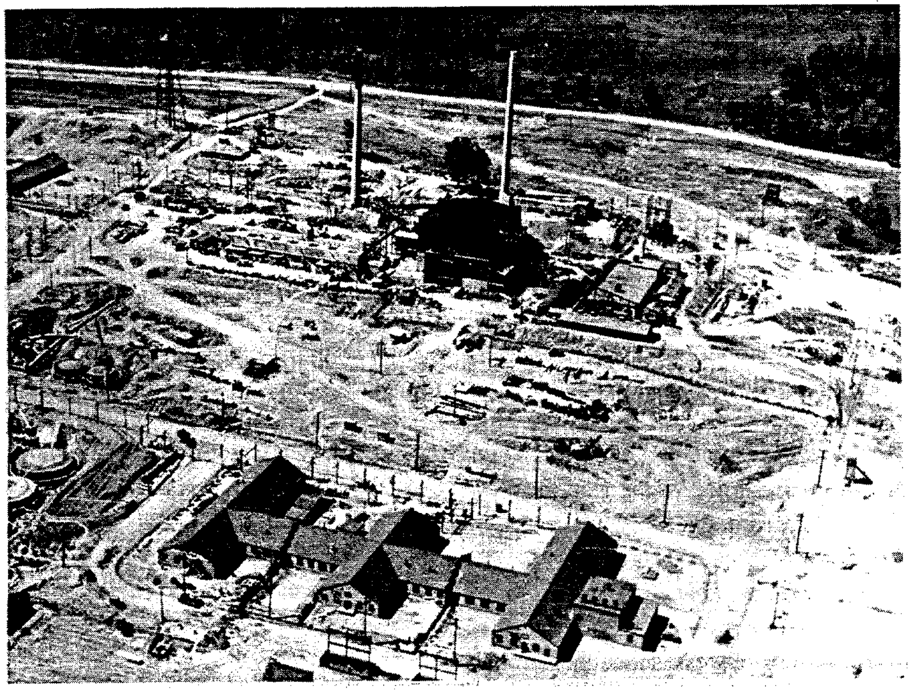


Fig. 1.1. Clinton Engineer Works, X-10 site, about September 1943, looking northwest from the main research building (709-A) In the foreground toward the graphite reactor (large dark building at upper center) and chemical separations pilot plant (to the left of the graphite reactor). Note the cells or cubicles with thick, heavy concrete shielding plainly visible in the roofless pilot plant under construction. Note also the tall stack behind the graphite reactor to release airflow used to cool the reactor and the tall stack (with the top painted black) to release exhaust gases from chemical processing in the pilot plant.

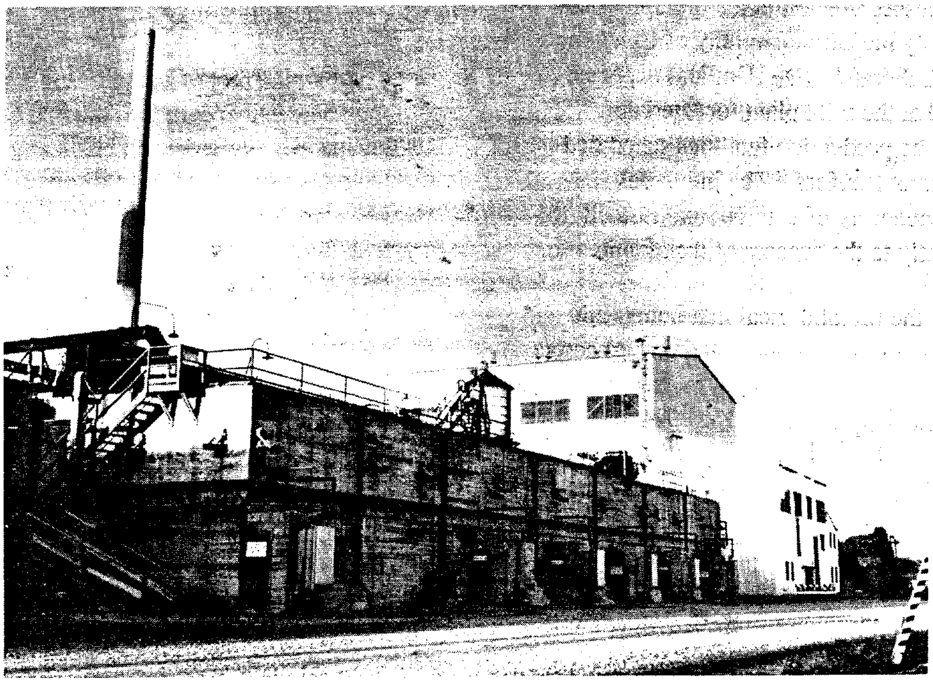


Fig. 1.2. Chemical separations pilot plant, late 1944, with graphite reactor in background.

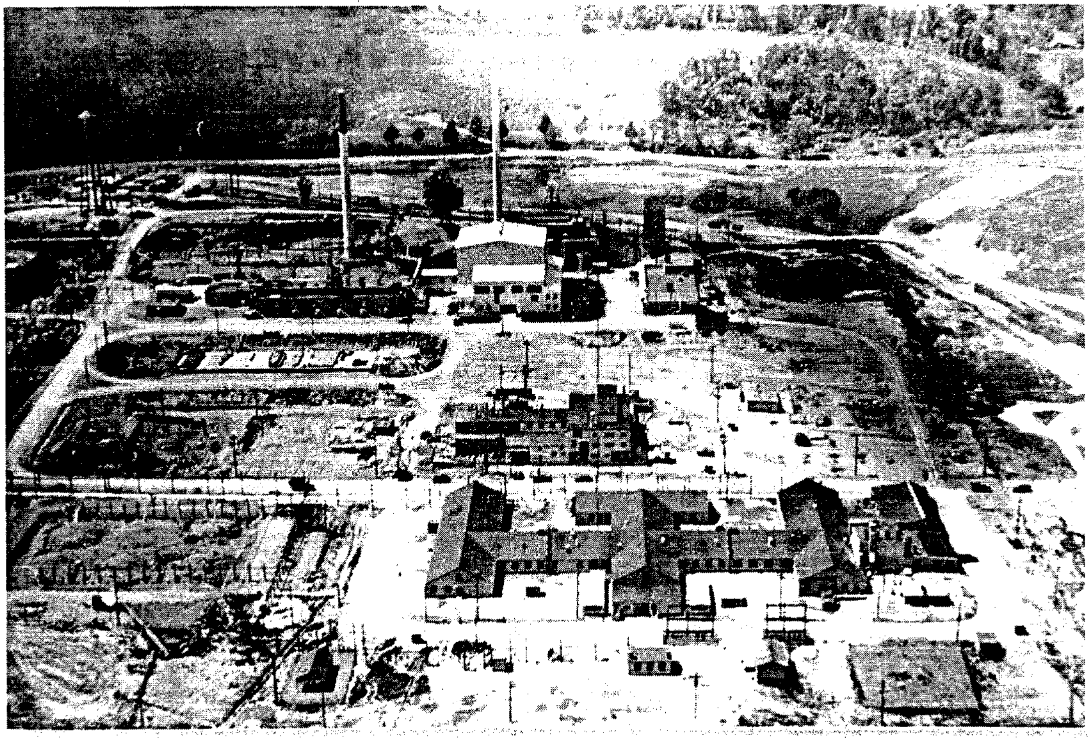


Fig. 1.3. Clinton Laboratoriesin May 1943, now X-10 site, looking northwest from the main research building (709-A) in the foreground toward the graphite reactor (large **building** at upper center now painted a metallic silver) and separations pilot plant (to the left of the graphite reactor).

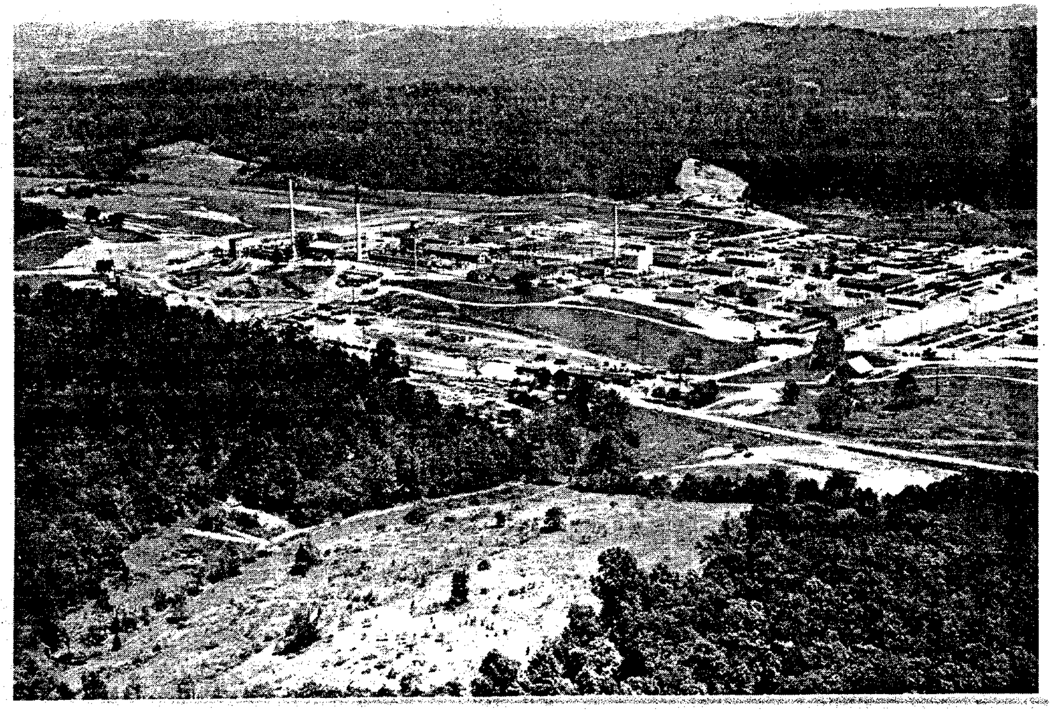


Fig. 1.4, An **overall** view, May 1947, of the Clinton National Laboratory (X-10 site) looking southeast showing the surrounding topography.

and purifying plutonium was to be tested (Figs. 1.5 and 1.6). The chemical operations would involve handling radioactive materials on a scale never before attempted. The design and construction of the pilot plant was an engineering achievement of the first magnitude. All equipment for the operations was enclosed in “hot cells” that were surrounded by 5 ft of concrete. Remote control was required for even the simplest operations. Most processing was accomplished by workers who could not even see the operation they were performing. Television, a relatively new development itself, was used at one critical stage of the process to allow observations of the operation. Performance of the equipment was determined by monitoring a bank of instruments in the operating gallery (Fig. 1.7).[1] Processing wastes were accumulated in nearby storable tanks (Fig. 1.8).

Operational testing of the pilot plant started in December 1943 using uranium irradiated by the cyclotron at Washington University in St. Louis. The low content of fission products and radioactivity associated with this irradiated uranium sample permitted relatively safe shakedown testing and troubleshooting. From the initial test runs, a few milligrams of plutonium were produced and shipped on December 30, 1943, to the University of Chicago for research use. Following the initial test runs, the system was further tested with uranium irradiated at low power in the graphite reactor. Processing of uranium irradiated at full power rapidly followed.[1]

1.1.4 Successful Plutonium Production and Isolation

Chemical separations of plutonium from the irradiated aluminum-clad uranium metal using the bismuth phosphate process at Clinton Laboratories, X-10, were so successful that plutonium needed for research was being shipped to Los Alamos by early summer 1944.2 By the end of January 1944, the chemical pilot plant was processing 0.3 ton per day of irradiated uranium and had produced several grams of plutonium by March 1944.[1] The Hanford chemical separations plants, completed in late 1944 and early 1945, were built based upon experience developed in the chemical pilot plant at Oak Ridge. The Oak Ridge pilot plant reported that bismuth phosphate was not suitable for the final plutonium concentration process at Hanford.2 The final concentration process (known as the cross-over cycle) at the pilot plant went to lanthanum fluoride precipitation, and peroxide precipitation was used to produce PuO_2 as the final product.[3] Accordingly, the Hanford separations plants adopted the lanthanum fluoride as the final concentration process as well.[2]

The Oak Ridge National Laboratory had its beginning in the pilot plant Clinton Laboratories designed to test plutonium production and separation methods. The need for purified fissionable material was urgent in order to accomplish the R&D necessary to produce a bomb. Clinton Laboratories supplied the first gram quantities of plutonium for such research and also had wartime responsibility for producing thousand-Curie amounts of the radioisotopes barium and lanthanum that were used at Los Alamos for research on high radiation sources.*

1.1.5 Monsanto Chemical Company, Reactors, and Fuel Processing

On July 1, 1945, the Monsanto Chemical Company assumed responsibility for the operation of the Clinton Laboratories, which had about completed its mission as a plutonium production pilot plant and was to begin its evolution into a nuclear energy R&D laboratory.[4] Because of the unique pilot plant and research facilities, as well as the high caliber and experience of the scientific team, the end of World War II did not also mark the end of operations at the Clinton Laboratories as originally expected. In early 1945, Oak Ridge scientists gave serious consideration to determining the most important and valid R&D activities necessary to make the greatest contributions to the new fields of nuclear science and technology. Under Monsanto, the reactor development efforts increased in scope. A major effort was devoted to R&D work leading to the design of a high-flux experimental reactor and to large-scale production of radioisotopes.

Even as early as 1943, there was interest in the possibility of developing reactors for power and research.[4] A small effort was begun in April 1944, and by July, a graphite-moderated reactor had been conceptualized that used stainless-steel-clad uranium-carbide fuel and high-temperature, high-pressure water to generate steam for powering turbines.4 As early as 1944, a homogeneous reactor using uranyl sulfate, enriched in ^{235}U, as fuel with a thorium reflector was proposed as a research tool to investigate breeder concepts (irradiating thorium to produce ^{233}U) and, also, to determine the

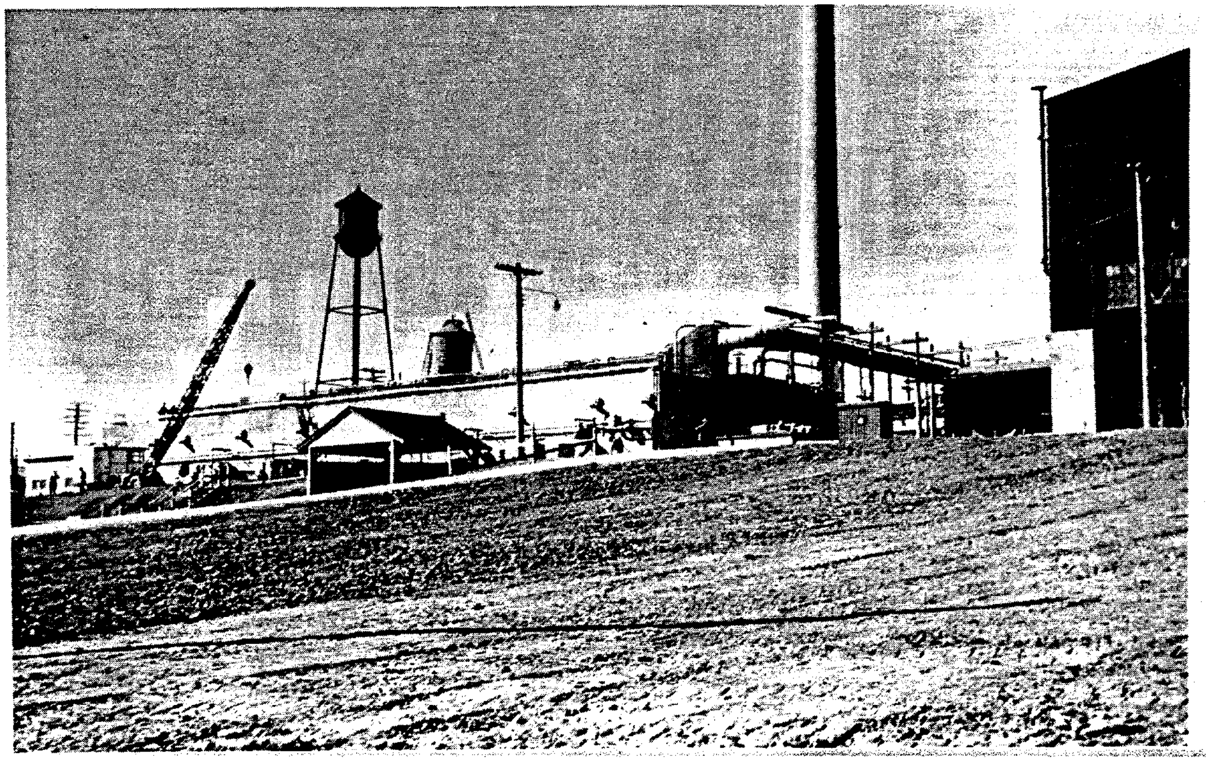


Fig. 1.5. A November **11, 1943,** view (looking northwest uphill) of the pilot plant, Building 205. A corner of the graphite reactor building is visible on the right and the tall exhaust stack behind the **pilot** plant.

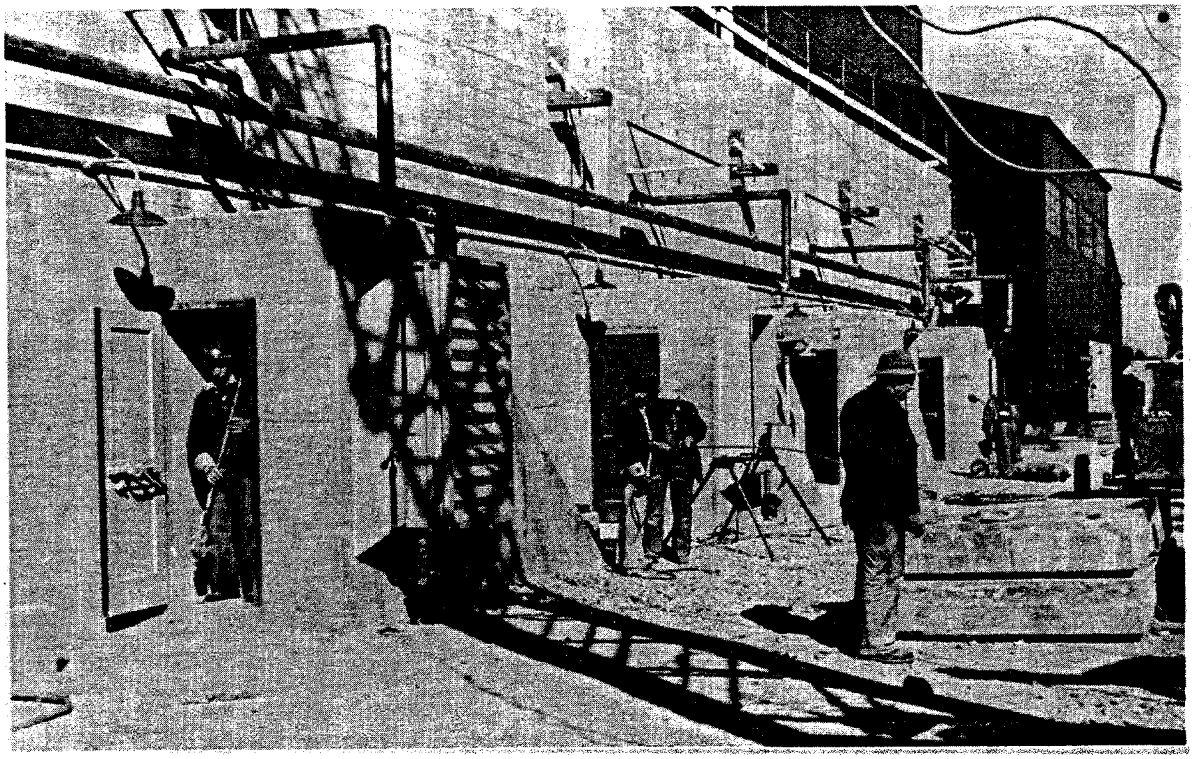


Fig. 1.6. A **close-up** view of pilot plant cell doors and construction **activities** (November **11, 1943**). The graphite **reactor** building is visible on the right.

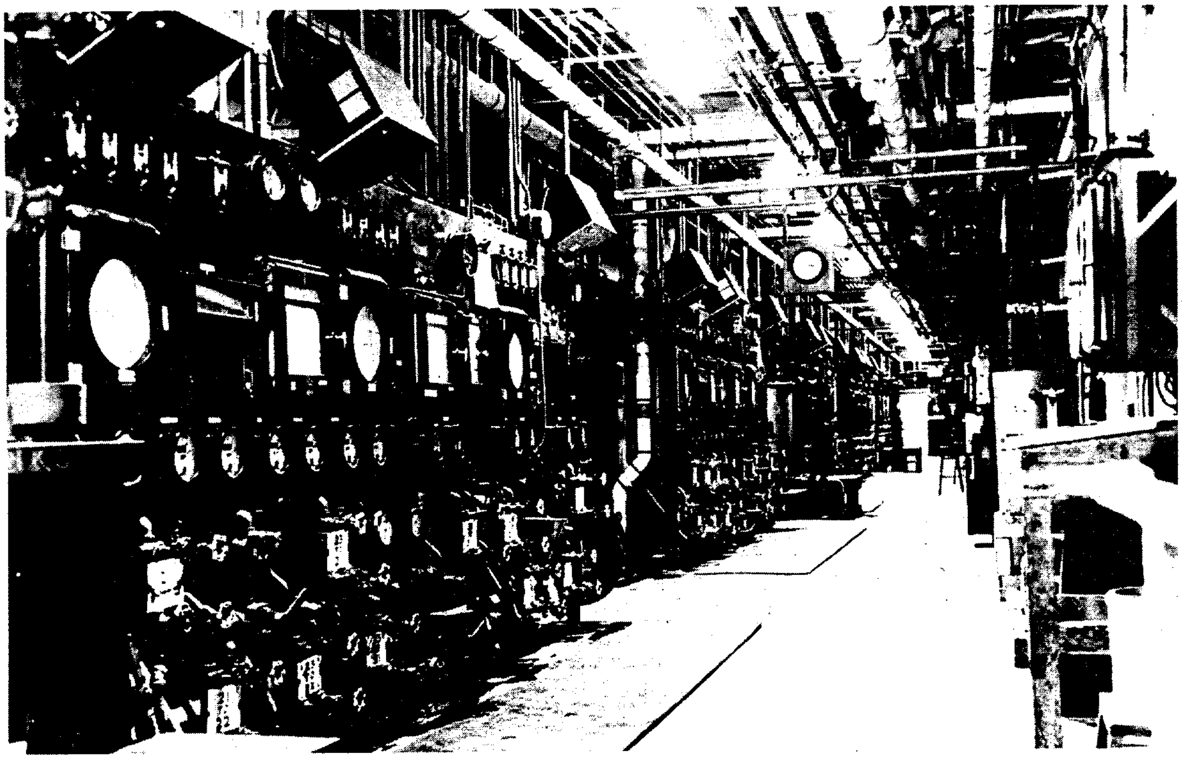


Fig. 1.7. Main control room for the chemical separations pilot plant.

Fig. 1.8. A July 14, 1943, view (looking northeast uphill) of the process waste storage tanks during construction. The completed tail exhaust stack, the pilot plant under construction, and the graphite reactor building under construction are visible In the upper left of the picture.

general feasibility of homogeneous reactors.[1,4] Work on the homogeneous reactor was discontinued in 1945 in favor of developing a heterogeneous high-flux reactor. During 1946 and 1947, a substantial development program was undertaken in support of the high-flux reactor design.[4]

The work on separations processes at the Clinton Laboratories was shared between the, Technical and Chemistry divisions and involved the following:

- Separation, decontamination, and recovery of ^{235}U from fission products
- Separation of ^{233}U from irradiated thorium
- Recovery of uranium and plutonium from large volumes of stored waste solution that resulted from the dissolution and chemical processing of graphite reactor slugs

The separations research involved solvent extraction, ion exchange, radiation effects, and analytical methods.'

1.1.6 The Atomic Energy Commission and the Clinton National laboratory

On January 1, 1947, the atomic energy activities were transferred from the Manhattan Project (Manhattan District of the U.S. Army Corps of Engineers) to the newly created Atomic Energy Commission (AEC). The name of the Clinton Laboratories was changed to the Clinton National Laboratory. In December 1947, the AEC consolidated reactor development activities at the Argonne National Laboratory (ANL) and announced plans to maintain the Clinton National Laboratory as a center for basic research, applied chemical research, and isotope production and research. The AEC program included a vigorous effort in applied chemical engineering directed toward the solution of current problems that were vital to the atomic energy operations. The Clinton National Laboratory was to be developed into a center of chemical technology for atomic energy activities.[1] However, moving all the reactor development work to ANL proved to be impractical, and design and development work on the Materials Testing Reactor (MTR) to be built in Idaho continued in Oak Ridge until the reactor was completed in early 1952.[4]

1.1.7 The Oak Ridge National laboratory and Union Carbide

On March 1, 1948, the name of the laboratory was changed to the Oak Ridge National Laboratory (ORNL), and Carbide and Carbon Chemicals Company (a division of the Union Carbide and Carbon Corporation) assumed responsibility for its operation. Carbide had a strong corporate background in chemical engineering research and development[1] and was already the operating contractor for the gaseous diffusion plant (K-25) and the electromagnetic separations plant (Y - 12). In early 1950 a mockup of the MTR reactor was operated at ORNL at very low power to measure neutron and gamma fluxes. This mockup evolved into the Low-Intensity Test Reactor (LITR).[4] In 1949, Alvin Weinberg, director of the ORNL 'Technical Division and also associate director of ORNL, had suggested that a new look be taken at aqueous homogeneous reactors. Thus, construction of Homogeneous Reactor Experiment I was begun in September 1950, and the reactor was completed in January 1952. At approximately the same time, a new program of reactor development, the Aircraft Nuclear Propulsion (ANP) program, began. By 1950, the programs in the ORNL Technical Division included the three reactor programs as well as chemical processing. On February 1, 1950, the Technical Division was divided into the Reactor Technology Division, with Alvin Weinberg as director, and the Chemical Technology Division, with Frank Steahly as director!

1.2 ESTABLISHMENT OF THE CHEMICAL TECHNOLOGY DIVISION

1.2.1 Background

As facilities became available during the summer and fall of 1943, a few people were transferred from the Metallurgical Laboratory at the University of Chicago and from some of the Du Pont plants to Clinton Laboratories to staff an Engineering Development Section headed up by Miles Leverett, Section Chief. Early in 1944, the industrial staff was supplemented by personnel from the Special Engineering Detachment of the Army. The Engineering Development Section collaborated with physicists and metallurgists to upgrade the graphite reactor performance and, through simulation with the graphite reactor, studied the performance anticipated from the Hanford reactors under construction. The design group of the section collaborated with the Separations Development Section that supported the operation of the chemical processing plant Collectively they designed a process and equipment for ion-exchange extraction of plutonium from uranyl nitrate solutions. A mock-up was built and tested. In April 1944, it was decided that the uranium should be recovered from the process wastes that had been discharged by the chemical processing plant into underground storage tanks. Design of a solvent extraction process and pilot plant was initiated. The chemical processing plant had fulfilled its mission of demonstrating the bismuth phosphate process for extracting plutonium from reactor fuel and was being shut down. Personnel were being sent to operate the processing plant at Hanford. In October 1944, the remaining staff of the Separations Development Section, responsible for the operation of the chemical processing plant, and the Engineering Development Section were joined to form the Clinton Laboratories' Technical Division, comprised of about 50 chemists and engineers. Jim Lane was assistant director of the division, with M. D. Peterson as chief of Process Development (Section I), R. B. Briggs as chief of Engineering Development (Section II), R. Ward, as chief of Process Design (Section III), and Don Reid, as chief of the Pilot Plant. Miles Leverett was the division director. (See Appendix A for a 1948 organization chart of the Technical Division.[5])

On February 1, 1950, the Chemical Technology Division (Chem Tech) was established at ORNL, and Frank L. Steahly was selected as its first director.6 The personnel comprising the new division formerly functioned as the Chemical Technology Department of the Technical Division. By mid-1950, the new division employed 110 people and consisted of four sections: Laboratory, Frank R. Bruce, Section Chief; Unit Operations, J. 0. Davis, Section Chief; Process Design, Floyd L. Culler, Section Chief; and Pilot Plants, D. G. Reid, Section Chief. Many of the early Chem Tech staff achieved national and international prominence in the nuclear energy area as well as rising to important leadership positions at ORNL.[7,8] As an aid in tracing the historical development and changing missions of the division, organization charts for 1950, 1960, 1970, 1981, and 1991 are presented in Appendix B.

The initial major activities of the new division included design of the Idaho Chemical Processing Plant (ICPP), development of the tributyl phosphate (TBP) process for the recovery of uranium from Hanford metal waste, development of the Purex process for the recovery of uranium and plutonium from Hanford-irradiated metal, development of the RaLa process for separation of barium from fuel units, homogeneous reactor fuel studies, and design and construction of the ORNL Metal Recovery Plant (Building 3505).[7-9]

This brief description of initial work involvements suggests that Chem Tech was at the forefront of international activities in nuclear energy studies, and indeed it was. These initial endeavors established a solid scientific basis for separations processes used not only nationally but also internationally in the fledgling nuclear energy industry.

1.2.2 Management and Organizational Evolution of Chem Tech

Metallurgical Project. The minutes of the Metallurgical Laboratory and Project Council meetings provide considerable insight into the early organization and management of the Manhattan Project activities under the purview of the Metallurgical Laboratory. The activities were complex, diverse, and widely separated geographically. They included those at the University of Chicago: the X site (Clinton, Tennessee); the Argonne, Illinois, site; the Ames, Iowa, site; and the W site (Hanford, Washington); some activities at the Universities of California, Notre Dame, Indiana, Washington (St. Louis), Princeton, and Columbia; and included coordination with DuPont and the U.S. Army.[10,11]

Arthur H. Compton, recipient of the Nobel Prize in Physics in 1927, was selected Director of the Metallurgical Laboratory and Project. Besides being technically competent, he was apparently a sound administrator and good businessman. Compton guided the research-oriented university-type team toward very practical considerations necessary for plutonium production. For secrecy pruposes, plutonium was referred to as "49" in the University of Chicago reports.

Engineering Considerations become Principal" Concern of the Metallurgical Project. In a January 1943 memorandum concerning organizations of the "Metallurgical Unit," Compton defined the Metallurgical Laboratory Organization and duties of the various leaders. Soon after, Du Pont accepted the contract for design, construction, and operation of "the production unit associated with our project." Compton stated that

> we are now engaged in cooperation with Du Pont upon the project of producing "49" in quantity and quality suitable for military use at the earliest possible moment . . . . Since the chief claim for the practical consideration of our project in comparison with others lies in the possibility of swifter completion, the direction which the design, construction, and operation program must take must be determined by engineering considerations based on the studies we have already completed. . . . our major responsibility from now on must be to supply the Du Pont engineers and operators with the technical information they need. . . and to maintain constant liaison with their staff while our work progresses to insure that the design at all stages meets the nuclear physics requirements. In order to meet our new responsibilities, it will be necessary for us to operate under a more specifically defined organization and under a more rigidly observed procedure than has been necessary or advisable in the past. Both the Army organization and that at Du Pont are of necessity complex. Unless we have a well-defined organizational procedure which is thoroughly understood by both the Army and by Du Pont, serious delays due to confusion and misunderstanding are bound to occur.

The Metallurgical Laboratory was directly responsible for its research program through the chairman of the S-l committee to the director of OSRD. This provided administrative flexibility and convenience of an ORSD contract to the University of Chicago as compared with a direct Army contract. Matters of general policy were cleared through Compton to A. V. Peterson, the Army representative for the Manhattan District. Work at Du Pont had been set up in a new Division of Security Materials (DSM) under the Du Pont Explosives Department. Matters of policy were to be cleared through Roger Williams, assistant general manager of the Explosives Department, who was in charge of the DSM division, and C. H. Greenewalt was named technical director of the division. It was agreed that all official information and decisions from the Metallurgical Laboratory would channel through Compton to Greenewalt and, conversely, requests for information through Greenewalt to Compton.' 1

Laboratory Council as Management Team. A laboratory council was established consisting of laboratory and division directors and the Du Pont and Army representatives. The council had two major functions: (1) keep the administrative staff informed concerning the current status of the Laboratory's program and (2) make recommendations on policy matters. The council held a weekly meeting on policy issues and a weekly meeting on technical issues.11

Analysis of Factors Contributing to Project Success. A large measure of the success of the Manahattan Project is attributable to this group of dedicated scientists and engineers who were able to pull together and integrate the many disciplines and areas into the warp and woof of a finished fabric. The group was successful because each member was (1) technically sound, (2) a good manager, and (3) a team player dedicated to the accomplishment of assigned objectives. These attributes also seem to be the principal elements of the early successes achieved by the AEC and the national laboratories.

The organization of the Metallurgical Laboratory11 is presented in Appendix A. It is interesting to note that the organizational concept of overall director, associate director: and divisions with division directors, section chiefs or heads, and group leaders is still essentially the same organizational style used by the ORNL and Chem Tech. M. D. Whitaker, who later served as director of the Clinton Laboratories (now ORNL), is shown as Sub-project Director for Site X; R. L. Doan, who later served as Associate Director for Research at Clinton Laboratories, is shown as Chief Administrative Officer for Laboratory Operations: and E. P. Wigner, Research Director of Clinton Laboratories in 1946 and recipient of the Nobel Prize in physics in 1963, is shown as the section chief of theoretical physics. Miles Leverett, who later became director of the Technical Division (the forerunner of the Chemical Technology Division) for Clinton Laboratories, is shown as the section chief of Development Engineering; and C. D. Coryell and George Boyd, who later served as group leaders in the Chemistry Division at Clinton Laboratories, are shown as section chiefs of By-Products and Analytical Chemistry Control, respectively.

"X" Research Program. In July 1943, in order to facilitate the setting up of a comprehensive research program at "Site X," the following program activities were established:[12]

- Operation and interpretation of the X pile
- Design, construction, and operation of the W pile
- Operation of the X separation plant
- Design, construction, and operation of the W plant
- Concentration of products
- Study of physical problems
- Study of fission products
- Study of new separations processes
- Study of the effect of radiations and products on biological tissue
- Study of the health of all employees

Future Chem Tech Staff Members. All personnel involved in the Metallurgical Project were listed in an August 1943 report.[13] Research workers were scattered all over the University of Chicago quadrangles, including the north and west stands of Stagg Field, an old ice house, and a former beer brewery. Site X personnel included M. D. Whitaker, Director: R. L. Doan, Director of Research; Lyle Borst, Assistant to Director of Research; Henry Newson, Physics Section Chief; Warren Nyer, Research Assistant: J. G. Stangby; and Pearl Margolis. Eight employees had already moved from Chicago to Site X: Jean Ashin, Imgrad Boeder, Melba Johnston, Haydn Jones, Wilcox Overbeck, Gerard Pawlicki, Howard Parsons, and E. Shapiro. Some future Chem Tech scientists as well as several ORNL staff members were working as research assistants in various sections: John P. McBride in the Chemistry Division, C-II group; George Parker in the Biology Division working with Waldo Cohn; Al Rom in the Technology Division, Section T-III: and Ed Frederick in the Technology Division, Section T-VII.

Cooperation and Communication. Management of the multi-disciplines and multi-sites involved in the Metallurgical Project became increasingly complex with the growth in worker numbers and sites. Compton and the council stated clearly the need for the closest possible cooperation between Chicago and Clinton, with information flowing freely. The following statement was prepared for the laboratories at Clinton, Ames, and the P-9 Project.

> The Laboratory Director will be a member of the Project Council and will regularly present program to council for their information and recommendation. It shall be his duty to arrange a procedure subject to the approval of the Project Director (i.e., Compton) whereby the men in his organization shall make full use of information and guidance by leaders in their technical fields elsewhere in the Project.

The statement helped directors to obtain a clear concept of what was expected of them, notified the Army that full transfer of information was needed, and emphasized availability of assistance from the most experienced and knowledgeable scientists and engineers.[14]

Clinton Laboratories Semi- Works, Chemical Engineering Section, and Separations Development Division. By October 1943, Whitaker reported to the council that the laboratory would need a ton of "Argonne metal" by November 15, and Fermi said it would be available. Leverett was investigating large-scale production of radioactive xenon. Compton scheduled for the next meeting further discussion on Health Group reports and the large level of effort required. 0. H. Greager reported to the council that 40 men were in the semi-works groups at X with about 15% of the work on alternate processes. [15]

The initial report of the Chemical Engineering Section at Clinton Laboratories dealt with dissolution, extraction, decontamination, and concentration of plutonium and uranium along with wet fluoride investigations and miscellaneous engineering process studies. 0. H. Greager was director of the newly formed section, M. F. Acken was the section chief, and R. S. Apple was associate section chief. In 1944, many staff members from this section, which later became the Separations Development Division, went to Hanford to operate the large fuel processing facility,[16] and the remaining staff members were merged with the Technical Division.[5]

Chemistry Division. Warren C. Johnson, division director, was selected by Compton and the Metallurgical Laboratory Council as the strong chemistry leader necessary for the project to succeed at Site X. Johnson pulled together teems in five major areas: Section C-I, chemistry of plutonium and uranium, Spafford English and Ray Stoughton as section chiefs: Section C-II, fission product chemistry, C. D. Cot-yell, T. H. Davies, and H. A. Levy as section chiefs: Section C-III, separations processes, George Boyd as section chief; Section C-IV, preparation of radioisotopes, Waldo Cohn as section chief; and Section C-V, analytical chemistry, D. N. Hume as section chief. In a March 1945 progress report, it was noted that several future Chem Tech staff members were working in the Chemistry Division: Section C-I, John Blomeke was working on Hanford process problems and Frank Steahly and John McBride on the isolation of ^{233}U from thorium; Section C-III, Bob Klotzbach and E. Nicholson on the preparation of active lanthanum; Section C-IV, George Parker on the preparation of radioisotopes from fission materials; Section C-V, Frank Bruce, Clair Scherston, Ed Beauchamp, and George Sadowski on analytical service and research and development.[17]

Technical Division. Elements of each of the two major divisions, Separations Development Division and Chemistry Division, were merged into the Technical Division from which the. Chemical Technology Division later evolved. In an October 1944 progress report, it was noted that several future Chem Tech staff members were working in the Technical Division. D. G. Reid was studying the corrosion of Hanford aluminum tubes and slugs underradiation; Dave Overholt was studying the disposal of stack gas; Fred McCullough was studying the detection of blisters on fuel elements and also the preparation of ^{140}La; M. D. Peterson was providing general plant assistance; and Warren Eister (Group Leader) and Joe Savolainen were working in the semi-works product recovery.[18]

A year later, future Chem Tech staff members were reported as follows: Savolainen, ^{140}Ba chemical development; Eister, ^{140}Ba equipment development and semi-works; Leuze and Frederick, ^{140}Ba analytical service; Peterson, chemical development: Reid, extraction process engineering development: Nicholson, extraction process chemical plant design; Eister, Blomeke, Klotzbach, Morse, Overholt, Steahly, and Savolainen, extraction process chemical plant development; and McCullough, pile design.19

1.3 AN EARLY PERSPECTIVE

1.3.1 Clinton laboratories-the War Years, William E. Thompson[20]

Fifty years ago, on February 1, 1943, construction work was started at the X-10 site. Looking back to those days, we can only feel amazement at the boldness with which the wartime atomic energy projects were planned and at the speed and success with which they were carried out.

In 1942, even before the first nuclear chain reaction had been achieved, the Corps of Engineers purchased 92 square miles of land in the area between Clinton, Oliver Springs, and Kingston, under the guise (for security reasons) of establishing the Kingston Demolition Range. This area was given the name Clinton Engineer Works and was intended for the large-scale production of enriched ^{235}U and Pu needed for atomic bombs. (The name Oak Ridge was not adopted until June 1943.) The army had originally planned to carry out all atomic bomb project activities at this site. Later, it was decided to locate the plutonium production facilities at a more remote site on the Columbia River near Hanford, Washington.

Chronology of Significant Events. The following chronology illustrates the rapid pace of early activities:

- In August 1942, the Manhattan District was organized under the Corps of Engineers to carry out the large-scale construction and production activities of the atomic bomb project.
- In September 1942, Brigadier General Leslie R. Groves was placed in complete charge of the

Mr. William E. **Thompson,** 1973

Manhattan District. General Groves selected the Du Pont Company to design and **construct the** Clinton Laboratories plutonium pilot plant and the **production** facilities at Hanford.

- On December **2, 1942,** Enrico Fermi and his colleagues on the Metallurgical Laboratory staff achieved the **first** nuclear **chain** reaction in the Chicago Pile.
- On December **16, 1942,** the nation's Military Policy Committee recommended to President Roosevelt that a plutonium production pilot plant be **constructed at** Clinton Engineer Works. The President approved.
- On January **15, 1943,** E. I. du Pont de Nemours and Company of Wilmington, Delaware, was selected to design and construct the plutonium pilot plant facilities in Tennessee, to be designated as Clinton Laboratories. The Du Pont Company arrived at the specifications of the Clinton Pile on the basis of recommendations **made by the** Metallurgical Laboratory **staff,** particularly by Eugene Wigner and Alvin Weinberg of the Theoretical Physics Group.
- By February **1, 1943,** Du Pont was breaking ground for the "Clinton Pile" (now the Graphite Reactor) at a rural site which was more than **five miles from the nearest electric power** line.
- By February 1943, Du Pont engineers were designing the plutonium separation pilot plant, and in Match, excavation for the building foundations was under way.
- The pile was complete in less than eight months, and the University of Chicago physicists had begun pre-operational testing. The plutonium separation pilot plant was nearly complete, and testing of the equipment was started in that facility also. Frank Bruce (later a Chem Tech staff member and Associate Director of ORNL), who at that time worked in the analytical lab in the pilot plant building, remembers being impressed by the smoothness and the businesslike manner with which the Du Pont people carried out the pre-operational tests and check-out procedures. This first large-scale radiochemical processing facility had 5 ft of concrete shielding around all the processing equipment, making it **necessary** to employ remote controls and to operate the system without being able to see it; but they made it work, right from the start.
- The pile started up in November 1943, reaching criticality at **5:00** a.m. on November 4, almost exactly nine months after ground was broken. Arthur Holly Compton, director of the Met Lab, was spending considerable time in Oak Ridge in those days. Ernie Wollan recalls coming down by train from the Met Lab for the pile start-up with a group that included Enrico Fermi and Norman Hilberry. Wollan looked for neutron leakage **through the** top of the shield and found the shielding to be adequate, as predicted. He also checked for neutrino emissions, which were expected to pass easily through the shield, if they were present. He established that the highest possible number of **neutrino** emissions anticipated could not present a **radiation** hazard.
- On December **19, 1943,** after the pile had been operating a few weeks, the first "hot" runs to separate plutonium in the pilot plant were conducted. Among those operating the pilot plant at the time were John Gillette, Harris Blauer, Roscoe Pressley, Stanley Rimshaw, Harvey Mahlman, D. C. King, and Claude **Keck** (later a Chem Tech staff member). The early research and development on fission, uranium isotopes separation, plutonium production, and **on** related matters had been performed mainly at Columbia University, the University of Chicago, the University of California, and Iowa State College under the **Office** of Scientific Research and Development, directed by Vannevar Bush.
- The first batch of irradiated fuel **slugs was** taken from the reactor to the chemical processing dissolver on December 20, 1943.
- On January 3.1944, the first plutonium (1.54 mg) was shipped from Clinton Laboratories to the Metallurgical Laboratory. Although the chemical processing proceeded in batches, it was continuous in the sense that a new batch was started as soon as the previous batch had been moved to the next step in processing. The **final** purification of plutonium was done in the Chemistry Building by I. Perlmau's group, which included Ray Stoughton, John McBride (later a Chem Tech staff member), Ed Bohlmann, and Joe **Halperin.**
- By the end of January **1944, 1/3** ton per day of irradiated fuel from the reactor was going through the pilot plant, although the low pile power level and short operating time had not allowed the plutonium concentration to build up to the planned levels. By **March the**

production rate was up to about 8 to 10 g per month of purified plutonium. Through May 1944, shipments had totaled 30.737 g, and soon afterward, higher production rates reflected the increase in pile power.

- From December 1943 to January 1945, the pilot plant processed 299 batches of slugs: about 100 tons at 1/3 ton per batch. An objective had been set to produce about 300 g of plutonium, and by the end of 1944, shipments had totaled 271.396 g of plutonium. Accordingly, plans were made for closing down the chemical pilot plant early in 1945, by which time more than the originally planned 300 g would have been produced. The final regular shipment in January 1945 brought the total plutonium up to 289.438 g, and additional plutonium reclaimed and purified in the process of closing down and cleaning out the pilot plant equipment was shipped in February 1945, bringing the grand total of plutonium production from the Clinton Laboratories pilot plant to 326.390 g. Two years after the start of construction, the objectives had been accomplished far beyond the original expectations.
- The project cost was $12 million for construction of all facilities at Clinton Laboratories, plus $12.5 million for all operations through June 1945.

Historical Footnotes, The first of the production buildings to go into operation at Clinton Labs was the graphite machining ship, where craftsmen took the extruded graphite bars as they came from the manufacturers and machined them to final specifications for stacking in the pile. Finished graphite pieces went into exponential piles, from which physicists could then measure neutron diffusion lengths using indium foil. Exponential pile experiments took up July and August; in September, crews began stacking the 676 tons of graphite in the pile by hand. This took three weeks.

The design power level of the Clinton Pile, 1000 kW, had been chosen with the knowledge that it would produce about 1 g of plutonium per d at that power level. Designers wanted the concentration of plutonium in the irradiated uranium fuel to be at least 1 ppm. Expecting the pile to contain about 60 tons of uranium (approximately 60 million grams), they figured that after a couple of months at 1000 kW the fuel, would contain 1 ppm of plutonium. They planned to process the irradiated fuel in 1/3-ton batches at the rate of one batch per d, so the nominal plutonium production capacity of the pilot plant at Clinton Laboratories was 1/3 g per d.

Higher airflows, coupled with the other improvements, permitted routine operation at power levels up to 4000 kW. This increase to four times the design power level gave a corresponding increase in plutonium production. Even at this power level, there were no difficulties with the operation of the pile. In ease of control, steadiness of operation, and reliability of performance, the Clinton Pile achieved an impressive record. There were no failures attributable to mistakes in design or construction-a remarkable fact, considering that this plant was designed on the basis of the meager data available in 1942 and was constructed without previous experience.

Stone and Webster Engineering Corporation had originally been selected as the overall engineering and construction firm for the Manhattan District, but it soon became apparent that the various parts of the work were too widely separated physically and too complicated technically to be handled by a single company. Stone and Webster built the Y-12 Plant and some of the townsite facilities, Du Pont built the X-10 facilities, and J. A. Jones Construction Company the K-25 complex. Many subcontractors came in to build the houses, dormitories, and miscellaneous building of the town. Roane-Anderson Company, a special subsidiary of Turner Construction Company, was the rental and maintenance agent for the houses, dormitories, and the commercial property on the Clinton Engineer Works townsite.

On that February day in 1943 when construction started, several farms still occupied the X-10 site. The Bethel Valley road was in existence, along its old route north of the present road, but it was not paved. Solway Bridge and the old wooden-planked Edgemoor Bridge were standing, but there was only a ferry at the White Wing (State Highway 95) crossing of the Clinch River. J. A. Jones Construction Company installed a pontoon bridge to replace the ferry in 1942, and this bridge continued in use, with some modifications and improvements, until the present bridge was built in 1963. Railroad spurs were built by the Louisville and Nashville (L&N) Railroad to serve the townsite and the Y-12 Plant and by the Southern Railway to serve the K-25 Plant, but there was no railroad spur to the X-10 site. All materials for the Clinton Laboratories had to come in by truck. Before Oak Ridge's rail spurs were built, Byington, Tenn., a small community near Kams, was the railroad destination of many L&N shipments.

Finding enough construction workers to build all the plants of the Clinton Engineer Works plus the town was a chronic problem in 1943. The construction fell behind schedule by several months because of the shortage of workers. Since part of the trouble was lack of living quarters close to the job, the Scarboro School was once again brought into use for a while as a barracks for workers. To recruit a labor force, John Fiser, at that time a Clinton Labs personnel officer, drove a bus through rural areas of Georgia, Alabama, Mississippi, and Tennessee, not only signing up people to work in Oak Ridge, but bringing them back with him in the bus as well.

The mud and dust of 1943 and 1944 are outstanding characteristics of Oak Ridge in everyone's memories; Larry Riordan recalls that in 1943 the Medical Department issued face masks to workers who experienced difficulties because of the dust. It was not until after the war was over and a more permanent role for Oak Ridge began to emerge that paving of the roads was started. Prior to that, Roane-Anderson leveled the town's streets with road graders from time to time, treating them with calcium chloride to allay the dust and with gravel to combat the mud. The roads on the Clinton Labs site itself were paved as a part of the program started in 1948 to make the place more permanent.

There was no air-conditioning of buildings during the war years, not even with window units. There were lots of electric fans, but when the dust outside made it impossible to have the windows open, the summer heat and humidity could be pretty oppressive. Some laboratories and instrument rooms were air-conditioned to protect electronic components from the temperature fluctuations and the high humidity of East Tennessee, but it was not until the 1950s that AEC construction criteria permitted the air-conditioning of office areas. Summer temperatures above 100°F in office and work areas were not rare. I can remember papers sticking to sweaty arms while working at my desk. We used lots of onionskin paper for making carbon copies, and it stuck to damp skin worst of all. Carbon copies of everything was the rule in those days, and often the number of carbons would be as high as possible. It took really strong typing fingers to make eight or ten carbons. When electric typewriters became generally available in the early 1950s, they could only be obtained to replace manual machines if there were special factors to justify the higher cost. The frequent need to make carbon copies was the justification most people used to try to get an electric typewriter.

1.3.2 Miles C. Leverett, Director, Technical Division 19431948, ORNL[5]

In the beginning, the Clinton Laboratory was created primarily in response to the recognition by the chemical engineers at the Metallurgical Laboratory and those in Du Pont that there had to be a pilot plant in which the chemical separation of plutonium from the irradiated uranium and fission products could be carried out. These engineers were quick to realize that many new, unique, complex engineering problems would arise in transferring processes from the laboratory to the plant; these would have to be solved before the Hanford plant could be designed and run successfully. Therefore, the centerpiece of the laboratory in its early years was the chemical separation plant The graphite reactor was a necessary adjunct, since no other source of irradiated uranium was available: however, the Hanford reactors could have been and largely were designed with little reference to the graphite

Mr. Miles C. Leverett, 1949

reactor. The chemical laboratories and their staffs were likewise vital support for the successful operation and problem solving in the 200 Area as the separation plant facilities were known, but chemical research was not otherwise a raison d'etre for the Clinton Laboratory. Similarly, physics was a supporting function for the graphite reactor, not an independent function of the laboratory in those early days. I do not, of course, mean to demean the enormous contributions of the chemists and physicists at the Metallurgical Laboratory, where the leadership in chemistry and physics resided in this period; I wish rather to emphasize that the initial function of the Clinton Laboratory was primarily a chemical engineering one.

A few illustrations of the kinds of chemical engineering problems faced may be helpful.

- Equipment capable of performing liquid transfer, liquid/solid separation, solids dissolution, evaporation, toxic off-gas disposal, etc., remotely, had to be designed, built, and tested
- Instrumentation capable of remotely measuring volumes, densities, temperatures, etc., in a lethally radioactive environment had to be found or developed.
- Procedures for remotely sampling, transporting, and analyzing liquids and solids remotely had to be devised, built, and proven practical.
- Techniques for separating, in relatively pure form, sub-gram quantities of solids from volumes of liquid thousands of times larger had to be perfected.
- Previously unknown effects of intense radiation on solvents such as were used in the process had to be coped with when they suddenly appeared.
- Heat from radioactive decay could create thermal convection currents which interfered with sedimentation processes.
- Techniques for maintaining, decontaminating, and disposing of contaminated equipment had to be devised.
- The problem of safely storing unprecedented quantities of radioactive waste had to be addressed.

Problems of these types were of little importance as long as only low-level radioactivity was being handled on a laboratory scale, but they were of great importance when the chosen separations processes were scaled up to plant scale. Suggestions for ways of dealing with these problems came from many sources, but it was the chemical engineers who had to make the tough choices and devise the ingenious solutions.

The design, construction, and successful operation of a facility for producing ^{140}Ba in kilocurie quantities (in Building 706D) in just a few months time was a job handed to me in 1945. Barium- 140 was urgently needed at Los Alamos. With great support from the Chemistry Division, my team of chemical engineers and I met the desired schedule and continued to produce the product as long as it was needed. I believe that this was the first production of a radioisotope on a large scale. Associated with me was Walton Rodger, among others.

During the war, a large contingent of young, technically trained soldiers was assigned to Oak Ridge and was used in numerous capacities of a technical nature. Some members of this group went on to achieve notable success in nuclear work, including Sam Beall, Beecher Briggs, Bill Bigler, Bernard Manowitz, and Milton Levenson. These men were students who had been drafted, while still in school, into the Army Specialized Training Program (ASTP). Several of them worked in my group and did more than their share.

The Du Pont people assigned to Oak Ridge for training in preparation for their later transfer to Hanford were of unusually high caliber. They deserve special mention in any account of the early success of the laboratory. Several of those people were assigned to my group.

Although, as noted already above, there was no Chemical Technology Division as such until one was created by Warren Johnson, there was, from the first days of 1943, a group of chemical engineers charged with carrying out semi-works-scale developmental and trouble shooting work on the chemical separations processes. For much of the time, this work was directed by Merlin Peterson; later it became my responsibility.

I arrived at the laboratory in September 1943, and I was director of the Technical Division when I left the laboratory in 1948. For this reason, my reminiscences, as reflected here, are of the period 1943-1948. Since that period, the character and mission of the laboratory, as I have observed as a visitor and consultant on numerous occasions, have changed substantially. I leave it to others to describe the changes, but I cannot close this letter without suggesting that, even now, chemical engineering is one of the stronger disciplines at ORNL, although it may well be applied to problem types quite different from those of 1943-1948. A 1948 organization chart for the Technical Division, valid at about the time I left the laboratory, is presented in Appendix A.

1.4 PERSPECTIVES OF FORMER ORNL DIRECTORS AND ASSOCIATE DIRECTORS

1.4.1 Alvin Weinberg, Director, ORNL, 1954-1974[21]

On December 31, 1947, the ax fell on Clinton Laboratories. Jack Franklin, the AEC's manager of Oak Ridge Operations, wrote to Prescott Sandridge, Monsanto's Executive Director for Clinton Laboratories: 'The Atomic Energy Commission, after very careful consideration, has decided to relocate reactor development to the Argonne National Laboratory . . . activities concerned . . . the high flux reactor . . . and a reactor for useful power will become part of. . . Argonne.. . Clinton will undertake a strong program of research on chemical process problems and on the chemical and chemical engineering phases . . . of nuclear energy."

At the time, the High Flux Pile (as the MTR was then called) was Clinton's centerpiece. Wresting it from Clinton was a bitter pill for everybody at the Laboratory-that is, everybody except the chemical engineers. For them, designation of chemical process problems as a central mission of ORNL was welcomed as a great opportunity.

Chemical development was conducted as part of the Technical Division—first under Miles Leverett, then Merlin Peterson, and, after splitting off as the Chemical Technology Division, under Frank Steahly and then Floyd Culler. The laboratory, with its war-built concrete cells (Building 205). was uniquely able to test Redox on a pilot scale, in preparation for its use at Hanford. Redox was followed by Purex, Thorex, and other processes. The Chemical Technology Division's reputation as the'leading center for development of solvent extraction in radiochemical processing became widely recognized.

Although the MTR was built in Idaho, not in Oak Ridge, Chem Tech remained responsible for the chemical reprocessing plant at Idaho Falls. This plant was entirely an Oak Ridge development. Floyd Culler directed the project, and its completion gave the division confidence in its ability to carry out big projects.

Chem Tech participated in the laboratory's efforts to develop liquid fuel reactors. Though these efforts did not culminate in successful breeders or aircraft engines, they did yield important by-products. Most notable was the sol-gel process, an outgrowth of Chem Tech's attempts to make thorium slurries for use in aqueous homogeneous reactors.

As I look back from the vantage point of forty years, I realize that Chem Tech was one of the most successful of the ORNL's divisions. It combined an admirable practicality with sophisticated chemical insights and imaginative engineering. In my later years at ORNL, when I found myself at growing odds with AEC's Division of Reactor Development, the great confidence Chem Tech enjoyed in Washington helped ORNL weather the difficult transitions of the early 1970's. I am forever

Dr. Alvin Weinberg, 1983

grateful to Chem Tech for its accomplishments. They have been a source of satisfaction for the members of the division and a source of strength for ORNL.

14.2 Herman Postma, Director, ORNL, 1974-1989[22]

The principal mission of ORNL during my tenure as director was to help ensure national security in energy independence through R&D; 'to be a steward of large and important national physical and biological facilities: and to perform research about the environment that would allow energy development and conservation to proceed in the best possible way.

The principal missions of the Chemical Technology Division were, of course, important aspects of the ORNL mission. At the start of my tenure, Chem Tech was essentially the only place in the country that could do R&D in several areas of importance, such as reactor technology and fuel processing. ORNL and Chem Tech had the kinds of experienced people and unique facilities that could accomplish such R&D. In the beginning, Chem Tech performed research in chemical engineering technologies, particularly those pertinent to nuclear technology. It also operated very difficult, high-priced nuclear facilities for the purpose of producing rare isotopes, particularly transuranic elements. Later, Chem Tech started to convert from solely chemical engineering to include biological and biochemical engineering. It still retained an important mission in the supplying critical isotopes but had essentially gotten out of fuel reprocessing, at least the production and hardware aspects, except for some activities in the research end. The Chem Tech mission then began to change dramatically by becoming involved in alleviating waste problems, such as the replacement of waste, producing processes with benign processes and, also, waste minimization. Chem Tech used its chemical and engineering expertise in developing methods to deal with the already existing wastes. Such methods generally involved chemical or geological means.

Dr. Herman Postma, 1996

There was one other mission that nobody ever really stated but that truly existed. Chem Tech managed to develop some of the very best people that ORNL ever had in terms of what they did in science, engineering, and management. I don't think they thought of that as a mission, but in fact that was one of the division's biggest contributions (i.e., training and exporting these people throughout the laboratory).

In terms of actual accomplishments, in one sense, Chem Tech founded ORNL. Essentially, the first mission of the whole laboratory was in chemical processing. We built a reactor for the purpose of producing crucial radioisotopes. So Chem Tech's fundamental contribution, which is the basis for all the processes used worldwide, was to separate the isotopes. That was started here. So that's a fundamental contribution. We created Savannah River processes. We created, essentially, the techniques used at Hanford. And, of course, we produced a lot of isotopes for research.

More recent accomplishments include conversion of biomass into alcohol, biological process development, and separation techniques. The latter has always been one of the more fundamental areas in which Chem Tech prevailed. Separations in the beginning used ion-exchange columns, but as that technology evolved, even bacterial columns were used to perform some exchanges. I think, basically, Chem Tech's overall contributions were generically in separation sciences-separations first chemically, later biologically, and sometimes physically (e.g., isotopes). Just a steady flow of improvements came along as a result of that kind of research.

What are important directions for ORNL in the future? The laboratory as a whole must provide responsibile stewardship of national facilities in the physical and biological sciences. Other important ongoing missions include the following:

- Coal energy conversion, such as the conversion of coal into liquids, or conversion of biomass into gases or liquids for transportation
- Energy conservation
- Improved integrated utilization of nuclear materials, waste management, and separation management, including fission and fusion reactor development
- Technology transfer
- Educating future engineers and scientists through cooperative agreements with universities, colleges, and even high schools

But the subtle focus in all this is basically energy and the environmental questions that surround it.

Chem Tech will play a central role in the future missions of the laboratory, for example, the waste management effort. The size of the waste problems that the Department of Energy (DOE) has is in perhaps the $100 billion range. The leverage that research may give to managing waste problem areas faster and more efficiently can be significant. Because the basic mission of Chem Tech is chemical and biological processing, the division will have an important role in waste research, and waste management will be a central theme for some time. Chem Tech will also continue to play an important role in the conversion of fuels.

Because of the emphasis on waste and environmental areas, it may be difficult to maintain a balanced approach with other research areas. The problem is that the people in the waste areas are not currently putting very much R&D money into waste research. Research dollars are a vital necessity. Getting the money so the people can do the right kind of basic and applied research on the separation sciences is very important to the overall waste program.

Chem Tech has been involved extensively in Work-for-Others efforts. This is an important effort. Research dollars are going to benefit a lot more than just the agencies that supply them. Work-for-Other agencies (e.g., the U. S. Environmental Protection Agency and the Department of Defense), besides meeting their R&D needs, help cut the cost to the DOE because they utilize facilities that have essentially fixed costs. It provides spin-offs in other research areas. It provides a multitude of sources of money. Some of these sources of money may go up and others may go down, but the balance is pretty good. Whereas if you just have one source of income and the budget is slashed, then you're swinging in the wind like everybody else. Work-for-Others efforts make for good science, as well as having a good science rationale.

Chem Tech has been around longer than, I guess, almost any other division. As a division, it started around 1950, a spin-off from the Technical Division. Since that time it has retained its integrity, although many things have changed. Chemistry, fuel reprocessing, and isotopes have drifted in and out from time to time. But the core division may have been there longer than any other core division. Now, whether it's first, or second, or third, it has always been a very fundamental part of the laboratory. That is recognized purely by the fact that it has retained that status since the beginning. That means it is flexible. That means it does very good things. Otherwise, it would not have lasted as long as it has. In addition, it has been one of the safest divisions of the laboratory. As a division that deals with a lot of very sophisticated hazardous substances, it has set a standard of not having had a serious accident in 35 years. That is remarkable.

ORNL and the Chemical Technology Division were created in response to the great atomic energy effort, and they are continuing to play active roles in that area. I foresee that there will be an increase in atomic energy use in the future. The first thing that has to be done is that nuclear energy around the world, not just the United States, has to operate safely with minimum occurrences of any sort for a long enough period so that people become more comfortable with it. People sooner or later will recognize the overall environmental concerns and risks with all methods of energy production, and they will be able to make the t&de-offs that are necessary. So nuclear energy will look pretty good compared with some other alternatives once all the facts are in. I think it will all come back probably within a decade, but I don't think very much sooner than that. It was thought that fusion energy was only ten years away when I started in the fusion program 30 years ago. It is safe to say that fission will make a revival before fusion will come in as a complementary energy source.

1 A.3 Donald Trauger, Associate Director, ORNL, 1970-1986[23]

The Chemical Technology Division is an example of dynamic enterprise that extends to the very beginning of the ORNL. Although the division was not officially organized until 1950, its origins trace to the initial program for which the laboratory was founded during World War II. The project to develop the separation process for recovering plutonium from uranium and fission products established a major thrust for the laboratory. Thus chemical engineering, the main discipline of the division, has been and remains a major component of ORNL programs.

A most important feature of a national laboratory is the ability to anticipate and respond to national challenges. The Chemical Technology Division has exhibited such capability starting with the improvement of military nuclear fuel cycles extending through the cold-war period. These processes have proven adaptable to civilian nuclear programs, and ORNL's technologies are in use wherever nuclear fuel reprocessing is employed throughout the world. The division has continued to anticipate and respond to needs in many fields from biological technologies to improved use of fossil fuels. Although early nuclear fuel cycle work contributed to the laboratory's legacy of environmental problems, the division also was a national leader in developing improved practices and corrective measures.

Mr. Donald Trauger, 1990

The importance of nuclear'waste disposal was recognized early by Chemical Technology Division staff members, and much of the technology employed and that anticipated for high-level-waste disposal facilities had their beginnings here. The adaptability of the division is exemplified in its successful roles in many fields of waste technology. These include hazardous and mixed wastes, as well as high-and low-level radioactive materials. The breadth of the programs in this area was effectively articulated in the recent Showcase lecture on bioremediation for the treatment of organic-type wastes.

The excellence of Chemical Technology Division programs extending from bioengineering to operation of major hot cell complexes represents a major example of successful management of complex operations. This extends from the development of analytical systems for body fluids and studies of photosynthesis to the building and operation of large chemical separation columns.

Not the least of Chemical Technology Division accomplishments is the outstanding safety record of a great many years without a lost-time accident. This is a most impressive record in view of the nature of the hazardous materials used and the processes developed involving reactive chemicals, large amounts of radiation, and biological material. Many of these required complex engineering facilities. This accomplishment reflects the capability to adapt good practices from one technology to another. Although the aggressiveness of the division's operations has produced some near misses for serious accidents, the record is impressive and the safety discipline has continually improved.

The division also has maintained a dynamic enthusiasm and entrepreneurship. For example, when it was deemed necessary to move a section to bolster another division, and on another occasion to move a major function to create a new division, the Chemical Technology Division rallied and filled the voids with new work. The division exemplifies an appropriate level of aggressiveness in finding new and needed areas of R&D.

The years spent working closely with the Chemical Technology Division, especially when it reported to my associate director position, were pleasant and rewarding. They also were a challenge since my training and prior experience were light in chemical engineering. The division staff served as excellent teachers and consultants. Chemical Technology is a great division, poised for continued good work and predictably should be reflected well when a 100-year history report is written.

14.4 Frank Bruce, Associate Director, ORNL, 1970–1978”

I came to the Clinton Laboratories-later to become ORNL-in December 1943 with the Du Pont Company as an analytical chemist. In 1945 I transferred to the Technical Division that later became the Chemical Technology Division (Chem Tech). Chem Tech’s mission at that time-and for many years that followed-was chiefly the reprocessing of irradiated material, to separate uranium and plutonium from fission products and from each other. I left Chem Tech in 1959 to become Assistant Deputy Director of ORNL. Accordingly, most of my very fond recollections of Chem Tech cover the period 1950–1959, a time when Floyd Culler served as division director and I as associate division director.

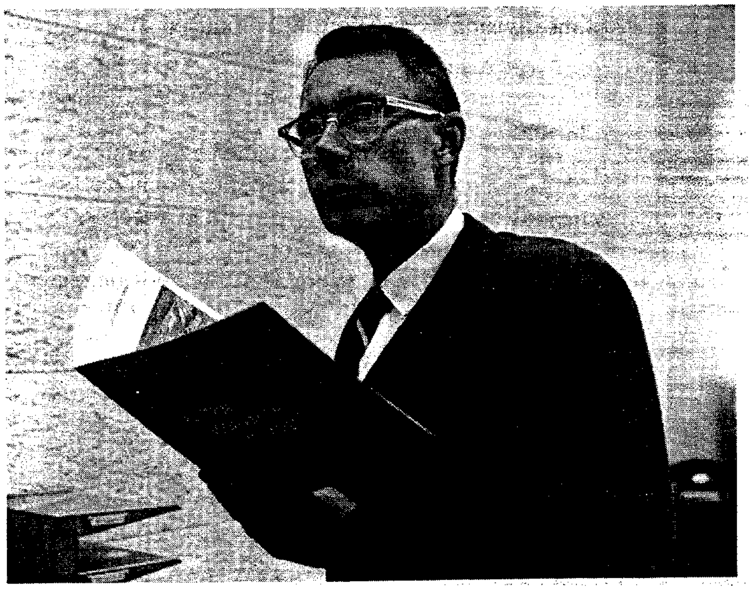
Looking back on those years, two things characterize Chem Tech in my mind. First, the division was highly integrated, conducting its own chemical research and development, unit operations work, &sign, and pilot plant operations. I believe that this integration resulted in better communications and cooperation and in better coordination of effort than I have observed more recently in projects that employ “contemporary” organizational styles. Second, Chem Tech had a “can do” attitude that just wouldn’t quit! And seldom did the division fail in accomplishing its mission. A few of these missions seem worthy of mention.

Chem Tech developed the process that was used for the recovery of enriched uranium from spent MTR fuel and other research reactor fuel as well. It designed the reprocessing plant that was built at Idaho Falls, supervised the plant construction, and managed its startup. This project was a ***tour de force*** of Floyd Culler.

Soon after that, Argonne National Laboratory (ANL) developed the Redox solvent extraction process that was to replace the bismuth phosphate precipitation process used at that time to recover plutonium from irradiated fuel at Hanford. There existed at ORNL the pilot plant that had been used originally to demonstrate the bismuth phosphate process before it was installed at Hanford. Chem Tech was asked to modify the pilot plant and to demonstrate the Redox process in it. During the demonstration, Chem Tech discovered that the effectiveness of the ANL flowsheet could be increased more than tenfold by modifying the chemical composition of the feed solution. This modified process was chosen by Hanford as the replacement for the bismuth phosphate process.

A few years later, Chem Tech developed a much improved process for uranium and plutonium purification using TBP as the solvent. This process, the Purex process, was soon installed in Hanford as a replacement for the Redox process. When the plutonium production plant at Savannah River was built, it too used the Purex process. Chem Tech subsequently applied modified Purex processes to other separations problems, including the recovery of ^{233}U from irradiated thorium (Thorex process); the recovery of uranium from Hanford bismuth phosphate wastes; and certain recovery problems at the Femald Feed Materials Production Center. More recently, when ORNL and Chem Tech became the center for transplutonium element production, the division developed new solvents and designed and built the TRU facility for the job.

No discussion of Chem Tech’s accomplishments would be complete



Mr. Frank Bruce, 1975

without mentioning the very important work that was done by Keith Brown and his. co-workers on processes for the recovery of uranium from domestic ores. At the time of peak production, practically all of the mills in the United States used these processes.

It is safe to say that Chem Tech has played the leading role in solving the nation's reprocessing problems. When Alvin Weinberg was ORNL's director, he used to say that one purpose of the laboratory was to undertake big projects of national importance that others could not handle. Chem Tech's achievements are testimony to that and have earned the division a lasting place in the history of the country's atomic energy program:,

1.5 CHEM TECH LEADERS, PROGRAMS, FACILITIES, AND ACCOMPLISHMENTS

Successful organizations seem to be governed by the maxim that the whole is always greater than the sum of its individual parts. It is also true that the most critically important elements in the success of any enterprise, whether research, education, commerce, industry, government, agency, or military, are the individual people staffing that organization. From its inception, Chem Tech seems to have been favored with capable staff members possessing a rich variety of experience, training, and education. The success of Chem Tech is in no small part attributable to the individual staff members and their collective esprit de corps.

Most of the current Chem Tech staff members (as of early 1992) are shown in Figs. 1.9 to 1.13.

Many of the technical accomplishments of Chem Tech during the past four decades have been achieved because of. the effective. use of collective -"groups or research teams. This fact is a tribute not only to the research teams. and individual members, but also to the technical knowledge and organizational skills of the Chem Tech leaders. The evolution of the organization and its leaders are embodied in Appendix C.l.

The numerous and diverse activities of a large and complex division such as Chem Tech are difficult to summarize. A litany gleaned from technical progress reports reveals some of that complexity (see Appendix C.2). Later in this history, selected programs and projects will be emphasized in greater detail. Many Chem Tech activities were of considerable national importance to DOE programs. The changing missions and evolving activities are summarized in the appendix.

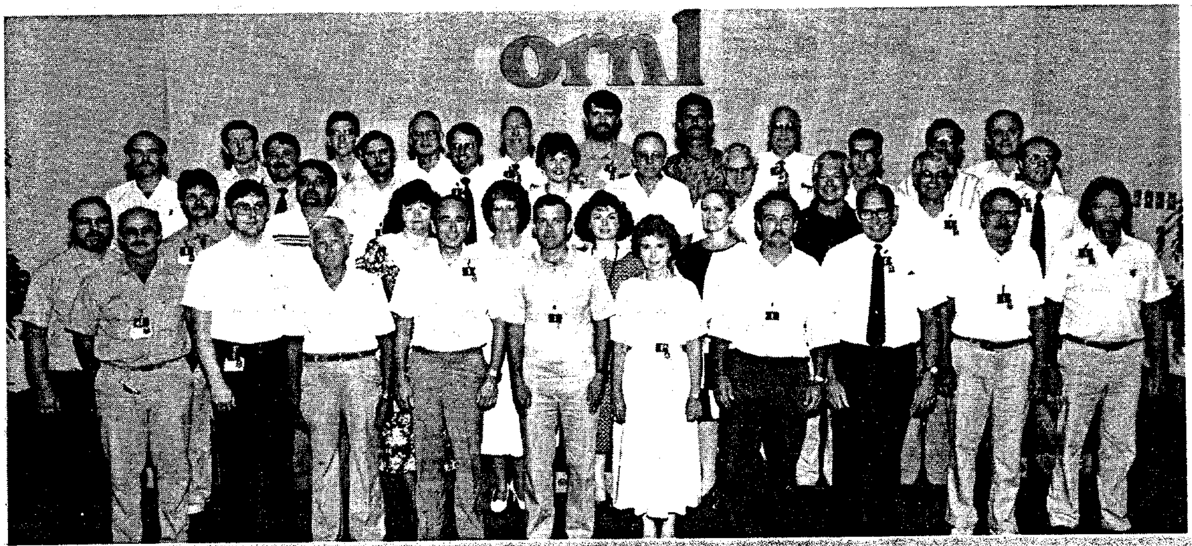


Fig. 1.9. Isotope Technology Section. First row (left to right): C. W. Evans, L. K. Felkner, D. H. Newman, D. B. Owsley, D. F. Williams, B. R. Phifer, T. T. McConnell, E. D. Collins, R. R. Laxson, and G. F. Galloway. Second row (left to right): A. E. Wayland, P. D. Bailey, M. E. Littleton, S. L. Lay, S. C. Owen, C. M. Smoot, L. J. Cotter, J. B. Knauer, J. E. Beaver, R. M. Wham. Third row (left to right): R. T. Barnett, G. L. Johnson, J. C. Glover, W. A. Brooke, J. A. Posey, J. T. Wiggins, C. E. Roberts, R. J. Vedder, S. E. Shell. Fourth row (left to right): R. G. Stacy, T. L. Turner, G. D. O'Kelley, R. G. Ross, G. D. Owen, J. W. McNeely, J. E. Bigelow, F. R. Chattin.

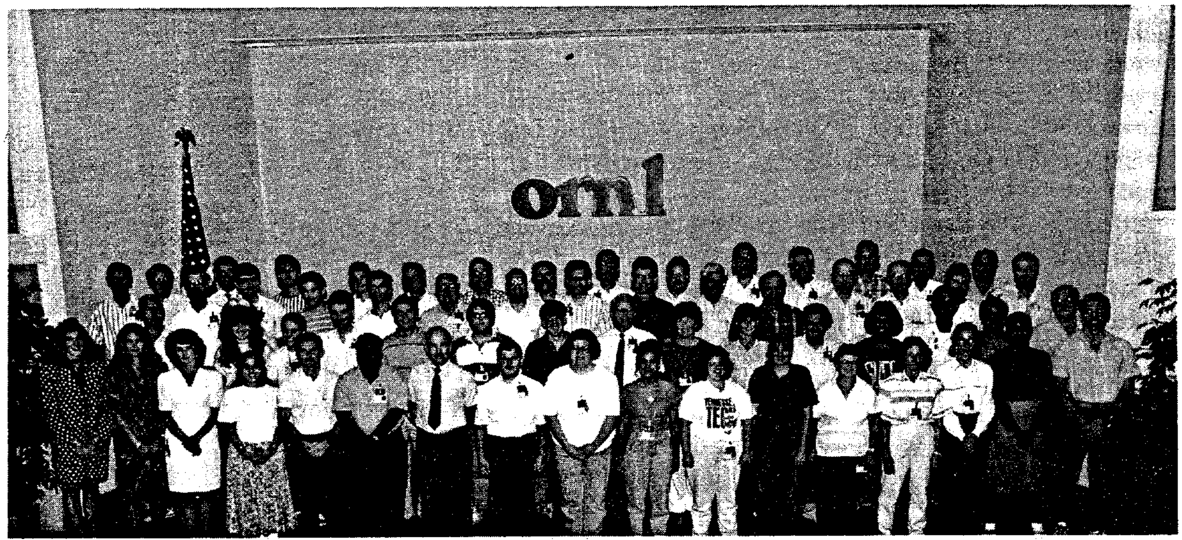


Fig. 1.10. Energy Research Section and Chemical Development Section. First row (left to tight): Gail **McNabb,** Joan Taylor, Brenda Light, unidentified (summer student), Osman Basaran, Mike Harris, Paul **Haas,** Ed Wilkes, Teresa Takacs, Omaria **Melendez** (summer student), Robin Bright (**TTU** student), Debbie Davidson, **Mitcha Petek, Charlene** Woodward, Jean MacInnis, Brenda Faison, Stan Cooper. Second row (left to right): Randy Gibson, unidentified summer student, Lourdes Hemandez (summer student), Rodney Hunt, David **DePaoli,** Bill Chase, Brian Dodson (summer student), Jimmy Bell, Giner **Tevault,** Donna Young (summer student), Charlie Webster, Rachelle Hess (summer student), Kwang-Fu Tsai (visitor from Taiwan), Chung-Shin Lee (visitor from Taiwan), Tom Dillow. Third row (left to tight): Jim Thompson, Doug Lee, Brian Davison, Diego Lopez (summer student), David Londono, Richard Shoun, Eli Greenbaum, Johnathan Woodward, John Cosgrove, Hank Cochran, **Charles** Byers, Jack Mrochek, Jack Collins, Chris Lockwood, Allen Boatman. Fourth row (left to right): Terry Lindemer, Dave Pruett, unidentified summerstudent, unidentified summer student, George Parker, Bill Pattison, Steve Blankinship, Mac Toth, Tim Scott, Warren Sisson, Morris Osborne, Jim Travis, Jim **Mailen,** Ron Brunson.

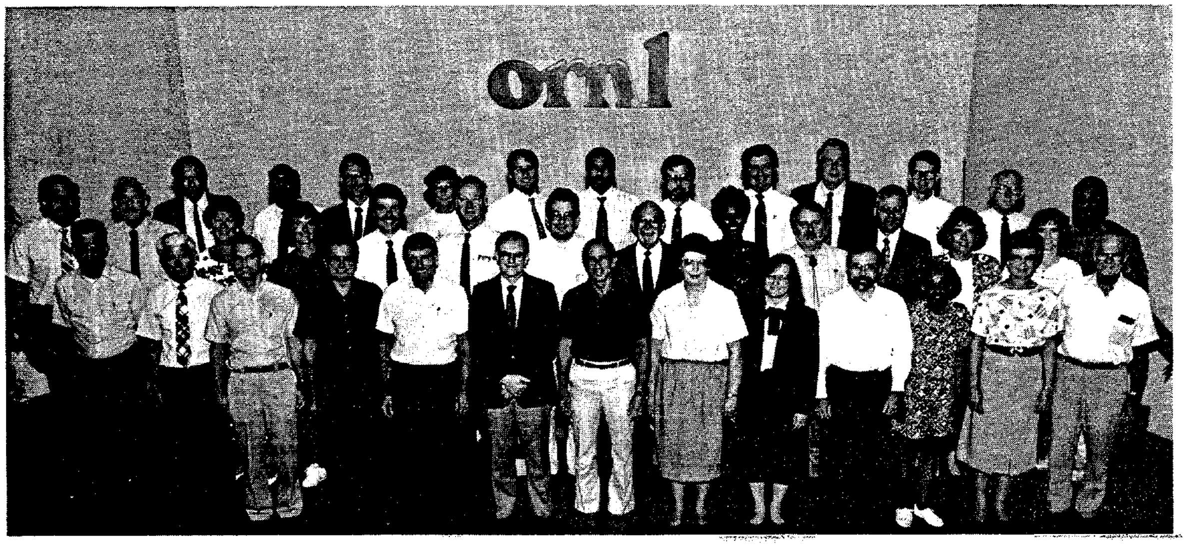


Fig. 1.11. Engineering Coordination and' Analysis Section; First row (left to right): Ralph **Andermann,** Brad Welles, Joel Shor, Don Box, Bill Rodgers, Max **Wankerl,** Mike Morris, Martha Dawson, **Vicki** Green, Tim Welch, Charlene Patrick, Veneeta King, Bob **Jolley** (member of the Waste Management Technology Support Group). Second row (left to right): Bill Reich, Joe Armento, Mimi Welch, Donna Brooksbank, John Begovich, Larry **Shappert,** Juan Ferrada, Karl Notz, Vanessa Brown, Ron Pope, Francis Kovac, Brenda Morrow, Robin Oody, Irvin Osborne-Lee. Third row (l'eft to right): Royes Salmon, **Suman** Singh, Rick **Rawl,** Sue McDaniel, Scott Ludwig, Leonard Dickerson, Jerry Klein, Phil McGinnis (member of the Office of Technology Transfer and Work for Others Oversight), Charles Forsberg, Steve **Storch,** Earl McDaniel (member of the Engineering Development Section).

Fig. 1.12. Resource Systems Management Section. First row (left to right): Sam Clinton (member of the Office of Safety and Operational Readiness), Allen Doucet, Allen Croff, Anita Sims, Brenda Breeden, Stan Kimmett, Donna Reichle, Debbie Stevens, Rita Camp, Tom Bayles, Norman Lee. Second row (left to right): Donna Ault, Kaye Johnson, K. H. Lin (member of the Radiochemical Processing Program), Jackie Jernigan, Chris Flannery (member of the Radiochemical Processing Program), Jerry King, Kathryn King-Jones, Danny Cochran. Third row (left to right): Ed Kosinskt (summer student), Richard Genung, Alice McWilliams, Bob Hightower, Brian Copeland (summer student), Dave Holladay, Jack Maguire, Vic Pardue, Ed Benson.

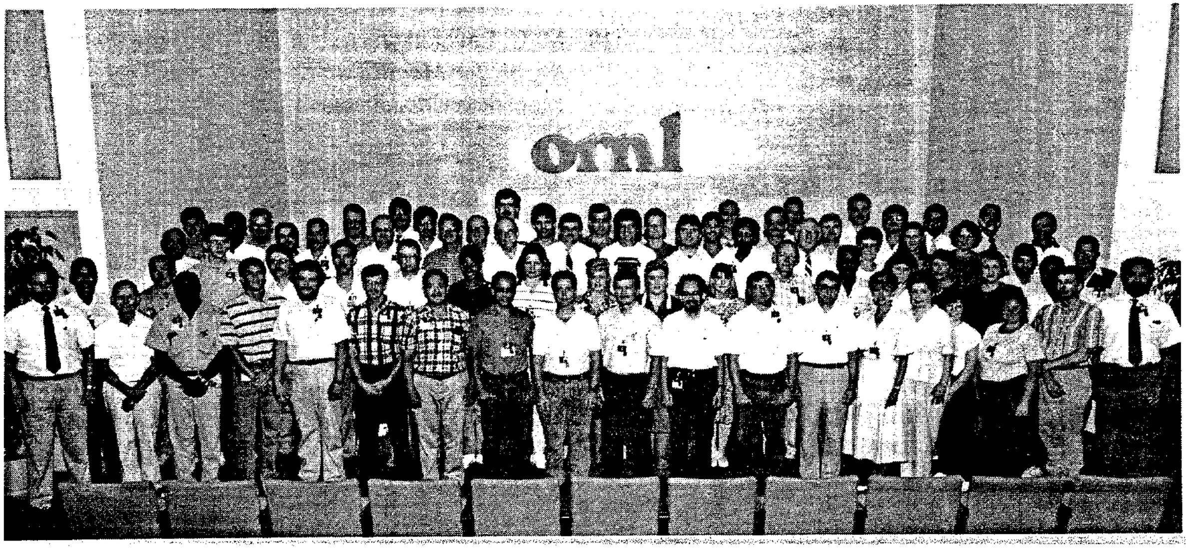


Fig. 1.13. Engineering Development Section and The Radiochemical Technology Section. First row (left to right): R. E. Rosenbaum, J. W. Snider, H. X. Phillips, S. L. Loghry, I. L. Morgan, T. D. Hylton, G. W. Strandberg, J. D. Hewitt, H. L. Jennings, J. H. Wilson, T. L. Donaldson, G. R. Herald, J. J. Perona, L. J. Fields, I. Beaty, S. M. Robinson, L. L. Farr, B. W. Stames, A. M. Krichinsky. Second row (left to right): R. Hall, A. W. Hensley, D. E. Newton, R. L. Cummins, unidentified staff member, co-op student, B. S. Evans, D. S. Cooper, K. D. Snyder, T. C. Loftis, F. G. Kitts, I. A. Conway, A. B. Walker, M. A. Sedlmeier, C. I. Radcliffe, P. A. Taylor, S. M. Fuqua, C. W. Leinart. Third row (left to right): B. L. Byrum, M. Johnson, R. G. Grubb, C. L. Francis, S. A. Richardson, C. H. Brown, Jr., E. L. Youngblood, B. D. Patton, L. R. Conner, J. W. Shaw, Jr., J. Woodward, E. W. McDaniel, J. M. Lynch, J. M. Butler, N. D. Johnson, B. Z. Egan. Fourth row (left to right): I. G. Gross, D. Foster, Jr., unidentified staff member, S. C. Osborne, R. E. Eversole, unidentified staff member, R. C. Lovelace, M. E. Reeves, A. J. Luccero, L. A. Zevenbergen, unidentified staff member, T. D. Clure, unidentified staff member, J. R. Parrott, J. R. Gibson, R. D. Spence, M. W. Burgess, T. M. Gilliam, R. K. Kibbe, L. G. Hill.

A large organization with varied missions requires many different types of facilities. Some of the major facilities that Chem Tech has been involved with are listed in Appendix C.3.

Chem Tech researchers have received many honors and awards and have produced numerous patents. Honors and awards are summarized in Appendix D, and patents are listed in Appendix E.

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2. THE FORMATIVE YEARS: NUCLEAR FUEL REPROCESSING

> Chem Tech possesses several attributes that contributed in a large part to its success as an ORNL division. First is that we started out small but we learned like crazy. . . . Second and probably the most important is the spirit of Chem Tech. I have never thought of a better word than "gung-ho." . . . Third is that we teamed with other divisions as routinely as we worked with our own. . . . Everybody felt an urgency and somehow we managed to keep it alive. . . . The reason is we kept getting big problems-one after another.
>
> *Floyd L. Culler*
> *June 18, 1992*

2.1 EARLY HISTORY

Even when first established in 1950, the Chem Tech Division and staff were located in several buildings because of the size and diversity of activities, namely, the director's office and the Laboratory Section, Building 3550 (formerly 706-A); the Design Section, Building 2067 (formerly 703-A Annex); the Unit Operations Section, Buildings 3502 and 3503 (formerly *706-I-D* and 706-HB); and the Pilot Plant Section, Building 3019 (formerly 205).[1] Figure 2.1 provides an aerial view of the X-10 site taken about 1944 showing Buildings 2067, 3503, 3019, and 3550. In 1951, Building 3505 (newly 'constructed Metal Recovery Plant) was occupied and Building 3508 for work with high alpha activities was occupied in 1952.[2] Buildings 3505 and 3508 are shown in Fig. 2.2, a photograph of the X-10 site taken about 1960.

Essentially all the activities of the Chemical Technology Department of the Technical Division were transferred to the newly created Chemical Technology Division. The major activities assigned to the new division dealt with the design of the Idaho Chemical Processing Plant (ICPP); the development of the metal recovery TBP process for the recovery of uranium from ORNL and Hanford" metal waste: the development of the Purex process for the recovery of uranium and plutonium from Hanford irradiated metal: the development (with Isotopes Division) of the RaLa process for separation of barium and lanthanum from fuel units; homogeneous reactor fuel studies; and the design/construction of the ORNL Metal Recovery Plant (Building 3505).[1,2]

Chem Tech work in the early 1950s was focused almost entirely on fuel reprocessing and the design and construction of particulate cleanup facilities for the graphite reactor. In addition to studying the chemical problems associated with the Purex process and the RaLa process, the Laboratory Section was concerned with the solvent extraction separation of fission products, Volatility process, TBP process, and the recovery of thorium by the Thorex process from 23 Process waste. The Process Design Section conducted studies on liquid and gaseous waste disposal for ORNL and, also, design work for the ICPP, the ORNL Metal Recovery Plant; and a RaLa plant modification. The ICPP was designed to separate ^{235}U from spent fuel from the MTR, the Enriched Breeder Reactor, and other reactors proposed to use ^{235}U for fuel. The Unit Operations Section started chemical engineering studies of radiochemical processes involving evaporation, solvent extraction, and ion exchange and continued unit operation-scale studies of the TBP Process and the RaLa Process. The Pilot Plant Section completed programs on the 23 Process, recovery of plutonium from the IBP waste resulting from the Redox pilot plant study, and recovery of plutonium from Chalk River irradiated metal.[1,3,4]

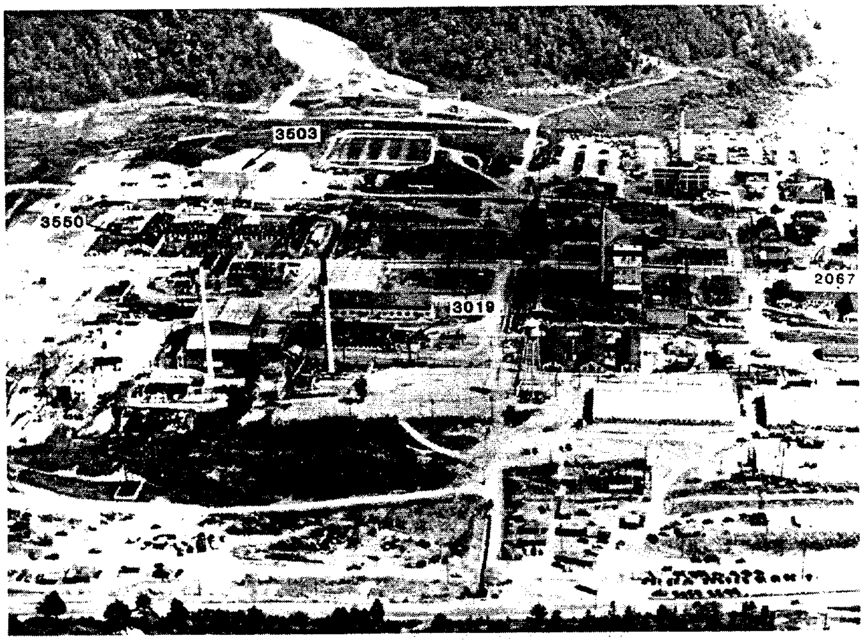

Fig. 2.1. An aerial view taken during the fall of 1944 of the Clinton Engineer *Works,* X-10 site, looking southward. Several buildings housing Chem Tech predecessor groups are identified: Laboratory, Building 3550 (formerly 706-A); Design Section, Building 2067 (formerly 703-A Annex); Unit Operations Section, Buildings 3502 and 3503 (formerly **706-HD** and **706-HB);** and the Pilot Plant Section, Building 3019 (formerly 205). Building 3502 (formerly **706-HD)** had not yet been constructed but is located **immediately** east (to the left) of 3503.

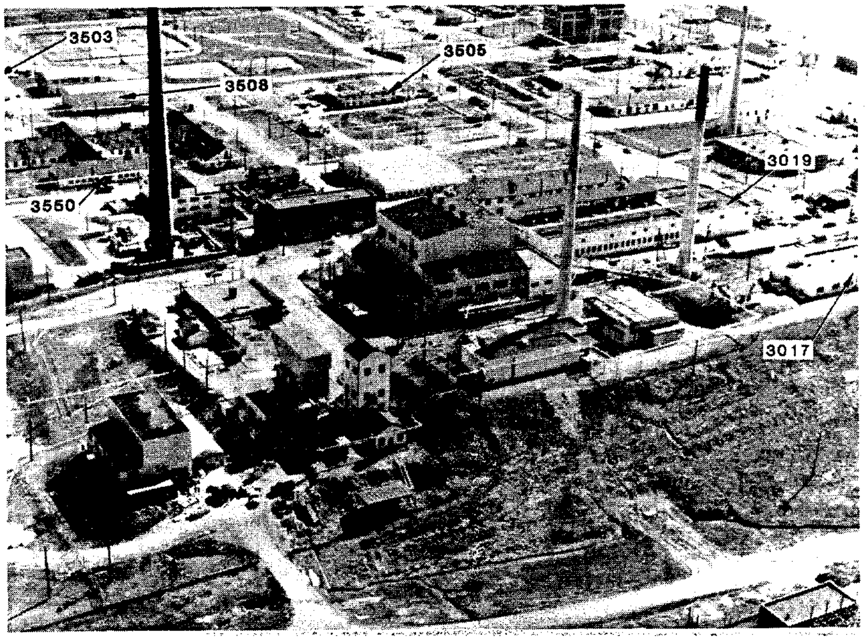

Fig. 2.2. An aerial view **taken about** 1960 of the Oak Ridge National. Laboratory, X-10 site, looking southwesterly. The photograph shows Building 3505 (Metal Recovery Plant) and Building 3508 (High Alpha Laboratory). Also identified are **3503, 3550, 3019,** and 3017 (later to be enlarged and occupied by Chem Tech personnel).

2.2 OVERVIEW OF NUCLEAR REACTORS

Even though the major objective of the wartime nuclear reactor program was production of the weapons materials, the potential use of nuclear reactors for production of thermal and electric power did not escape the attention of farsighted engineers and scientists. With the cessation of wartime activities, nuclear R&D evolved rapidly toward the goal of nuclear electric power. Production reactors generally used aluminum-clad natural or low-enriched uranium fuels coupled with carbon or graphite to moderate the energy of the neutrons produced during fission of uranium, plutonium, and other tissile fuel materials. However, many different electric power reactor concepts were soon proposed, including solid fuel element, molten fuel, and fluidized-bed systems.

The worldwide preponderance of power reactors appears to employ solid fuels and may be divided into four classes: (1) pressurized-water reactors (PWR) and boiling-water reactors (BWR) that use light water (H_2O) as both coolant and moderator: (2) heavy-water (D_2O)-moderated, pressure tube reactors that may use light water, heavy water, or other materials as coolants; (3) gas-cooled graphite-moderated reactors: and (4) fast reactors cooled by sodium or by helium?

Light-Water Reactors. BWRs and PWRs use UO_2 pellets enriched to 2 to 4%. The pellets or pins (1 cm in diam.) are clad in zircaloy and arranged in a square lattice with up to 200 pins per fuel assembly. PWRs and BWRs are depressurized every 12 to 18 months for partial refueling. BWRs operate at about one-half the pressure required in PWRs. Reactivity is controlled by burnable poisons (thermal neutron absorbers) and control rods?

Heavy-Water Reactors. Heavy-water reactors use natural uranium and, consequently, must be continually refueled. Fuel assemblies containing UO_2 pins clad with zirconium are arranged in series within pressure tubes. Reactivity is controlled by moderator levels and control rods.[5]

Gus-Cooled Reactors. Natural uranium can also. be used in graphite-moderated gas-cooled reactors, such as the carbon dioxide-cooled Magnox reactor developed by France and Great Britain. The Magnox reactor was named after the magnesium-based cladding used on the uranium-metal fuel elements. Gas-cooled reactors must be refueled on a continuous basis. The high-temperature gas-cooled reactor uses helium coolant with slightly enriched uranium carbide/thorium carbide fuel in a ceramic core.[5]

Fast Breeder Reactors. Fast breeder reactors use a fast neutron spectrum system fueled with plutonium and/or uranium-235, with uranium-238 as the fertile material. The fuel elements are uranium-plutonium oxide pellets enriched 15 to 20% in stainless steel cladding measuring about 0.25 in. in diameter. The reactor core is surrounded by a blanket of uranium oxide pins. Both sodium and helium have been proposed as coolants.[5]

Nuclear Power Reactors and Supporting Industry. Nuclear power reactors requite a large supporting fuel cycle industry. For example, the principal components of the complete light-water fuel cycle are the following:[6]

- Mining and milling to obtain a uranium oxide concentrate
- Refining the ore and converting it to UF_6
- Uranium isotopic enrichment
- Fabricating UO_2 fuel elements
- Reprocessing the irradiated fuel to recover fissionable materials (uranium and plutonium) for recycle
- Long-term waste management

Chem Tech was historically involved in all aspects of the nuclear fuel cycle. However, in April 1977, U.S. President Carter implemented a new nuclear policy that deferred indefinitely the commercial reprocessing of nuclear fuels and recycle of plutonium in light-water reactors and also delayed a decision on the use of plutonium in the liquid-metal fast breeder reactor (LMFBR) then under development.[6] One effect of this policy was a decrease in national emphasis on R&D in nuclear fuel reprocessing.

2.3 BRIEF HISTORY OF NUCLEAR FUEL REPROCESSING

Because the history of nuclear fuel reprocessing and Chem Tech are so intertwined, this section presents an overview of fuel reprocessing prepared by Chem Tech staff members J. R. Hightower and R. E. Brooksbank, Sr.[7]

The first large-scale nuclear reactors were built during World War II. These reactors were designed for the production of plutonium for use in nuclear weapons. The only chemical reprocessing required, therefore, was the extraction of the plutonium, free from fission product contamination, from the spent natural uranium fuel. In 1943, several methods were proposed for separating the relatively small quantity of plutonium from the uranium and fission products. The first method selected, a precipitation process called the bismuth phosphate process, was used at ORNL during the period 1943 to 1945 to produce quantities of plutonium for evaluation and use in weapons programs.

The bismuth phosphate process was first operated on a large scale at Hanford, Washington, in the latter part of 1944. It was successful for plutonium separation in the emergency situation existing then, but it had a significant weakness, namely, the inability to recover uranium. However, the first kilogram quantities of plutonium were produced in the half-ton-per-day pilot plant in Building 205 (now Building 3019) at ORNL.

Even before the bismuth phosphate process was chosen as the basis for the design of the Hanford plutonium separations plant, research on other methods for treating spent fuel-namely, volatility, adsorption, and solvent extraction-had been initiated. Significant advances in chemical reprocessing methods were made during the immediate postwar period, particularly with methods using solvent extraction. The basic principle upon which solvent extraction is based is that the nitrates of uranium and plutonium in the higher oxidation states are readily soluble in certain organic liquids which are immiscible with water. The nitrates of fission products are, in general, essentially insoluble in these liquids.

The first successful solvent extraction process for the recovery of both uranium and plutonium in decontaminated form was developed at Argonne National Laboratory (ANL) soon after World. War II. Methyl isobutyl ketone (hexone) was used as the organic solvent, and aluminum nitrate was added to the aqueous phase to improve the separation. Pilot plant testing of this process, the Redox process, was carried out with available equipment at ORNL (Building 3019) from 1945 to 1951, and large-scale operation began at Hanford in October 1952. The Redox process offered advantages over the bismuth phosphate process of (1) continuous operation; (2) a large decrease in waste volume: and (3) the ability to recover uranium as well as plutonium.

From 1948 to 1950, while the Redox process was under development, laboratory studies were being made on an improved solvent extraction process. This new method was called the Purex process and employed TBP as the organic solvent and nitric acid rather than aluminum nitrate in the aqueous phase. The Purex process was developed by ORNL and Knolls Atomic Power Laboratory (KAPL) and was carried through the pilot-plant stage at ORNL (Building 3019) from 1949 to 1960. This process offered four significant advantages over the Redox process:

1. a reduction in waste uranium,
2. greater process flexibility,
3. decreased solvent fire hazard, and
4. a decrease in operating costs.

This new process was put into operation at the Savannah River site at Aiken, South Carolina, in November 1954 and at Hanford in January 1956. All foreign reprocessing plants use the TBP process.

Since 1944, reprocessing has been practiced under the auspices of the government at one or more of the defense installations at the Savannah River Site near Aiken, South Carolina: at Hanford, Washington; and at Idaho Falls, Idaho. However, the growth of nuclear power generation in the 1960-1970 period prompted the government to encourage the entry of commercial firms into the reprocessing sector of the fuel cycle to recover unburned uranium and plutonium from fuel assemblies discharged from commercial power reactors. Accordingly, the first commercial reprocessing plant was constructed, and a provisional operational license was granted in 1966 to Nuclear Fuel Services (NFS) for a plant in West Valley, New York. During the period, other fiis became active in pursuing commercial reprocessing of irradiated fuel from the nation's reactors. These firms include General Electric Company (GE), Allied Chemical Corporation, National Lead Company, Atlantic Richfield Company, the Gulf Oil Corporation, and Exxon Corporation. Based on a series of studies, GE elected to build the Midwest Fuel Recovery Plant (MFRP), a l-metric ton of heavy metal (MTHM)/d plant in Morris, Illinois, employing the Aquafluor process, which differs considerably from the standard Purex process employed in other plants. A third plant of appreciably larger size (5 MTHM/d) was then constructed by Allied General Nuclear Services (AGNS) in Barnwell, South Carolina.

The NFS plant successfully operated for a period of 6 years, during which time a total of 641 tons of irradiated fuel was processed. In order to become more competitive, the plant was shut down in 1972 to increase its capacity from 1 to

5 MTHM/d. As a result of a series of new and retroactive regulations placed on the reprocessing sector by regulatory groups, mainly in the seismic area, the owners of the plant concluded that the cost of compliance with the new regulations for an expanded plant could not be justified and decided not to reopen the plant. Under terms of its operating permit from the state of New York, plant ownership reverted to the state.

In the case of the MFRP, the company decided not to proceed with the operation of the as-built plant following a series of operational difficulties experienced after a lengthy testing period. The difficulties were associated with the operation of a new process employing complicated equipment operating in a remote mode behind heavily shielded walls. No radioactive materials were involved in checkout testing.

The third commercial plant, built during the 1971-1975 period and owned by AGNS, has been completed but has yet to operate. Efforts to license and operate this plant were terminated by a commercial reprocessing moratorium in response to proliferation concerns expressed by President Carter.

Exxon was designing a modem reprocessing plant that was scheduled to be built in Oak Ridge, Tennessee; however, because of the moratorium, it was also canceled.

In spite of the dormant conditions of commercial reprocessing in the United States, separations technology has continued to evolve throughout the world and fuel reprocessing activity has advanced in several nations. The processes used are derived from development and pilot plants demonstrated in the Chemical Technology Division. Plants currently exist in the United Kingdom, France, Japan, India, Belgium, West Germany, China, and the Soviet Union. In-addition to the processing of spent fuels from light-water reactors, development of the technology necessary for the reprocessing of fast reactor fuels is advancing at a rapid pace in these nations. The United States has also been active in the development of this technology by way of research carried out in universities and government-owned laboratories.

2.4 THE CHEMICAL TECHNOLOGY DIVISION AT ORNL FROM 1943 TO 1961: Recollections of Warren Eister, September 1991[8]

Division Name. On the way to the cafeteria, I suggested to Frank Steahly we name our new organization the Chemical Technology Division. There were several reasons: Our chief competitor at Argonne had already adopted the name of Chemical Engineering; second, we had a strong group of chemists as well as chemical engineers; but foremost, Frank had just pulled together three

Chem Tech staff members Warren Eister (left) and Joe Matherne (rlght) with a Chemical Technology Division information Meeting guest (about 1960).

individualistic groups that needed a clear statement of who they were going to be. I do not recall the year, but it probably was when Monsanto took over from DuPont. My following recollections will lack similar chronologic and factual precision.

Organization. The nucleus of Chem Tech was the chemistry lab and semi-works and to these parts were added the pilot plant and process design groups. These all had been part of the Technical Division that had responsibilities for both process and reactor development under Miles Leverett. To split the reactor and process responsibilities, Frank Steahly was brought in from the Chemistry Division to provide a Ph.D. in the new organization. While Steahly was our first director, I don't know who originated the action.

At the beginning, Frank Bruce had Chemistry and I directed the Semi-works, with both of us reporting to Steahly. Then Don Reid joined the organization with responsibility for the Pilot Plant and Floyd Culler for Design. Most of us had remained behind when DuPont moved to Hanford, but Floyd hired in from Y-12. Then Jim Davis joined us from the Synthetic Rubber Program and took over the Semi-works and I became Steahly's assistant. By this time I had changed the name of the Semi-works to Unit Operations, but it might have more properly been named Unit Processes.

2.4.1 At the Beginning

The Start. Going back to creation, the Technical Division, the parent of Chem Tech, had been established by DuPont at the Clinton Laboratories, X-10 for the engineering development of

- safety procedures for nuclear operations;
- graphite nuclear reactor for production of plutonium: and
- bismuth phosphate process for the recovery of the plutonium from irradiated reactor fuel,

My First Encounter. I came to X-10 from the Indiana Ordnance Works where DuPont manufactured smokeless powder. At that location, in 1942, success meant moving back to Wilmington, Delaware. In 1943 folks were going to Chicago, and in May of that year Ted Arehart and I were the first from Du Pont to be sent to Knoxville. Our instructions were "Stand on the street comer in front of the Farragut Hotel at 7:00 a.m. on Monday and someone will pick you up. You'll contribute more than in the army." I was there on time, and at midnight that same day I trudged through rain and mud to an open-sided building to look for pimples on aluminum-jacketed slugs. I went to The University of Tennessee library on my first day off with the suspicion that uranium was in those slugs. I found a lot of information on uranium including ^{235}U separation in cyclotrons and driving a car around the world using a pellet as the fuel. The day after, I exchanged this knowledge with a few fellow inspectors. The next day, in a room with a drawn blind, three straight-backed chairs, and an old, bare, battered oak desk, I huddled with Julian Ellett and Jack Frame. There, in a few minutes, they introduced me to plutonium and the bomb. The slugs were the fuel to go into the graphite reactor, the pile. This revolutionary revelation frightened me! It was the easiest secret I ever kept.

Bismuth Phosphate (BiP) Process and the Semi-Works. In August, I went to the semi-works to work for Don Johnson. There were about five of us on each of four shifts. With one craftsman from maintenance, Glen Ricker, we put together a uranium dissolver followed by six cycles of precipitators and centrifuges. When I arrived, there was a two-story room (about 60 ft by 60 ft) with a balcony and a row of four concrete hot cells on the west wall. The room was crowded with a bunch of stainless steel tanks, iron drums, pipe, plastic tubing, centrifuges, valves, and other equipment.

Our group came mostly from the smokeless powder plants. There were also military servicemen who included Tex Blomeke, Nelmar Rigstad, Ed Johnson, George Havorka, and Joe Savolainen. They were chemists or engineers who had been drafted and then sent to help. Of the civilians, Irwin Higgins stands out in my memory as our welder, although he was actually a biochemist raised on a potato farm in Maine. Marshall Aken, our chief, and Joe Work, the data analyst, had been members of Du Pont's group of six that evaluated the governments's request to participate in the Manhattan Project. Crawford Greenwalt, later Du Pont's CEO, was the group leader. In addition, Luther Perry, Dick Apple, and Vance Cooper carried out special assignments for Aken. When DuPont returned for the Savannah River Project, Lu Perry was the Du Pont on-site coordinator at Chem Tech. Joe Work and I were associates in the DOE high-level-waste disposal when he retired. For Joe's retirement party, I found Marshall Aken in a retirement village in Chevy

Chase; Marshall was to be our mystery guest. While neither of us had seen Marshall for 40 years, Joe recognized him immediately when Marshall standing behind him said, "Hello, Joe."

It seems that by Christmas, the system was operating with natural uranium and simulated plutonium and fission products. The hot cells were closed with concrete blocks except for No. 2, which had a lead door that I don't recall ever using. In addition, there were three concrete block cells in the middle of the room. Then the "hot" tests started using irradiated uranium slugs from the reactor. Using a 10-ft cherry picker, we hustled the slugs, one at a time, from a lead-shielded container on a flat bed truck and dropped them into a funnel to the dissolver. We all wanted our turn driving the truck up to the reactor. Until that time, only Maintenance had trucks.

The fission product decontamination factors for the semi-works plutonium separations were terrible, and I remember John Swartout watching me on a midnight shift as I was enthusiastically braking the centrifuge. He suspected, correctly, that I was "throwing the baby out with the dishwater.*' The final demonstration employeda significant portion of the country's supply of plutonium split four ways. (I still wonder where that plutonium came from!) Everyone was there to watch the start of the operation. The plant manager, in a small midday ceremony, added the first batch to the first precipitator. His representative was there for the second batch, and our group leader for the third batch on the night shift. I don't remember the fourth batch since the third was, for me, a personal disaster. There were now three people on our midnight shift plus two recent trainees for the pilot plant. The shift leader was off at the time and his number 2 man was in his office almost incapacitated because of a severe foot problem. Most of the cells were operating, and I was directing the activities of the trainees while running a couple myself. A trainee centrifuging Run No. 3 in Cell No. 4 called over to me that he was not getting a reading on the catch tank manometer. Many of the manometers had air leaks necessitating higher airflow, so I told him to increase the airflow. After several increases-in airflow and more than half of the feed discharged, the awful truth was learned—Cell No. 4 was the only location discharging to the waste tank. The next day, Run No. 3 was scrapped. While I had dumped 25% of the world's supply of plutonium into the waste tank, it may have upgraded our controls.

Following this BiP work, DuPont departed and planned to send me back to Indiana Ordnance for rocket powder manufacture. Pete Peterson convinced me to stay on and we started work on the thorium cycle, RaLa, and Redox processing. It was during this time that Don Ferguson came to work with me in the semi-works plant from Tennessee Tech. Having to work shifts and with the group leader having the only office made it a very undesirable place to work. It was known as the "salt works." Don soon wondered to me whether he shouldn't just move on. I suggested he stick it out a bit longer. In time he moved on to Frank Bruce's chemistry group where they had more desks and day work.

Redox Process and Chicago Advenhues. A major development! Seaborg's group at Chicago had come up with a solvent extraction process that separated uranium as well as plutonium from the irradiated uranium. X-10 was again going to do the semi-works and pilot plant development. Here was a chance for some chemical engineering calculations of theoretical stages in a countercurrent solvent extraction column. BiP had none of this sophistication. Also, there was now an excuse to see the West Stands, University of Chicago, where the Chicago work was being done.

On my first trip to the West Stands, I got the chance to run a model of the Fermi Pile. Using a servo, the control rod was raised and I could follow the neutron flux on a mirror-type potentiometer. However, no one at Chicago who knew anything about theoretical stage calculations in the solvent extraction process. Steve Lawroski, a Ph.D. chemical engineer from Penn State, had departed for the reactor school starting up at X- 10. It was a relief to find Chicago as uninformed as we were. I went out with the Chicago solvent extraction group for a terrific Chinese lunch and we became "birds of a feather."

Acid-Deficient Redox Process and Three-Dimensional Designs. Nelmar Rigstad, at a drafting table in the operating area of the semi-works plant, created a three-dimensional design of the first Redox cycle for Cell No. 2. I had recently seen something on three-dimensional design and had bought the appropriate triangles. It took Riggie about two weeks to design and Glen Ricker and a couple of helpers about three months to install. It came in over budget, costing $20,000 instead of my estimate of $10,000.

In the meantime, Frank Bruce's group had investigated the effect of pH on the decontamination factor (DF). By partially neutralizing the aluminum nitrate salting agent, the DF was increased from about 100 to about 1000. We had invented the acid-deficient Redox process. It made us feel much better about our research. We soon developed modifications for the recovery of enriched uranium from test reactor fuel (the 25 process) and uranium-233 from irradiated thorium (the 23 process). When I started the design of the semi-works unit for the 23 process, I assigned the job to Bob Klotzbach and Bob Milford. They revolted when asked to use the three-dimensional approach and insisted on taking the job to the plant design group. They did a good job. The unit worked well, but the job took more than a year, and this installation alone cost over $100,000. They never returned to the Semi-works group.

Hexone vs Tributyl Phosphate and Purex Revisited. The first society-type technology exchange meeting was held at the Chicago Museum of Science and Industry. Attendance was restricted since classified data was to be reported. At the meeting I met Glen Seaborg and, more importantly, Ray Fisher from the Ames Laboratory at Iowa State University. After trying to impress Ray with our contributions to the Redox process, which was replacing BiP at Hanford, Fisher asked why we hadn't used tributyl phosphate (TBP) for the solvent instead of the hexone, used in Redox since TBP was stable in nitric acid. We had to use aluminum nitrate with hexone, and this loaded up the waste tanks.

Our Chem Tech forces were devoted to Redox/hexone systems, and it was several years before TBP was seriously considered both for the recovery of uranium from the BiP waste and for application at Savannah River for plutonium recovery. It seems to me that uranium recovery came first. Charles Runion, Charles Ellison, and Bill Lanham were recognized on the patent. It was exciting to have DuPont select the Purex process for Savannah River. Manson Benedict headed the selection team. He had gained recognition in the development of the gaseous diffusion process for ^{235}U separation, and I looked forward to meeting him when his team came to X-10 to review our Purex studies. I almost missed him. He was the quiet fellow taking copious notes and occasionally asking questions in this impressive group. I never did actually meet him!

Mixer-Settlers vs Pulsed Columns. It was a severe personal disappointment when they selected mixer-settlers instead of the pulsed columns used in our Chem Tech pilot plant for the solvent extraction operation. Pots Chambers was the technical leader of the DuPont team and visited monthly to review the status of our Chem Tech activities. Afterwards, we usually went to the Park Hotel in Clinton for steaks. It was very pleasant; however, one day in the 703A conference room he said he wanted to tell us something we were not going to like-Savannah River was going to use the mixer-settlers being developed at KAPL. Maybe that was the night at the hotel when steak juices were dumped on Pots's new dacron suit. It didn't bother him a bit. He scrubbed it with wet rags, and it dried before we got home. He wore it the next day.

Purer Model for Geneva. When the first Atoms for Peace meeting was being planned for Geneva, I recommended, designed, and took an operating model of Purex to the meeting. While Tom Cole's operating reactor exhibit received the most attention, our Purex unit with Admiral Strauss operating the manipulators was the background for the TV program, "Youth Wants to Know."

24.2 Chem Tech Accomplishments

Purex Process and Solvent Extraction. During my time, the development of the Purex process was Chem Tech's most important achievement. Earlier, the other significant development (but attributable to DuPont's management and the University of Chicago's chemistry) was the semi-works and pilot plant phases of the BiP process for the first large-scale plutonium recovery at Hanford. This was followed by the first solvent extraction processes, Redox-25 and -23 processes.

Idaho Chemical Process Plant. The most complete job related to the ICPP needed to recover the enriched uranium from test reactor fuels. This work included the chemistry and process engineering development, along with the design of the plant. The design assumed direct maintenance of the equipment as employed in the Chem Tech pilot plant. While the ICPP is still operating, remote maintenance seems more attractive to achieve reliable performance. However, at this time

I am not aware of a comparative analysis of direct vs remote maintenance for radioactive operations.

Isotope Separations and Production. In addition, Chem Tech made numerous essential contributions to most of the other radiochemical process requirements of the young nuclear industry. These included the separation of thorium, ^{235}U and ^{233}U, $^{238}Pu_2$, ^{140}La, and the transuranic elements through ^{252}Cf. The isotopic separation of lithium was a major activity, but a Y- 12 process, COLEX, was chosen over Chem Tech's OREX process. Waste treatment, while the incentive for the Purex process, seemed always to be a secondary interest. Of course, for me, a spectacular event was the evaporator explosion at the pilot plant since, at the time, I was the Division Safety Officer.

Other Oak Ridge groups, now part of Chem Tech, contributed to uranium ore processing and radioactive and stable isotope production. Radioisotope production was a Semi-works task until Art Rupp came back to take this. work and, along with Eddie Beauchamp, form the Isotopes Group.

Ion Exchange and Fuel Element Shearing. The Higgins continuous ion-exchange column and the shear to prepare the stainless-clad power reactor fuel for dissolution seem to be the major process equipment developments of the division. Irwin Higgins developed the ion-exchange unit, patented it when the government gave him the rights, and established a company to apply and sell the technology. Clyde Watson led the shear development efforts that seem to have provided the basic technology now in use.

2d.3 Changing lime6

The Explosion. Upon arriving at the office one morning, our secretary told me that Floyd Culler had been there since about 4:00 AM following an explosion at the pilot plant. It turned out to be an evaporator in the plutonium cycle. They were decontaminating it for repair using a proprietary reagent. The reagent was very effective, but we had failed to convince the manufacturer to tell us what was in it. Their instructions warned not to boil it with nitric acid, which the operators were doing on the night shift with the cell door open. The reagent contained an organic material which, under the conditions, had been nitrated and subsequently detonated. At first, a nuclear event was suspected, but the absence of short-life activity quickly disproved that.

It was a small explosion. However, with the cell door open, the pilot plant and the immediate outside area were contaminated-an estimated 3 grams of plutonium were expelled from the pilot plant. Roofs of adjoining buildings, roads, and the general environment were contaminated. Floyd Culler assigned Jim Bresee to the cleanup, which took about a year and a lot of absorbent materials. When the costs were added up, we found that the total cost of the accident was about $1 million per gram of plutonium. As the division's continuing safety officer, I took one trip through the "dressage" that the cleanup crew did every day. Alvin Weinberg transferred Frank Bruce from our division to his staff in order, according to Weinberg, "to ensure nothing like that happened again!" Frank took Ted Arehart and Tom Hungerford with him.

Atomic Energy Commission. It wasn't long before I was in Washington working for the Atomic Energy Commission. This was in May of 1961, and I became familiar with criticality analysis, radioisotope production, reactor effluent system analysis, waste solidification, and finally disposal of radioactive wastes in deep mined geologic repositories. In 1955, returning from the Atoms for Peace Conference, I reviewed the waste solidification work at Harwell. Based on this work, Floyd and I discussed with Clarence Larson a project to solidify the wastes and remove tanks from the middle of ORNL. At that time, we had other priorities!

2.5 THE CHEMICAL TECHNOLOGY DIVISION—A UNIQUE TEAM: Personal Comments of Floyd Culler, June 1992[9]

Chemical operations and fuel reprocessing were very important factors contributing to the worldwide renown of Oak Ridge. The Chemical Technology Division (Chem Tech) received international recognition for its role in the development and use of nuclear and chemical technologies and is among the key divisions at the Oak Ridge National Laboratory. Strong continuity in engineering helped provide the impetus for the development of strong analytical chemistry capabilities, provided a focal point for the emergence of environmental work, and provided a basis for accomplishing all the AEC reactor analyses (we did 70 of them).

Mr. Floyd Culler as ORNL Deputy Director, about 1970.

2.5.1 Attributes of Chem Tech

Chem Tech possesses several attributes that contributed, in a large part, to its success as an ORNL division.

Learning and Growth Oriented. First is that we started out small, but we learned like crazy. We built more than almost anyone in the laboratory.

Chem Tech Spirit. The second feature, and probably the most important thing, is the spirit of Chem Tech. I've never thought of a better word than "gung-ho." Consequently, we were able to start up and to make contributions early on in order to get support. I think that spirit has prevailed until even now.

Teaming with Other Divisions and Industry. The third thing is that we teamed with other divisions as routinely as we worked with our own. Almost every division at ORNL has had joint programs with Chem Tech, except perhaps for the very fundamental ones, and we even worked closely with some of them. For example, we were close partners with the Chemistry Division. The Analytical Chemistry Division grew totally in parallel, and their competence grew as a result of Chem Tech's activities. The Metals and Ceramics Division was closely aligned with Chem Tech, and there were several formal agreements between John Fry and I about how we would manage.

Organizationally, we set up major and minor projects. The first major project was power 'reactor fuel processing. Jack Ullmann started this. From 1953 through around 1956, the Chem Tech Long-Range Planning Group had worked out the requirements for fuel reprocessing before we really got started in that area. By the time we were ready to talk about it, we had already done the primary calculations on what the actinides would be. We started the chemistry to separate them from the fission products. And when Seaborg asked to have a reactor built, we had the chemistry, not proven, but pretty well in hand. That was the plus that allowed the system to go. The consequences of these efforts at teaming were that we were able to either be a part of somebody else's program or they were a part of our program when multidisciplinary efforts were required.

The organizational structure that we set up with some forethought was successful. We instituted meetings once a month, including everybody in control or working on a project. The major projects were managed by people such as Murray Rosenthal or others who had the power of an associate laboratory director. The minor projects were managed by division directors. There were two rules: First of all, there was to be no arguments about territorial prerogatives. Fry and I agreed that we would cut people salaries, if necessary, and fire them after three such arguments. Everybody had the sense that there would be no territorial arguments, and if they started, either John or I would move in and adjust them. And there were not many. The other rule was that all questions that were raised, either technically or administratively, would be resolved at the meeting following the meeting at which they were raised. If the question was carried over to a third meeting, it had to be reported to the division director for resolution. We had very few that required the latter.

The volatility project, for instance, was run within our division. The section chief in charge of the pilot plant was also in charge of the fluoride volatility program. At some point the volatility project merged with the recovery of the Molten Salt Reactor (MSR) fuel. But the decisions regarding the volatility program were a little difficult. There was considerable debate about how to go, what to do, or how good the process was. I finally was called in one time to resolve an issue. I knew what was going on because I got reports from people involved in the project.

But that mechanism of being able to pair up-to team-was important. We brought in architect-engineers (AEs) from major industries. We generally had two or three AEs in residence at one time in the Design Section. And they too fitted in. They became essential parts of the laboratory and were very well integnted by the time they left. It was this process of being able to team up with other divisions that helped us achieve our objectives. For example, we just automatically said to the Instrument Department that we wanted two guys assigned who will coordinate for your division. And it worked marvelously. The ability to team is an important attribute.

Management Acceptance. The fourth attribute of Chem Tech was that the top laboratory and DOE management accepted reasonable ideas about what we should do. The long-range planning group kept us ahead. By 1958, we were ready to do conceptual designs of power reactor processing plants and had started processing.

2.5.2 Personal Comments

My great advantage, I think, is that I truly like working with people. The nice thing about working at Oak Ridge and the Electric Power Research Institute (EPRI) is that the people enjoy working with me, and I don't know exactly why. If I did, and if I knew how to generate the spirit that we had in Chem Tech, I would be a billionaire. I think one little tiny inkling of what it is is that I always looked on management as a necessary 'evil. And I was willing to do that and to make decisions if they were clearly indicated as necessary. If they were not, I would put off making decisions until I got more information. But at other times it was necessary for me to work as part of the team. That was a real distinction between what I, was doing in my mind as a manager and what I was doing to try to wrestle the problems. In that, I have been blessed with a good memory for details and numbers. When information came out of the labs in weekly or monthly reports, I'd read and say, 'That's a new number." I'd call the responsible people and ask, "What do you think this means?" I could usually remember the pivotal numbers about when something would work. I remember when information came out of the laboratory on the effectiveness of ferrous sulfamate as a reductant. That was the key, incidently, to a successful TBP process. I saw those data and said, "'We've got the process now; it doesn't have any salting agent and it cleanly separates uranium. We've got a holding reductant that works at least, and we've got a world beating process." The TBP process immediately assumed major importance; in three months, it was the process of choice. I remember that one very well.

But I did have this fortunate ability to remember details for long enough to make sure that we focused ourselves in directions that looked to be profitable.

I was always curious about everything. That's a virtue, I guess, but it may not be. But in this whole business of management, however, I think it is necessary to have people recognize that you have power but then not to exercise it. And, as I say, work as part of the group whenever it's possible.

I'm not sure that's an accepted method of management, but it worked pretty well.

It allowed everybody to think they were an essential and productive part of the team. I never quite understood. Everyone felt so responsible for seeing that their work was done right and quickly, and that good sense was being used. They were willing to work with each other, which is something that doesn't happen too often I discovered after I left Chem Tech. We really need some kind of an emulsifier to work together to common objectives, in teams and such. I think it was carryover from the war years. Everybody felt an urgency, and somehow we managed to keep it alive for a long time in Chem Tech. The reason is we kept getting big problems-one after the other.

2.5.3 Y- 12 Experience

Solvent Extraction. I transferred to ORNL in 1947. I came out of Y-12. I had just finished building the final product building for fully enriched uranium. I built Building 9212, and, basically, I operated the solvent extraction system. Actually John Strohecker and I built the first solvent extraction column, and neither one of us knew anything about it. It used glass columns about 8-in. in diameter, and we were using diethyl ether.

The extraction of uranium from nitric acid is possible if the uranium is salted out. of solution. With hexone you salt it out with aluminum nitrate. With our Y-12 solvent, we salted it out with copper nitrate. We had an absolute first call on all the pure electrolytic copper in the United States.

When we built 9212 we had the full output of the Coming Glass Works for six months. And we built it in 9 months. I started on it in March 1945, and it was completed and running in November 1945.

Criticality. Actually the last thing that I worked on at Y-12 was criticality. Ray Murray, now a retired physicist from the University of North Carolina, was familiar with criticality. Dixon Callahan was working on that too. The chemical systems at Y-12 for the calutrons, the big ones, always used big tanks, and the tanks were probably unsafe (in terms of criticality) above 6% enriched uranium. But we didn't know that for sure. The decision as to whether to use K-25 as a bottoming cycle for the calutron was made in early 1945. It was decided to go on up to full enrichment in the cascade. At that point, I'd been using the Fermi Age Equations for criticality calculations, along with Ray, who was teaching me this stuff. I did mostly the mechanical work and not too much of the physics. But we went over the whole plant to decide what was useful and what was not. We concluded that it would have to be totally rebuilt to go above even 6% enrichment, from what we knew. That sort of made the decision. That is when Y-12 shut down, and 25,000 people or so left.

Weapons Work and Lithium Isotope Separation. The weapons work had just started. John Strohecker was responsible for designing most of the weapons complex. There were two people who were really at the roots of it. John Googin, who is the only resident genius in Oak Ridge, and Strohecker who did most of it. Stmhecker and Googin were essentially the abiding geniuses behind the lithium isotope work. John Strohecker was the best engineer I ever worked with. Stroh could do things in his head that nobody else could do. At least that's the way it seemed to me. He and I built the first solvent extraction column. He had more nerve than I did.

Tennessee Eastman. Stmhecker and I were the only two engineers left in the Engineering Department. I was doing the airflow balances for the buildings. One of our friends, Merv Jones, along with Bill Hawkins, had left Y- 12 to work for Eugene Wigner in the early layout studies for the MTR. Merv kept telling me, "My God, you ought to come over here. All this stuff you've never heard of. It's a marvelous place to work. You really ought to come over." Well in 1947, as Tennessee Eastman was pulling out of Y-12, I had to make a decision as to whether or not I would go back to Rochester. I had worked there at the Eastman-Kodak home office for six months after I got out of Johns Hopkins. I had been working, as a neophyte, in engineering design. Eastman-Kodak sent me to Oak Ridge.

I stayed at Y-12 until Tennessee Eastman left. Strohecker decided to stay behind at Y -12, and I decided to go to X-10.

2.5.4 X-l 0 Experience

First Year. After several year's experience at Y-12, I transferred to ORNL in 1947. The thing that really helped me, and there are only two people who ever got to do it, was that the Laboratory had decided that they were going to train new technical people using a "Loop" course. I was allowed to work in Analytical Chemistry, Metallurgy, Reactor Technology, and in Unit Operations on shifts for Warren Eister. And it was . . also a year or so before the Oak Ridge School of Reactor Technology (OSORT.) started. But they were giving ORSORT-type lectures with Alvin and others lecturing. So I got to attend those.

For eleven months I did nothing but attend lectures and work with different groups, making reports once a month about what I was doing. My notebooks were embargoed as classified stuff, in some respects, at Oak Ridge. However, I used them all of my life there.

Technical Division and Design Work. I went to work for Frank Ward in the Design Section of Miles Leverett's Technical Division. I was assigned responsibility for all chemical plants under Jim Lane. Jim and I got to be great, good friends. At that point it was decided to go ahead with building the MTR and a chemical processing plant in Oak Ridge. It was before the decision to put it in Idaho. I started to do the materials balance flowsheet for the 25 hexone separation, including materials, heat, and activity balances. I did most of those myself.

U-235 Project/Chemical Processing Plant Design Group. As time wore on, it became obvious that the chemical processing plant was not going to be built in Oak Ridge but in Idaho. John Swarthout and I went along with the team that inspected the Idaho site before there was anything there, except the beginning of construction for the Experimental Breeder Reactor (EBR 2). Harold Lichtenberger was in charge of that. I remember the first trip-we walked all over that desolate high desert, but we picked a site for the chemical plant and started in with diligence on that. At that point, I essentially got a group to work on the enriched plant there. It became a formal design section under Frank. The transition from Miles Leverett to M. D. Peterson as director of the Technical Division occurred about the same time as the decision to move the reactor and processing plant. That occurred in the 1948 to 1949 time period.

M. D. Peterson had been in charge of a large group under Leverett. Essentially it functioned as an independent unit, as did the Metals and Ceramics Group. Metals and Ceramics was under Miles, as was Chem Tech and Analytical—everything except Chemistry, which was an independent division. But all the technical stuff was enfolded under Miles Leverett, and Peterson was the head of the Chemical Technology group. It was a big effort and became the Chemical Technology Division in about 1950.

Design Section. I essentially worked with a group on the ^{235}U project. The group grew almost immediately into the Design Section. About the time Peterson left the division and Steahly took over as Chem Tech Director, I was 'assigned the Design Section.

Chemical Technology Division. I became the Chem Tech Director in 1953. By then we had finished the design of the ICPP, and it was operating in 1953. I was busy like crazy during those days. We were doing a conceptual design for a thorium plant. It was thought that the military might want ^{233}U because of its lower critical mass, The thorium plant had problems, but at least we were making plans to produce large quantities of thorium, which we did. We produced the first kilogram quantities of ^{233}U in the pilot plant. The thorium was irradiated at Hanford, shipped back, and we processed the thorium.

Anecdote Concerning Processing Very Short-Cooled Irradiated Thorium. I was truly frightened when Earl called me to come to the pilot plant and we looked at the meter reading. Earl Shank was in charge of the irradiated thorium processing at the pilot plant. During the course of processing the relatively short-cooled, irradiated thorium slugs, the meter monitoring the off-gas filter in the penthouse of Building 3019 began showing extremely high radiation fields. Samples indicated the presence of radioisotopes usually found only during fission events. We were concerned that we might have a criticality event occurring. Intense research and discussion revealed that the particular isotope in question was also in the normal decay chain and not necessarily indicative of a criticality event. Thus, we were not dealing with a criticality event-just intense radiation from the short-lived activity.

2.5.5 Idaho Chemical Processing Plant

The ICPP was an important accomplishment. We had built a good team for that effort-a team with a gung-ho attitude.

Goeller and Radioactive Particulate Filters. During that period, no right after that, we built the first filters for radioactive particulates. Hal Goeller sketched them out over a weekend, and we had the , steel up the next week for the pile filter building! Goeller was great at long hand. He could do sketches fast. He was so accurate, and he sized it.

The decision to put a filter house on the graphite pile was made sometime around 1953 or 1954.

Expansion* of *Plant. We were also studying the expansion of capacity of the ^{235}U plant in Idaho to take care of enriched Hanford piles. Pure ^{235}U slugs were put in the Hanford piles to increase production, and they came to Idaho for processing. Milford joined the group when we started looking at thorium.

Foster Wheeler and Bechtel. We had Foster Wheeler and Bechtel engineers in residence at that time. For both of them, it was one of the very first exposures of the AEs to nuclear power. Bechtel was working on the fast reactor design with Harold Lichtenberg. Harold and I basically got to know the Bechtels pretty well, and they offered jobs to both of us in 1953.

ICPP Design/Construction Management. Six months into the design of the plant, the division of responsibility was clear. We had overall technical authority, and my signature had to be on the bottom of every drawing, which was unusual. It's the only time we ever had that requirement, I think; but the idea of criticality control and how to manage it was such new stuff, and I did know a little bit about it from my Y-12 background. It allowed us basically to get on with that fully enriched plant pretty quickly. We had material balance flowsheets, the sizes of all the vessels, the schematic piping diagrams with all the sampler points marked, the size of the cells and all the penetrations, and everything ready to go in less than six or seven months.

New York Assignment. At that point, the problem became one of translating the information onto the construction drawings, and to accomplish that, Goeller took about ten people to New York and worked for almost another year. We got checkprints every night back in Oak Ridge that had to be back in New York in three days. We flew them up to New York.

Checkprints. Steahly decided that he wanted to look at the checkprints. There were big rolls of them. Klotzbach was in New York too. Klotts sent me three copies of every checkprint, and we'd get a roll 6 in. in diameter. Everybody worked checking them out, and we checked everything. We wrote up the specs, we developed new standards for stainless-steel welding-ah that stuff. It was fun!

Steahly said that he wanted to see the checkprints before they were sent back to New York. I said okay. We put them all together for two days, and at 8 o'clock in the morning, we placed them on Steahly's desk and told him that we had two days turnaround and this was the second day and he had to'look at them and get them out. They came back about 15 minutes later.

Klotzbach had a great sense of humor. We sent big rolls of prints back to them in rolls, and he didn't have any way to file them. So he sent down the message, "Please send no more prints in the rolls." I guess the Idaho work made a big impression on me.

ICPP Team. I had a marvelous group of people working together-experienced engineers and good nuclear people. People like Paul Robertson, an old hard-headed piping engineer, and Bill Kersley. All of these were older people who were on the staff, and they all worked like crazy.

2.5.6 long-Range Planning Group

In 1953 we created a means of staying reasonably far ahead in thinking-the Long-Range Planning Group. We actually had the best functioning long-range group in the entire laboratory, and we kept the same four people on it all the time. Over a period of five to seven years, they learned how to calculate anything. For example, they started the ORIGEN code (Sect. 4.5).

Bob Klotzbach was on it, and Ferguson for a very brief time. Klotzbach went to Union Carbide. He was the first employee that the corporation took from the laboratory. He was familiar with handling radioactive materials and could do good material balances. He knew the ore chemistry pretty well, as did Keith Brown. Incidentally, Warren Grimes was a huge contributor to the chemistry effort.

2.5.7 Building

We learned about building by working with a relatively large number of AEs, in situ. We made arrangements to have their principal designers come and stay with us for awhile. This was Bechtel and Foster Wheeler first, then Kellogg, and later Catalytic Construction. We did conceptual designs of many things, including three repositories along with the Health Physics Division. Again, a teaming took place.

2.5.8 Seminars and information Meetings

We had weekly seminars and annual information meetings. The seminars didn't take long, and everybody had an opportunity to talk about their work each year or even less than a year. The annual information meeting was a more formal presentation to ORNL and AEC management, to invited guests from industry and universities, and to the Chem Tech staff. It provided an opportunity to inform management and interested people concerning Chem Tech work and activities and to permit Chem Tech staff to rub elbows with important guests.

2.5.9 Challenging Work and Careful Workers

Chem Tech helped develop the formulation for the concrete used in the Transuranium Processing (TRU) facility. The concrete was designed to optimize radiation protection. The problem was that of building a reasonable shield for both neutrons and gamma radiation. Neutrons go through most materials. Adsorption of neutrons requires something with high hydrogen content. The gammas required high concentrations of heavy metals. We used iron punchings and little disks that were suspended in the concrete, along with a mineral from Idaho that was an iron compound with six waters of hydration. The mineral was stable in a high radiation field, and we put a large amount in the mix. The concrete had a sickly yellow cast. The TRU work that was done then was fascinating-with the metallurgy, the refabrication of the fuel, all of the remote handling gadgets, and the very dangerous materials involved. They were very careful workers, the entire crew, and all of them worked like crazy on that project.

2.5.10 Atoms for Peace

The Atoms for Peace conferences were born in 1955. Prior to that, there was one or so meetings in the United States, with selected countries on reprocessing and reactor technology and such. Early on, of course, the British were our partners during the war years, and there were British scientists in residence at ORNL during the war years on chemical separations and other aspects of nuclear technology.

2.5.11 International Obligations

In 1953, we started getting ready to meet an obligation that the United States had with Belgium. In return for the pitchblende ore from the Belgian Congo that supplied all the early uranium for the United States, we had promised to provide Belgium with processing information. Consequently, several Chem Tech staff members participated in a very important workshop in Belgium, essentially giving them all the desired information.

About the time we were finished with Idaho, we had people from France and Sweden visiting, and we gave them overviews of the kinds-of technology that we were doing. In 1954, there was a conference that preceded the Geneva conference at which some of the reactor work was revealed. The reactor design work on all of the reactor types that were then current-information on the light-water reactor, the breeder, the gas-cooled reactor, the aqueous homogeneous reactor, and others-was made partially available to the Europeans on a selective basis, and the Canadians were always there.

But the international business really started heating up in return for the Belgian uranium, since the reactor work in 1955 at the Geneva conference was totally declassified. We gave descriptive papers only at Geneva. We outlined, but did not give details, of the reprocessing. For example, we did not tell *them* what the reducing agent was. We didn't give the mole ratios and all the necessary process information. Generally the solvent extraction flowsheets were given in semi-cartoon-like review without specifics.

The United States government negotiated with the Belgians and released details of the reprocessing, plutonium recovery and isolation, the uranium recycle, and what we knew about waste as repayment to them for the natural uranium from the Congo. About ten months of intensive declassification, much previously classified information, including all the Hanford data, manuals, and layouts, was bundled up, and twenty of us went to Belgium for three weeks. In three or four weeks' time we described everything to the Belgians and the other OECD countries. At the time the Europeans were forming an economic community under France, Germany, and Great Britain to build the European Common Market, but this was a manufacturing community initially. As a result of our agreement with the Belgians, their atomic energy fuel processing programs were started.

During the meeting in Geneva, three people came and asked me if I thought the United States would be willing to provide assistance in the construction and formation of a central reprocessing facility. I said that I would check with Washington. AEC Headquarters said they thought it would be a good idea. With two AEC people, I went to France and it was agreed then that there would be an attempt made to form essentially the European Processing Plant and later the King of Belgium gave the land at Mol. The whole effort got started. And along with that, the same group was working toward setting up the International Atomic Energy Agency (IAEA), which came into official existence in 1957.

So it was an interesting period of emergence. Immediately after the Geneva conference, we starting getting foreigners in residence-a very large contingent from France, a relatively large one from Germany, and a few from Belgium and Holland. There were many from India, probably at least 150 to 200 Indians who went through Chem Tech. We also had the Japanese in large numbers in later years. John Grover, United Kingdom, sent over people to work in Analytical Chemistry, a few in Metallurgy, and a large number in Chkm Tech. We looked at essentially all of their flow sheets for building their chemical plants. Those type of visitations continued for some time. At the same time, around 1955, Du Pont was beginning the design of Savannah River, and we had probably 50 to 60 Du Pont people in residence all the time.

2.5.12 Biotechnology and Continuing Education

The initiation of biotechnology work at Oak Ridge started in an odd way. Around 1950, and the dates could be corroborated, Frank Bruce and I set up a week-long meeting in Gatlinburg to which we invited 40 separation experts from all over the world. We were finished with the war. We had tried every separation method that had been devised by man including electrophoresis, for example. All of these had been tried and screened. We'd invented 400 new solvents that had varying capabilities for complexation. Our object was to see what we might be able to do with the powerful separations tools we had developed. I said that I thought we had the science and, in part, the technology to separate anything from anything, including the adjacent rare earths and the big macromolecules, in an opening comment at the meeting-a foolish comment.

Now the interesting thing was that John Slusher from England, who was at the meeting with several others, went back and quoted the comment in Nature. Alex Hollander, director of the ORNL Biology Division, read about it and came back and asked me if I had really meant it. I said, "Yes." And he said, "Well, why don't we do something about it." I replied, "Well, why not." This was a little later I suppose, probably 1951 or 1952—no, it was after 1953 because I wasn't working as hard on Idaho.

Culler's College. I had already decided that many Chem Techers had been out of school for as long as I had, and I was getting rusty as all get out. There were marvelous new techniques coming out, and I figured if we didn't become educated, we'd rapidly become outdated. So I set up some courses that had to be attended by people that had been out of college for ten years.

Well, there were foolish rules that I enjoyed. First of all, you could beg, borrow, cheat, or steal or anything to answer any of the homework questions. There were no quizzes. All you had to do was work all the homework; if you didn't, you were in trouble with me, but nobody ever reneged. The questions were hard, but I told everybody that I didn't care how they obtained the answers.

Incidently, that's the way Johns Hopkins is operated, except for a few courses. You could even go in some of the classes, for example, physical chemistry, and talk to your instructor during quizzes. The questions were hard; there weren't any multiple-choice questions.

Biotechnology Courses. Now what happened was that I said to Alex Hollander, "Alright now, I think biotechnology is advancing very rapidly and Waldo Cohn and several others have been separating the amino acids by ion exchange. There are techniques you are using that we could help with. If you will agree to set up a school that will train ten people every year in the fundamentals of molecular biology, I'll assign ten people out of Chem Tech to he trained in biology. But they must have the very best people lecturing in this effort." And, in a small way, that was the beginning of the post-doctoral biomedical school, and it worked.

Dave Novelli was assigned to coordinate the educational program. Dave was working on the purification of transfer RNA. That project became our major focal point. Later, Norm Anderson came in. He was well along in the zonal gradient centrifuge program. The group picked that technology up. But it started in this "little round robin" fashion back to Chem Tech through Alex.

We had already decided that one of the areas that was important was "big" biology. Although the biologists were geniuses at inventing separation methods, they were terrible slop-jar chemists. They did the right thing, but they didn't engineer them correctly. They always got mixtures of impure products. They didn't have the controls and the understanding of separations. So we put together a group of three or four physical chemists and three or four engineers. I have forgotten, but there may also have been a physicist in that first group. I have forgotten the names of these individuals.

Continuing Education. The school for training had other prerequisites. After you finished the school, and that was a full year, you had to agree for the next three years that you'd take some courses in the extension service at The University of Tennessee (UT). I indicated that I didn't care. what courses were taken. You could study archaeology or animal husbandry, just as long as you enrolled in something. But most of the people went back and took additional biotechnology. It was a marvelous setup. I never expected it to work as well as it did.

One thing I do remember about the "retread" program was that at the Chem Tech dance (the first year it was on St. Patrick's Day), all the wives came up to me and said they were going mad. Their husbands were working these problems all the time. They stayed up until 12:00 o'clock at night, and the wives were really angry. Some of them were really put out. They said, "You're ruining my marriage!" I said that was too bad, I feel sorry for you, but it's necessary. After it was all over, I think it made a little better impression. But the little incidents like that were interesting.

In the last decade, math had progressed and the computers were coming in. So, I decided that refresher courses, using our own people as teachers, would be useful to hone some of the mathematical skills of the Chem Tech staff. Some, of the more recent hires were quite skilled in mathematics, for example, Jere Nichols and Gene McNeese. And the courses turned out reasonably well. But the biology training was laced into that because both "schooling efforts" were going on at the same time.

The biotechnology efforts were a very important part of the division's history. I don't think Chuck Scott has ever quite got the recognition he deserves for his accomplishments in that area. There were many others who have also worked in it, but the whole program did not attract enough attention, a common failing of many advanced technologies.

Biology Division. At the height of the Biology Division's achievements, AEC Chairman Schlessinger described the division as being the crown jewel in the AEC's programs-a really impressive statement. All of those good people from Biology are either gone or dead, except Bill Russell and Lea. However, there are still some awfully good people over there.

2.5.13 Environmental Impact Statements

Chem Tech played an important role in experimentation and report preparation for the environmental impact statements. Several Chem Tech people were involved in calculating release functions. There is a little bit of background that is important about the generation of impact statements. Oak Ridge, because it had a large number of engineers and the very largest Environmental Division, was assigned 55 or 60 of the impact statements that had to written. In a relatively short time, we essentially formatted an approach to impact statements, and it brought us into close contact with the Environmental Sciences Division. The Reactor and Metals & Ceramics divisions also were deeply involved.

Basically, each of the impact statements was assigned to individuals as leaders in that particular program. Tom Row cut his teeth on environmental impact statements. He was in the Reactor Division at that time. Sam Beall was also involved.

2.5.14 Fuel Reprocessing

The emergence of the Reactor Fuel Reprocessing Division under Bill Burch was important, as well as the work that led up to it. It was the last remnant of chemical processing and technology that existed in the United States, except for rebuilding Savannah River. There is no real development now in the United States, but at some point in time, it's going to be necessary.

2.5.15 Nuclear Energy

Nuclear energy is now a very emotional issue. We have to be able to separate the actual hazard of radioactivity from radioactive materials better than we have. One interesting comment about the state of nuclear energy, made sometime in the last two years, is that all the people who have worked on the effects of radiation now have grey hair, and none of the modem techniques of medicine and biology have been used as measurement tools in the area. Twenty-five years have elapsed with no new work.

I think we're working on trying to achieve some new data. Basically, there are techniques like blood indicators that may be able to provide us with a better measure of radiation effects than we now have and better correlations with radiation damage. We are going to, I hope, begin to sponsor workshops among biomedical professionals. The Department of Energy has cut back, almost to nothing, studies on the effects of radiation. In fact they were doing most of the effects work on solvents and other materials, and that work has also been cut back. The ORNL Biology Division has been decimated.

Well, hopefully there is a new group being formed in the private sector. It may emerge as something called the Annapolis Center for Environmental Quality. Initially, they received a limited amount of seed money from the U.S. Manufacturer's Association. The man who is president is also on the Board of Directors at Houston-Power. He was head of the University of Texas, University of North Carolina, University of Maryland, and deputy head of the University of Chicago. He will be retiring in about six months from the Houston Board, but he's the new president of this Annapolis Center for Environmental Quality. A basic precept of the center is to see whether it's possible to foster good science and to respond to the exaggerations-to begin to treat the perception with the reality. It's a very difficult operation. Chauncy Starr (former President of EPRI) and I have been trying for seven years essentially to set up a center for evaluation. Basically, it can't be within the government, and it can't be contained within one of the national laboratories because that's also within the government. It can't be set up within EPRI either because even though we are reasonably objective, we're labeled as part of the establishment.

We first attempted to set up the medical and biological engineering departments at Stanford as such a center, using Oak Ridge for information data collection. The independent center was chartered to develop policy for handling resources for the future. We (EPRI) funded that at $500,000 per year for a few years. That got Started, and it's doing very well, but the important parts-the illumination with new information, the scientific analyses, and understanding what epidemiology is-these are only beginning.

2.6 THE CHEMICAL TECHNOLOGY DIVISION, FROM WARTIME SECRECY TO THE COMING OF THE RUSSIANS: Personal Recollections of Ray Blanco, May 1992[10]

I arrived in Oak Ridge in 1944 in the army and was assigned work at Y-12. Previously, I worked at an oil refinery and then served in the infantry. Eight people were selected from my unit at Fort McClelland and sent to a secret installation (no leave and mail censorship) to work in our professions (a great day!). First, we were given two weeks of lectures by British and U.S. scientists on

A 1958 photo of Ray Blanco (extreme right) with Wes Lewis (center), Chem Tech Pilot Plant manager, and Euratom representatives.

the chemistry and processing of the element T (uranium), and then we started work on process development, material balances, and as shift supervisors. The barracks at Oak Ridge were new and like heaven compared to other army camps. They were located on the site of the present Oak Ridge Shopping Mall.

I started at X-l.0 in 1946 and joined Frank Bruce's group developing the Redox and 25 solvent extraction processes. We were in the Technical Division in the section headed by M. D. Peterson. Experimental work was simpler then. We used lead brick barriers in the hoods or on the floor and used significant amounts of radioactive tracers or irradiated U dissolver solution. Arlene Kibbey, Jim Farmer, and I built an enclosure for an ion-exchange column against the exterior building wall. Of course, we were careful to wear our radiation exposure pencils and rings and use our "cutie pie" hand-held monitor.

In 1946 or 1947, the "powers that be" decided the technical staff needed more training in basic nuclear physics-probably so we wouldn't kill ourselves in a criticality event. We were asked to attend 1- to 2-h lectures each day for about two weeks by the eminent physicists that were then present on the staff, including Dr. Wigner and Dr. Weinberg. We learned about criticality, cross sections, isotope and fission products production, decay schemes, and much more.

In some of the earliest waste treatment development work, we collaborated with Irwin Higgins and Bill Shockley to develop a process for reducing the volume of wastes to. be stored. The early processes used aluminum. nitrate as a salting agent and produced large volumes of raffmate wastes. We developed an ion-exchange process to separate the aluminum from the fission products (FPs) so that the FP waste could be evaporated to a small volume for storage or solidification. A patent was issued for this process.

RaLa Process. About 1950, we started the development of an improved RaLa process. RaLa is the separation of kilocurie amounts of ^{140}Ba from irradiated uranium dissolver solution. Barium-140 radioactively decays with a 12.8-d half-life, thus producing ^{140}La, which in turn radioactively decays with a 40-h half-life, forming stable cerium. Los Alamos wanted the ^{140}La with its intense gamma radiationin the bomb development work. The early process, carried out in 704 D Building, used lead as a carrier to precipitate the sulfates of Pb, Ba, and Sr. Subsequent precipitation and separations steps using organic solvents were hampered by heating and solvent radiation decomposition problems.

Ion exchange seemed a good alternative to produce a $Ba(NO_3)_2$ product from the Pb-Ba-Sr sulfate precipitate of the old process. The process developed included (1) metathesis of $Pb\text{-}BaSO_4$ precipitate to Pb-BaC03 with K_2CO_3, (2) dissolution in $HNO_3\text{-}NaNO_3$, (3) adsorption on a column of Dowex-50 resin, (4) elution of Pb with NaOH, (5) elution of Sr with EDTA (ethylenediamine tetraacetic acid), (6) elution of Na with 1 *M* HCl, (7) elution of Ba with 9 *M* HNO_3, and (8) precipitation of $Ba(NO_3)_2$ from 85% ***HN03.***

The process equipment was installed in new cells in 704 D Building and was used successfully to produce products containing up to 40,000 Ci of ^{140}Ba, as I recall. I remember working all night during some of the runs and watching the recorder show the large radiation peaks for the elution of the Sr and Ba components.

Bill Unger and his co-workers designed the cells and equipment and got the job done successfully. He did express some resentment that chemists specified such "friendly" chemicals as HCl and fuming HN03. Such chemicals appeared to cause a materials of construction problem.

Lithium Isotope Separations. In 1952, we began the development of a process for separation of the lithium isotopes ^{6}Li and ^{7}Li. We first tried ion exchange based on the pioneering work of Taylor and Urey using zeolites (sodium aluminosilicates) and LiCl. We also tried other zeolites, ion-exchange resins (Dowex 50, Duolite C-6), LiEDTA, lithium acetate, and fixed columns versus continuous, moving columns of resin. It was concluded that the systems were impractical because of the low separation factors. J. Tom Roberts, Arlene Kibbey, Fred Land, Dennis Helton, Irwin Higgins, Bill Shockley, and I participated in this work, which resulted in the publication of a paper in ***Progress in Nuclear Energy,*** volume 4, 1961.

The OREX Process. After the ion-exchange effort forseparation of the lithium isotopes, the emphasis turned to contacting lithium amalgams with solutions of LiCl in organic solvents. Previous investigators had shown that acceptable separation factors could be obtained in these systems. Of the many available, we chose LiCl in propylene diamine (PDA). A method was devised for contacting the amalgam with LiCl-PDA in a packed column. The crucial issue was the development of a successful method for recycling the mercury and PDA at each end of the system. Thus, we had a true continuous countercurrent system with a relatively high separation factor operating in a packed column-the OREX process was born. In contrast, the COLEX process, developed at Y- 12, contacted lithium amalgams with aqueous LiCl in a countercurrent mixer settler system. A race developed between the two processes. This effort to produce ^{6}Li for thermonuclear devices had national priority, and both systems were rushed into pilot plant tests. I was a participant in the chemical development along with Ray Wymer, Arlene Kibbey, Fred Land, and Dennis Helton.

A design/construction contractor was hired to work with the Design, Pilot Plant, and Unit Operations sections of the Chemical Technology Division and rush a large pilot plant to completion. Dick Lindauer was in charge of the pilot plant operation.

As I recall, enrichments of up to 15 to 18% were obtained, indicating the theoretical success of the system and the potential to obtain full enrichment. However, the system failed because of practical problems. Because of the urgency, used and unsuitable equipment (pumps, etc.) had to be used along with available new equipment; also, the system could not be maintained in an anhydrous condition (moisture caused decomposition of the amalgam and loss of separation efficiency). Finally, the engineering problem of pumping mercury to the top of tall columns and general problems with just containing mercury became horrendous. Thus, the COLEX process won the race.

R&D Center for Fuel Reprocessing and Waste Management. From the late 1950s to the early 1970s, Chem Tech was the principal R&D center for fuel reprocessing and waste management studies in the United States. It became the focus for foreign visitors. Hanford and Savannah River were highly classified production sites, and foreign visitors were discouraged. A sampling of foreign guests Chem Tech hosted included visitors from the United Kingdom, Belgium, Italy, Japan, South Africa, West Germany, Sweden, Norway, Finland, Yugoslavia, Australia, and India. Some countries sent guest scientists and engineers to work in the Chemical Development and Pilot Plant Sections. Most guest assignments lasted 6 to 12 months, but some were up to 2 years. Countries assigning guest scientists to Chem Tech included South Africa, Spain, India, Italy, France, West Germany, Japan, Australia, and the United Kingdom.

Sidelights. An interesting sidelight occurred at the 1958 Geneva conference. I was talking shop with some British and French delegates when suddenly the Britisher said, "You know we recently bought a large amount of LiCl on the open market for our ^{6}Li/^{7}Li separations program and found that the ^{6}Li content was depleted." Of course, they suspected the U.S. was the culprit, but I could only smile-it was highly classified work at the time.

Another time at the 1958 conference, I visited the Russian exhibit. They had a number of interesting nuclear exhibits, but the featured item was a full-scale Sputnik highlighted in'the middle of the room. There was nobody there, so I looked for a Russian to ask some questions. Finally, someone came out of a back room and I asked, "Where is everybody?' "Oh," she said, "They are all over at the U.S. exhibit taking notes."

Symposium on the Reprocessing of Irradiated Fuels, 1957. In 1957, the U. S. AEC agreed to present information on the reprocessing of fuels to its World War II allies Belgium, France, the United Kingdom (UK), the Organization for European Economic Cooperation (OEEC), and the Euratom nations. This was a very important meeting because the U.S. revealed for the first time the engineering details of the reprocessing methods developed in this country. The 1955 Geneva conference revealed much previously classified information, but little detail. It appeared to me to be a debt payment to the Belgians for the uranium provided from the Belgian Congo as well as to the UK for their technical support.

It was a momentous occasion for all concerned, with the Prince of Belgium opening the ceremonies-champagne and all. Speakers were present from Hanford, Idaho Falls, Argonne, Knolls Atomic Laboratory, and ORNL. Papers were presented by F. L. Culler, F. R. Bruce, G. I. Cathers, J. Ullmann, and me from the Chem Tech Division. The proceedings were published in TID-7534. Sessions included aqueous reprocessing, auxiliary processes, disposal of plant effluents, non-aqueous processing, and engineering and economics.

After the meeting, the speakers and wives were guests at tours of the French reprocessing plant at Marcoule, the UK processing plant at Windscale, and the UK feed materials plant at Springfield, England.

As further assistance to the Belgians and OEEC, Ed Nicholson and Earl Shank were sent by Floyd Culler to Belgium in order to assist the conceptualization and design of their new reprocessing plant at Mol. Ed had done similar work with Floyd for the Idaho Chemical Processing Plant, and Earl had pilot plant experience.

Visits with the Russians, 196465. In 1964–65, the United States engaged in a cultural exchange program with the U.S.S.R. As part of the program, the State Department and the AEC sponsored technical exchange visits by U.S. and U.S.S.R. scientists on nuclear waste management, but only low-level wastes, since high-level wastes were produced by fuel reprocessing and were classified secret.

ORNL was asked to participate in a U.S. visit to the U.S.S.R. in December of 1964. Floyd Culler selected Frank Parker from Health Physics Division and me from Chem Tech to go. Joe Lewin from the Thermonuclear Division also went as interpreter. Other delegates included three from AEC Washington, one from Hanford, and one from Brookhaven (Fig. 2.3).

Fig. 2.3 The final 1964 technical exchange conference with the Russians took place in the Moscow office of Professor Academician Spitzen (back to camera). The delegates (clockwise from Dr. Spitzen) are Joe Lieberman, AEC; Al Platt, Hanford; L. P. Hatch, Johns Hopkins University, Alex Perge, AEC; Madame Breshneva, USSR; Ray Blanco, ORNL; Walter Belter, AEC; Joe Lewin, ORNL; and Frank Parker, ORNL.

The group departed from Paris on a Russian jet for Moscow but, because of fog, landed in Leningrad. The train to Moscow had 2 bunks in each compartment and was very crowded. One of the group was to share a compartment with a woman traveler—a common occurrence in Russia. The U.S. traveler, when asked if he would be embarrassed, said he would be (the woman indicated otherwise). Eventually, separate compartments were obtained.

The group visited the Atomic Research Institutes and the nuclear power reactors at Beloyarsk (near Sverdlovsk in Siberia), Moscow, Leningrad, Obninsk, Novovoranezh, and Karkov. In each case the group observed and discussed the low-level waste processing and storage facilities. They were studying scavenging-precipitation ion-exchange processes and incorporation into asphalt, the same as in our country. They were very interested in our results and freely presented their information. They even demonstrated for the U.S. group their cold pilot plant for solidification of intermediate- and low-level wastes into glass. Of course this process could be used for high-level wastes also (Fig. 2.4).

Fig. 2.4. The IAEA panel at the 1965 Low-Level Waste Conference, Subna, Russia, included W. H. Hardwick and R. H. Burns of the United Kingdom and Ray Blanco from far left to right; Professor Academician Spitzen, standing; and J. Rodler and C. Sombret of Francs (with faces showing) at the right of the conference table.

Sidelights. The Russians were extremely gracious hosts and provided us with seats at the Bolshoi and Kirov ballets, shopping and sightseeing tours, and visits to the Kremlin and Hermitage museums. Representatives from the U.S.S.R. State Committee of International. Relations were assigned to us as Visit Coordinator (A. A. Serov) and interpreter (G. V. Volkov). Fortunately, we had our own interpreter, Joe Lewin, who had lived in Leningrad until age 7. Thus, we could hear everything that was said at the conference tables. We called Serov "the man who walked on water." For example, as he led us into a very crowded department store, the crowd parted, put down the four hats they were looking at, and made room for us, with the clerk's immediate attention. Not a word had been spoken that we could detect. Special treatment on the trains was also accorded-with only two bathrooms on the sleeping cars, the eight Americans were assigned one while all the Russians had to use the other.

On arriving at Sverdlovsk by air, we loaded onto an army-type bus to ride to the nuclear power reactor at Beloyarsk—one Russian and one American per seat. Suddenly, my companion said, "See that field but there, that's where we shot down Gary Powers. A farmer reported the downed plane and was told to be quiet but he got drunk and talked to everyone. Why do you fly over our country with spy planes?" I said, "President Eisenhower has proclaimed the freedom of the skies for everyone including Russians.'* Then I countered by asking, "Why are the Russians helping the Chinese build an atom bomb?" He exclaimed, "We only help them with peaceful work-such as building bridges, etc.," but he didn't sound too sure of this. All this was said loud enough so that the entire bus heard it and a deathly silence set in until we reached our destination. All , in all, they were extremely friendly and served a, marvelous banquet in the evening, with toasts and speeches from both sides.

Russian Visit to ORNL, 1965. The return visit of the Russians to discuss waste management occurred in June 1965. They were a party of ten, headed by Professor V. I. Spitsyn, a member of the U.S.S.R. Academy of Science. Frank Parker and I acted as the immediate contact hosts.

They arrived at ORNL from the Holiday Inn and were welcomed by Alvin Weinberg. A series of presentations were ma& in the conference room, followed by lunch and a tour of the waste facilities, including the low-level waste treatment plant, the hydrofracture plant, the Operation Division's waste monitoring and control building, White Oak Creek White Oak Lake, and burial grounds. The tour route had been carefully planned as to what they could see or photograph. Much classified work was still in progress at that time.

A number of items of interest occurred. The morning meeting had barely started when a secretary came in with a message to the meeting coordinator (I believe it was Jim Cox, Operations Division Director) that a large amount of money had been found under the pillows in one of the visitors' hotel rooms (about $7000, as I recall). This caused quite a stir, and one of the Russian visitors left immediately to get it. I guess he carried the money for the entire group.

When we arrived at the White Oak Creek bridge, every Russian raised his camera for permission to shoot. This creek was-famous worldwide after the American papers at the Geneva conference had described it as the outlet for ORNL-treated low-level wastes.

I took them on a bus tour of the city of Oak Ridge. On passing the old Oak Ridge Associated Universities site, I said that the building contained a large ^{60}Co source for cancer therapy. Their question was "Did the patients have to pay for their treatment?" "Yes, they did," I answered. The Russian said that they didn't have to pay for medical treatment and smiled. We then went to Pennsylvania Avenue and east on Cuter Drive. I could hear them talking in Russian, and I gathered that they were commenting on all the cars, boats and the individual homes—not apartments.

Alvin Weinberg hosted a dinner at the Holiday Inn. The Russians had produced a quart of caviar as a gift, and when it wasn't served promptly, a Russian went into the kitchen to see if it had disappeared, it was only delayed. Afterwards, the visitors and ORNL participants were divided between three homes for a visit-Frank Parker, Joe Levin, and me. Spitsyn was in my party. I remember him saying to my wife, "I have only one request. My wife wants me to see if you have a phone in your kitchen." Fortunately, we could show him one.

Alvin Weinberg hosted a final meeting in his conference room and presented them a token of . Tennessee hospitality (the encased Indian arrowheads) as a symbol of friendship and good will. At the post-visit meeting, we all decided that what our guests had liked best was the pop-open-type beer cans-just opening the cans was more fun than drinking the beer.

2.7 IDAHO CHEMICAL PROCESSING PLANT—DESIGN, CONSTRUCTION, AND STARTUP: Personal Recollections of Hal Goeller, April 1992[11]

Conception and design of the MTR had begun at ORNL toward the end of the 1940s. It was the first reactor to employ a fully enriched (93% ^{235}U) fuel. Since the fuel cannot be completely "burned up" during reactor operation, it must be removed from the reactor after being partially consumed and chemically processed for re-enrichment and reuse by removal of the fission products and transuranic elements generated by burning. Such processing could not be done in the existing facilities at ORNL and Hanford, Washington, because criticality, as well as radioactive shielding, had to be considered in its design: hence, a new facility was needed. The new facility was to have been built on the side of the hill south of and facing the site of the present-day 4500 buildings. The AEC ruled, however, that a new area be set up for the MTR and the processing plant. Selection was soon narrowed to either Fort Peck, Montana, or Arco, Idaho, with the latter becoming the final choice. This area belonged to the U. S. Navy as a test firing range for 16-in. guns being made at Pocatello, Idaho. One reason for its selection was the existence of fairly extensive central facilities.

Conceptual design was started in early 1950 at ORNL on chemical process flow sheets for dissolving the fuel and solvent extraction removal of fission products and transuranics. Preliminary design of the equipment and of a processing building followed. The process dictated that all operations be done remotely behind 5 ft of concrete shielding.

Foster Wheeler Company in New York City was selected to do the detailed design from the preliminary designs we provided them. In order to expedite this effort, about six of us worked with them in New York from April through July 1951 until detailed design was completed. In the meantime, the Bechtel Company of San Francisco was selected to do site preparation and construction, which began early in 195 1. Alex MacIntosh, an ORNL architect, supervised construction of the main buildings.

Nearly a dozen of us followed in August 1951 to supervise fabrication and installation of, equipment and startup of the plant by American Cyanamid Corporation, who was selected by the AEC to be the plant operator. The last of us returned to ORNL during the summer of 1953. After a year on the job, Cyanamid elected to leave, and operations were taken over by Phillips Petroleum Company, who was already operating the MTR at the Idaho site.

The ORNL Idaho crew included Frank Browder, Ed Frederick, Hal Goeller, Frank Harrington, Bill Kearsley, Bob Klotzbach, Ed Nicholson, Frank Peishel, Paul Robertson, Al Rom, and John Ruch. Everyone thoroughly enjoyed their Idaho stay despite the fact that we were working a six-day week and then a seven-day week for a while, Four or five "Idaho offspring" were born during the stay.

The plant went "hot" in late 1953 with the processing of enriched slugs from Hanford. In addition, there were some enriched materials from Savannah River and from MTR fuel (Fig. 2.5). Later on, the facility processed Experimental Breeder Reactor (EBR-1) fuel. Incidentally, the EBR-1 was the fist reactor in the world to produce electricity, though a minimal amount, namely, one light bulb. The Navy also built its nuclear submarine prototype reactor in Idaho during our stay (Fig. 2.6). The ICPP is still operating, and very successfully!

2.8 THE CHEMICAL TECHNOLOGY DIVISION FROM 1953 TO 199 1: Personal Recollections of Ray Wymer[12]

I joined Chem Tech in October of 1953 in Chemical Development Section B. This section was one of three chemical development sections and was headed up by Ray Blanco. There was, as I recall, also a design section, a unit operations section, a pilot plant section, and a long-range planning group.

In 1953 the 4500 Building had just been completed and occupied. Up until then, the Chemical Technology Division Office was housed in Building 3550. Parts of the division were located in several other buildings around the ORNL site. Then, as now, Chem Tech was scattered all over ORNL.

Floyd Culler became Chem Tech division director in 1953. Frank- Bruce was Associate Director. Don Ferguson, Ray Blanco, and Bill Lanham were the section chiefs of the Chemical Development sections. I believe that Kurt Jackson was section chief of the Pilot Plant Section, and Warren Eister was section chief of the Unit Operations Section. (Back in those days we called them chiefs instead of heads.)

OREX and Mercury Amalgams. The big deal in Chem Tech in 1953 was development of a lithium isotope separation process called OREX. Y-12 was developing acompetitive process called COLEX. There was a life-and-death competition between ORNL and Y-12 over which of the two competing isotope separation processes would eventually be used. The process was ultimately to be installed at Y-12, and perhaps, not surprisingly, the Y-12 process won the competition. (I am perhaps showing a little provincial bias in the way I worded that last sentence. Actually, COLEX was probably the better process from a practical point of view, but OREX was more elegant and imaginative.)

Both isotope separation processes used equilibration of lithium isotopes between two liquid phases. The separation took place without any net transfer of moles of lithium between the phases. Only the isotopes exchanged phases. One process fluid in both processes was a liquid lithium amalgam. The other liquid was a solution of lithium chloride. The ORNL and Y- 12 processes differed in the nature of the solvent used to dissolve the lithium-chloride. (Lithium isotope separation using lithium amalgams was first suggested and successfully used by G. N. Lewis back in the middle 1930s. He used equilibration between a lithium amalgam and lithium chloride dissolved in diethyl ether.)

The interesting thing about this isotope-separation process development was that neither ORNL nor Y-12 had any previous experience with handling, pumping, storing, or carrying out any operations with amalgams. Of course the specific gravity of the amalgams was about 13. This is "a little" beyond the experience of most chemical engineers. The OREX pilot plant was in Buildings 4505 and 4501. It was not uncommon to go into those buildings and see pumps leak amalgam as they strained to lift and move them, or to see pipes at the bottom of a long vertical run burst under the extreme pressure head. Amalgam showers were frequent.

Not only were the chemical engineers not accustomed to handling mercury and its amalgams, neither were the medical nor the industrial safety

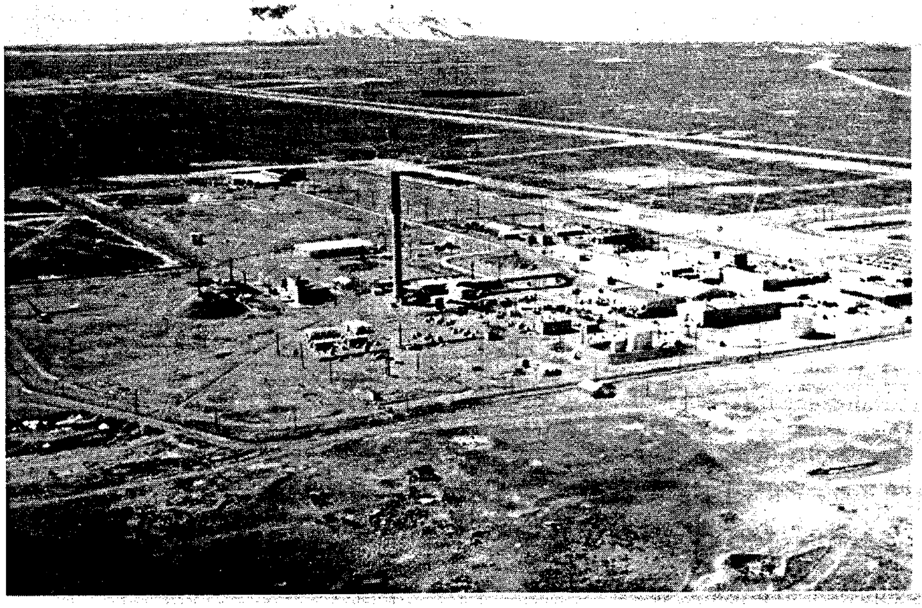

Fig. 2.5. The Idaho **Chemical** Processing Plant In this 1976 **aerial** photograph shows the plant essentially unchanged from Its original design and construction. The photograph was provided through the courtesy of Lloyd McClure, Idaho National Engineering Laboratory.

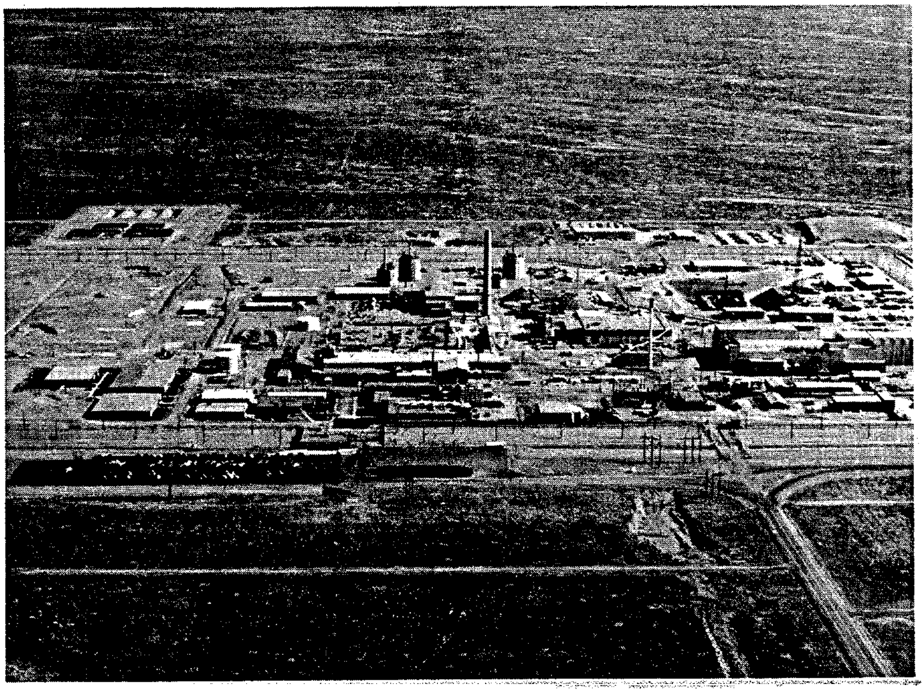

Fig. 2.6. This 1989 aerial photograph of the **Idaho Chemical** Processing Plant shows **considerable modification and change** of the original installation that were **required** to modernize and update the processing **plant. The** photograph was provided through the courtesy of Lloyd McClure, Idaho National Engineering Laboratory.

departments accustomed to protecting the people who worked with them. My co-workers and I went down to a local dispensary in the 4500 Building once a week and wrote our names on a sheet of paper. Our signatures were compared with our earlier signatures to see if there were any signs of palsy! That was the test used to see if we were getting mercury poisoning! We protected ourselves from mercury vapors in the laboratories by sprinkling "flowers of sulfur" around on spills. This was supposed to convert the mercury to non-volatile mercuric sulfide. I suppose it did-eventually.

Geneva Conference. The next thing of importance that I recall is the first Geneva conference. This took place in the mid-1950s. ORNL was importantly involved. Members of Chem Tech wrote a handful of papers, and we contributed several operating exhibits, including a solvent extraction column that Fred Land and I put together that was supposed to illustrate the principles involved in solvent extraction purification of uranium and plutonium. A big secret in those days was how we took iodine out of the off-gas from a reprocessing plant. Any reference to that technology was carefully excised from the papers submitted. Presumably it was possible to take iodine release data and calculate plutonium production rates. Now, of course, we lean over backward to make sure that everyone knows how to retain iodine so that radioiodine is not released to the environment.

Reactors. Ail during those early days, ORNL was working on one or another of the advanced reactor programs being proposed at that time. A very early program was the Aircraft Reactor Experiment. This was a reactor that was supposed to propel an airplane. The crew was to be shielded rather than the reactor. Presumably the, thing would have to land over a tunnel through which the crew would escape.

Weinberg and his associates always had a new reactor concept they were espousing. These concepts were all similar in basic outline but quite different in implementation. Weinberg used to refer to them as "a pot, a pipe, and a pump" to stress their supposed inherent simplicity. In concept they were simple. Materials problems were their undoing. I never worked on any of those programs, but a lot of Chem Tech people did. The first big one for Chem Tech was the Aqueous Homogeneous Reactor Program. People like Don Ferguson, Bob McNees (who later went on to be the mayor of Oak Ridge for many years), Chuck Schiiiing, John McBride, Bill Pattison, Leon Morse, and many others were in the middle of those programs.

Another reactor program that was big for Chem Tech was the Molten Salt Reactor Program. Chem Tech people a little more contemporary were involved in this program. These included Bob Hightower and Gene McNeese, to name two who went on to bigger and better things. All of these reactor concepts were based on the thorium-uranium fuel cycle. Since there is a huge amount of thorium in some granite rock, Weinberg talked about "burning the rocks." (He is a great phrase maker.)

Fluoride Volatility Process. An early non-aqueous process for reprocessing reactor fuels was the Fluoride Volatility process. ORNL was championing the use of fluorine gas to volatilize uranium as UF_6. ANL was championing the use of ClF_3 or ClF_5 for the same purpose. This difference lead to an intense rivalry between the chemical engineering divisions at the two laboratories, which has only in the last 5 or 6 years been finally and completely laid to rest. At that time, Chem Tech and a process engineering group at K-25 were collaborating in a sort of guarded way. K-25 had a lot experience in handling UF_6 and felt that they had considerable to offer ORNL. Strong personalities at both sites made collaboration rather difficult. Process equipment for a Volatility pilot plant was partially installed in Building 3019, but the AEC cut off funding before the program was completed.

Waste Disposal. Starting in the 1960s, Chem Tech was playing a prominent role nationally in the waste disposal business. Geological disposal was recognized very early as the only long-term solution to disposing of high-level radioactive wastes. Chem Tech was deeply involved in a pilot project in a salt mine in Lyons, Kansas. A prominent Kansas legislator strongly objected to the idea saying, "Let those who have feasted on the atomic turkey bury its bones." However, when it was learned that the area was riddled with oil-prospecting drill holes, it became obvious that the proposed disposal site was not and could not be isolated from water. This was not only the death knell for the Kansas site, but also for geological disposal for the time being. Chem Tech involvement in the project was such that its shiny reputation got a little tarnished.

High-Temperature Gas-Cooled Reactor. At about this time frame, the High-Temperature Gas-Cooled Reactor (HTGR) came into the ascendancy. It was an AEC program whose principal advocate was General Atomic. Partly because it was based on the thorium fuel, a cycle to which ORNL had a major commitment, a large part of the program was given to ORNL. We worked on fuel fabrication and reprocessing, as well as having a significant role in fuel testing. (Morris Osborne and others are still doing similar testing work to this day.) Pete Lotts had the programmatic responsibility for part of the fuel fabrication work. I led the effort in Chem Tech. The whole activity was under the direction of Don Ferguson at first, and then under Paul Kasten. This work came a cropper when General Atomic couldn't get the prototype 300-MW(e) HTGR at Fort St. Vrain to operate well. Although General Atomic had accepted several orders for large HTGRs, they reneged on the orders. That was the end of the program for all practical purposes, although AEC had a collaborative program with Germany for quite a few years after that.

HTGR fuel was to be reprocessed by burning the graphite fuel elements to liberate the uranium and thorium particles, which were the actual fuel and blanket, respectively. Bob Lowrie was a key player in this aspect of the program. As I recall, Chuck Scott also had some role to play, as did Ron Canon.

Burning the graphite produced a carbon dioxide off-gas that contained ^{85}Kr that had to be removed. Ron Glass worked on this problem, along with Vic Fowier and others. For a time, Bob Merriman also worked on this part of the program. The off-gas work and the sol-gel operations were a success, even though the patient died.

Sol-Gel ***Process.*** It was the HTGR fuel fabrication work that got Chem Tech into the sol-gel process development work. Some related work had gone on before in connection with the aqueous homogeneous reactor, but it was the HTGR fuels work that brought people like Claude Haws, Paul Haas, Al Irvine, John McBride, Ken McCorkle, and many others into the sol-gel fold.

TRU Programs. About 22 years ago, the High-Flux Isotope Reactor (HFIR) and Transuranium Processing Plant (TRU) were built. TRU design and construction was a major achievement of Floyd Cuiier, with a major assist from people like Bill Burch, Frank Peishei, Orion Yarbro, Bill Unger, Hal Goeiier, and many others. TRU [now called Radiochemical Engineering Development Center (REDC)] operations have been a mainstay in Chem Tech for more than twenty years, and TRU continues to be a major Chem Tech activity center to this day. The basic processes used in TRU in the early days are essentially the same ones that are used today. People like Rex Leuze, Russ Baybarz, Milt Lloyd, John Chiiton, John Bigeiow, Dave Campbell, Walt Bond and many others played key roles in developing the processes used.

Fuel Reprocessing Activities. About fifteen years ago, Bill Burch convinced Don Trauger and others that the fuel reprocessing activities in Chem Tech should be split off and put into a new division under Burch. At that point Chem Tech lost what had been a major part of its responsibility from its inception. Partly to compensate for that loss, and partly because it was the right time to do it, Chem Tech turned toward waste and the environment as major areas for its involvement. Those are major thrusts of the division to this day.

Biotechnology. Also, about that many years ago (my memory is dim on exactly when), Chem Tech started a modest effort in biotechnology under Chuck Scott. That area has been slow aboming but has been a steady activity for many years. At the present it appears to be gaining in strength.

Apologia. There are many areas that I have given short shrift to. But time available to think about the past is short, and so is memory. At any rate, this is a personal account, not a disciplined attempt to be thorough and accurate. Please excuse the oversights and errors. Perhaps the recollections of others will fill in the gaps I have left and correct the errors.

2.9 OVERVIEW OF EARLY CHEM TECH PROGRAMS

Most of the early activities and programs of Chem Tech were continuations of the important R&D activities of the Chemical Technology Department of the Technical Division. The major programs of national importance focused on reactor fuel reprocessing and the recovery of uranium, plutonium, and other radioisotopes. The major programs are presented chronologically in Table 2.1 and discussed briefly below. Selected facilities and other Chem Tech activities are also, discussed as appropriate to help present a balanced picture of the division involvement.

Table 2.1. Chronology of ORNL/Chem Tech reprocessing activities

Period	Process
1943-1945	Bismuth phosphate
1945-1951	Redox
1945-1952	RaLa
1946-1948	Hexone-25
1946-1948	Hexone-23
1948-1949	Uranyi ammonium phosphate
1948-1958	Metal recovery
1948-1953	TBP-25[a]
1949-1960	Purex
1949-1968	Fluoride volatility[b]
1949-1976	Fuel preparation[c]
195 1-1976	Raw materials[d]
1952	TBP-Interim-23
1952–present	Thorex
1953-1959	Feed materials[e]
1955-1976	Head-end[f]
1%l-1976	TRW

[a] Includes homogeneous reactor fuel processing.

[b] Includes ARE and MSRE fuel reprocessing.

[c] Includes aqueous sulfate fuels (HRE), sol-gel, carbide-graphite-oxide spheres (HTGR, EGCR, Rover), and molten salts (MSRE, ARE).

[d] Includes SLURREX, AMEX, DAPEX, MONEX, and other processes.

[e] Includes EXCER, METALLEX, FLUOROX, DRUHM.

[f] Includes mechanical methods, DAREX, ZIRCEX, ZIRFLEX, Voloxidation, etc.

[g] Includes TRAMEX, CLEANEX, BERKEX, PLURIX, and others.

Source: R. E. Brooksbank, Sr., ***Historical and Programmatic Overview of Building 3019,*** ORNL/CF-91/298 (July 17.1991).

ORNL/Chem Tech Pilot Plant. Since early 1943, the ORNL/Chem Tech-managed pilot plant, Building 3019 (formerly known as Building 205), has served as a pilot plant in the development of several radiochemical processes that have found plant-scale application in government and commercial facilities both nationally and internationally. In addition to the process development role, the facility's operations have also produced large quantities of product materials (plutonium, uranium of ail isotopes, thorium, and special isotopes) while processing highly irradiated fuel. The Chem Tech pilot plant‘ was used for process studies and production for the following processes: Bismuth Phosphate, Redox, Purex, Fluoride Volatility, Fuel Preparation, and Thorex. The pilot plant and operations are described in detail in Sect. 4.17.

Redox Process. The Redox process separated and recovered uranium and plutonium from irradiated fuel using hexone as a solvent and $Al(NO_3)_3$ as salting agent.[13] The solvent extraction process was developed by ANL for separating and purifying uranium and plutonium. The process promised to be more simple and economical than the bismuth phosphate process. Soon after the war, ORNL was requested to test and improve the Redox process in its pilot plant facility. ORNL modified the process and demonstrated production of sufficiently pure uranium. In 1952 a new $60 million chemical plant using this process was constructed at Hanford.[14]

RaLa Process. Chemists in the Technical Division had developed a laboratory-scale process for extracting ^{140}Ba from fission products to provide a source of ^{140}La with its high energy radiation for use in Los Alamos studies. Engineering-scale equipment was designed and installed in hot cells of Building 706C. By September 1944, curie quantities of ^{140}Ba were being produced and design was begun on equipment for producing 1000-Ci batches of ^{140}Ba. In the first half of 1945, Building 706D and its hot cells were built, equipment was installed and tested, and two production runs were made. The second run produced 1180 Ci ^{140}Ba. The Technical Division operated the plant until 1946, when it was turned over to the Operations Division.[15] A major problem in the RaLa process was the intense radioactivity resulting in radiolysis of the water. The first RaLa process involved precipitation with gravity settling, which did not work well because of the radiolytic gas formation.[16] The experimental work sometimes involved high levels of radioactivity that glowed in the darkened laboratory.17 In 1949 ion-exchange runs were made with large amounts of radioactivity, demonstrating the feasibility of an improved RaLa process.[18]

"25" Process. The MTR at Arco, Idaho, required a new chemical process to recover the highly enriched uranium from irradiated uranium-aluminum alloy fuel elements. Chem Tech staff chemists and chemical engineers were principally responsible for development and pilot plant testing of the "25" process.[14,18] The process recovered uranium from irradiated fuel by solvent extraction using hexone.[13] In 1949 the Chem Tech pilot plant had successfully completed the final development of the "25" process through the first and second cycles and was in the final stages of development on the isolation cycle.[18] The "25" process formed the basis for chemical separations used in the ICPP.[14]

Idaho Chemical Processing Plant. Initiated in 1950, the effort to develop a fuel reprocessing plant to be sited in Idaho rapidly developed into a major program area. The Design Section of Chem Tech was given the responsibility for the process design of ICPP with Foster Wheeler Corporation as AE and Bechtel as the construction engineering firm. It was one of the earliest experiences of these AEs with nuclear technology. Design was essentially complete in 1951-1952. Management of the complex design effort involved coordination with. several AEC offices, the Foster Wheeler Corporation, and other ORNL divisions. To assist in the design of the ICPP, a team of Chem Tech staff members was assigned to the New York office of Foster Wheeler. Construction of the plant had progressed to the point in early 1952 that Chem Tech staff members were then assigned to Idaho to assist in the field engineering, testing, and startup of the plant.2 The total capital cost of the ICPP was $31,106,000.[19] The philosophy of direct maintenance was used in this plant for the first time, all previous plants having used remote-control maintenance. The plant, completed in 1952, and process were sufficiently versatile to routinely process fuel elements from MTR, EBR, STR, and other reactors."

Solvent Extraction Recovery of ***^{233}U.*** During 194649, the Technical Division processed irradiated thorium oxycarbonate in 706A (part of Building 3550 that has now been razed). This work was done in equipment designed by John "Tex" Blomeke which was installed in one of the four hot cells in the Semi-works area. Some of the solvent extraction work to isolate ^{233}U was carried out in one of the hot cells in the, Chem Tech Pilot Plant Building 3019 (formerly 205). One of the solvents tested was diisopropyl ether.[21]

"23" Process. The "23" process was developed to separate ^{233}U from irradiated thorium by hexone extraction. Chem Tech pilot plant tests were initiated in 1949.[18] The Interim-23 process for isolated ^{233}U from irradiated thorium and fission products was developed for the production of kilogram quantities of weapons-grade ^{233}U. Slugs irradiated at Hanford were shipped in special shielded carriers to the ORNL/Chem Tech pilot plant and processed at 70 kg Th/day. In 1952, a total of 2.5 kg ^{233}U was isolated.[14]

Purex Process. The Purex process used solvent extraction with TBP and HNO_3 for salting out effects to, separate and recover uranium and plutonium from irradiated natural uranium fuel.[13,18] By the summer of 1949, the process was shown to be feasible and more economical than previous processes. An additional advantage was the separation and isolation of fission products which simplified radioactive waste storage problems. In 1950, the Purex process was selected by DuPont for use in the Savannah River Project. The major chemical development work for the process was done at ORNL. In 1954 the Purex process was used in two separations plants at Savannah River costing $75 million each. In 1956 a modified Purex plant was constructed at Hanford costing $78 million.[14] The Purex process is the process of choice for use in all present and planned reactor fuel reprocessing plants throughout the world.[22]

TBP Process. In 1949, ORNL initiated the development of a process for the recovery and purification of uranium from its metal wastes stored in the tank farms.[14,18] Many tons of metal wastes were stored in tanks at ORNL and Hanford. For example, the ORNL metal waste tanks contained 155 tons of uranium precipitated as a carbonate or hydroxide form.[23] The process developed used TBP for extraction of uranium and plutonium from the waste. An early complication of the plutonium recovery from ORNL metal waste was the discovery that 40–60% of the plutonium had polymerized on storage in the alkaline metal waste and had precipitated as a sludge. Digestion at 70°C in 3.5 M excess nitric acid for 4 h solubilized 95% of the plutonium.23 The TBP metal recovery process was installed at Hanford at a cost of $35 million. Chem Tech designed and constructed the Metal Recovery Plant to use the TBP process.[14]

Metal Recovery Plant (Building 3505).

The Metal Recovery pilot plant (Building 3505) was constructed in 195 1 at a cost of $500,000. The facility was designed to demonstrate a chemical process for recovering uranium from Oak Ridge National Laboratory and Hanford tank-farm wastes by continuous extraction with TBP in a kerosene-type diluent (Amsco). The plant was expanded at a cost of $300,000 to include a dissolver for solid materials, two additional solvent-extraction cycles, plutonium isolation equipment, and piping changes to permit processing of various feed materials for recovery of plutonium, americium, and neptunium as well as uranium. These included metallurgical wastes resulting from the reduction and calcination of plutonium compounds; ash residues from the conversion of UO_3 to UF_6; sodium diuranate carbonate sludge and supematant accumulated in ORNL tanks; uranyl nitrate solutions in tank car lots and plutonium nitrate from Chalk River; and spent fuel elements from Chalk River, Clementine (Los Alamos Scientific Laboratory), Graphite, Brookhaven National Laboratory, and ANL reactors, All the fuel elements were uranium clad in aluminum except the Clementine, which were nickel-coated plutonium alloy encased in mild, steel. The maximum capacity of the plant is 450 kg of uranium per day, which is reached in the processing of natural-uranium fuel elements.[24]

The basic design philosophy for the Metal Recovery pilot plant was that the equipment would be remotely operated but directly maintained. Remote operation of radiochemical plants is necessary to prevent exposure of personnel to radiation, and in earlier radiochemical production plants, maintenance was also carried out remotely. In the Oak Ridge National Laboratory Metal Recovery pilot plant, process equipment is arranged according to its function and activity level so that only the equipment associated with that requiring maintenance must be decontaminated. The greatest source of radioactivity, the dissolver, is in a cubicle by itself. Cells containing the rest of the equipment are isolated from each other, and each contains only related parts of process equipment. Certain vulnerable pieces of equipment which might require frequent maintenance are unit-shielded with lead. The exposure of maintenance men to radioactivity in 7 years of operation averaged 60 mrem/week, which is 20% of the permissible exposure. Because of limited shielding; the plant is restricted to the processing of materials of intermediate irradiation levels with long cooling times.[24]

As stated above, the ORNL Metal Recovery Plant used the TBP process. The new facility was first used in the AEC high-priority program for separating uranium and plutonium from Chalk River reactor fuel elements.[14] Subsequently, ORNL metal waste was processed and it was reported that 95 tons of uranium and 208 g plutonium had been recovered from the waste after operating for one year. The plant was converted to process Hanford slag and crucible waste, and in three weeks of operation it had recovered 1500 g plutonium and 0.1 g americium.[20] By September 1954, The Metal Recovery Plant had recovered 67 kg plutonium and 10 g americium from Hanford slag and crucible waste. Although the initially separated americium was contaminated with 3000 g lantbanum, the newly developed ion exchange–citrate elution process reduced the lanthanum contamination and yielded an americium product with a lanthanum-to-americium ratio of 1:100.[19] The facility was also used to recover uranium and plutonium from the' Brookhaven National Laboratory reactor, the ANL

CP-2 and CP-3 reactors, and the “Fermi pile” fuel.[14,16]

Thorex Process. The application of TBP extraction to thorium recovery was named the Thorex Process.[18] The Thorex process was developed to separate ^{233}Pa, ^{233}U, thorium, and fission products from each other. Major objectives of the process included isolation of ^{233}U for weapons development, separation of ^{233}U from thorium blankets in breeder reactors, and identification of a source of ^{233}Pa and isotopically pure ^{233}U.[13,14] The Thorex process pilot plant was installed and tested in 1954 on both the pilot plant and semi-production scales[14,19] During the development work, approximately 80 kg ^{233}U was separated along with 45 metric tons of thorium. The separation of ^{233}Pa as a specific product (an early objective of the Thorex program) was not achieved. However, isotopically pure ^{233}U resulting from the decay of ^{233}Pa was recovered by reprocessing the waste.[14]

> The Thorex pilot plant was installed in existing facilities (Building 3019) in 1955 to demonstrate the recovery of thorium and ^{233}U from irradiated thorium, decontaminated from radioactive ions, as aqueous products suitable for further direct handling. The original equipment was designed for material irradiated to 1500 g of ^{233}U per ton and decayed 200 days; this equipment was installed at an initial cost of $1,250,000. The plant was expanded from a one-cycle process[25] to a multicycle process, providing considerable experience with plant decontamination and equipment maintenance. The subsequent modifications made to permit processing of material irradiated to 4000 g of ^{233}U per ton decayed 30 days and recovery of isotopically pure ^{233}U after decay storage of the ^{233}Pa cost $750,000, bringing the total capital investment to $2,000,000. The annual operating cost has been about $1,100,000. The plant has also processed the Idaho Materials Testing Reactor assemblies for ^{237}Np and ^{235}U recovery and is considered suitable for processing thorium oxide slurries used in homogeneous reactor development work. The maximum capacity of the plant is 150 kg of thorium per day with thorium metal slugs.24
>
> Like the metal recovery pilot plant, the Thorex pilot plant is remotely operated and directly maintained. Major pieces of equipment are individually shielded by either cubicles or unit shielding and are easily accessible from a nonradioactive area. In addition, major pieces of equipment and cubicles are provided with built-in decontaminating facilities. Sample conveyors are shielded. Unique equipment items are concatenated pulsed columns, special pulse generators, and airlift-operated radioactive solution samplers. With 200-day-decayed material, radiation exposure to operating personnel averaged 10% of permissible: even during processing of short-decayed material, exposure was below permissible amounts.[24]

Early Fluoride Volutility Studies. As early as 1943, the Metallurgical Laboratory initiated scouting studies at the X site (Clinton Laboratories) of the Dry Fluoride Process for separations purposes.[26,27] Sometime around 1949, fluoride volatility studies were again started. Direct reaction of uranium metal with elemental fluoride was studied, and some work was carried out to investigate the reaction of uranium in bromine trifluoride. There may have been some early work in the 706A Semi-works (part of Building 3550 that has since been razed) that was later transferred to a laboratory in Corridor A of 4500N.[21]

Semi-Continuous Ion Exchanger. Chem Tech staff member I. R. Higgins developed a semi-continuous ion-exchange system.2 The ion-exchange system was used for a variety of separations, for example, see Sect. 2.10, Raw Material Processing.

Chalk River: Recovery of ***Plutonium in Chem Tech Pilot Plant.*** In 1950 the Chem Tech pilot plant began separating plutonium from Chalk River-irradiated uranium using the Redox flow sheet.[23] Within several months, it was reported that the plutonium loss across the Redox process fuel cycle and second plutonium cycle ranged from 0.8 to 2.5%, with gross fission product decontamination factors of 5×10^5 to 1.8×10^6. The pilot plant phase of the campaign was completed, yielding a plutonium product which contained 0.2 to 0.4 g of uranium per gram of plutonium. This product was ready for final concentration and purification.[4] The ion-exchange isolation step for recovery of plutonium from Chalk River-irradiated metal was called the SCRUP program.2

HOPE. The original Hope Project study, which was made in the summer of 1953,[28] was reactivated for a two-year Hope program during 1954–1955.[29] The original objective was to think of innovative and very inexpensive ways to reprocess reactor fuels.[30,31] The Chem Tech participants in that study directed by Eugene Wigner and Bob Charpie were Hal Goeller, Bob Klotzbach, and Ed Nicholson. The original Hope study assumed that a thermal reactor power economy would use reactors with ^{233}U fuel.

The purpose of the revitalized program was to study in greater detail all ideas presented in the original work.29 plus all others that showed promise of effecting reductions in radiochemical processing costs. Much of the effort was directed toward testing features of the Hope type of design for a Savannah River ^{235}U separation facility. The overall objective of the program was to demonstrate that enriched ^{235}U could be recovered from irradiated fuel elements at a cost that was compatible with production of economically competitive power from nuclear reactors.[28,32] The areas of radiochemical processing that appeared most susceptible to cost reduction and consequently were studied in the program were a new approach to criticality (i.e., always-safe equipment); underwater maintenance; feasibility of radioactive waste disposal methods other than storage in stainless steel tanks; a more reasonable approach to fissile material accountability: reduction of plant inventory costs: development of a continuous slug charger; and development of samplers and pumps. The Chem Tech staff members participating in the new Hope program were Baird Bottenfield, Al Irvine, D. 0. Darby, G. W. T. Kearsley, J. P. Jarvis, and Florence Isenhour.[29,32]

Special Equipment Tests. Much equipment development and testing were required to ensure safe and reliable operation of the equipment under adverse environmental conditions such as high levels of radioactivity, a corrosive environment, and high temperatures. Examples of equipment tested for resistance to radiation include valves, pipe dope, hoods, lead shot transfer jets, pulsafeeder pumps, steam traps, plastic polymers, paint surfaces, pipe and tubing fittings, magnetic-induction flowmeters, spray nozzles, electrostatic precipitators, and filters.[23,33,34]

2.10 OVERVIEW OF NUCLEAR REACTOR FUEL REPROCESSING

Chem Tech has been involved in fuel preparation or reprocessing studies on most, if not all, the reactor programs that ORNL either participated in or directed. Such programs included support work for the MTR and principal responsibilities for the homogeneous reactor series, the molten salt reactor, and the aircraft reactor experiment. Later reactor-related studies included safety analyses, fuel materials and fission product behaviour, and support work for the HTGR Program.

2.10.1 ORNL Reactor Studies

Brief summaries of the major reactor programs follow.

Materials Test Reactor (MTR). In 1948, ORNL reactor development efforts were concentrated entirely on the design of the MTR, which was to be constructed at the new National Reactor Testing Station in Idaho. The MTR concept of parallel plates with water moderator and coolant had a profound influence on the development of power reactors.[14]

Homogeneous Reactor (HR). The homogeneous reactor concept had been dropped in 1945 due to what had been thought to be insurmountable engineering problems, such as the formation of bubbles in the fuel solution as a result of the decomposition of water in the strong radiation field (this was later solved using dissolved copper as a catalyst to recombine the radiolytic hydrogen and oxygen). Greater engineering experience and new conceptual designs for homogeneous reactors appeared promising in 1949. Therefore the AEC gave approval for R&D leading to construction of a homogeneous reactor (HRE- 1). I-IRE- 1 was completed in January 1952 and operated successfully until 1954. Design of HRE-2 was started in January 1954, and the reactor went critical December 27, 1957, operating at full power in February 1958. The homogeneous reactor test (HRT) operations achieved a unique record for reactors at that time by operating continuously for 100 days. Phase separation problems in the uranyl sulfate solution and resulting corrosion of reactor materials at the hot spots resulted in AEC discontinuing the aqueous homogeneous reactor program, although it continued support for thorium breeder technology. A thorium breeder reactor. development became the objective of the new thorium utilization program initiated in 1961.[14]

Aircraft Reactor Experiment (ARE). On September 1, 1949, the AEC authorized ORNL to establish an Aircraft Nuclear Propulsion program (ANP). An experimental aircraft reactor (ARE) was completed and operated in October 1954. The ARE operated at full design power for 100 hr, as planned. The fuel consisted of $NaF\text{-}ZrF_4\text{-}UF_4$, and operational temperatures of 1200–1500°F were required. The use of molten salt fuel and reliable operation at such elevated temperatures were remarkable technological achievements. Early in FY1958, the ANP was discontinued because of costs as well as changing military needs, and ongoing work was phased out by 1961.[14]

Molten Salt Reactor (MSR). The molten salt reactor (MSR) concept originated out of the ANP research. The MSR was attractive for civilian power production because of low fuel cost and high thermal efficiency.14 In addition to R&D efforts on the MSR, ORNL actively engaged in the development of a molten-salt breeder reactor, (MSBR) that could produce low-cost power while producing its own fuel—^{233}U from ^{232}Th in amounts larger than it consumes.[35] It would use a molten fluoride salt, $LiF\text{-}BeF_2\text{-}ThF_4$ (76-16-12 mol %), as a fluid fuel and graphite as a moderator. The MSBR would succeed as a breeder only if the ^{233}Pa (27.4-d half-life) could be isolated at a rate significantly higher than its decay rate and if the rare-earth fission products were removed on a cycle of between 30 and 100 d. Chem Tech received the responsibility of developing an appropriate MSBR fuel process.[35]

High-Temperature Gas-Cooled Reactor (HTGR). *The* HTGR was considered to be a promising thermal reactor for the conversion of ^{232}Th to ^{233}U and the production of economical electric power. The Ft. St. Vrain reactor was the only HTGR reactor operated in the United States. The HTGR fissile fuels consisted of $^{235}UC_2$ particles, and fertile fuels consisted of $(U,Th)O_2$ and ThC_2 particles. The design for HTGR fuels included the use of coated (e.g., pyrocarbon layers) particles bonded into graphite sticks and inserted into holes in large graphite blocks of hexagonal cross section.36 Attaining the goals of economical power and improved utilization of uranium and, thorium resources required the development of an efficient fuel cycle, including fuel reprocessing and fabrication of recycle fuel.36

Liquid-Metal Fast Breeder Reactor (LMFBR). The Liquid-Metal Fast Breeder Reactor uses plutonium rather than ^{235}U as the major fissile fuel. The content of fissionable material in the LMFBR is higher by a factor of 7 in the core se&on as compared with light-water reactors, and the specific power of the LMFBR is higher by a factor of 5. The reactor name is derived from its use of unmoderated or fast neutrons for fission and breeding, as well as its use of a liquid metal (e.g., sodium) for the primary coolant. The fission product content of the irradiated fuel is higher than that of light-water reactors (locally by three times higher). Because of the high fissionable material content of the reactor core (and consequent high inventory costs) processing of the LMFBR fuel after only short decay periods will be advantageous.[37] Although not directly responsible for the design of the LMFBR, ORNL and Chem Tech were intimately involved as technical support in many areas.[38]

High-Flux Isotope Reactor (HFIR). The Transuranium Processing Plant (TRU) and the High-Flux Isotope Reactor (HFIR) were built at ORNL to produce large quantities of the heavy actinide elements as part of the AEC Heavy Element Production Program. These materials were used in basic research in laboratories throughout the country. In 1966 ^{242}Pu target irradiation was started in HFIR, and TRU began hot operations. During the first year of operation, more than 40 shipments of transuranium elements were made to other national laboratories, universities, and industries in this country. Shipments were also made to three foreign countries. The program phases managed by Chem Tech were operation of TRU, final isolation of the hansuranium elements, and development of chemical separations processes and equipment[38] (Sect. 4.10).

2.10.2 Nuclear Fuel Processing and Reprocessing

The development of reprocessing methods for nuclear power reactor fuels has been a major effort at ORNL and in Chem Tech. The principal work prior to 1961 covered dissolution procedures for stainless steel and zirconium-clad fuels. The resulting solutions were processed by established solvent extraction procedures.[14] An excellent and detailed summary of Chem Tech reprocessing experience is provided in Tables 4.1–4.5 of sect. 4.17.

Shear-Leach Process. The shear-leach process consists of shearing stainless steel or Ziicaloyclad tubular UO_2-bearing fuel elements and leaching the UO_2 from the sheared fuel tube with nitric acid in preparation for solvent extraction. The process was developed by Chem Tech staff member Clyde Watson. It was used for nuclear fuel preparation by Nuclear Fuel Services at the first commercial fuel processing plant in the United States at West Valley, New York.39 All power reactor processing plants worldwide use the shear-leach process.[30]

Darex Process. The Darex process was developed for stainless steel-jacketed fuels and used a mixture of boiling hydrochloric and nitric acids (aqua regia) to dissolve the stainless steel jacket.[14]

Sulfex Process. An alternate process for stainless steel-jacketed fuels was the Silfex process that dissolved the stainless steel jacket in sulfuric acid and the fuel materials in nitric acid.14 The only plant-scale use of the Sulfex process was at the Eurochemic Plant in Mol, Belgium.[30]

Zirflex Process. Zirflex was a similar process for zirconium-jacketed fuel using ammonium fluorideammonium nitrate solution to dissolve the zirconium jacket followed by nitric acid dissolution of the core. Such procedures were especially applicable to fuels with uranium and thorium oxide cores such as Commonwealth Edison, Yankee Atomic, and Consolidated Edison.[14]

Zircex Process. The Zircex process, using HCI gas to remove the zirconium cladding as $ZrCl_4$, was successfully tested on unirradiated zirconium-bearing fuel elements.[14]

Excer Process. The uranyl nitrate product of solvent extraction processes such as Redox and Purex is converted to UF4 in preparation for gaseous diffusion recycle as UF_6. This was usually accomplished by costly reduction with hydrogen and hydrogen fluoride. The Excer process involved aqueous phase hydrofluorination of uranium to UF_4. However, the process did not have sufficient economic advantage to replace the existing gas phase plants.[14] A modification, the Excer-Moving Bed process, for converting uranyl nitrate to UF_4 consisted of denitration of the uranyl nitrate to UO_3, reduction to UO_2, and hydrofluorination to UF4. The process was successfully demonstrated in bench-scale experiments in 1954.[19]

Fluorox Process. The Fluorox process involved reaction of UF_4 with oxygen to produce UF_6 and uranyl fluoride. The process promised some economy by using oxygen instead of elemental fluorine to convert UF_4 to UF_6.[14]

Metallex Process. The Metallex process for conversion of thorium tetrachloride to thorium metal ingots involves dissolving thorium tetrachloride in anhydrous propylene diamine (PDA) and reducing the thorium by contact with sodium or lithium amalgam at temperatures below 100°C. A button of thorium metal is formed from the resulting thorium quasi amalgam by filtering, cold-pressing, and melting the metallic product.[19] The process appeared to be competitive with other proposed reduction methods.[14]

Druhm Process. The Druhm process involves reduction of UF_6 by sodium or lithium amalgam. Yields of 80-85% were obtained.[14]

Homogeneous Reactor Chemistry/Fuel and Blanket Processing and Development Studies. Processes for Removal of Plutonium from Homogeneous Reactor Blankets. R&D directed at, removal of plutonium from HRE blanket solutions was started sometime around 1954 or later. In 1957, some of the laboratory-scale developments were reported These studies were made in Building 3508 where some steel cells were built to protect personnel in case the high-pressure equipment failed. A uranyl sulfate solution containing dissolved plutonium was heated to 250°C under an overpressure of mixed hydrogen and oxygen. The plutonium precipitated as oxide, but considerable amounts deposited on stainless steel coupons suspended in the solution. Plans were to remove the plutonium oxide as a concentrated slurry from the HRE blanket solution with a hydroclone.[21]

ARE Fuel Aqueous Processing. In 1954 fused salt fuel from the ANP-ARE was processed to recover the uranium. The fuel containing 4 kg ^{235}U in open-top cans was processed in the ORNL Metal Recovery Plant (Building 3505). The uranium was extracted from an aqueous aluminum nitrate solution of the ARE fuel by TBP.[20]

ARE Fuel Volatility Process Study. A molten salt dissolution fluoride-volatility process for preparing fresh ARE fuel from the used material was demonstrated in preliminary tests conducted in 1954. The molten $NaF\text{-}ZrF_4\text{-}UF_4$ salt was fluorinated, forming UF_6, which was volatilized, condensed in a cold trap, resublimed, and, subsequently, dissolved in fresh $NaF\text{-}ZrF_4$. The uranium recovery was greater than 99.95%, and decontamination factors (DF) of 4000–5000, sufficiently high for refabricating ARE fuel, were achieved. It was believed that DFs for uranium of 20,000 could be achieved by the process.[19]

Fluoride Volatility Studies. In 1957 laboratory-scale development of a volatilization process was reported at the First Nuclear Engineering Science Congress in Cleveland, Ohio. Experimental work was carried out by G. I. Cathers in which uranium was dissolved at 675°C in a fused salt composed of Zr, K, and Na fluorides. Anhydrous I-IF was bubbled through the salt containing a piece of uranium metal to form UF_4, which dissolved in the fused salt. The salt containing the UF_4 was treated with elemental fluorine to distill out UF_6.[21] Previous laboratory-scale studies had been conducted on the dry fluoride process using chlorine trifluoride as the fluorinating agent instead of elemental fluorine.

The purpose of the fluoride-volatility program in the United States was to develop an alternative to conventional aqueous processes for recovering uranium from spent nuclear fuels. Apparent advantages of volatility processes, compared with aqueous processes, included a high degree of separation of the uranium from its fission products, ease of processing certain refractory fuels, and increased nuclear safety. Principal disadvantages appeared to be the use of hazardous chemicals (e.g., fluorine, hydrofluoric acid) and high operating temperatures combined with extremely corrosive chemicals required the use of exotic metals and unique materials of construction. The. Chem Tech studies progressed through all R&D stages from the laboratory in 4500N, to hot-cell-level radioactivity pilot plant in Building 4507, to unit operations studies in Building 3592, and to full-scale successful operation of the Fluoride Volatility Process Plant in Building 3019 (see sect. 4.11).

Molten Salt Reactor Fuel Processing. The method proposed in 1960 for processing fused $^7LiF\text{-}BeF_2\text{-}UF_4$ fuel was fluorination of the UF_4, volatilization of the product UF_6, and separation of fission products from the residual $LiF\text{-}BeF_2$ carrier salt by dissolution in HF. Economically the fluoride volatility process appeared adequate for uranium recovery from both fuel and blanket salts, and the HF dissolution process for ^7Li-salt recovery, including replacement of fuel-carrier salt on a long cycle, appeared adequate for fuel-salt poison control. The first required process application would be for uranium recovery from both fuel and blanket salts which, with fuel salt replacement, would provide adequate processing for a converter reactor. Decontamination of the fuel-carrier salt would be required only with breeder reactors. It was proposed that thorium recovery and blanket-salt decontamination would not be needed for decades. Laboratory studies indicated that decontamination of the total fuel in one step was also feasible using 5 mol % NO_2 in anhydrous HF to dissolve the LiF, BeF_2, and UF_4 at 25°C and separating the materials from the insoluble rare-earth and thorium fluorides. The decontaminated fuel could be recycled after distilling off the NO_2 and HF.[40]

Chem Tech was challenged with the responsibility for developing an on-site, compact, low-inventory, high-performance, economical processing plant to process MSBR fuel. The process would be required to isolate ^{233}Pa (27.4-d half-life) at a rate significantly higher than its decay rate and remove the rare-earth fission products on a cycle of between 30 and 100 days. The process proposed employed, in addition to fluorination and UF_6 recovery, liquid-liquid extraction of the reactor salt with a bismuth phase containing reductants.[35]

Work in support of the MSBR concept was interrupted from January 1973 to January 1974 when the Molten Salt Reactor Program was discontinued for a one-year period.[41,42]

Reprocessing for HTGR Fuels. The reprocessing of HTGR fuels requires the burning of graphite blocks containing coated fuel particles in the form of fuel sticks. Chem Tech studies were primarily hot-cell tests with irradiated HTGR fuel specimens, development of burner technology using unirradiated fuel, and development of methods for decontaminating the burner off-gas. Assistance in the areas of equipment flow sheets, layouts, and cost information was also provided to the ICPP in the planning of HTGR fuel reprocessing.[43] Some work on development of reprocessing methods for HTGR fuel was reported in 1973. This included burning the fuel and separating krypton from the carbon dioxide off-gas by fractional distillation.[21]

LMFBR Fuel Reprocessing. As early as 1967, Chem Tech was involved in studying mechanical fuel disassembly and shear-leaching, methods for disposal of residual sodium coolant, and solvent extraction processes in addition to conducting economic analyses.[38]

The preparation of LMFBR fuel for solvent extraction is more difficult than that of light-water reactors fuel because of increased heat generation, greatly increased amounts of radioiodine (if the fuel is processed after short decay periods), and the presence of substantial quantities of relatively difficult-to-dissolve plutonium and fission products. The Purex process appeared to be applicable. The treatment of off-gas is a major consideration in the processing of fuels that have been cooled less than 120 days. Removal efficiencies for radioiodine of 10^8 are required for large-scale plants which process fuel that has been cooled for about 30 days. By 1971 Chem Tech's LMFBR studies had expanded significantly, including shipping of fuel: heat transport; head-end' processing of fuel including dismantling and shearing; deactivation of sodium; removal of volatile fission products: dissolution of UO_2 and PuO_2 fuel; solvent extraction: volatilization of radioiodine; off-gas treatment for removal of iodine; and radiation, shielding, and criticality studies.[37]

2.10.3 Early Nuclear Calculations

Nuclear scientists and engineers were quick to take advantage of the newly developing computer technology (e.g., the Oracle, an electronic digital computer developed for ORNL), which appeared to have considerable potential for calculating fission product yields as well as the buildup of uranium and transuranic elements through neutron adsorption and decay reactions. Chem Tech engineer John "Tex" Blomeke initiated calculations of heavy isotope buildup[44] and was assisted by Mary F. Todd, of the Mathematics Panel, in the voluminous calculations required for ^{235}U fission product production.[45,46] These calculations were the forerunner, and laid the initial groundwork, for the development of the Origen computer code (see Sect. 4.5).

Heavy Isotope Buildup. Neutron irradiation of uranium results in formation of a number of uranium and transuranic isotopes, some of which have nuclear properties that render them undesirable as reactor constituents. The effect of these isotopes on reactivity, based on single, long-term irradiation of fuel of various enrichments, has been considered by many investigators. Chem Tech engineers Jack Ullmann and Ed Arnold investigated the buildup of ^{236}U, ^{237}U, ^{237}Np, and ^{238}Pu in recycled ^{235}U fuels and showed how, even with partial removal of ^{236}U in each cycle, these products could grow to such levels as to influence the type and frequency of chemical processing and fuel fabrication.[44,47]

Calculations were initially made by Blomeke using an analog computer. The calculations permitted rapid estimation of heavy isotope concentrations in uranium fuel of any likely initial isotopic composition. Thus the calculation methods were especially suitable for studying buildup in fuel recycled a number of times through chemical processing, fabrication, and irradiation. Results were presented in the form of curves expressing growth of individual isotopes from pure ^{233}U, ^{234}U, ^{235}U, ^{236}U, and ^{238}U during irradiation at constant flux. Curves were computed for seven values of thermal neutron flux between 10^{12} and 10^{15} neutrons/cm^2/sec and irradiations up to 3×10^{21} neutrons/cm^2.[44,47]

Fission-Product Calculations. The production of fission products during reactor operations is an important consideration in almost every phase of atomic energy operations. The presence of fission products must be considered in the design and operation of nuclear reactors because of their contribution to neutron poisoning. However, fission products are extremely important in chemical processing facilities. Fission products and their concentrations determine the type of chemical processing necessary for separation of unused fissionable or fertile material as well as the shielding, off-gas treatment, and waste disposal methods required for successful and safe operations. Extensive information must be available on the chemical and nuclear properties of fission product mixtures to be encountered during fuel reprocessing. The fission product level in an irradiated reactor fuel is a function of three parameters: reactor operating power level, irradiation time, and decay time or time elapsed since reactor shutdown (or removal of fuel from operating reactors). Blomeke and Todd developed computer programs that computed levels of fission products resulting from thermal fission of ^{235}U in reactor fuels over a wide range of reactor operating conditions and decay times. Values for approximately 300 fission products were calculated, including gross totals for activities, radiation powers, and thermal neutron poisoning data.[45,46] In their voluminous report, Blomeke and Todd acknowledge the contributions of C. P. Hubbard, S. G. Campbell, and C. L. Gerberich for Oracle code and operations assistance: H. E. Goeller, W. H. Sullivan, H. S. Pomerance, H. E. Williamson, R. A. Char-pie,

R. W. Stoughton, and J. Halperin of ORNL; and L. E. Glendenin and E. P. Steinberg of Argonne National Laboratory.

2.11 OVERVIEW OF RAW MATERIALS PROCESSING

Raw Material Processing. The discovery that certain solvents and reagents could extract uranium from the sulfate solutions that were used to leach uranium from ores was an important technological achievement. The Dapex process used dialkyl phoshoric acid for both uranium and vanadium recovery. The Amex process uses a long-chain alkyl amine for uranium recovery. These solvent extraction procedures increased uranium recovery and decreased chemical costs.[14] The Monex process used TBP to extract thorium from unclarified Brazilian sludge leach liquor. Operationally tested through engineering-scale studies, the Monex program was terminated when the AEC requirements for thorium were reduced.[13] For uranium recovery from ore leach liquors, the Higgins continuous ipn-exchange system also showed significant reduction of cost as compared with conventional processes. The ion exchanger was successfully tested at the pilot plant scale. However, because existing uranium processing plants were operating below their capacities, the system was not further evaluated.[14]

Slurrex Process. In 1950, the AEC requested ORNL to make a preliminary evaluation of various solvents for recovery and purification of uranium from ore concentrates. Ethyl ether, a highly volatile and flammable solvent, had been used for this purpose. ORNL showed TBP to be a promising alternative. The Slurrex process using TBP as a solvent was subsequently developed in collaboration with Mallinckrodt Chemical Works and the Catalytic Construction Co.[14] The process was demonstrated at ORNL and consisted of extraction of nitric acid slurries of uranium ore with 30% TBP, scrubbing with 0.1 volume of hot water (80°C), and stripping the uranium with equal volumes of hot water.[1] A $20 million plant using the Slurrex process was constructed for use at the Feed Materials Production Center, Femald, Ohio.[14]

2.12 ISOTOPES PRODUCTION AND SEPARATION

Because of early and major involvement in separations of uranium, plutonium, and fission product isotopes, the continuing interest and involvement of Chem Tech in isotopes separations evolved in a natural way. The major programs are summarized briefly below and discussed in some detail in Sect. 4.

Around 1954, research and process development was started on separations of alpha-emitting radioactive materials. Solvent extraction and ion exchange were the primary techniques used: however, some precipitation methods were also developed.

- ***Americium.*** By 1958, 40 g of ^{241}Am had been recovered from a concentrate prepared by Los Alamos from plutonium metal reduction slag. The material was processed in the Metal Recovery Building by solvent extraction and purified in Building 3508 by ion exchange.[21]
- ***Neptunium.*** By 1958,670 g of ^{237}Np had also been recovered from Paducah fluorination ash. The initial recovery was made by solvent extraction in the Metal Recovery Building. A precipitation method was used for the final purification in Building 3508. This was accomplished by alternate oxidation and reduction of the neptunium in a fluoride solution.[21]
- ***Pluronium.*** In October 1958, plans were under way to irradiate ^{239}Pu to high burnup. These plans included irradiation of an MTR fuel element fabricated from plutonium and irradiation'of 12 kg Pu-239 at Savannah River.[21]
- ***Curium.*** In 1963-1964, ^{242}Cm was prepared jointly by Chem Tech and the Isotopes Division for use in heat sources. Chem Tech processed irradiated pellets of americium oxide and aluminum in the Building 4507 hot cells. The purified solution of americium and curium was delivered to the Isotopes Division.[21]

Preparation of ^{232}U. In 1962, ^{232}U$_2$ was prepared by irradiating about 40 g ^{231}Pa oxide loaned to us by the British. The irradiated material was processed in Building 4507 to produce the products shown in Table 2.2. Subsequently, a report was published on the redetermination of the ^{232}U half-life. This was a joint study between Chem Tech and the Analytical Chemistry Division.[21]

Table 2.2. Separation of ^{232}U from irradiated ^{231}Pa

Product	232U (mg)	^{233}U (ppm)	^{235}U (ppm)
U-l	4.29	209	92
u-2	1.51	317	112
u-3	21.6	129	97
U-4	5.49	206	32
u-5	1040	7,300	14

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3. SWORDS TO PLOWSHARES

The competitive economic position of nuclear power compared to other sources of energy will depend in large measure on how the chemical problems in chemical processing are ultimately solved.

Glenn T. Seaborg
Foreword, Progress in Nuclear Energy, 1956

The transition from wartime development of atomic energy to peaceful uses of nuclear energy was welcomed by essentially everyone. The world seemed receptive to the use of seemingly limitless atomic energy to achieve anticipated energy requirements for both developed and developing nations. All that was needed was sufficient scientific knowledge and technical know-how. To foster the transition, the United Nations sponsored a series of international conferences on the peaceful uses of atomic energy in 1955, 1958, 1964, and 1971. Chem Tech staff members played important roles in those conferences. In addition to the conference proceedings,[1–4] four books in the area of process chemistry resulted from the conferences and associated endeavors.[5–8] Frank R. Bruce served as an editor for the first three volumes,[5–7] and Al T. Gresky served as an editor for the fourth volume.s These collective “Geneva papers” encapsulated much of the R&D work in the chemistry and chemical engineering required for nuclear fuel development and reprocessing until the early 1970s. Chem Tech and ORNL became the Mecca for nuclear fuel reprocessing.

The era of the Geneva conferences perhaps represents the halcyon peak of peaceful applications of nuclear energy, the beating of swords into plowshares. Exciting concepts were proposed and seemed within the grasp of society, for example, large agro-industrial complexes; supplying the increasing world population with needed freshwater through nuclear desalting of seawater; low-cost production of electrical power through “burning rocks” (i.e., use of ubiquitous thorium and uranium): international cooperation in nuclear fuel reprocessing (e.g., the Eurochemic plant at Mol, Belgium): use of nuclear explosives for excavation, mining, recovery of gas and oil, and as research tools (U.S. Plowshare Program): the use of radioisotopes in medicine (“the Humane Atom’?; generation of electricity by direct thermoelectric conversion from the decay heat of radioisotopes: and practical applications of radiation in industry.[9]

In Seaborg’s address to the delegates at the 1964 conference he stated, “With continued attention to reactor safety and waste management, I firmly believe that we can achieve the potential benefits of nuclear power and at the same time protect or even improve our general standards of public health and safety. The increasing use of nuclear power may indeed help to lessen atmospheric pollution, a frequent result of the widespread use of fossil fuels.’[9]

3.1 FIRST GENEVA CONFERENCE, 1955

The first United Nations-sponsored International Conference for the Peaceful Uses of Atomic Energy was held in Geneva, August 8-20, 1955. The introductory words to the resulting volume on process chemistry are revealing.

> Chemical processes form an important aspect of nuclear technology. They are involved in a number of the stages involved in the utilization of nuclear energy; for example, in the extraction, purification and preparation of natural uranium and thorium from their ores; in the preparation of the materials used in reactors; and in the production of radioisotopes and in waste disposal.10

Apropos to the conference theme of peaceful uses of atomic energy, Glenn T. Seaborg, a conference facilitator, stated, "The competitive economic position of nuclear power compared to other sources of energy will, in many parts of the world, depend in large measure on how the problems in chemical processing are ultimately solved."[11]

The need for chemical engineering and economical fuel reprocessing was also stressed by Sir Harold Hartley, another conference facilitator.

> Although nuclear energy is essentially a physical process and physical data are basic for its utilization which is dependent on the most advanced engineering techniques, two of the major limiting factors in the construction and operation of economical power reactors are chemistry and chemical engineering. Success is so largely dependent on the manufacture of the materials of the pile-fissile elements, moderators, cladding and bonding of materials-to new standards of ultra-high purity to avoid wastage of neutrons. It depends too on economical methods of processing the fissioned elements to recover the fissionable materials and nuclear fuels as quickly as possible to minimize idle capital and to dispose of the radioactive products of the process. This is an even more difficult problem on account of the radioactive risks it involves. [12]

3.1.1 Prophetic Words on Waste

As is now known, one major stumbling block, as perceived by the general public, to acceptance of the peaceful use of atomic energy is environmentally safe nuclear waste disposal. This subject was debated even at the first Geneva conference.

> Few problems encountered in the atomic energy program have captured the interest and aroused speculation of the general public as much as the problem of radioactive waste disposal. This is, perhaps, as it should be, for the safety and welfare of both the present and the future generations may well depend on the establishment of safe and reliable disposal practices. The object, then, of waste processing is to modify the wastes in such a manner as to render them more suitable and less hazardous for ultimate or permanent confinement in some site or sites outside man's immediate environment. This may take the form of processes for separating the most hazardous isotopes from the bulk of the wastes and concentrating them in packaged form suitable for shipment and storage, or it may include processes for converting the bulk wastes to insoluble ceramics in which the radioisotopes have been "fixed" by combination with aluminosilicates. [13]

3.1.2 Chem Tech Exhibits and Papers

Significant contributions to the success of the 1955 Geneva conference were made by Chem Tech technical papers and exhibits. Chem Tech exhibits included a mock-up of a solvent-extraction pilot plant complete with remote cell operation and manipulators.[14] The conference papers collectively present many of the major Chem Tech accomplishments up to that time concerning chemical processing aspects of atomic energy. They provide simple vignettes, if you will, or glimpses into the complex and many faceted early history of the Chemical Technology Division. The Chem Tech papers presented at the conference included the following:

D. 0. Campbell, "'Removal of Fission Products from Stainless Steel," Paper 548.[15]

J. W. Landry, "High Level Sampling Devices for Radiochemical Plants," Paper 549. [16]

G. I. Cathers, "Radiation Damage to Radiochemical Processing Reagents," Paper 743.[17]

D. L. Foster, J. E. Savolainen, and R. G. Wymer "Nuclear Reactor Fuel Dissolution," Paper 547.[18]

F. R. Bruce, 'The Behavior of Fission Products in Solvent Extraction Processes," Paper 719.[19]

F. L. Culler, "Reprocessing of Reactor Fuel and Blanket Materials by Solvent Extraction," Paper 822.[20]

J. R. Flanary, "A Solvent Extraction Process for the Separation of Uranium and Plutonium from Fission Products by Tributyl Phosphate,*' Paper 539.[21]

F. L. Culler, "The Processing of Uranium-Aluminum Reactor Fuel Elements,*' Paper 541.[22]

A. T. Gresky, 'The Separation of ^{233}U and Thorium from Fission Products by Solvent Extraction," Paper 540.[23]

D. E. Ferguson, 'The Processing of Aqueous Homogeneous Reactor Fuel," Paper 55 1.[24]

Most of the Chem Tech papers (with the exception of Papers 548 and 549) were quoted in the first volume of the process chemistry series.[5] Summaries of the Campbell and Landry papers (548 and 549) and annotated abstracts of the quoted papers follow.

Removal** of **Fission Products** from Stainless **Steel. Decontamination of equipment used for handling solutions containing fission products presents a difficult problem. Dave Campbell's paper deals with the decontamination of stainless steels, a commonly used material of construction for radiochemical process equipment. Decontamination of large apparatus, such as that used in chemical plants for processing irradiated materials and reactor fuels, is necessary for modification of the equipment or for direct maintenance. Remote maintenance, an alternative, is generally more expensive and less flexible than direct maintenance. Successful decontamination of stainless-steel type 347 using cyclic treatment with 2 ***M*** nitric acid followed by caustic-tar&ate-peroxide solutions is reported in the paper. Oxalic acid solutions were also effective. Decontamination factors (DFs) of up to 10^4 were achieved.[15]

High-Level Sampling Devices** for **Radiochemical Plants. Sampling of liquids in a radiochemical plant is necessary for process control and for accountability. Analysis of samples is the principal means for following process variables such as density, acidity, radioactivity, contamination, corrosion, viscosity, separation of phases, and presence of suspensoids or insolubles. Successful remote sampling using airlifts and jets and minimum personnel exposure to radiation is described in John Landry's paper.[16]

Radiation Damage to Radiochemical Processing Reagents. George Cathers' paper deals with a vitally important consideration in chemical processing of materials associated with high levels of radioactivity, that is, radiation damage to process reagents. It is obvious that knowledge of such radiation damage is necessary to design and operate chemical processing units. The abstract follows:

> The use of organic reagents in radiochemical processes is limited by the destructive effects of radiation. The loss of capacity of ion exchange resins that have been irradiated is dependent on the type of resin; it has been examined. for two polystyrene resins and also for a sulfonated phenolic resin. Exposure of ethylenediamine tetraacetate to more than 0.1 watt-hr/mL radiation results in decreasing effectiveness of the material as a complexing agent for metallic ions. The radiation of tributyl phosphate produces hydrolytic reaction products that are deleterious in a solvent extraction process. These products lead to the retention of fission products, plutonium and uranium after the usual cycle of a solvent extraction process. The implications of this solvent breakdown in the processing of a thorium breeder blanket, after a decay of one day, is considered, together with the implications for processing the core of such a reactor by ion exchange resins.17

The paper cites contributions of other Chem Tech scientists, including R. E. Blanco, D. E. Ferguson, I. R. Higgins, A. H. Kibbey, R. G. Mansfield, and R. P. Wischow.[17]

Nuclear Reactor Fuel Dissolution. The Foster, Savolainen, and Wymer paper gives a summary of the principal dissolution methods. and equipment used through 1955. The abstract follows.

> The dissolution of heterogeneous nuclear reactor fuel elements is the usual first step in chemical processing to recover fissionable and fertile materials. A change of state is thus brought about from solid fuel to liquid feed solution. In this paper, chemical and engineering data are presented for dissolution systems for representative nuclear reactor fuel elements. The materials considered are. uranium metal, uranium-aluminum alloy, aluminum jackets, thorium metal, and zirconium-clad uranium-zirconium alloy. Batch pot dissolution techniques may be most satisfactory for many applications. However, continuous dissolution methods with intermittent charging of metal, continuous additidn of dissolvent, and continuous removal of product are of interest.
>
> The dissolution chemistry of nuclear fuel elements is concerned with reaction rates for various fuel materials in various dissolvent systems. Nitric acid dissolution has been used for uranium, uranium-aluminum alloy and thorium. Sodium hydroxide could be consider& for dissolution of aluminum jackets and uranium-aluminum alloy. Fuel elements containing zirconium can be dissolved in hydrofluoric acid.

> The equipment used in dissolution includes three major units: (1) the fuel charger, (2) the vessel, and (3) the off-gas system. The chargers could be shielded casks capable of controlled discharge of fuel elements, or a remote crane-type unit to charge the fuel elements from buckets. Conveyor-type chargers may be used with both batch and continuous dissolvers. Dissolver vessels are of three general types-pot, column, and slab. The choice depends on the critical mass of the fuel being processed and the shape of the fuel element. Pot-type dissolvers are operated either batchwise or continuously: column and slab dissolvers are continuous-type units. The off-gas system is concerned primarily with disposal of the gaseous radioactive fission products and removal or recovery of the chemical reaction products.[18]

The Behavior of Fission Products in Solvent Extraction Processes. Frank Bruce's paper states that much information exists on the behavior of fission products in solvent extraction processes for fissile and fertile materials. Such processes generally employ methylisobutyl ketone or TBP as solvents, although many solvents have been investigated (e.g., pentaether, diisopropyl ether, tertiary alcohols, dibutyl cellosolve, theonyl trifluoroacetone, dibutyl carbitol). Among the fission products, cerium, zirconium, niobium, ruthenium, and iodine tend to be extracted along with and are most difficult to separate from uranium, plutonium, and thorium.19 The abstract follows:

> The variables which influence the extraction of cerium, zirconium-niobium, iodine, and ruthenium into methylisobutyl ketone and tributyl phosphate are considered. In the case of methylisobutyl ketone extraction the important variables are: solvent purity, salting agent and nitric acid concentration, and temperature. In addition to the preceding variables, uranium saturation of the solvent and tributyl phosphate concentration influence the extraction of fission products into tributyl phosphate.19

Reprocessing* of *Reactor Fuel and Blanket Materials by Solvent Extraction. Floyd Culler states that the single most important reason for chemical reprocessing of nuclear reactor fuel is to recover the fissionable materials produced by neutron capture. Solvent extraction procedures were developed to separate plutonium from natural uranium: enriched uranium from aluminum and other reactor fuel element/cladding materials: and to separate ^{233}U, protactinium, and thorium. The most important features of major solventextraction processes were presented. The abstract follows:

> This paper is a survey of liquid-liquid solvent extraction as used for the processing of irradiated reactor fuels and fertile material. Descriptions are given of the fuels for which solvent extraction processes have been developed: the times required for decay of radioactive contaminants before processing; required decontamination factors: and fission product decontamination. Solvent extraction systemsare summarized and chemical processing flow sheets presented. A brief description is given of the requirements for radiochemical solvent extraction plants and the lines upon which equipment and plant may be designed?

The paper cites contributions of other Chem Tech scientists, including E. D. Arnold, J. R. Flanary, W. B. Lanham, A. T. Gresky, D. C. Overholt, A. C. Jealous, H. E. Goeller, W. G. Stockdale, R. W. Stoughton, S. W. Peterson. F. R. Bruce, D. 0. Campbell, H. K. Jackson, and D. G. Reid.[20]

A Solvent Extraction Process* for *the Separation* of *Uranium and Plutonium from Fission Products by Tributyl Phosphate. Jim Flanary states that after irradiation, nuclear fuel rods contain a nearly equivalent weight of plutonium and mixed fission products that collectively represent only a small fraction of the uranium present. The abstract follows:

> A continuous solvent-extraction process has been developed which uses tri-n-butyl-phosphate (TBP) as the solvent and nitric acid as the salting agent for the isolation of uranium, plutonium, and fission products from irradiated metallic uranium reactor fuel. Tributyl phosphate is less volatile and has a higher flash point than methylisobutyl ketone used in earlier processes. Nitric acid can be distilled off and reused in the process; this yields a lower waste volume than when aluminum nitrate is used as the salting agent[21]

The Processing* of *Uranium-Aluminum Reactor Fuel Elements. In this paper, Floyd Culler succinctly summarizes chemical processing for irradiated enriched uranium fuels. Enriched uranium fuel elements are generally stored underwater before chemical processing to allow for decay of fission product activity. The period of decay is usually determined by the quantity of ^{237}U produced by neutron capture from ^{235}U. The ^{237}U decays with a 6.75-d half-life to ^{237}Np. Because the ^{237}U will be present in the product uranium, it must be allowed to decay before uranium of sufficiently low background activity can be produced. The uranium product must also be sufficiently decontaminated from fission products to allow direct handling during refabrication of fuel elements. This requires gross decontamination factors on the order of 10^8, hence cooling periods of 100–140 d. Because of economic value, loses of enriched uranium should not exceed 0.1%. Because of the small quantity of plutonium produced from irradiation of highly enriched ^{235}U fuel, the plutonium is allowed to follow the fission-product waste streams in the chemical processes developed for such fuels. Also, contamination of the uranium product by such nonactive impurities as aluminum, iron, and sodium must not exceed several thousand parts per million.22 The abstract follows:

> The recovery and decontamination of enriched uranium fuel elements of the type used in the Material Testing Reactor, in the ORNL Swimming Pool Reactor, or from any reactor which uses fuel elements of aluminum-clad uranium-aluminum alloy can be accomplished by organic solvent extraction from nitric acid solution. The unconsumed enrich@ ^{235}U must be chemically separated from fission products, inert fuel diluents and impurities, and heavy elements resulting from neutron capture by fertile materials present in the fuel mixture and by parasitic capture of neutrons by the fuel itself. This chemical purification has been accomplished by the use of selective organic solvents such as methylisobutyl ketone (hexone) and tributyl phosphate (TBP) dissolved in an inert organic diluent such as aromatic-free kerosene. A chemical description of these two processes follows. Each process will decontaminate enriched uranium from fission products to background activity level and will separate plutonium by factors of 10^4 to 10^6.[22]

The Separation of ^{233}U and Thorium from Fission Products by Solvent Extract&n. Al Gresky's paper describes the development and technological aspects of a solvent-extraction process for the chemical recovery and radioactive decontamination of ^{232}Th, ^{233}U, and ^{233}Pa from neutron-irradiated thorium. Althpugh designed primarily for processing aluminum-clad thorium metal slugs, it may be modified for use with other reactor materials, such as thorium oxide or oxycarbonate. The process uses nitric acid as the thorium dissolution agent, tri-n-butyl phosphate (TBP) as the extractant, and aluminum nitrate and nitric acid as the aqueous salting agents. The ^{233}U is finally isolated by ion exchange.[23]

The paper cites contributions of other Chem Tech scientists, including E. D. Arnold, M. R. Bennett, W. T. McDuffee, J. E. Savolainen, R. P. Wischow, F. L. Steahly, and D. C. Overholt.[23]

The Processinq of Aqueous Homogeneous Reactor Fuel. Don Ferguson concludes that the successful operation of the ORNL experimental homogeneous reactor demonstrated the basic feasibility of aqueous homogeneous reactors. The paper's abstract follows:

> Two-region aqueous homogeneous reactors are of considerable interest for the production of economic power. A potential advantage of this type of reactor is simple, economical chemical processing. Fission products and corrosion products may be removed from the fuel solution by ion exchange or by taking advantage of the low solubility of many fission and corrosion products in the reactor fuel. Possible methods of thorium blanket processing include ion exchange, solvent extraction and precipitation. Schematic flow sheets for homogeneous reactor fuel and blanket processing are presented and the economics of the various approaches discussed.24

The paper cites contributions of other Chem Tech scientists, including R. A. McNees, I. R. Higgins, M. E. Whatley, A. T. Gresky, W. E. Tomlin, and F. R Bruce."

3.2 THE INTERIM

The first volume5 in the process chemistry series relied largely upon the information presented at the Geneva conference of 1955. The second volume,[6] published in 1958, reported the progress after 1955 and also widened the scope of subjects covered. The period covered was the interim, as it were, between the first and second Geneva conferences. Much new information was included in volume 2, for example, chapters on recovery processes associated with treatment of ores and feed materials for reactors, fuel cycle costs, and promising alternatives to solvent extraction.[25]

The following Chem Tech scientists and engineers prepared chapters for ***Process Chemistry,*** Volume 2:

K. B. Brown and C. F. Coleman, "Solvent Extraction in Ore Processing"[26]

0. C. Dean, "Reduction of Thorium Chloride by Alkali Metal Amalgams"[27]

J. W. Ullmann, "Factors Affecting Fuel Cycle Cost"[28]

R. E. Blanco, "'Preparation of Power Reactor Fuels for Processing by Solvent Extraction"[29]

E. M. Shank, "Operation of the Thorex Pilot Plant With Highly Irradiated Thorium"[30]

F. R Bruce, 'The Concentration and Purification of Uranium and Plutonium by Ion Exchange"[31]

0. C. Dean, "Mercury Processing of Uranium and Its Alloys"[32]

J. R. Flanary and G. W. Parker, "The Development of Recovery Processes for Neptunium-237"[33]

Annotated abstracts of the Chem Tech contributions to Volume 2 follow. Collectively, they represent significant chemical processing history.

Solvent Extraction in Ore Processing. The very intense search for uranium production with little consideration given to cost gave way to a concerted effort to build an efficient and strong industry based on efficient processing plants close to ore sources and matching steady production rates with long-term consumption rates.[25]

The paper of Chem Tech staff members Keith Brown and Charles Coleman summarizes significant breakthroughs in the recovery of uranium and thorium from raw materials. Nearly all uranium ores are processed by hydrometallurgical methods. The two principal methods for dissolution of the uranium from the ores involve leaching with sodium carbonate solution or with sulfuric acid. Customary solvent extraction procedures using ethers, ketones, and esters gave the best performance on aqueous feeds highly salted with nitrate and were not directly applicable to ore-leach liquors.[26]

Brown and co-workers evaluated hundreds organic compounds for ore processing purposes and found the most suitable characteristics in certain classes of alkylamines and organophosphorus compounds. This finding represented a significant technology breakthrough. The abstract of the paper follows:

> The extraction and stripping properties and other essential chamcteristics of several uranium extraction reagents are presented, with special attention to the long chain alkylamines and alkylphosphoric and dialkylphosphoric acids. Uranium recovery processes with these reagents are described, some of which are in commercial use and others imminent, and other proposed and potential processes are noted. Recovery of the associated metals thorium and vanadium is also considered.26

The paper cites contributions of the following Chem Tech scientists: K. A. Allen, K. B. Brown, C. F. Coleman, C. A. Blake, D. J. Crouse, A. D. Kelmers, D. J. Denis, J. G. Moore, A. D. Ryon, W. D. Arnold, R. S. Lowrie, B. B. Klima, H. M. McLeod, R. R. Wiethaup, and V. L. Saine.[26]

Reduction of Thorium Chloride by Alkali Metal Amalgams. Breeder reactors that use thorium metal blankets require metals of high purity and density. This paper by Chem Tech scientist 0. C. Dean discusses one reduction. process for preparing highquality thorium metal The abstract follows:

> A new semi-continuous, low-temperature process for producing thorium metal is described. Thorium tetrachloride is reduced with sodium amalgam to thorium metal which forms a quasi amalgam. Unreacted sodium and thorium tetrachloride are removed from the amalgam by washing with hydrochloric acid. The thorium is concentrated in the amalgam by filtration, mercury is removed in a vacuum retort and, finally, the thorium is arc melted. Laboratory studies indicate that the process will produce reactor-grade metal at a cost substantially less than the conventional process employing calcium reduction of thorium tetmfluoride.27

This paper also cites the work of Chem Tech engineer John Chandler.

Factors Affecting Fuel Cycle Cost. In this paper by Chem Tech engineer John Ullmann, the components of overall fuel cost for nuclear reactors are analyzed and the effects of several parameters discussed. The factors include reactor variables (e.g., burnup, specific power, thermal efficiency, load factor, anddegree of enrichment), elements of net fuel cost (e.g., fabrication, inventory, burnup, reprocessing, transportation, new fissile material, by-products, and net fuel cost), fuel cycle type (e.g., decontamination level, throw away concept), choice of fuel element (e.g., fuel type, fabrication, diluent, cladding material, bonding material), and reprocessing variables (e.g., plant size, site, amortization, operating efficiency, waste disposal, transmutation). The paper cites contributions of the following Chem Tech scientists and engineers: E. D. Arnold, F. L. Culler, J. 0. Blomeke, W. G. Stockdale, C. E. Guthrie, J. W. Ullmann, and H. R. Zeitlin.[28]

Preparation of Power Reactor Fuels for Processing by Solvent Extraction. All current radiochemical solvent-extraction reprocessing systems use nitrate solutions; therefore, preparation of fuels for solvent extraction implies their conversion to nitric acid solution. Many reactor fuels have aluminum or magnesium jackets that are easily removed mechanically or by caustic solutions before solution of the uranium core in nitric acid. Newer fuel types designed for high-temperature operation use nitric acid-insoluble cladding materials (e.g., zirconium, Zircaloy-2, niobium, or stainless steel). The fuel cores of these types may contain alloys of uranium with molybdenum, niobium, zirconium and silicon, uranium oxides, or mixtures of uranium oxide with stainless steel or thorium oxide. Head-end methods for processing these newer fuels are discussed in this paper by Ray Blanco. The abstract of the paper follows:

> Power reactor reprocessing problems are discussed from the viewpoint of converting heterogeneous fuels to a nitric acid solution suitable for purification by solvent extraction. Current and proposed fuels are classified into types based on reprocessing principles. Flow sheets are presented for the solution of aluminum-clad uranium or thorium fuels in nitric acid along with procedures under development for processing zirconium and stainless steel clad fuels. Methods under development include mechanical processing: solution in sulfuric, hydrofluoric or dilute hydrochloric-nitric acids: hydrochlorination; electrolytic solution: oxidation: and carburization.29

Contributions of the following Chem Tech staff members are noted: F. L. Culler, R. E. Blanco, H. E. Goeller, C. D. Watson, J. R. Flanary, A. T. Gresky, F. G. Kitts, J. J. Perona, E. 0. Nurmi, J. C. Bresee, D. L. Foster, J. E. Savolainen, and R. G. Wymer.[29]

Operation of the Thorex Pilot Plant with Highly irradiated Thorium. The abstract of the paper by Chem Tech engineer E. M. Shank follows:

> The Thorex solvent-extraction flow sheet was converted from a one-cycle to a multi-cycle system to increase the product decontamination when processing highly irradiated thorium. The new “co-decontamination” flow sheet provides two solvent-extraction cycles for thorium and three such cycles plus an ion-exchange step for uranium.
>
> The Thorex Pilot Plant is mostly installed in concrete-shielded cells. It is directly maintained and remotely operated. Process equipment is unit-shielded, provisions for equipment decontamination are built in, radiochemical sampling equipment is remotely operated, and concatenated pulsed columns are used for the extraction.
>
> The plant has been in operation for 3 years, including 17 months with the co-decontamination flow sheet. The thorium and uranium products from thorium irradiated to 4000 g of ^{233}U per ton and decayed 400 days met radioactivity specifications. Products from material decayed 30 days contained somewhat more fission products than specifications for direct refabrication to metal fuel permit. Ionic contaminants in all products were sufficiently low for satisfactory subsequent processing. Solvent-extraction losses were higher than desired. Equipment performance was generally satisfactory.30

Contributions of the following Chem Tech staff members are acknowledged: W. T. McDuffee, J. R. Parrott, R. H. Vaughan, R. E. Blanco, F. R. Bruce, H. G. Duggan, J. W. Landry, E. J. Frederick, A. T. Gresky, R. H. Rainey, G. S. Sadowski, J. W. Ullmann, E. M. Shank, and W. E. Unger.[30]

The Concentration and Purification of Uranium and Plutonium by ion Exchange. Chem Tech scientist Frank Bruce states that chemical processes for the separation of fissionable material from fission products usually produce a dilute solution of the product in nitric acid that must be concentrated and further purified. Accomplishment of the concentration and purification may involve processes such as evaporation, solvent extraction, precipitation, or ion exchange. This paper discusses the use of ion exchange. The abstract follows:

> Cation exchange may be used to concentrate the dilute ^{233}U or plutonium products which are obtained from solvent-extraction processes. Losses of fissionable material are less than 0.01%, and some separation of the products from fission and corrosion products is. achieved.31

Contributions of the following Chem Tech staff members are noted: J. R. Flanary, A. T. Gresky, I R. Higgins, J. T. Roberts, S. H. Jury, R. E. Leuze, V. C. A. Vaughen, W. T. McDuffee, W. L. Albrecht, B. R. Olander, D. C. Overholt, F. E. Tober, and D. C. Orth.[31]

Mercury Processing of Uranium and Its Alloys. Chem Tech scientist 0. C. Dean states that many refractory materials of interest in the construction of reactors (e.g., uranium, thorium, titanium, zirconium, niobium, rare earth metals) are reduced with difficulty, as well as with considerable expense. The loss of half the metal as scrap in the fabrication of fuel or structural elements is common. Much of the scrap is below specification and must be recycled to the refinery for purification. The method of refining metals discussed in this paper appears to be cost-effective. The abstract follows:

> A process for the removal of impurities from natural and irradiated uranium by solution in and recovery from mercury is described. Solution rates and solubilities of uranium in mercury and amalgams were determined. The extent of purification of natural uranium and decontamination factors for the higher-yield fission products were evaluated.32

Contributions of the following Chem Tech staff members are acknowledged: R. E. Blanco, B. H. Morrison, E. Sturch, and R. G. Wymer.[32]

The Development of Recovery Processes for Neptunium-237. Chem Tech staff members Jim Flanary and George Parker review the efforts to recover neptunium from several processes. In the first intermediate-scale effort at ORNL, 40 g neptunium was recovered from a waste stream in the TBP Metal Recovery system processing irradiated natural uranium. Neptunium was also recovered from enriched uranium fuels and from residue (ash) resulting from the fluorination of uranium to produce uranium hexafluoride. The abstract of the paper follows:

> Gram quantities of neptunium-237 were recovered from irradiated natural uranium, irradiated enriched uranium, and fluorination ash from depleted uranium by extraction with tributyl phosphate. The flow sheet from natural uranium is a modified Purex flow sheet, with simultaneous extraction of plutonium, uranium and neptunium(VI) and partitioning by acidity adjustment. Final concentration and purification are by ion exchange. For enriched uranium the Neptex process, which is similar to the 25-TBP process, is used. There is no plutonium present and neptunium is extracted in the tetravalent state. The process used with fluorination ash is similar to the Neptex second cycle.33

Other Chem Tech contributors cited in the paper are as follows: F. L. Culler, H. K. Jackson, G. S. Sadowski, W. H. Lewis, W. A. Brooksbank, P. M. Lantz, and W. J. Martin."

3.3 SECOND GENEVA CONFERENCE, 1958

As in the first conference, significant contributions to the success of the 1958 Geneva conference were made by Chem Tech technical exhibits and papers. Chem Tech exhibits included a motion picture on fuel reprocessing. The film opened with a view of the Hanford crane and canyon. The film also included Chem Tech Metal Recovery Plant operations during plutonium runs in which operators would dress out, go into the cells, disconnect plutonium product bottles, and move the bottles containing plutonium from the cells. This was probably the only motion picture made of the operation.14 Of the 2135 papers submitted to the second Geneva conference held in 1958, approximately 200 were papers concerning chemical processing aspects of atomic energy.[34] Only a few papers were approved in ***Process Chemistry, Volume*** 3. The book contained only three Chem Tech contributions, the first dealing with production of UF_4, the second dealing with the recovery of UF_6 from irradiated reactor fuel, and the third a review paper on process chemistry.[7]

F. R. Bruce, "'Process Chemistry at the Second International Conference on the Peaceful Uses of Atomic Energy," Review Paper.[34]

I. R. Higgins, W. J. Neill, and L. E. McNeese, 'The Excer Process-An Aqueous Method for Production of Pure Uranium Tetrafluoride from Crude Uranium Sources," Paper 506.[35]

G. I. Cathers et al., "'Recovery of Uranium from Highly Irradiated Reactor Fuel by a Fused Salt-Fluoride Volatility Process,** Paper 535.[36]

Although cited by Bruce,[34] the following Chem Tech papers were not quoted in Process *Chemistry, Volume 3*[7] but were published in the Proceedings?

C. A. Blake, C. F. Baes, K.B. Brown, C. F. Coleman, and J. C. White, "Solvent Extraction of Uranium and Other Metals by Acidic and Neutral Organophosphorus Compounds,** Paper 1550.[37]

C. F. Coleman, K. B. Brown, J. G. Moore, and K. A. Allen, "Amine Salts as Solvent Extraction Reagents for Uranium and Other Metals," Paper 510.[38]

K. B. Brown, C. F. Coleman, D. J. Crouse, C. A. Blake, and A. D. Ryon, "Solvent Extraction Processing of Uranium and Thorium Ores," Paper 509.39

F. L. Culler and R. E. Blanco, "Dissolution and Feed Preparation for Aqueous Radiochemical Separation Processes," Paper 1930.[40]

F. R. Bruce, E. M. Shank, R. E. Brooksbank, J. R. Parrott, and G. S. Sadowski, "Operating Experience with Two Radiochemical Processing Pilot Plants," Paper 536.[41]

E. G. Struxness and J. 0. Blomeke, "Multipurpose Processing and Ultimate Disposal of Radioactive Wastes,'* Paper 1073.[42]

The Chem Tech papers, summarized below, discuss selected major Chem Tech contributions to the atomic energy programs through 1958.

Process Chemistry at the Second International Conference on the Peaceful Uses of Atomic Energy. At the time of the 1958 conference, amines and dialkyl phosphates had gained wide acceptance as solvents for extraction of uranium. from ore leach liquors. Their use had resulted in significant savings in both investment capital and operating cost. Solvent extraction recovery of spent reactor fuel using TBP was firmly established. Outstanding advances had been made in decreasing process waste volumes and product losses through use of waste recycle and improvement of process efficiency.[34]

Chem Tech scientist and reviewer Frank Bruce very perceptively concluded

> Although the major research and development effort in waste disposal has centered on the handling of high activity level wastes, it becomes increasingly apparent that low and intermediate level wastes constitute an equally serious economic problem, particularly in European countries where the current United States practice of low level waste disposal to the environment is not acceptable. Consequently, the major recent advances in low level waste treatment have been made in the European countries.34

Chem Tech staff scientists and engineers cited in this review paper include C. A. Blake, C. F. Baes, K. B. Brown, C. F. Coleman, J. C. White, J. G. Moore, K. A. Allen, D. J. Crouse, A, D. Ryon, I. R. Higgins, W. J. Neill, L. E. McNeese, F. L. Culler, R. E. Blanco, F. R. Bruce, E. M. Shank, R. E. Brooksbank, J. R. Parrott, G. S. Sadowski, G. I. Catbers, W. H. Carr, R. B. Lindauer, R. P. Milford, M. E. Whatley, E. G. Struxness, and J. 0. Blomeke.34

The Excer Process-An Aqueous Method for Production of Pure Uranium Tetrafluoride from Crude Uranium Sources. The Excer process, developed to produce pure UF_4 from crude uranium sources, was studied by Chem Tech through advanced stages of engineering development. In the process, crude uranium (VI) is isolated and partially purified by ion exchange, reduced to uranium (IV), precipitated as hydrated UF_4, and dehydrated to pure UF_4. The Excer process requires fewer process steps than existing production methods for UF_4 and is adaptable to various crude uranium sources including sulfate, nitrate, and chloride systems. The paper is authored by Chem Tech staff members I. R. Higgins, W. J. Neill. and L. E. McNeese and cites work by I. R. Higgins, J. T. Roberts, and J. A. Marinsky.[35]

Recovery of Uranium from Highly Irradiated Reactor Fuel by a Fused Salt-Fluoride Volatility Process. The abstract of this paper, collectively authored by Chem Tech staff members G. I. Cathers, W. H. Carr, R. B. Liidauer, R. P. Milford, and M. E. Whatley, follows:

> Uranium may be dissolved in a fused fluoride bath by hydrofluorination to tetravalent uranium and volatilized from this bath as uranium hexafluoride after fluorination. The hexafluoride may be purified by an absorption-desorption step using sodium fluoride. The operation of this process on a laboratory and pilot plant scale is described.36

The fused salt-fluoride volatility process appeared to hold considerable promise for recovering uranium from heterogeneous reactor fuels containing zirconium and from molten fluoride fuels. The principal advantages included small volumes of fission product waste in solid form, product as UF_6 for convenient diffusion plant feed or reduction to metal or UF_4, and essentially no criticality problems. Disadvantages included high temperatures and equipment corrosion.[36]

The ORNL Fused Salt-Fluoride Volatility Pilot Plant was constructed in Building 3019 to recover enriched uranium from irradiated Aircraft Reactor Experiment (ARE) fuel and to serve as a development facility for studying the processing of molten salt reactor fuels and zirconium-uranium metallic fuel elements. The ORNL process was unique in that it involved use of sodium fluoride for absorption and desorption of the UF_6 product. Processing of 40 g enriched uranium (90% enrichment) in irradiated ARE salt produced 39.4 g product (96.92% yield), with an overall material balance of 98.91%.[36]

This paper cites the contributions of several Chem Tech scientists and engineers, including G. I. Cathers, W. H. Carr, R. B. Lindauer, R. P. Milford, M. E. Whatley, M. R. Bennett, J. E. Bigelow, F. N. Browder, L. M. Ferris, A. E. Goldman, R. W. Horton, R. L. Jolley, R. B. Keely, J. T. Long, S. Mann, F. W. Miles, J. B. Ruch, C. L. Whitmarsh, R. G. Wymer, and R. E. Leuze.[36]

Solvent Extraction of Uranium and Other Metals by Acidic and Neutral Organophosphorus Compounds. Charlie Blake and co-workers systematically explored the use of organophosphorus compounds such as solvent-extraction reagents. The dialkylphosphoric acid extraction @APEX) process was developed during that study. This paper presents a portion of that study dealing with acidic and neutral organophosphorus reagents, The use of organophosphorus extractants is especially important in uranium raw-material processing. The reagents also extract a number of additional elements (e.g., alkali metals, alkaline earth metals, lanthanide elements, titanium, zirconium, and hafnium). This paper references several other Chem Tech scientists and engineers, including D. J. Crouse, A. D. Ryon, and W. J. Ross.[37]

Amine Salts as Solvent Extraction Reagents for Uranium and Other Metals. Since 1952, the extraction properties of a wide range of amines and related organonitrogen compounds were studied at ORNL, at first principally for the recovery of uranium from ore leach liquors and later for wider application to general separations. This paper by Coleman and co-workers is a significant contribution to solvent extraction chemistry. Extraction performance was evaluated with respect to the chemical nature of the amine, molecular size and structure, acidity and basicity, concentration, and diluent. Many similarities with resin adsorption were observed. This paper references several other Chem Tech scientists and engineers, including D. J. Crouse, C. A. Blake, J. 0. Denis, W. D. Arnold, R. S. Lowrie, W. J. McDowell, C. F. Baes, and A. D. Ryon.[38]

Solvent Extraction Processing of Uranium and Thorium Ores. This paper describes the then most representative U. S. uranium-recovery processes, which included the use of long-chain alkyl amines for extraction of uranium from sulfate leach liquors (Amex Process) and the use of organophosphorus acids, especially di(2-ethylhexyl)-phosphoric acid (Dapex Process), also for extraction of uranium from sulfuric acid leach liquors. Process flow sheets were presented and discussed with respect to chemistry, reagent costs, equipment, and the extent of commercial application. This paper references several other Chem Tech scientists and engineers, including K. A. Allen, J. 0. Denis, W. D. Arnold, J. G. Moore, K. 0. Johnsson, B. B. Klima, R. H. Guymon, W. T. Ward, R. R. Wiethaup, C. F. Baes, and A. D. Ryon.[39]

Dissolution and Feed Preparation for Aqueous Radiochemical Separation Processes. Solvent extraction was the proven and most versatile method for complete decontamination of plutonium and uranium fuels from fission products and other contaminants. The development of head-end techniques for converting zirconium- and stainless-steel-bearing fuels and homogeneous reactor fluids to nitric acid solutions suitable for solvent extraction was a major effort in Chem Tech. This Culler and Blanco paper thoroughly reviewed mechanical processes for clad fuel element and fuel assemblies (i.e., disassembly and dejacketing) and laboratory data (chemical dejacketing and dissolution of fuel elements) for the preparation of satisfactory aqueous feeds for solvent extraction processes. Homogeneous reactor fuel and blanket processing is also reviewed. The authors acknowledged the contributions of other Chem Tech staff, including W. D. Burch, W. E. Clark, D. E. Ferguson, L. M. Ferris, J. R. Flanary, T. A. Gens, J. H. Goode, P. A. Haas, F. G. Kitts, R. A. McNees, E. L. Nicholson, J. J. Perona, J. E. Savolainen, W. E. Unger, and C. D. Watson and cited references by D. L. Foster, R. G. Wymer, G. A. West, C. V. Chester, A. H. Kibbey, I. R. Irvine, A. C. Schafer, G. W. Parker, W. D. Bond, A. T. Gresky, and J. L. English.@

Operating Experience with Two Radiochemical Processing Pilot Plants, This paper by Frank Bruce, Earl Shank, Bob Brooksbank, John Parrott, Sr., and George Sadowski presented the 4 and 7 years operating experience with the Metal Recovery and Thorex radiochemical processing pilot plants, respectively. The two facilities incorporated design principles derived from experience with three earlier installations that had been dismantled following completion of their intended use. In the Metal Recovery plant, various waste solutions, scrap, and miscellaneous fuel elements had been processed for the recovery of uranium, plutonium, neptunium, and americium. In the Thorex pilot plant, irradiated thorium was processed for the recovery of thorium and ^{233}U. The three pilot plants that had been dismantled were the 25, Redox, and Purex plants, each of which had been operated some 5 years to obtain process chemical data for the design of larger production facilities. For example, the experience gained from the three dismantled plants was used in the design of the ICPP, and the experience gained from the Metal Recovery plant and also from the ICPP plant was used to design the Thorex pilot plant The Metal Recovery and Thorex plants are discussed in detail. The chief way in which the design of a radiochemical processing plant differs from an ordinary plant is in the provisions that must be made to protect operating personnel from overexposure to radiation. Direct and remote maintenance, equipment reliability, decontamination, process control, and waste control are discussed relative to plant operations. The paper acknowledges contributions of Chem Tech staff, including R E. Leuze, W. H. Lewis, W. T. McDuffee, and R. H. Rainey and cited references by F. L. Culler, A. T. Gresky, and E. D. Arnold.[41]

Multipurpose Processing and Ultimate Disposal of Radioactive Wastes. In 1958 the treatment and disposal of high-level radioactive wastes were mainly in the conceptual and formative stages. There had been little treatment other than neutralization, evaporation, and storage of concentrated salt solutions in tanks. This paper by Ed Struxness of the ORNL Health Physics Division and Chem Tech engineer Tex Blomeke discussed several possibilities for ultimate disposal in geologic repositories (e.g., salt domes and deep wells). Problem areas discussed included chemical compatibility and heat dissipation. The authors acknowledge the contributions of F. L. Culler, W. J. Boegly, R. E. Blanco, W. de Laguna, F. W. Parker, L. Hemphill, I. R. Higgins, A. F. Messing, F. M. Empson, W. J. Lacey, E. E. Eastwood, 0. H. Myers, and M. 0. Sealand.[42]

3.4 THIRD GENEVA CONFERENCE, 1964

The theme of the third conference was Reactors and Nuclear Power, Sigvard Eklund, Director-General of the International Atomic Energy Agency in 1964, stated that the frank exchange of views that occurred at the conference on nuclear reactors and systems “should have beneficial effects on the growth of atomic energy as a source of electricity.”[9]

According to Glenn T. Seaborg, Chairman of the U.S. Atomic Energy Commission in 1964, many of the conference delegates viewed nuclear power as developing in three phases. The first phase had already occurred in the development of economic competitiveness of three types of reactors, namely, graphite moderated and gas cooled; heavy-water moderated and heavy-water cooled; and light-water moderated and light-water cooled. The second phase would consist of the development of advanced converter reactors, including near breeders. The third phase would consist of the development of breeder reactors, both the fast breeder using the plutonium and ^{238}U fuel cycle and the thermal breeder fueled on the thorium and ^{233}U fuel cycle.[9]

34.1 Chem Tech Papers

Two Chem Tech staff members, Floyd Culler and Don Ferguson, presented papers at the 1964 Geneva conference?

F. L. Culler and R. E. **Blanco**, "Advances in Aqueous Processing of Power Reactor Fuels," Paper **249.**[43]

D. E. Ferguson, 0. C. Dean, and D. A. Douglas, 'The Sol-Gel Process for the Remote Preparation and Fabrication of Recycle Fuels," Paper **237.**[44]

The Ferguson paper was quoted in ***Process Chemistry, Volume 4,***[8] along with several detailed review papers also prepared by Chem Tech staff. A listing of the Chem Tech review papers published in Volume **4**[8] follows:

L. M. Ferris, "Head-End Processes for Graphite-Base and Carbide Reactor **Fuels."**[45]

C. F. Coleman, "Amine Extraction in Reprocessing,"*

R. E. **Leuze** and M. H. Lloyd, "Processing Methods for the Recovery of Transplutonium **Elements,"**[47]

Summaries of the papers presented at the third Geneva conference and the detailed review papers follow. The papers discuss selected major Chem Tech contributions to the atomic energy programs through 1964.

Advances in Aqueous Processing of Power Reactor Fuels. This paper by Floyd Culler and Ray **Blanco** presents a thorough review of aqueous reprocessing methods for fuels and for the recovery of valuable radioactive materials which by 1964 had reached **the** stage of either pilot plant or production plant. The methods reviewed provided promise of overall processing capability of great diversity and also relative simplicity. The separation processes used organic solvent extraction and ion exchange and were based primarily on well-established processes using TBP (e.g., Purex, Thorex, TBP-25) for recovery of the fissionable and fertile materials. Inorganic adsorbents, ion-exchange resins, **amines,** and phosphate extractants were also used for special recovery processes (e.g., fission products, transuranium elements). Power reactor fuels generally had metal-clad ceramic core fuels (e.g., Zircaloy-2 or stainless-steel-clad UO_2 or ThO_2-UO_2). A major effort for reprocessing power reactor fuels had been required in the development of feed dissolution and preparation methods. These head end processes included shear-leach or chop-leach followed by dissolution in boiling nitric acid: the **Zirflex** process in which Zircaloy jackets are dissolved in boiling 6 *M* NH_4F-1 *M* NH_4NO_3; the Sulfex process in which stainless steel jackets are dissolved in refluxing 4 *M* H_2SO_4; electrolytic dissolution; the Darex process in which stainless steel jackets are dissolved in aqua **regia** (thus requiring titanium equipment): combustion followed by ash dissolution (used for graphite matrix fuels). The paper summarizes major separations developments: the Purex and Thorex processes: recovery of protactinium, plutonium, and neptunium: tmnsuranium separations: and fission product recovery. Culler and **Blanco** discuss safe plant operation and emphasize criticality control and secondary containment as necessary safety concerns. The paper also reviews fuel cycle costs and economies of plant **scale.**[43]

This paper references publications of many Chem Tech staff members including L. M. Ferris, E. L. Nicholson, R. H. Rainey, J. W. Ullmann, C. D. Watson, B. C. Finney, B. A. Hannaford, G. A. West, A. H. Kibbey, J. R. Flanary, J. H. Goode, M. G. **Baille,** F. G. **Kitts,** W. E. Clark, D. E. Ferguson, J. G. Moore, W. F. **Schaffer,** T. A. Gens, F. G. Baird, M. J. Bradley, R. W. Horton, G. I. **Cathers,** R. L. Jolley, E. C. Moncrief, T. Hikido, M. E. Whatley, W. M. Wood, C. A. Blake, W. Davis, J. M. Schmitt, A. B. Meservey, R. G. Mansfield, A. T. Gresky, J. R. Oliver, J. R. Meriweather, R. E. Brooksbank, W. T. **McDuffee,** C. F. Coleman, F. A. Kappleman, B. S. Weaver, W. D. **Burch,** E. D. Arnold, A. Chatham-Strode, W. E. **Unger,** B. F. Bottenfield, F. L. **Hanon,** R. D. Baybarez, H. B. Kinser, D. E. Homer, D. J. Crouse, K. B. Brown, J. P. Nichols, and C. E. **Guthrie.**[43]

Sol-Gel Technology in the Nuclear Reactor Fuel Cycle. This paper by Don Ferguson acknowledges the writing assistance of fellow Chem Tech staff members K. H. **McCorkle,** P. A. **Haas,** W. D. Bond, A. L. **Lotts,** R. E. Brooksbank, L. M. Ferris, and R. G. Wymer. The paper stresses that economical nuclear power depends in part on developing low-cost methods for producing and recycling reactor fuels. Except for fluid fuels such as aqueous solutions or molten salts, reactor fuels and fertile materials (i.e., for breeding) are metals or ceramics. Efficiencies achieved at high temperatures and high **burnups** (i.e., high percentage fuel consumption before recycle) provide incentives to use ceramic fuels, and fertile materials. The sol-gel process represents a technological breakthrough in the preparation of ceramic fuels and materials. Colloidal solutions (sols) and gels are important intermediatesin the process, hence the name of the method. The paper discusses several methods to precipitate metal hydroxides, remove the anions to cause hydrolysis and sol formation, and thermally treat gel to form the desired ceramic materials. The paper discusses preparation of thoria and urania materials, with particular emphasis on the Kilorod project. The Kilorod Facility was designed, constructed, and operated at ORNL to advance the technology of the sol-gel process from a laboratory to an engineering scale. Approximately 1000 Zircaloy-clad fuel rods (hence the name "Kilorod") containing a mixture of 3% $^{233}UO_2$ and 97% ThO_2 were prepared, and much information was obtained on radiation levels and personnel exposures encountered in fabrication of ^{233}U-bearing fuels, as well as scaleup and engineering information applicable to the thorium fuel cycle.[44]

This review cites the work of many Chem Tech staff members, including D. M. Helton, D. E. Ferguson, C. J. Hardy, C. C. Haws, J. L. Matherne, F. W. Miles, J. E. Van Cleve, E. D. Arnold, W. S. Ernst, A. L. Lotts, J. D. Sease, R. E. Brooksbank, A. R. Irvine, F. W. Davis, F. W. Miles, E. M. Shank, J. J. Varagona, J. P. Nichols, J. P. McBride, R. G. Haire, M. H. Lloyd, E. J. Kosiancic, P. A. Haas, S. D. Clinton, A. T. Kleinsteuber, S. R. Buxton, J. L. Kelly, and 0. C. Dean.[44]

Head-End Processes for Graphite-Base and Carbide Reactor Fuels. Graphite-base nuclear reactor fuel elements are designed primarily for use in HTGRs. The fuels are generally small particles of uranium and thorium carbides or oxides dispersed throughout a graphite matrix as the major structural component. For graphite-base fuels, two processing methods, burn-leach and grind-leach, are concluded to be most advantageous. Other methods briefly discussed include pressurized aqueous combustion, chemical disintegration, electrodisintegmtion, combustion-volatility, and pyrochemical processes. Unirradiated uranium, thorium, and plutonium carbides hydrolyze rapidly in water, forming hydrogen and hydrocarbons. Consequently, fuel containing carbides in massive forms (e.g., pellets, rods) would most likely be jacketed. Thus treatment processes would involve removing or penetrating the protective jackets and reacting the metal carbides with aqueous agents. This review by L. M. Ferris cites, in addition to his own, research conducted by other Chem Tech staff, including A. H. Kibbey, M. J. Bradley, R. E Blanco, G. I. Cathers, E. L. Nicholson, J. T. Roberts, K. S. Warren, J. W. Ullmann, J. R. Flanary, J. H. Goode, H. 0. Witte, V. C. A. Vaughen, C. D. Watson, R. H. Rainey, W. D. Bond, A. L. Uriarte, M. E. Whatley, G. C. Wall, P. A. Haas, J. B. Farrell, R W. Horton, R. P. Milford, T. A. Gens, M. D. Pattengill, G. A. West, and W. F. Schaffer.[45]

Amine Extraction in Reprocessing. ORNL staff member Fletcher Moore (Analytical Chemistry Division) introduced amine extraction to ORNL in 1952, and it quickly entered intensive study in the Raw Materials Group under K. B. Brown. Amine extraction soon developed into one of the major categories of solvent extraction systems. The abstract of this review paper authored by C. F. Coleman follows:

> In the eighteen years since amine extraction was first reported, it has developed into one of the major categories of solvent extraction systems. Its industrial use, particularly in the hydrometallurgical processing of uranium and related metals, has both stimulated and benefitted from the commercial production of an increasing range of high molecular weight amines, especially secondary and tertiary amines. Amine extraction appears to promise special advantages in several aspects of nuclear fuel reprocessing by virtue of high extraction power, selectivity, and reagent stability. This review presents recent developments and current status in amine extraction systems as pertinent to their potential use in nuclear fuel reprocessing, including both studies of the extraction systems per se and their adaptation to particular process uses. Proposed chemical flow sheets are cited for a number of processes in and auxiliary to fuel reprocessing. Literature references are arranged so as to facilitate access to previous reviews and bibliographies and to specialized studies of the amine systems.46

This review paper cites work of several Chem Staff scientists, including C. A. Blake, Jr., K. B. Brown, C. F. Coleman, D. E. Homer, J. G. Moore, D. J. Crouse, F. G. Seely, D. 0. Campbell, F. J. Hurst, F. A. Kappelmann, W. Davis, and J. M. Schmitt.[46]

Processing Methods for the Recovery of Transplutonium Elements. Chem Tech staff members R. E. Leuze and M. H. Lloyd collaborated on this review paper outlining the history of TRU and transplutonium element production and detailing the process chemistry and engineering involved in the separation and isolation of the elements. Chem Tech personnel had a very important role in the TRU studies that have been conducted in the last several decades. The abstract of the review paper published in 1970 follows:

> Within the last few years, several programs have been initiated in the United States to significantly increase the availability of the tmnsplutonium elements. Although many process methods used for isolating small quantities of these elements are still in use, a number of special separations methods have been developed to meet the larger-scale processing requirements. Processes that have actually been used for large-scale separations of the transplutonium elements are described, and with brief descriptions of the process equipment.[47]

The paper discusses the three major tmnsplutonium element programs in the United States, namely, the AEC Transplutonium-Element Production Program, the ORNL Curium Program, and the Savannah River Laboratory pilot program to produce 3 to 4.5 **g** ^{244}**Cm** for the development and demonstration of SNAP **systems.**[47] The two ORNL and Chem Tech programs are discussed below:

- AEC Transplutonium-Element Production Program. In 1959, the AEC Transplutonium-Element Production Program was established. Kilogram quantities of ^{239}**Pu** were fabricated into plutonium-aluminum alloy rods at Hanford. The rods were irradiated at moderate neutron fluxes in Savannah River reactors. The **irradiated** rods were processed at Savannah River to recover ^{242}**Pu and to** concentrate the americium and curium along with rare-earth **fission** products. In each of two campaigns, about 10 kg ^{239}**Pu,** 1 kg ^{242}**Pu,** 300 **g** ^{243}**Am,** and **300 g** ^{244}**Cm** were produced, The ^{242}**Pu** in oxide form was shipped to ORNL for incorporation in **HFIR** targets, and the concentrates of americium, curium, and rare earths in nitric acid solution were sent to ORNL for subsequent isolation of the americium and curium. Two new facilities were constructed at ORNL for this production program: **HFIR** for the irradiation of ^{242}**Pu,** ^{243}**Am,** and ^{244}**Cm** at very high neutron fluxes, and TRU (cost **$8,700,000),** for the fabrication of HFIR targets and the processing of irradiated targets. Both facilities started operation in **1966.**[47]
- ORNL Curium Program. The objectives of this program were to isolate gram amounts of ^{242}**Cm** and ^{244}**Cm** for use in the initial development and testing of SNAP heat sources. Processing was carried out in the Curium Recovery Facility **(CRF)** installed by Chem Tech in the High-Level Hot Cell Facility (Building 4507). The CRF was designed to test TRU processes at high activity levels. Curium-242 was recovered from ^{241}**AmO$_2$-Al cermets** irradiated in the MTR and the Oak Ridge Research **Reactor. Part** of the Savannah River concentrate was processed to recover ^{244}**Cm.** The CRF operations conducted by Chem Tech staff resulted in the successful isolation of about 35 **g** each of ^{243}**Am** and ^{244}**Cm** and 25 **g** of ^{242}**Cm.**[47]

This review cites work of many Chem Tech staff members, including J. E. Bigelow, D. E. Ferguson, A. Chetham-Strode, J. R. **McWherter,** F. L. Culler, W. D. **Burch,** E. D. Arnold, L. J. King, J. L. **Matherne,** B. F. Bottenfield, F. L. Harmon, R **McCarter,** C. A. Haws, F. L. Peishel, 0.0. Yarbro, A. L. Lotts, M. K. Preston, J. D. **Sease,** J. E. Van Cleve, Jr., R D. Baybarz, B. S. Weaver, R. G. **Haire,** C. J. Hardy, S. R Buxton, V. C. A. Vaughen, F. A. Kappelmann, R. E. **Leuze,** M. H. Lloyd, R. H. Rainey, R. E. Brooksbank, **W.** T. **McDuffee,** and H. B. **Kinser.**[47]

3.5 FOURTH GENEVA CONFERENCE, 1971

The Fourth International Conference on the Peaceful Uses of Atomic Energy was convened in Geneva on September **6–16, 1971**, under **the** Presidency of Glenn T. **Seaborg.** Over 4,000 participated in the fourth Geneva conference sponsored by the United Nations and the IAEA. The conference was somewhat broader in scope than the previous three conferences. It again proved to be an appropriate forum for exchange of information on the discoveries, projects, and problems of developed and developing nations. Three Chem Tech papers were included in the proceedings which were published in 15 volumes:

- D. E. Ferguson, C. R. Cooley, E. B. Shelton, and D. S. Webster, "Recovery of Liquid-Metal Fast-Breeder Reactor (MFBR) Fuels: Development of Techniques"[48]
- R. Salmon, J. T. Roberts, A. L. Lotts, T. N. Washburn, and W. II. McVey, "Price Forecasting and Resource Utilization for the Fuel Cycle Industry of the United States"[49]
- F. L. Culler, J. 0. Blomeke, and W. G. Belter, "Current Developments in Long-Term Radioactive Waste Management"[50]

Recovery of Liquid-Metal Fast-Breeder Reactor (LMFBR) Fuels: Development of Techniques. The abstract of the Chem Tech Division Director Don Ferguson follows:

> Nearly every major nuclear-fuel processing facility in existence today employs the Purex process. Its favorable features—unexcelled separation, versatility, ease of scale-up, and vast operating experience—also apply to the treatment of liquid-metal fast-breeder reactor (LMFBR) fuel and make Purex an obvious choice for use in processing this fuel. Certain modifications to the existing techniques are necessary, however, to make the process economical for LMFBR fuels. Because of the higher plutonium contents, a factor of 10 as compared with light-water reactor (LWR) fuel, there is a large economic incentive for processing at shorter decay times. This, coupled with the fact that LMFBR fuels will operate at higher power densities, 150 vs 35 kW/kg for LWR fuel, and to burn-up of the order of 80,000 MWd/t, creates a serious heating problem in handling the fuel. These same factors increase the amounts of volatile fission products, such as iodine, xenon, krypton, and tritium, which must be handled and contained in the plant A comprehensive development program is being carried out in the United States of America to solve these and. other unique problems associated with the processing of LMFBR fuels. Methods are being developed for the deactivation and removal of sodium. Handling techniques are being developed to provide reliable cooling of the spent fuel elements. Two alternative head-end processes are being studied. One uses molten zinc to remove the stainless steel cladding. In the other, the fuel is chopped and the volatile fission products are removed by a new process, called voloxidation, which involves oxidation of 500 C Dissolution and feed preparation for the high-plutonium-content, high bum-up fuel are also being studied. Solvent extraction flow sheets to accommodate the higher plutonium and fission-product concentrations have been developed and are being tested. Both pulsed columns and centrifugal contactors are being evaluated. Methods are being developed and tested for reducing radioactive effluents, especially such volatile fission products as iodine, from the plant to near zero.48

The paper cites several Chem Tech progress reports and Chem Tech engineers V. C. A. Vaughen, J. G. Moore, and D. J. Crouse.[48]

Price Forecasting and Resource Utilization for the Fuel Cycb Industry of the United States. In 1966, the AEC began a long-range systems analysis study to determine the optimal utilization of various types of nuclear reactors in an expanding nuclear power economy. The Fuel Cycle Task Force working group, chaired by Chem Tech Division Director Don Ferguson; was made responsible for the development of a model for the long-range projection of fuel-cycle costs. The FUELCO model was developed for this purpose by Chem Tech engineer Royes Salmon. This paper describing the model and its use in the AEC study was presented at the fourth Geneva conference. The FUELCO model typically showed individual (fuel cycle) plants earning 1 to 25% on equity. Small plants generally earned the lowest rates,. Large plants coexisting with smaller competitors tended to show the highest rates of return.[49]

Current Developments in Long-Term Radioactive Waste Management. The abstract of the paper by ORNL Associate Director Floyd Culler and Chem Tech engineer John "Tex" Blomeke follows:

> The safe disposal of radioactive wastes is possibly the most important and controlling problem in the large-scale use of nuclear energy. Radioactive wastes with very long half-lives require processing to chemically stable form and storage in an isolated natural environment, an environment that remains protected from natural phenomena for periods of several hundreds of thousands of years. The

radioactive fission products and long-lived fissile and fertile isotopes of alpha emitter, such as plutonium, set the requirements for chemical fixation, heat release, shielding, containment, shipment, and ultimate disposal. The main thrust of work in the USA radioactive-waste program over the past 15 years has been directed toward the development of processes for solidifying the wastes and in establishing the suitability of natural salt deposits as an ultimate repository. As a consequence of the advances made in this work, the USAEC has established the industrial requirement for conversion of all high-level liquid wastes to a solid form suitable for interim on-site storage, shipment, and disposal in a few centralized repositories.

The choices for long-term disposal or management are few. Starting in 1950 the problems of ultimate disposal were considered by various committees of the National Academy of Sciences at the request of the USAEC. Disposal in thick-bedded salt in the central part of the United States was recommended for study. A pilot project for high-level disposal was initiated by the Oak Ridge National Laboratory for the USAEC in 1963 and was satisfactorily completed in 1968. The location of this test was at Lyons, Kansas, in a portion of an old salt mine located in an extensive, 1000-ft-deep, 300-ft-thick salt layer. As a result of this demonstration and extensive supporting development, a national repository for radioactive wastes will be constructed at this site. It will serve both as an expanded demonstration project and a disposal site for radioactive wastes originating from the nuclear industry and from USAEC operations. Both plutonium-bearing wastes and fixed, canned high-level fission products will be stored, monitored, and evaluated. The initial project site will have a sufficient capacity to handle fixed high-level fission products and alpha-emitting wastes produced in the United States until near the end of this century.[50]

The paper cites Chem Tech engineers John Blomeke, W. C. McClain, and R. L. Bradshaw as well as an ORNL staff report on citing for fuel reprocessing and waste management facilities.50

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4. EXPANDED AND CHANGING MISSIONS

The work that took place at the Graphite Reactor had ramifications for many fields of science. This was the place, for example, where mammalian radiation biology in its modem sense really originated. The reactor had important implications for the development of nuclear power, both for naval submarines and for electric utilities. It had great influence, from the very beginning, on materials research, particularly the fields of neutron diffraction and radiation damage to solids. And then, of course, it was the first place where isotopes were produced for every science you can think of. . . . There were other missions. The most important, I suppose, was the production and distribution of radioisotopes. Much of the work that led to unraveling the genetic code could not have been done without them. They are used in the diagnosis and treatment of cancer., They are used in agriculture to trace phosphorus in fertilizers, and in innumerable other ways. Science couldn't continue today without them. If God has a golden book and writes down what it is that Oak Ridge National Laboratory did that had the biggest influence on science, I would guess that was the production and distribution of radioisotopes.

Alvin M. Weinberg
The Graphite Reactor, 1992

During the past half century, national energy-related goals and objectives have changed. In response to these changes, the work tasks and missions of ORNL and Chem Tech have been redirected and expanded. Some of the major Chem Tech projects and programs-are described in the following sections.

4.1 SEPARATIONS DEVELOPMENT FOR RAW MATERIALS,

Charles F. Coleman, David J. Crouse, and Fred J. Hurst

In recognition of the urgent need for improvement in domestic uranium production, a Raw Materials Section under the direction of K. B. Brown was established in 1948 within the Y-12 Research Division, which soon became the ORNL Materials Chemistry Division (Fig. 4.1). Initial emphasis was placed on improving the leaching and precipitation methods that were then being used to recover uranium from the sandstone carnotite ores located on the Colorado Plateau. Because the known domestic reserves were

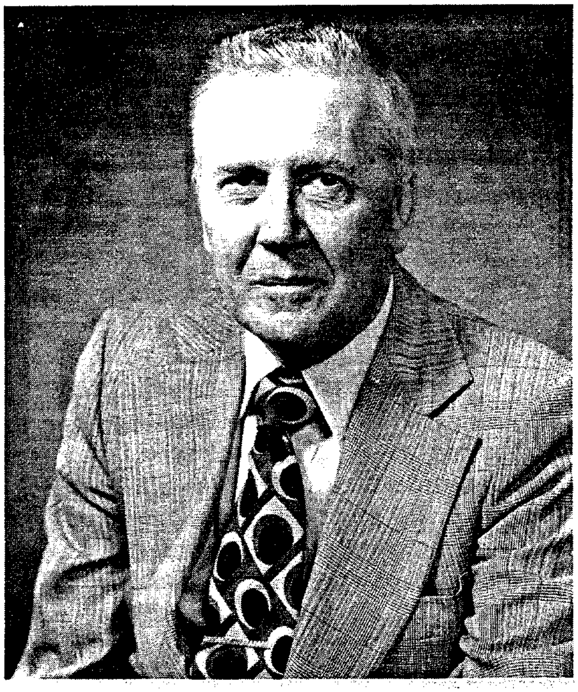

Fig. 4.1 Keith Brown, long-time leader of the separations chemistry group, was Assistant Division Director of Chem Tech in this 1978 photograph.

extremely limited, investigations also were conducted on the recovery of uranium (and in some cases thorium) from a variety of low-grade sources such as phosphates, shales, lignites, granitic rocks, and monazite sands.,

A limiting problem in the development of suitable recovery processes was efficient recovery of uranium from the complex process liquors. A liquid-solid ion-exchange process, already placed into operation by some other organizations, was carefully studied and evaluated. However, conviction grew that liquid-liquid extraction (solvent extraction) offered the most promise if systems could be found that would extract uranium from sulfate solutions since the extensive and growing uranium nitrate extraction systems (Sect. 3.1) could not tolerate sulfate.

A program to identify suitable extractants was initiated, and the development of separation technology soon became the principal effort of the research group. As a result of this effort, two major processes, DAPEX and AMEX, were developed. DAPEX uses a synergistic mixture of organophosphorus compounds, and AMEX uses long-chain alkyl amines to extract the uranium. These processes have been used successfully in many domestic and foreign uranium mills since the mid-1950s. Development of these processes was described at the second Geneva conference in 1958 and earned a Technical Achievement Award from *Mining World* and a Certificate of Merit from the American Nuclear Society.

As the Raw Materials Section grew, it was organized into specialized interacting groups: Exploratory/Descriptive Chemistry, Fundamental Chemistry, Process Development, an engineering group for scaleup studies, and, for a critical period, a synthetic organic chemistry group to synthesize potential extractants that did not yet exist.

In 1956, this separations development group was transferred to the Chemical Technology Division and became known as Chemical Development Section C. During the ensuing years, the raw materials separations technology developed in this section spread into many different areas. The special reagents and processes and the personnel skilled in the uses of those reagents and processes have been effective in many applications superficially remote from raw materials. Some of these became major projects within this section, notably the Chem Tech-Biology Division cooperative project that isolated pure tRNAs (see Sect. 4.19). Others were started and turned over to other users or were pursued jointly, including separation of transplutonium elements from rare earths in chloride or carboxylic acid solutions, recovery of uranium-neptunium-technetium from refinery wastes, recovery of uranium from sulfuric acid-fuel solution and from fluoride-fuel solution, recovery of cesium and strontium from fission-product waste solutions, and recovery of nonradioactive metals such as beryllium, cesium, and vanadium from their ores (see Sects. 2.9-2.12). One very successful program was the identification of granites as an almost unlimited supply of thorium for use in a thermal breeder system that was being studied. A considerable effort was expended on the Plowshare program to, define the behavior of radionuclides in rubble cavities formed for natural gas stimulation, copper oxide leaching, and recovery of oil from shales. A study of many difficult metal separations such as Zr-Hf, Ta-Nb, and Co-Ni was carried out using solvent extraction and ion-exchange techniques. The Co-Ni and Zr-Hf separation techniques were later used in continuous chromatography studies. After many years of frustration, a successful process @EPA-TOPO) was developed to recover uranium from wet-process phosphoric acid (Fig. 4.2). This process is currently being used by several companies in the United States and in several., foreign countries. This development was embroiled in a bitter patent dispute which was won by ORNL and DOE and received significant attention for technology transfer. It was also selected for a prestigious Kirkpatrick Chemical Engineering Honor Award in 1979 and an IR-100 award in 1980 (Figs. 4.3 and 4.4).

Many advances in analytical methods were developed to meet the needs in separations research. In particular, a highly sensitive and versatile alpha spectrometry system (PERALS) received an IR-100 award in 1981 and is gaining increasing commercial acceptance.

While not feasible for enumeration here, it should also be noted that the open-literature publications from wide ranges of fundamental and descriptive-chemistry topics, related to and suggested by the separations studies, have been important contributions (Fig. 4.5).

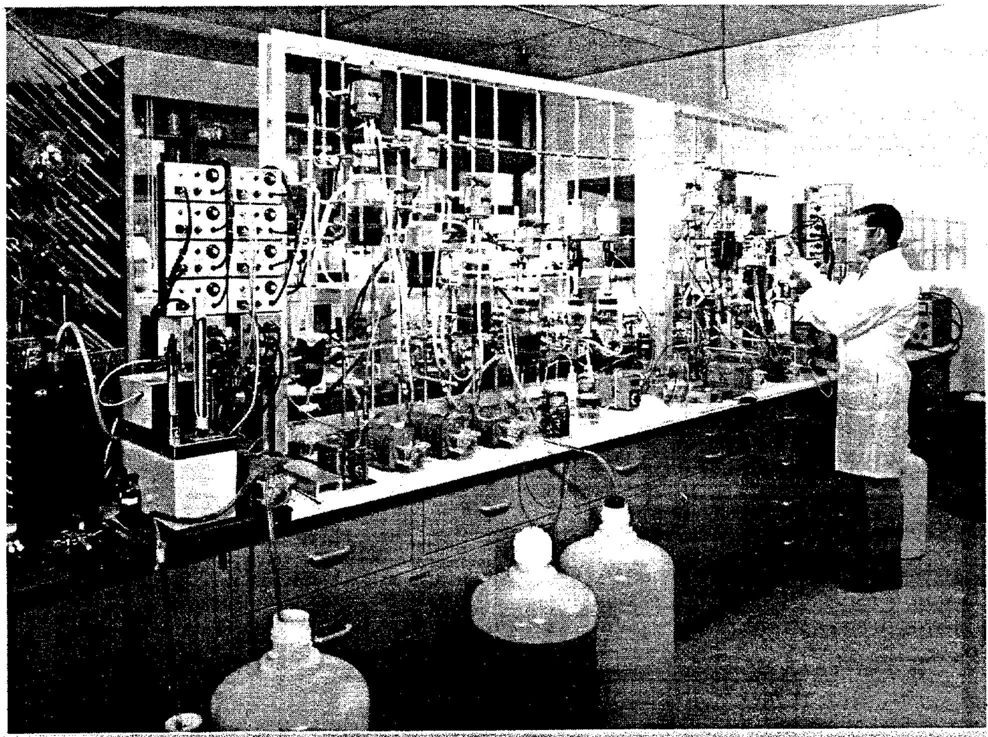

Fig. 4.2. Fred Hurst monitoring the bench-scale process for solvent extraction recovery of uranium from wet-process phosphoric acid.

Fig. 4.3. The Chem Tech R&D team with the 1980 Chemical Engineering Honor Award for developing a Solvent Extraction Process for Recovery of Uranium from Wet-Process Phosphoric Acid. Shown (left to right) are Charles Coleman, Iran Thomas, David Crouse, Bill Howerton, Fred Hurst, Bill Arnold, Charles Baes (Chemistry Division), and Al Ryon.

Fig. 4.4. Fred Hurst (center) receiving the Chemical Engineering Honor Award for developing A Solvent Extraction Process for Recovery of Uranium from Wet-Process Phosphoric Acid from Calvin Cronan, Editor of *Chemical Engineering* (left) and Herman Postma, Director, Oak Ridge National Laboratory (right).

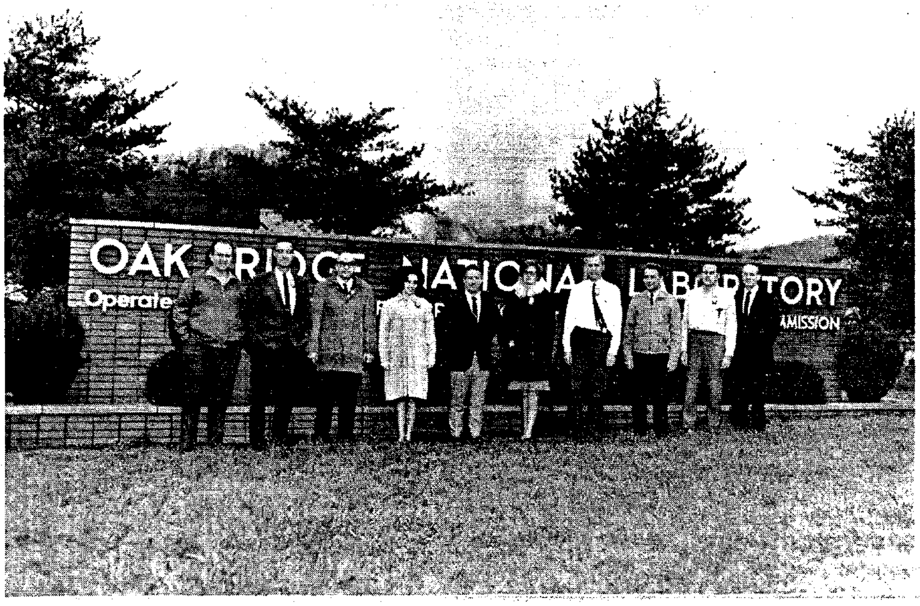

Fig. 4.5. An early 1970s photograph of the Chemical Development Section A staff involved in fundamental studies on solvent extraction processes (left to right): P. B. Orr, Ray Wymer, Rex Leuze, Carolyn Gheen, John Fardy (guest scientist from the Australian Atomic Energy Commission), Bea Garber, Boyd Weaver, John Chilton, Richard Shoun, and Charles Coleman.

4.2 FUEL PROCESSING: THE SOL-GEL PROCESS, Walt Bond and Paul Haas

4.2.1 Early Sol-Gel R&D

In 1959, work was begun on a method for preparing dense fuel particles which came to be called the "sol-gel" process. The early effort was led by Don E. Ferguson (then Section Head, Chemical Development A), who was the first to recognize the advantage of sol-gel processes in regard to their adaptability to fully remote processing methods and to their potential in the manufacture of dense, homogeneous, high-strength ceramics. The initial work by Ferguson, Ken McCorkle, Chuck Schilling, 0. C. Dean, and Todd Kleinsteuber quickly demonstrated the technical feasibility of the sol-gel approach to ceramics manufacture.

4.2.2 Angular Sol-Gel Particles

In the early 1960s, an engineering-scale demonstration project (called Kilorod) was carried out semiremotely in Bldg. 3019 in which 1000 Zircaloy-clad fuel rods were manufactured and tested. The angular fuel particles were vibratorily packed into the tubular cladding. This effort required a considerable team of chemists, chemical engineers, ceramicists, and technicians. At this point, Claude Haws, Bob Brooksbank, and Jim Snider joined the sol-gel team previously mentioned along with John Sease and John Van Cieve from the Metals and Ceramics (M&C) Division.

This early work created considerable interest throughout the nuclear community, both nationally and internationally. The sol-gel process was revealed internationally at a conference in Rome, Italy, on June 13–15, 1961. The exhibit and the ORNL exhibit preparation team are shown in Fig. 4.6.

4.2.3 Spherical Particles

Concurrent with the angular particle development, Paul Haas, Todd Kleinstueber, and Sam Clinton demonstrated that spherical particles could be prepared by simply extracting water from aqueous sols of colloidal oxides. This was then followed (1962– 1973) by the development of processes for preparing spherical particles of ThO_2, UO_2, PuO_2, and their binary oxide combinations

Fig. 4.6. Soigei exhibit with ORNL research participants and exhibit preparation t&m. Front row (left to right): Chem Tech staff members 0. C. Dean, Bob Brooksbank, Jim Snider, Paul Haas, and an unidentified engineer. Back row (left to right): Jim Bresee, Bill Gronier, Sam Clinton, Glenn Williams, unidentified engineer, Tom Gale, Pete Lotts, and an unidentified engineer.

for high-temperature gas-cooled reactor fuels. It was also during this period that work was performed that showed that spherical, dense ceramic oxide powders could be made from nearly all elements that form highly insoluble oxides. This included the important industrial ceramic powders such as alumina, titania, and zirconia and also specialty oxides such as those of the rare earths, americium, and curium. During this period, Ray Wymer, Rex Leuze, Walt Bond, Jim Snider, Milt Lloyd, John McBride, Leon Morse, Al Ryon, Russ Baybarz, Dick Haire, Paul Haas, Fred Kitts, Karl Notz, Albert B. Meservey, Ray Buxton, and Sam Clinton were heavily involved. Examples of good and bad microspheres are shown in Fig. 4.7. Laboratory-scale equipment used for the resin loading process, an alternative microsphere-making process, is shown in Fig. 4.8.

4.2.4 Fuel Particles for Light-Wafer and Breeder Reactors

Work ceased on sol-gel processing in 1972 and was not resumed until 1977, when considerable interest arose in its potential for manufacturing spherical fuel particles for the Light-Water Reactor (LWR) and in the early 1980s for oxide fuels for Advanced Breeder Reactors (ABR). Also, work was initiated during this period on the fixation of nuclear wastes, improving the properties and performance of ceramics in nonnuclear applications, and on improving gel properties for chromatographic separations. At the encouragement of the Exxon Company, improvements were made in the process flow sheets and equipment for LWR-grade UO_2 and ABR-grade UO_2-PuO_2 and subsequently demonstrated on the pilot plant scale (1 kg/h) (Fig. 4.9). Incorporation of nuclear waste in zirconia-based ceramic was found feasible. Key Chem Tech players in these developments were Karl Notz, Roy Norman, Paul Haas, Milt Lloyd, Jack Collins, Vic Fowler, Sam Shell, Claude Haws, John Vavruska, Roger Spence, Al Ryon, Rex Leuze, Emory Collins, Sharon Robinson, Bruce Finney, Bill Arnold, Fletcher Daley, John Begovich, Dave Williams, and Wait Bond.

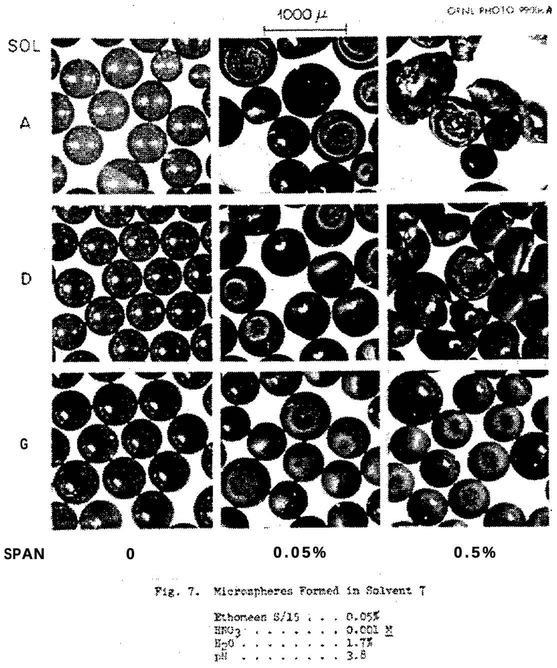

Fig. 4.7. Sol-gel microspheres. it took many tests (and failures) to determine the proper conditions to make good UO_2 sol-gel microspheres. This is a very small sample of test specimens prepared by Karl Nob and Albert B. Meservey.

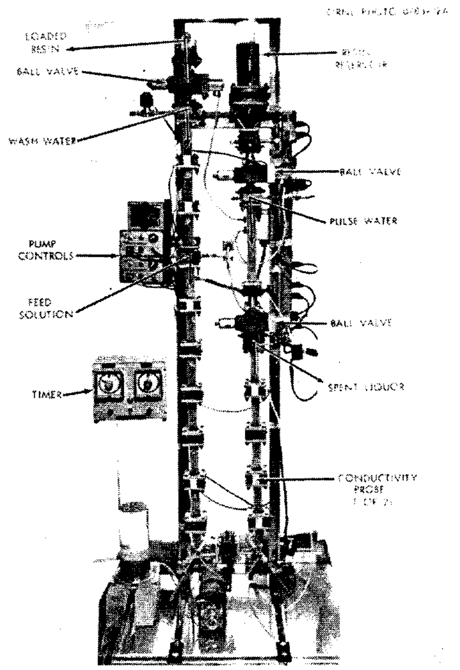

Fig. 4.8. Resin loading process. Another route developed for making UO_2 microspheres is to first load uranium on spherical, weak-acid ion-exchange resin beads. This column, designed and operated by Karl Notz and Charles Greene, accomplishes the loading operation on a pulse-continuous basis. The concept is an adaptation of the Higgins column.

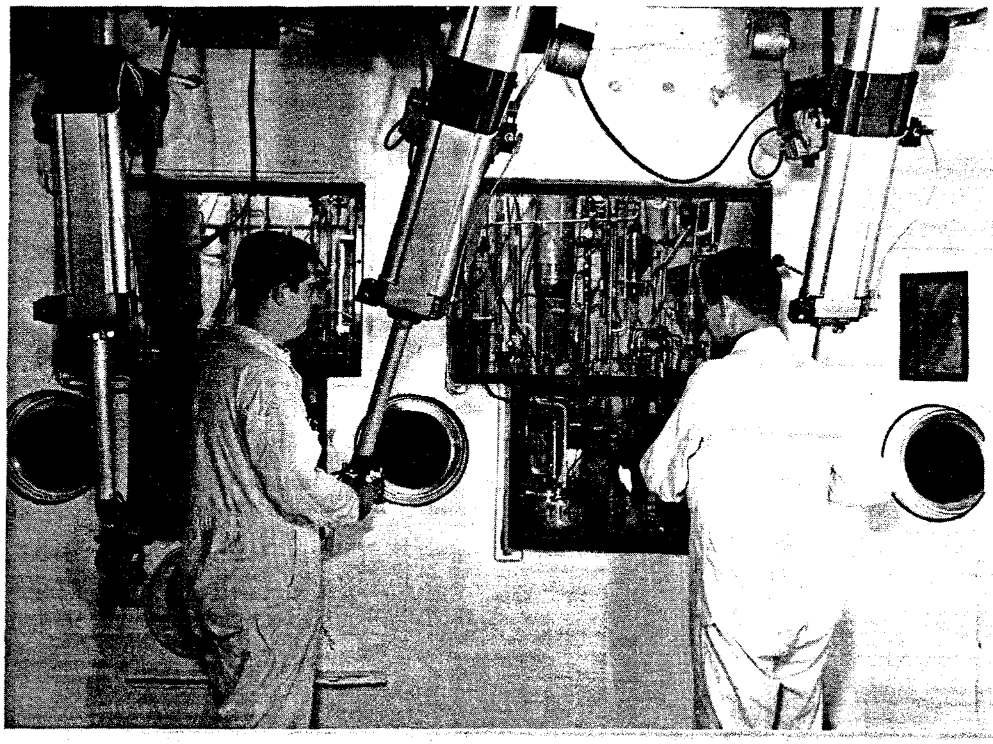

Fig. 4.9. The plutonium sol-gel pilot plant was located in a hot cell requiring that operations be conducted by glove or master-slave manipulators.

The following personnel from the ORNL Metals and Ceramics Division also made major contributions to this program: Ron Beatty, Ron Bradley, Jack Lackey, and Dave Stratton. Figure 4.10 shows Vic Fowler adjusting a prototype of the 1-kg/h sol-gel production unit.

4.2.5 Sol-Gel-Derived Metal Oxides

Sol-gel work on nuclear fuels and waste applications was ceased in 1985. However, work is being continued to the present day on the applications of sol-gel processes to improve the properties of ceramic materials so that their range of applications may be extended. This work showed that tougher ceramics, better electrical surge arrestors, and superior performance can be attained in chromatographic separations using sol-gel-derived metal oxides. Major contributors to this work are Paul Haas, Jack Collins, Walt Bond, and George Davis.

4.3 PLOWSHARE PROGRAM,

Walt Bond and Bob Jolley

In the mid 1950s, the AEC initiated studies on the peaceful applications of nuclear explosives, which came to be known as the Plowshare Program. All types of applications were proposed for the explosives, ranging from using the blast effect to dig canals or crush mineral deposits for easy mining to utilizing the high neutron flux to produce valuable isotopes such as tritium and transuranic elements. The Chemical Technology Division became involved in the studies to develop flow sheets for the recovery of isotopes or metals from the crushed ore deposits. A necessary part of these studies was to obtain samples of the various underground nuclear detonation tests and determine the chemical species that resulted. This knowledge was then used to design chemical flow sheets. John Landry spearheaded the sampling program associated with the underground nuclear detonations. He designed the sampler system that was employed to obtain gas samples a few milliseconds after detonation and also the system for collecting solid debris. John was assisted by Baird Bottenfieid in the early days of this work. Various people were involved in the determination of the “nuclear explosion chemistry” and chemical flow sheets for the recovery of isotopes or metals (such as copper) from ore bodies. Early on, the program focused on isotopes and Walt Bond, Walt Clark. Al Ryon, and Ray Wymer were involved. Floyd Culler maintained a keen personal interest in

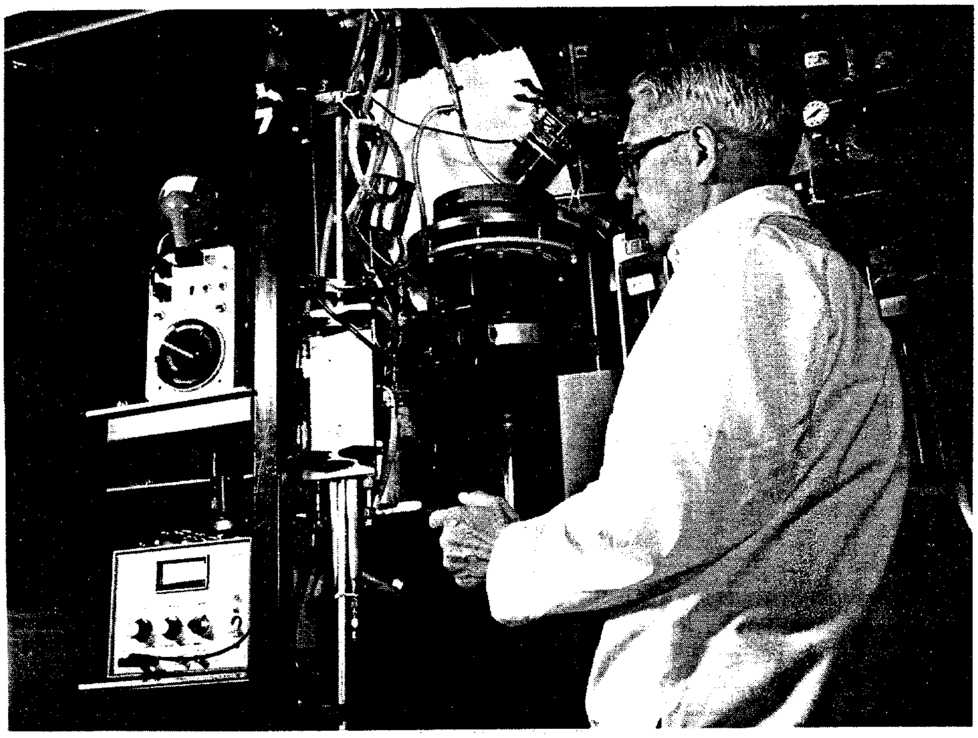

Fig. 4.10. Vic Fowier adjusts the prototype of the l-kg/h production unit during preparation of UO_3 sol-gel spheres.

the program because of (I believe) the challenging chemistry and formidable engineering problems it posed.

The person that I remember most on this program was John Landry. Figure 4.11 shows John Landry in conversation at an annual Chem Tech information meeting. John was not only ORNL's "ambassador" to the Plowshare Program, but also a character in his own right. John's mind is in the genius category, and he was interested in nearly everything going on in the universe and how it worked. This also explains how he sometimes got distracted from the main task at hand. Because of his nature to be distracted, John had a way of suddenly disappearing from the group he was traveling with. However, he always could find the group later (from a few minutes to a few hours). The following personal story illustrates the point.

John, Floyd, and I were attending a Plowshare meeting in San Francisco, and for dinner that evening, we decided to walk down to Fisherman's Wharf for seafood, along with some of the other attendees. Along the way we noticed John was missing (not unusual), so we just went on to the wharf and selected a restaurant. About 20 minutes later, John arrived at our table (we had not yet been served) and ordered his dinner. He then proceeded to describe in great detail how the drive systems for the famous San Francisco cable cars are housed, constructed, and operated. Unbeknown to us, John had noticed the cable house on our route and had talked a security guard into letting him in the cable house to see the chief engineer. The chief engineer gave him a technical tour of the cable house. John was also known for tuning up his motorcycle in his dormitory room in Oak Ridge at 1:00 a.m., which included timing by ear-1 won't go into that!

Participation in the Plowshares Program ended in 1974 with experimental studies on tritium behavior during in situ recovery of oil shale and a paper describing studies of radionuciides (principally tritium as tritiated water) in copper recovery from copper ores fractured by explosives to facilitate in situ leaching.' Examples of the breadth of the Chem Tech involvement in Plowshares follow.

4.3.1 Tamalpais Experiment and Field Sampling Tests

The Tamalpais underground nuclear experiment, Project Coach, Project Gnome, and CANE (for Chemical Application of Nuclear Explosives) were some of the Plowshare projects

Fig. 4.11. John Landry (on right) in conversation with J. P. Blakely and another ORNL staff member at an annual Chem Tech information meeting.

for which Chem Tech staff provided technical support.

Perhaps one of the more interesting accounts of this technical support was supplied by Landry, who designed and helped install instrumentation and sampling devices for the Tamalpais project. The sampler after design was fabricated by ORNL and installed at the AEC Nevada Proving Ground. The sampling device incorporated explosion-operated valves and special features for fast removal of, the samples (Figs. 4.12 and 4.13). The Tamalpais event of October 8, 1958, consisted of the explosion of an atomic device equivalent to 72 tons of TNT at a depth of 330 ft. The purpose was to investigate an isotope-producing nuclear explosion in a salt medium. Samples were to be drawn at time intervals of 0.01, 0.11, 1.1, and 11 s, and also cumulatively. Recovery of the samples and equipment was delayed for over two months. The explosive gas hazard prevented entry to the sampling room in the first two weeks, following detonation. More importantly, the atomic bomb testing schedule had been accelerated to meet the October 31, 1958, ban on nuclear testing, and the effects of subsequent neighboring detonations caused additional delay. Landry stated that he "had hoped to be present for the recovery of the samples and inspection of the equipment but left the proving ground a few hours after seeing one of the succeeding events breach the mesa near the location of the sampling room."[2] University of California Radiation Laboratory personnel recovered the samples in late December. A total of six large samples were collected. One of the 11 -s sample vessels obtained no sample due to failure of the admission valve to fire. One 1. l-s sample was lost due to leakage during the 2-month recovery period.[2]

4.3.2 Prompt Sampler Studies

A hypervelocity jet sampler and a bubble-tapping sampler were conceptualized and tested by John Landry. The hypervelocity jet sampler was designed to recover specimens after they had been irradiated with neutrons about 1 m from an underground-exploded nuclear device. The sampler was designed to recover the irradiated specimen before it could be engulfed in the nuclear explosion and resulting debris.[3] Copper and iron targets were jetted successfully4 and demonstrated that at least 50% of a target was formed into a jet in about 15 μs. The jet traveled at a velocity equal to, or faster than, the estimated velocity of a nuclear shock wave in rock.4 The non-nuclear jet tests

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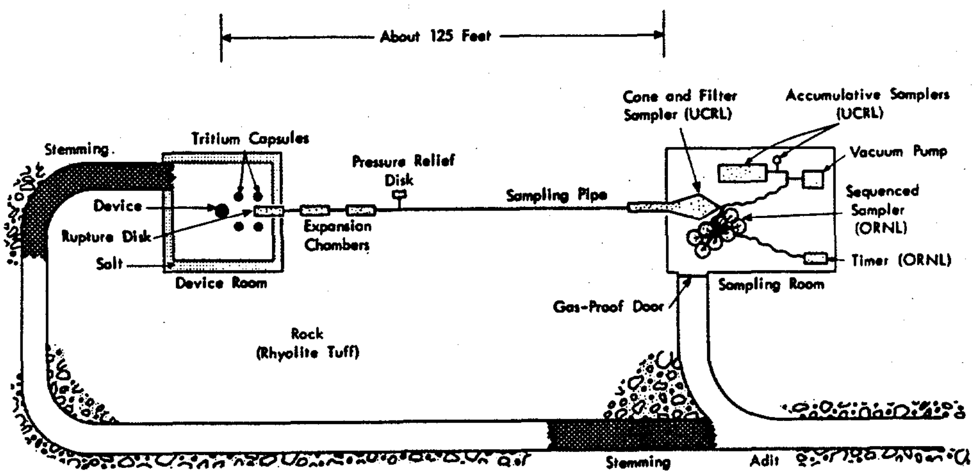


Fig. 4.12. Plan view of Tamalpais sampling arrangement. [Source: J. W. Landty, *Oak Ridge National Laboratory Samples for the Tamalpais Underground Nuclear Detonation Experiment,* ORNL-2881 (June 30, 1960)]

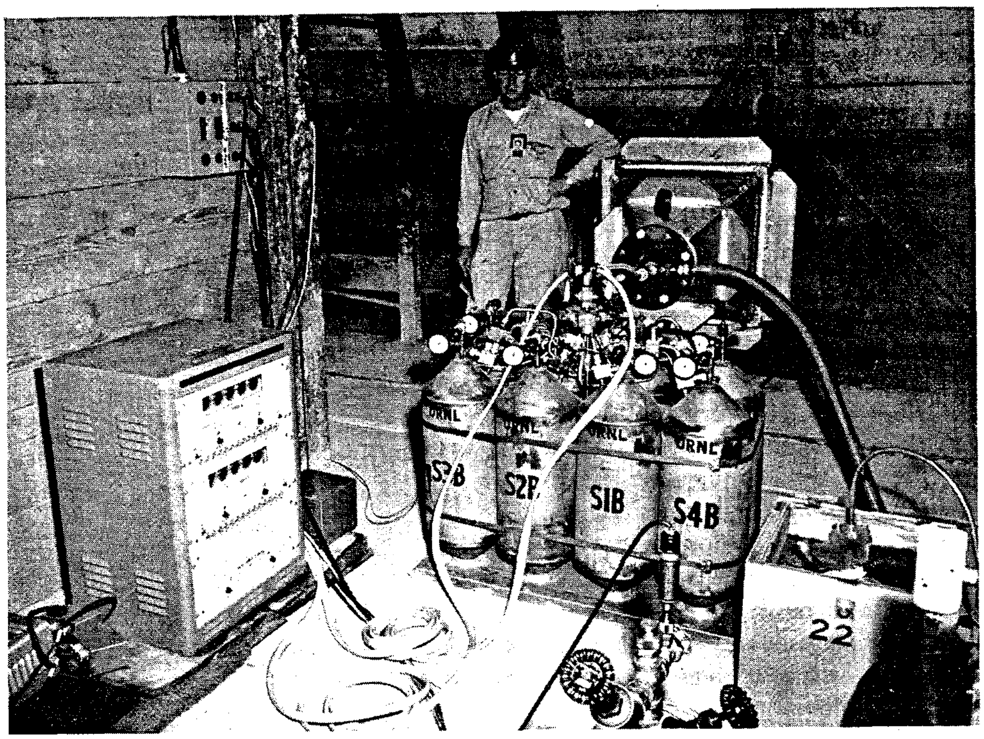


Fig. 4.13. Tamalpais sampling equipment. [Source: J. W. Landry, *Oak Ridge National Laboratory Sampler for the Tamalpais Underground Nuclear Detonation Experiment,* ORNL-2881 (June 30, 1960)].

were conducted at Frankford Arsenal, Vincentown, New Jersey, in an evacuated flight chamber 18 ft in diameter by 55 ft long.5

Also, a bubble-tapping sampler was conceptualized, but not tested, to sample the gaseous and particulate explosion products from an underground nuclear explosion, providing useful knowledge about underground detonations.[3]

4.3.3 Project Coach

In Project Coach it was proposed that a 5- to 10-kiloton nuclear device designed for producing maximum nuclear fluxes be detonated underground in a bedded-salt formation near Carlsbad, New Mexico, for producing milligram or larger quantities of transcurium elements. The isotopes produced would have been dispersed in 10,000 to 35,000 tons of salt debris that would have been mined a year after detonation to allow time for fission product decay. A tentative process flow sheet was developed and tested on kilogram samples of the Project Gnome shot debris. This debris contained high concentrations of silicates and was similar in content to the debris expected from Project Coach. Flow sheet tests indicated expected transplutonium element recoveries of 80 to 95%.[6]

4.3.4 Project Gasbuggy

Stimulation of gas production from wells by nuclear devices was thought to be one peaceful use of nuclear explosives.6 It was proposed that Project Gasbuggy use a IO-kiloton fission device with a 10,000-Ci tritium spike to study gas stimulation and tritium-hydrogen exchange in the water and gas, as well as possible tritium contamination of the methane product[4]

43.5 Production of Oil from Shale

The use of a nuclear device to crush Green River oil shale in place was proposed under the Plowshare Program. The recovery of oil from the crushed shale would involve in situ retorting. Chem Tech staff conducted bench-scale studies on radionuclide-spiked crushed shale samples.[4] It was determined that tritium was the major potential contaminant of the recovered oil. Contamination was determined to be more rapid when the shale was exposed to tritiated water or tritiated hydrogen than when exposed to tritiated hydrocarbons.[5]

4.3.6 Copper Ores

The Division of Peaceful Nuclear Explosives of the AEC proposed the crushing of copper ores as a possible peaceful use of nuclear devices. The controlled explosion would be followed by in situ leaching of the shot debris to recover the product. The process flow sheet proposed by Chem Tech staff members included percolating dilute sulfuric acid down through the nuclear-broken ore to dissolve the copper, collecting the leach liquor at the bottom of the ore body and pumping it to the surface, recovering the copper concentrate from the solution by cementation on iron, and recycling the barren solution (after adding more acid) for use in the leaching step. Radiotracer studies with ^{106}Ru were conducted to determine possible ruthenium contamination of the copper product.[7]

4.3.7 Magnesium Ores

Based on discussions with the Tennessee Valley Authority and the U.S. Geological Survey, it was concluded that recovery of magnesium, and possibly nickel and chromium, from olivine deposits might be possible. The large olivine deposits contained hundreds of millions of tons of olivine (48% MgO).[6]

4.4 NUCLEAR REGULATORY ASSISTANCE PROGRAMS,

Ray Blanco

In 197 1, the Nuclear Regulatory Commission (NRC) requested assistance from ORNL in reviewing and supplementary construction and operating license applications for commercial nuclear power plants. This was a new field of work for Chem Tech and other personnel at ORNL. By 1972 this grew into a larger program for all nuclear installations. and I moved from Section Chief for Chemical Development to Director of NRC programs in Chem Tech.

A series of cost benefit chemical engineering surveys were prepared to assist the NRC in defining the phrase “as low as reasonably achievable” (ALARA). This philosophic approach was of prime importance to the NRC in developing more stringent release limits for nuclear power and processing plants. The surveys determined the cost in dollars to reduce the amounts of radioactive materials released to the environment by adding successive treatment steps to the waste streams released to the environment. An evaluation of the decrease in radiation dose to the population surrounding the plants for each increment of treatment was also prepared. Graphs of the effect of added waste treatment steps on total body and thyroid dose versus annual treatment cost were developed. The abscissa of the graphs showed the dose and the ordinate the annual cost in dollars. ALARA was judged to be the area where increasing waste treatment steps and cost produced little decrease in dose.*

The first survey served as a part of the technical basis for the environmental statement for the guidelines for limiting the releases from LWR.[8] This, in turn, was the basis for the NRC to issue a revised ***Code of Federal Regulations,*** Title 10, Part 50, Appendix . This became the rule for licensing all nuclear power stations.

During the period 1972-78, similar surveys were issued on (1) reprocessing irradiated LWR fuel, (2) reprocessing irradiated HTGR fuel, (3) fabrication of LWR fuel-containing uranium, (4) fabrication of LWR fuel-containing plutonium, (5) milling of uranium ore, (6) fabrication of HTGR fuel, (7) conversion of yellow cake to UF_6, and (8) conversion of uranium to UF_6.[8,9]

A final paper in this series was published in ***Science*** entitled "Radiologic Impact of Airborne Effluents of Coal and Nuclear Plants."[10] Coal contains up to about 43 ppm of uranium and 48 ppm of thorium in equilibrium with radium and other decay products. The summary from the paper states "Radiation doses from airborne effluents of model coal-fired and nuclear power plants (1000 megawatts electric) are compared. Assuming a 1 percent ash release to the atmosphere (Environmental Protection Agency regulation) and 1 part per million of uranium and 2 parts per million of thorium in the coal (approximately the U.S. average), population doses from the coal plant are typically higher than those from pressurized-water or boiling-water reactors that meet government regulations. Higher radionuclide content and ash releases are common and would result in increased doses from the coal plant." The study does not assess the impact of nonradiological pollutants or the total radiological impacts of a coal versus a nuclear economy. Of course, any ash ***not*** released in the fly ash is present in the bottom and precipitation ash and, in general, is released to holding ponds. The mere fact that coal-fired power plants release radioactive materials to the environment in the fly and bottom ash (radium is the principal contributor of dose to the surrounding population) and that coal-fired plants typically release more radioactive materials in airborne effluents than LWR power plants which meet government regulations (10 CFR Pt. 50 Appendix 1) caused considerable comment in the nuclear and coal industry and prompted a call from the EPA. However, the issue was soon forgotten. The paper received an Award of Merit from the Society for Technical Communications through the assistance of Cathy Shappert, our technical editor.

Chemical Technology collaborators for these surveys were B. C. Finney, E. J. Frederick, A. H. Kibbey, H. W. Godbee, F. G. Kitts, W. Davis, Jr., R. B. Lindauer, G. S. Ryon, J. W. Roddy, J. P. McBride, and R. E. Blanco. Environmental Sciences, Health Physics, and Metals and Ceramics Divisions provided co-authors.

4.5 NUCLEAR ANALYSES: THE HISTORY OF THE ORIGEN COMPUTER CODE,

Scott B. Ludwig

The ORIGEN computer code; used throughout the world as the starting point for a wide variety of nuclear analyses, was created by the Engineering Coordination and Analysis Section (EC&A) (formerly Process Design) of the Chemical Technology Division at Oak Ridge National Laboratory. ORIGEN is an acronym for "Oak Ridge Isotope GENeration." ORIGEN and ORIGEN2 have been made available to users worldwide through the Radiation Shielding Information Center (RSIC), and since the initial release of ORIGEN in 1973, about 1000 users have acquired either ORIGEN, ORIGEN2, or ORIGEN2-PC. In addition to ORIGEN and ORIGEN2, others have created their own versions of ORIGEN, including KORIGEN (Karisruhe, Germany's version), ORIGEN-JR (Japan Research), SANDOR (Sandia Labs), and ORIGEN-S (part of SCALE package developed by ORNL's Computing and Telecommunications Division). When one mentions the name ORIGEN, Chem Tech staff may remember the original ORIGEN code created by Mike Bell in the early 1970s or may think of Allen Croff's ORIGEN2 code created in the late 1970s. Whichever version comes to mind, the creation of ORIGEN actually occurred in the 1960s, and many staff within Chem

Tech played an important role in the development of what is commonly called ORIGEN.

Of the thousands of computer codes created by the scientific community each year, only a few are so blessed as to take on a life of their own, to grow and mature over the years, and to endure the test of time. ORIGEN is one such code and is one of the more famous codes used in the nuclear industry, both in the United States, and worldwide.

4.5.1 Beginnings

ORIGEN was created to predict the concentrations and radiological characteristics of individual isotopes in nuclear reactor fuel and the products (including wastes) of processing spent fuel when their initial compositions and the burnup characteristics of the reactor are known. ORIGEN requires as input a library of nuclear data containing half-lives, decay schemes, cross sections, fission yields, and disintegration energies. The ORIGEN code was first mentioned in the Chem Tech annual progress report for 1969.[11] ORIGEN was the first code to deal with a large matrix (10,000 by 10,000) encompassing over 1000 nuclides undergoing simultaneous transmutation, decay, and flow in ten separate compartments. Most neutronic codes only dealt with a handful of the most important radionuclides in a single compartment. The calculation of such a large transition matrix allowed ORIGEN to determine the concentration of actinides, fission products, and light elements within the Molten Salt Reactor Experiment (MSRE), since the concentration also determined the radioactivity and thermal power from the decay of the radioactive species. The decay heat was a controlling parameter for the design of the reactor. From this start, the ORIGEN code became an important tool in the design and analysis of reactors, reprocessing plants, shipping casks, and waste disposal facilities.

4.5.2 The Players

ORIGEN first came into existence in Chem Tech in the mid-1960s. Jere Nichols initiated the development of ORIGEN and was responsible for the development of the recursion technique for solution of the sparse matrix (matrix exponential method) used in ORIGEN to solve the set of first-order linear differential equations that describe the transient concentrations of radionuclides within the reactor. Jere Nichols also created the now famous ORIGEN flux parameters THERM, RES , and FAST and derived (by hand) the cross sections for the first 1000 nuclides using existing compilations of integral cross section and flux spectra and the Westcott method. The initial reactor models in ORIGEN (LWR, HTGR, LMFBR, and MSBR) were all developed (by hand) from estimates of the values of THERM, RES, and FAST and analytical solutions of the reactor physics of each type of reactor based on the assumption that the neutron flux distribution is represented by a combination of a Maxwellian, a $1/\nu$, and a fission spectrum. Bill Nestor created the indexing technique used in ORIGEN to identify the non-zero terms in the matrix. Much of the decay data, cross sections, and photon yield data were developed by E. D. Arnold and Frank Soard. Mike Bell's first assignment in Chem Tech (under Jere Nichols) was ORIGEN. Mike was responsible for turning ORIGEN from an "in-house" code into the version known around the world Mike developed the initial ORIGEN documentation and implemented the Bateman equations used for some forms of radioactive decay. Soon after ORIGEN was first released through the Radiation Shielding Information Center (RSIC), Mike Bell left ORNL for a position at the NRC. Charles W. Kee assumed the role of ORIGEN custodian. In the first in a series of "ORIGEN Newsletters" dated June 1977, Kee describes the version numbering of ORIGEN, with version 1 .O being the original Mike Bell version and 1.1 representing the version being distributed by RSIC at that time. Another version, numbered 2.0, was in use within Chem Tech's EC&A Section and included features dealing with the chemical reprocessing of nuclear waste. Kee also mentions the existence of two other versions--one by Bill Hermann of C&TD that employs the FIDO input method and uses a coupling code to access AMPX multigroup cross sections (p&cursor to ORIGEN-S) and another developed by Allen Croff of Chem Tech that uses a much more flexible problem description which allows recycling calculations and more flexibility concerning reprocessing. At this point, further development of ORIGEN appears to have ceased in favor of the Bill Hermann and Allen Croff versions, namely, ORIGEN-S and ORIGEN2. Kee was also responsible for extensive revisions to the cross section data for light elements.

ORIGEN-S became an integral part of the SCALE system. ORIGEN2 developed a series of reactor models using more detailed reactor physics calculations. Under Allen Croff from the mid-1970s through the early 1980s, ORIGEN2 models for PWRs and boiling-water reactors (BWRs), CANDUs, and LMFBRs [including the Clinch River Breeder Reactor (CRBR) and the Fast Flux Test Facility (FFTF)] were developed. Croff also did extensive work updating the decay and photon libraries and determined the compositions of the fuel and structural components in the reactor, including the distribution of various impurities in each material. In this work, Croff was assisted by Mike Bjerke, Chuck Weisbin, Lester M. Petrie, and Wayne Morrison. In addition to all the work on data libraries for ORIGEN2, Croff created ORIGEN2's unique "command oriented" input structure. Using a few simple commands, the user can specify complex flow sheets undergoing a series of irradiation, decay, and reprocessing steps. Croff passed the role of ORIGEN2 "guru" to Chuck Alexander, then to Ron Ashline, and most recently to Scott Ludwig. ORIGEN2 was adapted to the IBM PC in 1986 by the late Jim Marable. In the late 1980s, ORIGEN2 code revisions and additional reactor models for LWRs were developed by John-Paul Renier and Scott Ludwig. Most recently, an ORIGEN2 version capable of working on 80386 and 80486 PCs was developed.

4.5.3 ORIGEN-Supporting the Development of Nuclear Energy in the United States

ORIGEN and ORIGEN2 have been used extensively to support the development of nuclear energy concepts within the United States. ORIGEN was first used to support the siting report for fuel reprocessing in 1968.[12] During the 1970s and 1980s, ORIGEN or ORIGEN2 results have found their way into many major documents for DOE. ORIGEN/ORIGEN2 have also been routinely used by both license applicants and regulatory authorities to determine radiological properties. Fuel cycle areas included are reactors, spent fuel storage, transportation casks, reprocessing, fuel fabrication, and waste treatment/disposal.

4.5.4 Bibliography

A brief topical listing of documents that include ORIGEN/ORIGEN2 results is given below.

Reactor Safety

- "Reactor Safety Study-An Assessment of Accident Risks to U.S. Commercial Nuclear Power Plants", Appendix VI, WASH-1900 (NUREG 75/014).

Inventory Projections

- J. 0. Blomeke, C. W. Kee, and J. P. Nichols, ***Projections of Radioactive Wastes to be Generated by the U.S. Nuclear Power industry,*** ORNL/TM-3965 (1974).
- C. W. Alexander, C. W. Kee, A. G. Croff, and J. 0. Blomeke, ***Projections of Spent Fuel to be Discharged by the U.S. Nuclear Power Industry,*** ORNL/TM-6008 (1977).
- J. A. Klein et ***al., Spent Fuel and Radioactive Waste Inventories and Projections as of December 31, 1980,*** DOE/NE-0017 (1981) and subsequent annual updates numbered DOE/RW-0006 prepared by the ORNL Integrated Data Base Program.
- C. W. Forsberg, C. W. Alexander, and G. W. Morrison, "Integrated Data Base Projections," ***Trans. Am. Nucl. Soc.*** **41**, 83 (1982).
- ***Characteristics of Spent Fuel, High-Level Waste, and Other Radioactive Wastes Which May Require Long-Tenn Isolation,*** DOE/RW-0184, vols. l-6 (December 1987) and vols. '7-8 (June 1988).

Reprocessing

- ***Correlation of Radioactive Waste Treatment Costs and the Environmental Impact of Waste Effluents in the Nuclear Fuel Cycle—Reprocessing Light-Water Reactor Fuel,*** ORNL/NUREG/TM-6 (1977).

Environmental Impacts

- ***Final Generic Environmental Statement on the Use of Recycle Plutonium in Mixed Oxide Fuel in Light-Water Cooled Reactors, NUREG-0002*** (1976)
- ***Statement of Position of the United States Department of Energy in the Matter of Proposed Rulemaking on the Storage and Disposal of Nuclear Waste (Waste Confidence Rulemaking),*** DOE/NE-0007 (1980).
- ***Final Environmental Impact Statement - Management of Commercially-Generated Radioactive Waste,*** DOE/EIS-0046F (1980).
- ***Technical Support of Standards for High-Level Radioactive Waste Management, Volume A-Source Term Characterization,*** EPA 520/4-79-007A (1977).

4.6 SPENT FUEL AND RADWASTE DATA BASES, Karl Notz

Projections of future quantities of spent nuclear fuel and other radioactive wastes, along with their radiological properties, are the basis for planning future requirements regarding the handling and eventual disposal of these materials. This future planning includes systems alternatives such as spent fuel reprocessing, by-product recovery, assembly consolidation, various immobilization technologies, and also transportation and temporary storage as well as final disposal. One of the earliest applications of the ORIGEN and ORIGEN2 codes (see Sect. 4.5) was to calculate the radiological properties of spent fuels and high-level waste from commercial reprocessing. Three major reports by "Tex" Blomeke and Jere Nichols in 1973; Charlie Kee and Nichols in 1974; and Chuck Alexander, Kee, Allen Croff, and Blomeke in 1977 were milestone works in this regard. Since that time, many changes have occurred in national policy that drastically effected technological implementation, including, for example the following:

- Commercial reprocessing has been halted in this country.
- The preferred repository geology has been changed from bedded salt to volcanic tuff (the Yucca Mountain site in Nevada, still under extreme contention).
- DOE has created a geologic disposal site for defense TRU waste [the Waste Isolation Pilot Plant (WIPP) facility near Carlsbad, New Mexico, still waiting for judicial approval to open on a test-only basis].
- Those sites currently still being used for shallow-land burial of low-level waste are in the process of being phased out (to be replaced by new sites to be developed by "compacts" of states).
- Commercial spent fuel is now being stored on-site by each utility in ever-increasing quantities.
- DOE is trying to locate a site for a centralized away-from-reactor storage site for LWR spent fuel (a Monitored Retrievable Storage facility (MRS)).
- An earlier proposal for large-scale consolidation of LWR fuel assemblies is being re-evaluated.
- Defense production reactors and defense reprocessing have been shut down.
- Start-up of the first two vitrification plants (one each for commercial and defense high-level waste) continues to be delayed
- P/T (Partitioning/Transmutation) is, once again, under serious consideration.

One thing has remained constant, however, and that is the need to know the fundamental properties of the starting materials. DOE will eventually be directly responsible for all of the nation's spent nuclear fuel and already has jurisdiction over high-level waste, TRU waste, remedial action waste, and much of the low-level waste. The ready availability of comprehensive and self-consistent data on inventories, projections, and characteristics of these materials is clearly an essential component of all aspects of dealing with these materials. Toward this objective, the DOE has funded the creation of two major data bases, the so-called Integrated Data Base and the Characteristics Data Base. Both were conceived within the Chemical Technology Division and implemented by Chem Tech staff. Both draw extensively on data sources external to ORNL and depend strongly on cooperative interaction with other national laboratories and other DOE organizations. Both have acquired a well-deserved reputation for thoroughness and integrity of technical data. Both are highly regarded by their many users. Each is described briefly in the following paragraphs.

4.6.1 Integrated Data Base

The Integrated Data Base, referred to as the IDB, provides domestic spent fuel and radioactive waste inventories, projections, and characteristics of spent fuel, high-level waste, TRU waste, low-level waste, remedial action waste, mill tailings, and mixed waste. Thus, the IDB covers all radioactive materials, which necessarily limits the level of detail. These data are assembled in a one-volume report. It was first published in its present form in 1981 and is updated annually. The latest (199 1) edition is report number DOE/RW-0006, Revision 7. Along the way, a PC data base of summary data was added using a menu-driven format written in dBASE. This was one of the first significant applications of PC technology and matching data base management software within DOE. Among its many users, the IDB report is often referred-to as "the blue book" because of its blue cover. Development of the IDB was led by Karl Notz and Blomeke. The original IDB staff included Herschel Godbee, Lloyd Carter, Arlene Kibbey, Alexander, Bruce Finney, Charles Forsberg, and Wayne Morrison. The IDB is now under the guidance of Jerry Klein and Steve Storch.

4.6.2 Characteristics Data Base

The Characteristics Data Base, or CDB, covers only those materials that will, or may, be eventually disposed of in a geologic repository (such as Yucca Mountain). This includes LWR spent fuel, immobilized high-level waste, non-LWR spent fuel, and miscellaneous wastes (which are largely sealed isotope capsules and greater-than-Class-C low-level waste). These materials are characterized in extensive detail, including physical, chemical, radiological, and thermal properties, as well as inventories and projections, The CDB was first issued in 1987 and 1988 as eight volumes plus five PC menu-driven data bases covering LWR quantities, assemblies, hardware, and radiological properties and high-level waste. The first revision will be released in 1992 as report number DOE/RW-0184, Revision 1, and has an additional PC data base on LWR assembly serial numbers. The CDB was originated under Karl Notz, and the CDB staff included Royes Salmon, Al Irvine, Tim Welch, Bill Reich, and Scott Moore (a local consultant from Automated Sciences Group) plus some dBASE programmers. A forerunner to the CDB was a 1985 report and a PC data base by Bill Roddy et al. on the physical and decay characteristics of LWR spent fuel.

4.7 TRANSPORTATION STUDIES,

Larry B. Shappert

The Transportation Group in the Chemical Technology Division got its beginning in the early 1960s. At that time, drop and puncture testing had been carried out at ORNL primarily in support of AEC (and now DOE) packaging programs. The work was initiated in 1960 by Larry Shappert to investigate the damage that would be accrued by a package if it were subject to the test requirements of the federal regulations. At that time, there was very little experimental evidence as to how packages designed to ship radioactive material would behave under the stringent regulatory requirements. Many of these activities were originally documented in different volumes of a master report, number ORNL/TM-1312. This series continued through Volume 19, which was published in 1977.

In order to carry out the drop testing of packages, a small impact pad was constructed in an area close to the X-10 steam plant in the early 1960s. Above the impact pad, a 40-ft-tall tower structure was built for use by experimenters to raise small packages to an elevation of 30 ft above the impact pad and drop them. Testing of packages has continued and has supported the preparation of many Safety Analysis Reports on Packagings (SARPs). The SARPs were prepared by Shappert, Bart Klima, Don Box, and others, depending upon the package and its use. These and related activities ultimately led to the development of the ***Cask Designers Guide*** (ORNL-NSIC-68) in 1970 by ORNL staff and other experts in the field of packaging design.

In subsequent years, the facilities, equipment, experience base, and scoped activities have continued to expand. As packagings have gotten larger and heavier, the ORNL staff, including Box and R. D. Seagren, surveyed various locations at X-10 with an eye to increasing the lifting capability at a fixed facility, rather than rely on dropping test pieces from a mobile crane. The Tower Shielding Facility (TSF) was found to be such a place: the towers can lift weights in excess of 25 tons and, with some modifications to the lifting system, may be increased up to 100 tons (Fig. 4.14). As a result, a large impact pad was installed at the TSF in the mid-1970s and a number of packages, some weighing up to 25 tons, were drop tested (Fig. 4.15).

In the late 1970s the Transportation Group moved into Bldg. 3019. Dave Joy and John Marshall of Chem Tech and Paul Johnson of the Computing and Telecommunications Division (C&TD) initiated work on a truck routing model and then expanded the effort to include a rail/barge routing model. These activities included the development of mapping capabilities which could be meshed with transportation routes generated by a computer and, ultimately, population density information within the continental United States (Fig. 4.16). Joy and B. Hudson developed a stochastic model of a cask- receiving facility using the GPSS code and a spent fuel logistics model that simulated the flow of waste material through DOE facilities. At about the same time, the Transportation Group became involved with Sandia National Laboratories in the design of a

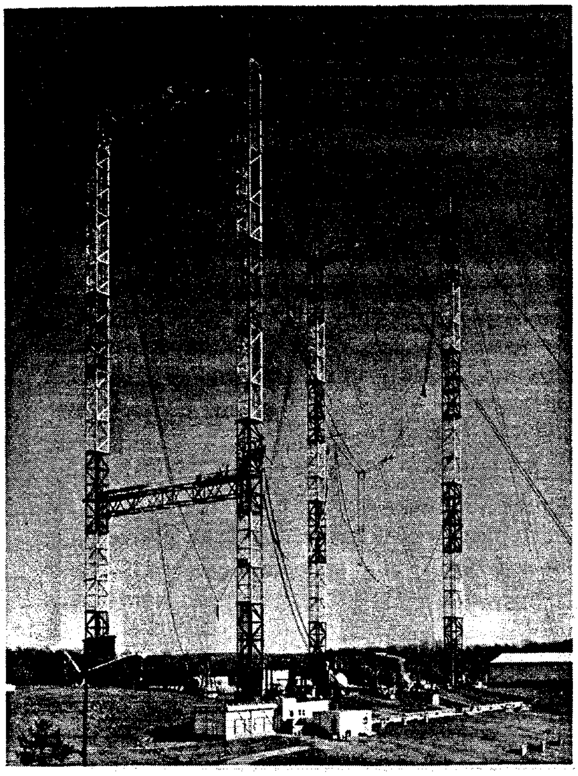


Fig. 4.14. The towers of the Tower Shielding Facility have been fitted with cables and special instrumentation for drop-testing of waste transportation casks. To facilitate testing, the concrete pad on which the casks are dropped was heavily reinforced with steel bars.

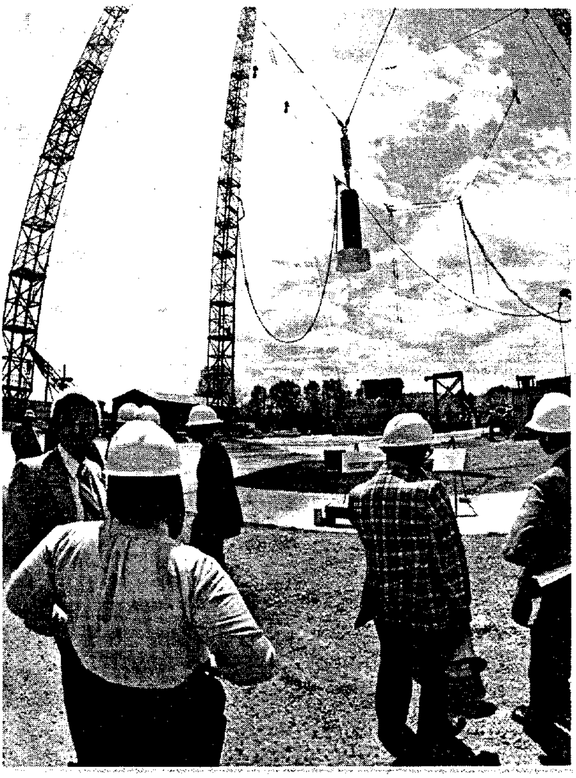


Fig. 4.15 The first in a series of drop tests conducted on a 22-ton spent-fuel shipping cask at ORNL's Tower Shielding Facility, which was normally used to suspend the Tower Shielding Reactor il. The photo was taken using a wide-angle (fish-eye) tens, which creates a distorted image of the steel towers.

Fig. 4.16. Dave Joy and Paul Johnson studying newly created routing maps produced on the computer.

cask for shipping CRBR fuel. That work continued until the CRBR concept was abandoned

More recently, the Transportation Group has continued to expand both its personnel and its technical capabilities. In the mid-1980s, rapid expansion in the transportation area began, which included assisting DOE in planning and developing its system for transporting spent nuclear fuel from the nation's 122 nuclear reactors. The transportation activities in CTD were merged under R. R. Rawl, who was a key individual in promoting support of the transportation needs throughout Martin Marietta Energy Systems. A concept for a Cask Maintenance Facility was developed, the functions of transportation were delineated in detail, the transportation system was described, and numerous engineering and trade-off studies were performed The spent nuclear fuel shipping efforts grew in the early 1990s to include institutional and economic and systems analyses, and the work in transportation expanded even further to include support in environmental health and safety, waste management, and an NRC-sponsored study for specification packages. Those who supported these efforts included Leonard Dickerson, Juan Ferrada, Ruth Gove, Vicki Green, Francis Kovac, Ron Pope, Joe Ratledge, Rick Rawl, Larry Shappert, Max Wankerl, Brad Welles, and Mimi Welch of Chem Tech; Paul Johnson of C&TD; Glen Harrison of Energy Division; and Reid Attaway, Larry Medley, Mark Rennich, and Andy Williamson of Central Engineering.

Testing of packages continues and has been. carried out for both the DOE as well as companies from the private sector. The data and information generated in the tests have been instrumental in obtaining DOE, NRC, and Department of Transportation (DOT) approvals of the package designs, meeting all necessary technical and QA requirements.

4.7.1 Drop Test Facilities

Test facilities were developed to test the heavy packages that are used to shield highly radioactive material.

Impact (Target) Pads. There are two drop test facilities that have been used to test packages. The smallest is the Small Test Facility (STF) that utilizes a concrete pad and has an impact surface of armor plate. This facility was modified in 1990 to provide a larger impacting surface than was available in the original pad

The concrete and steel in the original pad weighs approximately 40 tons: its top surface is about 11 ft by 10 ft and has an 8-ft-square armor plate surface imbedded in it. Recently several 6-in.-thick pieces of armor plate were added, which effectively cover the entire pad and overhang about 2 ft in one direction. The additional armor plate is welded to the original plate and adds approximately 20 tons of weight, bringing the total weight of the pad to approximately 60 tons. However, it has a significantly larger effective mass since the bulk of the pad rests on a 3-ft-diam concrete column that was sunk into bedrock approximately 10 ft below grade.

A much larger pad was built at the TSF. This weighs approximately 670 tons and has an armor-plate impact surface which is approximately 8 ft wide and 20 ft long. It was designed to accept the impact of a 100-ton cask dropped from a height of 30 ft.

4.8 REACTOR SAFETY RESEARCH, BUILDING 4501, R. A. Lorenz

4.8.1 George Parker and the Manhattan Project

The pioneer of reactor safety research in the Chem Tech Division is George Parker. While George worked for DuPont Powder Works in Louisville, Kentucky, in 1942, he noticed that key scientists were leaving the Works without saying where they were going or why they were leaving. Another indication that something interesting was going on was that a lecturer from Purdue University commented to George that while in Knoxville he had seen several Nobel Laureates together and that he had seen the word "uranium" on a blackboard. George learned that one of the "missing" DuPont people had been seen at the University of Chicago. Since George was looking for something more interesting to do than making gunpowder, he wrote to the University of Chicago saying that he believed that they were doing something special for the war effort and that he would like to get in on it. He immediately received a telegram asking him to come up to the university and to be prepared to stay. The first day or so after arrival consisted of a grilling from the FBI concerning how and what he knew about what was happening at the university. George told them he thought that they were involved in making a bomb. As you can imagine, George was hired on the spot;

he came to Oak Ridge in 1943. Here he pioneered in fission product separations, which included the discovery of technetium. This work was conducted in Bldg. 706-C (now Bldg. 3026).

F. R Bruce has told how personal decontamination was done in the early days: go home and wash the dishes. Frank claimed that the method worked well, but he did not say whether he had hands-on experience.

4.8.2 Early Fission Product Release Tests

G. W. Parker, G. E. Creek, and W. J. Martin began fission-product release testing in about 1955. Parker was the planner; Creek, the calculator (and often glassblower); and Martin, the equipment specialist. The earliest tests were with metal-clad research-reactor-type fuels to provide safety information for reactors such as the MTR. Uranium slug-type fuel was also tested for Hanford reactor safety studies. Uranium dioxide fuels were studied to provide LWR safety information for TID- 14844, the AEC's first reactor safety analysis. Tests were first performed with trace-irradiated fuel. In the early 1960s, 'high burnup" fuel became available: at that time, high burnup was 4 MWd/kg U (now high burnup is anything > 40 MWd/kg U).
R. A. Lorenz joined Parker's group in ~1961 . One of the few unpleasantries associated with working in Bldg. 4501 in the late 50's and 60's was the odor of propylene diamine (PDA) that permeated parts of the building. This material was used earlier in the OREX process researched by Chem Tech in Bldg. 4501 (Sect. 2.4).

Parker used several methods to reach high temperatures, including the melting point of UO_2: arc-image furnace (focused light from a carbon arc), a tungsten V-filament, and induction-heated tantalum, and tungsten crucibles. Parker and Lorenz pioneered a technique using electrically heated tungsten rods inserted through the center of stacks of UO_2 pellets. This method was further developed in the 1970s by researchers at Karlsruhe, Germany, to heat bundles of i-m-long rods in the CORA tests, a very sophisticated and successful apparatus that is still in operation. The above work was summarized in ORNL-398 1, the "Bible" of early fission product release. C. J. Barton assisted with this and other report writing.

In 1959, R. A. Lorenz was asked to design a fission product release experiment in which UO_2 fuel would be heated to the melting point from fission heat in the Oak Ridge Research Reactor (ORR). He chose to design an irradiation facility for the test packages that would allow for installation of the package into the reactor core and adjust its position while the reactor was at full power. Dick asked for a design-draftsman to help with the project, and a young man was sent. He volunteered to work out the design of a piston to seal the bottom of the facility insert and to be driven by ORR core cooling water. Dick thought this to be a little ambitious (compared with a mechanically operated shutoff), but he said O.K. The facility and experiment package worked flawlessly for more than 40 tests. The talented design assistant-John E. Jones-is now Director of the Engineering Technology Division.

Tests of fission-product transport behavior began in 1964 with the operation of the Containment Mockup Facility (CMF). The Containment Research Installation (CRI) was built in 1966 to provide support for the LOFT (Loss of Flow Test) program. LOFT was a LWR at Idaho Falls built especially for accident testing. Tests in the CRI showed rapid deposition of simulants HI and I_2 as well as fission product iodine on the walls of the vessels, slow settling of a fraction of the iodine with aerosols, slow growth of organic iodides, and a high liquid/gas iodine partition coefficient. George had good connections with the AEC in Washington. He was famous for going up there and returning with money to continue his various fission product release and behavior programs.

Two types of in-reactor fission product release and fuel behavior tests were performed in the 1960s. Lorenz and Parker ran 12 tests in the TREAT reactor to study release under rapid heatup (to UO_2 melting) and cooldown. The first visiting scientist from Germany, Juergen Wilhelm, assisted with these tests. Juergen later became director of the Laboratory for Aerosol Physics and Filtration Technology (aerosol and iodine behavior) at Karlsruhe and became known by some as the "Iodine Pope." Other visiting scientists from Germany who worked in fission product studies were Heinz Buchholz, Horst Feuerstein, and Ernst Hoinkis.

M. F. Osborne joined Parker's group in 1967. LWR safety research was shifting away from fission product behavior in containments to accident-induced fuel rod rupture, cladding expansion (ballooning), and cladding embrittlement caused by oxidation of the cladding. Osborne and Parker, assisted by Bill Martin, tested the failure characteristics of highly irradiated Ziicaloy cladding. They found slightly less expansion than with unirradiated cladding. A bundle of 25 pressurized fuel rods was heated with internal tungsten lamp heaters. The rupturing fuel rods sounded like popcorn popping.

Lorenz and Parker performed two tests with 'I-rod bundles of l-m-long fuel rods in the TREAT reactor. These tests simulated behavior during a controlled loss-of-coolant accident (1200°C maximum temperature). In addition to fission-product release information from an irradiated center rod, these tests showed that the pressurized rods ballooned and ruptured in the same way as in out-of-reactor tests. One of the fuel rods from the second bundle test was accidently broken during post-test handling in the hot cell. This fact was used to bolster other ORNL data presented at the Emergency Core Cooling System hearings which showed that oxygen embrittlement of Zircaloy was important. When Parker and Lorenz visited Westinghouse in June 1967 to discuss a possible contract, the Westinghouse representative asked how the test fuel rods had been pressurized and sealed. Approximately two years later, the news was out that some commercial fuel rods were being pressurized with helium in order to maintain good thermal conductivity. Apparently, Westinghouse was trying to learn if ORNL had a better method for seal-welding pressurized fuel rods.

4.8.3 The Chemical Development Section

Most of the reactor safety research described above had been part of Reactor Chemistry Division programs. In 1973, this division was dissolved and the nuclear reactor safety research group and facilities became part of the newly formed Chemical Development Section in the Chemical Technology Division. A. P. Malinauskas was the new section chief.

Light-water reactor safety research was at a low point at this time. In 1971, Milton Shaw, Director of Reactor Development and Technology, had decreed that no further government-funded research was warranted, if additional safety information was needed, reactor vendors and utilities should pay for it. Of the Bldg. 4501 group, G. W. Parker, G. E. Creek, and even W. J. Martin were working at least part-time on environmental impact statements. M. F. Osborne was working on a gas-cooled reactor project to investigate the sorption of iodine on iron oxide.

R. A. Lorenz was working with Chem Tech's first visiting scientist from Japan, Hiroyuki Nagao, on the ignition of charcoal by decay heat from radioactive iodine. George Parker had suggested looking at ^{130}I as the radioactive isotope to use instead of ^{131}I because of its shorter half-life and the possibility of generating large quantities by activation of ^{129}I. This turned out to be feasible, and 1000-Ci amounts were generated in the HFIR for each test conducted in cell A, Bldg. 4501. It was found that heat from the oxidation of the charcoal was as important as the heat from radioactive decay, and desorption from the ignited charcoal bed was surprisingly slow, especially when the charcoal contained an excess of potassium over iodine.

In late 1973 and early 1974, G. W. Parker, assisted by G. E. Creek and somewhat by R. A. Lorenz, contributed fission product release rates and behavior to WASH- 1400, the AEC report on severe reactor accidents. Back in 1961, George had contributed almost all of the needed fission product information for TID-14844, the AEC reactor safety report on which the AEC Regulatory Guides for power reactor design requirements were based. George said that most of the information he supplied was given via telephone. It is interesting to note that the "Reg-Guides" resulting from TID-14844 are still current, but Ed Beahm is reexamining iodine behavior for a probable Reg-Guide revision (Sect. 4.8.13).

4.8.4 Restart of Fission-Product Release Testing

After about a three-year layoff from LWR safety research, the AEC began fission product release research at ORNL in the fall of 1974. R. A. Lorenz and M. F. Osborne were assigned the task; A. P. Malinauskas was the Project Manager and Ray DiSalvo was the AEC sponsor. When M. F. Osborne went to Germany as first technical liaison for the USAEC-FRG agreement on core melt research, Jack Collins joined the project from the Isotopes Division where his experience with the chemistry of small quantities of various elements including uranium and plutonium made him a natural for this work. As part of his isotopes work, he had operated calutrons left over from the war years. Morris Osborne was on assignment in Karlsruhe, Germany, from October 1975 to

August 1977. Technicians on this project have been S. R. Manning, R. L. Towns, 0. L. Kirkland, J. R. Travis, and C. S. Webster.

This work has been funded off and on for 17 years. Work progressed along these lines: "control" tests with fission product simulant species in the absence of fuel (1975), Knudsen cell tests with cesium species (1975), "implant" tests with fission product species implanted in the pellet-clad gap space (1975). low burnup (LBU) tests with low-burnup (1 to 2 MWd/kg) fuel (1976), high burnup (HBU) tests with high burnup (20 to 30 MWd/kg) fuel between 500 and 1200°C (1976). high temperature (HT) tests with high burnup fuel between 1300 and 1610°C (1978), and tests with BWR fuel (-10 MWd/kg) between 800 and 1200°C (1979). Peaks of activity occurred in 1977, when seven tests were run, and in 1979, when five tests were completed.

In September 1980, more than a year after the TMI-2 accident, NRC began discussing plans for higher-temperature tests. The furnace was redesigned and the HI (Horizontal Induction Heated) tests were run from 1982 to 1984 (1400–2000°C). Tests with Karlsruhe simulant fuel (HS Series) were run in 1984 (2000–2425°C). A major design improvement was made in 1985. The furnace was oriented vertically, and two additional sequentially operated fission-product collection systems were added. Six VI series tests Were performed (1725–2425°C) between 1985 and 1991.

The Knudsen cell work performed by Collins in 1975 and 1976 showed that the vapor pressure of both CsOH and Cs_2CO_3 was substantially reduced in the presence of UO_2. The simulant studies (implant tests) performed by Lorenz and Collins at that time showed that the vapor pressure of both cesium and iodine were reduced by factors of 10 to 100 when CsI and CsOH were deposited on UO_2 pellets inside the Zircaloy cladding.

A low point in morale came in the summer of 1976 when an abstract of a paper covering the "implant" tests was submitted for an international meeting in Norway. A Swedish reviewer commented that ORNL should be working with real irradiated fuel and not with simulants. This was taken seriously in both Washington and at ORNL, where consideration was given to withdrawing the paper or changing the authorship. The paper was well received and the simulant (implant) work provided the foundation for the LOCA Source Term Model. It was the opinion of Chem Tech's fission-product release team that these and later radioactively traced simulant tests, dollar-fordollar, provided ten times as much information as the expensive yet very essential tests with commercial irradiated fuel.

The LOCA Source Term Model by Lorenz, Collins, and Malinauskas was published in early 1978 after inclusion of data from the first 11 tests with high-burnup commercial fuel. The model covers the release of cesium, iodine, and fission gas from ballooned and ruptured fuel rods in the temperature range of 500 to 1200°C. The model was confirmed by results from tests later run with high gap-inventory BWR fuel. It is still in use for low-temperature accidents such as shipping or fuel bundle handling accidents, in addition to the early stage of LWR accidents.

4.8.5 Hearings and Investigative Support

In early 1975, the AEC was divided into ERDA and NRC. Funding for Chem Tech came from the NRC. At about this time, George Parker was called as a witness at a licensing hearing for a proposed reactor since it was known that an "intervenor" was going to claim high cesium releases from fuel shipping accidents. George testified that cesium would not be released in significant amounts at the temperatures cited. The intervenor then was called to the stand and asked what he knew about cesium. He replied that everything he knew he had learned from George Parker. The "intervenor" was dismissed from further testimony.

Also about this time there was a very serious effort to obtain NRC approval of a proposed floating off-shore nuclear plant design. George Parker and Frank Binford were assigned to evaluate the safety of the plant and decided that it was not safe enough. George said that the original floating plant design looked like a good way to spread radioactivity all along the East Coast.

4.8.6 Response to the TMI-2 Accident

The TMI-2 accident occurred on March 28, 1979. Preliminary f&ion-product release estimates released a few days after the accident showed nearly equal percentage amounts of the cesium and iodine inventories in the primary cooling water. This was exactly the behavior that the fission-product release group had seen in their experimental results: equal percentage releases of cesium and iodine, with the iodine behaving like cesium iodide. When Lorenz heard about the TMI-2 releases, he commented to Jack Collins, 'There's our cesium iodide.*' This was not necessarily a correct deduction, of course, because most of the released fission products were still confined to the primary system, and all chemical forms of iodine would be there.

Babcock and Wilcox called for a meeting of specialists to discuss the accident. On April 4, Lorenz and two others from ORNL took a charter flight to Lynchburg. Lorenz presented evidence that showed that the temperature must have been above 2000°C for a period of time. Some "industry" representatives talked in terms of 2000°F (1093°C) or possibly 2800°F (1538°C), which were impossibly low temperatures. That evening the ORNL group had dinner with a metallurgist from B&W. His private opinion was "I'm willing to concede the upper third of the core." He was very realistic in his estimate made only 8 days after the accident.

Extensive assistance was provided to TMI-2 by other Chem Tech members in the days and years after the accident. Floyd Culler, director of the Electric Power Research Institute (EPRI), quickly called for assistance with the highly radioactive primary system water that had escaped to the TMI-2 Auxiliary Building, a building that was not designed for containment. Bob Brooksbank formed a team with Orlan Yarbrough, Jim Snider, and Frank Harrington, who immediately went to the reactor site to help with radioactive waste management projects. Snider and Harrington were involved in various engineering projects, while Yarbrough analyzed the potential release of ^{131}I and methods for preventing or mitigating such a release. They found that the charcoal traps in the TMI-2 Auxiliary Building had deteriorated and arranged for replacement traps to be flown in from Hanford by the Air Force (although this didn't occur until approximately 6 weeks later). A few days after the accidents, Les King, Emory Collins, and Bill Shannon joined the on-site team. They collected and analyzed data on ^{131}I evolution and determined that the release was only 15 Ci, even though the reactor fuel contained 10^7 Ci at the time of the accident.

A few weeks later, Bob Brooksbank was selected as a member of the first Technical Advisory Group (TAG), and Chem Tech began work on designing a decontamination process for the high-activity water generated by the accident. Dave Campbell, Emory Collins, Les King, and Joe Knauer were co-developers of this process, which was first used in 1981 to decontaminate the water. A variety of other cleanup projects were performed by Joe Knauer, Walt Bond, P. B. Orr, Lew Bird, Don Box, Herschel Godbee, and Tim Scott.

Brooksbank became the first Program Manager at ORNL for TMI-2 Assistance, which included projects in Chem Tech and other ORNL divisions. He was followed by Tony Malinauskas and Emory Collins during the 1980 to 1989 time period. Dave Campbell was the first appointee to the TMI-2 Technical Assistance and Advisory Group (TAAG), beginning in 1982. He was later joined by Malinauskas and Collins. The TAAG assisted with defueling and decontamination projects and devised the "quick look" examination, during which a television camera was first lowered into the core region.

Lorenz wrote a report analyzing the amount of ^{85}Kr that might be released from intact fuel rods during core removal operations. Lorenz and Jack Collins developed a model of cesium transport which showed that desorption of cesium from primary system surfaces was causing the very slow increase in cesium concentration in the primary water for up to 2 years after the accident.

4.8.7 The President's Commission (Kemeny Commission) on the TMI-2 Accident

President Carterappointed the Kemeny Commission to investigate the TMI-2 accident. A committee from the commission, headed by Tom Pigford, visited ORNL on June 11, 1979. Lorenz used Parker's fission-product release results obtained in the early 1960s, which showed that the fuel temperature must have been very high, probably for a long time. Volatile fission-product release estimates from TMI-2 measurements ranged around 50%, and steam-Zircaloy reaction estimates were ~60%. Since the zirconium oxidation occurs at very much lower temperatures than fission-product release, Lorenz said that the best explanation for these results was that 60% of the core got very hot and 40% remained cooled. He suggested that a possible mechanism for unusually high fission-product release might be the result of reaction between zirconium cladding and the UO_2 fuel. Pellet-cladding interactions had been under study at several laboratories, but only at low temperatures and only because of concern over cladding integrity. No fission-product release data had ever been obtained at high temperature with unoxidized Zircaloy-clad UO_2. The reaction between Zr and UO_2 came to be known as "liquefaction," but tests at ORNL in the HI and VI series showed that this reaction did not result in a significant enhancement of volatile fission-product release.

A. P. Malinauskas and D. 0. Campbell worked on a committee for the President's commission to investigate the behavior of iodine during the TMI-2 accident. The committee reported that the iodine released from the fuel, if not already in the iodide form, encountered a chemically reducing environment which converted it to iodide. The iodine subsequently went into solution as iodide when it contacted water. The CRI tests by Parker, Creek, and Martin had shown a similar end point for I_2 placed in oxidizing steam-air atmospheres for which liquid/gas partition coefficients of $\sim10^4$ were rapidly obtained.

4.8.8 Cesium, Iodine, and Cesium Iodide Before and After the TMI-2 Accident

The Chemical Technology Division has provided most of the LWR safety study information about the chemical and physical behavior of fission product iodine. The fission-product release studies (Sect. 4.8.4) provided information about the time period of -1 s after releases from the fuel. The aerosol reaction/sorption studies (Sect. 4.8.14) showed significant interaction with several aerosol components that would take place at high temperature in the first minute following release. The low-temperature iodine studies (Sect. 4.8.13) evaluated the complex behavior of iodine in the containment atmosphere and water pools.

The challenge to the fission-product release group was to determine whether fission product iodine existed in volatile forms (I_2, HI, or CH_3I) immediately after release from the UO_2 fuel or in much less volatile forms, such as CsI. The chemical form could not be positively identified because the high radioactivity and intimate mixing with other chemical species interfered with methods such as X-ray diffraction. The question was resolved by using activated charcoal, which showed that only trivial amounts of iodine existed in the volatile form.

By 1977, the Knudsen Cell, Implant (Simulant) tests, and HBU tests had shown interesting behavior of cesium and iodine. The vapor pressures of cesium and iodine were less than expected for CsOH and CsI in the presence of fuel and cladding, there was less release of cesium than iodine at, , lower temperatures, and released iodine behaved much like CsI when releases were above the trace level. In late 1977, T. M. Besmann and T. B. Lindemer performed thermodynamic calculations which confirmed the observed behaviors.

By November 1978, the evidence for "release" of iodine as CsI was convincing. In a paper by Malinauskas, Lorenz, Collins, and Osborne presented at the Sixth Water Reactor Safety Research Information Meeting, they stated that "the release data obtained in the High Burnup Fuel Test Series suggests that CsOH and CsI were the species of cesium and iodine that were released in the steam atmosphere tests," and "in a purified helium test, the iodine was collected as CsI." The importance of these observations is that CsI is much less volatile than I_2 or CH_3I. Thermodynamic calculations consistently showed that CsI was the most stable form of iodine under most accident conditions, but convincing experimental evidence for this fact previously had been lacking.

The HT and BWR series of tests performed in 1978 and 1979 provided more evidence for the existence of fission product iodine as CsI and the absence of highly volatile forms of iodine such as I_2 and HI. For iodine releases above 1% of inventory, the amount of volatile iodine was always $<$ 1% of the amount released. In the ORNL apparatus, collection occurred within 1 s of release from the fuel. Papers were presented at four national and international meetings between September and December of 1979 in which the authors explained the test results which showed that the released iodine behaved like CsI and was not in a volatile form. It was thought that iodine behavior results of the Chem Tech staff were being understood and accepted worldwide.

In July 1980, more than a year after the TMI-2 accident, A. P. Malinauskas and D. 0. Campbell attended a meeting at which they were disappointed to find that most of the reactor safety community was ignoring real iodine chemistry as demonstrated by the ORNL experimental results, by thermodynamic calculations, and inferred from iodine behavior at TMI-2. They were taking the simple "conservative" approach of assuming that released fission product iodine was in the volatile form I_2. W. R. Stratton (Los Alamos National

Laboratory), who was also dissatisfied with this approach, along with Campbell and Malinauskas, wrote a letter to the NRC commissioners explaining the evidence for their belief that fission product iodine "emerged from the fuel as cesium iodide" and requested that the true iodine chemical behavior be verified and applied to NRC Regulatory Guides, accident analyses, and guidelines for emergency evacuation. The letter had a big impact. A hearing was held by the NRC, with the result being that iodine chemistry was taken more seriously.

4.8.9 Post-TMI LWR Research

A. P. Malinauskas continued as Chemical Development Section Head. Contrary to what was expected, there was a lull in new reactor safety research after the TMI-2 accident. This is probably because NRC concentrated its resources on analyzing the accident. In early 1980, R. P. Wichner, who had been managing the HTGR studies, began building/rebuilding an LWR safety research program. The major activities and principal investigators in this program were fission product release, M. F. Osborne and R. A. Lorenz; iodine chemistry, L. M. Toth and E. C. Beahm (Fig. 4.17); Severe Accident Sequences Analysis Program (SASA), R. P. Wichner; fission product adsorption on aerosols, Roger Spence; iodiie leakage from steam generator tubing, S. D. Clinton and E. C. Beahm; core-melt studies, G. W. Parker (Fig. 4.18); aerosol transport, A. L. Wright; and iodine absorption by water sprays, Mike Albert, University of Tennessee, MS thesis. In 1983, J. R. Hightower became section head of the Chemical Development Section. He was followed by J. T. Bell in 1988.

4.8.10 Fission Product Release from Fuel

In the summer of 1980, M. F. Osborne began the design of a furnace that would heat LWR fuel in steam atmospheres to temperatures as high as 2300 K. J. R Travis and C. S. Webster joined the project in December 1980 and began preparing the hot cells for the new apparatus and setting up computerized data recording equipment. Travis and Webster came from hot cell Bldg. 4507, where they had worked with J. H. Goode, R. G. Stacy, and V. C. A. Vaughen on head-end reprocessing studies and parametric voloxidation studies. R. A. Lorenz assisted with design of the new apparatus and with safety analyses. Jack Collins returned from TRU in 1983 specializing in chemical interpretations of the data and to perform laboratory tests using radioactively traced fission product simulants (See Fig. 4.19). Six tests (HI-l to 6) were performed before the apparatus was converted to the vertical orientation in 1985. K. S. Norwood, a visiting scientist from the United Kingdom, contributed significantly to the HI test series (Fig. 4.20). The vertical apparatus featured higher temperature capability (2700 K), improved temperature measurement, more uniform temperature along the length of the 15-cm fuel specimen, three sequentially operated fission-product collection systems, and a hydrogen measuring system. Six tests (VI-l to 6) have been performed to date with both steam and hydrogen atmospheres using fuel from LWR reactors with burnups up to 47 MWd/kg U. Fission-product release rates obtained from this program provided the data worldwide for fission-product release models.

Test VI-4 provided a surprising outcome after a difficult start. NRC and ORNL program managers required that test VI-4 be run with a steep axial temperature gradient of ~400 K from the center to the top of the fuel specimen in order to provide an exact duplication of an in-reactor fission-product-release test run at Sandia. The ORNL experimenters maintained that such a temperature gradient makes it impossible to calculate accurate release rates as a function of temperature. Duplication of the temperature gradient required 6 months and 26 heatup tests before the proper temperature gradient and temperature calibration was obtained. Early in the actual test, the fuel specimen collapsed down into the uniform temperature zone, thus permitting the measurement of accurate release rates as a function of time and temperature. To this date, the experimenters refuse to reveal how they caused the fuel to collapse.

4.8.11 Modeling of Fission Product Release at High Temperature

In late 1980, the NRC instigated a high priority project to establish the technical bases for estimating fission product behavior during LWR accidents, which resulted in the famous NUREG-0772 report. This was done to improve upon the methods used in the 1974 WASH-1400 report ***Reactor Safety Study.*** R. P. Wichner and R. A. Lorenz wrote the section on fission product

Fig. 4.17. Members of the Spectrophotometric Studies Group from the Chemical Development Section consult with George Begun (Chemistry Division) on a collaborative investigation in which the structure of urania and zirconia hydrous polymers is being elucidated by means of Raman spectroscopy. Mac Toth (left) and Kevin Felker are on the back row. In the front row left to right are Karen Dodson, Susan Sherrow, and George Begun. This work was supported by DOE through the Office of Basic Energy Sciences, Division of Chemical Sciences.

Fig. 4.18. George Parker (left) and Al Sutton are at the control panel of the Chem Tech CORE-MELT Facility checking the heating rate to be used in melting a 10-kg simulated LWR nuclear reactor core. These unique experiments allow researchers to learn about phenomena that occur during those nuclear reactor accidents in which portions of the reactor core melt. Results from these experiments helped explain findings in the damaged reactor vessel at Three Mile Island. This work was performed in Chem Tech as part of ORNL's NRC program.

Fig. 4.19. Chem Tech members and visiting scientists in front of a hot cell used for fission product release studies. Top row (left to right): Jack Collins, Bob Hightower, Take Nakamura, Tony Wright, and Jim Travis. Bottom row (left to right): Morris Osborne, Stephen Daish, Y. -C. Tong, Charlie Webster, and Dick Lorenz.

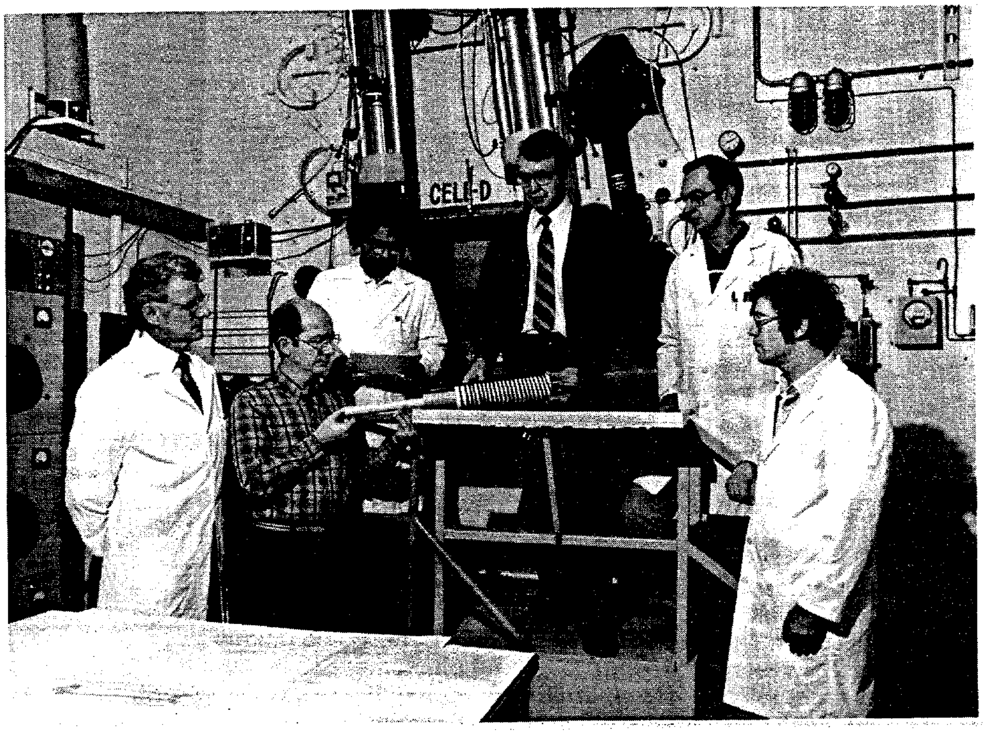


Fig. 4.20. Left to right, M. F. Osborne, J. L. Collins, C. S. Webster, R. A. Lorenz, J. R. Travis, and K. S. Norwood (United Kingdom) examine a furnace used in fission-product release tests.

release from fuel and devised the fractional release rate model which became known as CORSOR. The model is simple and easy to use, which accounts for much of its popularity. Although not as accurate as recent, more complicated models, it continues to be used and is the basis for comparison with all other fission-product release models. J. T. Bell, L. M. Toth, D. 0. Campbell, and A. P. Malinauskas assisted with the "Chemistry of Cesium and Iodine" chapter. The authors concluded that "the stability of CsI makes this compound the predominant iodine species for most conditions" in the gas phase.

In 1983, NRC asked ORNL to examine fission-product tellurium release rates. Commercial interests claimed that tellurium was released at the same rate as cesium, iodine, and fission gases, but the ORNL summary in NUREG-0772 indicated significantly lower release rates. The importance of this is that tellurium released late in the accident might actually have a better chance of escaping from containment. Lorenz, Beahm, and Wichner found that tellurium release was delayed by retention in unoxidized Zircaloy cladding but was released when the cladding became essentially completely oxidized. Jack Collins later performed tracer test C-9, which dramatically proved this to be the case.

R. A. Lorenz showed in 1985 that classical diffusion equations accounted for the time dependence of fission product release better than the CORSOR model. Takehiko Nakamura, a visiting scientist from Japan, correlated the ORNL test results using diffusion equations and developed the ORNL Diffusion Release Model. In 1991, the NRC changed the original CORSOR model to a diffusion-type model.

A. P. Malinauskas and R. A. Lorenz recently used the ORNL LOCA Source Term Model, mentioned previously, to calculate fission-product release during low-temperature shipping and fuel handling accidents. This model was also used in 1991 to determine when the first radioactivity would be released from fuel in order to know how fast reactor containment isolation valves would need to operate.

4.8.12 Severe Accident Sequence Analysis (SASA)

R. P. Wichner, assisted by R. A. Lorenz. W. Davis, Jr., A. L. Wright, C. F. Weber, and A. D. Mitchell, directed an investigation into fission product release, chemistry, and transport during specific reactor accidents. (Previous accident studies were much more generic in nature.) Steve Hodge's group in the Engineering Technology Division provided the time, temperature, atmosphere, and leakage rate data. The first accident studied was a station blackout (complete loss of electricity except for storage batteries) at the Browns Ferry- 1 BWR. The study was a landmark in that it demonstrated the importance of individual power plant designs and the importance of iodine chemistry. All reactor safety analyses now use the SASA approach of using exact design details for the plant being studied. J. W. Nehls and E. C. Beahm worked on later SASA studies, which continued to 1985. The work of Beahm and Weber led to the development of the TRENDS model for iodine behavior in reactor containment buildings.

4.8.13 Iodine Chemistry and Volatility Studies

L. M. Toth, K. E. Dodson, and 0. L. Kirkland began studying iodine chemistry in 1981 using absorption spectrophotometry at temperatures up to 150°C. They measured the production of volatile iodine species as a function of gamma radiation and solution characteristics. Ed Beahm and Bill Shockley began the study of organic iodide formation in 1984. E. C. Beahm and C. F. Weber expanded the studies of chemistry and transport of iodine in reactor containment that was started in the SASA program. They were supported by W. E. Shockley, S. J. Wisbey (U.K. visiting scientist), and Y.-M. Wang (visiting scientist from Taiwan). Additional work was performed by M. Brown (U.K.) and T. S. Kress (Engineering Technology Division). This led to summarizing the iodine behavior work in the computer program TRENDS (Fig. 4.21).

E. C. Beahm has become the NRC's iodine behavior expert and is currently evaluating the chemical forms of iodine in containment spaces for a revision of the *NRC Regulatory Guides,* the safety guidelines for reactor design and siting. The TRENDS models for iodine chemical forms and other behavior in containment that have been developed by Beahm et al. include the effect of radiation on iodine in water pools, the effect of pH, calculation of pH, the effects of nitric acid formed in water by radiation, the effect on pH of HCl from certain decomposed plastics, and the effects of

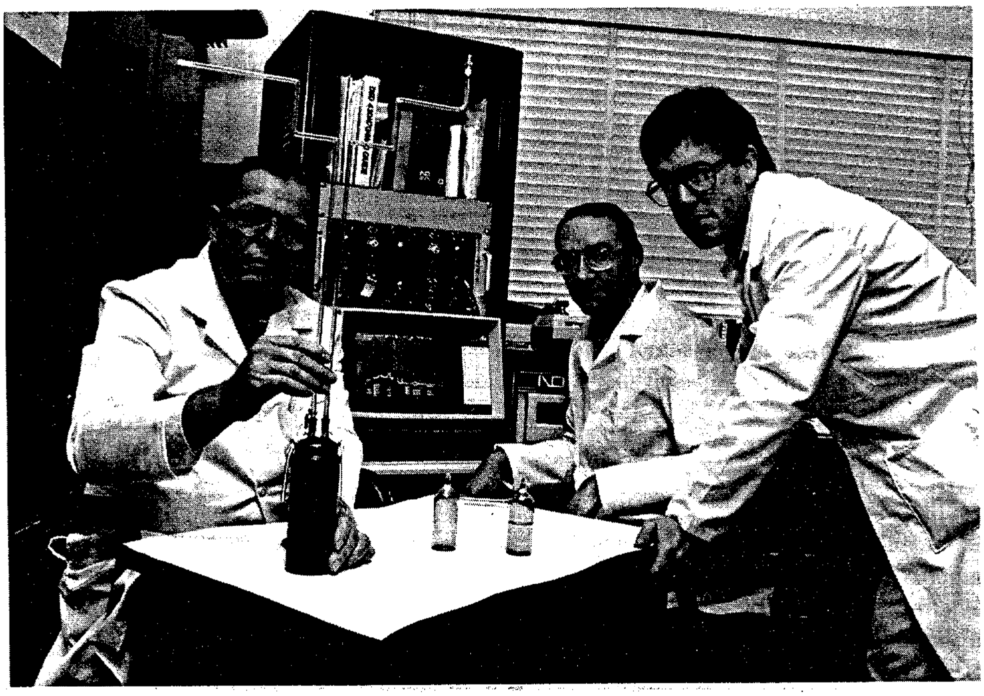


Fig. 4.21. Bill Shockley (left), Ed Beahm (center), and Simon Wisbey (guest scientist from AERE, Harwell, England) prepare an experiment to study the formation of volatile forms of fission product iodine under conditions that are predicted to occur in the containment building of a nuclear reactor that has undergone a severe accident. Because intense radiation fields would be present under such circumstances, these experiments are conducted in a ^{60}Co irradiator to study the effects of gamma irradiation. This work was performed in Chem Tech as part of ORNL's NRC program.

silver and temperature. Another iodine volatility study, "Iodine Leakage from Steam Generator Tubing," was performed by S. D. Clinton, E. C. Beahm, and W. E. Shockley (Fig. 4.22).

4.8.14 Aerosol Studies

G. W. Parker began aerosol studies for LMFBR safety in 1974 by forming high-concentration UO_2 aerosols using a metallurgical cold hearth furnace especially designed for the purpose. Parker also used a plasma torch for aerosol generation. George, Minton Kelley, and Jim Rochelle (I&C Division) performed another LMFBR aerosol study by developing a capacitor-discharge UO_2 pellet vaporizer. One of the secrets to this very successful project was electrical preheating of the pellet, which increased the electrical conductivity for the capacitor discharge.

LWR aerosol studies were started in 1982 by A. L. Wright and W. L. Pattison. They used Parker's plasma torch method to generate LWR accident-type aerosols (Fig. 4.23). They studied both settling/plateout of high-concentration aerosols flowing at low velocity and the resuspension of deposited aerosols. They found much higher deposition velocities than predicted by the NRC's TRAP-MELT CODE.

Roger Spence and Frank Dyer (Analytical Chemistry Division) began studying the sorption and reaction of CsI and CsOH with a variety of LWR accident aerosol components. These reactions could take place in the primary system at temperatures above 500°C. They found interesting reactions, but all the aerosol programs were terminated in late 1986 or early 1987 just as they were producing interesting and useful results.

4.8.15 Response to the Chernobyl Reactor Accident

The Chernobyl accident occurred April 26, 1986. R. A. Lorenz and Toshiyuki Yamashita correlated the gamma count results from several hundred air, fallout, and smear samples of radioactivity collected at locations all over the world. In combination with ORIGEN inventory estimates, they used the ^{134}Cs/^{137}Cs ratio to

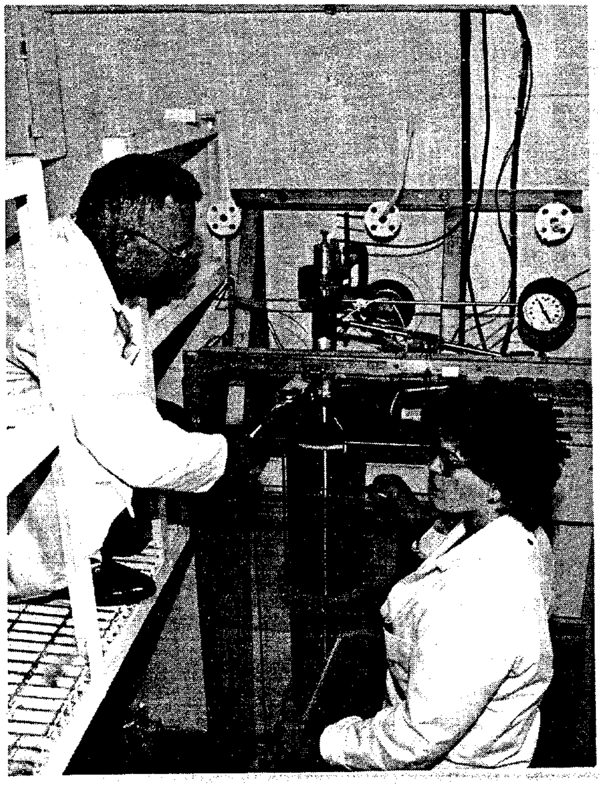


Fig. 4.22. Barn Clinton (left) and Cathy Simmons are **preparing** for an experiment that **will** determine the partitioning of iodine between the vapor and liquid within the environment of a simulated pressurized water reactor steam generator as it experiences a break in one of the tubes. The partitioning is **being** studied as a function of coolant **pH, iodine concentration,** and iodine chemical form. This work **was** performed in Chem Tech as part of **ORNL's** NRC program.

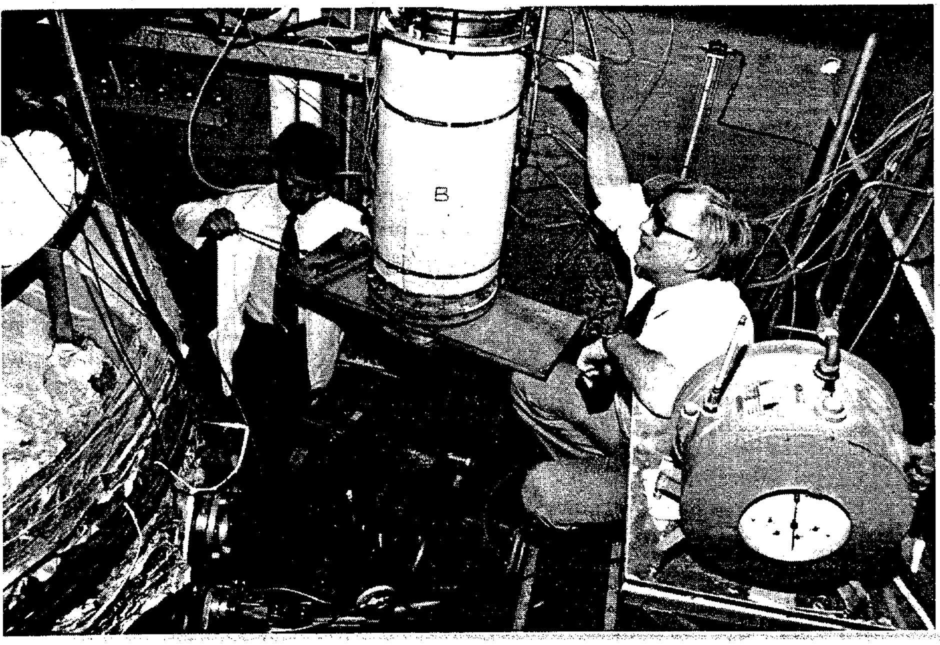


Fig. 4.23. Tony Wright (left) and Bill **Pattison** prepare for an aerosol transport experiment in equipment simulating the upper plenum portion of an LWR nuclear reactor vessel. These experiments are designed to determine whether the computer code TRAP-MELT accurately predicts the transport and deposition of metallic and metal oxide aerosols that would be generated during a nuclear reactor accident. This work was performed in Chem Tech as part of **ORNL's** NRC program.

determine average **burnup** and, more importantly, the $^{133}I/^{131}I$ ratio to determine the time of reactor shutdown and whether or not the reactor had been at **normal** power at the time of the accident. The challenge of doing this detective work was taken away about 2 weeks after the accident when the Soviets admitted to the accident and identified the reactor, the time of the accident, etc. Lorenz and Yamashita also detected increased releases of ruthenium and tellurium in material that was released 2 or 3 days after the accident, an indication that the reactor atmosphere had shifted from reducing or neutral to oxidizing. They also concluded that what appeared to be increasing releases of iodine was undoubtedly the result of the volatilization of previously deposited iodine as organic iodides.

4.8.16 HTGR Programs, 1964-l 991

Shortly after the Peach Bottom gas-cooled reactor shutdown in November 1964, R. P. Wichner, L. Fairchild, and F. F. Dyer (Analytical Chemistry Division) entered the reactor building and gamma-scanned reactor piping and components to determine fission product distribution. They continued related work until 1979 by examining Peach Bottom samplers and graphite components for fission product behavior. Bob Wichner and others calculated tritium and ^{14}C balances for the Peach Bottom Reactor.

In the early **1970s,** Bob Evans, assisted by Roy Towns, used sophisticated techniques to measure gas diffusion and permeation through various types of graphite. When a visitor arrived one day, and Roy introduced himself as Dr. Evans, he said he had to leave and turned the visitor over to his technician (the teal Dr. Bob Evans). I'm sure that Bob made a good impression as the knowledgeable "technician."

Under the direction of H. J. de Nordwall, potential accidents in **HTGRs** were studied E. L. Compere analyzed leakage paths from the reactor and the critical transport pathway to the human thyroid outside the reactor exclusion area and calculated potential doses to the thyroid. In an experimental project initiated by Ernst Hoinkis, a German visiting scientist, Morris Osborne studied the sorption and desorption of iodine onto/from HTGR materials, both in helium and under vacuum. In order to better simulate HT'GR conditions, sorption data were obtained at much lower partial pressures of iodine, $\sim 10^{-11}$ atm, than had been achieved previously. As expected, iodine sorption was shown to be proportional to iodine partial **pressure** and inversely proportional to surface temperature for a wide range of conditions. Maximum iodine covet-ages of -2 x 10^{14} and -1 x 10^{14} atoms/cm2 were found for Fe_3O_4 and Cr_2O_3, respectively: graphite cover-ages were less by a factor of -100. Of particular significance to reactor safety was the fact that small partial pressures of water vapor were found to enhance iodine desorption.

In later studies under Wichner, Dick Lorenz, Frank Dyer, and Roy Towns measured the sorption of iodine on graphite at higher temperatures (up to **800°C)** and **Osborne** measured the sorption of iodine (at 200 to **400°C)** on low-alloy steels used in HTGR steam generators. Very small amounts of surface oxide and small differences in gas chemistry were shown to strongly affect iodine sorption characteristics.

Bob Wichner and Lou Fairchild measured low-rate oxidation of graphite using a gas **chromatograph** to follow the oxidation rate. 0. K. Tallent measured the sorptivity and diffusivity of both uranium and plutonium in graphite. Bob Fellows completed this work and analyzed the materials. Jim Wilson measured the diffusion coefficients of several fission products through graphite.

In work directed by Jim **Mailen** in the 1990-9 1 period, Sam Clinton and Randy Gibson investigated the sorption of iodine on steel at low iodine concentrations. Randy and Dick Lorenz investigated the sorption **of iodine** on Inconel and cesium on low-alloy steel.

T. B. Lindemer and Ray Pearson conducted research on the chemical design of HTGR fuels. One phase of their research was the "amoeba effect," the problem of fuel kernels migrating inside of the small coated fuel particles. This research led to a quantitative description of oxide and carbide fuel migration, a description that permitted a matching of fuel behavior to reactor operating characteristics. Also, they found that unit-radiated fuel particles doped with synthetic fission products matched real fuel particle behavior and used them to explore the chemistry of UO_2/UC_2/**fission** product mixtures typical of different **burnups.** This resulted in the improved chemical design of the UO_2-UC_2 fuel. Ted Bessman, Ed Beahm, Terry, and Charlie Culpepper performed Basic Energy Science fuel-related research with actinides including uranium, thorium, and plutonium carbides and oxides. Terry also performed original research on the high-temperature chemistry of the urania/gadolinia system and developed a unique chemical thermodynamic model, useful for predicting the effect of lanthanide fission products on UO_2.

In 1990, a cooperative effort with Metals and Ceramics Division to measure fission product release from HTGR fuels under accident conditions was begun. This project was sponsored by DOE. A graphite element furnace, capable of temperatures well above 2000°C, was purchased, modified, and installed for remote operation in Hot Cell A of Building 4501. Morris Osborne and Jack Collins, along with Jim Travis and Charlie Webster, participated in the design, installation, and operation of this effort. The irradiated fuel specimens have included both groups of individual microspheres mounted in graphite holders and fuel particle-graphite compacts that are planned for use in the HTGR Principal test parameters are temperature (1400 to 1800°C) and time (up to 1000 h) at temperature.

4.9 THE STABLE ISOTOPE ENRICHMENT PROGRAM: THE EARLY YEARS, Gene Newman

In October 1973, Floyd Culler appointed a committee to “evaluate the future strategy of the stable isotopes program at ORNL.” This committee included Don Ferguson as chairman, Stan Auerbach, Gene Lamb, and Fred Maienschein. This action was prompted by concern on the part of Laboratory management that the Isotope Program, including Stable Isotopes, Radioisotopes, and the Isotope Research Materials Laboratory in the Isotopes Division, was faced with severe funding and management problems. The committee enlisted the aid of Program Planning and Analysis (PP&A), under the direction of Bob Livingston, to provide staff support. The Chem Tech staff assigned to this project from PP&A included John Bigelow, Gene Newman, and Colin West.

The PP&A staff generated a report, transmitted to Culler by the committee in July 1974, entitled ***The Electromagnetically Separated Stable Isotopes Program at ORNL: Analysis and Recommendations.*** This report emphasized the benefits of the stable isotopes program to the then AEC, made note of the fact that the Laboratory gained recognition from distributing highly enriched stable isotopes to the worldwide research community, and made recommendations for implementation by both the Laboratory and the AEC to address the business aspects of the program.

In June 1975, Laboratory management dissolved the Isotopes Division and transferred the management and operation of the Stable Isotopes Program to the Chemical Technology Division, Radioisotopes Production and the Isotope Sales Office to the Operations Division, and the Isotopes Research Materials Laboratory to the Solid State Division. Chem Tech elected to create a new Stable Isotopes Section, appointed Gene Newman as section head, and nurtured the program through the perennial budget problems and changes directed toward establishing a viable business-oriented operation.

Initially the Isotope Section reported to Ray Wymer in his role as one of the two associate directors of the division. When Gene McNeese rejoined the division as the third associate director, the section was transferred to his area, and when Gene accepted another assignment at ORNL, the section once again reported to Wymer. Chem Tech reassigned activities involving the fixation of waste in concrete and borehole plugging in order to increase the R&D component of the Isotope Section at the Y-12 site.

One of the best, and probably the last ever, isotopic enrichment of plutonium was made in the contained facility during this time. Approximately 1 kg of plutonium with ^{242}Pu assay of almost 95% was dropped off at Oak Ridge from a larger shipment on its way to Los Alamos. Joe Tracy and H. T. Milton reactivated the actinide facility, nursed two of the calutrons back to operational status after a stand down of at least 5 years, and designed and built the ion sources and receivers. George Banic tweaked and tuned the high-voltage and magnet power supplies, and Bill Bell and Al Veech established the operational parameters. Jim Barker and Frank Scheitlin reactivated the actinide chemistry facility and did the feed preparation, product recovery, and recycle chemistry. The product recovered from running 1.6 kg feed was more than 250 g of ^{242}Pu with an isotopic purity of 99.932%. Portions of this material have been characterized and certified as mass-spectrometry standards.

One of the continuing challenges of the section was to prepare unique samples for research. Several interesting projects were accomplished, and one notable effort was the preparation of a

1-kg bar of **99.9+%** enriched ^{56}Fe for use as a neutron **filter** at the Brookhaven National Laboratory reactor. Purifying the material, converting it to metallic form, and then casting the bar without reintroducing any chemical contaminants sounds simple, but molten iron is almost a universal solvent. Hugh Caudill worked out a procedure that kept the impurity level to the few ppm range, and the bar was sent off to Brookhaven.

Another unique research material was prepared by Joe Paehler. The request was **from** the National Bureau of Standards for 10 μCi of ^{229}Th (the daughter of ^{233}U) with less than 10% activity of the other thorium isotopes. Starting with a 100-g cow of enriched ^{233}U containing less **than** 18 ppb of ^{232}U, Joe extracted and purified the daughter products for the National Bureau of Standards, who, in turn, characterized the sample as a standard.

The nature of the business continued to evolve from one of producing enriched isotopes of all the elements for the research community to use primarily in the measurement of nuclear properties to answering the demand generated by the nuclear medical health-care delivery field. As the pattern of demand changed from one of small quantities of all of the approximately 230 isotopes in the periodic table to one of large quantities of a very few isotopes, so did the funding and the R&D picture.

Although the budget problems were never completely solved, due mainly to the unique accounting policies associated with return of only a fraction of the isotopes sales credits to offset operating expense, the Chem Tech management did succeed in reestablishing the fact that operation of the Calutron Enrichment Program was beneficial to the research mission of the DOE and to the national nuclear medical health-care delivery sector. Furthermore, the morale of the personnel continued to improve under the leadership of the division.

In 1983, Laboratory management again reviewed the operation of the various components of the total Isotopes Program. The conclusion was that each of the activities had benefitted **from** the initial fragmentation and that, in the interests of unified program management, it was time to recombine the program in one division. The Operations Division was selected as the new home, and the Stable Isotope Program was transferred out of Chem Tech. The Concrete Fixation **Program** remained within Chem Tech but was transferred to another section, and the Isotopes Section was eliminated.

July 1988 brought another change in the stewardship of the Isotope Program. The Operations Division was eliminated, and this time the responsibility for the entire Isotope Program was transferred to Chem Tech.

4.10 THE TRANSURANIUM ELEMENT (TRU) PROCESSING PROGRAM IN THE CHEMICAL TECHNOLOGY DIVISION,

John Bigelow

On October 27.1957, Dr. Glenn T. **Seaborg** wrote to Lewis L. Strauss, then Chairman of the AEC, to stress the need for a very high flux reactor and a program to produce berkelium, califomium, and einsteinium in substantial (multimilligram) quantities. This topic was then brought up at meetings of the directors of the various national laboratories, and a plan was developed whereby new facilities specifically designed for this purpose would be constructed at Oak Ridge (Fig. 4.24) and the resulting products would be allocated by the **AEC** to the participating laboratories based on the advice of a Transplutonium Program Committee.

ORNL was undoubtedly selected because of the many years of demonstrated capabilities in reactor design and development, as well as in the development of many chemical processes and the safe processing of reactor fuels and radioisotopes on the pilot-plant scale. The dedication of the transuranium research laboratories was attended by Glenn **Seaborg.** (Fig. 4.25)

During the design and construction of the Oak Ridge facilities, hvo lots of ^{242}Pu **feed** were prepared at Savannah River by long-term irradiation of approximately **10-kg** batches of ^{239}Pu, and ***then*** an extra push was given to the program by irradiating some of the ^{242}Pu at Savannah River to provide feed to the processing facility before the HFIR could bring targets to maturity. Thus, initial criticality at the HFIR occurred in 1965; however, after a substantial testing program, the first targets were loaded into Cycle 3 and operated at 90 MW on August 7.1966. The first hot **processing** campaign was completed in the Transuranium Processing Facility (now REDC) in November 1966 on prototype **HFIR** targets irradiated at Savannah River. That

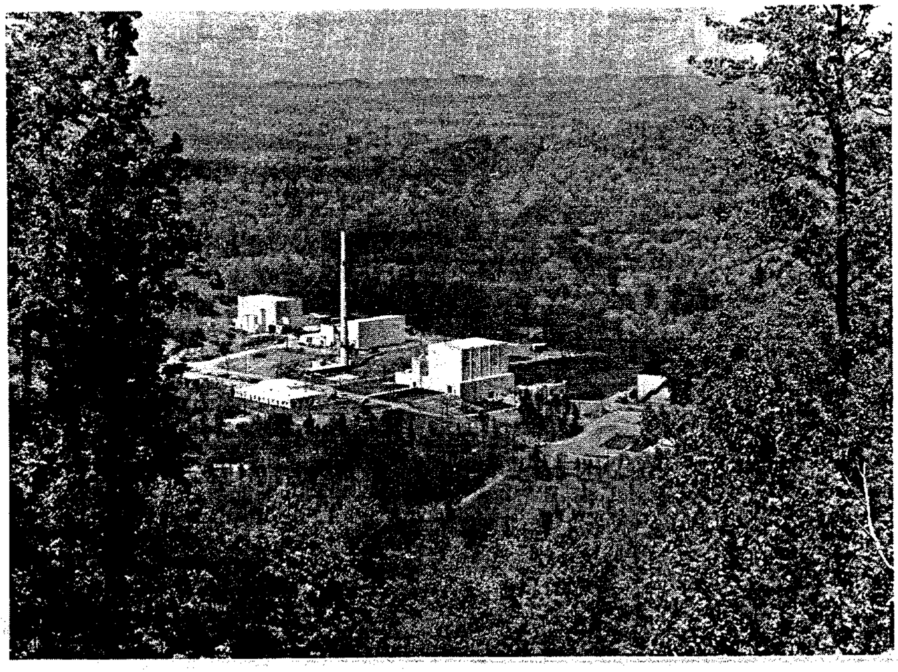
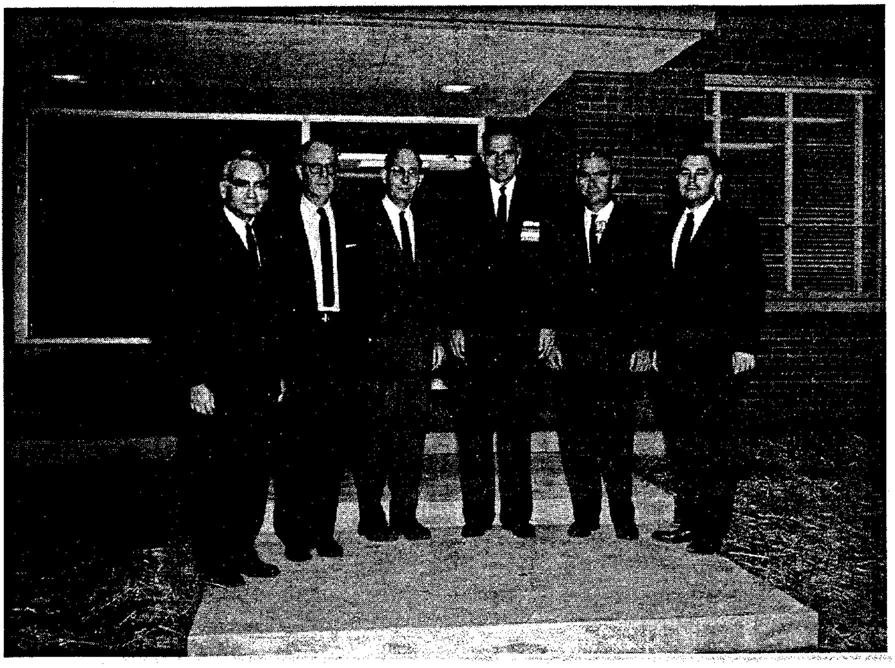


Fig. 4.24. In this photograph of the **TRU** Complex, the High-Flux Isotope Reactor Is located in the building at the right center **with** the reactor's cooling tower to the far right; the Transuranium Processing Plant is located in the center behind the smokestack; the **HFIR** office building is located In the **left** foreground; and the Thorium-Uranium Fuel Cycle Development Facility is located at the extreme left.



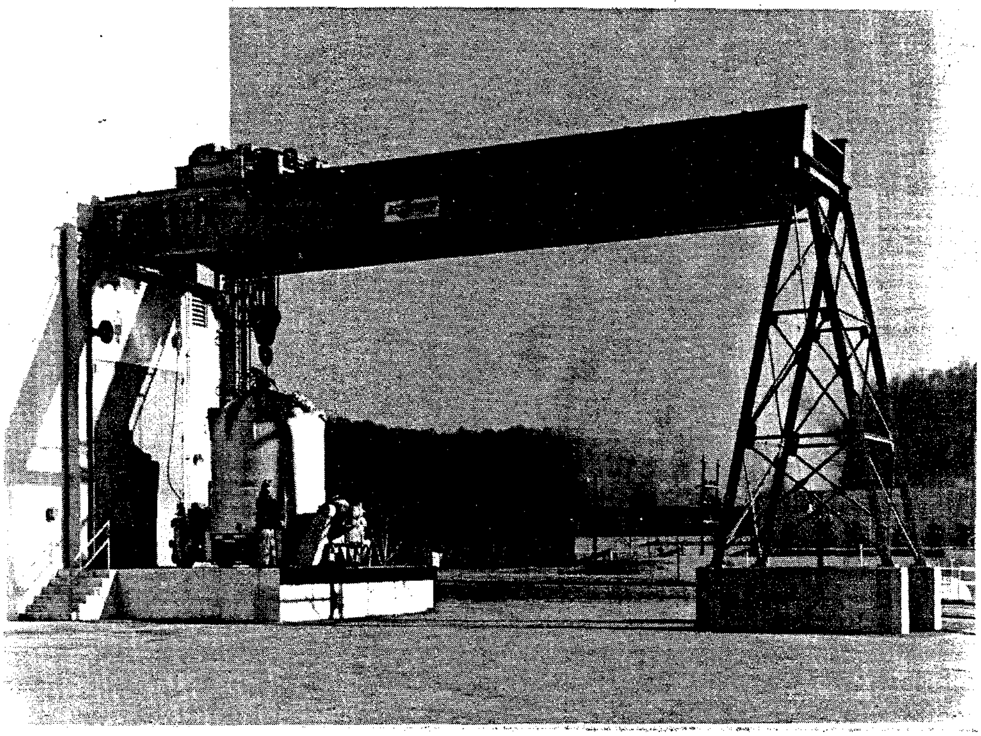
Flg. 4.25. Glenn **Seaborg, discoverer** of plutonium and Chairman of the Atomic Energy Commission, assisted in the dedication of the newly constructed transumnlum chemistry research facilities. Pictured (left to right) are Alvin Weinberg, Director of ORNL; George **E.** Boyd, Associate Director of ORNL; **Ellison** Taylor, Director of the Chemistry Division; Chairman **Seaborg**; Don Ferguson, Director of the Chemical Technology Division; and Lou Keller, Manager of the TRU laboratory facility.

campaign yielded about 180 g of ^{252}Cf (Fig. 4.26).

Savannah River continued to boost the National Transplutonium Element Program by producing 3 kg of ^{244}Cm for the Space Power Program and then embarking on a Califomium-I Campaign to produce multigram quantities of ^{252}Cf for a Market Evaluation Program. As it turned out, the market for ^{252}Cf did not develop as rapidly as anticipated, the Multipurpose Processing Facility (MPPF) being built at Savannah River was never completed, and about one-third of the irradiated materials were shipped to ORNL for processing. The 720 mg of ^{252}Cf that ORNL recovered was returned to Savannah River for the Market Evaluation Program, but eventually ORNL was allowed to include the recovered curium fraction (rich in ^{246}Cm and ^{248}Cm) in feed to the HFIR. This advanced recycle feed allowed ORNL to reach and sustain a level of production approaching 0.5 g/year beginning in 1972. It also allowed some time at the REDC to undertake other activities and some space in the HFIR target island to undertake irradiation testing activities and production of certain high-specific-activity radioisotopes.

4.10.1 The Earty Development Years

The chemical processes for separating and purifying the transplutonium elements had been demonstrated on the microscale by the scientists involved in the initial production and discovery of these elements and their various isotopes. These processes mostly involved ion exchange using various complexing ions to hold the materials in solution and help to differentiate between adjacent elements with nearly identical chemical properties. A group under Rex Leuze in the Chemical Development Section was assigned the responsibility for scaling up these techniques to the macro scale involving grams of curium and multimilligrams of transcurium elements. This group already had experience in handling ^{233}U, ^{235}U, ^{239}Pu, and ^{241}Am on the gram scale and believed that they could handle ^{244}Cm that way also, with some unit shielding in their glove boxes. Consequently, a super glove box line was designed for use in water-filled caves (there was a small concrete shielded cell to dissolve the targets and remove the fission products). However, as the design work proceeded, it became apparent that this scheme was rather marginal. Indeed, when ^{254}Cf was first produced in a reactor and cross sections weremeasured, Alfred (Chet)



Fig, 4.26. The HFIR Target Cask and Chem Tech engineer are shown with reactor cooling towers in the background.

Chetham-Strode, leader of the Transplutonium Group in the ORNL Chemistry Division, predicted that the neutron and gamma radiation from the ^{252}Cf produced in the HFIR would. be doubled by the intense spontaneous fission rate in the ^{254}Cf that accompanied it. (Actual experience shows that at discharge about one-third as many neutrons come from the ^{254}Cf as from the ^{252}Cf.) This information was the "straw" that caused a total reevaluation of the processing plans.

The result was a 12-cell, 12-laboratory version of Building 7920 that was estimated to cost $14 million. Laboratory Director Alvin Weinberg vetoed this idea and set a ceiling of $8 million. The CTD Process Design Section team headed by Bill Unger went on a six-day week for the remainder of the summer and part of the fall to downsize the plans to the current nine-cell, eight-laboratory building. Other members of the team included Bill Burch, Orlan Yarbro, Baird Bottenfield, and John Bigelow, who had just returned from a three-year stint with the AEC. Also helping was co-op student Ben Crump.

The Process Design Section advised the Chemical Development Section (principally chemist Rus Baybarz and his technician Chick Wiggins) to concentrate on the development of solvent extraction processes to isolate and purify the transplutonium elements. The ratiqnale was that in ion exchange, the active site is a specific functional group bound to a polystyrene matrix. The nuclide-emitting radiation is chemically bound to that active site: thus, the radiation source is precisely located to do the most damage to the chemical structure needed to do the processing. In solvent extraction, the radiation source is dissolved in the organic phase and "theoretically" this dilutes the effect of the radiation and allows processing at a much higher radiation density. Accordingly, Baybarz developed the TRAMEX and HEPEX processes for separating the heavy elements, respectively, from the rare-earth fission products and from each other. The layout of Bldg. 7920 was based on these processes.

With the basic process established and with the advent of congressional approval of the line items for the reactor and processing plant, development work for these facilities began in earnest. The Unit. Operations Section began to study designs of pulse columns that could be mounted on the new equipment racks. The Process Design Section, working with ORNL Engineering, developed a complete armory of modular equipment (e.g., pumps, valves, samplers, and filters) that could be easily mounted and dismounted from these racks using master-slave manipulators. However, along with the equipment rack concept, it was necessary to devise a series of gadgets that facilitated the entrance and removal of materials from the cell bank while maintaining positive alpha containment. These included the transfer case, the inter-cell conveyor system, the plastic bucket sealing system for solid-waste removal, and the concrete waste cask to accumulate plastic buckets and protect them during transfer to the burial ground.

The Chemical Development Section continued to work on the flow sheets, studying the parameters that affected the process, and providing the engineers with data to optimize the curium separation. This included a crude, small-scale batch countercurrent extraction test of the TRAMEX process at full activity level-10 W/L.

It was decided to build a test facility in the Building 4507 hot cells that, in addition to testing the processes, would provide the function of purifying some of the ^{243}Am and ^{244}Cm that had been produced at Savannah River as co-products with the ^{242}Pu destined for the initial feed to the HFIR. These materials would be the first multigram-scale distributions to the heavy-element -community. However, before the Curium Recovery Facility (CRF) got into operation, a new mission developed: to back up the private operation to recover ^{242}Cm for the Space Power Program. It was necessary to upgrade the CRF to achieve a higher degree of reliable operation to support this latter mission. In fact, as the time approached, the ORNL facility became the prime recovery facility, and the Martin Company Hot Cells at Quehanna, Pennsylvania, were shut down.

The redesign and installation of the upgraded CRF equipment was under the supervision of Frank Peishel. Frank had come to ORNL as a draftsman with the Foster Wheeler Corporation architect-engineers for the ICPP. After returning to ORNL, he attended UT night school and took a year's leave of absence to complete his bachelor's degree in mechanical engineering. The CRF was Frank's first major project after becoming a professional engineer. In December 1963, the first shipment of raffinate solution was received from Savannah River and the CRF went hot. Vic Vaughen was in charge of the operation, which started out as a group-within the Chemical Development Section but later was transferred to the Pilot Plant Section. Other personnel attached to the project included John Bigelow as data analyst, Fred Chattin, Tom McDuffee, Fred Kappelmann, and Bill Whitson as shift engineers and John Brock, Bill Lindsey, Bill Bryan, and Bill Bostic (and others) as shift technicians.

Over the next two years, several batches of ^{243}Am and ^{244}Cm were isolated and purified and distributed to various users. Then a series of irradiated ^{241}Am slugs was processed to recover ^{242}Cm for materials compatibility studies and an environmental test of a prototype space power supply. A typical 6-g batch would generate over 700 W of radiation power. This stuff required very careful handling!

Besides producing useful products, the CRF served well as a pilot plant for several of the processes that were being installed in Building 7920, the Transuranium Processing Plant, or TRU as it was known in those days. The staff learned of several problems resulting from corrosion, impurities in drum-quantity reagents, and the problems of operating small mixer-settlers remotely. They also learned that high-radiation power densities did not have a significant first-order effect on solvent extraction processes, but they did cause loss of acid by radiolysis and resulted in corrosion and impurities. This information resulted in two significant design changes at TRU: Hastelloy C, which contained molybdenum, a bad actor, was replaced by Zircaloy-2 in all process applications but was retained for use in waste tanks. Later some process vessels were upgraded again to tantalum when it was discovered that Zircaloy-2 corroded rapidly upon coming in contact with strong HCl at high power densities.

During the last few campaigns to produce ^{242}Cm, operational support from Building 7920 (TRU) was required to provide solution makeup and off-shift delivery services. This was necessary because some of the CRF crew had been taken away to form the nucleus of the TRU operating group. The final ^{242}Cm campaign was completed in May of 1966, at which time additional personnel moved over to TRU, leaving only a day crew for design and development.

4.10.2 Start-up of Building 7920 (TRU)

The organization of an operating staff for TRU began in the fall of 1965 with a formal, three-week training program commencing on October 18, 1965. This group was under the direction of Bill Burch, with Orlan Yarbro serving as Chief of Operations. Other personnel assigned to the project included John Bigelow, Fred Chattin, Emory Collins, Waldo Evans, Les King, Joe Matherne, and Jim Turley. John Van Cleve, from the Metals and Ceramics Division, was to be in charge of target fabrication. The operating group moved into the new facility on November 1965 and began cold checkout of equipment and calibration of tanks and flowmeters. Some of the development laboratories were placed in radioactive service in January 1966, and the first activity was introduced into the hot cells in June 1966. The first hot processing campaign was completed in November 1966 on prototype HFIR targets that had been irradiated at Savannah River. About 180 g of ^{252}Cf (the yardstick for heavy-element production) had been recovered in this inaugural campaign at ORNL. After final purification by Russ Baybarz, this material was distributed to Argonne National Laboratory and Savannah River Laboratory, as recommended by the Transplutonium Program Committee. Figure 4.27 shows the cell area of the Transuranium Processing Plant, and Fig. 4.28 shows operations being conducted within the cell behind a 54-in.-thick window.

In that first campaign, and indeed many that followed, the staff had to face and solve a host of problems. Some were chemical, some mechanical. Of the chemical problems, most were related to the unexpected behavior of the 92 “regular” elements. The transplutonium elements did what was expected, provided the common elements were kept under control. One exception was berkelium, but in the presence of holding reductants, it could be maintained in the Bk(III) state. In the mechanical arena, the same sort of situation applied. Gadgets had to be modified and some new ones invented, but the basic equipment rack concept, the equipment transfer case, and indeed the idea of total replaceability engineered into the building worked beautifully. As shakedown continued and processing of HFIR-irradiated materials began, our output increased to milligrams, tens of milligrams, and eventualiy hundreds of milligrams of ^{252}Cf per campaign.

While this was going on, the HFIR began having problems with the target rods. They were beginning to split open, although, fortunately, relatively small fractions of their contents were being released to the primary coolant. A very intensive effort to determine the cause ensued.

Fig. 4.27. The cell area of the Transuranium Processing Plant. The cells at the right are used in preparing target rods for Irradiation and for separation of elements produced by irradiation. The panel boards at the left control the processes.

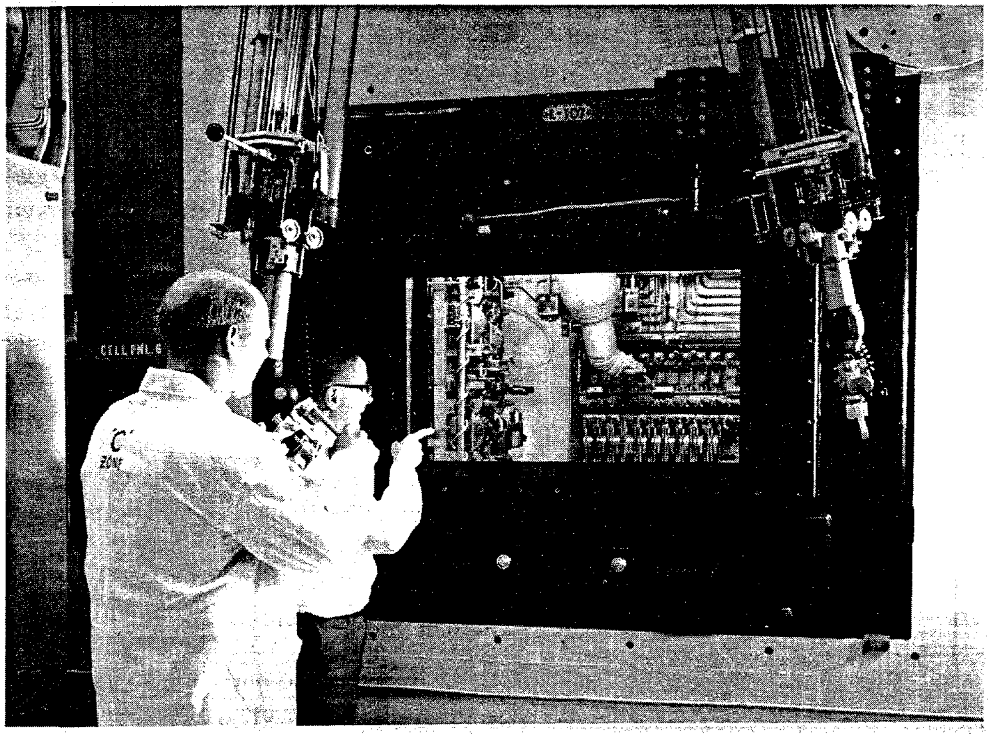


Fig. 4.28. **Orlan Yarbro** and a co-worker conduct cell pperations **with** slaved manipulators behind the **54-in.-thick cell** window.

Apparently, the aluminum alloy cladding had embrittled at the very high fluence (flux multiplied by time) experienced by these targets. Seventeen had been irradiated in a high-flux demonstration run at Savannah River and then, after inspection and new shrouds at TRU, were among the first set of targets irradiated in the HFIR. By the summer of 1967, they had received a greater neutron exposure than any other aluminum samples yet studied. New mechanisms for embrittlement were uncovered: aluminum was being transmuted to silicon, nickel (particularly ^{59}Ni, itself an activation product) was undergoing an (n,α) reaction to produce significant amounts of helium, and the high fast-neutron flux was causing a large number of knock-on protons to end up in the outer layers of the cladding. All three of these impurities exceeded their solubility in aluminum, so they migrated to the grain boundaries, where they reduced the tensile strength and ductility of the aluminum. At the same time, the ^{242}Pu in the targets was being'transmuted to ^{243}Am, ^{244}Cm, and ^{245}Cm, a nuclide with a very high fission-to-capture ratio. The fissioning of ^{245}Cm, augmented by lesser amounts of the other heavy nuclides present, accounted for a fissioning of over 65% of the initial heavy-metal atoms present. There simply was no room for all of these fission products inside the target assembly, and since the ductility was lost, the expanding pellets split the jackets. The solution was simple: leave more space inside the pellet by pressing to only 80% density instead of the initially specified 90%.

Fortunately the reactor operators and the AEC allowed the staff to continue the irradiation of the stock of 90% dense targets either till failure was detected or until the planned irradiation was reached. None showed the spectacular split that the first failures had shown, but most had a few hairline cracks when they were discharged. Of the subsequent recycle targets, only one showed cracks but no other indication that it was leaking.

In addition to solving the target failures, the staff learned to build targets containing ^{244}Cm. Originally, the first batches of curium oxide microspheres were made in Bldg. 4507 by Jim Hoeschele. Based on his process parameters, an equipment rack was designed, built, and installed in TRU to carry out the process. Although the process worked, a significant recycle stream (-30%) was generated, so Russ Baybarz and Joe Knauer of the Chemical Development Section came up with a resin-ldading process that produced good oxide particles with very little rework. Dick Haire made a suggestion on the firing cycle that improved the process more. This process has been used ever since. Research on development of plutonia microspheres was conducted in the TRU glove box facilities shown in Figs. 4.29 and 4.30.

4.10.3 Californium-I Campaign

The experience that Savannah River had in preparing ^{242}Pu, ^{243}Am, and ^{244}Cm for the National Transplutonium Program, plus a new Curium-I and II series of irradiations to produce 3 kg of ^{244}Cm for the Space Nuclear Programs, suggested that they could also produce ^{252}Cf in commercial quantities at an attractive price. A Market Evaluation Program had been under way at Savannah River ever since the earliest quantities of ^{252}Cf had become available and the time seemed right to produce 2 to 3 g of ^{252}Cf. Accordingly, they extended the Curium-II irradiation to make 4.5 kg of ^{244}Cm and then fabricated the excess Am-Cm and ^{242}Pu into targets for ^{252}Cf production. Because their new MPPF was not yet in operation, they asked TRU to undertake the processing of some of the irradiated Cf-I targets. ORNL built two new shipping containers on Savannah River's account and also installed a new dissolver to handle some of the Cf-I outer housings. TRU campaigns and Cf-I campaigns were alternated, and by June 1973,720 mg of ^{252}Cf for Savannah River and 571 mg for the national heavy-element research program had been recovered. As a bonus, Savannah River allowed ORNL to use the high-quality intermediates (^{244}Cm, ^{246}Cm, and ^{248}Cm) recovered from the Cf-I targets to enhance the production rate in the HFIR. By then, ORNL was able to trim back the staff somewhat and maintain a production rate of about 500 mg per year. This was sufficient for the majority of needs for the research community and for a supply to Savannah River to carry on the Market Evaluation Program. Since the needs of this program were not growing at anywhere near the rate anticipated when the Cf-I campaign was initiated, plans to complete the MPPF and process the remaining 65 Cf-I outer housings were put on indefinite hold. The 65 outer housings were eventually discharged, cropped to a length too long to tit into the Q-Ball Shipping Container, and stored in the RBOF. Perhaps these housings will be shipped to ORNL for processing someday.

Fig. 4.29. Dick Haire (left) and O. K. Tallent researched development of plutonia microspheres in the glove box laboratory at TRU.

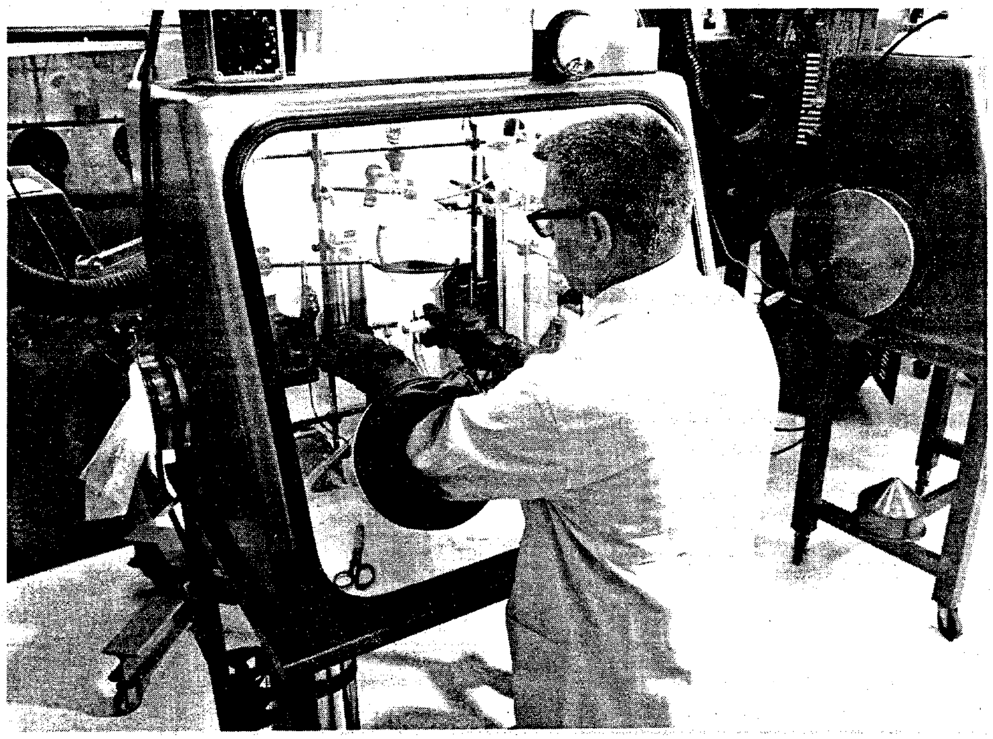


Fig. 4.30. O. K. Tallent is shown conducting a solvent extraction separation process to purify a plutonium solution for preparation of plutonium microspheres.

4.10.4 Maturity of the Transuranium Element Processing Program

The years following the Cf-I campaigns were years of fruitful maturity. Bill Burch accepted another position for Union Carbide, and Les King became manager of TRU (Fig. 4.31). New equipment racks were installed that incorporated several years of operating experience. Processes underwent little change from campaign to campaign. Transplutonium Program Committee meetings became much less acrimonious because more “goodies” were available and didn’t have to be fought over. Schedules could be prepared a year in advance and met within a few days. During this time, TRU established a substantial reputation as a reliable supplier of main-line isotopes ^{249}Bk, ^{252}Cf, ^{253}Es, and ^{257}Fm, as well as the secondary products ^{248}Cm, ^{249}Cf, and ^{254}Es. Preparation of special isotopes or special purities of standard isotopes to meet the needs of researchers was also undertaken. The details of the technology and program activities were described in a series of papers listed in Sect. 4.10.8.

This technology is available to continue a heavy-element program as long as it may be needed, provided the HFIR or Advanced Neutron Source (ANS) continues to operate and as long as the supply of makeup curium holds out.

4.10.5 Other Spin-Offs from the Heavy-Element Program

The CFRP funded a series of Purex Flow-Sheet Tests in TRU to help determine the effectiveness of this process on very high burnup fuels. Special equipment racks were installed in Cubicle 5 of Building 7920, with a set of dedicated vessels in Tank Pit 5. This special equipment was designated the Solvent Extraction Test Facility (SETF). The equipment in the cubicle included a dissolver, feed adjustment tank, feed head tanks, three each 16-stage mixer settlers and an anion exchange column for plutonium purification. Various catch tanks and evaporators were in the tank pit. In a series of campaigns, several batches of LWR fuel were processed and, as it became available, small samples of oxide fuel from EBR-II and FFTF were processed, eventually working up to 100,000 MWd/ton FFTF fuel. The tests showed that the solvent extraction flow sheet could deal with the high burnup fuel, provided that adequate feed clarification is obtained. The SETF was closed down in 1986 because of severe funding cuts in the CFR Program. The equipment in the SETF was kept because it was thought that it might come in handy someday.

Fig. 4.31. John Bigelow (left) and Les King plan the TRU facility production in order to meet the national and International needs of scientists who use its heavy element products as research tools.

The LEAP (Large Einsteinium Activation Program) was more of a very special project than a program in its own right. For several years, ORNL had been considering efforts to increase the supply of ^{254}Es (typically 4 μg per campaign) because this isotope was so valuable in doing research in the transactinide region. Following the National Academy Workshop in 1983, a group of four national laboratories collaborated on a proposal for a major thrust (LEAP) to increase the supply of ^{254}Es by an order of magnitude and focus the research using that isotope into four particular areas. The proposal requested funding for new instrumentation and equipment to do the research, as well as for out-of-pocket funding for the difficult production effort. The program was never funded, and preparatory work ran down when the HFIR was shut down in 1986.

The Mark-42 Processing Program is designed to recover ^{243}Am and ^{244}Cm from Savannah River-irradiated Mark-42 assemblies for use by Los Alamos National Laboratory (LANL) and Lawrence Livermore National Laboratory (LLNL).

However, Savannah River cannot prepare the assemblies because their MPPF is not available. This program, which supplies a significant part of the REDC funding, has been in planning since 1985 and has reached the point that one segmented assembly is in the REDC hot cells and a second segmented assembly is at Pacific Northwest Laboratory (PNL) awaiting shipment to ORNL. Portions of the first assembly have been dissolved, and processing began in the spring of 1992. It will be necessary to recover the valuable ^{242}Pu also., Researchers at ORNL are hoping that some of the ^{244}Cm will be considered excess to the program and can be applied to the Transuranium Processing Program.

The Califomium-252 Industrial Sales/Loan Program was transferred to ORNL in 1987. ORNL continues this program of supplying ^{252}Cf to industry by sale and to DOE and DOD installations and contractors by loan, which was begun at Savannah River 25 years ago. ORNL furnishes the same source forms developed and furnished by Savannah River. ORNL also carries out medical and educational loans but does not, as yet, have the capability for fabricating medical sources. At the present time, there is an annual demand for about 50 mg of ^{252}Cf for sale and about 130 mg for all other uses. Excess ^{252}Cf (if any) is stockpiled as a "cow" for production of the ^{248}Cm daughter, a valuable research isotope.

Other radioisotopes are now being produced using (in part) REDC facilities. These include ^{103}Pd and ^{63}Ni, with ^{192}Ir to be phased in shortly.

4.10.6 The Radiochemical Engineering Development Center and Its User Community

The Radiochemical Engineering Development Center (REDC) is comprised of two adjacent hot cell structures and ancillary facilities. It is operated around the clock by a staff of some 50 members of the Chemical Technology Division who are assisted by about 25 members of in-house support divisions-Analytical Chemistry, Plant and Equipment, Instrumentation and Controls, and Environmental and Health Protection. The principal programs carried out at the REDC are the Transuranium Element Processing Program, the Califomium-252 Industrial Sales/Loan Program, and the Mark 42 Processing Program.

Transplutonium Element Processing Program. The REDC is the production, storage, and distribution center for the DOE heavy-element research program. Transplutonium element targets fabricated at the REDC and irradiated in the High Flux Isotope Reactor are returned to the REDC for processing to recover and purify all transplutonium elements from americium through fermium.

The mainline products ^{243}Am, ^{244}Cm, ^{249}Bk, ^{252}Cf, ^{253}Es, and ^{257}Fm, along with the derived products ^{248}Cm, ^{249}Cf, ^{254}Es, and ^{255}Fm, are routinely supplied to 6 DOE Sites: ANL, LANL, LBL, LLNL, ORAU, and ORNL and occasionally to other laboratories in the US and overseas (4 in the past 6 years). In addition, special products can be prepared in collaboration with the researchers to suit the specific needs of their experiments. Examples of such materials furnished in the past 6 years include ^{240}Pu, ^{242}Cm, and ^{250}Cf. We have also made ^{24}Na by bombarding ^{27}Al with fast neutrons from ^{252}Cf. It should be noted that several of the DOE laboratories host collaborative research activities with nearby educational institutions.

Research using these transplutonium elements is conducted in the following fields:

- Nuclear Reactions and Synthesis of New Species
- Nuclear Properties of the Transplutonium Elements
- Chemical Properties in Solution of the Elements Americium through Fermium
- Spectroscopy of the Heavy Actinide Elements
- Solid-State/Magnetic Properties
- Industrial and Other Applications
- Biomedical Applications

In a number of the above areas, significant progress has been made in recent years. Among the unexpected and exciting findings and new capabilities are:

- The discovery of bimodal fission in some nuclides with $Z \geq 100$;
- The longer than predicted spontaneous fission half-lives in some relatively neutron-rich nuclides;
- Chemical studies of Lr, element 104, and element 105 by one-atom-at-a-time techniques;
- Formation of Bk(II) by pulse radiolysis of Bk(III) solutions;
- Development of a technique for detecting Am^{3+} at a concentration level of 10^{-10} M;
- Elucidation of the photophysics of Bk^{4+} and Cm^{4+}.

One of the products listed above (^{252}Cf) is a rather poor subject for research because of its intense neutron emission. However, for this same reason, the material has a great many applications as discussed in the next section. In the years since the program began, the Transplutonium Element Processing Program has provided to the Califomium-252 Industrial Sales/Loan Program over 2 g of ^{252}Cf (out of the -8 g produced).

Californium-252 Industrial Sales/Loan Program. Californium-252 and ^{252}Cf neutron sources have been made available to DOE and other U.S. Government agencies since 1967 as a coproduct of the DOE heavy-element production facilities of the High Flux Isotope Reactor (HFIR) and the Radiochemical Engineering Development Center (REDC) at ORNL. A program to evaluate the market potential of ^{252}Cf, based on its value as an intense neutron source, was established in 1968 at the Savannah River Site. The Market Evaluation Program continued until 1978, when a ^{252}Cf Sales/Loan Program and a Medical/University Loan Program were instituted to continue supplying ^{252}Cf to those sectors that had found it useful. In 1987, the DOE transferred responsibility for these programs to ORNL, where they were integrated into the activities of the REDC. Special neutron source forms for medical applications will continue to be fabricated at the Savannah River Laboratory.

As of June 30, 1991, there were 102 formal loan agreements in force with 71 institutions covering 239 neutron sources containing 227.3 mg of ^{252}Cf. The 7 1 institutions consist of 17 DOE units or integrated contractors, 9 U.S. Government agencies, 43 educational and medical institutions (including 1 in Japan), and 2 private research institutions that do contract research and development for various federal agencies.

In addition to the above formal loans, there were in earlier years a number of ^{252}Cf neutron sources furnished to DOE integrated contractors on a “product voucher” basis without formal loan agreements. We have identified 28 such sources containing 2.5 mg of ^{252}Cf manufactured at ORNL and delivered to 11 different sites. Similarly, we have so far identified 34 such sources containing 32.6 mg manufactured at SRL and delivered to 11 different sites. We have begun the process of “reclaiming” these sources where practical and requesting their return to ORNL so that they can be made available to other users under the Califomium-252 Sales/Loan Program.

The 102 loan agreements are broken down as follows: 7 for medical research or treatment, 39 for classroom instruction or demonstration, and the remaining 56 for research and development and/or industrial-type applications.

DOE Production and Military Applications of ^{252}Cf Neutron Sources. Neutron sources are on loan to 17 DOE or DOE contractor facilities and to 8 military installations. A large source has been used at Mound Laboratory for 100% nondestructive inspection of explosive fill in detonators since 1971; a replacement source for this purpose, containing 50 mg of ^{252}Cf, was shipped in October 1989. In addition, the Savannah River production reactors require ^{252}Cf sources for startup; four 10-mg sources were furnished in 1990.

Sources have been used for neutron gauging of reactor components at the Hanford N Reactor for burnable poison content, for neutron activation analysis at Savannah River, and for assay of high-level waste glass at Westinghouse Hanford Company.

Systems designed by Los Alamos for assaying “TRU” waste require a ^{252}Cf neutron source. Such systems are in operation at Los Alamos National Laboratory, ORNL, Portsmouth Gaseous Diffusion Plant, Savannah River Site, and WINCO.

Neutron radiography of F- 111 and F- 15 military aircraft at McClellan Air Force Base serves as a diagnostic tool for locating areas of corrosion of the aluminum honeycomb and debonding of composites. The facility (which includes an X-ray bay as well as the neutron bay) was completed in 1988 and stocked with an initial loading of 108 mg of ^{252}Cf. After a thorough checkout of the system, routine operations began in January 1990. The operations are going very well and the Navy has developed an interest in the procedure. They are negotiating with the Air Force to get their F-14s inspected at McClellan and are interested in acquiring a comparable facility of their own on the east coast. The first two replacement sources for McClellan AFB were delivered in October of 1990 and the second two are scheduled for January 1992.

The Naval Ocean Systems Center uses ^{252}Cf as a substitute reactor, since test reactor facilities are becoming very scarce. The neutron fields are used to expose infrared devices to test the resistance of those devices to neutron damage. The initial loading was 190 mg of ^{252}Cf; the first 50 mg replacement source has now been ordered.

Medical and Health-related Applications of ^{252}Cf Neutron Sources. Neutron therapy followed by X-my or gamma ray therapy has been shown to be particularly effective in the treatment of advanced, hypoxic tumors of the cervix. Altogether, six hundred fifty eight patients have been treated at the University of Kentucky Medical Center for this and other types of cancer. Survival rates have been significantly improved over that for conventional radiotherapy. Proposals have been submitted to NCI to fund development of boron neutron-capture therapy for malignant gliomas using a ^{252}Cf source. Sources are also being developed as an ***in vivo*** neutron activation tool for ultimate clinical use in diagnosis and monitoring of several medical conditions involving abnormal quantities of lithium, aluminum, or nitrogen.

The Food and Drug Administration has ordered 200 mg of ^{252}Cf to be used as a neutron activation source for determining the sodium content of certain foodstuffs. This technique would allow them to simultaneously detect tie presence of toxic heavy metals, such as arsenic or mercury.

Commercial Applications of ^{252}Cf Neutron Sources. Fit used as a reactor startup source in 1973, ^{252}Cf is currently the industry standard for this application. About 50% of ^{252}Cf is sold for this purpose. The second major use is for 100% nondestructive examination of power reactor fuel rods. This is both a quality control tool and a safeguards measure.

In some applications where reactors are not available, ^{252}Cf is used for neutron activation analysis. No other single analytical method gives as complete a profile of elemental composition. Instrument calibration, dosimetry studies, and industrial gauging also utilize about 10% of the ^{252}Cf sold. The fact that neutron fields generated by ^{252}Cf sources are now dosimetry standards is significant. DOE as well as civilian facilities are now setting up calibration ranges using ^{252}Cf neutron sources.

A new and exciting application is the thermal neutron analysis (TNA) technique for detecting hidden explosives in air passengers' luggage. Monitors have been installed at New York (JFK), San Francisco, Los Angeles, Miami, Gatwick and Frankfurt, while two have been installed in air freight centers in San Diego. The unit at San Francisco was unharmed by the earthquake in the fall of 1989.

Mark 42 Processing Program. The REDC has been asked to process a number (-10) of Mark 42 assemblies that were irradiated at the Savannah River Site (SRS) as part of a larger program for the Los Alamos National Laboratory. The reason for processing these assemblies at ORNL is to permit recovery of -100-g amounts of ^{243}Am and ^{244}Cm from the irradiated targets. Facilities at SRS are not set up to accomplish this task. The recovered ^{243}Am and some of the ^{244}Cm will be shipped to Los Alamos. By using these materials for diagnostic purposes, the nuclear chemistry staffs of Los Alamos and Lawrence Livermore National Laboratories will be able to derive mote information from each weapons test than would otherwise be possible.

The first of these assemblies is now at the REDC. Processing will begin upon completion of the current Transuranium Element Processing Campaign. The unused portion of the ^{244}Cm may become available for use as makeup feed for heavy element production in the HFIR, thus benetitting the Transuranium Element Processing Program.

4.10.7 The Future of REDC

Bob Wham took over from Les King as Manager of the REDC in March 1991. Les remained as associate section head with responsibilities for environmental, safety, and health documentation. The personnel roster has increased by about 50% to cover all responsibilities in this area. The DOE Office of Energy Research is continuing to bear these increases in cost while oversight of the operations is now under the Assistant Secretary for Nuclear Energy. The REDC is on track to meet the new requirements of DOE for excellence in operation and continues to have a mission in the preparation of radioisotopes to help meet the nation's needs, including some that are not heavy elements. It is expected the effort on the former to grow, but hopefully not at the expense of the latter.

4.10.8 Bibliography

The details of Chem Tech technology and transuranium processing program activities were described in the following series of papers:

J. E. Bigelow, B. L. Corbett, L. J. King, S. C. McGuire, and T. M. Sims, "'Production of Transplutonium Elements in the High Flux Isotope Reactor,*' ACS Symposium Series No. 161, ***Transplutonium Elements--Production and*** Recovery, Ed. J. D. Navratil and W. W. Schulz, 1981.

L. J. King, J. E. Bigelow, and E. D. Collins, "Experience in the Separation and Purification of Transplutonium Elements in the Transuranium Processing Plant at Oak Ridge National Laboratory,'* ACS Symposium Series No. 161, ***Transplutonium Elements-Production and*** Recovery, Ed. J. D. Navratil and W. W. Schulz, 1981.

J. E. Bigelow, E. D. Collins, and L. J. King, 'The 'Cleanex' Process: A Versatile Solvent Extraction Process for Recovery and Purification of Lanthanides, Americium, and Curium," ACS Symposium Series ***No.*** 117, ***Actinide Separations,*** Ed. J. D. Navratil and W. W. Schulz, 1980.

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D. E. Benker, F. R. Chattin, E. D. Collins, J. B. Knauer, P. B. Orr, R. G. Ross, and J. T. Wiggins, "Chromatographic Cation Exchange Separation of Decigram Quantities of Califomium and Other Transplutonium Elements," ACS Symposium Series ***No.*** 161, ***Transpiutonium Elements Production and Recovery, Ed.*** J. D. Navratil and W. W. Schulz, 1981.

F. R. Chattin, D. E. Benker, M. H. Lloyd, P. B. Orr, R. G. Ross, and J. T. Wiggins, "Preparation of Curium-Americium Oxide Microspheres by Resin-Bead Loading," ACS Symposium Series ***No.*** 161, ***Transplutonium Elements-Production and Recovery, Ed.*** J. D. Navratil and W. W. Schulz, 1981.

4.11 FLUORIDE VOLATILITY PROCESSING, Bob Jolley, Wilson Pitt, Lloyd Youngblood, and Bob Hightower

The purpose of the volatility program in the United States was to develop an alternative to conventional aqueous processes for recovering uranium from spent nuclear fuels. All fluoride-volatility methods used the volatility of UF_6 to achieve a high degree of separation of the uranium from its fission products. Other advantages of volatility processes, compared with aqueous processes, were (1) the formation of a dry, highly concentrated waste, (2) the greater ease of processing certain refractory fuels, (3) fewer processing steps between the original fuel element and UF_6, and (4) increased nuclear safety. The dependence on a physical property (i.e., volatility), rather than a chemical one, made possible the recovery of uranium for return to production channels.13 Disadvantages included the following: (1) customary engineering practices were generally oriented toward aqueous processing: (2) fluorine, hydrofluoric acid, and other chemicals used in the non-aqueous fluoride volatility process can be extremely hazardous; (3) the volatility processes frequently involved the use of high operating temperatures: and (4) high temperatures combined with extremely corrosive chemicals required the use of exotic metals and unique materials of construction.

Although studied at most national laboratories, the primary emphasis in the ORNL/Chem Tech work was on a molten-salt fluoride-volatility process for use with spent zirconium-based fuels containing highly enriched uranium, The Chem Tech process progressed through all R&D stages from the laboratory in 4500N, to hot-cell-level radioactivity pilot plant in Building 4507, to unit operations studies in Building 3592, and to full-scale Fluoride Volatility Process Plant in Building 3019. The process consisted basically of three steps: (1) dissolution of the metal or oxide by hydrofluoxination in molten salt (~500°C), (2) volatilization of the uranium as UF_6 from the molten salt by fluorination, and (3) purification of the product UF_6 from contaminants by adsorption and desorption from beds of NaF and MgF_2.

Early R&D. The earliest R&D in the so-called dry fluoride process was conducted by Rex Leuze as problem leader with the assistance of a small group. Members of the group varied from time to time but included C. P. Johnston, Chuck Schilling, Bob Bennett, Brooks Graham, and A. B. Green. However, with the advent of work on Aircraft Nuclear Propulsion (ANP) and anticipated processing of the unique highly enriched fuels of the Aircraft Reactor Experiment, increased R&D became necessary. Consequently, ORNL and Chem Tech increased the level of activity in this area. In 1954 Don Ferguson assumed the role of Problem Leader of the Volatility Studies, ANP Fuel Recovery Process and was assisted by Bob Bennett, George Cathers, and Chuck Schilling.[14] In March 1955, Don Ferguson became Assistant Section Chief of the Chemical Development

Section and George Cathers became the ANP Studies Problem Leader.15

Laboratory Bench-Scale Studies. In the mid-1950s, George Cathers became the Group Leader of the Fluoride Volatility Processing Group in the Chemical Development A Section. George was principally responsible for the chemical flowsheet development that eventually led to the pilot plant and full-scale process plant work. His R&D group consisted initially of Bob Bennett, Bob Jolley, and Bob Duff. Bennett pioneered dissolution of uranium metals and fuel element components in molten salt, and Jolley initially studied the reaction of NaF with UF_6.

In a 1956, Cathers and Jolley determined that UF_6 could be desorbed thermally and reversibly from the unique NaF–UF_6 complex. Subsequently, Jolley, using plutonium tracer at the 2-ppm level, determined that PuF_6 sorbs "irreversibly" on NaF at temperatures at which UF_6 sorbs "reversibly.'* This led to a patent of the NaF volatility process for separation of UF_6 from PuF_6. Using mirrors and long-handled tongs, a heavily shielded bench-scale "pilot-plant*' in a walk-in hood (Laboratory B-25, 4500N) was constructed to test the complete engineering flow sheet from dissolution to volatilization and isolation of the decontaminated UF_6 product. The system was successfully operated by Cathers, Jolley, and Duff. Decontamination of the facility and metallurgical examination of the hydrofluorinator and fluorinator vessels provided much needed corrosion data.

With rapidly expanding responsibilities such as bench-scale studies, processing demonstrations in a walk-in hood (B-25, 4500N) and hot cell (Cell 4, 4507), and technical support for pilot plant design, the group soon expanded to include Dave Campbell, Stan Kirslis, E. C. Moncrief, Frank Soard, Calvin Shipman, and Tom Crabtree. Sidney Katz and Jim Mailen joined the volatility group in the early 1960s. Katz studied extensively the adsorption reactions of UF_6 and NaF and expanded the adsorption studies to include the hexafluorides of several chemical elements including tellurium, tungsten, technetium, and molybdenum.

Hot Cell Pilot Plant. The bench-scale studies of the process flow sheet led to the design and successful operation of a larger scale pilot plant in Hot Cell 4, Building 4507. With the leadership of Cathers, Dave Campbell was principally responsible for the design of the remotely operated facility, and Jolley was principally responsible for the operation. Crabtree, Shipman, Soard, Bennett, Kirslis, Moncrief, and Jolley worked essentially "around the clock" during many operational runs. The remote system operated flawlessly except for one hitch in which a plugged line forced some contaminated liquid through a compressed gas line (used for remote control of valves) into the control panel outside the cell. Even Don Ferguson, Section Chief of Chem Development A, assisted in the relatively minor cleanup operation. Such hot cell work was greatly assisted by the cadre of excellently trained ORNL health physicists such as Bruce Walters, Got-man Hill, Ed Kuna, and many others. After the hot-cell pilot plant work was finished, Shipman and Jolley decontaminated and disassembled the system (blacksmithing via slave manipulators-not an easy task), first remotely, then manually. Decontamination of the cell required dressing out in several layers of protective clothing, including fully self-contained breathing apparatus and, later, gas masks. The cell was cleaned in preparation for the next activity-R&D for separations process development in the fledgling TRU program led by Victor Vaughen.

Unit Operations Studies. The Unit Operations volatility studies increased dramatically in the late 1950s. In 1960 the group included Group Leader Bob Horton and Gene McNeese, R. J. McNamee, S. H. Stainker, Jack Beams, and F. N. McClain.[16] Shortly thereafter, Bob Hightower, Bob Lowrie, Wilson Pitt, Bill Woods, Vic Fowler, Bruce Hanaford, and F. G. Kilpatrick joined the Unit Ops Group.[17] Operations studies centered on (1) operation of engineering-scale molten salt dissolution, fluorination, and NaF sorber vessels in order to evaluate vessel designs: (2) determination of reaction rates: (3) development and testing of monitoring instrumentation and control systems; (4) evaluation of autoresistance-heated transfer lines and spray nozzles: and (5) evaluation of corrosion of test coupons and equipment under actual process conditions.

The studies of Gene McNeese and Jack Beams on sorption of UF_6 by NaF led to the development of a mathematical model that considered the effects of temperature, concentration of UF_6, gas flow rate, and NaF pellet characteristics. Such information proved useful in the design of the pilot plant and process flow sheet. Engineering-scale studies on the process were conducted in Building 3592 in the early sixties. Short sections of full-size zirconium- and later aluminum-clad fuel elements were dissolved in fused fluoride salts and the salt subsequently fluorinated. Fuel dissolution and fluorination rates were determined as well as corrosion rates on the proposed materials of construction. Proven monitoring and control concepts were also evaluated.[17]

Although most of the unit operations (UNOP) studies were carried out in support of the Fluoride Volatility Process (FVP) for zirconium- and aluminum-based fuels, Wilson Pitt and Vic Fowler determined the vapor liquid equilibrium of the UF_6 and NbF_5 system and the critical constants of NbF_5. This was in support of the possible use of FVP to process the niobium-based Rover (Nuclear Rocket Prototype Reactor) fuels. Bob Hightower and Wilson Pitt also attempted to develop *W* and IR photometric monitors for UF_6 in fluorinator and NaF sorber off-gas. A major problem in that effort was the tendency for UF_6 to quickly coat all window materials tested.

Process Design Activities. In the late 1950s, Bob Milford with the assistance of W. E. Dunn assumed responsibility for the fluoride volatility process in the Process Design Section. His group was shortly joined by John Ruch.[18] Milford and Ruch were principally responsible for designing the pilot plant constructed in Building 3019.

Fluoride Volatility Process Plant, Building 3019. The first fluoride volatility pilot plant was constructed and operated between 1956 and 1958 to recover fully enriched uranium from the Aircraft Reactor Experiment (ARE) fuel. The extensive operation recovered 97.93% (134 kg) uranium as UF_6 product and 1.76% uranium from the NaF trap cleanout. No fission product activity was detected in the product. Principal operating difficulties were plugging of molten salt lines and the NaF beds and, also, maintenance of a gas-tight system.[19]

Although the principal responsible engineers were Bill Carr, F. W. Miles, Dick Keely, Sid Mann, R. G. Nichol, John Ruch, Frank Browder, C. L. Whitmarsh, and John Bigelow, many others were also involved. The magnitude of this initial pilot plant effort is evidenced by the following quotation.[19]

> In a program of the magnitude of this one it is impossible to acknowledge the assistance of all persons who contributed. Such a program inherently requires, at various stages, the devoted efforts of a large percentage of the total number of workers at the Laboratory. However, even at the risk of inadvertently omitting acknowledgment of major contributions, the authors wish to recognize the efforts of the following: A. P. Litman, A. E. Goldman, and other members of the Metallurgy Division for continuing advice and assistance on corrosion studies and on construction and maintenance specifications: personnel at Battelle Memorial Institute for corrosion studies: W. A. Bush and other members of the Engineering and Mechanical Division for their work on design: C. L. Fox, J. B. Henry, B. E. Van Horn, and other members of the Engineering and Mechanical Division for their work on construction and maintenance; B. Lieberman, L. H. Chase, W. J. Greter, G. P. Hinchey, and other members of the Instrumentation and Controls Division for the design and maintenance of the instrumentation: C. E. Lamb, M. J. Gaitanis, W. R. Laing, E. I. Wyatt, L. T. Corbin, and other members of the Analytical Chemistry Division for development of analytical methods, for consultation, and for the many "rush" analyses: G. J. Nessle, F. A. Doss, J. P. Blakely, R. E. Thoma, and other members of the Reactor Chemistry Division for special analytical services and for supplying fluoride salt mixtures; R. B. Lindauer, H. K. Jackson, W. H. Lewis, and F. L. Culler for over-all direction and coordination . . . ; G. I. Cathers and his Chemical Development group; J. T. Long, M. E. Whatley, R. W. Horton, and other members of Unit Operations; R. P. Milford and his design group, and C. E. Guthrie for development, design, consultation, and direct pilot plant support work: J. E. Bigelow, F. N. Browder, R. B. Keely, S. Mann, F. W. Miles, R. G. Nicol, J. B. Ruch, and all the Co-op and Summer employees, technicians, and operators for the actual plant operations.

The 1960 Chem Tech organization chart (Appendix B) shows a large group responsible for the fluoride volatility process:[16] Dick Lindauer (Fig. 4.32). Group Leader, Bill Carr, Problem Leader, and Sid Mann, F. W. Miles, R. G. Nichol, John Ruch, C. L. Whitmarsh, and Lloyd Youngblood. Technicians and operators were H. C. Thompson, V. R. Young, Bob Duff, J. H. Gibson, M. C. Hill, W. T. Henry, Z. R. McNutt, and A. V. Wilder. The group later included engineers Fred Kitts, Bob Lowrie, Bob Shannon, and E. C. Moncrief, technician

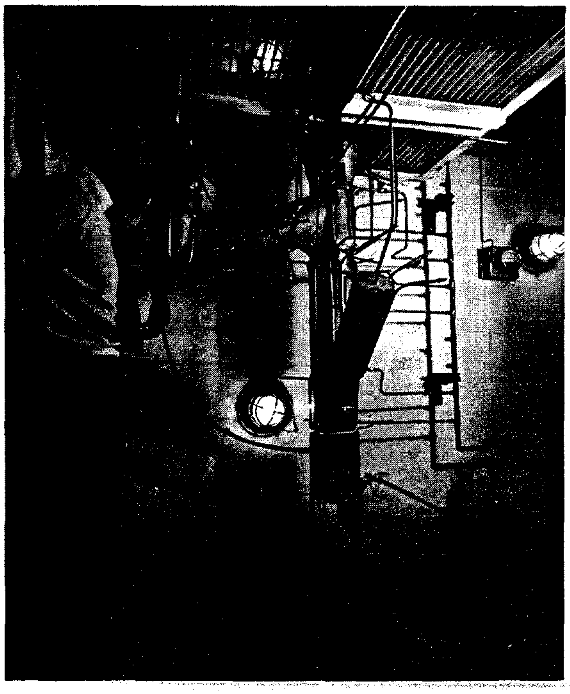

Fig. 4.32. Dick Lindauer examining the UF_6 product trap for the Fluoride Volatility Process Plant (about 1975).

M. C. Hill, and operators Ed Brantley, C. W. Boatman, J. H. Brock, W. J. Bryan, H. S. Caldwell, C. H. Jones, and G. R. Thompson.[18]

Following the ARE fuel processing, the pilot plant was completely redesigned to permit processing of zirconium fuel of the type used in navy submarines. This involved HF dissolution of the fuel in molten salt. A magnesium fluoride trap was also added to trap the volatile fluorides of technetium and neptunium. The redesigned pilot plant was operated from about 1960 to 1964. After a series of cold runs, the pilot plant processed long-cooled Nautilus submarine fuel. Generally, one or two fuel elements were processed per run, and a run required about a week to complete.

During the operation of the 3019 fluoride volatility plant, the capability for sustained operation was demonstrated with losses of only a fraction of a percent and with excellent decontamination. Decontamination factors (DFs) were among the highest ever reported for a radiochemical process, including those for three cycles of solvent extraction. DFs ranged from 10^6 to 10^{10} (e.g., a Zr-Nb DF greater than 5 x 10^{10} was achieved in one of the runs).[13]

After the campaign to process zirconium-clad fuel, the pilot plant was decontaminated and used to demonstrate the processing of aluminum-clad fuel of the type used in the LITR and ORR. Four runs were made. In the last run, an ORR fuel element was processed that had cooled for only four weeks. The processing of such short-cooled fuel represented a landmark accomplishment. DFs ranged from 10^5 to 10^8, with the exception of ^{99}Mo and ^{125}Sb, which were 36 and 500, respectively. Nonrecoverable uranium losses were only 0.4%, but the uranium product had a high radioactivity level due to the presence of ^{237}U.[19]

The report by Bill Carr, Les Ring, Fred Kitts, Tom McDuffee, and F. W. Miles acknowledges the assistance of others as follows:[20]

> R. E. Brooksbank, Chief of the Pilot Plant Section of the Chemical Technology Division, had overall responsibility for this pilot plant. R. P. Milford was responsible for coordinating the Volatility Project. Other supervisors of the Volatility Pilot Plant who made major contribution were: R. S. Lowrie, S. Mann, R. J. Shannon, and E. L. Youngblood.
>
> Many people in other parts of the Laboratory provided aid and assistance that made the operation and maintenance of this pilot plant a success; chief among these were: G. E. Pierce and R. P. Beard of the Plant and Equipment Division, W. J. Greter of the Instrumentation and Controls Division, E. I. Wyatt and C. E. Lamb of the Analytical Chemistry Division, 0, J. Smith of the Inspection Engineering Department, and C. H. Miller and W. A. McLoud of the Health Physics and Safety Division. The assistance of these, and of many other people at ORNL and at other AEC installations, is gratefully acknowledged.
>
> We also take this opportunity to express appreciation to Martha G. Stewart for her invaluable editorial assistance.

Program Closure. The molten salt fluoride process was very successful in processing and recovering uranium from fuel. However, it was very difficult to remove the plutonium from the salt, and long fluorination times gave excessively high corrosion rates. To permit processing of plutonium-containing fuel, the molten salt equipment was removed and work was begun to install a fluidized-bed volatility system based on bench-scale work done at Argonne National Laboratory. A considerable amount of equipment was installed in the Building 3019 cells during the period 1965 and 1966, but the project was canceled in 1966 when it appeared that it was not economically competitive with the Purex process for uranium and plutonium recovery.

4.12 CHEMICAL PROCESSING FOR THE MOLTEN-SALT REACTOR PROGRAM, Bob Hightower

The ORNL molten-salt reactor program was an outgrowth of the aircraft nuclear propulsion (ANP) program to make a molten-salt reactor power plant for aircraft. The first molten-salt reactor, the ARE, was operated at ORNL in 1954 as part of the ANP program. The objective of the molten-salt reactor program was the development of nuclear breeder reactors that used fluid fuels consisting of solutions of fissile and fertile materials in suitable carrier salts. Fuel for this type of reactor would be $^{233}UF_4$ dissolved in a molten mixture (solution) of ^7LiF and BeF_2. The fertile material would be ThF_4 dissolved in the same salt or in a separate salt of similar composition. The breeder reactor concept included an on-line continuous processing plant to remove the fission product poisons and to isolate the capture product ^{233}Pa, allowing it to decay to ^{233}U outside the neutron flux. This reactor concept and the associated on-line continuous processing was rich in chemistry and chemical engineering challenges, and the Chemical Technology Division played a major role in this program from the early days.

The underlying chemical processing technology for the molten-salt reactor program had been developed in the old Reactor Chemistry Division under the leadership of Warren Grimes, Charlie Baes, Charlie Barton, and others and in the Fused Salt Fluoride Volatility Program. Extrapolations of these early developments for use in continuous processing needed for a breeder reactor were operation in 1965 of a bench-scale continuous fluorinator by Gene McNeese and development of continuous on-line measurement of UF_6 in F_2 streams by gas chromatography and infrared spectroscopy of Gene McNeese and Wilson Pitt in the Unit Operations Section. A concept of fluorination of salt sprays to remove uranium was tested by Jim Mailen and George Cathers in the Chemical Development B Section. The concept for converting UF_6 directly to UF_4 in the salt phase (fuel reconstitution) by initial formation of the intermediate UF_5 and subsequent reduction by H_2 was a joint development by Gene McNeese and Chuck Scott. Experiments using rapid vapor condensation were conducted by Chuck Scott and. Warren Sisson to measure relative volatilities of the rare-earth fluorides in support of the design for a molten-salt distillation concept. These studies were refined and completed by Bob Hightower and Vic Fowler using small recirculating equilibrium stills operating at 1000°C and 0.5 mm Hg.

A major program activity through 1969 was the operation of the Molten-Salt Reactor Experiment (MSRE). The initial fuel was 0.9 mole % $^{235}UF_4$, 5% ZrF_4, 29% BeF_2, and 65% ^7LiF. After a period of operation with this fuel mixture, the $^{235}UF_4$ was separated and recovered and then replaced by $^{233}UF_4$. The processing plant for the removal of ^{235}U by fluorination of the reactor core and recovery of the ^{235}U by sorption on NaF beds was designed by Dick Lindauer of the Pilot Plant Section, who also supervised its construction, installation, and operation. The fuel loading of $^{233}UF_4$ was prepared in Building 3019 by John Parrott, John Chandler, Ralph Nicol, Bill Shannon, and Bob Shannon in the Pilot Plant Section. An experiment to test the separation of the rare-earth fission products by distillation was designed and built by Lloyd Carter of the Process Design Section. The experimental equipment was installed and operated at the MSRE by Bob Hightower, Hank Cochran, Bruce Hannaford, Gene McNeese, Vic Fowler, Ralph Payne, and Jack Beams, and it utilized remote condensate sampling equipment designed by Luther Pugh.

As the molten-salt reactor program matured, the chemical processing effort in the Chemical Technology Division grew substantially. Early processing developments for the molten-salt breeder reactorwere guided initially by Marvin Whatley, later by Gene McNeese, and still later by Bob Hightower in the Unit Operations Section. The processing concepts and technology improved steadily, and the early flow sheets based upon fluorination and distillation were replaced by fluorination, fuel reconstitution, and molten bismuth extraction for ^{233}Pa isolation and rare-earth removal. Flow sheet alternatives were synthesized and analyzed by Gene McNeese and Charles Kee using sophisticated computer codes, which they developed. The definitive process design for the reference processing concept was done by Lloyd Carter and Ed Nicholson of the Process Design Section. The hydrodynamics of bubble columns for the fluorinator was studied and defined by Jack Watson. Concepts for frozen salt corrosion protection in fluorinators were studied by Bob Hightower, Dick Lindauer, Pete Counce, and Ralph Payne. Hank Cochran, Bruce Hannaford, and Vic Fowler constructed a facility and developed engineering experience and information on extraction from molten salt mixtures into molten bismuth using packed columns and stirred contactors; these studies were later taken over by Cliff Brown. These studies were complemented by Jack Watson's fundamental work on countercurrent flow through packed columns of fluids with large density differences. Bill Schaffer of the Process Design Section designed and initiated fabrication of equipment made from molybdenum to test operations and reliability of equipment made from the preferred material of construction. Herman Weeren, Jerry Klein, and Cliff Brown conducted fundamental studies of mass transfer at liquid-metal interfaces to support equipment design and experimental interpretation. Don Kelmers and Bob Bennett of the Chemical Development B Section studied the chemistry of the conversion of UF_6 to UF_4, and Pete Counce designed and built engineering-scale processing equipment (gold lined) for scale-up and testing. Mike Bell studied oxide precipitation as an alternative to fluorination for uranium removal. Lloyd Youngblood, Clif Savage, and Jack Beams tested the Metal Transfer Process for extracting the rare earths into bismuth, back-extracting them into molten LiCl, and ultimately isolating them in bismuth for disposal. The basic chemistry of these extraction processes and other alternative process concepts, including the reductive extraction chemistry of the actinides (uranium, neptunium, plutonium, americium, and califomium) and the rare earths, was defined by studies headed by Les Ferris and his group in the Chemical Development B Section. This group included Fred Land, Mildred Sears, Chuck Schilling, Fred Smith, Jim Mailen, C. T. Thompson, Jack Lawrence (Australia), and Eduardo Nogueira (Spain).

The molten-salt reactor program was suspended from 1973 to 1974 and finally terminated in 1975 because of an ABC/ERDA decision to concentrate resources on LMFBR development.

4.13 NUCLEAR WASTE PROGRAMS, Herschel Godbee and Tom Lomenick

The Chem Tech waste treatment and disposal development program was designed to develop a comprehensive waste management system for nuclear wastes, including their final disposal, and to estimate the cost of operation. The effective, economic, safe management of radioactive effluents is a prerequisite to the natural growth of a nuclear power industry.[21]

In 1964 two methods were being developed in Chem Tech for the conversion of high-level radioactive wastes (HLW) to solids, the Potcal (pot calculation) and Potglass (fixation of wastes in glassy solids in a pot) processes.2225 The calcined or glassy solids were to be contained in 88-in.-long, 24-in.-diam stainless steel "pots" (pipes), which are designed to be closed by welding prior to shipment and ultimate disposal. Since there should be no off-gasses, future surveillance requirements during ultimate disposal were thought to be negligible. Glassy solids, rather than porous calcines, were thought to be desirable because of their higher thermal conductivity and high degree of insolubility. End products with these properties were thought to yield increased economy and safety.

Low-level radioactive wastes (LLW), such as cooling water, canal water, flood-drain water, and other low salt content waters, were treated to remove radionuclides prior to discharge to the environment. Chem Tech participated in the development of two general treatment methods for LLW: the scavenging-precipitation ion-exchange process[26,27] and the scavenging precipitation foam separation process.[28,29] An improved scavenging-precipitation ion-exchange process was successfully tested by Chem Tech, where ORNL process waste was decontaminated to less than 2% of the then allowable permissible levels for human exposure and also reduced costs approximately 15%.[25]

Intermediate-level radioactive waste (ILW) consisted of materials such as residues from processing LLW, evaporator concentrates, and concentrates from second and third solvent extraction cycles from processing nuclear fuels and other high-salt content wastes. Such wastes are categorized as either LLW or transuranic (TRU) waste under current definitions. Chem Tech studies incorporation of such wastes in asphalt and/or concrete.[28]

Chem Tech was also involved in economic analysis of HLW management and storage in ultimate disposal sites such as salt mines.[30,31] The economic analyses were in addition to the collaborative R&D with the Health Physics Division on waste-solidification systems and geologic disposal sites.[21]

Pot Calcination. The Chem Tech involvement in waste studies and management began at an early date (the late 1950s) with exploratory studies in calcination of HLW. Chuck Hancher and Herschel Godbee studied the pot calcination of actual Hanford waste in Building 4505. During the 1958-1959 time period, Tom Roberts, Ray Blanco, and Godbee continued waste studies in Laboratory B-13, Building 4500N. One of the initial problem areas was the volatility of ruthenium during waste calcination. A process was developed that used formaldehyde to reduce ruthenium (RuO_4) to a less volatile chemical form (RuO_2). Corresponding studies at Hanford used sugar instead of formaldehyde. These initial studies were conducted in stainless steel pipes 4-in. in diameter by 2 ft. At about this same time period, Hancher successfully conducted full-scale pot calcination in the high bay area of Building 4505. He controlled ruthenium by recycling the acid condensate.

Glass Formation. The waste group, soon joined by Walt Clark and George Davis, began studying use of sodium hypophosphite to reduce the volatilization of ruthenium during calcination. Subsequently, the group developed a phosphate glass process for treatment/disposal of waste. The glass process was commonly called "rising level" glass formation because the level of glass in the pots rose as the pots were filled and glass formed at -900°C. After Curt Fitzgerald joined the group, the studies were expanded to include formation of borosilicate glasses at -900°C. Phosphate glasses were highly and unpredictively corrosive. The corrosivity was known as the "zipper effect" in which stainless steel vessels would develop holes during the calcining/glass formation process. Dialogue was maintained with glass manufacturing firms concerning borosilicate glasses. The waste group started adding zinc oxide to the borosilicate glass to improve properties. Hanford developed a spray calcination process that produced a very fine waste oxide powder. The Chem Tech waste group developed a process that mixed this powder with a low-melting glass. The product contained the oxide powder (discontinuous phase) dispersed in the glass (continuous phase). Hanford took over the HLW glass studies after a Chem Tech management decision was made that most of the HLW problems had been solved. Hanford developed a continuous glassmaking process based on passing an electric current through the glass to generate heat for glass formation. West Valley and Savannah River are scheduled to use this technique to solidify their HLW.

FVETAP and Concrete Studies. Because concrete has considerable potential for waste treatment/disposal, the FUETAP program was initiated in 1970 by Chem Tech. The project ended about 10 years later when the Chem Tech group moved back from Y-12 to Building 3017 at ORNL. High-level waste produces an excellent ceramic, and the FUETAP group [John Moore, Gene Newman, Earl McDaniel, Marv Morgan, Les Dole (after returning from a year assignment studying waste treatment/disposal in Germany), and Mike Gilliam] conducted studies in developing such ceramics.

The FUETAP program takes its name from the phase "Formed Under Elevated Temperature and Pressure." Concretes that are formed under elevated temperature and pressure (i.e., autoclaved) are called FUETAP concretes. The accelerated curing process produces strong, durable, relatively impermeable solids. FUETAP concretes use the thermal power of radioactive waste to accelerate the curing process. Heat can also be applied externally. More than 98% of the unbound waste is driven off from the final product, and problems of radiolytic decomposition and thermal expansion become negligible. FUETAP concretes are less susceptible to weathering or degradation than normally hardened cement pastes.32

High-Level Waste Form. The DOE appointed a committee to help determine the best waste form to select for disposal of HLW. After reviewing the several options that existed at that time, the committee recommended to DOE that borosilicate glass be selected.

Scavenging ***Precipitation-Ion Exchange Process.*** In the early 1960s, Chem Tech developed the scavenging precipitation-ion exchange process (SP-IX) to treat and remove ^{90}Sr and ^{137}Cs from large volumes of low-activity waste or process waste.[33] The treatment plant, still in use, is currently managed by the Waste Operations Division. Process waste over 40 dpm/mL is treated and stored before treatment in a storage pond or equalization basin. The waste goes through a precipitation step, solids settling, and an ion-exchange process. Tom Roberts suggested using Duolite CS 100 resin. Zeolite is under consideration for replacement of the resin ion exchanger. Laboratory-scale work by Roland Holcomb and Bill Shockley confirmed the feasibility of the scavenging-ion exchange process. John Holmes designed a pilot plant which was operated successfully by Bob Brooksbank.

Hydrofracture. A study for disposing of ILW (4 mCi/gal to 5 Ci/gal) based on the oil field technique of hydraulic fracturing was initiated in 1959. Experimental injections were made with a grout mixture tagged with ^{137}Cs, and core drillings with gamma-my loggings were used to verify that the grout sheets followed the bedding planes and, also, that the fractures were essentially horizontal. Subsequent injections result in safe disposal of 1,600 m^3 waste containing 11,500 Ci total activity. Routine disposal began in 1966. A well is drilled into the geologic formation and cemented to prevent groundwater from entering the well. Water is pumped into the well under pressure and fractures the formation. The ILW wastes are then mixed with a cement-base blend of dry solids to produce a slurry that is injected into the impermeable shale formation at depths of 700 to 1000 ft. In the original grout formation, radiocesium was retained by illite clay and radiostrontium retained by cement and fly ash.[13]

Wally deLaguna and Tammy Tamura, then in the Health Physics Division, initiated the hydrofracture process and developed the initial grout recipe, respectively. Chem Tech staff members John Moore and Herman Weeren developed the grout formula for use in the later phases of the program. Illite clays had good cesium ion-exchange properties. However, when the source of illite clay used in the hydrofracture program was closed or shut down, Weeren discovered that pulverized Conasauga shale was even better than illite clay for sorbing cesium. However, no commercial source for large quantities of pulverized Conasauga shale was available. Basically, the Edisonian approach was used, and many different commercially available materials were tested, Red Indian pottery clay was found to be the best commercially available substitute for illite clay.

Asphalt. In 1965, the Belgians began disposing of LLW and ILW in an asphalt waste form using a batch process (heated pot with stirrer). Asphalt processes were being studied also by the English, French, and Germans. Chem Tech initiated studies in this area, with Herschel Godbee, John Holmes, and George Davis conducting the laboratory R&D. The Edisonian approach was again used, and the group developed and subsequently patented the use of emulsified asphalt for waste disposal in a continuous process (Fig. 4.33). The group also ‘developed a wiped film evaporator for use in the process, but were not permitted to patent that aspect of the process. The wiped film evaporator was tested successfully in the high bay area of Building 4505 under the direction of Ed Frederick (Fig. 4.34). The Chem Tech group also uncovered the incompatibility of nitrates and asphalt. Their studies indicated that nitrates in the waste solutions were not compatible with the burnable asphalt mixtures and that fires, and even explosions, could result from the nitrate-asphalt mixtures with high loadings of nitrate (>10–15 wt%). The French started using the wiped film evaporator process developed by Chem Tech for some waste types

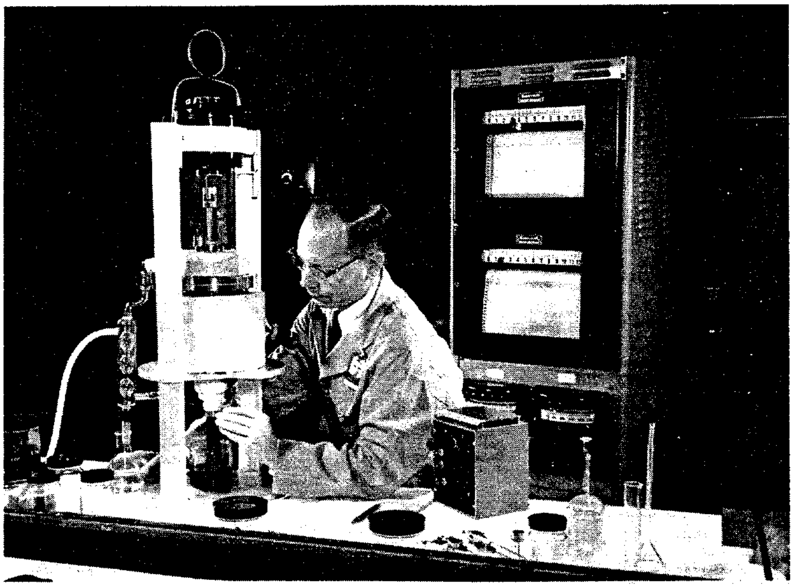

Fig. 4.33. Herschel Godbee conducting an emulsified asphalt experimental run with simulated waste.

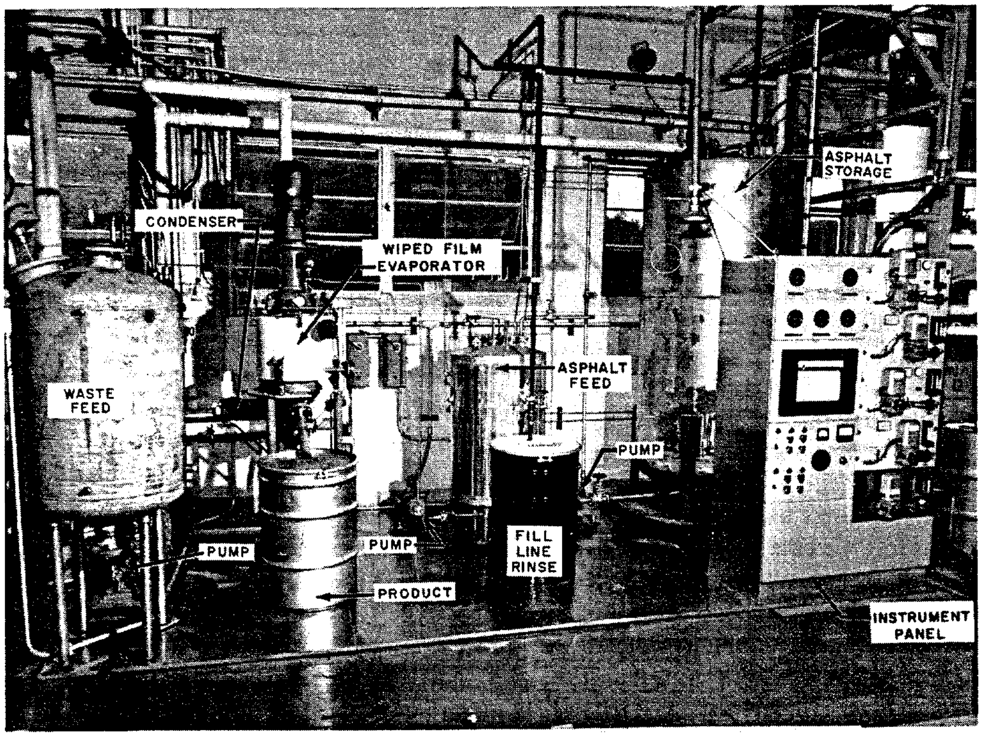

Fig. 4.34. Waste-asphalt process installation in the high bay area showing the wiped film evaporator equipment.

while continuing to use the screw-extruder process (continuous) which they had developed.

Disposal in Salt Domes. In the early 1970s, Chem Tech was involved in a large effort headed up by the Health Physics Division to establish a national repository for radioactive wastes in a bedded salt formation located near Lyons, Kansas. Chem Tech staff participated in almost all phases of the study but had principal responsibility in (1) conceptual design and safety analysis of the repository, (2) a survey of the sources and characteristics of TRU-contaminated solid wastes, and (3) experimental investigations concerning criteria for *packages* for storage of the *TRV wastes.* John Blomeke headed up the Chem Tech effort and was assisted by Baird Bottenfield, Frank Browder, R. S. Dillon, Ed Frederick, Frank Harrington, John Holmes, Joe Perona, and Bill Ulrich.[34]

Later Bill McClain and Tom Lomenick (who transferred into Chem Tech from the Health Physics Division) were involved in the study of the feasibility of using rock salt deposits for the disposal of radioactive waste. The state of Kansas was clearly the focus of this first field investigative period, with the highly successful experimental testing during Project Salt Vault being conducted at Lyons, Kansas, followed by unsuccessful efforts to site a "demonstration" repository at the same mine used in that project, and concluded by unsuccessful additional studies to locate and evaluate other sites within the state for a similar demonstration facility. Possibly the most noteworthy realizations made during this period concerned (1) certain technical issues regarding rock salt and its dissolution by groundwater and (2) increasing interface problems between the federal and state governments.

With the successful efforts of Chem Tech and Hanford to convert liquid I-ILW into solids for geologic disposal, the AEC requested Chem Tech to conduct a demonstration test in a suitable salt formation in an effort to establish the practicality of using salt deposits for the disposal of solidified wastes. The engineering and scientific objectives of this test, which were largely carried out by McClain, Blomeke, Bottenfield, and Lomenick, included (1) the demonstration of waste-handling equipment and techniques: (2) the determination of gross effects of radiation (up to 10^9 rads) on factors such as hole closure, floor uplift, and salt-pillar deformation within a temperature range of 100 to 200°C; (3) the determination of the radiolytic production of chlorine; and (4) the collection of data on the plastic flow of salt at elevated temperatures. The latter information was to be used in the design of an actual disposal facility.

The Project Salt Vault demonstration was carried out in the Carey Salt Company mine located at Lyons, Kansas. Fourteen irradiated Engineering Test Reactor fuel assemblies contained in seven canisters served as the radiation sources. Individual experiments commenced in mid-1964, and testing extended until late 1967.

In view of the uncertainties identified in selecting and constructing a geologic repository at Lyons, Kansas, as well as at any of the eight supplemental study areas located in that state, the AEC and Chem Tech enlarged the federal waste-disposal program in March of 1972 to include investigations of rocks other than salt and alternative disposal methods. The most dramatic alternative to geologic disposal was provided by an engineering approach termed the Retrievable Surface Storage Facility (RSSF), which was pursued by Blomeke. This was to be a part of a broad program of studies and assessments to investigate alternatives for both the near- and long-term storage of high-level and alpha-contaminated wastes. Specifically, the pilot-repository program included investigations by McClain, Lomenick, and others of various formations such as bedded salt located outside Kansas, in particular, the potash mining area of the Permian Basin in southeastern New Mexico and other impermeable rock types that might be suitable for the disposal of waste.

The overall objective of this program was to identify specific locations where pilot repositories could be constructed so that in situ demonstrations could be conducted that would provide confirmatory evidence of waste-rock compatibility. In order to accomplish this objective in the time allocated, studies were to be concentrated in areas where the necessary geologic and hydrologic data were already available or could be obtained readily. Godbee determined the thermal stability of alpha-containing waste and concluded (1) combustible wastes in combustible containers should not be accepted at a repository, (2) combustible wastes sealed in noncombustible containers (e.g., DOE 17C and 17H) should be acceptable, and (3) the quantity of gases evolved during heating can be reduced substantially by adding “getters” such as CaO (lime).[35] Other Chem Tech laboratory studies included the diffusion of ruthenium, cesium; and plutonium in salt (NaCl). Involvement in the Lyons, Kansas, project was terminated after it, was determined that the salt deposits might not provide adequate containment because they had been penetrated with numerous holes as a result of oil and gas drilling operations.

TRV Waste. Chem Tech staff members Jere Nichols and Herschel Godbee helped develop and set the disposal criteria for TRU waste. The DOE sponsor, Harvey Sole, desired to have the disposal criteria set at a TRU element concentration that produced >1 nCi/g waste. The NRC would have accepted the limit set at >500 nCi/g. The DOE initially set the limit at >1 nCi/g. After several years, it became apparent to DOE that the limit was set much lower than necessary for protection of the public and the environment. At the request of DOE, ORNL conducted a survey that eventually resulted in the limit being set at 100 nCi/g.

Leaching Tests and ANS 16.1. The characterization of phosphate, borosilicate, and dispersion glasses required the use of leaching tests. This laboratory work by Godbee and Moore led to the national development of the standardized ANS 16.1 leach test. This test was the result of a committee of ~40 people including Oswald Anders of Dow Chemical Co., John Mendall of Westinghouse Hanford, Bob Nielsen of Idaho National Engineering Laboratory (INEL), and Ed Compere and Godbee of ORNL.

Polymers. In the 1971 Chem Tech progress report it was reported that ILW was incorporated in polyethylene. From 19 to 32% waste solids were incorporated in low-melting polyethylene (softening point, 100°C) with material balances of 95 to 100%. Waste form leach rates were favorable.[34] Chem Tech staff members Godbee, Fitzgerald, Davis, and Ken McCorkle patented the process for incorporating waste in polyethylene.

4.14 WASTE MANAGEMENT TECHNOLOGY CENTER, Bob Jolley and Suman Singh

During FY 1986, Martin Marietta Energy Systems, Inc., and DOE/ORO established a new Waste Management Technology Center (WMTC) to serve as a technology component of a comprehensive DOE/ORO waste management coordination effort referred to as the “Oak Ridge Model” (later changed to the DOE Model). The responsibility for the development and management of the WMTC was assigned to Richard Genung of Chem Tech. By April 1987, Genung, as manager, had staffed WMTC with a

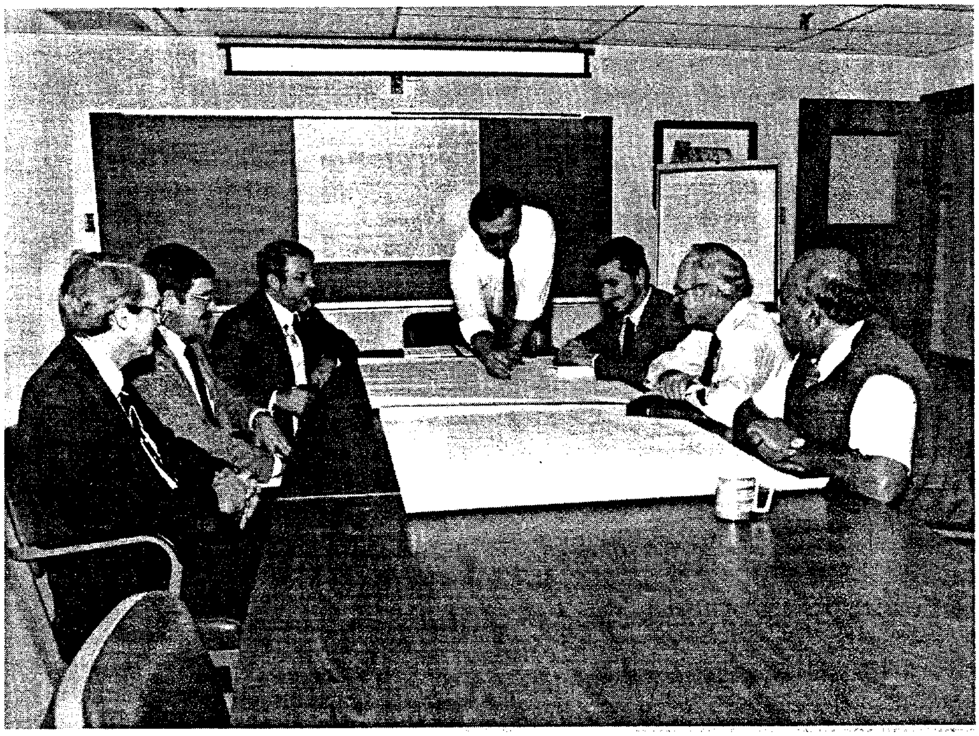

Fig. 4.35. Richard Genung leads a 1987 WMTC brainstorming session on the development of an integrated management scheme for low-level and mixed radioactive wastes. Left to right: Bill Brooke, John Kenneriy, Les Dole, Richard Genung, Angel Rivera, Bob Joiiey, and Suman Singh.

small team of engineers, scientists, and technical support personnel: D. S. Brown, L. R. Dole, R. L. Jolley, J. M. Kennerly, M. J. Emmett, G. D. Humphrey, A. L. Rivera, and S. P. N. Singh.[36] C. A. Proaps, G. E. Butterworth, P. E. Hollenbeck, T. F. Lomenick, M. I. Morris, C. P. McGinnis, and L. K. Hunt joined WMTC later (Fig. 4.35).

When Genung became Chem Tech Division Director in 1988, the WMT'C was combined with the other Chem Tech groups located at the K-25 site (Grout Technology and Engineered Waste Disposal Technology Groups) to form the Waste Management Technology Support Group (WMTSG), with Jolley as manager.[37] The WMTC was effectively dissolved in 1991 when its major programs were assimilated into other Energy Systems divisions (Environmental Restoration and Central Waste Management) and the Waste R&D Program.

The WMTC charter included providing special support in a centralized manner to various waste management organizations in the DOE/ORO-managed complex. The charter encompassed demonstrations of treatment, storage, and disposal technologies relevant to the management of transuranic, low-level, hazardous, and mixed wastes and provided staff support for the development and analysis of technical information supporting remedial actions and compliance strategies by DOE/ORO contractors, including Energy Systems (Fig. 4.36).[36]

The WMTC managed several major programs: the Low-Level Waste Disposal, Development, and Demonstration (LLWDDD) Program; Hazardous Waste Development, Disposal, and Demonstration (HAZWDDD) Program; demonstration of hazardous waste management technologies on DOE/ORO sites: Historically Black Colleges and Universities (HBCUs) waste program; and technical workshops.

Low-Level Waste. Disposal Development and Demonstration Program. The fust major program managed by the WMTC was the LLWDDD Program. Genung assumed management of this program in 1985. Butterworth became the manager in 1987 and managed the program until it became a corporate division in 1990. The LLWDDD had responsibility for (1) defining and demonstrating improved waste management technologies for

Fig. 4.36. Les Dole (left), Bob Jolley (center), and Angel Rivera discuss current DOE-managed sites.

low-level solid wastes generated within the ORO complex; (2) siting new disposal facilities to be used by plants on the Oak Ridge Reservation; (3) coordinating the preparation of an Environmental Impact Statement for new disposal facilities on the Oak Ridge Reservation: and (4) designing and constructing large-scale disposal demonstration units within the new facilities. Several LLWDDD demonstration projects were initiated during 1986 and 1987. These included above-grade, earth-mounded disposal systems based on information exchanges with the French using their experience with similar technology, the evaluation of various waste packaging materials and technologies, and the assessment of mechanical volume reduction technologies based on super-compaction processes. All of these projects involved participants from the private sector, through competitive procurement processes, with emphasis on the selection of innovative mobile technologies for waste treatment.[36]

Hazardous Waste Disposal, Development, and Demonstration Program. The HAZWDDD Program managed by Phil McGinnis was initiated in late 1987 to integrate hazardous and mixed-waste treatment, storage, and disposal needs. Major responsibilities included developing an overall corporate strategy plan covering hazardous and mixed waste, including all five Energy Systems managed sites; identifying and coordinating needed development, demonstration, and technology transfer projects; and serving as a communications link among the five Energy Systems facilities, the Environmental and Safety (ESA) Organization, Central Waste Management Organization (CWMO), and WMTC.[38]

Technical Demonstrations. The next major program managed by the WMTC concerned the demonstration of hazardous/mixed waste management technologies on DOE/ORO sites. John Kennerly, Suman Singh, Mike Morris, and Paul Hollenbeck were actively involved in demonstration planning, management, and execution. Demonstration projects were awarded to DOE field offices by the DOE National Hazardous Waste Remedial Action Program (HAZWRAP) and, in some cases, were also supported by the sites themselves. The WMTC identified candidate projects for DOE/ORO, assisted in presenting these candidate projects to HAZWRAP for competitive evaluation and funding, and worked with DOE/ORO sites to manage these demonstration projects.[36] The WMTC worked closely with Energy Systems sites to facilitate demonstrations.

In 1990, demonstration management became a corporate function under the Waste R&D Programs managed by Tony Malinauskas.

From 1986–1990, WMTC managed, coordinated, or assisted in conducting the following demonstrations. All of the projects involved participants from the private sector, through competitive procurement processes, and emphasized selection of innovative technologies for waste treatment (Fig. 4.37).

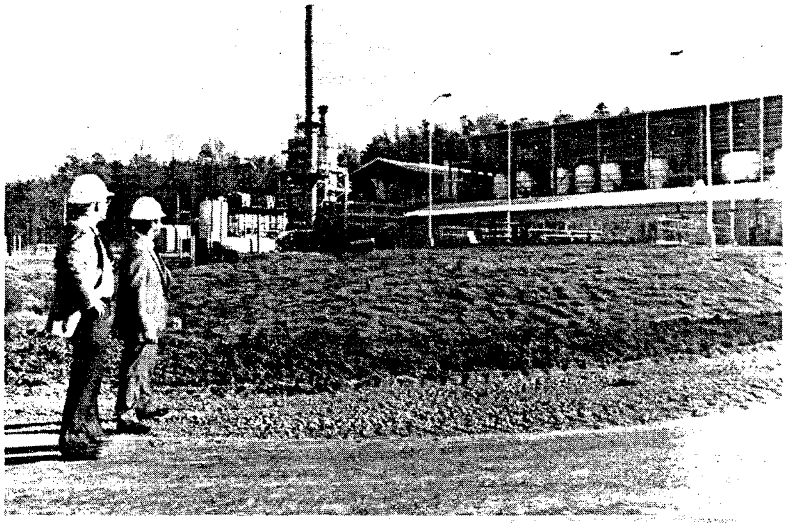

Fig. 4.37. The TSCA Incinerator located near the K-1037 office of WMTC was technically supported by Suman Singh and Angel Rivera, WMTC staff engineers. In this 1987 photograph, Bob Jolley (left) and Angel Rivera **discuss** Angel's support role for the **Incinerator.**

- Evaluation of Vendor, Process, and Waste Form/Packaging of Oak Ridge Reservation Waste Streams: Waste Isolation Technology Project. The objective of this LLWDDD demonstration managed by Mike Gilliam, Mike Morris, and Jennifer Kasten was to evaluate the conditioning, treatment, and immobilization of selected waste streams. A major emphasis was the identification and demonstration of innovative technologies for processing four specific waste streams: Y- 12 wastewater treatment sludge containing heavy metals and depleted uranium: ORNL wastewater-treatment water-softening sludges containing Cs, St, Co, and trace rare earths; Y-12 metal plating sludges containing heavy metals and chlorinated phenolics; and ORNL wastewater treatment concentrates containing very high quantities of nitrates, Cs, Sr, and h-ace rare earths.38
- Supercompaction/Grouting of ORNL Solid Low-Level Waste Drums. The objective of this LLWDDD project was to demonstrate supercompaction and grouting of solid waste in drums after real-time radiogmphy. The demonstration was coordinated by John Kennerly and conducted by L. C. (Red) Williams, R. W. Morrow, and R. L. Jeffers.[38]
- ORNL Tumulus Disposal Demonstration for Low-Level Waste. The objective of this LLWDDD project was to evaluate above-grade disposal of short-half-life radionuclides using the tumulus concept developed in France for disposal of LLW. The demonstration was coordinated by John Kennerly and managed by Dirk Van Hoesen, Sid Garland, Red Williams, and Tom Scanlan.[38]
- Supercompaction of Y-12-Baled Waste. The objective of this LLWDDD demonstration was to evaluate volume reduction of previously baled solid waste using super-compaction. The demonstration was coordinated by Kennedy and Beth McDougald.[38]
- Incineration of Y-12 Combustible Waste. The objective of this Y-12-sponsored demonstration was to evaluate volume reduction of Y-12 combustible wastes. The demonstration was conducted by a commercial vendor and coordinated by Mike Morris for Jim Bailey and Rod Kimmitt.
- Demonstration of Improved Operations Disposal at ORNL SWSA-6. The objective of this LLWDDD demonstration was to evaluate greater confinement disposal using below-grade concrete silos. The demonstration was coordinated by John Kennerly for John Van Cleve, Red Williams, and Tom Scanlan.[38]
- Treatment of PCB-Contaminated Soil. The objective of this Energy Systems/private sector sponsored demonstration managed by Mike Morris was to evaluate the thermal decontamination (calcination) of soils

contaminated with uranium and PCB. This demonstration was conducted at the commercial vendor site.38

- Y-12 Sludge Detoxification, The objective of this HAZWRAP demonstration managed by Paul Hollenbeck and John Kennerly was to evaluate the thermal treatment of Y-12 mixed waste sludges contaminated with high concentrations of phenolic materials. Detoxification of waste is defined as the removal or degradation of chemically hazardous materials. This demonstration was conducted by Chem Nuclear at the K-25 site.[38]
- Closure of Old Hydrofracture Surface Impoundment. The objective of this Energy Systems demonstration coordinated by Tom Lomenick and Mike Morris was to evaluate in situ processes for stabilization/fixation of impoundment residues.[38]
- Pyroplasma Technology for Destroying PCB-Containing Fluids (Westinghouse Pymplasma Demonstration Project). In late February 1987, Westinghouse Plasma Systems (WPS), a subsidiary of Westinghouse Electric Corporation, sought DOE/ORO assistance to conduct a demonstration of their innovative Pymplasma waste destruction process. Suman Singh coordinated Energy Systems support for conducting the demonstration to evaluate the high-temperature thermal destruction (Pyroplasma) of PCB-containing liquid wastes. The demonstration was conducted by WPS at the K-25 site.[38]

Historically Black Colleges and Universities. With support from DOE/ORO, in 1987 the WMTC also established a special program to involve staff members and graduate students from Historically Black Colleges and Universities (HBCUs) in waste management activities on the Oak Ridge Reservation. HBCUs were later to be known as the Minority Educational Institutes (MEIs). The WMTC worked with waste management organizations at ORNL, K-25, and Y-12 and with HBCU coordinators for Energy Systems and DOE/ORO to define potential problems to be addressed, identify and match HBCU staff members with Energy Systems technical project monitors, and develop contractual arrangements.[36] Les Dole and later Bob Jolley coordinated the HBCU waste program.

Technical Workshops. The expertise available in several of the research and development organizations within Energy Systems was also used in planning and evaluating technology demonstrations, in evaluating the associated implications for the protection of human health and the environment, and in complying with regulations. These planning and evaluation exercises were supplemented by a series of technical workshops conducted by the WMTC to assist in evaluating issues and in assessing potentially useful technologies within the broad subject context."

Workshops conducted by WMTC/WMTSG included the following:

- Leaching Test Workshop series managed by Herschel Godbee and Bob Jolley,[37,38]
- Uranium-Bearing Waste Workshop managed by Tom Lomenick,[38]
- RCRA/CERCLA Treatment Alternatives for Hazardous Waste workshop managed by Bob Jolley,[38]
- Contaminated Soils Workshop managed'by Tom Lomenick,[37]
- Off-Site Release Criteria Workshop managed by Suman Singh,[37] and
- Alternative Treatment Technologies Workshop for Superfund Sites managed by Bob Jolley.[37]

4.15 THE CEUSP PROJECT: A LARGE-SCALE CONVERSION AND SOLIDIFICATION OF HIGHLY RADIOACTIVE LIQUID WASTE,

C. P. McGinnis, R. A. Jacobus, and L.. H. Bell[39]

The Consolidated Edison Uranium Solidification Program (CEUSP) project was a large-scale project to convert and solidify stored, highly radioactive liquid waste at ORNL. The project was made necessary because highly radioactive liquid waste had been stored for many years at ORNL in a facility where no provisions were made for the eventual chemical conversion of the solution to a form more suitable for long-term storage or disposal. ORNL's liquid radioactive waste solution resulted from a 1960's burn-up of experimental nuclear fuel consisting of mixed thorium oxide and fully enriched uranium oxide in the Consolidated Edison Indian Point I nuclear reactor in New York. The depleted experimental fuel was subsequently reprocessed at the Nuclear Fuel Services Plant in West Valley, New York. During reprocessing, the fuel was dissolved and passed through a Purex solvent extraction line to separate uranium from the thorium and fission products.

The reprocessed uranium solution was shipped to ORNL in late 1968 and placed in a 19,000-L storage tank located within a stainless-steel-lined catch pit and surrounded by an additional 1 m of concrete shielding. The stainless steel tank was filled with ordinary borosilicate glass Raschig rings to absorb neutrons, and cadmium nitrate and gadolinium nitrate solutions were added to prevent criticality of the highly enriched solution. At storage time, the liquid waste contained 1047 kg of uranium as uranyl nitrate hexahydrate in 8000 L of nitric acid.

Immediately after reprocessing, the uranium solution was not highly radioactive and could be handled semi-remotely. However, during the years of storage at ORNL, the liquid's high concentration of ^{232}U (70-year half-life) and the resulting high concentrations of daughter decay products caused the solution to become highly radioactive. The waste contained a total estimated inventory of 20,000 Ci by 1978 and a radiation field at the tank of more than 120 rad/h. This intense radioactivity made remote handling a necessity during the CEUSP removal, conversion, and storage processes and introduced complications in design, construction, and operation.

In 1974, a DOE Safety Review Team visiting ORNL concluded that long-term storage of the fuel solution was unacceptable, and no uses for the material were identified. Scientists and engineers at ORNL led by John Parott, Sr., evaluated seven different disposal alternatives and eventually recommended a simple chemical and thermal decomposition and solidification process.

4.151 CEUSP Process and Facility Design

A preliminary conceptual design for the CEUSP waste processing plant was completed in 1979. The Conceptual Design Report called for a 3-year construction and fabrication schedule. An extensive process development study was conducted to determine the equipment modifications and processing conditions needed for successful process operation, including work by E. D. Collins, B. D. Patton, R. Hall, P. A. Haas, and R. J. Vedder on in situ solidification of fissile uranium, reaction of formaldehyde and nitric acid (denitration), and associated remote operations.

4.15.2 Preoperational Testing Failure

Construction of the CEUSP facility was completed in March 1984 (Fig. 4.38), and preoperational hydrostatic pressure testing and process shakedown were immediately done using water and then dilute acid. These initial system tests were successfully completed. Six weeks later, a second hydrostatic pressure test was done before introducing a simulated process feed material. Surprisingly, the second hydrostatic test showed the system had developed leaks. A detailed technical investigation revealed the cause to be a failure of the compression fittings used at pipeline joints.

Fig. 4.38. Phil McGinnis, Program Manager (far tight), and Ron Glass (far left) are shown escorting DOE visitors on a 1984 tour of the newly opened CESUP facility. Jim Cooke, DOE/ORO, and Fred Mynatt, ORNL Associate Director, are shown in the center.

4.15.3 CEUSP Process Operation and Results

Actual process-line operation began in April 1985. During the initial acid destruction/evaporation step of the CEUSP process, each 21-L batch of feed solution was heated to >95°C in the evaporator before an aqueous-methanol solution containing 37% formaldehyde was added. Additional heat was added to this exothermic acid-destruction reaction to evaporate each batch to a final volume of 8 L. Each batch of liquid from the evaporator was routed to one of three thermal denitration/solidification systems and fed directly into a can (the primary storage container) held in a small furnace. The solution was evaporated to dryness and the nitrates decomposed in situ, leaving a solid oxide cake in each can. The cans were then transferred remotely to another process area, where they were placed in secondary containment canisters and then welded shut. After sealing was completed, the materials were placed in a specially prepared shielded well for long-term storage. A “hard” (very energetic) gamma associated with the radioactive decay products of ^{233}U required special shielding and precautions. The CEUSP process successfully solidified 400 batches of highly radioactive liquid waste containing a total of >1000 kg of uranium.

Surprisingly, no unanticipated down-time occurred during the entire CEUSP processing. Several planned down-times were required to replace the waste concentrate feed pumps, which were known to have a limited service life when exposed to acidic radioactive solutions. Fortunately, no other equipment repairs or replacements were necessary. As a result, the operational phase was completed in early June 1986, almost two months ahead of schedule. During its 13 months of operation, the CEUSP facility converted 8000 L of the stored radioactive liquid waste to a more stable and space-conserving solid oxide form.

Don Ferguson was the CEUSP program director. The project was managed by Phil McGinnis. Operational and management assistance was provided by R. A. Jacobus, J. M. Baker, R. D. Manthey, Jim L. Snider, Emory D. Collins, and Rex E. Leuze.

4.16 CHEMICAL ENGINEERING RESEARCH, John Mrochek, Osman Basaran, and Tim Scott

Chemical engineering research has, since the inception of the division in 1950, been one of the primary foci of its activities. These activities have included studies on the fundamentals of engineering science and bridging research that utilized the results of the more basic chemical research. Incorporated in these endeavors has been the innovative development of new hardware, instrumentation, and/or process concepts in order to accomplish needed programmatic goals. R&D efforts of the division have always been at the cutting edge of an established or new technology. There has always existed a relatively fine line between fundamental or basic research and applied research. In 1965, chemical engineering research was described in the division annual report as “an aggregate of studies which, while generally pertinent to the Division’s applied programs, are fundamental in nature or pursue attractive new ideas. The fundamental studies usually arise from interesting effects noted during work on programs with more specific commitments.”[40]

Much of the activities of the division in chemical engineering research generally were focused on the fundamentals of separation methods and/or processes. In the beginning, such separations were based upon solvent extraction equipment and processes. The development of new contactors and investigations into their fundamental behavior were, of course, fair game (Figs. 4.39 and 4.40). Pioneering work on sieve-plate, pulsed columns for solvent extraction was noteworthy, as was the work on a stacked hydroclone contactor for high-speed extraction: design and development work on these new contactors continued for many years. Of interest also were fundamental studies of mass transfer, especially across two-phase interfaces associated with solvent extraction. Another important factor in the design of solvent extraction equipment, namely, the coalescence and the effect of ionizing radiation on it, was being investigated by division staff members during the period 1962–65.

The need for the production of small, uniform microspheres having particle sizes on the order of 20 led to investigations on the breakaway of droplets from a jet or nozzle in conjunction with an

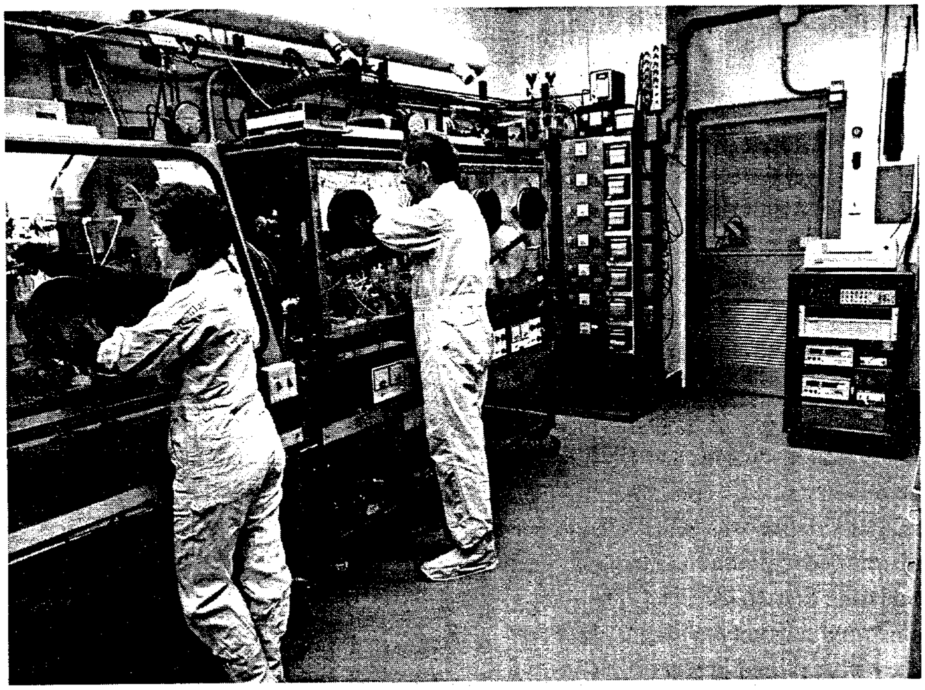

Fig. 4.39. D. F. Green (left) and George Davis work in a glove box system for plutonium flow sheet development using miniature mixer-settlers. The left glove box contains solution receivers, In-line samplers, and In-line density meters and spectrophotometers. The right glove box contains three 16-stage mixer settlers for extraction, partitioning, and stripping of plutonium and uranium. In the background are the metering systems for the feed solutions and a portion of the Hewlett-Packard data acquisition and control system.

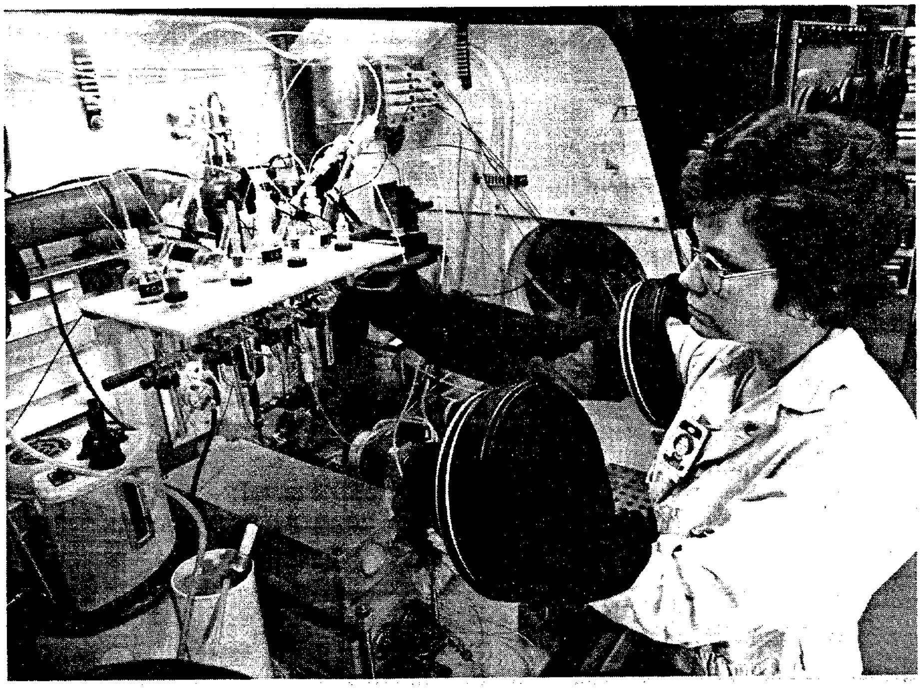

Fig. 4.49. D. F. Green is withdrawing a Vacutainer containing a 1-mL sample of the plutonium flow sheet process stream. Two remote density cells are visible In the center foreground. A miniature spectrophotometer containing a flow cell is visible in the center background.

applied program associated with sol-gel microspheres. Nonresonant and resonant ultrasonic devices were employed to cause droplets to break away from the injection nozzle at exactly the correct time to form droplets of the desired size. This work, initiated in 1969, was a prelude to fundamental studies on the mass transfer of water from sol droplets and its effect on droplet deformation in the sol-gel process in the early 1970s. The hydrodynamics of bubble formation, dispersion, and collapse was an important fundamental investigational area associated with separations processes.

Frequently, during the 1960s and 1970s, more fundamental studies of chemical engineering problems were reported as a part of the applied programs with which they were associated. Such programs, while not enumerated here, contributed much to the fundamental knowledge associated with various separations processes.

Developments in the use of small-particle-size ion-exchange resins to improve separations were pioneered in a divisional biochemical technology program during the late 1960s. In 1971, the applicability of small resin particles in the size range of 10 to 50 μ to large-scale separations processes was proposed based upon experimental and economic studies. Even though the cost of the small ion exchangers was much higher, improved separations and the downsizing of equipment enabled their utilization to be cost-effective.

About 1975, chemical engineering research programs became more oriented toward fundamental or basic science and less driven by needs in applied programs. The annual report in 1975 listed the area as "Studies in Chemical Engineering Science" instead of "Chemical Engineering Research." In conjunction with coal conversion studies, research on three-phase fluidized-bed reacting systems was initiated together with some ongoing work on tritium permeation associated with fusion reactor work. Innovative work was begun in 1975 on a new separations concept known as continuous annular chromatography. An annular chromatograph was developed which, in 1978, won a prestigious IR-100 award as one of the 100 most significant new technical products of the year (Figs. 4.41 and 4.42).

During 1977, a number of different basic research efforts, including chemical engineering research, were, for reporting purposes, combined under the title Basic Science and Technology.

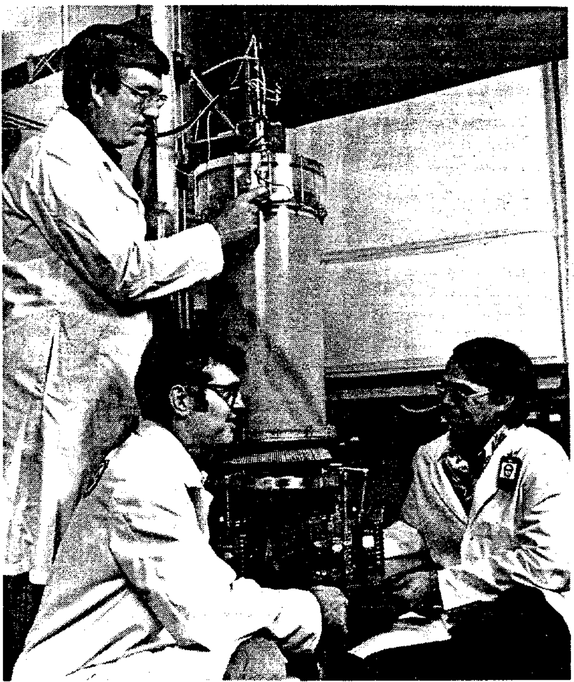

Fig. 4.41. Chem Tech development team members Warren Sisson (standing), Roger Spence (kneeling left), and Ron Canon discuss the performance of the continuous annular chromatograph for which they received the IR-100 award.

Fig. 4.42. John Begovich and Warren Sisson monitor the continuous chromatographic separation of test materials.

Included among these were Separations Science; Resource Recovery; Interphase Transfer Kinetics; Fusion Energy Studies; Actinide Oxides, Nitrides, and Carbides; and Chemical Engineering Research. Continuous chromatography continued to be an important focus of chemical engineering research. However, because of national concerns in the overall area of energy, research associated with fossil energy conversion processes was assuming increased national importance in 1977. Agglomeration studies of coal-derived particles in organic media began, and resource recovery employing energy-efficient processes assumed additional importance. The early 1980s saw continued emphasis on the investigation of new engineering concepts in separation and material science areas which related to advanced energy sources, the conservation of energy, or the energy-efficient recovery of important resources. Granular elecirofiltration was shown to be effective in removing small particles (<1 μ)m) from dilute suspensions in organic liquids in conjunction with coal conversion processes. Advanced concepts such as an absorption-fractionation process using liquid CO_2 for purification of methane from advanced coal gasification processes were conceived and tested.

During the period of 1983 to 1985, the continuous annular chromatography project was brought to a successful conclusion. Efforts to transfer the technology to industry were starting to bear fruit, with several companies building their own annular chromatographs either to solve their own problems or to commercialize the idea. Light scattering techniques were under development to measure fluid properties near the critical region of process fluids. Sorption properties were under investigation in electrically stabilized expanded beds of non-conducting granular sorbents. Also commencing were sedimentation studies on multimodal suspensions. Fundamental studies were begun on improving the efficiency of separations processes based on solvent extraction by enhancing mass transfer by causing drops to oscillate by means of a high-voltage pulsed electrical field. Basic investigations into high-temperature slagging were continued with the objective of recycling a large fraction of the nation's scrap steel, currently limited by lack of a technique for reducing its copper content.

Fundamental research in chemistry and chemical engineering within the Chemical Technology Division was supported by the Division of Chemical Sciences and the Division of Materials Sciences in the Office of Basic Energy Sciences (BES) within the Department of Energy. In 1984, most of this work was brought together in a newly formed section of the division called Energy Research Programs. A new area of research was initiated, a fundamental study of the molecular interactions between solvent and solute in supercritical solutions. The primary goal was to develop a predictive capability for use in supercritical separations employing both theoretical and experimental techniques (Fig. 4.43).

By the mid to late 1980s, it was realized that computational capabilities in the division were lagging far behind experimental capabilities. Thus new research activities in theoretical analysis, modeling, and computational science were initiated to close this perceived gap. By the early 1990s, computational techniques, which were primarily based on finite element methods, were being

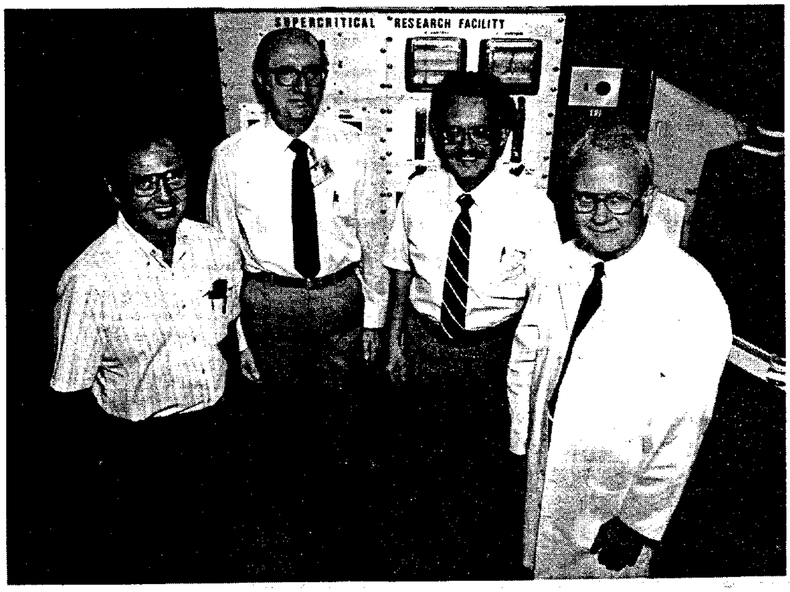

Fig. 4.43. From left to right, Charlie Byers, Jack Watson, Terry Donaldson, and Chuck Scott pose before the control panel for the supercritical research facility.

routinely used to analyze, without approximation, oscillations of free and pendant drops, flow and mass transport to or from liquid drops, and interactions between two drops. The calculations were becoming sophisticated enough that, in a few selected instances, they provided the initial knowledge base for performing the right experiments that ultimately led to the conceptualization of improved practical designs and even patent applications. Fundamental research on separations processes continued and was combined with the new computational thrusts in an overall initiative designed to seek understanding of the principles and develop methods for studying the influence of external electric fields upon mass transport and fluid mechanics. This program, primariiy concerned with droplet dispersion, coalescence, and mass transfer in two-phase systems, was to continue into the 1990s.

Continuing fundamental work in the above program during 1985-1987 resulted in the invention of a radical new concept in solvent extraction contactors, an electrically driven contactor. Two major problems which limit the use of solvent extraction in industrial applications are the efficient creation and control of mass-transfer surface area. In practice, interfacial mass-transfer surface area is usually created by a form of mechanical agitation. When attempting to maximize surface area production in an apparatus, mechanical agitation techniques tend to form polydisperse emulsions which are difficult to characterize and control in mass transfer operations. A method was required that would not only efficiently create large amounts of interfacial area for transport but also enable adequate control of phase disengagement once mass transport is completed. The use of electric fields to accomplish surface area generation and coalescence/phase separation enables multistage countercurrent solvent-extraction operations to be performed in vessels which are an order of magnitude smaller while requiring only a fraction of the operating energy of mechanical agitation. The initial patent on this new invention was granted in 1988; a subsequent patent was received in 1990.

The development path followed by this Emulsion Phase Contactor (EPC) illustrates the unique way in which government-funded research in national laboratories should be employed for enhancement of the public welfare. This invention demonstrates conceptual development of an idea under fundamental or basic research funding by the Chemical Sciences Division of the BES. Proof-of-principle funding to further develop the invention to a point ‘where industry could take over was provided by the Advanced Energy Projects Division of BES. Under the impetus provided by the Technology Transfer Program, the technology has now been licensed to two industrial firms. Both firms are proceeding with the development of commercial versions of the EPC, one in petroleum, rare-earth, and specialty chemical technologies and the other in analytical and pharmaceutical applications. A commercial version is expected to be on the market in 1992.

The early 1990s saw a continuation of the programs entitled Effects of External Fields on Multiphase Systems and Interactions of Solvents, Solutes, and Surfaces. Both of these programs are, of course, deeply rooted in the fundamentals of separations processes. The former is principally concerned with improving the energetics and efficiency of solvent extraction by initiating fundamental studies on the use of electromagnetic fields to enhance multiphase separations. Experimental and theoretical thrusts are on probing transport fundamentals in field-enhanced liquid-liquid, liquid-vapor, and fluid-solid separations. The latter program is primarily concerned with developing a predictive theory for separations conducted in the supercritical region of solutes and/or solvents. Fundamental experimental and theoretical studies are aimed at understanding the striking properties of supercritical solutions in terms of the underlying fluid microstructure and molecular interactions. Basic research is focused on relating macroscopic properties of mixtures to intermolecular interactions and molecular correlation functions for the highly asymmetric systems characteristic of extraction by and adsorption from supercritical fluids.

Over the 42-year history of the Chemical Technology Division, chemical engineering research has maintained a strong interest and involvement in separations (Fig. 4.44). During the early years, there appeared to be a very close coupling of basic or fundamental research with applied problems arising in other division programs. In later years, a clear decoupling of fundamental research from applied areas seemed to occur. This was most certainly due to constraints imposed by the various funding organizations. This is not to imply that such separation was undesirable because it may have resulted in enhanced progress in the fundamental areas. The division’s programs

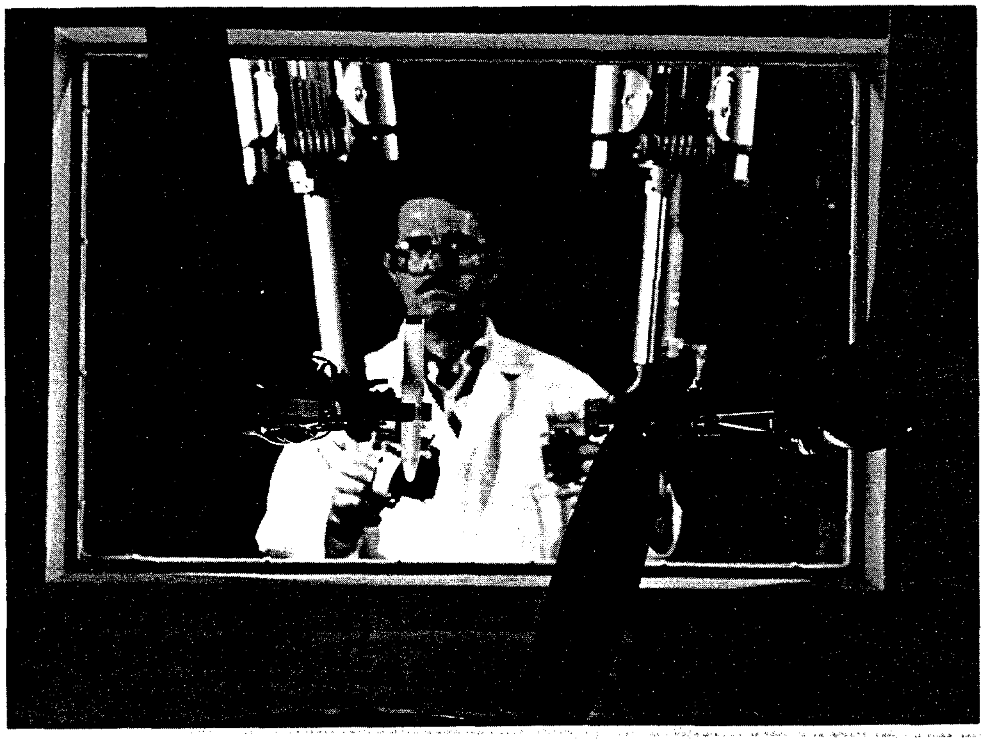

Fig. 4.44. Bob Fellows is using manipulators to prepare a solvent extraction **sample** for removal from the hot **cell.**

in chemical engineering research have **usually** been at the cutting edge of technology and have always served to enhance the division's image as a premier research division at the Oak Ridge National Laboratory (Fig. 4.45).

4.17 HISTORICAL OVERVIEW OF THE CHEM TECH PILOT PLANT (BUILDING 3019),

J. R. Hightower and R. E. Brooksbank, Sr.

In early 1943, as part of the Manhattan Project, plans were made to build an air-cooled nuclear experimental pile, a chemical separations pilot plant, and supporting laboratories on an isolated tract known as X-10. These major installations became the prime function of the Clinton Engineer Works, now known as ORNL. Since that time, Building 3019 (formerly known as Building 205) has served as a pilot plant in the development of several **radiochemical** processes that have found plant-scale application in both government and commercial facilities **on** a worldwide basis. In addition to the process development role, the facility's operations have also produced large quantities of product materials (plutonium, uranium of all isotopes, thorium, and special isotopes) while processing highly irradiated fuel.

The programs conducted in Building 3019 during its **48-year** history **have** had a major impact on the government's missions. The versatility of the facility has been adequately demonstrated, indicating that the building represents a valuable asset to future government programs.

4.17.1 Role of a Pilot Plant

A pilot plant is one operational step in the orderly plan of chemical process development. The usual function of a pilot plant is to bring out procedures arising from the integration of all phases of the process and to obtain adequate quantitative data for the design and operation of an economical production plant. In addition to being a development facility, a pilot plant serves as a small-scale production plant, having many of the characteristics of a full-scale production plant. In general, development programs in a pilot plant should accomplish the following primary **objec**tives:

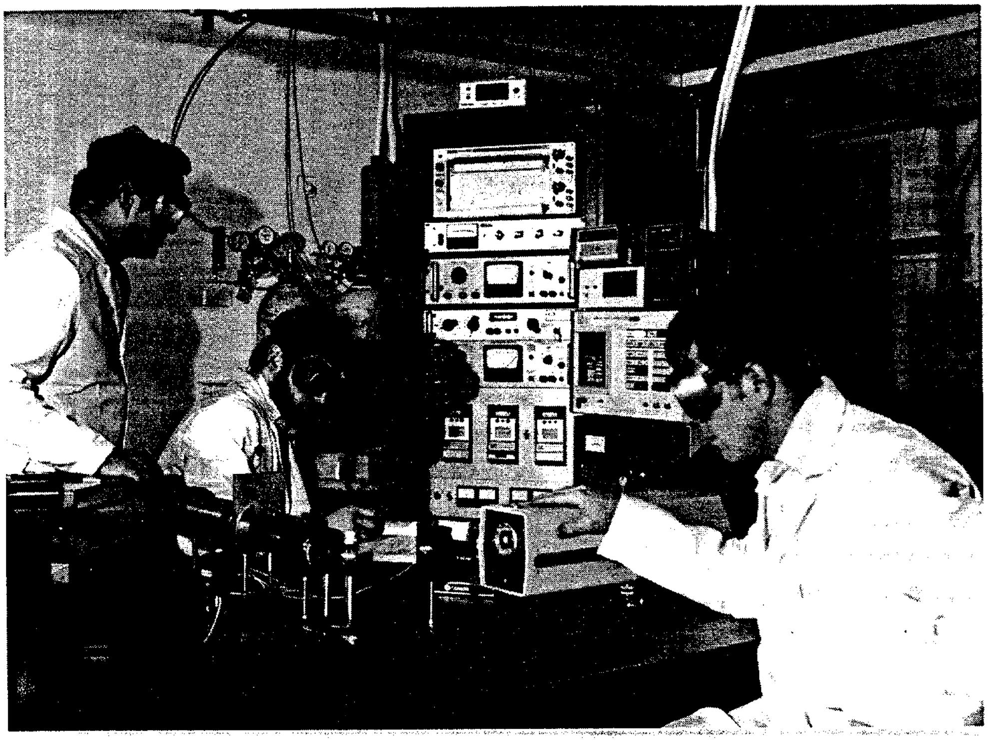

Fig. 4.45. Charlie Byers **(left)**, **Ron Brunson** (center), and Dave Williams **monitor** the laser systems being used to study fundamental structure and properties of solutions.

1. **Confirm** the feasibility of the proposed process.
2. Obtain quantitative engineering data necessary for the design and operation of a production plant.
3. Provide quantities of the product for large-scale evaluation at other sites.
4. Identify chemical and engineering problems that were not recognized in smaller-scale development work.

The Chem Tech Pilot Plant achieved these objectives very successfully.

4.17.2 Role of Building 3019 in Reprocessing Technology

The major programs conducted within Building 3019 in support of the government's missions during the period from 1943 to 1976 (the period of formidable development) are presented in this section along with the quantities of material recovered as the result of the building's operation. These materials were recycled into other government programs as required. In addition to the efforts expended in the handling of uranium-based spent reactor fuel, the reprocessing of irradiated thorium in the United States is discussed, indicating the quantity of the ^{233}U recovered to date. This uranium isotope is currently a major concern to the operations taking place in the building because the ^{233}U isotope has unique characteristics relating to criticality, shielding, and contamination control.

The first tens-of-grams quantities of plutonium were precipitated from tons of uranium and grams of fission products in Oak Ridge in 1943, just four years after Dr. Seaborg isolated a few micrograms from an accelerator target. Between 1943 and the present time, hundreds of thousands of tons of irradiated uranium have been processed, both in defense and commercial reprocessing plants, on a worldwide basis.

Since 1942, ORNL (formally Clinton Engineer Works) has been continuously engaged in process development of the nuclear fuel cycle. The basic process techniques in which ORNL participated, along with their chronology, are listed in Table 4.1. As can be seen from this table, Building 3019 has played a major role in this development effort. The role of a pilot plant in the major development

Table 4.1. Chronology of reprocessing experience at ORNL

Period	Process	Building No.
1943-194s	Bismuth Phosphate	3019
1945-1951	Redox	3019
1945-1952	RaLa	3026
19461948	Hexone-25	706A
1946-1948	Hexone-23	706A
1948-1949	Uranyl Ammonium Phosphate	706A
1948-1958	Metal Recovery	3505
1948-1953	TBP-25[a]	3505
1949-1960	Purex	3019.3505
1949-1968	Flouride Volatility[b]	3019
1949-1976	Fuel Preparation[c]	3019, 4505, 7930
1951-1976	Raw Material[d]	4500
1952	TBP-Interim-23	3503
1952–Present	Thorex	3019
1953-1959	Feed Materials[e]	4500
1955-1976	Head-End	4500N, 4505, 4507, 7601
1961-1976	TRUG[g]	3508, 4507, 7920

[a] Includes Homogeneous Reactor Fuel Processing.

[b] Includes ARE and MSRE Fuel Reprocessing.

[c] Includes aqueous sulphate fuels (I-IRE), sol-gel, carbide-graphite-oxide spheres (HTGR, EGCR, Rover), and molten salts (MSRE, ARE).

[d] Includes SLURREX, AMEX, DAPEX, MONEX, and other processes.

[e] Includes EXCER, METALLEX, FLUOROX, and DRUHM.

/Includes mechanical methods, DAREX, ZIRCEX, ZIRFLEX, Voloxidation, etc.

[g] Includes TRAMEX, CLEANEX, BERKEX, PLURIX, and others.

efforts undertaken in the reprocessing segment of the fuel cycle for the 1943-1976 period (the period of most development) is given in Table 4.2. The sites of full-scale plants that ultimately resulted from this development effort are also presented in this table. Major segments of reprocessing unit operations were also developed in pilot plants to formulate auxiliary processes for these large plants. Primary among these specific process development programs were the head-end operations necessary to dissolve the irradiated fuel and to remove the iodine and rare gases from the off-gas streams. Table 4.3 indicates some of the significant processes developed in this category.

For many years, ORNL and Chem Tech have been recognized worldwide as a leader in the development of reprocessing technology. In this regard, Building 3019 has played an integral part in each of the spent fuel reprocessing flow sheets used in plant-scale application within the United States (including both defense and proposed commercial facilities). Commercial firms that constructed reprocessing plants that would employ the Purex process, which was piloted in Building 30 19, included the Allied General Nuclear Services (AGNS) Plant at Barnwell, South Carolina, and the Midwest Fuel Recovery Plant (MFRP) at Morris, Illinois, operated by General Electric (GE). In the

Table 4.2. Reprocessing experience at ORNL—separation processes

Development dates	Process	Method	Hot pilot plant	Plant site
1943-1945	Bismuth phosphate	Precipitation for Pu only, from metal slugs	ORNL	Hanford
19461950	Redox	Solvent extraction: hexone for U and Pu from metal slugs	ORNL, Hanford, ANL	Hanford
19461950	Radioisotopes	Precipitation, ion exchange, solvent extraction, absorption, distillation	ORNL	ORNL, industry, Hanford
1946–1952	RaLa	Precipitation for barium/lanthanum	ORNL	ORNL, Idaho
1946-1948	Hexone-25	Solvent extraction for fully enriched U-Al alloy	ORNL	Idaho
1947-1950	Hexone-23	Solvent extraction for thorium and ^{233}U from metal slugs	ORNL	ORNL
1948-1949	Metal recovery	Solvent extraction with tributyl phosphate (TBP) for U sludges	ORNL (recovery of WWII U)	Hanford
1948–1953[a]	TBP-25	Solvent extraction for fully enriched ^{235}U-Al; homogeneous reactor fuel	ORNL	Idaho
194919600	Purex	Solvent extraction with TBP for U and Pu	ORNL (2 plants), KAPL, Hanford	Hanford, SRS , NFS, all foreign plants
1946[a]	Pu ion exchange	Product Pu, ^{235}U	ORNL	
1949-1968	Flouride	F_2 for ^{235}U recovery. Molten salt fuel, MSRE fuel, plate fuel	ORNL, ANL	None
1952	TBP-interim 23	Solvent extraction for ^{235}U recovery only	ORNL	NFS[b] for Con-Ed Spectral Shift, Hanford, SRS
1955-1956	Zirflex	Ammonium flouride dissolution of zirconium fuel	ORNL, Idaho	Idaho, UK
1952–1959[a]	Thorex	^{233}U and Th recovery (2 versions)	ORNL	Hanford, Savannah River
1965–1976	Sol-gel	Solvent extraction and precipitation to prepare ^{233}U	ORNL	LWBR ^{233}U fuel demo (Bettis)
1961–1976[a]	TRU processes	Solvent extraction, ion exchange, precipitation for Am, Cm, Bk, Cf, and Es	ORNL, SRS	SRS for ^{252}Cf

[a] Widely used process.

[b] Nuclear Fuel Services.

Table 4.3. Reprocessing experience at ORNL—head-end and dissolver off-gas processes

Development dates	Process	Method	Hot pilot plant	Plant site
1943–1976[a]	Chemical dejacketing, batch dissolver	Dissolution in *HN03*	All U.S. plants	All U.S. plants
1949-1952	Xe, Kr absorption	Removal of Kr by charcoal absorption; cryogenic distillation	ORNL	Idaho
1955–1976[a]	Mechanical dejacketing	Dissolution with Magnox-clad fuel	Britain, France	Britain, France
1963[b]	Mechanical dejacketing	Fast reactor fuel: Hallam and SRE, stainless clad metal	ORNL	None
1955–1976[a]	Zirflex HF dissolution	Dissolution of zirconium-clad fuels	ORNL, Idaho	Idaho, Eurochemie
1962–1976[a]	Chop-leach	Power reactor fuels	ORNL (cold)	NFS, AGNS, Britain, La Hague, Japan, India
1965–present	Continuous dissolution	Power reactor UO_2 fuels	ORNL (developing)	None; planned for LMFBR
1965–1976[b]	Crush, burn, leach	HTGR and graphite fuels	ORNL (hot cell): Gulf-GA (cold)	Proposed pilot plant for HTGR
1970-1976	Voloxidation and tritium	UO_2 to U_3O_8 for Kr, I_2, tritium	ORNL (hot cell, small scale)	None
1969-1976	Selective absorption of fission gases	Xe, Kr, Co_2 removal with freon scrubbing	ORNL, K-25 (full-scale cold)	None
1970-1976	Iodox, Ag zeolite, mercury nitrate, caustic and $Ag(NO_3)$ scrubbers	Increased ^{129}I, ^{131}I retention	ORNL (hot)	Caustic scrub in all plants; others in or planned

[a] Widely used process.

[b] Not for LWR fuel.

case of the **MFRP,** the flow sheet selected included both solvent extraction and fluoride volatility methods. Both of these concepts were demonstrated in pilot-scale efforts in Building 3019. Exxon was proposing to build a large reprocessing plant in Oak Ridge that would also employ Purex process concepts. Purex technology has continued to evolve throughout the world and has advanced in several nations. Plants currently exist in the United Kingdom, France, Japan, China, and the Soviet Union.

Although not stressed in this document, there are the many side benefits to the government from the experience gained from the operation of Building 3019. **A** partial listing of these benefits is presented below:

1. The training of the Du Pont operating staff assigned to operate the Savannah River plant took place in Building 3019. During the 1952 period, 26 key operations managers were assigned to the facility to train for the production plant operation.
2. Because of the extensive Purex and Volatility experience with irradiated fuel at Building 3019, key members of the building staff assisted the government in the training and testing of commercial reprocessing plant operators [Nuclear Fuel Services (NFS), GE,, AGNS]. In essence, all of the various operators in these plants that were granted government operating licenses were examined by Building 3019 personnel at the plant site. Each of the written examinations given by examiners for all operating licenses was prepared by the Building 3019 staff.
3. Interim production quantities of plutonium, uranium (all isotopes), thorium, and various special isotopes were provided from the operations in Building 3019 and recycled.
4. A large number of technical papers and reports were prepared and presented as the direct result of the experience gained in Building 3019.
5. Because of the experience gained in the building with the handling of highly radioactive materials, personnel from the building served as consultants to the government in numerous capacities. Included in this category are (1) the recovery of weapons debris from the crash of an aircraft in Thule, Greenland; (2) the safeguards evaluation of the **Tokai-Mura** plant in Japan; (3) major on-site assistance to the cleanup of Three Mile Island (TMI); and (4) other assignments too numerous to mention.

4.17.3 Early History and Operating Philosophy Evaluation of Building 3019

As is well known, the Oak Ridge site was selected as part of the famous Manhattan-Project. Hewlett and Anderson described the construction phase of Building 205 (3019) as follows.

> When the Hanford site was fmally selected in January 1943, plans were made to build an air-cooled experimental pile, a chemical separations pilot plant (Building 3019 or Building 205). and supporting laboratories on the isolated tract in Bethel Valley, known as X- 10. Since the Du Pont Company was charged with both the design and construction of X-10, only a few weeks elapsed between the decision to proceed and the'groundbreaking for the first building. Du Pont started the first temporary buildings February 2, 1943, and completed these and the utility installations in March 1943. At that time, sufficient data on the separations plant (Building 3019) were available to permit construction crews to initiate excavation. Two months were required to complete the foundation for the six large underground cells in which the plutonium would be separated from the uranium slugs. With concrete walls several feet thick, the cells would extend one story above ground and would be covered with mammoth concrete slabs which could be removed when replacing equipment. The first cell, linked to the pile building by an underground canal, contained a large tank in which the uranium slugs and their aluminum jackets could be dissolved. The next four cells were designed for the large stainless steel tanks, centrifuges, and piping for the successive oxidation-reduction cycles. The last cell served as a spare for storing contaminated equipment. Stretching alongside the cells was a one-story frame building used for the operating gallery and offices. By June, Du Pont had started the pouring of the cell walls. When the Bismuth Phosphate process was selected, the equipment design function was accelerated. The installation of piping and cell tankage began in September. The testing and extensive modification of process equipment required most of October, but the plant was ready to

operate when the first slugs were discharged from the pile (December 1943).[41]

Since this early beginning, numerous changes have been made to the Building 3019 pilot plant to accommodate the multitude of processes requiring demonstration. Considerable credit should be given to the original designers of the facility to permit this flexibility. Basically, the designers provided a facility and cell structure that could be tested with nonradioactive materials, demonstrate a process with irradiated fuel, collect data, decontaminate equipment to permit its removal, decontaminate the cells, and prepare for the installation of new process equipment. The successful programs accomplished in the building attest to this factor.

As with all maturing technologies, the processes conducted in Building 3019 required the facility to undergo numerous changes over the years. Demands made to improve safety, containment, criticality control, process control, safeguards, and data collection are among the requirements that have been responsible for these changes.

4.17.4 Building 3019 Programs

A listing of the major programs conducted in Building 3019 since it was commissioned in 1943 is presented in Table 4.4. In addition to these programs, other programs involving the development of the ion exchange of plutonium (^{239}Pu, ^{238}Pu) were conducted in the laboratories attached to the main building cells. Analytical procedure development and analysis of the pilot plant samples for process control were accomplished in the analytical cell block on the west side of the building. During the Thorex program, a remote sample withdrawal system was developed for the process system to decrease radiation exposure to the pilot plant operators and the analytical chemists. Samples were remotely transferred from the pilot plant to the analytical cells via a shielded conveyor system located on the roof of the building. For some programs where sufficient decontamination could not be attained in the building's solvent extraction cycle, Building 3505 was used, which contained three additional cycles along with the isolation system for the plutonium product. An underground pipeline was installed between the buildings to permit the processing of certain fuels under these conditions.

A concern regarding the long-range availability of uranium as a nuclear fuel was demonstrated from 1949 to the early 1950s, and the government turned its development efforts toward thorium. In this regard, Building 3019 has played a major and unique role. As is well known, irradiated thorium contains the isotope ^{233}U, which is also a fissionable isotope. Early pilot plant programs with irradiated thorium took place in the building in the 1954-1958 period, during which time 35 tons of thorium was processed as part of the development of the Thorex and Interim-23 flow sheets. A total of 55 kg of ^{233}U (containing 10–40 ppm ^{232}U) was isolated from this material, which required unique storage requirements. Because of the daughter products of ^{232}U contained in the ^{233}U, this material represents a serious gamma radiation hazard to personnel and requires shielding, especially when aged. As the result of the inventory of irradiated thorium in the reactors at both Hanford and Savannah River, the government wisely selected Building 3019 as the "^{233}U National Repository*' in 1962. The Purex plants at both Hanford and Savannah River modified their flow sheets to Thorex and processed a total of 870 tons of irradiated thorium during the 1964-1970 period. The 1400 kg of ^{233}U isolated from these programs was sent to ORNL for storage at Building 3019. Additionally, as instructed by the government, the NFS Plant at West Valley, New York, recovered 1019 kg of uranium from the processing of Consolidated Edison reactor fuel. This material also found its way to the storage facilities at Building 3019. A summary of the thorium-^{233}U processing in the United States is presented in Table 4.5.

4.17.5 Future Plans

Plans for the facility include the following:

- continue as the national repository and dispensing facility for ^{233}U,
- provide development services to the AVLIS program for demonstration of the product conversion by modified direct denitration,
- provide radiochemical laboratories in which waste treatment studies may be performed,
- provide a test bed for demonstration of novel decontamination techniques, and
- provide secure or bonded storage of other valuable radioactive materials, as appropriate.

Table 4.4. Building 3019 pilot plant program

Date	Program	Feed material	Process employed	Material recovered U (kg)	Pu (kg)	Np (g)	Am (g	Irradiation level (MWd/ton)	Cooling months	Remarks and/or references
1943–1945	Weapons	X-10 uranium slogs	Bismuth phosphate					Low		Recover Pu; demonstrate separation process; train personnel
1946-1948	Devcbpment	Enriched uranium	Redox 25 Pmcess					Low		Separate and recover enriched uranium
1950-1953	Purex	Uranium slugs	Purex	~7,500[a]	-7			-500	2–4	Demonstrate Purex process; recover Pu and U; train personnel; provide engineering data
1954-1958	Thorex	Thorium slugs	Thorex	~60[b]				500–5,000[c]	<1–30	Demonstrate Thorex one-cycle, two-cycle, and three-cycle process at hiih "g/t" levels and at shortdecay periods
	High-isotopic purity ^{233}U	Thaex short-decay waste	Modified Interim-23	0.9[b]					12	Demonstrate recovery process and recover ^{233}U containing <0.5 ppm ^{232}U
1958-1960	SCRUP-2	NRX reactor fuel	Purex	5,386	3.1			~400	24	Recover high-quality Pu
	SRPE	SRP fuel	Purex	1.4[d]	15			1,000	-12	Recover enriched U and Pu
	BNL-1,2	BNL reactor fuel	Purex	25,000[a]	18.3			-500	-12	Recover Pu and U; 3019/3-505 complex
	SNAP-A	SRP-U slugs	Purex	3,071[e]	33			~1,000	-6	Recover Pu high in ^{240}Pu; provide wastes fa FP recovery in 3019/3505 compkx
	H-240	SRP-U slugs	Purex	5,800[e]	7.7			-800	3	Recovery Pu hiihin ^{240}Pu in 3019/3505 complex
	S-240	SRP-U slugs	Purex	5,800[e]	13.7			-2200	3	Recova Pu high in ^{240}Pu in 3019/3505 compkx
1958–1960	MTR-1	Pu-Al MTR assemblies	Low TBP		0.5				>6	Recover high ^{240}Pu; 3019/3505 complex
		CP-2 reactor fuel	Purex	4,500				slightly	>12	

Table 4.4. **(continued)**

				Materiel recovered						
Date	**Program**	Feed materiel	Process **employed**	**U (kg)**	Pu (kg)	Np (g)	Am (g)	**Irradiation** kvel **(MWd/ton)**	**Cooling** months	Remarks **and/or references**
1958–1963	**Volatility**	ARE molten salt and fuel	Volatility	**40.6**[a]				slightly	**>12**	**Recovery** enriched **U,** to demonstrate **the** volatiity **process**
		Criticality assembly of molten salt	vohuiity	**72**[a]				**Slightly**	**>12**	**Provide engineering** data
		Zr-U fuel	**Volatility**	**23**[a]				B" -32%	3-7	**Demonstrate** the process with **Zr-clad** assemblies
19604964	**Kilorod**	$^{233}UO_2(NO_3)_2$ • $Th(NO_3)_4$	**U-solvent** ♏☒♦❒♋♍♦♓❑■▤ Th-steam demonstration; sol-gel **preparation; remote** fuel rod fabrication	**37**[b]				**None**	*NA*	To **fabricate** 1,100 **SS-clad** fuel rods **charged** with 3% $^{233}UO_2$**-97%** ThO_2
19694976	**LWBR**	233"NH and $^{233}U_3O_8$	**Purification** (solvent extraction, ion exchange); **oxide** conversion	**1,675**[b]				**None**	NA	To **provide ceramic-grade** $^{233}UO_2$ **of high quality** for fabricating LWBR **fuel**
		$^{233}UO_2$-ThO_2 hard **scrap**	**Thorex dissolution;** solvent **extraction;** ion exchange	**711**[b]				**None**	NA	To **recover** ^{233}U

[a] **Enriched** U.

[b] ^{233}U.

[c] g mass ^{233}U**/ton Th**

[d] **Enriched** U.

[e] **Depleted** u.

Table 45. Summary of thorium-^{233}U processing in the United States

Site	Date (year)	Thorium processed (tons)	^{233}U recovered (kg)	^{232}U content (ppm U)	Flow sheet employed	Remarks
			Irradiated fuel processing			
ORNL	**1954 and 1958**	5	8	**10–40**	Interim-23	Pilot-scale development
	1955-1958	30	47	**10–40**	Thorex	Pilot-scale development up to 4,000 **MWd/ton,** cooled 30 d
	Total	35	55			
SRP	1964-1965	14	107	225	Interim-23	Th discarded
	1965	9	19	38	Interim-23	Th discarded
	1966, 1968, 1969	193	412	6-9	Thorex	Th recovered, 1.5 *M* HNO_3, 0.25 *M* $Th(NO_3)_4$ AF, 30% TBP
	Total	216	538			
Hanford	1965	4			Interim-23	Th discarded, flow sheet test
	1966	250	270	6-10	Acid thorex	Th recovered, acid-deficient feed
	1970	400	589	6-10	Acid thorex	**HNO_3** added below HA column feed plate
	Total	654	859			
Nuclear Fuel	1969	17	1,019[a] (103)	125	Interim-23	15,800 **MWd/ton, Th** discarded, 4.3 M HNO_3, 112 g/L Th used as salting agent
			Unirradiated processing			
ORNL	1962	2	50	40	25% DSBPP	Rod fabrication, Th added to process; recycled
	1973, 1974, 1975	30	1,100	10	5.0% DSBPP-IX	Th added to process; discharged
	1957–continuing	9	225	3-250	2.5% DSBPP	Th added to process
	1957–continuing	9	1,100	3-250		
	Total	50				

[a] **Mixture** of ^{235}U and ^{233}U; ^{233}U is in parentheses.

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4.18 COAL TECHNOLOGY PROGRAM, Hank Cochran

The United States and the rest of the industrialized world were reminded of their dependence on petroleum when in 1973 the Organization of Petroleum Exporting Countries (OPEC) cartel first exercised production constraint in order to effect dramatic increases in the world petroleum price. At that time, ORNL was the only one of the AEC laboratories to have acquired a multiprogram mission—beyond nuclear science and technology-and it was to ORNL that the AEC turned first in seeking technical solutions to the oil crisis. ORNL was asked to focus its efforts on technologies leading to increased and broadened utilization of the nation's vast coal resources; conversion of coal to liquid and gaseous fuels and cleaner and more efficient coal-fired power generation were the objectives.

The AEC gave ORNL permission to "reprogram" available monies from the AEC Applied Technology Division to bring the AEC into the coal technology field. This field had been sustained-like western culture in a few monasteries during the Dark Ages-during two decades of fiscal deprivation at a few small laboratories of the Department of Interior's Bureau of Mines. Principal among these Bureau of Mines laboratories were those at Bruceton. Pennsylvania; Laramie, Wyoming: and Morgantown, West Virginia, Bruceton had developed a modem version of the Bergius process called Synthoil. Morgantown was conducting coal liquefaction research also. Laramie was exploring underground (or in situ) coal gasification.

At that time, Union Carbide operated ORNL for the AEC, Herman Postma was ORNL Director, Don Ferguson was Chem Tech director, and Gene McNeese, Ray Wymer, and Chuck Scott were Chem Tech associate directors. Postma and his associate director, Murray Rosenthal, turned to Chem Tech to create the coal technology program. Interior's Office of Coal Research was at the same time negotiating with Union Carbide for a government-funded "demonstration plant" utilizing Carbide's Hydrocarbonization process for coal conversion. Chem Tech promptly assigned key personnel to initiate reviews and research in coal conversion processes.

Jere Nichols (with Royes Salmon and John Holmes) in his Engineering Coordination and Analysis Section and Bob Hightower of Gene McNeese's Unit Operations Section completed a short engineering and economic survey for Jim Bresee's (formerly from Chem Tech) Applied Technology Division in AEC. The objective was to assess technologies for conversion to coal of Ohio Valley Electric Corp's Clifty Creek Power Station. The study concluded that the application of HRI's H-Oil process to coal liquefaction (H-Coal) appeared technically and economically attractive. Later, Royes Salmon was tasked to initiate a major engineering evaluation of the Synthoil process. John Holmes was tasked to undertake a comparable evaluation of the Hydmcarbonization process. Mike Edwards and Bill Rodgers initiated an evaluation of solid-liquid separation technology for coal liquefaction; this was to be followed by bench-scale experimental studies by Rodgers. John Holmes, Hank Cochran, and Dave Joy initiated a review of carbonization and hydrocarbonization technologies: this was to be followed by bench-scale experimental studies by Cochran (Fig. 4.46)

In Ray Wymer's Chemical Development Section, Jim Mailen reviewed coal conversion technology. Shortly later in Scott's Experimental Engineering Section (formerly McNeese's Unit Operations Section), Hank Cochran was tasked to initiate a bench-scale experimental investigation of Hydrocarbonization; Richard Forrester, to initiate a bench-scale experimental investigation of in situ

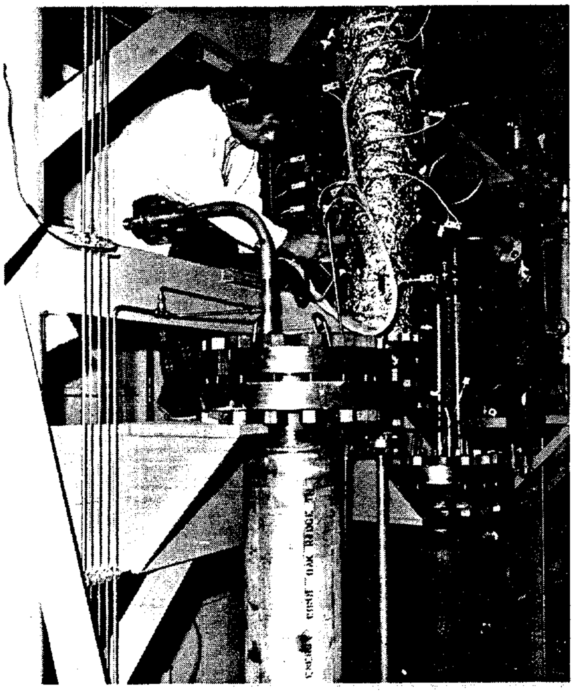


Fig. 4.46. Hank Cochran is testing the experimental hydrocarbonization reactor for leaks with a high-frequency acoustic leak detector, which moderates the supersonic noise made by the leaks into audible range.

gasification; and Bill Rodgers, to initiate a bench-scale experimental investigation of solid-liquid separation. Phil Westmoreland soon began studies of the pyrolysis of 6-in. right circular cylinders of Wyoming coal in support of in situ gasification, Fred Endeiman began atmospheric pressure studies of fluidized-bed coal pyrolysis, and Rodgers started batch filtration and antisolvent deashing studies.

The creation of ERDA (Energy Research and Development Administration) brought the Bureau of Mines labs and the AEC labs together within the same organization. ORNL, operated by Union Carbide, was performing engineering evaluation and bench-scale experimental study of hydrocarbonization, while Carbide was designing a commercial-scale, single-train hydrocarbonization plant. With several oil companies, HRI began construction of a large-scale H-Coal (H stands for the hydrogen used in the process) pilot plant in Catlettsburg, Kentucky. Smaller pilot plants employing variants of the Solvent-Refined Coal (SRC) process were operating in Wilsonville, Alabama, and Ft. Lewis, Washington, and employing different approaches to solid-liquid separation. Exxon and others began construction of a large-scale pilot plant in Baytown, Texas, employing the Exxon Donor Solvent (EDS) process. Our engineering study of the Synlhoii Process concluded that (1) the fixed-bed catalytic hydrogenation reactor was technically infeasible and (2) the use of high operating pressures, residue filtration, and large hydrogen recycle requirements, together with low product value, led to very unfavorable economics. This assessment contrasted sharply with previous published studies by the Morgantown laboratory and the Pittsburgh Energy Technology Center (PETC).

On December 4.1974, Herman Postma and about three dozen engineers and scientists from ORNL met at the Holiday Inn in Oak Ridge to celebrate the entry of ORNL into the coal conversion field. Don Ferguson and Chem Tech were commended by Postma for the proactive initiation of the Coal Technology Program. Jere Nichols had been named Director of the new Coal Technology Program in November 1974; he immediately set about to establish a fluidized-bed coal combustion program in the Engineering Technology Division and catalytic liquefaction studies in the Chemistry Division. Thus, the new coal program at ORNL became a multi-divisional effort.

In September 1977, when McNeese took over the program, it became the Fossil Energy Program, and before it was all over, dozens of Chem Tech engineers and scientists had been involved, including the following: John Begovich (gas-liquid-solid fluidized beds): Cliff Brown and Leonard Dickerson (wet-oxidation of coal conversion wastewaters and hydrocarbonization experiments) (Figs. 4.47 and 4.48); Ron Bnmson (resource recovery from coal ash): Ron Canon (resource recovery from coal ash); Ron Glass and Jim Snider (a myriad of engineering studies and subcontracts); Richard Genung and Teny Donaldson (environmental control technology studies) (Fig. 4.49): Randy Gibson (experimental liquefaction studies); Jim Hewitt (solid-liquid separation experiments); Bob Hightower (a collection of bench-scale experiments); Jerry Klein, Bob Jolley, and Jennifer Brand (several wastewater treatment studies): Doug Lee (both wastewater treatment and coal liquefaction studies) (Fig. 4.50); K. H. Lin (engineering analyses); Bill Rodgers, Bob Jolley, Leonard Dickerson, and Dave McWherter (bench-scale liquefaction and chemical characterization studies); Bill Rodgers and John Mrochek (chemistry of antisolvent

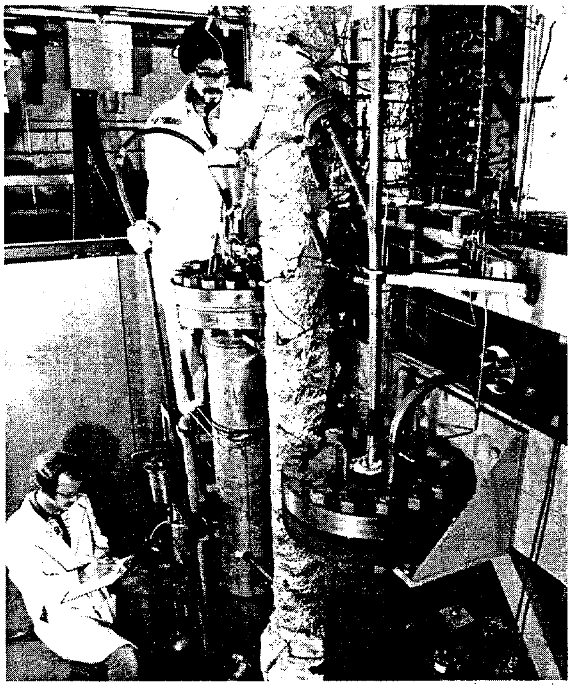


Fig. 4.47. Cliff Brown and Leonard Dickerson record data and adjust the loading of the Chem Tech "bench-scale" coal hydrocarbonization unit in Building 2528.

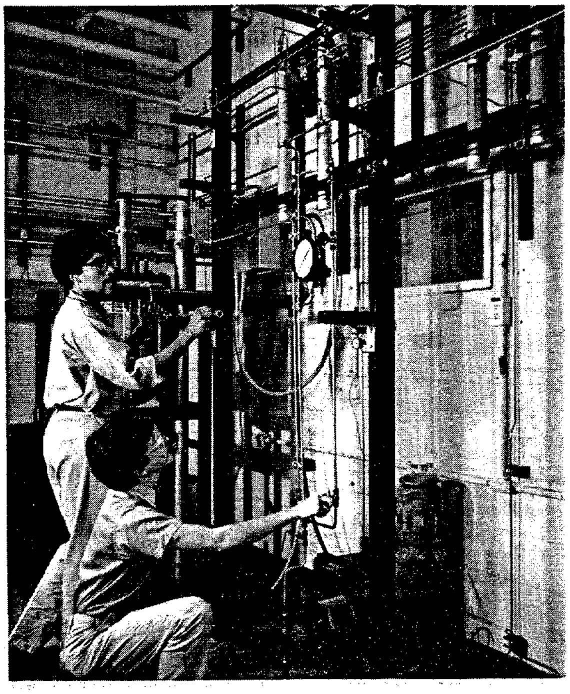


Fig. 4.48. Leonard Dickerson and another Chem Tech engineer adjust the gas flows to the coal hydrocarbonization unit.

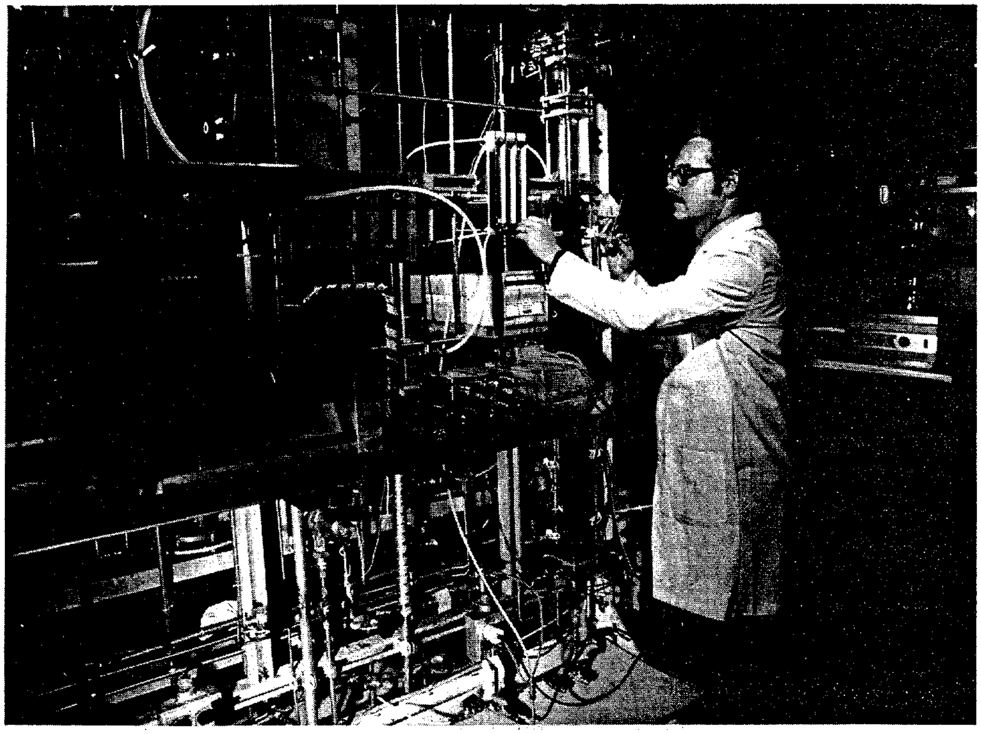


Fig. 4.49. Terry Donaldson adjusts test conditions on an experimental bioreactor used to treat coal gasification wastewater. Degradation rates for dissolved organics are substantially higher in this system than in conventional wastewater treatment systems.

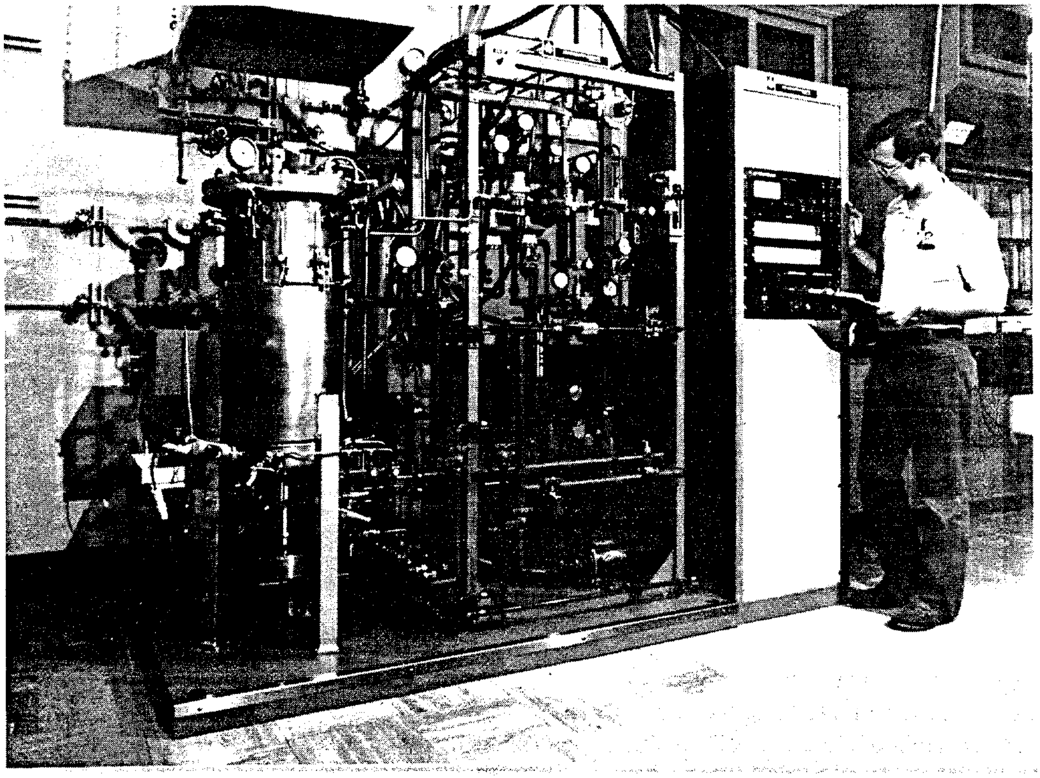


Fig. 4.50. Doug Lee monitors the Chem Tech experimental coal liquefaction unit in Building 4505.

deashing and thermal conductivity of coal liquids); Bill Rodgers and Sidney Katz (solids separations from coal liquefaction products); Royes Salmon (a member of technical and economic assessments); Terry Sams (exploratory liquefaction studies): Suman Singh (several engineering evaluations, particularly in effluent treatment technology); Suman Singh, Bob Jolley, and Leonard Dickerson (bench-scale coal desulfurization studies): Joe Walker (resource recovery from coal ash); Jack Watson (a collection of experimental studies); Bob Wham (engineering evaluations and exploratory liquefaction experiments): Jim Wilson (coal liquefaction experiments): and Lloyd Youngblood (coal liquefaction experiments). This listing omits (for brevity only) the contributions of numerous other former Chem Tech staff members. In addition, the substantial contributions to Chem Tech's program of the staff in Tom Pickel's Engineering group should not be forgotten.

In the early 1980s, interest in coal conversion processes waned with the initiation of petroleum prices being set by the market rather than by governments. From the beginning, the conservative Bureau of Mines laboratories thought of coal studies as within only their purview. There appeared to be some mistrust of the AEC laboratories which persisted through ERDA and DOE. In the end, two of the Office of Coal Research (OCR) laboratories (Bruceton and Morgantown, which had evolved through various names—PERC/MERC, PETC/METC, etc.) had taken over all of the process technology programs within the 'coal program from the former AEC national laboratories, including ORNL. At its peak in about 1980, the ORNL Fossil Energy Program centered around four major coal liquefaction projects, two SRC demonstration plant projects, and the two large H-Coal and EDS pilot plants managed by DOE's Oak Ridge Operations office. The program involved engineers and scientists and support personnel from at least ten ORNL divisions. Today, except for some exploratory microbial coal liquefaction studies in Chem Tech, the remnants of the Fossil Energy program are in the M&C Division.

4.19 BIOTECHNOLOGY R&D IN THE CHEMICAL TECHNOLOGY DIVISION, John Mrochek, Carl Burtis, Zane Egan, Elias Greenbaum, Bob Jolley, Jerry Strandberg, and Charles Scott

Chem Tech has maintained a significant competence in separations science and technology throughout its history. This was the primary reason for the initiation of biotechnology R&D in the division that occurred over 25 years ago. It all started in the winter of 1963 when Connie Chester and Don Kelmers met with Mel Stulberg of the Biology Division to discuss the possibility of large-scale production of several individual transfer ribonucleic acids (tRNAs) for use by the biomedical research community. It was proposed that current laboratory techniques for production, separation, and purification of these biochemicals be enhanced and scaled up and carried out in Chem Tech with consultation and assistance from Biology Division staff.

This new separations effort was subsequently funded by the AEC and the National Institutes of Health (NIH), and an R&D team was then put together in Chem Tech. This included Chester and Kelmers, as well as Chuck Hancher, Herman Weeren, Zane Egan, Joe Weiss, and Ray Pearson, along with several technicians. Over the next four years, the Chem Tech staff proceeded to establish a macromolecular separations effort in which hundreds of kilograms of *E. coli* cells were grown, nucleic acids extracted, and several tRNAs were separated and purified by advanced chromatographic techniques.

Another collaborative effort was begun between the Chem Tech staff and the Molecular Anatomy (MAN) Program, also at ORNL. Dr. Norman Anderson and co-workers in the MAN Program had developed prototype systems for high-pressure liquid chromatography of ultraviolet-adsorbing and carbohydrate constituents in urine. Anderson, Director of the MAN Program, and Chem Tech staff member Don Vissers initiated the collaborative effort early in 1965. Shortly thereafter, Don left the ORNL and Chuck Scott took up this responsibility. The initial thrust of the program was the development of high-resolution separations systems based on column chromatography to separate and quantitate molecular indicators of disease. About a year later, a viable R&D program entitled the Body Fluids Analysis Program was established with support primarily from the NIH. The Chem Tech research team was headed by Scott for several years with participation first by Bob Jolley and Maurice Freeman, followed shortly thereafter by Wilson Pitt, Ken Warren, Carl Burtis, Norman Lee, Stan Dinsmore, Sidney Katz, John Mrochek, and Bill Butts (Analytical Chemistry Division). This group, in conjunction with personnel from the Instrument and Controls Division, developed a series of advanced instrumental concepts that were used for biomedical research and the clinical laboratory. A broad range of support was engendered from several of the institutes of NM, DOE, EPA, and the National Aeronautics and Space Administration (NASA). In 1967 an annual symposium series entitled “Advanced Instrumental Concepts for the Clinical Laboratory” was initiated in cooperation with staff at the NM. This meeting series has since been administered by the American Association for Clinical Chemistry (AACC) and is approaching its 25th year.

Spin-off from the high-resolution chromatographic separations work included application to analysis of drinking water, wastewater, and cooling water. This research was initiated by Wilson Pitt and Sidney Katz, with wastewater analysis; and applied by Bob Joliey and Jim Thompson to chlorinated wastewater and cooling water. Pioneering research coupled the use of radioactive tracers for chlorination with analysis of chlorinated products using chromatography and proved conclusively the formation of chlorinated organics during the chlorination disinfection process. In 1975 a symposium series entitled the “‘Environmental Impact of Water Chlorination” was initiated by Jolley. The series has continued under the sponsorship of federal agencies with the objective of documenting national and international progress in the determination of the health effects of water chlorination.

During the mid 1970s, the biotechnology staff in Chem Tech began to investigate the use of advanced biological processing concepts for the removal or degradation of various types of pollutants from aqueous effluent streams. This was the beginning of the division’s bioprocessing research effort. Support for this effort was obtained from several offices within the ERDA and its successor, DOE. Chuck Scott, Chuck Hancher, and

Wilson Pitt were the early participants, but they were later joined by Dennis Chilcote, Wes Shumate, Richard Genung, and Jerry Strandberg. Within a few years, this bioprocessing R&D effort had expanded into investigation of many advanced bioprocessing concepts for the production of liquid and gaseous fuels and for energy-conserving environmental control technology. A broad, multidisciplinary R&D team evolved during the late 1970s and early 1980s that included biophysicists, biochemists, microbiologists, and biochemical engineers with the addition of Eli Greenbaum, Terry Donaldson, Ed Arcuri, Jonathan Woodward, Brian Davison, and Brenda Faison.

Several bioprocesses were developed and transferred to the industrial sector, and a new symposium series entitled “Biotechnology for Fuels and Chemicals*’ was started. This annual international meeting series is now in its 13th year and is cosponsored by the DOE, Solar Energy Research Institute (SERI), and various industrial firms. Research efforts have included a study of biophotochemical systems, advanced methods for enzyme-catalyzed reactions, innovative bioreactor concepts, biologically oriented environmental control technology, bioprocessing concepts for fuels and chemicals from biomass, and biological processing of fossil fuels.

4.19.1 Macromolecular Separations Program

The Macromolecular Separations Program was a collaborative effort involving Chem Tech, the Biology Division, and the Analytical Chemistry Division during the period from about 1963 through 1973. It was directed by G. D. Novelli of the Biology Division and Don Kelmers of Chem Tech. The program was sponsored by the National Institute of General Medical Sciences and NIH. Bench-scale studies were carried out in Chemical Development Section C, and the scale-up operations were conducted in the Unit Operations Section of Chem Tech; Keith Brown was head of Chemical Development Section C. Two refrigerated laboratories (“cold rooms”) were built for handling the biological materials.

At that time most of the work in the section was concerned with uranium recovery and fission product separation using solvent extraction. Based on this experience, Brown decided that one of the fust things to do was grow a ton of (bacteria) cells. (It should be noted that biochemists at the time were used to working with milligram amounts of material and usually measured ribonucleic acids in optical density units; one milligram was about twenty optical density units.) After recovering from the initial shock, Novelli became excited and enthusiastic about the program. Zane Egan, Ray Pearson, and Joe Weiss, with technician support from Perry Eubanks, Dale Heatherly, and Jim Overton, worked on the bench-scale development. Incidentally, all were trained as inorganic chemists, but they had some experience in developing separations methods. Dave Holladay, Chuck Hancher, Al Ryon, and Herman Weeren, with George Dinsmore and John Taylor, were involved in the scale up.

The overall goals of the program were twofold: development of separations methods and engineering scale-up for the production of purified tRNAs. After a considerable development effort, several reversed-phase liquid chromatographic columns and methodologies were developed to separate individual tRNAs, ribosomal ribonucleic acids, and oligonucleotides. The most successful chromatographic packing was prepared by coating a polychlorotrifluoroethylene powder with quaternary amines. (Interestingly, a similar material was studied at Hanford for the recovery of palladium from nuclear waste solutions.) The chromatographic columns were scaled up to 2 in. in diameter by % in. long and 4 in. in diameter by 30 in. long to produce the first gram quantities of tRNAs.

A small spinoff company was also formed to market the chromatographic packing for a short period. During the program, the supplier of the support material discontinued manufacture of the product, and ORNL became the sole source of the material. Later, similar packings were marketed by private industry.

Sources of the RNAs were calf liver, rat liver, and various strains of *E. coli* bacteria. Half-ton batches of the bacteria were grown in a fermentor in the Biology Division at the Y-12 site and then harvested and brought to Building 4505, where the RNAs were recovered and separated. The RNAs were characterized by the Analytical Chemistry Division using radioactively labelled amino acids, gel electrophoresis, and nucleotide analysis. The Analytical Chemistry support was critical to the success of the project.

In related studies, other separation techniques, including solvent extraction and gel chromotography, were evaluated, and the base sequences of some of the tRNAs were also determined. Many of the products were supplied to researchers in the Biology Division for studies on the structure and function of tRNAs.

Public announcement of the availability of the purified tRNAs, without charge, was made in ***Science*** and in ***Chemical and Engineering News*** in January 1970. Requests for samples averaged one to two per day for several months. The requests were reviewed by a committee of the National Institute of General Medical Sciences, and shipments were made in accordance with their recommendations. Over 14 grams of purified specific tRNAs were produced and distributed in a total of 752 shipments to 208 scientists in the United States and 12 foreign countries, including West Germany, England, the U.S.S.R., Poland, Japan, France, Norway, and Canada. The results of the research and development program at ORNL were reported at various national and international scientific meetings and in over 40 open literature publications. Several visiting scientists from universities in the United States, as well as Australia, Italy, and the U.S.S.R., spent extended periods in our laboratories learning the separation techniques developed during this program.

4.19.2 Body Fluids Analysis

The genesis of the Body Fluids Analysis (BFA) Program in Chem Tech began in 1965 with a collaborative effort between Don Vissers and Norman Anderson, Director of the ORNL Molecular Anatomy Program. Initially, work from this effort was located in the Power Plant at the K-25 Site. When Vissers left ORNL, Chuck Scott assumed the responsibility for the collaborative effort, working closely with Jim Attrill (Analytical Chemistry Division). Scott established the BFA program, which was moved to Chem Tech in 1965. The initial project of the program was the development of high-resolution liquid chromatography, which was followed by development efforts in centrifugal analysis and environmental monitoring.

High-Resolution Chromatography. As a result of Chem Tech expertise in the use of ion-exchange chromatography for separation of isotopes, the first thrust of the BFA program, which was funded by the National Institute of Health General Medical Sciences (NIGMS), was the application of this technology to the separation of biological compounds, namely, the uv-absorbing constituents found in human urine. An initial breakthrough was achieved by Chuck Scott and Norman Lee with the development of an elutriation process which provided ion-exchange beads of very small and uniform particle size. The use of the small particles as a chromatographic support provided excellent resolution but required high pressure to obtain a reasonable flow rate of eluent through the column. The uniformly sized particles lowered the necessary operating pressures to a reasonable 4000–5000 psi.

Consequently, a Chem Tech development group under the direction of Chuck Scott which included Dennis Chilcote, Maurice Freeman, Bob Jolley, Guy Jones, Norman Lee, and Wilson Pitt of Chem Tech: Wayne Johnson and Lou Thacker of the Instrumentation and Controls (I&C) Division; and Bill Walker of the Plant and Equipment (P&E) Division developed analytical systems that were capable of operating at the high pressures dictated by the use of the small-particle, ion-exchange resins. Two types of systems were developed. A urine analyzer that was capable of separating over 140 uv-absorbing constituents from a single urine sample and a carbohydrate analyzer that would separate over 30 sugars of physiological interest. Both systems included a high-pressure sample injection valve and miniature flow-through photometer which were developed by members of this group and become prototypes for high-pressure liquid chromatography (I-IPLC) systems.

To identify the compounds that were separated by this new technology, a large analytical effort was launched with valuable contributions being made by Carl Burtis, Stan Dinsmore, Bob Jolley, Sid Katz, John Mrochek, and Ken Warren of Chem Tech and by Bill Butts and Bill Rainey of the Analytical Chemistry Division. In fact, this group was one of the first in the country to use the emerging technique of mass spectrometry to identify compounds from human urine. A variety of applications were also pursued by this group, and a widely quoted paper authored by John Mrochek et al.[42] was one of the first to demonstrate the use of HPLC in the analysis of biogenic amines and their relationship to various types of cancer. This work engendered support from the National Cancer Institute (NCI).

Considerable interest in the biomedical community resulted from the publicity that this new technology received. Consequently, with NIGMS funding, several systems were fabricated and placed in research laboratories in prestigious institutions, such as the NIH and the medical schools of Duke and Johns Hopkins Universities. In addition, considerable commercial interest was also generated, which resulted in HPLC systems appearing on the marketplace, thus constituting one of the first examples of technology transfer from ORNL to the private sector. In 1971, the Body Fluids Analyzer was presented an IR-100 award as one of the most significant new technical products of the year. Subsequent application studies by Sidney Katz, Wilson Pitt, and Bob Jolley were funded by the Federal Water Pollution Control Administration [which became the Environmental Protection Agency (EPA) during the course of the Chem Tech study] and demonstrated the utility of HPLC in environmental water analysis. In 1973 Bob Jolley used the HPLC ultraviolet analyzer coupled with ^{36}Cl tracer studies to prove that chlorinated organic compounds are formed when wastewater and natural waters are chlorinated for disinfection and biofouling treatment. Also, during this period, a Body Fluids Analyzer (UV Analyzer) was fabricated by the group for the EPA Analytical Laboratory in Athens, Georgia.

Oak Ridge Conferences on Clinical Chemistry. In addition to its technical achievements, the BFA program has also had a significant impact on the practice of clinical laboratory medicine by organizing and starting a very successful conference series on advanced technology for the clinical lab. On November 30, 1967, the first conference entitled “Urinary Constituents of Low Molecular Weight’* was held at ORNL. This conference was sponsored by ORNL and NIGMS. It was organized and chaired by Charles D. Scott (ORNL) and Robert Melville (NIGMS), and the proceedings from it were published in ***Clinical Chemistry.***

This historic conference was the first of what has become an annual conference which is now known as the “Oak Ridge Conference.” The basic objective of this conference series has always been to present and discuss the advanced technology and concepts that could be expected to have an impact on the practice of clinical chemistry. Consequently, the theme of these conferences has become ‘Tomorrow’s Technology Today.” Reviewing the program of past conferences indicates the successful achievement of this objective, as most of the analytical techniques and instruments that are now routinely used in clinical laboratories were presented and discussed at earlier conferences. For example, various types of high-performance chromatography, mass spectrometry, sensors, advanced analytical systems, chemometrics,, advanced immunoassays, monoclonal antibodies, DNA probes, capillary electrophoresis, and robotics all were discussed at prior conferences.

A second objective and goal of this conference series has been to disseminate the information presented to a wide clinical audience in a rapid and timely manner. Consequently, the presentations that are given in early spring have all been published in peer-review journals in the same year as they were presented. With the exception of the 1969 conference, whose proceedings were published ***in*** the ***American Journal of Clinical Pathology,*** all of the previous conference proceedings have been published in ***Clinical Chemistry.***

As mentioned previously, the early conferences had a definite Tennessee “flavor” as they were organized, sponsored, and managed by Chem Tech staff at ORNL. However, various other organizations co-sponsored and helped organize the early conferences, including NIGMS (1967–1978), the Academy of Clinical Laboratory Physicians and Scientists, AEC (1974). ERDA (1975–1977), and DOE. In 1979, the AACC joined ORNL in co-sponsoring the conferences through 1984. In 1985, the AACC assumed full sponsorship of this conference series and is currently responsible for organizing and managing them. In 1993, the conference will celebrate its 25th anniversary and will be held in Oak Ridge, its original home.

Centrifugal Analysis. The genesis of the centrifugal analysis program in the Chem Tech, again, was traceable to the division’s collaborative efforts with Norman Anderson. A conference on the future instrumental needs of clinical laboratories was organized by Bob Melville of the NIGMS and held in Quail Roost, North Carolina in 1966. Shortly thereafter, Dr. Anderson invented a new approach to automated analysis which has become known as centrifugal analysis. Basic patents were filed for and several publications appeared, beginning in 1968, which described the technology.

Basically, a centrifugal analyzer is a multicuvette spectrophotometer or spectrofluorometer in which a unique multicuvette is mounted in a modified centrifuge. By spinning the rotor, the analyzer takes advantage of the centrifugal field generated to simultaneously transfer and mix several aliquots of sample and reagent into their respective cuvettes. Thus, an identical starting time is established for each and all reactions and factors such as time, temperature, and reactant composition which affect each will effect all reactions simultaneously. In addition, the rotary motion of the spinning rotor is used to move the cuvettes through a stationary optical system. Thus, the contents of each cuvette in the spinning rotor can be rapidly monitored at timed intervals as low as once every 50 milliseconds. An on-line computer is used to rapidly acquire and process the data generated by a centrifugal analyzer. (Note: this rapid generating, acquiring, and processing of the data produced by a centrifugal analyzer led them to initially being called centrifugal "fast" analyzers.) The first analyzers were also known as GEMSAEC analyzers to denote that the initial funding for their development came from the National Institute of Health General Medical Sciences and AEC (Fig. 4.51).

The first prototype of a centrifugal analyzer was fabricated in 1968 at K-25 under the direction of Norman Anderson, Don Hatcher, Dick Willis, and Warren Harris. In 1969, the program was moved to ORNL and Chem Tech under the direction of Chuck Scott and Carl Burtis. Over the years, several staff members worked on this project, including Bill Bostick, Richard Genung, Norman Lee, Roy Lovelace, Jim Mailen, John Mrochek, Jim Overton, Wes Schumate, Tom Tiffany, and Mel Watsky. In addition, valuable support was provided by Martin Bauer, Jim Jansen, Wayne Johnson, and Lou Thacker from the I&C Division and from Bill Walker from the Plant and Equipment (P&E) Division.

To demonstrate feasibility and utility, the first prototype centrifugal analyzer was placed in the clinical laboratory of the ORNL Health Division and several clinical assays were developed for it. It was then placed into routine operation where it and subsequent models were operated through 1986 (Fig. 4.52).

It became obvious immediately that centrifugal analyzers had many advantages over the currently available instruments used for clinical analyzers, and the American Instrument Co., Electro-Nucleonics, Inc., and the Union Carbide Corp. were licensed to produce them commercially. The collective sales of these three companies have been over 2000 units worldwide. The analyzer received an IR-100 award in 1969 as one of the 100 most significant new technological products of the year.

In 1972, NASA funded the development of a miniaturized version of the analyzer with the expectation that it would be used as an on-board analyzer in Skylab B (Fig. 4.53). Although Skylab B was canceled for funding reasons and the analyzer therefore never flew, the development was successful, as prototype units were fabricated and used in the clinical laboratories at the Johnson Space Center, ORNL Health Division, Albert Einstein Hospital, and the NIH, with funding provided by the EPA, the Food and Drug Administration, and AEC. Additional prototypes were fabricated and used for genetic screening in the research laboratories located at the University of Michigan and the National Center for Toxicological Research.

The miniaturized version of the analyzer also generated considerable interest from the private sector, with Electro-Nucleonics, Inc., Instrumentation Laboratories, and Roche Laboratories receiving licenses to produce the units commercially. It is estimated that over 4000 of these analyzers have been sold worldwide.

With continuing funding provided by NASA, a third-generation unit was developed which featured an on-board integrated computer and portability, and in 1977, this unit received an IR- 100 award. In recent years, rotors capable of processing whole blood samples and rotors useful for immunoassay have been developed. This technology has been licensed to Abaxis of Mt. View, California.

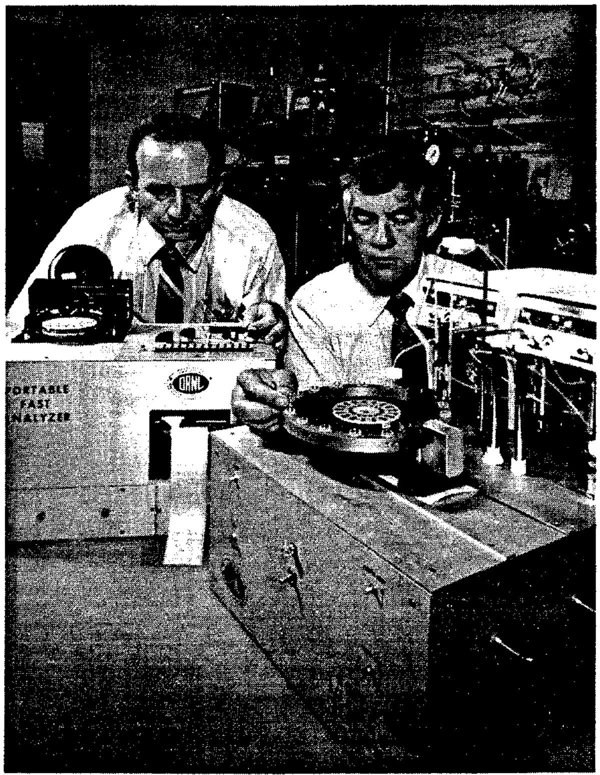
4.19.3 Environmental and Bio-Monitoring

As a result of active application efforts, whose goals were to expand and extend the use of the newly developed chromatographic and centrifugal technologies, the staff of the BFA program developed an interest and expertise in various types of environmental analysis. Consequently, when funding for the body fluids and centrifugal analyses was reduced or ended in the late 1970s, work continued on the development of environmental monitoring technology with funding from the DOE Office of Health and Environmental Research (OHER). A portable monitor, termed the spill spotter, and ultrasensitive bioassays to detect toxic chemical exposure were developed.

Spill Spotter. As a result of a discussion with a visitor, Wilson Pitt suggested to newly hired Chem Tech biophysicist, Don Shuresko, that it should be possible to remotely detect strongly fluorescent polycyclic aromatic hydrocarbons (PAHs). With

Fig. 4.51. Chem Tech development team members with early centrifugal analyzer prototype. Seated are Wayne Johnson (I&C Division) on the left and Chuck Scott. Standing from left to right are John Mrochek, Norman Lee, and Richard Genung.

Fig. 4.52. John Mrochek (left) and Carl Burtis adjust a centrifugal analyzer for clinical chemistry analyses In the ORNL Health Division.



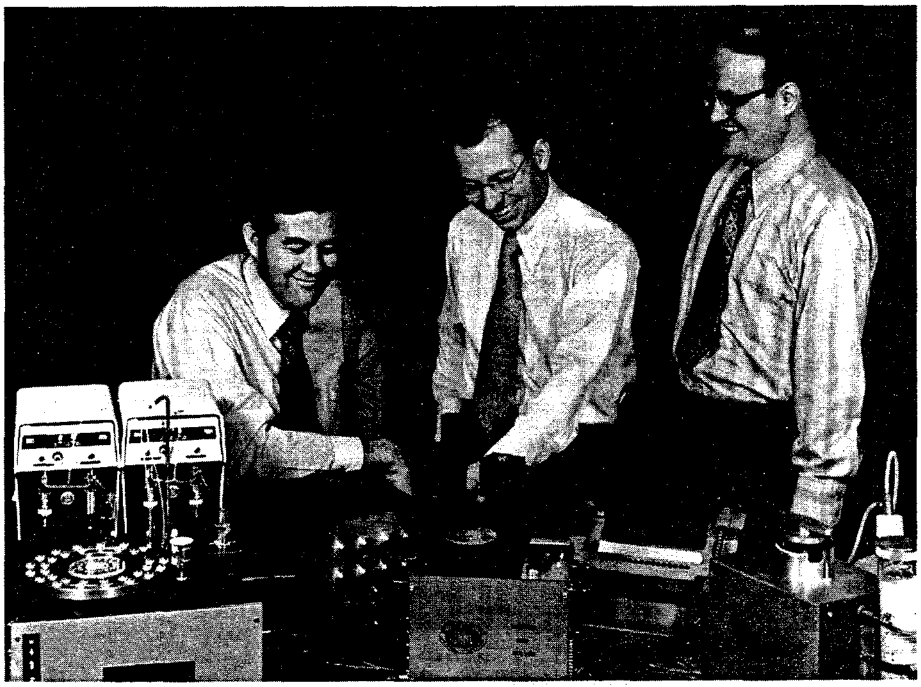


Fig. 4.53. Carl Buttls (left), Wayne Johnson (center), and Jim Mallen are pleased with the performance of the prototype miniaturized centrifugal analyzer develop&J for Skylab use.

Pitt's encouragement and suggestions, Schuresko worked out the basic concept. Under the guidance of Pitt and Chuck Scott, Schuresko led a multidivision effort that resulted in the development of a portable fluorescence monitor, termed a Spill Spotter, whose purpose was to detect surface contamination by PAHs resulting from liquid spills in coal conversion plants. In addition to Pitt, Scott, and Schuresko, other Chem Tech personnel involved in its development were John Mrochek, Zane Egan, Jim Overton, Norman Lee, Guy Jones, and Carl Burtis. Technical assistance was provided by Mike Blair, Gerry Schulze, David McNeilly, Richard Hutchens, and Martin Bauer (I&C Division), Bill Walker (P&E Division), and Mike Holland (Biology Division).

The Spill Spotter operates by projecting an exciting beam of ultraviolet (uv) light onto a surface. If PAHs are present, the emitted light resulting from their fluorescence is detected and quantified by the Spill Spotter. Prior to the development of this device, such monitoring was performed by turning off the overhead light and scanning the suspected surface with a uv ("black") light, a technique which was cumbersome, insensitive, nonspecific, and only qualitative. The Spill Spotter offered several advantages over this monitoring technique, including: (1) capability to operate outdoors in direct sunlight or indoors in the presence of a strong background illumination, (2) provide a quantitative measure of the fluorescing material, (3) discriminate between the fluorescence of organic and inorganic compounds, and (4) did not present a vision hazard to monitoring personnel.

Evaluation of the performance characteristics of the Spill Spotter by Chem Tech personnel demonstrated that it was capable of measuring PAH concentration at the levels where significant biohazards could occur. For example, the device was shown to have a linear dynamic range for perylene (a typical PAH) from 0.1 to 10 g at a distance of 50 cm with a lower limit of detection of 0.001 g. Since 10-g doses of several PAHs are cytotoxic, it was evident that the Spill Spotter had the prerequisite sensitivity for operation for levels expected to be found in coal conversion plants. An unexpected application of the device was demonstrated when John Mrochek and Ronald H. Hill of the Ashland Synthetic Fuels, Inc. (ASFI), H-Coal Pilot Plant Project demonstrated that it was capable of detecting and quantitating skin contamination by PAHs.

To demonstrate the monitoring utility of the Spill Spotter under field conditions, several prototypes were fabricated and tested at the SRC pilot plant in Ft. Lewis, Washington, at the coal gasifier plant in Duluth, Minnesota, and at the H-Coal Pilot Plant in Ashland, Kentucky. These field evaluations demonstrated that the Spill Spotter was capable of performing reliably in the environments for which it was developed. In 1980, the Spill Spotter received an IR-100 award as one of the most significant new technical products of the year.

Bio-Monitoring. An increasing concern about occupational and environmental exposure of personnel to toxic chemicals and potential carcinogens led a group consisting of Zane Egan, Carl Burtis, and Norman Lee in collaboration with, John Kao and Mike Holland of the ORNL Biology Division and Guy Griffin of the Health and Safely Research Division to develop instrumentation and ultrasensitive bioassays for detecting exposure to such compounds. PAH compounds have been implicated as primary carcinogens in coal-derived products. These compounds are metabolized in mammals by a cytochrome P-448 enzyme system, termed the mixed function oxidase (MFO) system. Induction of this system by toxic chemicals can be measured and used as an indicator of exposure. Consequently, a centrifugal analyzer equipped with an argon-ion laser light source was successfully used to quantify cytochrome P-448 MFO activity in mouse liver microsomes after exposing the animal to different doses of liquids derived from a coal liquefaction process. The P-448 MFO activity was determined kinetically by measuring the rate at which the highly fluorescent compound, resorufin, was produced by conversion of 7-ethoxyresorufin substrate. The 5 14.5-nm laser excitation beam was directed with a fiber optic bundle from the laser to the cuvettes of a specially designed and fabricated rotor consisting of a black, acrylic plastic center body with transparent acrylic cuvette windows. The emitted fluorescence, filtered through a 560-nm cut-on filter, was monitored with a photomuitipier tube located 90" to the incident beam. A dedicated computer was used for instrument control as well as data acquisition, computation, and interpretation. A dose-response relationship was demonstrated between MFO activity and the exposure of test animals injected with various materials, including coal liquids and Arocior mixtures. The use of a laser excitation source allowed very low MFO activities to be measured in physiologic samples. This work began in 1980 and was terminated in 1985, when funding by OHER ended.

4.19.4 Bioprocessing

The initial entry of Chem Tech into research and development efforts in bioprocessing research occurred in about the 1973-1974 time frame. Initial programs were in the areas of bioreactor development, particularly as applied to waste treatment technology, and more fundamental studies of photosynthetic hydrogen production. As these programs evolved and expanded, the number and diversity of the staff increased and also many new initiatives were undertaken. The program has been very dynamic, with the number and types of projects fluctuating as new ideas were rapidly assessed for their long-term potential and then further pursued or abandoned. Currently the staff includes biochemical engineers, microbiologists, a biochemist, and a biophysicist (Fig. 4.54).

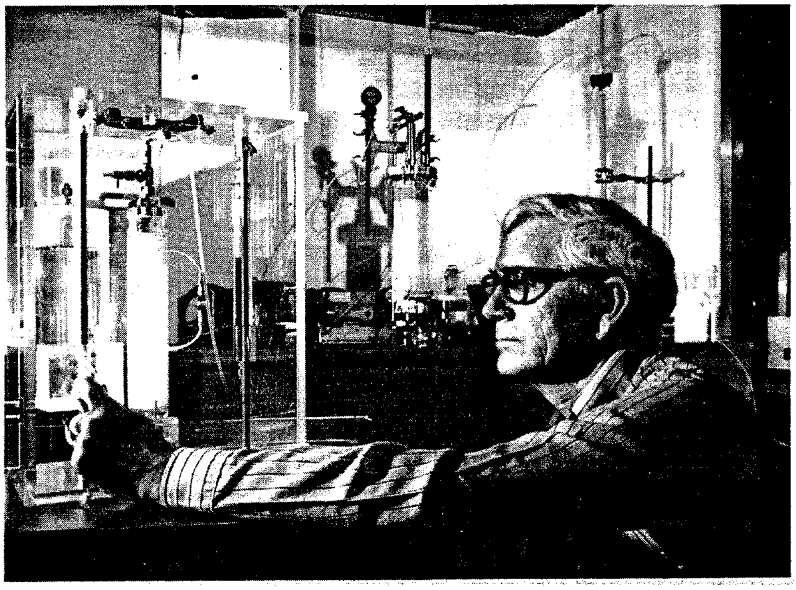


Fig. 4.54. Guy Jones adjusts the eluent flow through a chromatographic column during the separation of bioreaction products.

Bioreactors. The use of activated sludge bioreactors employing very large tanks or ponds and lengthy retention times was a well-known bioprocessing concept in the 1970s. However, Chuck Scott, Chuck Hancher, Dave Holladay, Dennis Chilcote, and George Dinsmore pioneered the application of tapered fluidized-bed bioreactors (TFBRs) to the cleanup of aqueous effluents from coal conversion processes and the denitrification of aqueous nuclear processing waste streams. Experimental comparisons of phenolic waste degradation by continuous stirred-tank, packed-bed, and TFBRs demonstrated the volumetric efficiency advantages for the latter two technologies. Continuing developments in the application of TFBRs to bioprocessing led to an IR-100 award in 1979 for Chuck Scott, Doug Lee and Chuck Hancher.

Photosynthetic Hydrogen. During the early 1970s, an increasing interest in a hydrogen economy where hydrogen was employed as the medium of energy transport resulted in enhanced efforts to develop alternative methods of production. Zane Egan, Perry Eubanks, and Chuck Scott initiated research efforts on a program to develop enzyme-catalyzed production of hydrogen. The initial emphasis of this BES-funded program was on determining the kinetics of hydrogen production using enzymes from ***Clostridia*** and the necessary components from plants such as spinach chloroplasts. The approach was to develop a non-living system which was more amenable to scaleup for hydrogen production. Some interesting tales evolved from the "cultivation" of spinach on B corridor of Bldg. 4500N under the watchful eyes of Egan and Eubanks.

This program and its agricultural efforts was passed on to Eli Greenbaum, a biophysicist who joined the division in 1979, coming from the Union Carbide Corporate Research Laboratory in Tarrytown, New York. Agricultural efforts were replaced by trips to the local grocery store to buy spinach as a source for chloroplasts. One of the key advances made by Eli working with Perry Eubanks was the first demonstration of simultaneous photoproduction of molecular hydrogen and oxygen in the chloroplast/ferredoxin/hydrogenase system. They also demonstrated that a simpler photosynthetic water-splitting system could be prepared by precipitating metallic platinum onto the surface of photosynthetic membranes. This work was featured on the cover of ***Science*** magazine in 1985. Another approach to this goal of hydrogen production, funded by the SERI and the Gas Research Institute (GRI), involved the use of intact microalgae for water splitting. Eli worked with Steve Blankenship, Duane Graves, Jean McInnis, Dave McWherter, Mike Morrissey, Mark Reeves, Ginger Tevault, and Jim Thompson on these projects (Fig. 4.55).

Jonathan Woodward, a biochemist/enzymologist, joined the division in 1980. His work on this program has focused on the stabilization of spinach chloroplasts for the photobiological production of hydrogen (Fig. 4.56). Glenna Shields, Jim Overton, Mike Morrissey, and Norman Lee collaborated with Woodward in this work. In addition, several undergraduate students, high school teachers, and college faculty have spent between three and eight months in Woodward's laboratory between 1980 and 1992. Many of these people have had positive experiences which have aided them in achieving career goals (Figs. 4.57 and 4.58).

Biodenitrification. As mentioned previously, one of the focal points of the division's entry into the bioprocessing research area included the development of TFBRs by Scott, Hancher, Holladay, Chilcote, and Dinsmore. These reactors employed biofilms of bacteria which naturally attach to a fluidizable solid matrix (in this case, anthracite coal). One of the first attempts at application of this bioreactor design to an existing problem was the biodenitrification of nuclear processing wastewaters. Based on earlier studies by Chet Francis in the Environmental Sciences Division (ESD), who showed that certain microorganisms could metabolize the nitrate in these wastes to innocuous nitrogen gas, this process was readily adapted to the TFBR. Continuous development efforts on this bioreactor over the years led to its successful application at the Femald facility and the Portsmouth Gaseous Diffusion Plant and, in 1989, to the startup of a pilot unit at Pacific Northwest Laboratory. Chem Tech staff were active in the design, startup, and where necessary, troubleshooting for these units. In addition to those mentioned above, several other staff members were involved at various stages in this development effort, including Terry Donaldson, Joe Walker, Hal Jennings, John Parrott, Jr., Tom Dinsmore, and Doug Lee.

Biotreatment of Wastewaters from Coal Conversion Processes. This development effort had its beginnings in the same time frame as the

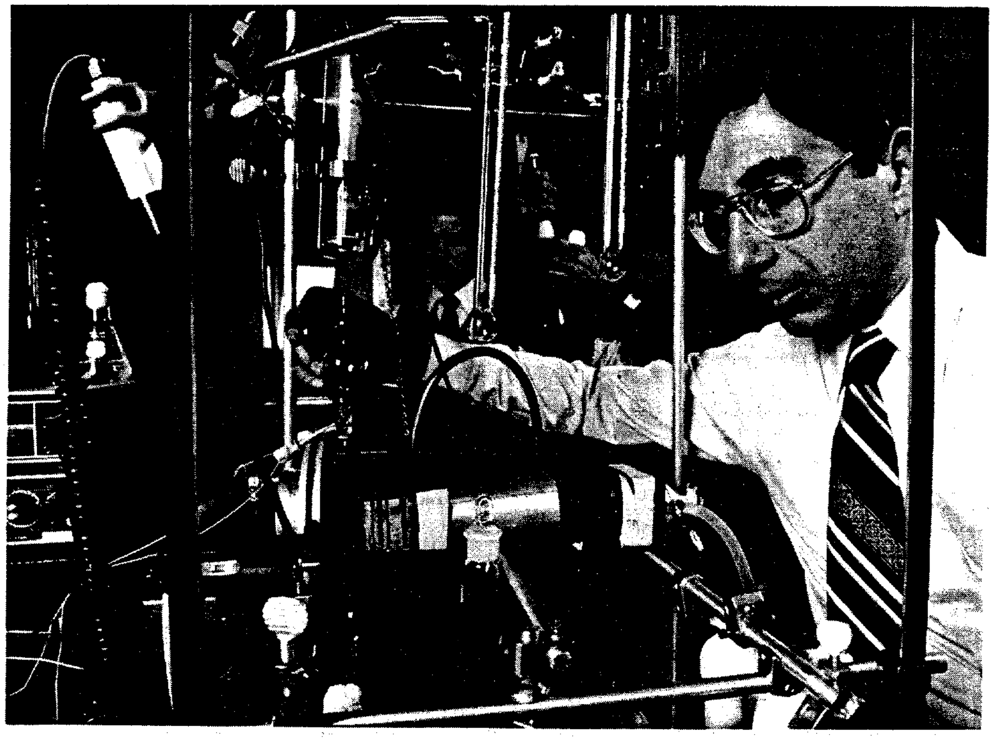


Fig. 4.55. Chem Tech researchers **Elias** Greenbaum in the foreground and Mark Reeves (left) with Jim Thompson in the background work in an experimental facility used to study photosynthetic splitting of water.

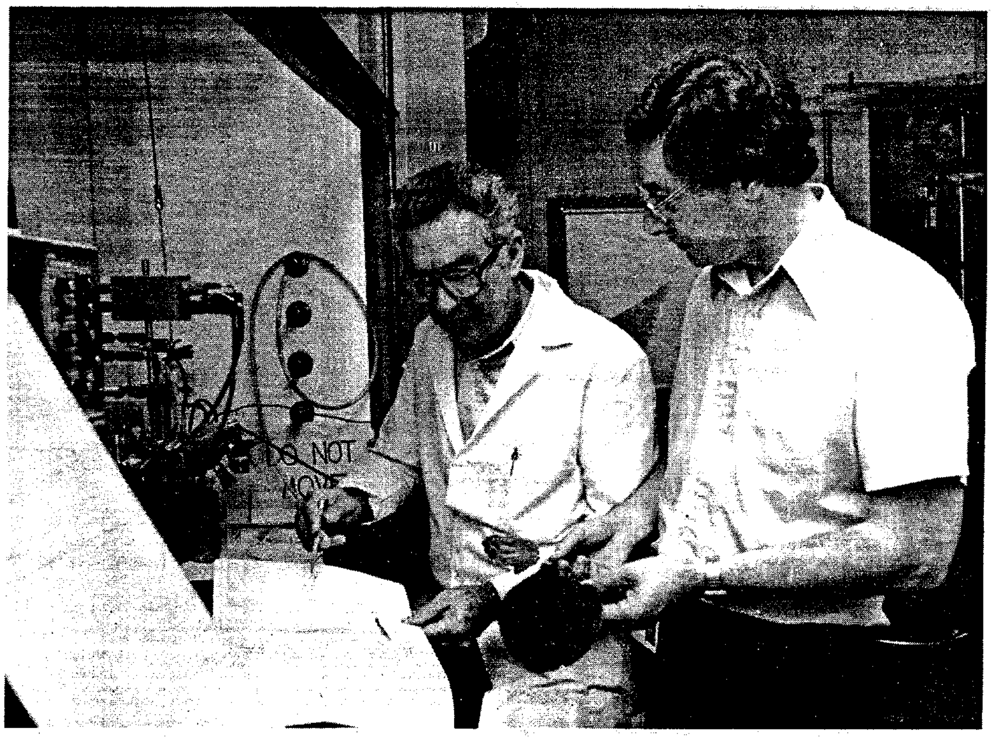


Fig. 4.56. Perry Eubanks (left) and Jonathan **Woodward** discuss data on the biophotolysis of water.

Fig. 4.57. Students and teachers under various **ORAU/DOE** programs working with Chem Tech scientists Norman Lee and Jonathan **Woodward** during the summer of 1989. From left: Steven **McNair,** Technology intern, Peliissippi State; Michelle Clark, Teacher Research Associate, Marquette University High School, Milwaukee, Wisconsin; Jonathan Woodward; Jeffrey Carmichael, ORSERS student, Slippery Rock University, Slippery Rock, Pennsylvania; Norman Lee; Kathy Kapps, American Chemical Society SEED student, .Ciinton High School, Clinton, Tennessee; Delia **Truett,** ORAU Summer Research Participant, Washington and Lee Unlversity.

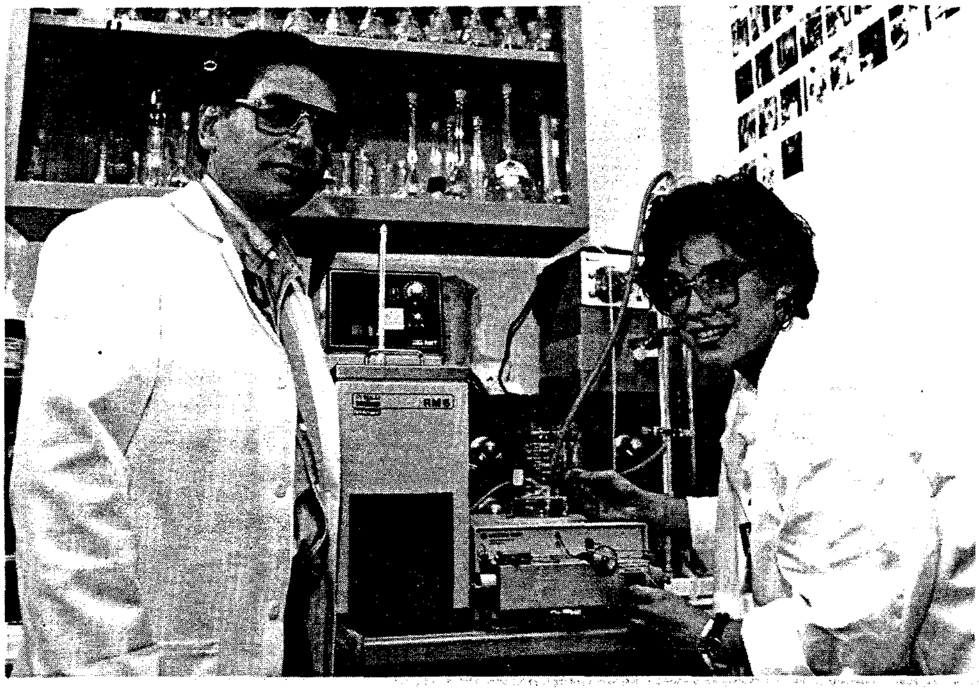


Fig. 4.58. Jonathan **Woodward** and SEED student Lourdes Hemandez, Central High School for the Visual Arts, Carolina, Puerto Rico, during the summer of 1991.

biodenitrification project. It too was an effort to extend the application of TFBRs. The initial work was conducted by Chuck Scott, Chuck Hancher, Dave Holladay, Dennis Chilcote, and George Dinsmore and included studies on phenol degradation in both a continuous stirred-tank reactor and a TFBR. Later, Doug Lee, Chuck Hancher, and Chuck Scott began looking at the biodegradation of a number of compounds, including phenolics, in coal conversion wastewaters using the TFBR. Around 1977-1978, the commercially available culture they were using suddenly lost its ability to attach to the anthracite coal support medium. Ed Arcuri, who had recently joined the bioengineering research effort, undertook a lab study of what causes these organisms to attach to solid matrices. Ed, along with Sherry Gibson and Dave McWherter, were able to reestablish an attached film and resumed studies on phenol degradation. Around 1980, Terry Donaldson and Jerry Strandberg took over the project. Personnel at Sybron Biochemicals, who provide commercial cultures, helped to provide a relatively easy method to induce most, if not all, cultures to attach to surfaces. It involved simply supplying the organisms with cheese whey and mineral salts during the attachment phase. This procedure greatly assisted later studies of fixed-film systems. With technical assistance from Jim Hewitt, Glenna Zachary (Shields) and Mark Worden (a UT chemical engineering graduate student), the study of the biodegradation of various phenolics in coal gasification wastewater (obtained from the Morgantown Energy Technology Center) was renewed in earnest. Doug Lee returned to the project around 1985 and extended the study to include the degradation of a variety of specific organic compounds in the wastewater. A lack of sponsor interest and funding ended CTD's involvement in this development project around 1986-1987.

ANFLOW. Another significant bioreactor development that began in the mid 1970s was the ANFLOW system or "anaerobic, up-flow" bioreactor for treating organics in municipal and industrial wastewaters. Rather than a TFBR, this system employed microorganisms attached to a stationary support matrix. It has the significant advantage that a by-product of biodegradation, methane, can be recovered and reused as a fuel, thus helping reduce operating costs. Alicia Compere and Bill Griffith carried out initial bench-scale studies. Progress was rapid and in November of 1976, in spite of record below-freezing temperatures, Chuck Hancher, Dan Million, Wilson Pitt, and George Dinsmore successfully started up the 5000-gpd pilot demonstration unit at the East End Sewage Treatment Plant in Oak Ridge. They undoubtedly recall recovering copious quantities of cow rumen contents, which served as a source of microbial inocuium for the bioreactor. This unit successfully treated raw sewage, essentially continuously, for two years, even though on two occasions "midnight" discharges from plating facilities caused month-long periods of deactivation of the Sewage Treatment Plant activated sludge. In 1981, Richard Genung, Chuck Hancher, Mike Harris, George Dinsmore, Hal Jennings, and Tom Dinsmore initiated the operation of a 50,000-gpd system at the Loves Creek Wastewater Treatment Plant in Knoxville. This technology proved quite successful and was eventually commercialized.

Biological Separation of Hydrogen Isotopes. In the mid 1970s, a U.S. Geological Survey (USGS) report claimed that a specific marine bacterium had the capability to preferentially utilize tritium as opposed to normal hydrogen. Under the intense interest and scrutiny of the sponsor (ERDA), a concerted effort was undertaken by Chuck Scott, Wes Shumate, and John Parrot, Jr., to verify this phenomenon and study its potential for removing tritium from aqueous nuclear wastes. Jerry Strandberg joined this effort in late 1976. After examining innumerable biological mechanisms capable of directly interacting with hydrogen, the somewhat frustrating conclusion was reached that biological systems (including the original USGS bacterium) tend to specifically exclude tritium from biochemical reactions when compared to the normal hydrogen isotope. From a process standpoint, this exclusion principle still offered a possible, though highly impractical, means of removing tritium from contaminated water. The project was discontinued after little more than a year's effort.

Biosorption. In the mid-1970s, existing processes for removing and concentrating radionuclides and heavy metals from waste streams were costly and relatively ineffective at low metal concentrations. Wes Shumate and Tom Dinsmore initiated studies on the use of microorganisms, which were known to be capable of accumulating certain metals, to remove uranium and strontium from nuclear processing wastewaters. The process was generally considered to be akin to an ion-exchange phenomenon and called "Biosorption." This effort was, like the denitritication development project, part of an overall ERDA-sponsored program for the "Removal and Separation of Radioactive Nuclides and Nitrates from Liquid Waste Streams." Around 1978, Jerry Strandberg and John Parrott, Jr., became involved in these studies. Unfortunately, in spite of ORNL's worldwide recognition for expertise in this area, there was a hiatus due to a cessation of funding for the program. Fortunately, renewed interest developed and work was resumed around 1987–88 by Brenda Faison, Brian Davison, and Jack Watson, with a particular effort toward the use of cells immobilized in gel beads. Studies have continued and have achieved an international flavor through a project directed at the removal of uranium from contaminated waste ponds in eastern Germany.

Bioprocessing of Coal. Chemical/physical processes for converting coal to liquid and gaseous fuels require extremes of temperatures and pressures. The discovery in 1982 by researchers at the University of Hartford of a fungus that could solubilize lignite coal led to a flurry of activity around the country and within the Chem Tech to further investigate this potentially important phenomenon as an alternative method of processing coal to useful fuels and chemicals. Chuck Scott, Jerry Strandberg, and Susan Lewis conducted the early studies which led to important advances including the extension of organisms known to solubilize coal to include other fungi and bacteria and in the understanding of the process, including a primary mechanism of coal solubiiization (i.e., the action of a microbiaily produced organic base). Unfortunately, a primary disadvantage to the use of the microbialiy solubilized product is its high oxidation state. Brenda Faison and Brian Davison continued these studies in the late 1980s, with a particular emphasis on the anaerobic microbial conversion of the oxidized product to methane. In part, because of the high oxidation state of the product, there has also been considerable interest in conversion of coal under anaerobic conditions, particularly by isolated enzymes functioning in nonpolar solvents. The products of such a conversion should be in a more reduced state and considerably more useable in terms of their fuel and chemical value. This has become the major thrust of current research by Chuck Scott and Charlene Woodward.

Enzymatic Hydrolysis of Cellulose. Jonathan Woodward joined the bioprocessing team in 1980. He immediately initiated a program on the enzymatic conversion of cellulose to glucose, an important substrate for various fermentation processes including ethanol production. Jonathan has maintained an active research program in this area over the years, and his work has provided several insights into the mechanism of cellulase activity. As indicated earlier, Jonathan has provided opportunities for a host of students and university faculty to conduct research at ORNL.

Anaerobic Digestion of Cellulosic Wastes. The rapid loss of available landfill space prompted an effort by Jerry Strandberg and John Parrott, Jr., in 1981 to investigate the reduction of volume of the cellulosic fraction of low-level contaminated waste by anaerobic microbial digestion. The idea was to "solubilize" the major portion of the waste and to dispose of the digested material via the hydrofracture waste disposal process. Successful bench-scale results led Doug Lee and Terry Donaldson to scale up the process to a 50-L fermentor. A 25,000-gal digester was designed and scheduled for installation at the east end of ORNL. The unfortunate problems with the hydrofracture facility around this time led to the consideration of disposal of the digest in the grout program. However, it was eventually decided to ***retain*** compaction and burial as the means of disposing of contaminated cellulosic wastes, and the anaerobic digestion program was discontinued

Ethanol Production. Around 1979-1980, Ed Arcuri, Wes Shumate, Chuck Scott, and Jerry Strandberg initiated bench-scale, continuous, bioreactor studies on ethanol production. An additional twist to this development effort was the use of the bacterium ***Zymomonas mobilis***, which has inherently better characteristics for ethanol production than the commonly used fermentation yeasts. The organism also had a propensity to naturally floc and provide its own mechanism for immobilization. A variety of bioreactor designs were investigated and substantially higher ethanol productivities were achieved by the combined use of bioreactor design and ***Zymomonas.*** Brian Davison, Jim Thompson, and Chuck Scott have continued these development efforts using more refined immobilization techniques, including entrapment in gel beads. They were also successful in establishing a cooperative venture with A. E. Staley Co., in Loudon, Tennessee. The use of gel beads as a means of cell and enzyme immobilization technique has proved quite successful. The scope of the work has also expanded to include bioreactor studies on acetone-butanol production and organic acid production.

Bioremediation. Chem Tech's most recent foray in biotechnology is in the area of bioremediation-the use of microorganisms not to treat waste streams, but to clean up "natural*' environments such as soil and groundwater aquifers contaminated with toxic materials. The effort began with a study by Terry Donaldson, Jerry Strandberg, Phil McGinnis, and Tom Ashwood (ESD) on the efficacy of the biological remediation of a jet fuel contaminated site at the McChord Air Force Base in Tacoma, Washington. Subsequently, with funding from Y-12, HAZWRAP, and the ORNL director's fund, and in cooperation with members of ESD, the Oak Ridge Research Institute, and The University of Tennessee, projects were initiated on the biodegradation of PCBs, the biodegradation of mixtures of organic compounds, and the use of bioreactors employing methanotrophic bacteria to degrade trichloroethylene and other chlorinated aliphatic compounds. Terry Donaldson, Jerry Strandberg, Linda Farr, John Cosgrove, Hal Jennings, and Andrew Lucero have all been involved in the latter project in conjunction with personnel from ESD which led, in 1991, to the establishment of a demonstration bioreactor for the treatment of contaminated seep water at the K-25 plant.

While aerobic microbial activity is effective against most of the lower chlorinated congeners of PCBs, there is little action on the higher chlorinated compounds. Mark Reeves and Betty Evans have, of late, been investigating the anaerobic microbial dechlorination of PCBs. Removal of the chlorine molecules renders the higher chlorinated congeners more susceptible to subsequent aerobic attack. A Cooperative Research and Development Agreement (CRADA) has also been established with General Electric.

Terry Donaldson, Bob Jolley, and Andrew Lucero are leading one of Chem Tech's most recent and largest efforts (in conjunction with The University of Tennessee, Oak Ridge Associated Universities, and the ORNL Environmnetal Sciences Division) directed at the remediation of diesel fuel contaminated soil on the Kwajalein Atoll in the South Pacific. Joe and Angie Walker are now temporarily located on Kwajalein conducting activities at the site.

Bioengineering Research User's Facility. In the early 1980s, the Bioengineering Research User's Facility was established in Building 4505 under the auspices of Chuck Scott, with assistance from Terry Donaldson and Brian Davison. The purpose of this facility is to enable universities and private industry the opportunity to conduct bioengineering research at ORNL and to utilize Chem Tech's expertise and equipment.

4.20 WATER CHLORINATION AND RELATED PROGRAMS,

Bob Jolley

Formation of Chlorinated Organics During Water Chlorination. In the 1960s Rachel Carson's book ***The Silent Spring*** focused national attention on chlorinated organics used as pesticides and herbicides. The newly awakened environmental conscience of scientists brought into question whether toxic chlorinated organic compounds might be produced during the chlorine disinfection of drinking water and wastewater. In a major effort to answer part of that question, in 1970 Bob Jolley chlorinated several wastewater samples using ^{36}Cl-tagged chlorine and separated the organic constituents from the chlorinated wastewaters using high-resolution anion exchange chromatography. Over 50 chlorine-containing organic constituents were separated from secondary wastewater samples, thus providing convincing evidence that such compounds are produced during wastewater chlorination. Follow-up studies using cooling water samples from several electric power production plants also indicated that chlorinated organics are formed during the cooling water chlorination, a common practice to prevent biofouling of heat exchangers (Fig. 4.59). At approximately the same time period, EPA researchers reported that chloroform and other trihalomethanes are produced during the chlorine disinfection of drinking waters. These collective studies led to an intense flurry of national and international research to identify chlorination by-products and to determine their toxicological properties.

Water *Chlorination Conference Series.* In 1975 Bob Jolley and Sidney Katz of Chem Tech, Bob Cumming of the Biology Division, and Carl Gehrs of the Environmental Science Division organized a conference, The Environmental Impact

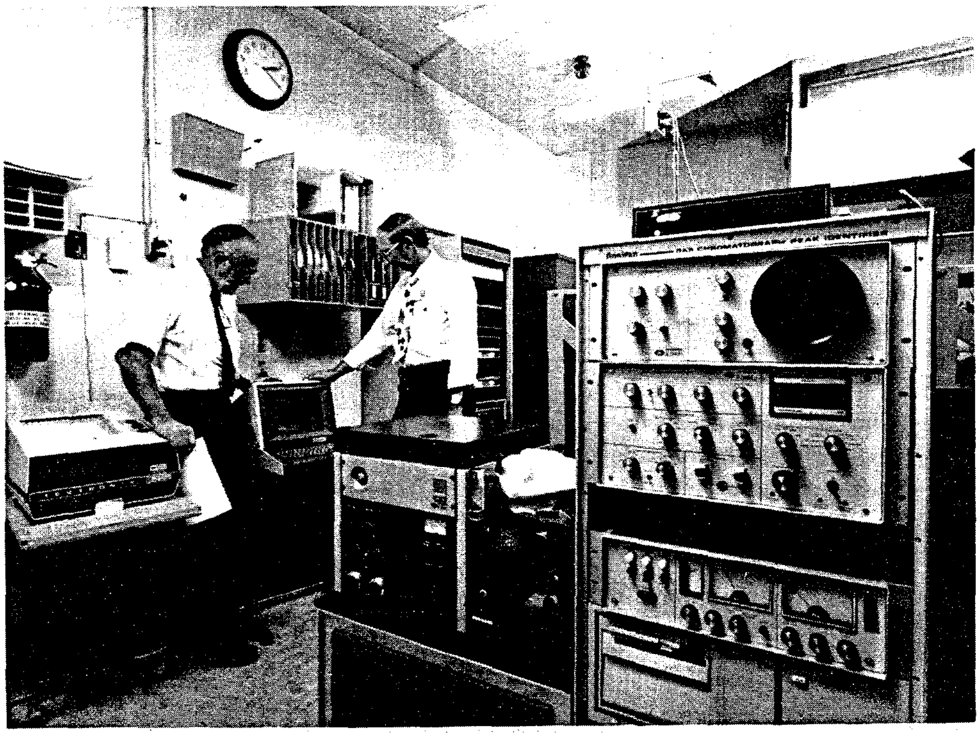


Fig. 4.59. Bob Joiiey (left) and Norman Lee discuss mass spectra of constituents separated by gas chromatography and liquid chromatography from chlorinated water samples.

of Water Chlorination, held October 22–24, 1975, at ORNL. The conference was highly successful, with over 300 attendees, and resulted in publication of ***Water Chlorination: Environmental Impact and Health Effects, Volume*** I .[43] Jolley convened six conferences on the subject between 1975 and 1987, resulting in the publication of six volumes totaling over 6600 pages.[43–48] The conferences and resulting books encapsulated approximately two decades of research in the area of disinfectant chemistry, engineering, and the environmental and health effects of disinfectants and disinfectant by-products. The collective volumes represent a significant national and international contribution to this important area of health and toxicology of drinking water, wastewater, and cooling water treatment.

Drinking Water and Goiter. Goiter is a common national and international disease. Untold losses to the world's reservoir of human intelligence and productivity result from goiter despite almost ubiquitous iodine prophylactic programs.[49] Approximately 1% of surgical operations in the United States in the early 1980s were reported to involve the thyroid gland (i.e., associated with goiter or goiter-related problems).50 During the late 1970s and early 1980s a small collaborative program between Chem Tech and the Universities of Alabama and Mississippi explored the use of high-resolution chromatography for identification of organic constituents in a variety of drinking water sources, including wells and springs as well as public water supplies.

About one-half of the children in a certain geographical part of Cali, Columbia developed goiter compared to IO-20% in other areas. This occurred despite a national supplementary dietary iodine program. In a classic epidemiology study conducted by Dr. Eduardo Gaitan, currently located at the University of Mississippi Medical Center, it was determined that the drinking water source for that geographical part of the city with high goiter incidence was of sedimentary geological origin and different from that for the remainder of the city. Using high-resolution chromatography for organics separation and gas chromatography/mass spectrometry for organics identification, Bob Jolley and Jim Thompson analyzed some water samples collected by Dr. Gaitan. In addition to many organic constituents, they identified resorcinol as present in the goitrogenic water. Resorcinol is a known potent goitrogen.[49,51] The collaborative work between Dr. Gaitan and Jolley was continued in an effort to determine similar compounds in selected counties in Kentucky that have high goiter incidence. Although resorcinol was not identified in these waters, a similar compound was identified.[49,51] Federal budget restraints prevented sufficient collaborative efforts to conclusively identify goitrogenic compounds in the United States water samples (Fig. 4.60).

4.2 1 BRANCHING OUT—STUDIES ON AGRO-INDUSTRIAL COMPLEXES, Hal Goeller

For the first 25 years of its existence, ORNL was renowned, pretty exclusively, for its expertise and activities in the physical and biological sciences. Looking back we all remember the 1960s as a period of social upheaval, mainly by the baby boomers who were beginning to reach early adulthood. In those times, among other things, disillusionment was setting in against science and many of the applications of science.

It seems only natural now that ORNL should have entered into the fray-and it did-by expanding its endeavors to include the social sciences as well as its more traditional activities. At that time, this was a rather bold move. This branching out began in 1967, when a special summer study was set up, at the urging of Alvin Weinberg and Floyd Culler, to begin examination of the many social problems which the physical sciences had long largely ignored.

The summer study ended, but the impetus led in time to the formation of ORNL's Energy Division, which included many economists, geographers, sociologists, and other social scientists who continue to examine the softer side of science. At first it was difficult for the physical and social scientists to understand, much less appreciate, each others viewpoints, but understanding did come as many scientists became generalists as well as specialists.

One of the first efforts of the 1967 summer study was the multi-disciplinary study of agro-industrial complexes using nuclear reactors to produce fresh water from seawater for irrigation agriculture and electricity for electro-intensive industries. This effort was designed to address the Eisenhower-Lilienthal plan to "make the deserts bloom" in water- and power-deficient developing nations. Some of those involved in this study included Cal Burwell, Hal Goeller, John Holmes, Jack Mrochek, John Michel, Irv Spiewak, Phil Hammond, Jim Lane, Bill Yee, Ed Mason from MIT, Perry Stout (one of the "fathers" of the green revolution) from the University of California at Davis, Clair Nader (sister of Ralph Nader), Art Squires an engineer from City College of New York, J. R. Chapman an engineer from Alcoa in Pittsburg, and M. Striplin and Glenn Blouin, engineers from TVA, Muscle Shoals, Alabama. Many members of the multi-discipline and multi-divisional study team are shown in Fig. 4.61.

At first the study was a general one, examining rough reactor designs, possible associated types of industrial chemical processes, and the most suitable crops to be grown. Later studies were directed to

Fig. 4.60. Dr. Nathaniel Revis (left), Director of Oak Ridge Research institute, Dr. Eduardo Gaitan (center), Chief of Endocrinology, Veterans Administration Medical Center at the University of Mississippi, and Bob Jolley discuss chemical spectra produced by analysis of drinking water samples collected by Dr. Gaitan in geographical areas of high goiter Incidence.

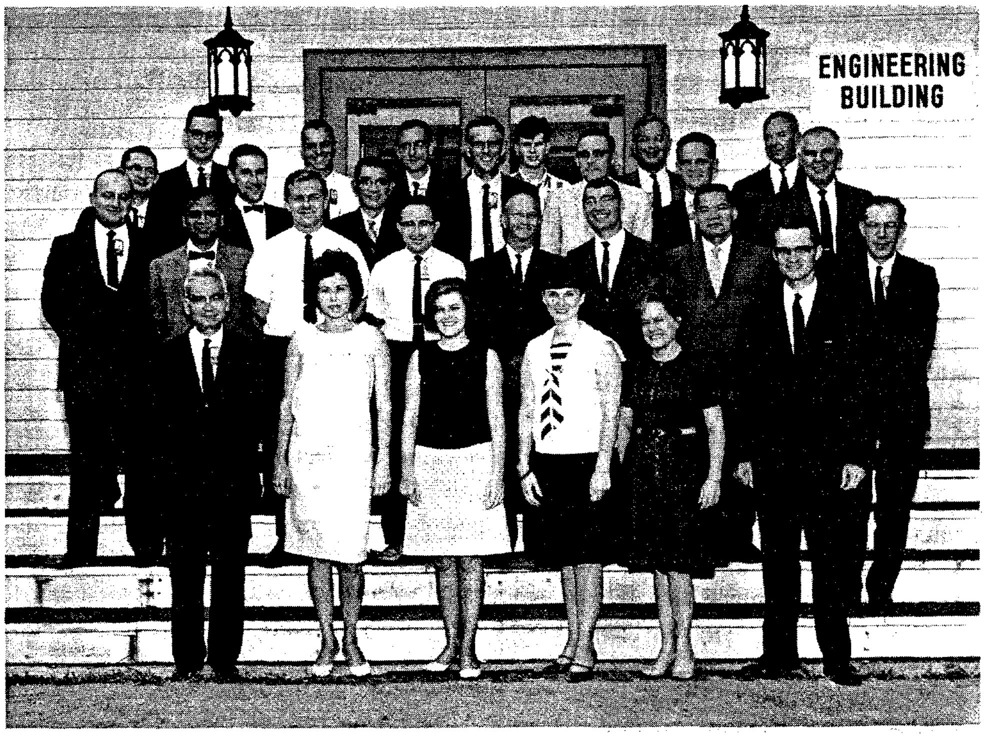


Fig. 4.61. Because of busy travel schedules and the use of a large number of consultants, not all members of the 1967 ORNL agro-industrial study team are shown. Members shown are (left to right): Front row, Alvin Weinberg, Virginia Lee, unidentified secretary, Kathy Gardner, Alice Maxwell, and Ed Mason. Second row, John Mrochek (Chem Tech), unidentified consultant, unidentified consultant, unidentified consultant, unidentified consultant, Marv Yarosh, Bill Yee (Chem Tech), and Hal Goeller (Chem Tech), Third row, John Holmes (Chem Tech), unidentified consultant, Tammy Tamura, John Michel, unidentified consultant, Ray Blanco (Chem Tech), and unidentified consultant. Back Row, unidentified consultant, Cal Burwell, Dick Philippone, unidentified Massachusetts Institute of Technology student, Floyd Culler (Chem Tech), and Gale Young.

selected countries and sites; the industrial processes were then selected to utilize local raw materials. In the case of Egypt, these proposed industrial processes included production of aluminum from domestic bauxite, phosphate-containing fertilizers from phosphate rock, and salt from seawater. Major studies were also done for India and Israel.

In addition, brief application studies were made for the Tokar Delta on the Red Sea in Sudan: the Danakil Depression in Eritria, Ethiopia: the Qattar Depression in Egypt; and the coastal desert in northern Peru. In the latter case, for example, reactors could be used for desalination of seawater or pumping water over the Andes from the Maranon River, a headwater of the Amazon located only 50 miles away.

The major studies were followed by extended trips to India and Israel in November 1967, Egypt in October 1968, and Israel in April 1969. Discussions were held with government agencies on all trips.

During the Indian hip, visits were made to agricultural research stations, to Bakhra Dam on the Sutlej River in the Punjab, and to the Tata Chemical Works on the Arabian Sea in Gujarat, as well as to Bombay (headquarters site for the Indian AEC), New Delhi, Agra, and Chandragahr. One of the favored applications of our idea was for the powering of 30,000 tube wells to raise water from the "third great river" of India, which runs 8 ft underground in Uttar Pradesh, in order to provide water to the area between the monsoon rains.

Discussions in Egypt were carried on mainly in Cairo and Alexandria. Visits were made to new model farms in the delta. We also toured the Mediterranean coast as far west as Mersa Matruh, Rommel's headquarters in World War II, and visited El Alamein, where Rommel was turned back. One problem for Egypt at the time of our visit was the fact that the High Aswan Dam produced so much electric power that no additional generating capacity would be needed for a long time.

The first trip to Israel was brief and involved, besides discussions in Tel Aviv, we travelled to Jerusalem and made a coastal trip to El Arish in the Sinai where we saw some Bedouin farming. The second Israeli trip was much more extensive-so extensive, in fact, that it is almost easier to say where we didn't go. One trip was to the south in the Negev, where we went first to Beersheba and then to Elat on the Gulf of Aqaba. There we saw desalination facilities. Then we traveled along the Jordan border to a desert farm and kabutz, where brackish water, desalinated by reverse osmosis, was used for trickle irrigation fanning. Moving northward to Sedom, we visited the Dead Sea Chemical Works, which produces salt and potash. Above the Dead Sea we toured a new town that was to be the site of a magnesium plant. Traveling westward we visited an orange grove and wheat farm on the edge of the Negev, an orange processing plant, and Ashdod and Ashqelon on the Mediterranean Sea. After visiting Jaffa and Jerusalem we headed north to Nazareth and the Sea of Galilee, which supplies most of Israel's fresh water. Our trip ended with visits to Haifa, Acre, and ancient Caesarea.

Unfortunately, no solid achievements appear to have ever resulted directly from the agro-industrial studies. In retrospect, this appears to have resulted from (1) the agro-industrial complexes were based on 1 mil per kWh power, and that was not really feasible; and (2) there was no real chance for private or U.S. government financing of the huge costs of such projects.

4.22 WORLD ENERGY CONFERENCE—1974 SURVEY OF ENERGY RESOURCES (1974 Enquete sur les Ressources Energetiqes), Hal Goeller

The World Energy Conference (WEC) is an international organization based in London that concerns itself with energy matters of all sorts. In the early 1970s, it had national committees in 69 countries. The WEC holds a conference every six years (now every three years), with the host country in charge. In 1974 the conference was held in Detroit, Michigan.

For each conference the WEC updates its survey of world energy resources for presentation to conference attendees in booklet form. Until 1974 (and also thereafter) the booklets contained only a few pages of statistical material prepared by the London office based on data provided by member nations plus estimates for non-members.

Early in 1973 the U.S. National Committee decided, with London's approval, to finance, write, and print a much expanded survey for the Detroit Conference. They then asked the AEC to take on this task, and in turn the AEC requested ORNL to do it. ORNL readily accepted the role.

The ORNL effort was a multi-phase activity for a variety of energy resources, including coal, oil, natural gas, tar sands, oil shale, nuclear fuels (uranium, thorium, and fusion fuels), hydropower, and other renewable resources (tidal, geothermal, solar, wind and ocean thermal gradients).

The first-phase effort involved the receipt, review, and processing of questionnaires that had been developed by others and sent out to both WEC member and non-member nations. Ultimately, completed replies were received from 54 member nations, including 27 of their dependencies, and from 11 non-member countries. Many replies provided data for numerous national subdivisions. All of these data were then entered into computer files for ultimate print out of a very detailed set of appendices, which finally occupied 115 pages (including introductory text). Data on the geology of fossil and nuclear fuels were also covered.

The second effort involved the writing of an appropriate text for each resource. These texts included discussions of geology, mining technology, fuel characteristics and uses, and environmental and societal problems associated with recovery and use of each resource. Summary resource tables were also provided for each resource, including data from independent sources for non-reporting countries.

Other efforts included preparation of detailed specific and general bibliographies and of world maps showing the location of resources. Finally the entire text was translated into French since the survey has traditionally been issued in bilingual form. In final form the book contained 400 pages plus eight maps.

Each attendee at the Detroit conference received a copy. In addition, President Gerald Ford, who gave opening remarks at the conference, was also presented with a copy.

Those at ORNL who contributed to the overall effort included Roger Carlsmith, Lloyd Carter, Hal Goeller, Miriam Gutherie, Frank Hammerling, and Pat Love. Ralph Perhac from UT provided geological coverage, and John Patterson from AEC Headquarters produced the nuclear resources text. I visited the WEC London office in 1986 and was informed that requests are still coming in for our book.

4.23 HISTORY OF THE CHEM TECH REPORTS OFFICE,

Martha G. Stewart

When I came to work in the Chem Tech Reports Office in 1966, I was on loan from the Information Division (now the Publications Division). After being interviewed by Don Ferguson, Division Director, and Ray Wymer, Head of Section A, I was selected to replace Howard Whetsel, Chem Tech's only editor, because he had decided to accept an assignment with the Nuclear Safety Information Center, which was located in the Y-12 area. Howard had initially said that he would train me for a month before leaving; however, as it turned out, he left after only 9 working days (Fig. 4.62).

I found that my career background was, indeed, quite suitable for the Chem Tech programs under way at that time: 1 year of work in heavy-metal chemistry in the Y-l2 Plant Laboratory, five years of work in radiochemistry and biochemistry at the Oak Ridge Institute of Nuclear Studies (now the Oak Ridge Associated Universities), and two years of training as an editor in the Information Division. Much of my editorial work had been concerned with Biology Division materials. Fortunately, I had also had the experience of serving as the lead editor for the 1965 Chem Tech annual progress report. When I was assigned to Chem Tech Reports Office in 1966, the division was in the throes of preparing the drafts for the 1966 annual progress report. In fact, I arrived just in time to coordinate the collection, editing, and finalization of these drafts.

At that time, the Reports Office (now called the Publications Services Office) was located in Room B-224 in the B Attic (4500N), adjacent to the Xerox Room. There were very few resource materials, one "ancient" typewriter, one desk-and-chair combination, and one bookcase. The noise from the Xerox machine was significant-almost intolerable at times. I felt isolated and uncertain as to how to deal with the large backlog that I had inherited, but the division office personnel (Don Ferguson, Nancy Beach, and Elaine Hickman) were very helpful and the section heads made me feel very welcome.

As is the case at present, the secretaries in the various sections were responsible for report preparation. However, the mode of preparation was slow and unwieldy since it involved use of the standard IBM typewriter. Corrections, which were tedious, were made by using correction fluid, strip-on rape, or paste-ons, or by retyping entire pages. Authors traditionally wrote the first draft in longhand on yellow, lined, legal-size tablets. Then, many additional typed drafts followed, as editorial comments and suggestions from reviewers were incorporated.

All documents were prepared on photomasters (ORNL and ORNL/TM reports) or duplimats (ORNL/CF memos). Duplimats were a glossy-surface medium that had to be corrected by erasing with a special type of pencil in order to correct errors. Thus, the preparation of reports and papers was a very time-consuming and expensive procedure.

Fig. 4.62. A 1991 photograph of Martha Stewart at a seminar.

As the years progressed, the work load of the Reports Office expanded astronomically and the editorial staff was increased accordingly: Vivian Jacobs, 1974; Cathy Shappert, 1976; Debbie Stevens, 1976; Amy Harkey, Leon Morse, and W. D. Norton, 1980; Luci Bell, 1981; Frank Scheitlin, 1983; Donna Reichle, 1984; Vicki Hinkel, 1986; Cindy Robinson, 1987; Mary Louise Conte, 1988; and Kathryn King-Jones, 1990. Four of these editors were eventually promoted to higher-level positions and were transferred to areas of greater opportunity within the Publications Division. Two others retired, and one individual moved to a different geographical location.

During this period, the mode of report preparation also advanced significantly. Several stages of new technology and improved efficiency evolved, successively, as the following types of equipment were installed and implemented: IBM Selectric typewriter, IBM Selectric II (Correcting) typewriter, various word processing systems (MagCard, Royal's CPT, Wang, etc.), and, finally, personal computers (primarily IBM clones and WordPerfect software). Each phase, or stage, in this sequence represented a "giant step" with regard to labor and cost savings-as well as a more attractive and readable finished product.

The Publications Services group has always been interested in achieving top-quality ratings for their editorial work; maintaining a high level of personnel interaction and liaison with the Records Management Department, Patent Office, Reproduction, and Central Publications services: meeting all clearance and editing deadlines with respect to papers for scientific meetings and ORNL or DOE milestones: and enhancing their capabilities commensurate with new technology. In this regard, the Publications Services staff members have won numerous Society for Technical Communication awards for journal articles, reports, and brochures: have been awarded the high honor of having the best "no-late" clearance record at ORNL (received "zero-late-clearances" award for 199 1); and have received numerous commendations for outstanding work on special projects. All in all, the Publications Services group has played an integral part in all the execution of the many important programs and projects carried out by Chem Tech for nearly three decades.

4.24 A MERCIFULLY BRIEF HISTORY OF ENVIRONMENTAL CONTROL TECHNOLOGY, John Parrott, Jr.

The core of what is now the Engineering Development Section (EDS) of the Chemical Technology Division originated in the late 1970s as the Environmental Control Technology (ECT) R&D Group within the Advanced Technology Section. This group, headed by Richard Genung, drew its inspiration from and built upon the pioneering Chem Tech environmental technology efforts of Chuck Hancher's biodenitrification project and Chuck Scott's biotechnology program.

The initial projects undertaken by the ECT group included monitoring and assessment of wastewater from fossil energy plants, design and development of a mobile pilot-scale wastewater treatment system for use at fossil energy plants, bench-scale evaluations of mutanegicity and toxicity phenomena associated with proposed wastewater treatment processes at fossil energy sites, and design and development of the ANFLOW (ANaerobic upFLOW) wastewater treatment system.

Mobile Wastewater Treatment. Planning for the mobile pilot-scale wastewater treatment system, which was to play an important role in future ECT activities, began in October 1980 with the following objectives:

- Evaluate advanced wastewater treament techniques to effect zero-stream discharge or meet future discharge regulations.
- Provide scaleup data for larger conversion plants.
- Assist in solving operational problems at existing wastewater treatment facilities.

The mobile pilot plant was constructed during FY 1981 and early FY 1982 and consisted of three semi-trailer vans--two housing the process equipment and one serving as an analytical laboratory (Figs. 4.63 and 4.64). These trailers were initially deployed to the H-Coal coal gasification plant in Catlettsburg, Kentucky, in the winter of 1981-1982, manned by George Oswald, Cliff Brown, Joe Walker, Jim Hewitt, Mike Harris, Steve DeCicco, and Jack Rose. The successful completion of this mission in September 1982 led to a similar expedition in October 1983 to the

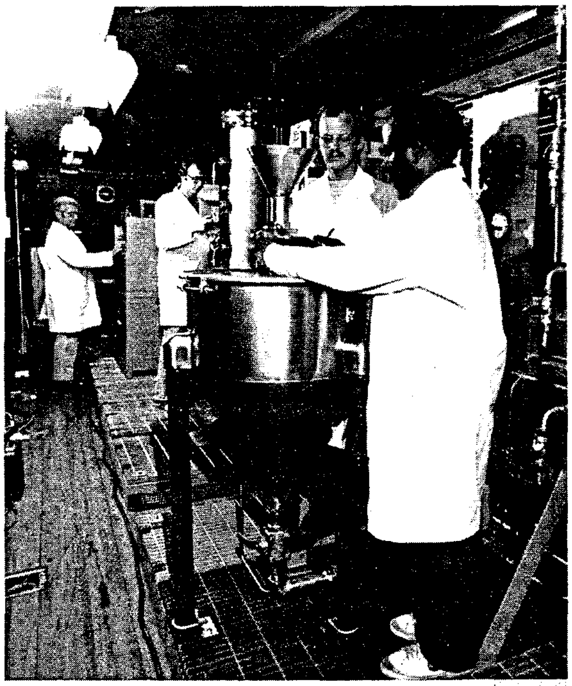


Fig. 4.63. Coal conversion wastewater treatment pilot plant operated at H-coal and Solvent Refined Coal Pilot Plants. Left to right are Jack Rose, Joe Walker, Cliff Brown, and Mike Harris.

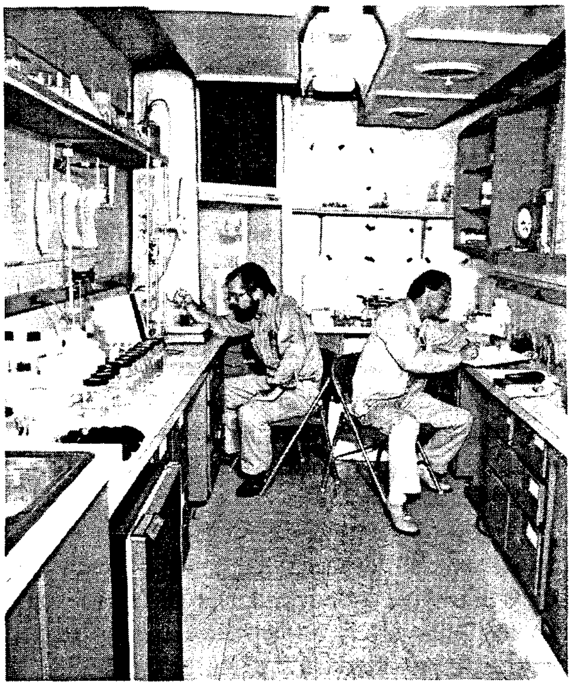


Fig. 4.64. Analytical support trailer for coal conversion wastewater treatment pilot plant showing Chem Tech staff members Don McTaggart (left) and Jim Hewitt.

Advanced Coal Liquefaction R&D Facility in Wilsonvilie, Alabama. At these sites, bench-scale unit operations (granular activated carbon, ozonation, and reverse osmosis) were evaluated for treatment of the process condensate from the coal conversion processes. The trailers were then sent to Y-12 where they successfully removed mercury contamination from wastewater (by reverse osmosis and ion exchange) to a level permitting the water to be discharged.

ANFLOW Wastewater Process. The ANFLOW sewage treatment process was developed in the late 1970s using facilities in the high bay of Building 4505. A 500-gpd system was then demonstrated at the West End Treatment Plant plant in the city of Oak Ridge. A near-full-scale ANFLOW sewage treatment pilot plant was operated by George Dinsmore and Hal Jennings at a Knoxville wastewater treatment facility from August 1981 through the summer of 1983 (Figs. 4.65).

Growth in Wastewater Treatment Experience: Nonradiological Wastewater Treatment Plant; Process Waste Treatment Plant. After these successful wastewater treatment campaigns, the ECT group grew to encompass increased responsibilities, providing support to the entire Oak Ridge complex by conducting wastewater treatment projects for sponsors at K-25, Y-12, and ORNL. Cliff Brown became manager of the group, which by mid-1985 had become the largest group within the new Engineering Development Section. John Begovich joined the ECT group and initiated programs to provide assistance and support to the ORNL Operations Division, which was under pressure to improve the performance of ORNL wastewater treatment facilities in the face of aging equipment and increasingly stringent environmental regulations. Operations Division support by the ECT group began when Chem Tech was asked to assess the proposed flow sheet for the new Nonradiological Wastewater Treatment Plant (NRWTP) for technical feasibility and to simulate the proposed treatment plant to provide design data for the full-scale plant. At about the same time, Operations Division requested that Chem Tech perform an evaluation of the Process Waste Treatment Plant (PWTP) to improve its efficiency and to define its relationship with the new NRWTP. To facilitate evaluations of the PWTP, an engineering-scale facility was designed and constructed to aid in the rapid solution of day-to-day operating problems at the PWTP as

Fig. 4.65. Tom Dinsmore (left) and Hal Jennings unload support rings from the ANFLOW Pilot Plant located in Oak Ridge.

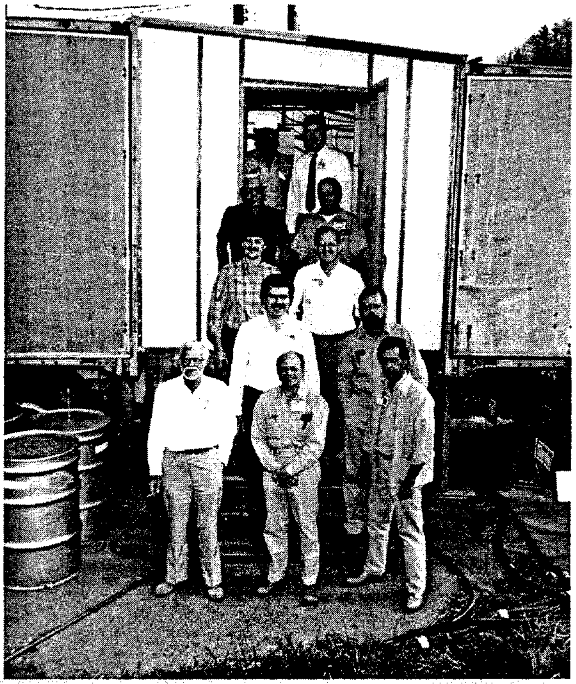


Fig. 4.66. Development team for ORNL Nonradiological Wastewater Treatment Plant. Bottom row: John Villiers-Fisher (left), George Dinsmore (center), Jim Hewitt. Second row: John Begovich (left), Don McTaggart. Third row: Brad Patton (left), Cliff Brown. Fourth row: Vic Fowler (left), Vaughn Justice (Plant and Equipment Division). Top row: Charles Kackler (left, Plant and Equipment Division), Warren Thompson (Engineering Division).

well as serving as a vehicle for testing new treatment concepts (Fig. 4.66). Meanwhile, experiments with new wastewater treatment technologies led to the design and operation of a zeolite column system at the PWTP. All of these projects were remarkably successful, with the pilot-scale zeolite columns leading to a system which is used at the PWTP today and which has become the basis for the new PWTP which will become operational in the late 1990s. The NRWTP studies led to the unit operations that were used in the actual plant, and the PWTP evaluations spawned process improvements such as the addition of the reactor/clarifier, which improved PWTP performance by decreasing LLLW generation and increasing the life of the PWTP ion exchange resin. The success of these projects laid the groundwork for the expanded role that CID now plays in strategic planning for Waste Management and Remedial Actions Division programs (Fig. 4.67).

Analytical/Treatment Technology. The ECT exportable treatment technology program continued during the mid-1980s. The analytical trailer was dispatched in the fall of 1984 to the KILnGAS® rotary kiln coal gasification project in Alton, Illinois, to characterize the wastewater generated by this process. Back at ORNL, the ECT group became involved in a wide variety of endeavors, including the Fossil Energy Technology Program which began in May 1983 under DOE sponsorship, experimental advanced treatment technologies such as the prototype wet-air oxidation system designed to remove organic constituents from wastewater, and the indirect liquefaction environmental control technology assessment project to assess the impact of designing a coal conversion plant to operate with zero aqueous discharge.

In 1986 the group, still heavily involved in support for ORNL wastewater treatment facilities,

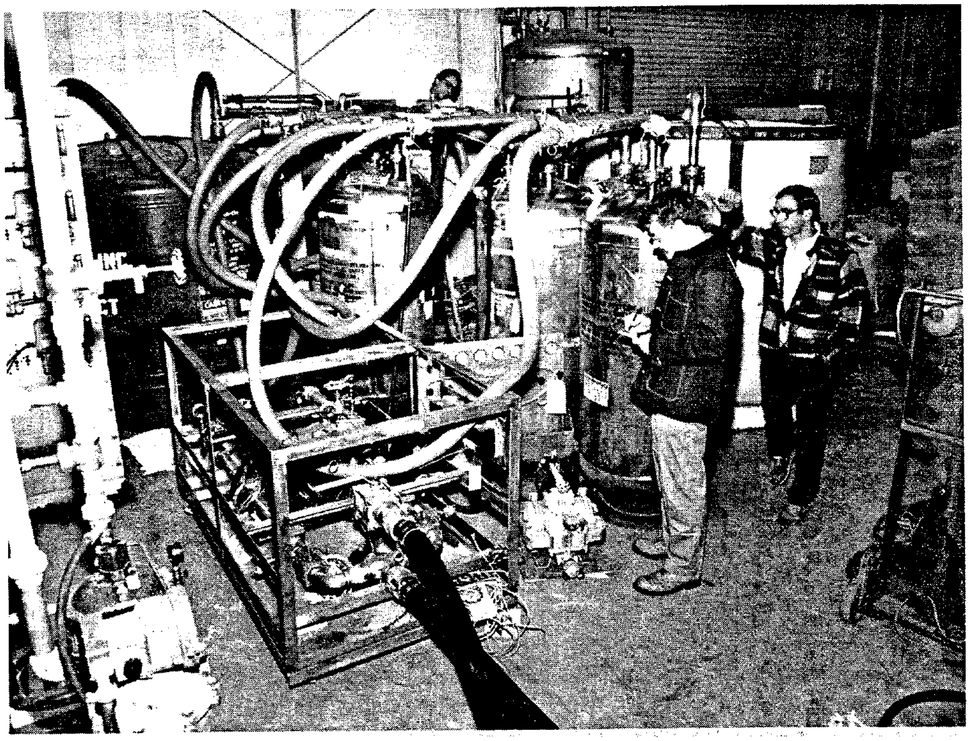


Fig. 4.67. Jim Hewitt (background), John Begovich, and Mike Morris (right) **monitor emergency treatment of ORNL process wastewater using a zeolite** ion-exchange-treatment system.

expanded its Operations Division support to include the following:

- Development and demonstration of the use of continuous automated metals monitors to characterize ORNL process waste streams (Dave DePaoli and Don McTaggart).
- Treatment of LLLW stored in the Melton Valley Storage Tanks (MVST) by filtration to remove the TRU isotopes to produce a lower-activity non-TRU waste for final disposal (Vic Fowler).
- Studies on the disposal of post-silver removal photographic wastes at the ORNL sewage treatment plant (John Parrott, Jr., and Jerry Strandberg).
- Treatability studies on five new wastewater streams for discharge to the coal yard runoff treatment facility (Joe Walker, George Dinsmore, and John Parrott, Jr.).
- Laboratory-scale tests to determine the feasibility of treating ORNL-contaminated laundry wastes at the PWTP (Sharon Robinson).

Bioremediation. ECT-related activities in 1985-1986 included the ongoing bioremediation technologies being conducted by the Advanced Technology Programs Group under the direction of Terry Donaldson (Fig. 4.68). These projects included bioprocessing support to the Feed Materials Production Center at Femald, Ohio, where a fluidized-bed biodenitrification plant was constructed based on the pilot work conducted at ORNL in the late 1970s and early 1980s and biooxidation of coal gasification wastewaters for the Morgantown Energy Technology Center. In June 1987, a project demonstrating bioremediation of PCB-contaminated soils was conducted with six lysimeters being installed at a PCB-contaminated site on the Bear Creek floodplain near Y - 12. Another bioprocessing project in 1987-1988 included bench-scale biological degradation of trichloroethylene in groundwater. An additional ECT project during this time period involved conducting water quality programs through an interagency agreement with the US Environmental Protection Agency.

Changing ECT Management. In 1987 John Begovich left the ECT group (and ORNL) on an l&month leave of absence to work on the AVLIS program at Lawrence Livermore National Laboratory. Jim Wilson and Jan Berry then came

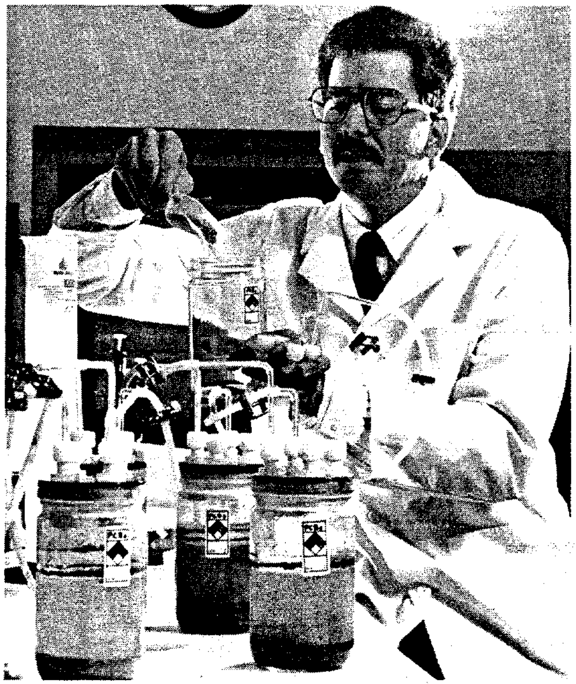


Fig. 4.68. Mark Reeves utilizes small-scale bioreactors to study PCB degradatlon.

into the section, joining Sharon Robinson as group leaders, and Cliff Brown replaced Wilson Pitt as the EDS Section Head when Pitt left ORNL for a faculty position at Texas A&M University. The four EDS groups were Work for Others Waste Technology (Wilson), Support to Operations Division (Robinson), Solid Waste Technology (Berry), and Advanced Technology Programs (Donaldson). Around the beginning of FY 1990, the Waste Soldification/Immobilization Group headed by Mike Gilliam joined the section when the Waste Management Technology Center ceased to exist.

Work for Others--Air Force. The Work for Others Waste Technology Group initially developed and demonstrated technologies to remove organic contamination from soil, obtaining contracts from the Air Force to demonstrate the ability to cleanse soil and/or groundwater contaminated with fuel spills. An air-stripping demonstration was conducted in 1987-1988 to remediate groundwater contaminated by diesel fuel at Eglin AFB near Pensacola, Florida, and a soil-venting demonstration was conducted at Hill AFB in Salt Lake City, Utah. Dave DePaoli, Hal Jennings, Andrew Lucero, and Archie Wilson were the primary investigators in these endeavors (Fig. 4.69). Other Air Force projects during this period involved environmental evaluation of fire training pits at several Air Force Bases by Tom Hylton and Joe Walker.

Work for Others-Navy. The group also. conducted a hazardous waste minimization project headed by Joe Walker which demonstrated innovative wastewater treatment technologies to mimimize chromium contamination in plating shop rinsewaters from the Naval Ordnance Station in Louisville, Kentucky.

Remediation Technology. Upon the termination of the Air Force contracts, this group merged with Terry Donaldson's group and was renamed the Remediition Technology Group, with Donaldson as its leader. In 1989 this new group conducted in-lab experiments on the bioremediation of PCB-contaminated soil and in 1990 and early 1991 evaluated the performance of a catalytic incinerator at Wurtsmith AFB, participating in a soil remediation demonstration at an old oil landfarm site at the Portsmouth Gaseous Diffusion Plant (in conjunction with ESD), and began what would be the group's farthest-reaching exportable technology demonstration when Joe and Angie Walker traveled to Kwajalein Island in the Pacific Ocean to conduct an in situ soil bioremediation project (Fig. 4.70). In late 199 1, Hal Jennings and Andrew Lucero began a treatment technology demonstration much closer to home, with on-site treatment of solvent-contaminated groundwater leaking from a burial ground at K-25 using cometabolic bioreactor technology.

Waste Management Problems. The Support to Operations Division Group under Sharon Robinson quickly expanded to address all types of ORNL waste mangement problems, changing its name to the Liquid and Gaseous Treatment Technology Group. This group grew from its initial mission of providing operational support to the PWTP and NRWTP to include strategic planning and coordinating/performing applied research programs in support of ORNL waste treatment system upgrades. The assistance to Operations Division (now the Liquid and Gaseous Waste Operations Division) has been ongoing, first with John Villiers-Fisher (now retired) then Paul Taylor and Tim Kent providing answers to daily treatment plant operating problems and performing treatability studies to improve plant operations.

Low-Level Radioavtive Waste. In 1988, John Parrott, Jr., in his capacity as the Laboratory Certification Official for Liquid Waste, spearheaded development of technology-based

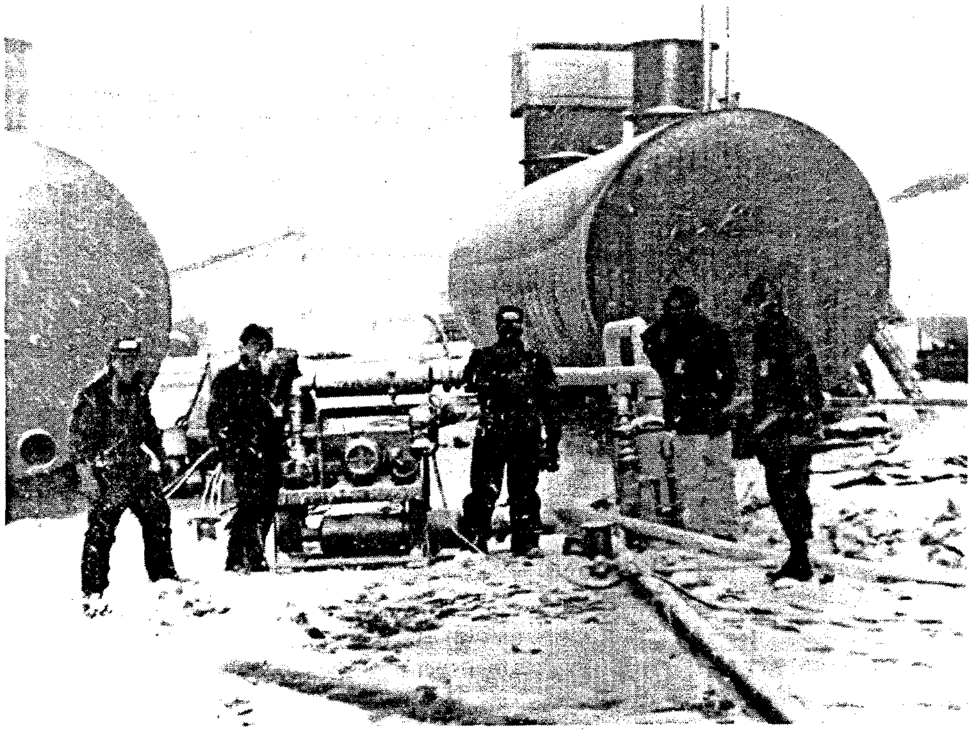


Fig. 4.69. The rigors of wintertime conductions of the soil venting demonstration at Hill Air Force Base, Utah. ORNL team members (left to right) are Hal Jennings, David Depaoli, Dan Gillespie, and Jim Wilson. U.S. Air Force Captain Ed Heyse is on the right.

Fig. 4.70. Angie Walker (right) and Alvin Ajen (a Kwajalein Island citizen) install the multi-cell in-situ bioremediation test system on Kwajalein Island, Kwajalein Atoll, Republic of the Marshall Islands.

waste acceptance criteria for all ORNL liquid waste treatment facilities and, in conjunction with other group members, developed and instituted a liquid waste certification program which encompasses training on LLLW bottling and liquid waste disposal policy in general. Also during this time period, Susan DePaoli and Angie Walker designed and implemented a comprehensive LLLW data base. In 1989, a sampling and analysis campaign to characterize ORNL LLLW was conducted to provide environmental assessment data as well as design data for Waste Handling and Packaging Plant (WHPP) and data in support of R&D activities in developing waste management alternatives. The group used this data to perform a systems analysis in 1989 for all ORNL liquid waste treatment and collection systems. The systems analysis is currently being used to identify and prioritize LLLW system upgrades needed to meet new environmental regulations. In 1990, Sharon Robinson developed the methodology for implementing the Federal Facilities Agreement to upgrade ORNL LLLW collection and treatment facilities and developed strategies for long-term treatment of all liquid waste streams generated at ORNL. Contingency planning for LLLW management was also carried out, in which methods to decrease the volume of LLLW concentrate stored in the MVSTs were investigated. These studies are continuing, with Joe Perona working on in-tank evaporation of the stored concentrated LLLW in the MVSTs and Matt Boring concentrating on the development of a wiped-film evaporator to dewater the sludge in the MVSTs. Innovative wastewater treatment methods investigated by this group have included a continous countercurrent ion-exchange system, operated by Reggie Hall and Jim Hewitt in FY 1989, which selectively removed a preferred component (in this case strontium) from a multi-component stream which also contained calcium and magnesium. Currently in the Liquid and Gaseous Treatment Technology Group, flow sheets are being developed to treat existing LLLW, newly-generated LLLW, process wastewater, and new liquid waste streams such as those generated by environmental restoration activities. These flow sheets focus on the development of improved physical and chemical separation processes to produce small volumes of segregated waste packages for long-term disposal

Solid *Waste.* The Solid Waste Technology Group began work in early 1987 in support of the proposed WHPP, performing research and development on the design of a system that would remove, treat, and solidify contaminated MVST liquid waste to prepare it for shipment to a repository in Carlsbad, New Mexico. In FY 1989, WHPP technology development continued with the creation of the WHPP Development Facility in Building 2528 to provide the engineering data needed for the design of the slurry processing systems plan to be installed at the actual WHPP. These included MVST sludge mobilization development studies to investigate methods of sludge removal from the MVSTs and conceptual designs for the evaporation by either conventional means or microwave energy of the sludge mobilized from the MVSTs. Since 1990, the expertise gained by this group in WHPP development has been expanded to address similar problems at K-25 and Hanford.

WasteSolidification/Immobilization. The Waste Solidification/Immobilization Group, although the last group to join the EDS, has been in existence for a long time, evolving from work starting in the early 1960s in the Waste Management Section of the Health Physics Division in support of hydrofracture disposal of liquid waste. This effort was headed by Dr. T. Tamura in the Health Physics Division until the responsibility was assumed by John G. Moore of the Chemical Development Section of CTD in FY 1972. Programmatically the effort was part of the Geological Isolation Program headed By Bill McClain. Initial tasks consisted of mix design for disposing of a grouted sludge via hydrofracturing and a borehole plugging program in support of sealing boreholes and mine shafts for geologic repositories.

In early CY 1976 the group became part of the newly organized Office of Waste Isolation (OWI). After several weeks CTD management decided that the group would return to CTD since OWI was a project management organization and did not want responsibility for development efforts within their organizational structure.

In December 1976 the group moved to Building 9204-3 at Y-12, expecting very rapid growth in support of the ever-enlarging OWI and became part of the Isotopes Section headed by Gene Newman. During this period the group tasks were hydrofmcture mix development, borehole plugging, and development of a FUETAP (fixed under elevated temperature and pressure) thermally treated waste form as a candidate for high level waste disposal.

When the Isotopes Section was transferred to the Operations Division in the late 1970s, the group was transferred to the Experimental Engineering Section of CTD and moved to Building 3017 at the X-10 site, where they remained until moving to Building K-1006 at the K-25 site in mid-1985.

In mid-summer 198 1 the first injection was made with the new Hydrofracture Facility (NHF). Support was provided for the two-year effort to dispose of all the sludge in the gunite tanks in the South Tank Farm. This effort was officially completed in FY 1984.

Grout Technologies. Beginning in FY 1982, support was provided to Rockwell Hanford Operations to provide cement-based waste form development technology for disposal of liquid wastes stored in a large number of tanks at the Hanford Reservation in southeastern Washington. This effort continues to the present, led by Earl McDaniel.

Recent projects by this group have included technical support to in situ stabilization of shallow-land burial trenches, technical support to Y- 12 and K-25 in grout formulation for immobilization of mixed organic-containing wastes, hydrofracture support, ORNL pond sediment stabilization study, MVST support, and studies of polymer impregnation of cement-based waste forms in CY 1986. In 1987 through mid 1988, the group worked on a transportable grout facility, solidification of MVST supernate, and evaluation of grouts under pressure (Fig. 4.71). Fiscal year 1989 projects included grout calorimetry, grout meter development, and in situ immobilization of volatile organic compounds. From 1990 to the present, the group has focused on providing Westinghouse Hanford Company with technical support to the Hanford Grout Technology Program and with technical support for the implementation of grouting technology as a remediation option for Femald waste.

EDS Today. The Engineering Development Section today is a viable, dynamic organization, providing real-world solutions to everyday waste treatment problems while constantly striving through research to improve the environment by the application of advanced technology.

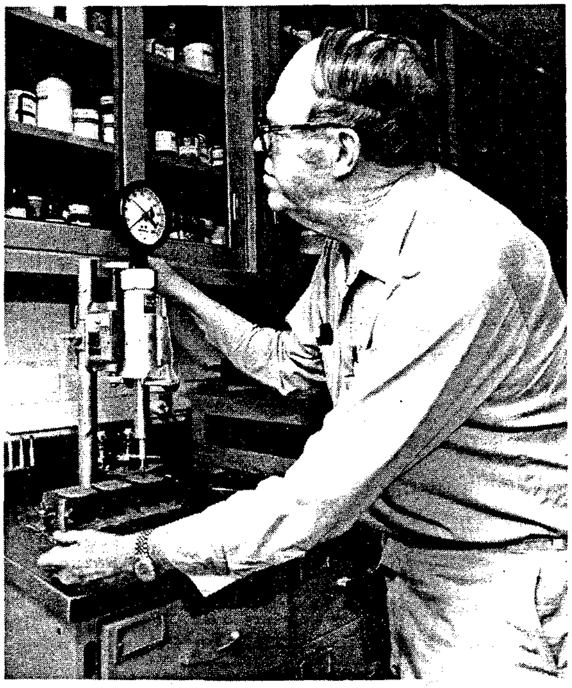


Fig. 4.71. Roy Lovelace measures penetration resistance of a grout sample.

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5. POTPOURRI: PERSONAL ANECDOTES, VIGNETTES, THE ST. PATRICK'S DAY DANCE, AND OTHER RECREATIONAL ACTIVITIES

There is really a sense of satisfaction when a process we have something to do with is actually used.

David 0. Campbell
Comments, 1992

Life is a theater.

Angel L. Rivera
Conversations, 1988

5.1 MILES C. LEVERETT, SEPTEMBER 28, 1991

One of the tasks for which I was responsible was the preparation of kilocurie quantities of carrier-free ^{140}Ba. The source of, the material was freshly irradiated uranium slugs, discharged from the air-cooled graphite reactor and transported immediately to Building 706D, which had been hurriedly built in a few weeks for just this purpose. Barium-140 (or, more precisely, its daughter ^{140}La) was urgently and unexpectedly needed at Los Alamos for some weapons development tests. Barium-140 has a half-life of 12.8 d, so it was necessary that the entire process of preparing each kilocurie batch be carried out in just a few days. The basic process was developed on a one-curie level by the Chemistry Division; it was my job to adapt the process to a three-orders-of-magnitude scaleup, develop and design the equipment, assemble and train a staff, put the plant into operation, and see that the whole thing was accomplished in the shortest possible time. An idea of the urgency that the Army attached to this project may be gained from the fact that when we needed some special alloy stainless steel, the Army diverted a carload of it from a train which was en route to Hanford. We also had lots of "help" from the Army in other ways; they were continually asking for progress reports and doing inspections of the work. At one point, we let it be known that if they were willing to go to work as pipe fitters they would be welcome, but otherwise not.

The process involved de-canning the irradiated uranium slugs by dissolving the aluminum cans in NaOH, dissolving the uranium in nitric acid, adding lead (as the nitrate I think), precipitating the barium and lead as the sulfates (the lead sulfate was a carrier), converting the lead and barium to a form in which the lead but not the barium would be soluble in ethyl ether, and performing an ether extraction which left the barium behind. The details of the process have become a little hazy in my mind, but the point is that we found out the hard way that the intense radiation field could cause the ether solution to solidify, thus filling our inaccessible and fragile extraction apparatus with a highly radioactive, immobile material and bringing the whole process to an untimely end. This happened several times before we found out how to avoid it-just one of the little surprises resulting from working in a new technology.

5.2 FRANK E. HARRINGTON, SEPTEMBER 12, 1991

I reported to work July 8, 1946, at Clinton Laboratory managed by Monsanto Chemical Company. I was assigned to the Building 3019 (then 205) Fission Product Analysis Laboratory and conducted analytical determinations of elements such as ruthenium, zirconium, and niobium for Frank Bruce and others in the Technical Division. At that time, management believed only college-trained chemists or chemical engineers could do this type analysis. The analytical group included Larry Corbin, Zeke Burros, Buddy Warren, and Oscar Bizzell.

I volunteered for assignment in shift operation of the first postwar pilot plant of the Technical Division to demonstrate the "25" process. Later the 3019 Pilot Plant was modified and expanded to demonstrate sequentially the "25," Redox, Purex, and Thorex processes.

Chem Tech was started from Section T-I, Process Development, and Section VI, Pilot Plant, of the Technical Division. At that time, Dr. M. D. Peterson was Director of the Technical Division. He was outstanding in many fields of endeavor and left to teach at Vanderbilt, then Princeton, and later Argonne. The Section VI Chief was Don G. Reid, who later went to Idaho. Reprocessing was the central mission of Section VI and became the central mission of the Chem Tech Division when the Technical Division sections became divisions. That central mission carried throughout the Laboratory and the Unit Operation and Pilot Plant Sections of Chem Tech. Prominent early in the Laboratory Section of Chem Tech were Ray Blanco, Frank Bruce, and Ray Wymer. Similarly, Unit Operations included Warren Eister, Jim Davis, Bill Unger, and Marvin Whatley. The Pilot Plant included Walt Rodgers, Milt Levensen, Hal Feder, Ed Nicholson, Nelmar Rigstad, Don Reid (Reid and Rigstad went to ICPP), and Curt Jackson. The Design Section was led by Floyd Culler, who transferred to Clinton Labs about November 1946 from Y - 12. Hal Goeller was Floyd's right-hand man.

The first Chem Tech Division Director was Frank Steahly, who left to lead Union Carbide Corporation's effort in the separation of coal processing products at South Charleston. Second was Floyd Culler. Floyd became the director at age 28 with a B.S. in chemical engineering. Floyd Culler's speeches at regular seminars were inspiring. He had the best informed division in ORNL. He also could get people to go the extra mile better than anyone I have ever known.

Working on the ICPP project was an exciting experience. The Chem Tech team assigned to conduct the ICPP work onsite in Idaho included Hal Goeller, Leader, and Bob Klotzbach, Ed Nicholson, Bill Kearsley, AI Rom, Frank Browder, John Ruch, Ed Frederick, and me. Can you imagine a Reprocessing Plant built for $30 million? The ICPP plant was based on the 3019 Pilot Plant process and experience and was initially directly maintained. Our group plus Tex Blomeke had responsibility for initial operation of the plant (American Cyanide refused to accept responsibility until after that period). Since then the plant has had a remote head end (and more) added. It was a thrill to me to visit the plant when it was approximately 30 years old to

Frank **Harrington (on the left) with several Chem Tech colleagues (from left to right): Karl Notz (Sect. 4.6), Al Ryon, Ray Wymer (Sect. 2.8), Claude Haws, and Paul Haas (Sect. 4.2).**

find much of original plant still operating as designed and installed.

5.3 ED L. NICHOLSON, MAY 26, 1992

RaLa Process. The RaLa process was run in Building 706C from 1944 through part of 1946. I was an Army GI and was involved in the design, installation, repair, and operation of the 706-C final purification part of the system. The RaLa process was then moved to Building 706D (3026D). However, the first runs in 706D were a disaster, and 706C was pressed back into service until the 706D system was operable.

Purex and Redox. I was discharged from the Army in 1946 and hired back into the Technical Division on March 13, 1946. After working on the last design phases of the 25 Process Pilot Plant for Building 205, I moved to the Pilot Plant Operations Group. With the able assistance of Frank Harrington, I converted the 25 Pilot Plant for Redox operations. Numerous changes were made, including facilities for filling 5-gal shipping containers (shielded) for sending the plutonium solution from the 1BP process stage to Argonne. After 2 or 3 years of Redox operation, the plant was returned to the 25 Process configuration and a few final verification runs were made using full-activity-level Hanford ^{235}U–Al alloy slugs. Al Rom did that work in about 195 1 to verify the ICPP system design.

AU in a Day's Work. In 1946-1948, the development runs for the pilot plant 25 Hexone Process were all with natural uranium slugs from the Graphite Pile. The final confirmation runs were made about 1951 with fully enriched ^{235}U–Al slugs from Hanford. I remember lifting the Hanford slugs out of the jammed slug charging funnel with long-handled tongs. All in a day's work! ! !

HOPE. During the summer of 1954 or 1955, Eugene Wigner pulled together and directed a team or "think-tank" group to try to develop innovative, very inexpensive ways to reprocess fuels. The team included Bob Char-pie. Hal Goeller, Bob Klotzbach, and I were the Chem Tech participants. Some of the wayout considerations included covering process equipment with sand for cheap shielding. Another was to install process equipment in a swimming pool in order to use the water for shielding. At that time the reactor fuels were enriched ^{235}U and not low enriched, as most fuels are now. Wigner wanted a project name that was indicative of being able to reduce fuel processing costs. The group considered the nature of the proposals, some of them were wayout, and came up with the name HOPE. Wigner laughed when we finally told him about our name for the project. To us HOPE stood for Hooty Owl Project Experiment, as in "crazy as a hooty owl." At the end of the summer, we had a closing celebration at either Wigner's or Char-pie's home in Oak Ridge.

5.4 BILL UNGER, APRIL 1992

Chem Tech staff member William E. Unger participated in the design and operation of the RaLa process. His comments concerning this important process follow.

Bear in mind that some 40 years have passed, and some of the technical details have grown a bit vague. I never did find out precisely what Los Alamos did with the lanthanum that they "milked" from the ^{140}Ba that we shipped to them. In those days it was considered in bad taste to be too inquisitive beyond a "need to know," but important was the high specific activity, the hard 1.2-MeV gamma and the short 40-h half-life. The tolerance specs on the lanthanum purity were inversely related to the half-lives of the contaminants.

The original plant (Walt Rogers gets that credit) dissolved the slugs in nitric acid after dejacketing

Bill Unger, 1960

in caustic, then a sulfate, precipitation of the barium using lead for a carrier. The lead was later removed by electroplating on a platinum screen electrode. The barium was redissolved in nitric acid and then precipitated in fuming nitric acid in which barium is uniquely insoluble, thus separating it from the contaminants by filtration in a 4-L Stange reactor. It was all a little unsophisticated, inefficient, and unreliable (the glass Stange reactor turned black from radiation), and a colorful part of every run was arguments among the operators, each taking a turn at the periscope, as to the level of the liquid in the reactor. Harris Blauer was the loudest and fortunately usually right.

The 706-D Modification Project (that title had to be lettered on every drawing and I never made that mistake again) used all of the dissolution and sulfate precipitation equipment but added new purification equipment that was located in floor pits outside the walls of the A Cell. This was to satisfy Los Alamos that the old equipment would continue to function until the new equipment was in place and operable. Stainless filters were enclosed in lead-shielded blisters in the cell wall and designed to accommodate Cellite filter aid.

Ray Blanco devised a neat ion-exchange isotope separation process which was followed by a fuming nitric acid precipitation on the eluted product stream, mainly to get rid of the iron, and a dilute nitric solution of the purified barium precipitate was drawn over into a platinum-lined 4-in. centrifuge cone and dried with hot air. The product was assayed by counting the fissions in a ^{235}U foil generated by the neutrons, moderated by a block of paraffin, that were knocked out of metallic beryllium by the 2.1-MeV lanthanum gammas and then back-calculating to the last separation time of the fuming nitric precipitation step.

One of the frustrating operating problems was "chasing" the pH of the ion-exchange column feed solution. The high radiation level caused real chemical problems. Once, I recall, we lost the product and turned out the cell lights to find it. The barium was all precipitated in a glowing ring on the inside of the feed tank wall.

The equipment consisted of ion-exchange columns, a feed tank, waste tank, tantalum filter, tantalum-lined nitric precipitation tank, ancillary flow monitors (an electromagnetic device that was later patented and marketed by others), radiation monitors, and a sampler that remotely took a measured 200-lambda (microliter) sample. All equipment was compactly mounted in a rectangular frame, 2 x 4 x 6 ft high, enclosed in a stainless steel shroud, and topped by a 22-in.-deep pan through which all the instrument lines were snaked. After this module was lowered into the floor pit, the pan was filled with lead shot for shielding. The building crane had a 3-ton capacity, and the lead shot could be moved around as needed using a modified commercial vacuum sweeper. This module concept later appeared in the neodymium racks at Savannah River, and throughout the process cells in the TRU Facility.

This was a "hot" job. Some 400 pounds of slugs were received at Oak Ridge just 5 days after discharge from the Hanford reactor. The plant was designed for a nominal 2200 Ci of ^{140}Ba. But it actually shipped in one shipment 74,000 Ci that, after the 2 days' shipment time, they could safely assume that the lanthanum daughter was in secular equilibrium with the ^{140}Ba. Our assay, as I recall, was 68,000, but Los Alamos' assay of 74,000 Ci was probably the more accurate.

The plant was a lot "'crankier" to operate than it sounds like here. It took the skill of operators like Bob Schaich to make it work. He could probably add a lot to what I have outlined here, as, of course, could Ray Blanco. Eric Wischusen would have some unbelievable stories too.

5.5 REX E. LEUZE, SEPTEMBER 1991 AND OCTOBER 1992

My Exciting Trip to Oak Ridge. Monday morning June 25, 1945, I was up early so I could go from 5555 Pershing Avenue through St. Louis and across the Mississippi River to my job by 8:00 a.m. at Monsanto's Heavy Chemical Plant in Monsanto, Illinois. When I accepted a position following graduation from Kansas State, I requested that I not be assigned to the analytical lab. However, they insisted that was the place to spend a short time to learn about all the activities at the plant. Now, more than a year later there were no prospects of ever moving from the lab.

Shortly after arriving that morning, I was called into the office and told that some of the top managers wanted to see me across the river in Monsanto's head office. Back I went with three other Monsanto employees, Page Buckley, Les Burris, and Phil Schnelle. There we were told about this important new work at a distant site. It was so secret that they couldn't describe the type of work nor the location. The question was, "Are you

This 1977 photograph shows Rex Leuze, Pilot Plant Section Head (right front), with the Pilot Plant secretarial and support staff. Front row (left and right): Jan Thomas, Brenda Light, Chris Flanaty, Jewell Ellis, and Rex Leuze. Back row (left to right): Homer Thompson, Charles Roberts, Jane Posey, Bob Laxon, Karen Thacker, and George Ford.

willing to go?” My choice was easy. Here was my chance to get out of the analytical lab, so my answer was, “Yes!!” The others also agreed. The next question was, “How soon can you go?’ There seemed to he such an urgency that we all said we could leave anytime. “Well, can you be ready by noon day after tomorrow?” they asked. After another brief discussion, we all agreed that we could go on that short notice.

Monday afternoon was spent packing all my belongings. I couldn’t take everything with me, so several boxes were packed to be sent back home to my mother in Kansas. That evening, I walked up the street to 5630 Pershing Avenue to see if Ruth Morris (who became my wife in 1948) would mail those packages for me, since I didn’t have time to go to the Post Office. She agreed to help me, so all I had left to do was to finish packing, pick up my belongings at the Lab on Tuesday, and travel to Union Station by noon on Wednesday.

Only Page Buckley had any idea where we were going. He had the train tickets for all of us and a sealed letter that wasn’t to be opened until after the train left St. Louis. We were all anxious to learn where we were headed and what we would be doing. We boarded the train for Cincinnati. Soon after leaving the station, we opened the letter but didn’t learn much. It said something about going to a secret government plant near Byington, Tennessee, which was close to Knoxville. It also said that a limousine would meet us at the L&N Station in Knoxville about 8:00 a.m. on Thursday morning.

You can’t imagine how excited I was. I had never been east of Chicago, nor very far south of St. Louis. Tennessee was almost like a foreign country to me. All I knew was that it was somewhere in the Deep South, wherever that was.

The train from Cincinnati arrived in Knoxville during the night and left our pullman car in the L&N Station. We arose early-very early-and walked up that impressive staircase to the waiting room. We didn’t realize that Knoxville was on Central Standard Time (it was in 1945). But an hour after we had expected the car to arrive for us, here came Jim Rule to transport us to Clinton Laboratory. We drove out Oak Ridge Highway to the Clinch River and across the old Solway Bridge.

There we stopped at the guard station. Since Jim Rule had passes for us, we were soon on our way again. Such dusty roads! My thought was, "Doesn't it ever rain here?" I learned about the rain a few days later when the dust turned to mud!

The first person we met at the laboratory was Larry Riorden. He gave us the standard lecture on security and on not talking about anything that went on at the lab. Soon we were ushered up the stairs into the office of Bill Thumbser. We learned that Monsanto was taking over operation of the lab effective July 1 and that Thumbser was the new lab manager. All at once he said, "I guess you know what we're doing here?" We all shook our heads and declared that we were completely in the **dark.** Then he said, "We're working on an atomic bomb!!" What a shock!

A short time later, we were transported to the Guest House near Jackson Square where we resided until arrangements could be made for us to move into one of the nearby dormitories.

Later that week, after all the details required for working at the lab were completed, I was shown to my new job. I was to work with Sam Reynolds in Building **706D** (now 3026D) in the ***analytical lab.***

How exciting and thrilling it has been during the 42 plus years I had the privilege of working at ORNL. Even though there have been some difficult times, the people and experiences have been just outstanding! How glad I am that back on June 25, 1945, I accepted the challenge of venturing into the unknown.

Outstanding ***Scientists.*** Before I retired, I collected all of my published and oral papers that I could find. One statement that I made in the Forward of the Oral Presentations is very appropriate for the Chem Tech History. It went like this:

"Accumulating and editing this information has been a refreshing way to re-live over forty years at Oak Ridge National Laboratory. My most striking impression was the truly outstanding work by scientists like Rus Baybarz, Dave Campbell, Fred Kappelmann, Milt Lloyd, Boyd Weaver, and others as they evolved methods for the recovery of the **transplutonium** elements. They were able to develop processes based on scouting work with lanthanides to represent the actinides or with only tiny traces of some of the actinides. Later, when significant amounts of the transplutonium elements became available for recovery at the Transuranium Processing Plant, these methods worked and they worked well!"

5.6 PRE–CHEM TECH ERA: THOUGHTS FROM CLAIR W. SCHERSTEN, NOVEMBER 11, 1992*

I have dug into the annals of **not-always-** reliable memory to try to recall some early history of Chem Tech and its precedent groups in the old Technical Division. Those were interesting and challenging days with many highly capable people and many "colorful" characters. Some were both. For many of us, lifelong friendships were forming, as we were starting our "'careers" and our families.

Enough philosophizing! I will try to relate some amusing or otherwise interesting incidents or anecdotes. Thinking now of the **1946–49,** pre-Chem Tech period, I have retained a lot from those early years. So much of it is not recalling specific incidents but remembering individuals and my impressions of them. Or it is recalling typical everyday happenings, relationships, or scenes. I will relate several occurrences that may be of interest.

Good ***Fences Make Good Neighbors.*** The time: circa **1948–50,** when Dr. Frank Steahly was over Cheinical Development in the Technical Division. **The** characters: Frank Steahly and Warren Eister, who was in charge of the Semi-Works. Location: the Semi-Works was a separated extension of Building 706-A (now Building 3550). located northward and at the far east end of Building 706-A. **The** Semi-Works was surrounded by a fence. Access to this fenced-in area was only through the Semi-Works. The situation: Eister was in Steahly's **office** discussing progress on a new unit. They decided to go on an inspection. (The plot thickens.) Why go all the way to and through the Semi-Works to get there when they could take a shortcut from a back door of 706-A, cross a little open ground, and climb over the low fence? Who would see them? What difference would it make? It was late afternoon and most employees had already left.

So the "innocent" action was taken and **nobody** the wiser. But an alert guard had observed them and hastened to the scene. Seeing who they were,

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***Clair** Schesten, Administrative Assistant to Floyd **Culler,** Director of Chem Tech, transferred in 1961 **from** ORNL to Union Carbide Research Division at Charleston, West Virginia.

and knowing their positions, he only admonished them. There was no **gunfire!** Of course, he dutifully included a report of the incident in his activity log. Who should learn of this terrible deed but Larry **Riordan,** chief Security Officer of Oak Ridge National Laboratory! It immediately became his solemn duty and responsibility to call in the guilty parties--acting in the serious interest of national security! Chagrined, they dutifully went. They went because they knew it was a necessary exercise. In the privacy of his office Larry charged them: "You ***broke security regulations!*** Furthermore, your irresponsible disregard for serious rules ***set a very bud example.*** Especially bad, because you hold responsible positions."

Needless to say, all of their employees soon **learned** of-and profited from-their supervisors daring but costly deed. The profit, it must be said, was mostly in a good laugh and the opportunity to point fingers of shame at Steahly and Eister!

Social ***Life*** at ***ORNL. No*** doubt any Chem Tech history will not neglect the contribution of the annual St. Patrick's Day dances to the social life of ORNL. I probably have all of the printed programs from 1951-61 stashed away somewhere in boxes of mementoes moved here from Oak Ridge, many **not** opened since. I did **run** across a few in a **file** folder, along with a script copy from one of the famous skits.

The first Chem Tech Dance was actually on St. Patrick's Day, March **17, 1951,** at the Oak Terrace in Oak Ridge. Most of the skits were authored largely by illustrious members of the Long-Range Planning Committee. I am not sure whether this responsibility was included in their job description or not. But they did pursue the task with zeal and with dedication-none of the **effort** ever **performed** on company time, of course! The primary writer was usually Al Gresky, I believe. His talented and enthusiastic co-authors were Eldon Arnold and Bob Klotzbach. Then **there** was a motley crew of kibitzers and volunteer collaborators of varying talents, who contributed ideas-many of them actually acceptable, some even great. Of course, the high level of success achieved in the presentation of these annual dramas was **also** due to the very talented actors and actresses and production crews! And we must attribute some of the successes to the illustrious ORNL personalities, in or outside of Chem Tech, who were characterized in these plays!

Illustrious Personalities. When I **first** arrived in Oak Ridge and Clinton Labs in April of 1944, there was a little weekly newspaper, ***The Oak Leaf, that*** was distributed in the area. It was Oak Ridge's first newspaper. I think I got my copies at the desk of my West Village Dorm. It was edited by A. Carleton Jealous. I wondered if that was a pen name. After **some time,** I learned that this guy was one of the hundreds (maybe a hundred or more would be better) of Army personnel, mostly tech sergeants, who had degrees in science and engineering and had been pulled from their units and assigned to the Manhattan **Project.** They lived in barracks in the midtown area, I think Carl worked in the Y-12 area first. My memory says he came to the Technical Division about 1948 and was assigned to Eister's Semi-Works. He became Assistant Section Chief in Unit Operations when Chem Tech was formed He was one of about four Chem Tech engineers who transferred to the Union Carbide facility at Sterling Forest in New York about the mid-fifties.

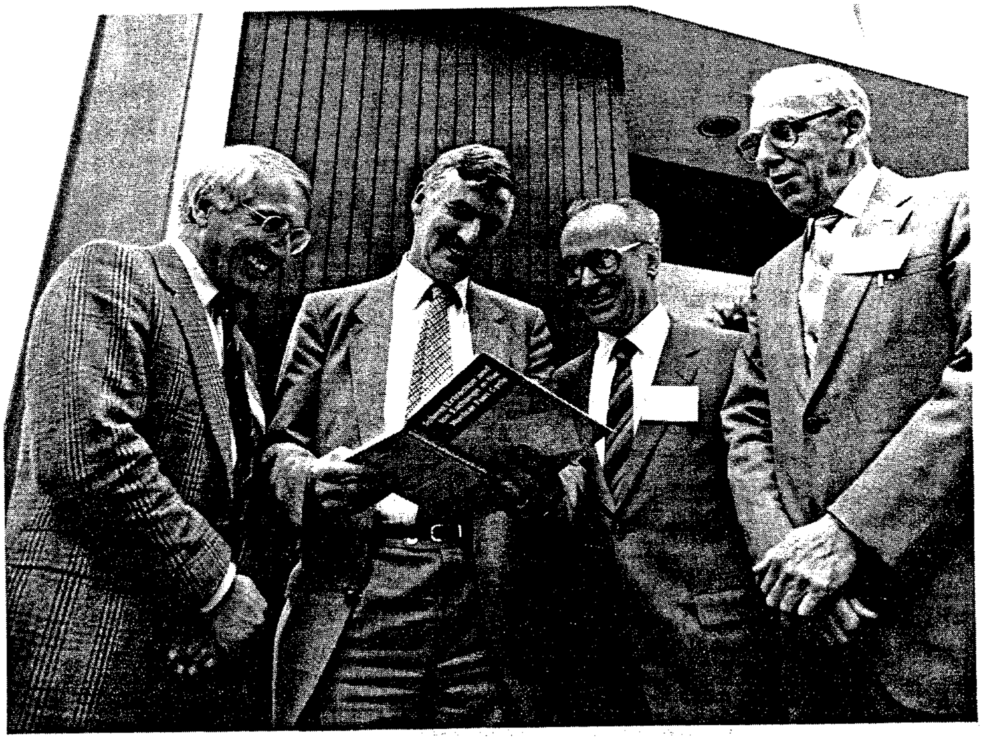
In the mid- 1940s other Chem **Techers** besides Carl Jealous had been in Army uniform. These included John Blomeke, Ed Nicholson, and Joe Savolainen. I hereby offer formal apology for any omissions or false inclusions! About 1946, two other guys hired in very soon after having been discharge&Don Ferguson from the Navy and Ray **Blanco** from the Army.

Chem Tech. We issued Chem Tech directories monthly. I'm quite sure that the original directory, February 1950, listed 110 personnel. Then, shortly thereafter, the Design Section under Floyd Culler was moved into Chem Tech. The Pilot Plant, under D. G.(Don) Reid, was in at the first.

Considering the successful contributions of Chem Tech and all its great personnel-and the more ordinary, but interesting, personnel-the historical treatise will **be** a boon to humanity! . . . Are you arranging to have it sent out in a capsule via rocket to unknown planets in outer space?

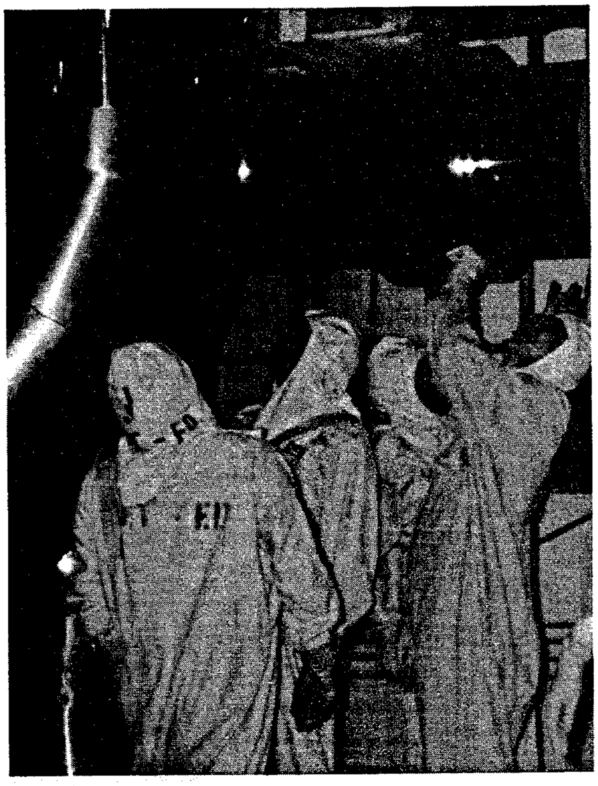
5.7 DAVID O. CAMPBELL, JULY 17, 1992

Unforgettable Colleagues. When I arrived at ORNL in 1953, Building 3508 was new and had just been occupied. There were several Du Pont employees developing processes and gaining experience for the Savannah River Plant, which was **almost** complete. Don Orth and Dave Karraker (still at Savannah River) were working on the



Dave Campbell (right) is shown in a 1985 visit to Harwell, United Kingdom, with (left to right) K. Ebert, Institut für Heisse Chemle Kernforschzentrum, Karlsruhe, Germany; Roy Nelson, Harwell; and L.Patarin, CEA, France.

Dave Campbell (looking up at a large pipe)i n the containment building at Three Mile Island during the TAAG inspection of the reactor system (after the nuclear meltdown incident).



ion-exchange process that was used at SRP for plutonium recovery, via the 3019 pilot plant. Several Du Pont people were working with John Cooper's analytical group to learn how to do the alpha analyses. The group leader was Don Overholt, about whom a book could be written. There simply are no characters in the lab anymore like he was, and they would not be tolerated if they were. Bob Rainey was there, and the technicians were Dave Hendrix, Jess Delozier, Brad Quincy, and Charlie Green. Collectively, this was the most interesting bunch of people I ever worked with.

Early Actinide Separations. My first job was equipment decontamination related to 3019. However, my introduction to actinide separations came in 1954. As always, the military wanted more plutonium. A program was set up at Oak Ridge to recover plutonium from some Hanford slag and crucible material. There was also about 10 g of americium in this material, and they wanted that also. So, the usual solvent extraction process for plutonium recovery was modified by adding an americium-recovery step, which Bob Rainey developed. All of the americium from the entire campaign was supposed to end up on a ***single*** cation-exchange column, from which it could be easily recovered. The process was carefully developed with simulants and tracers, and it looked great. They started up the plant, but after five or six days, americium started to break through the ion-exchange column. So they put on a back-up column, but it also broke through after only about five days. Before it was over, 22 columns were eluted, not just one.

Lanthanum Contamination in Americium. The teal problem turned out to be that there was about 3 kg of lanthanum, along with the 10 g of americium, that they wanted. The problem was to separate americium from rare earths, and on an impossible time schedule. They wanted 1 g in a few months and 5 g in a year. This is when I was called in and told to figure out how to separate americium from lanthanum, and real fast! At that time, no one had purified more that 1 g of americium, and that was at Los Alamos. I called Bob Penneman at Los Alamos and Bill Crane at Livermore. The most promising method, based on their experience, was HCl elution from Dowex 50 resin. We started out with that and quickly decided it was a hard way to make a living. Multiple cycles and lots of rework were required. I decided to continue that method in order to meet the commitment for 1 g but started looking for a better process for the rest of it.

The better process turned out to be chromatographic separation with citrate and Dowex 50 resin. Better complexing agents than citrate would appear later, but, at that time, Greg Chopin had not yet published about alpha-hydroxyisobutyrate. Using this method, we were able to recover and purify the remaining 9 g of americium and easily met the schedule.

In view of the present state of affairs, you might be interested in how this was done. I got a 5-ft-long piece of 6-in.-diam glass pipe and put a flow distributor screen in the bottom and a stainless steel flange on each end. It made a fine column that held 18 L of resin. We fed it from poly bottles using a finger pump with Tygon tubing (the rotary peristaltic pump had not been invented yet); the effluent came off in a Tygon tube to a jack-leg to control interface level, and finally to a poly product bottle. All this was done in a full-length hood-no glove box-by one technician and me. The hood had sliding Plexiglas doors like a bedroom closet.

Memorable Experience. One of my memorable moments occurred during the middle of the campaign. The gamma radiation from americium was a problem, so we kept as far from the equipment as we could. During an elution, we watched the column from across the lab. In this run, just as the americium peak was coming off the column, we noted that there was no solution running into the product bottle. Closer examination revealed that the effluent tube at the bottom of the column had split, and the product was running out on the floor of the hood. Fortunately, the floor was a stainless steel pan, but it was a memorable moment to observe a nice, pink americium solution, about 10 g/L, running into the pan. Although the leak was confined to the hood, it got the adrenalin flowing!

What did we do? We put on some extra gloves, yanked the split line off the column, and stuck a new one on. Throughout this work, carried out in open hoods, neither we nor the lab outside the hoods were contaminated to any serious extent.

Pressurized Ion-Exchange Separation of Actinides. It was some 20 years later when I started pressurized ion-exchange work, which has since been used in several countries for producing and purifying the heavier actinides. In the early nuclear days, there were at least two processes developed for every task. You may know that the bismuth phosphate process was used for the initial plutonium production at Hanford, but you may not know that cation exchange was the back-up process. It is a good thing bismuth phosphate worked, because cation exchange gave poor decontamination. However, there was a sizable program at Oak Ridge to develop ion exchange, and later on, they did a lot of work on ion-exchange chromatography.

One of these people was a biochemist, Waldo Cohn, who eventually went back to his field and became involved with the problem of separating transfer-ribonucleic acids (t-RNAs). Recalling his work on ion-exchange chromatography, he tried it for biochemical separations and it worked. Extreme resolution was required, and this could be achieved by using smaller resin particles. But as one moved in that direction, the flow rate decreased. The answer to that was to apply high pressure, which resulted in occasionally blowing up a column and splattering it all over the lab. Later on, better high-pressure components were developed, and Chuck Scott was instrumental in properly engineering them. It turned out that very dependable systems could be built and operated at 1000 to 1500 psi. These ion-exchange systems provided a large factor of improvement in performance-by a couple of orders of magnitude. This improvement could be taken advantage of in several different ways.

Now it turns out that these systems are admirably suited to actinide separations. It is interesting that development of this technique can be traced back to the wartime Manhattan Project work on ion exchange for separating fission products from plutonium, through biochemical separations, and finally back to actinide separations. The development for biochemical applications was driven by the need for extreme resolution. We needed good resolution, but the more practical problems were radiation damage to the resin and gas formation in the column because of radiolysis. With really high-activity actinides such as ^{242}Cm or Cf, gas generation often limited performance.

The high-pressure systems permitted much faster elution, thus reducing radiation damage. This high pressure, in itself, eliminated the gassing problem because gases are soluble and are carried out in the effluent solution. In spite of the obvious applicability of this approach, it was surprising how much opposition was encountered to using it with highly radioactive materials. In fact, the name had to be changed to "pressurized ion exchange," although the biologists called it "high-pressure ion exchange." The term high-pressure was too much of a red flag to get through a safety analysis.

However, I did demonstrate this method, and it was adopted and has been used extensively. Those of you who work on new processes know full well that nearly everything you do will never be used in practice, no matter how brilliant your work or how useful and practical the process. That is just the way life is in our business. Very few processes are ever used at all, and the way the decision is made to choose a particular process is weird, at best. So there really is a feeling of satisfaction when a process we have something to do with is actually used.

5.8 RAY E. BLANCO, JUNE 1, 1992

The "Retread" Campaign. In late 1963, Floyd Culler decided his "troops" were getting "rusty and worn" and that their skills needed rejuvenation. After all, most of them had received their degrees about 15 to 20 years earlier. Wallace Davis, Jr., was appointed professor for physical chemistry and Jere Nichols for mathematics. ORNL provided the books. We were to attend classes in the conference room on the second floor, Building 4505. The program soon became known as the "retread program." Classes met for 2 h once a week for 9 months. About 30 "students" signed up. The course was held twice-November 1963 to May 1964 and November 1964 to May 1965. Wallace led us through the trials of physical chemistry and Jere poured on the advanced algebn and calculus. Homework was assigned and faithfully completed, although Wally Davis said that I was remiss in some of my homework! We had a ball. Just like going back to old times. We even complained that the professors were too hard on us!

International Atomic Energy Agency (IAEA) Panel Meeting, Dubna, U.S.S.R. — 1964. I was fortunate to receive several interesting trip assignments-lucky to be in the right place at the right time. Joe Lieberman, Assistant Director for Nuclear Safety, AEC, Washington, called me at home on a Saturday to ask if I could attend an IAEA waste meeting at Dubna, U.S.S.R., as the United States representative. It was only about two weeks notice for a meeting to be held in December 1964. I said, "Sure could, if ORNL management agreed." They did agree.

Several Chem Tech staff and guests are pictured at an early 1960 conference in Gatlinburg, Tennessee. Left to right are Don Ferguson, Wilma Ferguson, Ray Blanco, Elaine Blanco, Edna Briggs, and Beecher Briggs (Reactor Division).

Dubna was a new city (since 1944) of apartment buildings on the banks of the Volga River, about 70 miles north of Moscow. It is the main center in the U.S.S.R. for high-energy research and employed about 4000 people. Excellent meeting facilities were available here. We were given a tour of the cyclotrons and other accelerators and their thermonuclear test unit. We were free to walk around the town and take pictures, as we wished. There were many people fishing through holes in the ice on the Volga.

The emphasis of the meeting was on the exchange of information on the incorporation of low- and intermediate-level wastes in various types of concrete and the new concept of incorporation into asphalt (bitumen in Europe). The Russians and others were extremely interested in the work being done in Chem Tech by Herschel Godbee, Arlene Kibbey, and John Moore. Of particular interest were the Chem Tech results of irradiation damage to the asphalt. Results of our work reached the Russians regularly through the IAEA, where the U.S.S.R. delegates combed the literature for new information.

The principal value of the meeting to us was the opportunity to hold direct discussions with the Russians since none of their written reports on waste treatment reached the United States. In addition to the IAEA representative, one official delegate was present from each participating country, that is, Belgium, Czechoslovakia, France, Germany, India, Japan, United Kingdom, U.S.S.R., and the United States. However, the Russians had about 20 unofficial “onlookers.”

On the bus back to Moscow on the final evening, it was again one Russian and one visitor per seat. I had a Russian lady chemical engineer as my companion. We had a nice, pleasant conversation, and I didn’t argue with her since she was much bigger than me! The group (a full bus) had become very friendly during the week of meetings, and there was a lot of loud shop talk-some direct and some through interpreters. The Russian leader passed down the aisle with 2 bottles of vodka, paper cups, and apples for those who wanted to celebrate the end of the meeting (the Russian lady chemical engineer did not). Soon the bus sounded like football fans going to a game-loud national songs and laughter. My lady companion sang like a valkyrie at the opera. The ‘IO-mile, nighttime trip to Moscow over a road covered with ice, snow, and potholes took a long time.

***Teaching at the University of São Paulo, Brazil* —** 1969. Professor Pedro Bento de Camargo of the Institute of Atomic Energy (IEA), São Paulo, Brazil (associated with the University of São Paulo) contacted Alvin Weinberg with a proposal for ORNL to provide post-graduate credit courses in nuclear technology. Negotiations were completed for financial backing and support from the U.S. AID, IEA, and the AEC in mid-1969. Three people were given the teaching assignment; that is, Paul Kasten and Bill Ergen, Reactor Division, were to give courses on reactor economics and reactor physics and Ray Blanco, Chem Tech Division, a course on radioactive waste management and nuclear fuel reprocessing.

The salary problem was resolved with the ORNL participants to remain on their normal salary and trip expense reimbursement. The IEA paid us their normal salary for professors in Brazilian currency, which we returned to ORNL-AEC.

I was very worried about the whole thing because Brazilians speak the Portuguese language, no textbook was available, and I didn't visualize myself as a professor. I finally rationalized that it could be no worse than a series of 50-min. lectures at the annual Chem Tech Information Meeting, so I set out working to prepare all of the lectures in advance, that is, the complete text and figures (copies of slides). I planned to hand them out in advance as a textbook. I was assured that the students read and understood English reasonably well, since most of their college texts were in English. Thus, I could speak using the slides as a guide, and using the blackboard, I could get the message across. I slaved for several weeks at ORNL and at home preparing the lectures and slides. I am forever grateful to Sue Damewood for typing and Jim Farmer for drawing up the figures and slides *on* a rush-rush basis. It seems that permission from the AEC always arrived late for foreign visits. I expected about 17 students and prepared 25 packages of lectures.

We arrived in late September and classes started immediately. I had shipped all of my reference literature and lecture packages by air mail in advance. Disaster struck immediately!! My box of lectures, etc., were being detained in the Brazilian Customs warehouse. They claimed that the package contained pornographic material but, in reality, they were extorting ransom from the University and Institute. Fortunately, I had brought my copy of the lectures and my slides in my personal luggage. I had to start and make the best of it for two weeks until the IEA got the shipment released They could not reproduce the lectures from my personal copy so that I could distribute them immediately because of a lack of reproduction facilities. In the end, it all worked out very well. One of the resident professors helped grade the exam papers since the students were allowed to write in either Portuguese or English. We returned to this country in early December. The lectures were published by the IEA in 1976 and 1977 in *three* volumes as ***Radioactive Waste Management—Informacão,*** IEA 53-l 1 CPRDO02.

The information I presented came from declassified literature publications which were referenced in the lectures. Many of the referenced authors were from Chem Tech and are listed as co-authors on the lecture series, including W. E. Unger, A. R. Irvine, D. J. Crouse, and C. D. Watson.

Professor Camargo asked us to prepare a preliminary survey of an Agro-Industrial Nuclear Complex for Brazil. A group in my Chem Tech Section had been active in that program at ORNL in studies on the production of fertilizer using nuclear power and developing a method for preventing scale formation on the evaporator tubes in the desalination distillation system (a patent was issued to Walt Clark, Bill Yee and me for the latter). An initial survey was completed in cooperation with Brazilian experts on agriculture, minerals, heavy industry, and hydrology. Northeastern Brazil is very dry and the location of a Nuclear Complex in that area to distill seawater (or to pump groundwater) and to produce fertilizer looked promising.

Sidelights. We stayed at a hotel in the center of São Paul0 (population approximately 7 million in 1970). Our balcony overlooked eight lanes of Volkswagens moving day and night-it was like living on Times Square. Leaving Brazil proved difficult. Our visas had to be stamped for approval to depart. The officials claimed a new law had been passed in the last month saying that you had to give them one month's notice to leave. Thus, it cost each of us an extra $10 to depart the country.

Delegation to the Institute of Nuclear Energy Research, Taiwan, 1979. In 1979, the U.S. State Department and the DOE decided to send a delegation of U.S. nuclear scientists to Taiwan as an assistance program. The U.S. had recently withdrawn its ambassador because of a problem with mainland China and relations were a little strained. A team was formed composed of S. Goldsmith (Fuels Development - Battelle Northwest), W. Schwartz (Quality Assurance and Control - DOE Idaho), W. Lapinski (Nuclear Reactors - Argonne National Laboratory), Ray Blanco (Waste Management - ORNL), and A. Lewis (Leader, Argonne). We spent a couple of days consulting in Taipai and the remainder of two weeks at the Nuclear Institute in the interior of Taiwan. The Institute is a smaller ORNL centered around a heavy-water-moderated, natural uranium reactor. Areas of interest to me were the counter-current bench-scale mixer-settler solvent extraction system, research and development in waste management, and waste treatment operations for the installation. They were using a process for incorpomtion of low- and intermediate-level radioactive wastes into asphalt and had developed a process using a mixture of polyethylene and asphalt to increase the melting point of the product. During the tours of the facilities, I found that there was a strong rivalry between the chemical engineers doing R&D in the Engineering Research Division and those doing the actual operations in the Operating Division. On my first morning of separate meetings with these people, I had a problem! I found myself at the head of the room facing six R&D engineers at a table to my left and seven operating engineers at a table to my right. A man from each side came to the front table and presented me with a glass of tea-one with sugar with a lotus flower floating in it and the other, plain (my "one-on-one" man was on the R&D side). They were sort of smiling and watching to see what I would do. I drank out of both glasses immediately and the tension evaporated. We had several sessions like this where they would ask about the work at ORNL and bring up problems for discussion. At the end of the visit, the Institute management indicated that they were pleased with the efforts of the delegation. They were very gracious hosts!

Sidelights. At that time, the Republic of China (Taiwan) considered itself to be still at war with mainland China. Soldiers with rifles and bayonets were stationed at all bridges, tunnels, airport, and other strategic locations.

My "one-on-one man" Dr. Chou spoke excellent English and had received his Ph.D. degree from the University of Louvain in Belgium using the Flemish and French languages! He and a friend took two of us to an evening theater performance about an old Chinese legend. The actors were 11 to 13 year old children who had made their own traditional Chinese costumes and performed in the old style with slow, stilted movements. It was very beautiful and expressive! Before the performance, I noticed some empty front seats in the theater. At the last minute, they were filled when several generals and civilians marched down the aisle accompanied by two soldiers with rifles and fixed bayonets.

We were taken on a tour of the huge National Museum. Chiang Kai-shek had moved most of the beautiful treasures from China to Taiwan. They change out the displayed treasures each six months, and they have sufficient numbers to ensure that nothing is repeated over a 30-year period.

5.9 ROBERT L. JOLLEY, AUGUST 7, 1992

January 2, 1956. My introduction to the strange and exotic world of fluorine, uranium hexafluoride, and molten salts occurred the day I started working in Chem Tech. I was assigned to George Cathers fluoride volatility group (Labs A-25 and B-25 in Building 4500) and was introduced rapidly to the unique oxidative powers of fluorine via the burning leather glove demonstration; thermocouples and potentiometers; pressure gauges and "needle" control valves: using an aluminum rod as stethoscope to detect the methodical "heart beats" of the hydrogen fluoride dissolver and the fluorinator; and radiation detectors and radioactivity. To a neophyte from an organic and analytical chemistry background, it was all an exotic adventure in physical chemistry—a wonderful new world to explore and learn.

Unforgettable Team. George, with a deep and practical understanding of physical and inorganic chemistry, was an excellent mentor. George's group also included Bob Bennett and Bob Duff. Both were astute experimentalists and superb craftsmen. Bennett was a master chess player and often served as the straight man for Lou Byrd, who worked in a lab close by. Regular visitors to our lab included Bill Ratledge, janitor for Corridor A, 4500, and Yankee supporter par excellence. Later Cather's group was joined by Dave Campbell, a genius in separations chemistry and kinetics who had recently returned from the Army: Stan Kirslis, who could have discovered the "impossible" reaction of fluorine with xenon; Tom Crabtree, a football referee by avocation with a heart of gold; Cal Shipman, a quiet, steady, and reliable team member; Frank Soard, thorough and competent and former baseball player: and Gene Moncrief, a young chemical engineer in training for volatility pilot plant adventures. Only years later have I realized how unique and talented the team was-a familiar refrain I am sure for most ORNL scientists and engineers, for ORNL was a meeting place for many of the nation's brightest.

Joule-Thompson Effect and the Fire Department. I had been in Cathers' volatility group perhaps half a year. It was fall and the trees were turning-flaming golds, crimson, and reds. The grass behind Building 4500 (there was no 4500S at that time) was a dead brown. George assigned me the task to dispose of the ClF_3 previously used by Rex Leuze and Chuck Schilling in fluoride studies. Chlorine trifluoride, a gas at room temperature, is stored as a liquid under slight pressure. Because ClF_3 is highly reactive, with properties similar to fluorine, George and I attached a 10-ft length of 0.25-in. copper tubing to the bottle to ensure that the gas would disperse sufficiently far from 4500 and personnel.

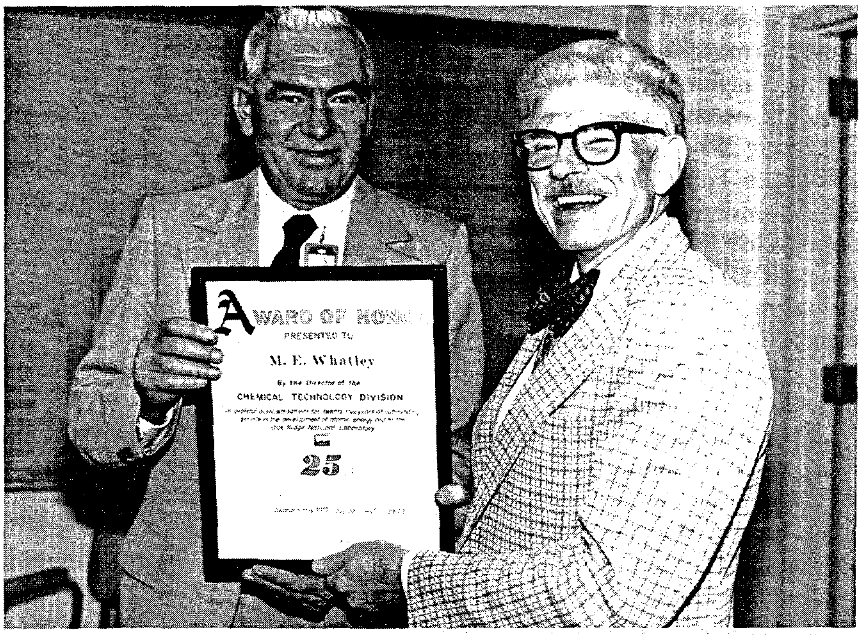
We carried the ClF_3 tank across the street behind 4500 and set it up not too far from the fence that ran between the street and the small creek behind 4500. We aimed the tubing away from 4500 and slowly cracked the valve to release the ClF_3 gas. Several minutes later we noticed what appeared to be a jet of colorless liquid shoot from the end of the tubing, arching beautifully several yards, and landing on the dry grass. Instantly, the grass caught fire. By the time we had closed the tank valve and removed it to a safe place, the fire was rapidly approaching the fence. The fire extinguisher, alertly brought by John Harris, Corridor B janitor, who also called the fire department, was effective except where the fire had passed through the chain length fence and appeared to be racing rapidly towards the woods.

The ORNL Fire Department soon arrived and positioned themselves for extinguishing the grass fire. When the pressure was cranked up on the tank truck, the top seal blew, and wet many of the onlookers and firemen. This was quickly corrected and the fire was extinguished.

A red-faced young chemist never forgot the Joule-Thompson effect again-that most gases, except hydrogen, cool on expansion. And ClF_3 liquifies at 11.3°C. The Fire Department did not forget either. For about a year, I received weekly calls and visits from the Fire Chief to ensure that I was focused on fire safety. To my knowledge, the ClF_3 may still reside undisturbed and undisposed within the hallowed walls of A-25, 4500N.

5.10 THE CREMATION OF JIM BRESEE'S R2-XG (A FANTASY): MARVIN WHATLEY, 1958

The following poem indicates that renowned scientists also have lighter moments. It is included by popular demand.



Don Ferguson, Director of Chem Tech, presents Marvin Whatley (right) with a plaque recognizing 25 years of company service in 1978.

THE CREMATION OF JIM BRESEE'S R2-XG

(A Fantasy)
by M. E. Whatley (about Xmas 1958)
with apologies to Robert W. Service

There are strange things done for a processing run
To recover reactor fuels,
And the engineers' wails hold their fearful tails
That would shatter your rational rules.
These fluorescent lights have seen strange sights,
But the strangest they ever did see,
Was the night on the brink of the awfullest stink
When we process the R2-XG.

Now the R2-XG is nine foot three
with fused U 0 2 for its core
plus T H 0 2 in quantities true
(Or perhaps just a little bit more)
Which at 5000 K was fired for a day
To make particles perfectly round
which are quickly dispersed in a manner rehearsed
And found quite philosophically sound.

This is floated in NaK and squeezed through a crack
Fifteen mils by six inches or so
That produces a plate, the most stable to date,
Which is clad by the method we show:
Take zirconium pure with some iron from the Ruhr
And a very thin layer of gold,
Then molybdenum, tin, and a platinum fin
With a delicate film of green mold.

A mega kilo watt year it is burned, then shipped here
Where its cooled sixty seconds or less.
"We'll process the thing in the Unit Op Wing
Without even making a mess."

So read our claim, out pride was to blame,
"Just think of the prestige we'll gain.
Recover the stuff and the waste well enough,
and I'll talk at Geneva again."
It was then that Bresee stated clearly his plea
Not to go off half-cocked in this race.
"Some few problems involved are yet to be solved,
It will take us a week at our pace."

Then the section took hold, you could tell by the cold
Quiet drive that pervaded its men,
And we wrote twelve reports of various sorts
And the coffee breaks ended at ten.
Oh the sweat that was shed, the technicians half dead
Under pressure fantastic to see.
But the group never mired, it was truly inspired
By its glorious leader, Bresee.

We'll dissolve it in salt while we whip it like malt
Using H F at one thousand K,
Or we'll chop it in bits and grind it like grits
And bake it real solid like clay.
The reagent to sell might by day H C L,
So the volatile chlorides we'll lose,
When the reaction stops the remains will be tops,
For an Aqueous process we'll choose.

The debate then grew hot: What to use and what not
Till the engineers' chins dragged the ground.
Bresee simply remarked (and the reaction sparked)
"A solution will surely be found."

Like a bright gleam of light to illumine the night
Where we stumbled and frequently fell,
From the janitors stall 'Why not just use them all?
The reaction will go bent for Hell!"
Thirty stories or more coming up from the floor
The dissolver was built in a day:
Pulsed plates for and aft and a super down-draft
Condenser to reflux away.

Wet, dry, hot and cold, with saws in the hold
And filter to take up the fines.
The still strong of heart were now ready to start,
The others were off in the mines.
Bresee gave the sign and the four-inch feed line
Gave a belch We were finally on stream.
Our handiwork ran, the reaction began
You could tell by the large cloud of steam.

There are strange things done for a processing run
To recover reactor fuels
And the engineers' wails hold their fearful tails
That would shatter your rational rules.
These fluorescent lights have seen strange sights,
But the strangest they ever did see,
Was the night on the brink of the awfullest stink
When we burned up the R2-XG.

5.11 CAREER OPPORTUNITIES, 1943*

George Parker had a hunch in early 1943. That hunch has had a lot to do with where he is in 1993 as a researcher in ORNL's Chemical Technology Division.

During the early, nervous years of World War II, the former University of Tennessee graduate student had taken a munitions industry job at the Indiana Ordnance Works.

The E. I. Dupont de Nemours Company then operated Indiana Ordnance, and shortly after Parker joined Dupont the company began to transfer employees. "It was all very secretive,' Parker said. "Nobody knew or would say where they were going or what they were going to do."

Meanwhile, Parker had also enrolled in a night class taught by a Purdue University engineering professor, who returned from a trip to the University of Tennessee with an interesting story for the class. "He said that he had just come from Knoxville where he visited UT's engineering and physics departments, and he was amazed to see four Nobel laureates in one group!" (Parker now speculates that the four were probably Arthur Compton, Harold Urey, Earnest Lawrence, and Sam Allison.)

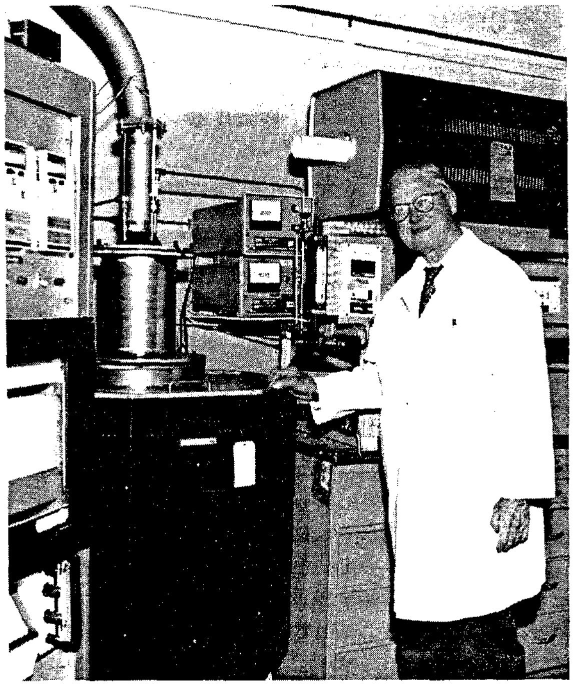
Parker's professor then told of a project getting under way close to nearby Clinton that might be connected to the plethora of brilliant scientists present at the school. In fact, someone at UT had put a note on a blackboard that said, "What's going on at the Clinton site?'

'The professor said that there were all kinds of wild guesses listed under it, such as chemical warfare," Parker related, but one speculation was noted as "uranium energy," and it clicked with the young chemist.

Parker recalled an article in ***Physical Review*** that announced that German scientists had fissioned uranium by exposing it to neutrons, yielding a very high energy content.

"I immediately associated it with the blackboard guess," Parker said.

At that time the selective service was inducting eligible young men at a rapid rate, and Parker's draft board in his hometown of Johnson City, Tennessee, suggested that if he intended to continue his new career uninterrupted, he should seek employment that was more vital to the war effort.



George Parker's canny observations on the purpose of a big wartime project In hls home state of Tennessee led to a 50-year career at Oak Ridge National Laboratory.

"I had a draft board at home in Johnson City who kept pretty good track of me and what I was doing, and they suggested I should find a more strategic job. One day a friend told me that he had seen so-and-so at the University of Chicago who was temporarily assigned there."

'That gave me an idea to write a letter to the University of Chicago registrar that I had reason to believe there was an important defense effort being conducted at the school, and I would like to be in on it. After about the third day I got a telegram that said simply 'come up for an interview prepared to stay."'

Parker resigned from the ordnance plant, packed his few belongings, and left for Chicago. Once at Chicago, Parker met with people from the "Metallurgical Project Laboratory" and was interviewed by gentlemen who he later discovered were FBI and military intelligence agents. 'They were extremely interested in what I knew." Parker told them how he had put his hunch together from the professor's reports and the journal article. He was told that his guesses were wrong but that they

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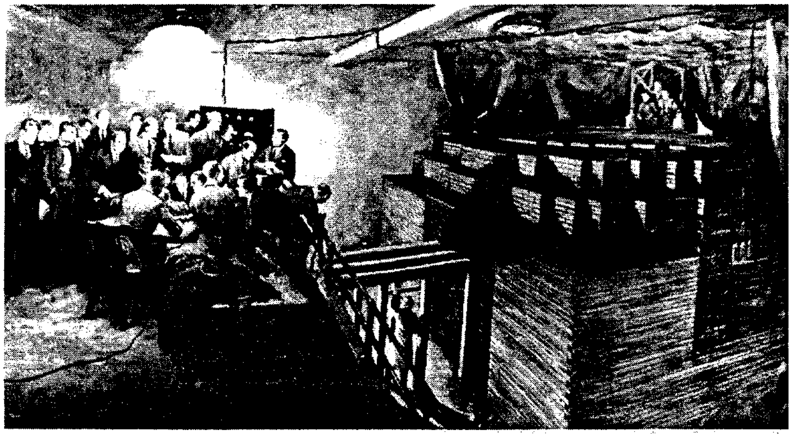
*Appeared as an article written by W. H. Cabage in ***Lab Notes,*** No. 26 (January 1993).

had a job for him, anyway, in the **biochemistry group** isolating **trace** quantities of **fission** products from uranium nitrate taken **from the** St. Louis cyclotron's shielding. The day after he started work **he was** told **that,** in **truth,** the agents were **actually** amazed at the accuracy of his speculations.

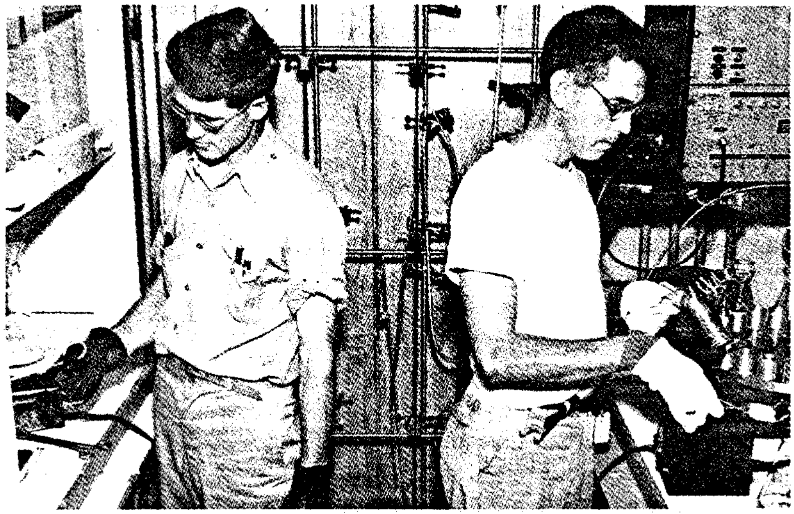
Parker was assigned to a laboratory next to the university's Stagg Field squash court, **the** site of **the** graphite pile of the first nuclear reactor. A slightly romanticized painting depicts the famous pile and the scientists **who** developed and operated it **standing on the** squash court's balcony. Parker's first uranium extraction lab was in a locker room adjacent to **the** balcony. "I could walk out and look at the pile any time," Parker said.

The pile by then had been **shut** down and a better **one was** being built, as Parker had suspected, near Knoxville. In October 1943 Parker was transferred to the barren East Tennessee site called Clinton Laboratories.

"I took a train from Chicago, checked into the laboratory's personnel office in the L&N **station,** and was instructed to get a bus to Oak Ridge and obtain quarters in **the** guest **house."**



George Parker's first laboratory at the University of Chicago was adjacent to the balcony, depicted In the painting, overlooking the historic Stagg Field squash court that served as the site of the first nuclear reactor.



This photo dating to around 1946 shows George Parker (left) and P. M. Lantz Isolating rare-earth elements from fission products This early separation work yielded valuable material for safety studies and served as a beginning of the Isotope Industry.

"After a bus ride down Highway 62, they checked our papers at **Elza** Gate and let us in. I asked the bus driver to indicate where the **guest** house was. Suddenly the bus driver stopped and said 'this is where you get out.' I got out and immediately sank in mud almost up to my knees."

"People who were knowledgeable wore high boots," Parker said.

Conditions at Oak Ridge were spartan, but eventually Parker, after experiencing the shady politics of waiting lists (he kept slipping down them), was able to obtain **housing** for himself and **his new** bride. **Entertainment** was available in Knoxville, which, since there were no cars, involved a journey on "atrocious buses." Transportation to **the** reactor site was via "cattle car," a **converted** car carrier equipped with a woodstove heater.

The Parkers became friends **with** several noted scientists. They babysat for the Arthur Comptons. Parker said he also knew Enrico Fermi, Harold Urey, and Edward Teller informally, along with Leo Szilard, Robert Oppenheimer, and General **Leslie** Groves, **who once came through** with a quartet of four-star generals in tow.

Such visits were not uncommon. Preparations for an anticipated visit by President Roosevelt, which never happened, included making sure all corridors were clear enough for a wheelchair to pass.

Parker also came to know several "strange and wonderful" individuals, including a foreign fellow who bathed in the quarters' community shower in his skivvies, then wrung them out and dressed. A mechanical wizard from Knoxville named Arthur Tripp once fashioned a radial engine out of tin cans.

Another fellow always wore a topcoat and a felt hat, even in summer. One day he failed to show up for work-he had disappeared. His mother demanded to know where her son was, but officials had no clue. Parker said the man's job was very ill-defined: although he was a scientist, no one knew exactly what he did. Parker never did find out what had happened to him.

There was no open discussion then about the Laboratory's mission, although Parker said that he was aware that Y - 12 was enriching uranium-235 and that the Graphite Reactor was a pilot plant for plutonium separation. An extremely important issue was neutron yield, Parker related, and it became apparent that plutonium would make a better bomb than uranium.

"I saw some of the first milligrams of plutonium that were separated at the Graphite Reactor. There was a microchemist from Berkeley, Louis Werner, who even though he was a microchemist was one of the biggest men I ever knew, about seven feet tall. One day he asked me to look at a small amount of blue-green liquid under a microscope. It was the first visible quantity of plutonium made in the Oak Ridge reactor, and from that tiny amount they made a lot of extremely important measurements.*'

Parker's work during the war was in the separation of radioactive materials for biological damage safety studies. "Our efforts were the basis for the radioisotope industry," Parker said. "All of our early work was with carbon-14, iodine-131, phosphorus-32, and sulfur-35, biologically important isotopes that were generated in the Graphite Reactor and separated in the old 706-C hot lab (now Building 3026)."

His first actual knowledge of the atom bomb came in July 1945, when he saw a report that a test bomb had been successfully fired in Alamogordo, New Mexico. At that point an in-house controversy had blossomed over the use of the atom bomb, Parker said. "Many well-informed individuals were against it."

However, almost half a century later, Parker sat in his office, which is computerless and stacked with reports and documents, and described his learning of the use of the atom bomb and the resulting end of the war as his best day at the Lab.

"The darkest day may he yet to come," he said, "but I've been lucky. I've had experimental work continuously."

Not a bad career just for putting two and two together.

5.12 CHARLES F. COLEMAN, MARCH 31,1993*

Extraction of uranium and thorium from geological materials (e.g., granite, sands, shale) is one of the most remarkable achievements of ORNL and Chemical Technology Division research and development. This work received international acclaim, and many of the extractants developed or studied by the group were used not only for commercial extraction process, but also found application in nuclear fuel reprocessing. Some of the unique accomplishments of the R&D team responsible for developing the separations chemistry were described in Sect. 4.1. That modest writeup provided a brief history of the R&D team. This anecdotal section includes some additional photographs of some Chem Tech personnel and visiting international scientists (Figs. 5.1-5.3).

5.13 HISTORY OF THE CHEMICAL TECHNOLOGY DIVISION SAINT PATRICK'S DAY DANCE, KAYE JOHNSON

From the very beginning, the staff of the Chemical Technology Division has always "worked hard." As a reward for their endeavors, they decided long ago that they should also "play hard," and what better theme for celebrations than St. Patrick, the patron saint of engineers. Approximately a year after the formation of the Chemical Technology Division, a celebration was planned for Saturday, March 17,195 1. Thus the first Chem Tech St. Patrick's Day Dance was held. The place was the Oak Terrace Ballroom in Oak Ridge, Tennessee. The music was provided by a

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*Prepared by Robert L. Jolley.

Fig. 5.1. international scientists visited the Chemical Technology Division for discussions of extraction and separations chemistry In the fledging nuclear fuel reprocessing area. This October 19, 1962, photograph shows (left to right): Andre Grieneisen, in charge of plutonium purification by solvent extraction, Direction Industrielle, Montrouge (Seine), France; Hal E. Goeller, Chem Tech; Claude Bernard, Chemical Engineering, Saint-Gobain Nucleaire, Courbevoie (Seine), France; Clyde D. Watson, Chem Tech; Andre Bathellier, in charge of plutonium purification by solvent extraction, Centre d'Etudes Nucleaires, Fontenay-aux-Roses (Seine), France; Al D. Ryon, Chem Tech; Michele Journet, Service Control Department, Saint-Gobain Nucleaire; Charles F. Coleman, Chem Tech; and W. (Bill) F. Schaffer, Chem Tech.

ten-person orchestra conducted by Bill Dexter. The dance chairman was Johnny W. Clark. His subcommittee chairpersons were Ruth Pennington, decorations; Clyde Watson, entertainment; and Taylor Napier, tickets. One of the highlights of the evening was a group called the Spark Plugs, who evidently functioned successfully without a leader. The Spark Plugs included such notable characters as Frank Rogers, Garrett Parker, Ed Johns, Al Gresky, Bob Klotzbach, Bill Unger, George Sadowski, Jim Lockmiller, Virginia Malone, Cathleen Foster, and Frances Bray. The evening festivities began at nine and lasted until one. In addition to the orchestra, entertainment provided by division staff members included songs, dances, Irish music, door prizes, a quartet contest, and what was to become the traditional skit. The first skit was written by Don Ferguson, presented by Don Ferguson and Company, and lasted 8 minutes. Obvious by omission in the historical records is any description or photographs of the skit. However, it must have made quite an impression on the attendees because there has been some type skit every year for the past 42 years. Ticket sales were ***strictly limited to*** 300; in the true Chem Tech tradition, 301 tickets were sold, at a cost of $1.00 each plus a $0.20 entertainment tax.

Historic records show that many successful managers were strong participants in the annual St. Patrick's Day Dance activities. On August 11, 1953, F. L. Culler was authorized to sign checks written on the Chem Tech Dance checking account. In 1953 he became the division director (Fig. 5.4). For many years after he left the division, he would RSVP his plans to attend the dance by telegram. Sadly, the records for 1952, 1953, and 1954 do not contain any descriptive materials about the skit. However, we do know that in 1954 the production

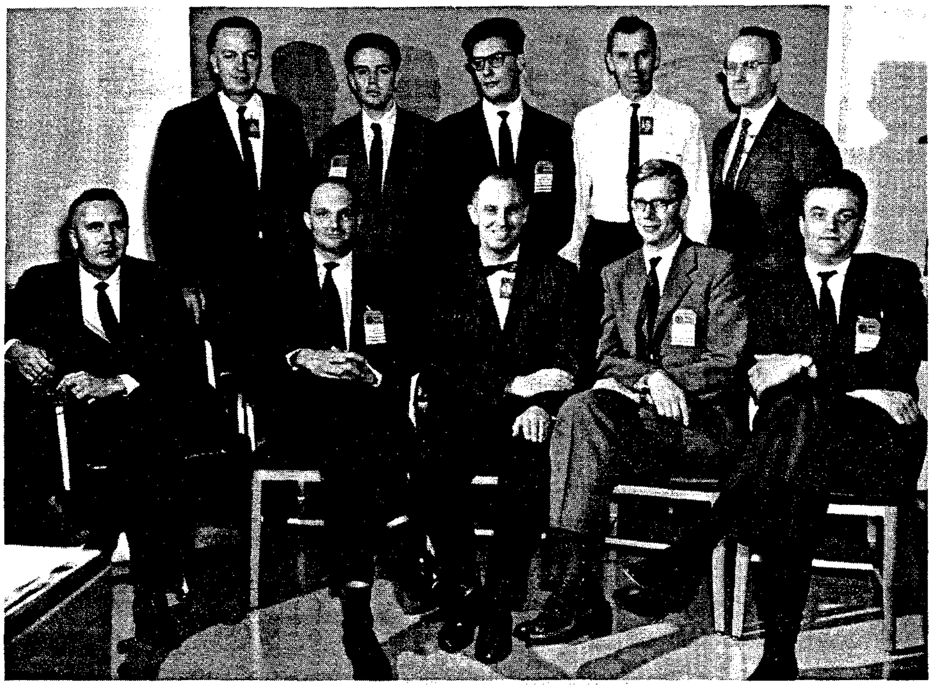


Fig. 5.2. international scientists visited the Chemical Technology Division for discussions of extraction and separations chemistry In the fledgling nuclear fuel reprocessing area. This October 30, 1962, photograph shows seated (left to right): Keith 8. Brown, Chem Tech; A. S. Kertes, Hebrew University of Jerusalem, Israel; Charles A. Blake, Chem Tech; Erik **Axel Strandell**, AB Atomenergi, Stockholm, Sweden; and Giacomo **Calleri**, CNEN, Rome, **Italy**. Standing (left to right): Dave Crouse, Chem Tech; G. Scibona, CNEN; F. Baronceiii, CNEN; Boyd Weaver, Chem Tech; and Charles F. Coleman, Chem Tech.

Fig. 5.3. Jim Hardy of the Atomic Energy Research Establishment (AERE), **Harwell**, England, was a visiting scientist in **Chem** Tech on a l-year assignment to study separations chemistry. This April 23, 1963, photograph shows (left to right) Charles Blake, Keith Brown, Jim Hardy, and Charles Coleman.

Fig. 5.4. Floyd Culier (third from right) with Chem Tech guests at an early St. Patrick's Day Dance. To Floyd's right is Larry Corbin and to his left are Mrs. Culier and Mrs. Browder.

company changed its name to the Atomic Thespians.

Other notable researchers and scientists made their debut in the 1957 production of ***Oak Ridge Revisited.*** Scene IV, entitled "Ballet Grotesque or The Dance of the Fairies," featured Ray Blanco, Bill Burch, Hal Goeller, Chuck Guthrie, John Landry, Dick Lindauer, Taylor Napier, Bob Milford, Kyle Stooksbury, and Jack Uliman (Fig. 5.5). Bill Burch, later to become Director of the Fuel Recycle Division, performed in his first ballet recital in Fig. 5.6.

In 1959 coaches were chartered and the dance was held at the Beaver Brook Country Club in Knoxville. WCTD, the division's "Thru Channels" Station, presented ***The Scissor-Frantics,*** starring Larry Shappert as Peter Gum, with Paul Haydon and Tom Roberts as the cigarette and checkroom girls (Fig. 5.7). Larry Shappert was not seen or heard of again until 1962, at which time he served as chairman of the dance committee.

The decade of the sixties brought America protesters to the Vietnam War, long-haired hippies, and Woodstock. It brought Chem Tech Brenda Light, Janice Shannon, Jimmy T. Bell, and Martha Stewart. The following is a list of the dance skits developed and presented during that decade:

1960	***Beauty and the Beasts***
1961	***Please Mr. Collar***
1962	***The KKK, Kollar's Kemical Kollege***
1963	***The Wonderful World of Collar***
1964	***School Daze***
1965	***Coldfinger***
1966	***A Night on CTD TV***
1967	***A Happening***
1968	***Do You Remember***
1969	***CTD Laugh-in or Misinformation Session of 1969***

It is hard to overlook the talent that Les King, Chairman of the 1962 Program Committee, discovered that year. First, there was Alice McWilliams in her famous fishnets (Fig. 5.8). Next, was Fred Kitts and Vic Fowler in their bold horizontal-stripe motif (Fig. 5.9). Last, but not least, was John Bigeiow in his virgin cells attire (Fig. 5.10).

As the Chem Tech Division grew in number of employees, patents, discoveries, publications, and technical expertise, the one thing that remained

Fig. 5.5. The ballet team featured in the Ballet Grotesque or *The Dance* of *the Fairies* are (left to right) Bob Miiford, Bill Burch, Taylor Napier, Kyle Stooksbury, Dick Lindauer, Chuck Guthrie, Hal Goeiler, Ray Bianco, Jack Uliman, and John Landry.

Fig. 5.6. Bill Burch performs in the first ballet recital at the St. Patrick's Day Dance.

Fig. 5.7. Paul Haydon (left) as a cigarette girl with Larry Shappert as Peter Gum in *The Scissor-Frantics* skit. In the background is Tom Roberts as a checkroom attendant.

Fig. 5.8. Alice **McWilliams** in her famous fishnets with Bruce Finney as Frayed L. **Kollar** and **Vic** Fowler as the doorman.

Fig. 5.9. Fred Kitts (left) and Vic Fowler as inmates in *Kollar's Kemical Kollege*.

Fig. 5.10. John Bigelow as Virgin Cells with Bob Rainey In the background in the 1962 ***Kollar's Kemical Kollege*** skit.

unchanged over the decades was the acting ability of the St. Patty's Day Dance casts and the caliber of the skits that were written.

The dance activities of the seventies were equal to previous years' events in every possible way. In 1971, the archives show that "203 people attended the social hour and consumed 2.0 gallons of unspiked punch, 10.3 gallons of spiked punch, and 16 pounds of potato chips and peanuts." The skit that year, one of the dance's earliest musicals, was entitled ***The Devil Made Us Do It,*** starring Eldon Arnold, George Dinsmore, and Steve Goldberg. The original, score was provided by John Holmes. The words to one of the songs sung in this production have proven to be ageless. Sung to the tune of "America, The Beautiful," the words are as follows:

Oh beautiful for smoggy skies,
Insecticided grain,
For strip mines, mountains, majesty
Above the asphalt plain.
America! America!
Man sheds his wastes on thee,
And hides the pines
With billboard signs
From sea to oily sea.

The Happy Hippy Flowerpot Dance Company, who also made their appearance that year, consisted of Linda Knauer, Donna Fitzgerald, Nancy Pope, Linda Loveday, Les Hubbard, Mike Hartsell, Bob Oliver, and Kyle Stooksbury.

Remarkably, only two Chem Tech St. Patrick's Day Dance skits have gone down in infamy at ORNL. The first was *M. Hartman, M. Hartman* which premiered Friday, March 18, 1977, in the Social Hall of St. George's Greek Orthodox Church in Knoxville, Tennessee. The idea was to produce the first (and, to date, final) full-length (25-min.) feature movie. The subject of the movie was a parody on lab activities. The cast consisted of Ron Glass, Ray Barker, Mike Gilliam, Jan Talbot, John Parrott, Jr., Brad Weil, Bill Eidridge, Tom Dinsmore, John Younghance, the remaining Chem Tech section heads, and the CTD associate directors. Rex Leuze only agreed to appear in the production because he was told that the last scene was to be a pie fight among the section heads, and Rex wanted to throw a pie in Chuck Scott's face. Chuck Scott did not show up for the filming. The movie cost $165, which included the developing of 20 rolls of film. The reason the film is legendary is because the pie fight was filmed in Building 4505 Conference Room and the janitorial crew was not pleased with having to clean up the mess. As a result, an official ORNL announcement was issued that effective immediately film crews were not allowed on the site without official permission. Postscript: Jim Snider won the door prize that year, a magnum of New York State champagne.

The second infamous film was ***Martina Marietta and the Seven Benefits,*** presented in 1984. In true Chem Tech tradition, this production was quite topical and timely in that it tried to capture all the concerns about the potential change in company benefits as a result of the Department of Energy changing its Oak Ridge Operating contractor from the Union Carbide Corporation, Nuclear Division, to Martin Marietta Energy Systems. The cast consisted of Beverly Sweeden in the title role and the seven benefits played by Suman Singh, Becky Hamley, Debbie Weaver, Karen Pannell, Donna Snow, Martha Dawson, and Mary Ann Neal, with Miliicent Drake, Otis Scates, Phil McGinnis, Ray Wymer, and Bob Hightower. The production was narrated by John Parrott and directed by Jonathan Woodward and Bob Wham. All preparations, plans, rehearsals, and costume preparations Were going well when suddenly, two days before the dance, an official complaint was registered with the ORNL Affirmative Action Coordinator. Ray Wymer, the division director at the time, was called and told to take down all posters and pictures advertising the dance. It seems that someone in another division felt that the women in the production were being exploited. And so, after 33 consecutive years of presenting a skit, the division officially scored poorly with the theatrical critics. The offensive picture is presented here for an objective opinion (Fig. 5.11).

We believe the Chemical Technology Division Annual Saint Patrick's Day Dance is, by far. the longest running social activity of any division at ORNL. In 1992 the division celebrated the 43rd annual event. A skit entitled ***Weekday at Begovich's*** proved beyond the shadow of a doubt that theatrical talent in the division has been maintained and that very little, if anything, has been lost in the tradition of skit writing. The purpose of the first and all subsequent skits was to allow the staff an opportunity to poke fun at the management of the division. This concept was the forerunner of what is know today as stress management.

Fig. 5.11. Clockwise from center ara Suman Singh, Mary Ann Neal, Donna Snow, Beverly Sweeden, Becky Hamley, Martha Dawson, and Karen Pannell as actors In the 1994 *Martina Marietta and the Seven Benefits* skit.

5.14 OTHER RECREATIONAL ACTIVITIES

Over the years, Chem Tech personnel participated in every recreational opportunity made available to ORNL employees. Although a thorough search of *ORNL* News items was not attempted, several examples follow.

Bowling. Many Chem Techers were avid bowlers. Figure 5.12 shows Bill Burch receiving an award for bowling a perfect game.

Softball. Both fast pitch and later slow pitch softball teams were sponsored by Chem Tech. A 1955 fast pitch team is shown in Fig. 5.13. A slow pitch Chem Tech Pilot Plant team was fielded in the late 1960s, as shown in Fig. 5.14.

White water. During the mid-1960s, several Chem Techers participated in white water activities sponsored by the Carbide Canoe Club. An especially favorite float was the IO-mile hip on the Emory River from Nemo Bridge near Wartburg, Tennessee, to Oakdale after heavy rains. John Landry and Arnold Lyle are shown in Fig. 5.15 amidst the roaring white waters of the Emory river on a July 17, 1967, float hip following a heavy rain in the Cumberland Plateau. The summer of 1967 had frequent heavy rains, and Arnold and John are shown on an August 5, 1967, Emory River float in Fig. 5.16. The end product or conclusion of that float is shown in Fig. 5.17. Bob Jolley is shown running a rapid solo on the July 17 Emory River run in Fig. 5.18 and with his son Richard on the August 5 Emory River Run (Fig. 5.19).

Fig. 5.12. Bill **Burch** (left) **receiving** a bowling award from the **ORNL Recreation Department** manager.

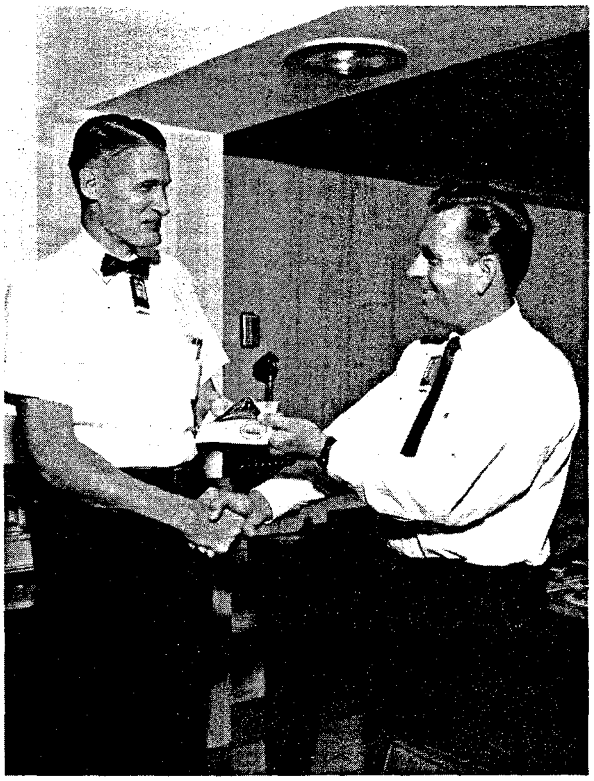


Fig. 5.13. The 1955 Chem Tech fast **pitch** softball team members were, front row, left to right, **Marvin** Whatley, unidentified, Vic **Fowler,** and John **Parrott**, Sr.; Back row, left to right, unidentified, unidentified, unidentified, Sam Clinton, Pete Newman, **Milt Lietzke,** Arky Vaughn, and Bob Blair.

Fig. 5.14. The Chem Tech **Pilot** Plant slow pitch team, approximately 1967, members were, first row, left to right, Fred **Chattin,** Jerry West, and Joe **Tinley** (I&C Division); second row, left to right, unidentified, Sam Shell, Fred Kitts, Bill Lindsey (team manager), Ron Cooper, Bob Shannon, and unidentified; Back row, left to right, Jerry Beeler (P&E Division), Rob Waters (K-25), Ray Cooper (Health **Physics** Division), Don Taylor, Ray **Bolden** (P&E Division), unidentified, **Bill** Stines (M&C Division).

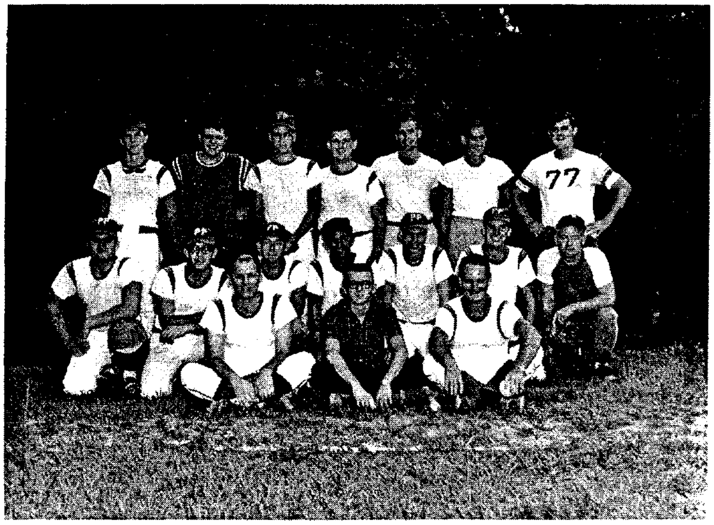


Fig. 5.15. John **Landry** (left) and Arnold Lyle approach a monstrous standing wave on the July **17, 1967,** Emory River float trip.

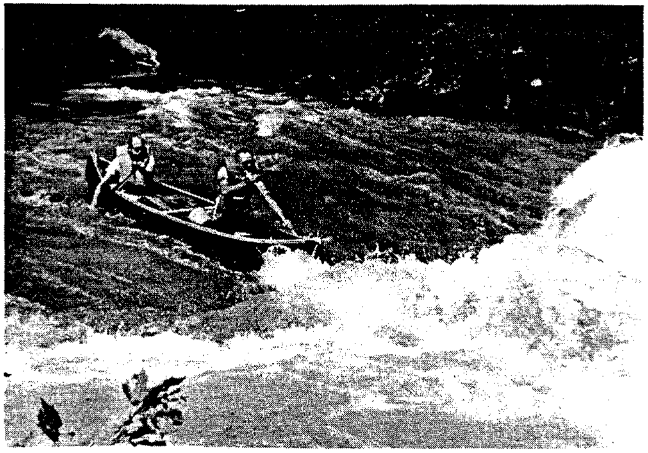
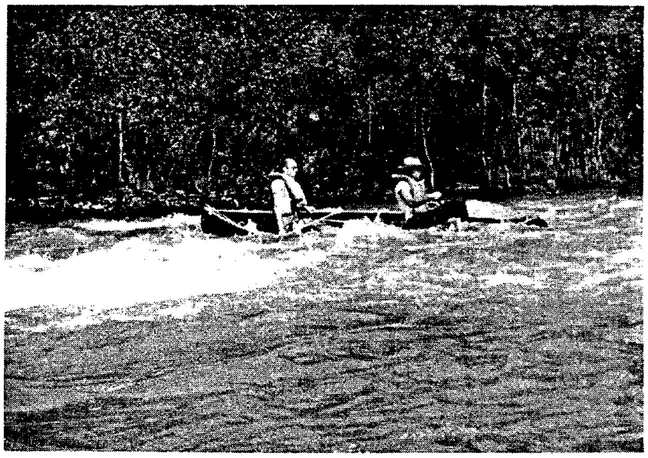


Fig. 5.16. John Landty (left) and Arnold Lyle on the August **5, 1967,** Emory River float.



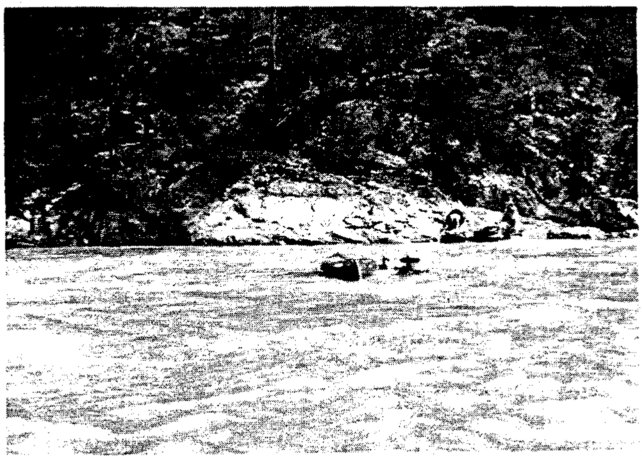


Fig. 5.17. John Landry and Arnold Lyle (still with hat) am shown with capsized canoe. Such a conclusion following a large rapid was often encountered.

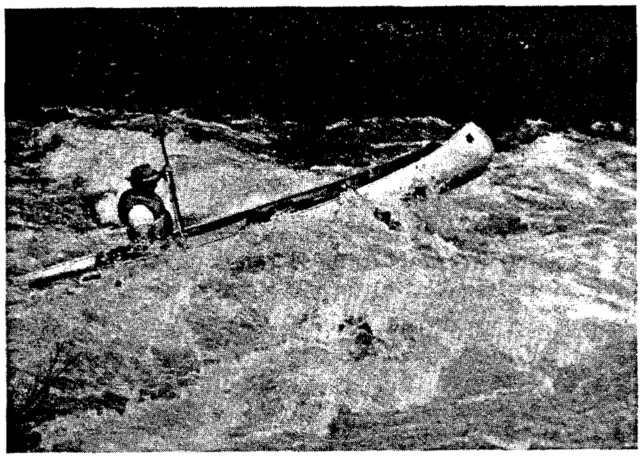


Fig. 5.18. Bob Jolley ran the 10-mile float solo on the July 17, 1967, Emory River float and encountered some heavy white water.

Fig. 5.19. Bob Jolley and son Richard Jolley negotiate a rapid on the August 5, 1967, Emory River float trip.

APPENDICES: THE CURTAIN REMAINS OPEN

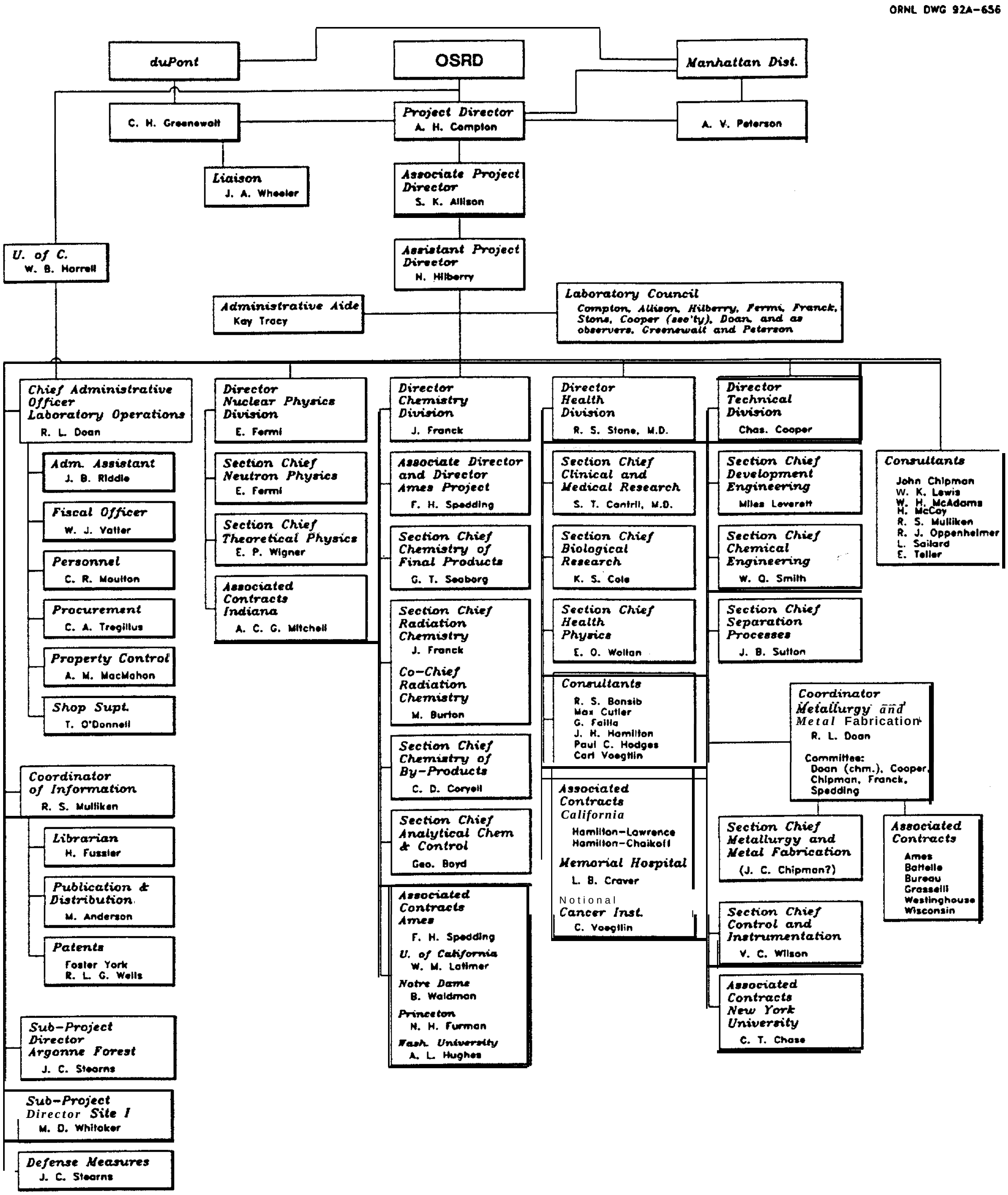
APPENDIX A

A.1 ORGANIZATION CHART FOR THE METALLURGICAL PROJECT,UNIVERSITY OF CHICAGO, JANUARY 23, 1943

A.2 ORGANIZATION CHART FOR THE TECHNICAL DIVISION, OAK RIDGE NATIONAL LABORATORY, 1948

APPENDIX A.1

ORGANIZATIONAL CHART FOR THE METALLURJGAL PROJECT, UNIVERSITY OF CHICAGO, JANUARY 23, 1943



APPENDIX A.2

ORGANIZATION CHART FOR THE TECHNICAL DIVISION, OAK RIDGE NATIONAL LABORATORY, 1948*

M. C. Leverett-Director	Ethel Tuttle, Sec. Louise Bond, Sec.
W. M. Hawkins, Assistant Director Administration	Eleanor Pippin, Sec. Evelyn Gilliland, Clerk. Jane Snow, clerk.
T. W. Hunaerford, Assistant to Director Projects & Safety	Eunice Greenway, Sec.
J. R. Whitlock, Administration Assistant Procurement	Virgil Reynolds, Clerk June Hale, Clerk
Stuart McLain, Special Assistant	Jean Kuna, Sec.
M. D. Peterson, Chief Section T-l Process Development	Marie Wolfe, Sec. Agnes Hair, Typist Ann Leinart, Sec.
C. W. Schersten, Assistant to Chief	
C. D. Watson, Assistant to Chief	
W. K. Eister, Assistant to Chief - Semi-Works	
F. L. Steahly, Assistant Chief - Laboratory and Plant Assistant	
D. E. Ferguson, R. E. Leuze, Group Leaders 23 Extraction (J. W. Gost) (C. V. Ellison)	J. L. Bamberg, Tech. G. C. Balock, Tech R. C. Lovelace, Tech. H. F. Soard, Tech
A. C. Jealous, F. R. Bruce, Group Leaders 25 Extraction F. N. Browder J. 0. Davis E. 0. Nurmi G. A. West W. A. Home - Shift Supervisor C. D. Hylton - Shift Supervisor G. S. Parker - Shift Supervisor	R. 0. Chambers, Tech. G. B. Diimore, Tech. J. E. Farmer, Tech. G. R. Guinn, Tech Guy Johns, Tech W. H. Luster, Tech F. L. Rogers, Tech. W. E. Shockley, Tech J. W. Smith, Tech.

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*This organization chart for the Technical Division was supplied through the courtesy of Dr. Miles Leverette. Director of the Technical Division, 1943-1948.

F. R. Bruce, Group Leader, 25 Extraction	A. Johnson, Janitress C. A. Clark, Tech.
R. E. Blanco Arlene Kibbey W. B. Lanham L. P. Morse A. T. Gresky - Special Assignment	L. A. Byrd, Tech. W. B. Howerton, Tech. Vannesse Orr, Tech. E. R. Jones, Tech. D. Q. White, Tech.
M. R. Poston, F. N. Browder, Group Leader Solvents	B. I. Bailey, Tech. Gladys Howser, Tech.
F. L. Steahly	J. M. DeLozier, Tech V. L. Fowler, Tech.
D. C. Overholt T. C. Runion	R. B. Quincy, Tech.
W. K. Eister	T. D. Napier, Tech.
L. R. Higgins J. B. Ruch C. D. Watson G. A. West	
R. B. Briggs. Chief, Section T-II Engineering Development	Thelma Sutton, Sec.
S. B. Beall, ***Leader*** - Control Elements Group	A. L. Davis, Tech J. J. Hairston, Tech
T. H. Mauney	
J. Reed, Leader - Corrosion Group	W. Kirkland, Tech. J. L. Stepp, Tech.
0. Sisman, Leader - Pile Irradiation Engineering Assistant Group	R. L. Townes, Tech.
R. Van Winkle, Leader - Scale Formation and Water Treatment Group	C. M. Burchell, Tech W. B. Krick, Tech. R. Smith, Tech.
C. D. Bopp J. B. Chtisney	
W. B. Allred, Leader - Strength of Materials Group	
H. C. Savage	
W. H. Stromquist - Special Problem C. E. Clifford - Special Problem C. P. Coughlen - Special Problem	G. H. Johnstone, Tech.

(B. W. Kinyon) - Research Shops Coordinator

. R. Huffman, Chief, Section T-Ill - Process Design

Mary Dougher, Sec.

C. F. West, Jr. - Administrative Assistant

D. Nicoll, Assistant Chief

C. E. Winters, Associate Section Chief

A. D. Mackintosh

J. A. Lane, Associate Section Chief

R. M. Jones, Joint Leader, Group A
Pile Proper

N. E. Hill
D. Nicoll
S. Scott, Jr.

C. W. Day, Draftsman
R. C. Allerbe, Draftsman
A. S. Ludlow, Draftsman
H. W. Watts. Draftsman
(C. A. Roberts), Draftsman
(Sue Eatherly), Clerk

J. T. Weills, Joint Leader, Group A, Pile Proper

W. S. Farmer
W. G. Stockdale

G. Hovorka, Leader, Group B - Pile Buildings

F. C. McCullough, Leader, Group C - External Systems

W. R. Gall, Leader, Group D - Pile Mockup

D. J. Mallon
J. R. McWherter
R. A. Long
W. E. Unger
F. C. Zapp

F. M. Culler, Leader, Group E - 1200 - 1300 Areas

G. Hanson
H. E. Goeller
R. L. Klotzbach
R. P. Milford

J. A. Kyger, Chief, Section IV **Engineering** Materials	Ruby Bullard, Sec. Susan Comisk Sec.
W. L. Cockrell, Assistant Chief	
F. A. Kocur - Individual Assignment	
C. D. Smith, Leader, Group A	F. Blackshere, Tech C. F. **Cutcher,** Tech. J. H. Day, Tech.
G. M. Adarnson H. Wallace	
F. W. Drosten, Leader, Group B	J. N. Hix, Tech.
J. T. Howe D. A. Lawson V. L. **McKinney**	
F. **Kerze,** Leader, Group C	C. C. Cooley, Tech
G. M. **Carlton** J. E. Cunningham W. H. Wilson	
J. L. English, Leader Group D	R. N. **Tench,** Tech.
A. R. Olsen S. H. Wheeler	
T. Rockwell, Leader, Group E	
F. J. Roehrenbeck	
(J. H. Erwin)	(Harry Seaman), Machinist
R. N. Lyon, Chief, Section V - Engineering Research	C. C. Hum, Tech.
D. G. Reid, Chief Supervisor, Acting Chief **Section VI - Pilot Plant**	Jeanne **Doran,** Sec.
H. M. Feder, Assistant Chief Supervisor	R. F. Benson, Operator H. S. Caldwell, Operator
H. K. Jackson	H. G. Chambers, Operator G. Davis, Operator
M. **Levenson,** Assistant Chief Supervisor	L. L. Fairchild, Operator C. A. Gifford, Operator

K. K. Kennedy, Senior Supervisor
W. H. **Carr**
F. M. **Grizzell,** Operator
J. H. **Groover,** Operator
W. Jenings, Operator
C. H. Jones, Operator

E. L. Nicholson, Senior Supervisor
G. Sadowski
J. Land, Operator
W. E. Ledbetter, Operator
J. F. Lo&miller, Operator
T. R. **McLellon,** Operator

N. J. **Rigstad,** Senior Supervisor
B. B. **Harrington**
J. R. Ogle, Operator
R. P. Purkey, Operator
M. Richardson, Operator
H. E. Sexton, Operator

A. M. Rom, Senior Supervisor
E. M. Shank
E. E. Shields, Operator
B. J. **Stradder,** Operator
G. **Tipton,** Operator
H. C. Thompson, Operator
J. T. Wiggins, Operator

(W. P. Bigler **- Argonne)**

Hired: Monthly **- Nurmi**
Weekly - Bond

<u>Teminated</u>: Monthly **-** Bigler, Bomwasser, **Burris, DeHaan,** Ward
Weekly **-** Allen (January) **Caraglin**

<u>Transferred In</u>: Leinard from Purchasing Department (Weekly)

Personnel on loan to the Technical Division from other departments as shown in parenthesis.

	2-4-48	**3-1-48**
Monthly (Technical)	105	101
Weekly (Non-technical)	87	**88**
	192	**189**

APPENDIX B

ORGANIZATION CHARTS FOR THE CHEMICAL TECHNOLOGY DIVISION

B.l ORGANIZATION CHART FOR 1950

B.2 ORGANIZATION CHART FOR 1960

B.3 ORGANIZATION CHART FOR 1970

B.4 ORGANIZATION CHART FOR 1981

8.5 ORGANIZATION CHART FOR 1991

APPENDIX B.l

ORGANIZATION CHART FOR THE CHEMICAL TECHNOLOGY DIVISION, JUNE 1, 1950*

- F. L. Steahly, Director
  - W. K. Eister, Special Assignment
  - C. W. Schersten, Assistant to Director
  - F. R. Bruce, Section Chief - Laboratory
  - F. L. Culler, Section Chief - Process Design
  - J. 0. Davis, Section Chief - Unit Operations
  - D. G. Reid, Section Chief - Pilot Plants

PERSONNEL DISTRIBUTION

	Monthly	Weekly	Hourly	Total
Admimistration	2	4	0	6
Laboratory	16	16	4	34
Process Design	13	1	0	14
Unit Operations	8	13	0	21
Pilot Plants	13	2	18	33
TOTALS: (6-1-50)	52	36	22	110
(2-20-50)	41	33	22	96

CHANGED SINCE FEBRUARY 20

	Monthly	Weekly	Hourly
Hired:	R. B. Lindauer J. W. Ullman	Mary Pembenon J. C. Suddath Virginia Wells	
Transferred in:	F. N. Browder F. L. Culler H. E. Goeller R. J. Klotzbach R. G. Mansfield F. C. McCullough R. P. Milford W. G. Stockdale W. E. Unger R. H. Vaughn	Lucille Kuykendall	
Terminated:		W. H. Luster	

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*Source: ***Chemical Technology Division Progress Report for Quarter Ending May 31.1950,*** ed. W. K. Eister, ORNL 763.

- F. L. Steahly, Director
  - June Parrott, Sec.
  - Agnes Hair, Sec.
  - Virginia Wells, Sec.
  - W. K. Eister, Special Assignment
  - C. W. Schersten, Assistant to Director
    - J. W. Clark, Tech.
    - A. Johnson, Janitress
  - F. R. Bruce, Section Chief - Laboratory
    - Mary Pemberton, Sec.
    - T. C. Runion, Problem Leader - Metal Recovery
      - C. V. Ellison
      - R. G. Mansfield
      - C. E. Schilling
      - J. L. Barnberg, Tech.
      - V. L. Fowler, Tech.
      - H. F. Soard, Tech.
    - W. B. Lanham, Problem Leader-Purex
      - D. 0. Darby
      - G. A. Eaton
      - J. R. Flanary
      - L. A. Byrd, Tech.
      - E. R. Johns, Tech.
      - W. B. Howerton, Tech.
      - C. F. Keck, Tech.
      - H. S. Caldwell, Opr.
      - C. H. Jones, Opr.
    - R. E. Leuze, Problem Leader - Dry Fluoride
      - C. P. Johnston
      - H. B. Graham, Tech.
      - A. B. Green, Tech.
    - R. E. Blanco, Problem Leader - RaLa
      - A. H. Kibbey
      - G. C. Blalock, Tech.
      - S. E. Farmer, Tech.
    - D. C. Overholt, Problem Leader - Isolation
      - D. E. Ferguson
      - W. E. Tomlin
      - J. M. Delozier, Tech.
      - R. B. Quincy, Tech.
      - J. F. Land, Opr.
    - L. E. Morse, Problem Leader Homogeneous Reactor
      - R. C. Lovelace, Tech.
- F. L. C ller, Section Chief - Process Design
  - Lucille Kuykendall, Typist
  - Draftsman
  - W. E. Unger, Project Engineer - RaLa
    - L. Brewer #
    - J. B. Ruch
    - R. H. Vaughn
    - E. Wischhusen #
    - R. L. Berry #
    - R. V. Foltz #

F. N. Browder -Liquid Waste

H. E. Goeller - "25"

R. J. Koltzbach - "25"

F. C. McCullough - Cost Analysis

R. P. Milford - Metal Recovery

E. L. Nicholson - "25"

A. M. Rom - "25"

E. C. Stewart - Metal Recovery

W. G. Stockdale - Gaseous Waste

On loan from Engineering.

J. 0. Davis, Section Chief
Unit Operations

Ruth Pennington, Sec.
C. J. Shipman, Tech.

A. C. Jealous,* Assistant Chief

T. A, Arehart[1] - Evaporation

M. R. Bennett

I. R. Higgins[2] - Ion Exchange

D. B. Masters, Tech.
W. E. Shockley, Tech

H. 0. Weeren[x]

A. C. Jealous*[3] - Solvent Extraction

w. L. Carter[x]
F. P. Pike #

P. 0. Payne, Tech.

C. D. Watson - Equipment Development

J. C. Suddath
G. A. West

E. 0. Nurmi, Shift Coordinator -Metal Recovery

F. Mills

G. B. Dinsmore, Tech.
G. R. Guinn, Tech.
G. Jones, Tech.
T. D. Napier, Tech.
F. L. Roger, Tech.
J. C. Rose, Tech.

D. G. Reid, Section Chief - Pilot Plants

Phyllis Groover, Sec.
R. B. Walters, Draftsman

H. K. Jackson.’ Associate Chief

H. K. Jackson,* Program Leader - **Purex**

- F. E. Harrington
- J. W. **Landry**
- W. H. Lewis

N. J. **Rigstad,** Problem Leader, CR Separations

G. S. Sadowski, Assistant Leader

- A. T. Gresky
- C. D. Hylton
- R. B. Lindauer
- J. W. Ullman
- R. **H. Winget**

R. F. Benson, Operator
J. H. Groover, **Operator**
W. Jennings, Operator
W. E. Ledbetter, Operator
J. F. Lockmiller, Operator
T. R. **McLellan,** Operator
R. E. Purkey, Operator
H, L. Sexton, Operator
E. E. Shields, Operator
R. C. Shipwash, Operator
D. E. **Spangler,** Operator
B. J. Strader, Operator
D. G. Summers, Operator
H. R. Thomas, Operator
H. C. Thompson, Operator
J. T. Wiggins, Operator

A. M. Rom. Coordinator - “25” Process Data

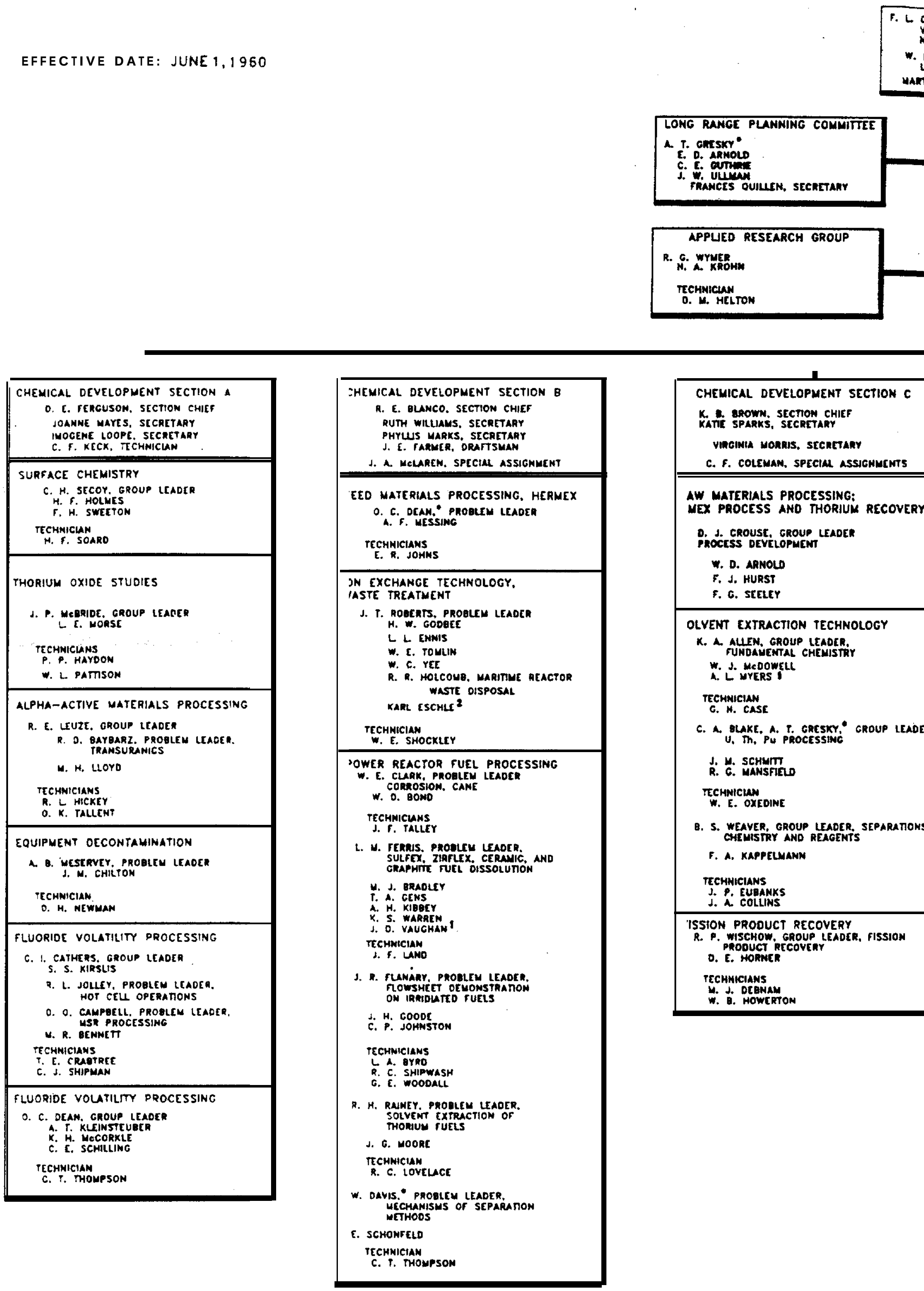
E. M. Shank, Problem Leader
Waste Processor

G. D. Davis, Operator
F. M. **Grizzel,** Operator

1 - responsible for Purex in this Section
2 - responsible for RaLa in this Section
3 - responsible for Metal Recovery in this Section
- Research Participant
* - dual capacity
x - on loan from Engineering

CHEMICAL T

EFFECTIVE DATE: JUNE 1, 1960



F. L. C

W.

MAR

LONG RANGE PLANNING COMMITTEE

A. T. GRESKY*
E. D. ARNOLD
C. E. GUTHRIE
J. W. ULLMAN
FRANCES QUILLEN, SECRETARY

APPLIED RESEARCH GROUP

R. G. WYMER
N. A. KROHN

TECHNICIAN
D. M. HELTON

CHEMICAL DEVELOPMENT SECTION A

D. E. FERGUSON, SECTION CHIEF
JOANNE MAYES, SECRETARY
IMOGENE LOOPE, SECRETARY
C. F. KECK, TECHNICIAN

SURFACE CHEMISTRY

C. H. SECOY, GROUP LEADER
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THORIUM OXIDE STUDIES

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E. R. JOHNS

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W. E. TOMLIN
W. C. YEE
R. R. HOLCOMB, MARITIME REACTOR WASTE DISPOSAL
KARL ESCHLE[2]

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K. S. WARREN
J. D. VAUGHAN[1]

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R. C. LOVELACE

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E. SCHONFELD

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MEX PROCESS AND THORIUM RECOVERY

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F. J. HURST
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D. E. HORNER

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M. J. DEBNAM
W. B. HOWERTON

TOTAL PERSONNEL

TECHNICAL	161
SCIENTIFIC	101
ADMINISTRATIVE AND CLERICAL	30
TOTAL	292
SUMMER EMPLOYEES	9

* DUAL CAPACITY
[1] RESEARCH PARTICIPANT
[2] FOREIGN RESEARCH PARTICIPANT
[3] EMPLOYEE OF RADIATION APPLICATIONS.
[4] CONSULTANT, U. T.

Source: Chemical Technology Division Annual ***Progress Report for Period Ending August 31, 1960***, ORNL-2993.

HNOLOGY DIVISION

APPENDIX 8.2

ORGANIZATION CHART FOR JUNE 1, 1960

ORNL DWG 92-660

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B. B. KLIMA*
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C. H. JONES
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CAROLYN GHEEN, SECRETARY

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W. L. PATTISON

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K. S. WARREN[6]

TECHNICIANS: C. W. GREENE
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R. C. LOVELACE

G. I. CATHERS, GROUP LEADER, IODINE RETENTION IN LMFBR FUEL PROCESSING
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RADIOIODINE CHEMISTRY, RADIATION STABILITY OF SOLVENTS IN LMFBR FUEL PROCESSING

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PLOWSHARE PROGRAM

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W. D. ARNOLD

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UNIT

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LI

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E. L

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C. W
D. W
H. C

TECHN

POWE

C. D.
S. D
R. L
W. S
H. F
R. S
G. A
W. I

TECHN

---

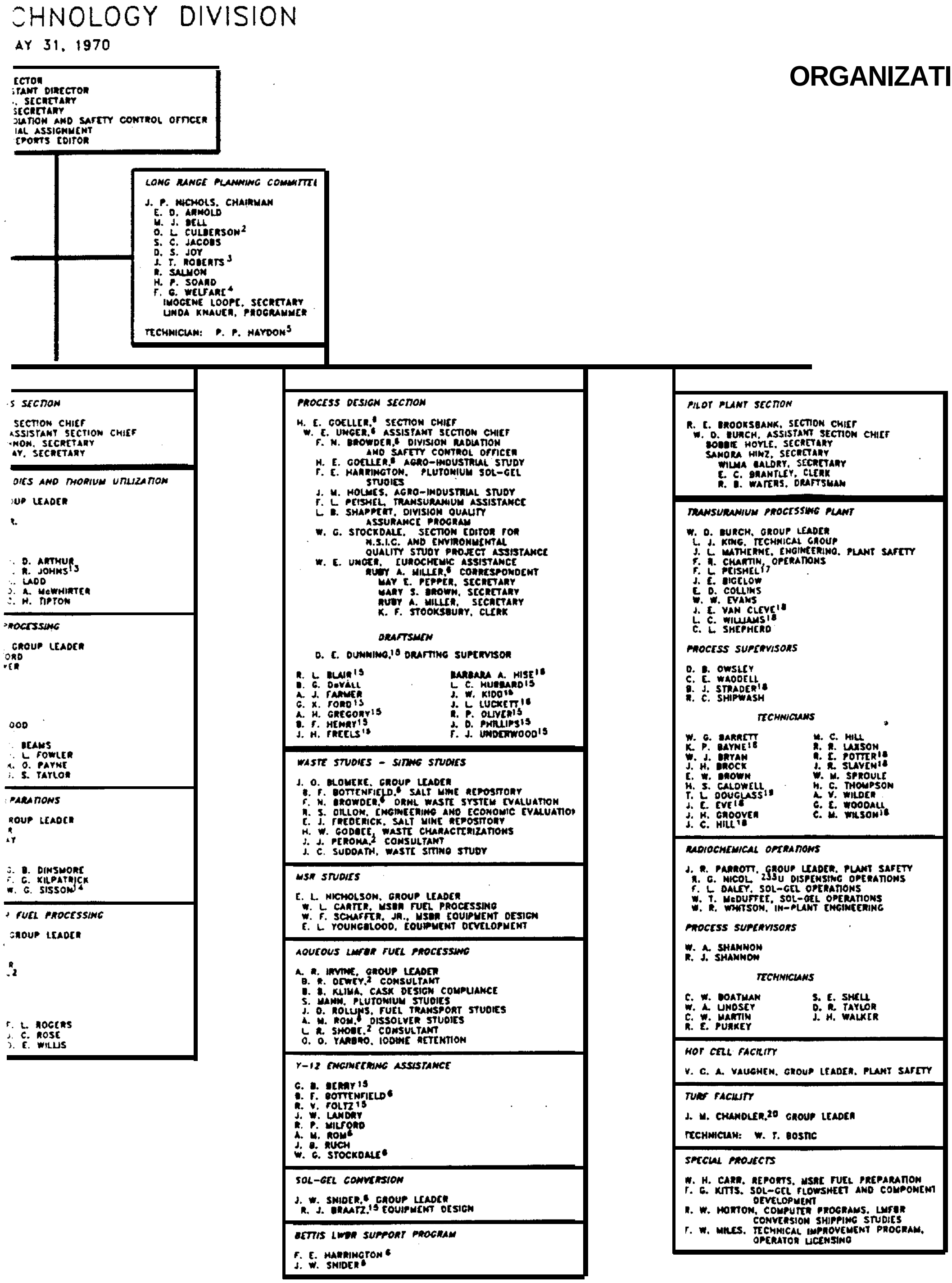
[1] ON LOA
[2] CONSUL
[3] ON LEA
[4] ON LOA
[5] ON LOA
[6] DUAL C
[7] ON LOA
[8] ASSIGN
[9] GUEST
[10] ON LOA
[11] PART-T
[12] GUEST
[13] ON LOA
[14] ON LOA
[15] ON LOA
[16] PART T
[17] ON LOA
[18] ON LOA
[19] ON EDU
[20] ON LOA

Source: Chemical Technology Division Annual Progress Report for Period Ending May 31, 1970, ORNL-4572.

ORNL DWG 92A-658

APPENDIX 8.3

ORGANIZATION CHART FOR MAY 31, 1970



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AY 31, 1970

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., SECRETARY
SECRETARY
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LADD
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ORD
ER

OOD

BEAMS
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O. PAYNE
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R
AY

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G. KILPATRICK
G. SISSON[14]

FUEL PROCESSING

GROUP LEADER

R
2

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E. J. FREDERICK, SALT MINE REPOSITORY
H. W. GODBEE, WASTE CHARACTERIZATIONS
J. J. PERONA,[2] CONSULTANT
J. C. SUDDATH, WASTE SITING STUDY

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E. L. YOUNGBLOOD, EQUIPMENT DEVELOPMENT

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S. MANN, PLUTONIUM STUDIES
J. O. ROLLINS, FUEL TRANSPORT STUDIES
A. M. ROM,[6] DISSOLVER STUDIES
L. R. SHOBE,[2] CONSULTANT
O. O. YARBRO, IODINE RETENTION

Y-12 ENGINEERING ASSISTANCE

G. B. BERRY[15]
B. F. BOTTENFIELD[6]
R. V. FOLTZ[15]
J. W. LANDRY
R. P. MILFORD
A. M. ROM[6]
J. B. RUCH
W. G. STOCKDALE[6]

SOL-GEL CONVERSION

J. W. SNIDER,[6] GROUP LEADER
R. J. BRAATZ,[15] EQUIPMENT DESIGN

BETTIS LWBR SUPPORT PROGRAM

F. E. HARRINGTON[6]
J. W. SNIDER[6]

PILOT PLANT SECTION

R. E. BROOKSBANK, SECTION CHIEF
W. D. BURCH, ASSISTANT SECTION CHIEF
BOBBIE HOYLE, SECRETARY
SANDRA HINZ, SECRETARY
WILMA BALDRY, SECRETARY
E. C. BRANTLEY, CLERK
R. B. WATERS, DRAFTSMAN

TRANSURANIUM PROCESSING PLANT

W. D. BURCH, GROUP LEADER
L. J. KING, TECHNICAL GROUP
J. L. MATHERNE, ENGINEERING, PLANT SAFETY
F. R. CHARTIN, OPERATIONS
F. L. PEISHEL[17]
J. E. BIGELOW
E. D. COLLINS
W. W. EVANS
J. E. VAN CLEVE[18]
L. C. WILLIAMS[18]
C. L. SHEPHERD

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C. E. WADDELL
B. J. STRADER[18]
R. C. SHIPWASH

TECHNICIANS

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K. P. BAYNE[18]	R. R. LAXSON
W. J. BRYAN	R. E. POTTER[18]
J. H. BROCK	J. R. SLAVEN[18]
E. W. BROWN	W. M. SPROULE
H. S. CALDWELL	H. C. THOMPSON
T. L. DOUGLASS[19]	A. V. WILDER
J. E. EVE[18]	G. E. WOODALL
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J. R. PARROTT, GROUP LEADER, PLANT SAFETY
R. G. NICOL, ^{235}U DISPENSING OPERATIONS
F. L. DALEY, SOL-GEL OPERATIONS
W. T. McDUFFEE, SOL-GEL OPERATIONS
W. R. WHITSON, IN-PLANT ENGINEERING

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V. C. A. VAUGHEN, GROUP LEADER, PLANT SAFETY

TURF FACILITY

J. M. CHANDLER,[20] GROUP LEADER

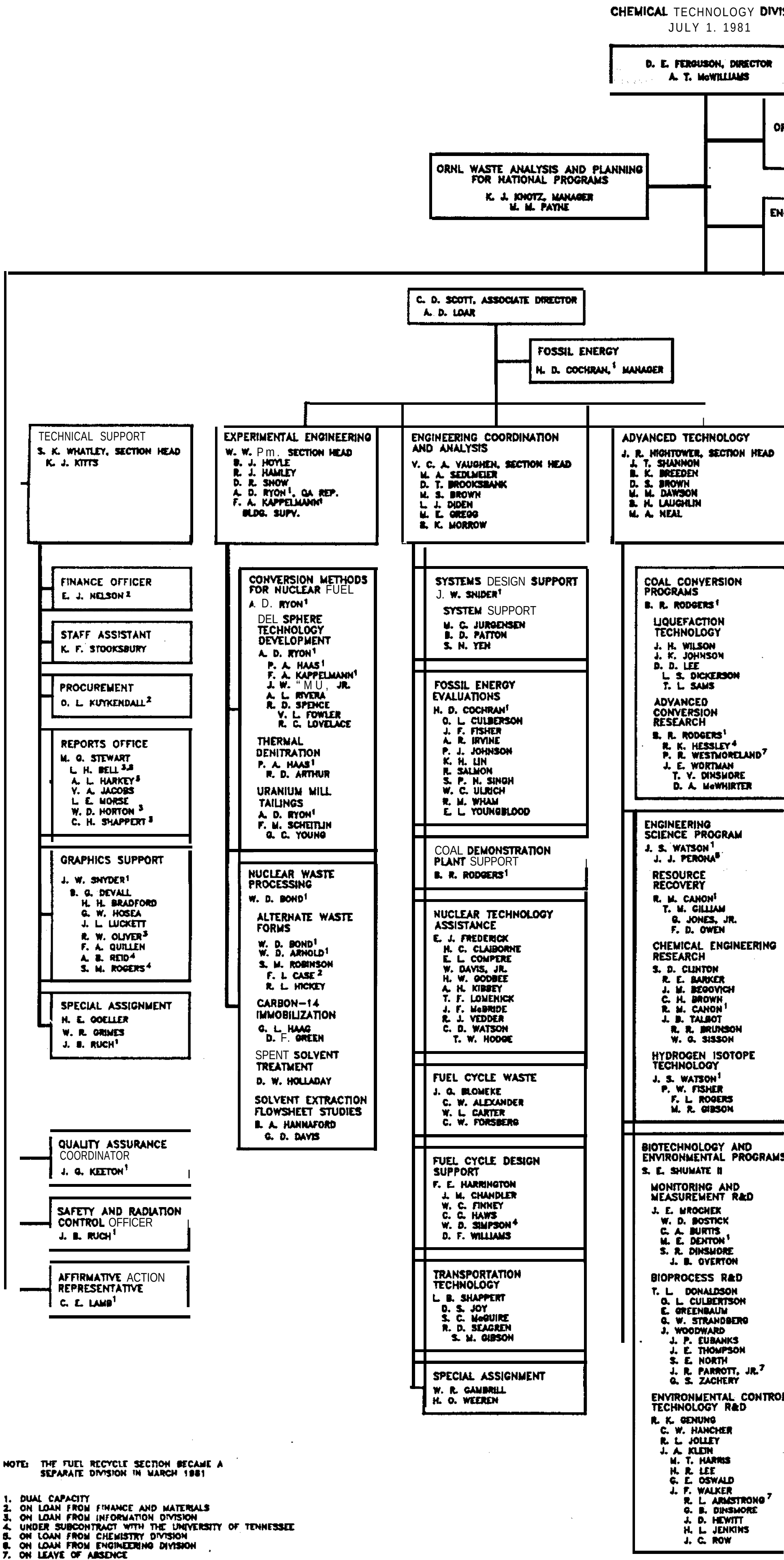
TECHNICIAN: W. T. BOSTIC

SPECIAL PROJECTS

W. H. CARR, REPORTS, MSRE FUEL PREPARATION
F. G. KITTS, SOL-GEL FLOWSHEET AND COMPONENT DEVELOPMENT
R. W. HORTON, COMPUTER PROGRAMS, LMFBR CONVERSION SHIPPING STUDIES
F. W. MILES, TECHNICAL IMPROVEMENT PROGRAM, OPERATOR LICENSING

ANT AND EQUIPMENT DIVISION
ERSITY OF TENNESSEE
ENCE
EACTOR DIVISION
2

EMICAL DEVELOPMENT SECTION B
PROBLEM BY THE ANALYTICAL CHEMISTRY DIVISION
ROM THE UNIVERSITY OF NEW YORK AT BUFFALO
-INDUSTRIAL SURVEY
EE
ROM SPAIN
EMICAL DEVELOPMENT SECTION A
MISTRY DIVISION
ENERAL ENGINEERING DIVISION
AN FROM NUCLEAR DATA PROJECT
OCESS DESIGN SECTION
TALS AND CERAMICS DIVISION
AVE
ALS AND CERAMICS DIVISION



CHEMICAL TECHNOLOGY DIVI
JULY 1. 1981

D. E. FERGUSON, DIRECTOR
A. T. McWILLIAMS

OF

EN

ORNL WASTE ANALYSIS AND PLANNING FOR NATIONAL PROGRAMS
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C. D. SCOTT, ASSOCIATE DIRECTOR
A. D. LOAR

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D. R. SNOW
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P. A. HAAS[1]
F. A. KAPPELMANN[1]
J. W. "MU, JR.
A. L. RIVERA
R. D. SPENCE
V. L. FOWLER
R. C. LOVELACE

THERMAL DENITRATION
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R. D. ARTHUR

URANIUM MILL TAILINGS
A. D. RYON[1]
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G. C. YOUNG

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W. D. BOND[1]

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W. D. BOND[1]
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R. L. HICKEY

CARBON–14 IMMOBILIZATION
G. L. HAAG
D. F. GREEN

SPENT SOLVENT TREATMENT
D. W. HOLLADAY

SOLVENT EXTRACTION FLOWSHEET STUDIES
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S. K. MORROW

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SYSTEM SUPPORT
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B. D. PATTON
S. N. YEH

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P. J. JOHNSON
K. H. LIN
R. SALMON
S. P. N. SINGH
W. C. ULRICH
R. M. WHAM
E. L. YOUNGBLOOD

COAL DEMONSTRATION PLANT SUPPORT
B. R. RODGERS[1]

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E. L. COMPERE
W. DAVIS, JR.
H. W. GODBEE
A. H. KIBBEY
T. F. LOMENICK
J. F. McBRIDE
R. J. VEDDER
C. D. WATSON
T. W. HODGE

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C. W. ALEXANDER
W. L. CARTER
C. W. FORSBERG

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F. E. HARRINGTON
J. M. CHANDLER
W. C. FINNEY
C. C. HAWS
W. D. SIMPSON[4]
D. F. WILLIAMS

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D. S. JOY
S. C. McGUIRE
R. D. SEAGREN
S. M. GIBSON

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H. O. WEEREN

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J. R. HIGHTOWER, SECTION HEAD
J. T. SHANNON
B. K. BREEDEN
D. S. BROWN
M. M. DAWSON
B. H. LAUGHLIN
M. A. NEAL

COAL CONVERSION PROGRAMS
B. R. RODGERS[1]

LIQUEFACTION TECHNOLOGY
J. H. WILSON
J. K. JOHNSON
D. D. LEE
L. S. DICKERSON
T. L. SAMS

ADVANCED CONVERSION RESEARCH
B. R. RODGERS[1]
R. K. HESSLEY[4]
P. R. WESTMORELAND[7]
J. E. WORTMAN
T. V. DINSMORE
D. A. McWHIRTER

ENGINEERING SCIENCE PROGRAM
J. S. WATSON[1]
J. J. PERONA[5]

RESOURCE RECOVERY
R. M. CANON[1]
T. M. GILLIAM
G. JONES, JR.
F. D. OWEN

CHEMICAL ENGINEERING RESEARCH
S. D. CLINTON
R. E. BARKER
J. M. BEGOVICH
C. H. BROWN
R. M. CANON[1]
J. B. TALBOT
R. R. BRUNSON
W. G. SISSON

HYDROGEN ISOTOPE TECHNOLOGY
J. S. WATSON[1]
P. W. FISHER
F. L. ROGERS
M. R. GIBSON

BIOTECHNOLOGY AND ENVIRONMENTAL PROGRAMS
S. E. SHUMATE II

MONITORING AND MEASUREMENT R&D
J. E. MROCHEK
W. D. BOSTICK
C. A. BURTIS
M. E. DENTON[1]
S. R. DINSMORE
J. B. OVERTON

BIOPROCESS R&D
T. L. DONALDSON
G. L. CULBERTSON
E. GREENBAUM
G. W. STRANDBERG
J. WOODWARD
J. P. EUBANKS
J. E. THOMPSON
S. E. NORTH
J. R. PARROTT, JR.[7]
G. S. ZACHERY

ENVIRONMENTAL CONTRO
TECHNOLOGY R&D
R. K. GENUNG
C. W. HANCHER
R. L. JOLLEY
J. A. KLEIN
M. T. HARRIS
H. R. LEE
G. E. OSWALD
J. F. WALKER
R. L. ARMSTRONG[7]
G. B. DINSMORE
J. D. HEWITT
H. L. JENKINS
J. C. ROW

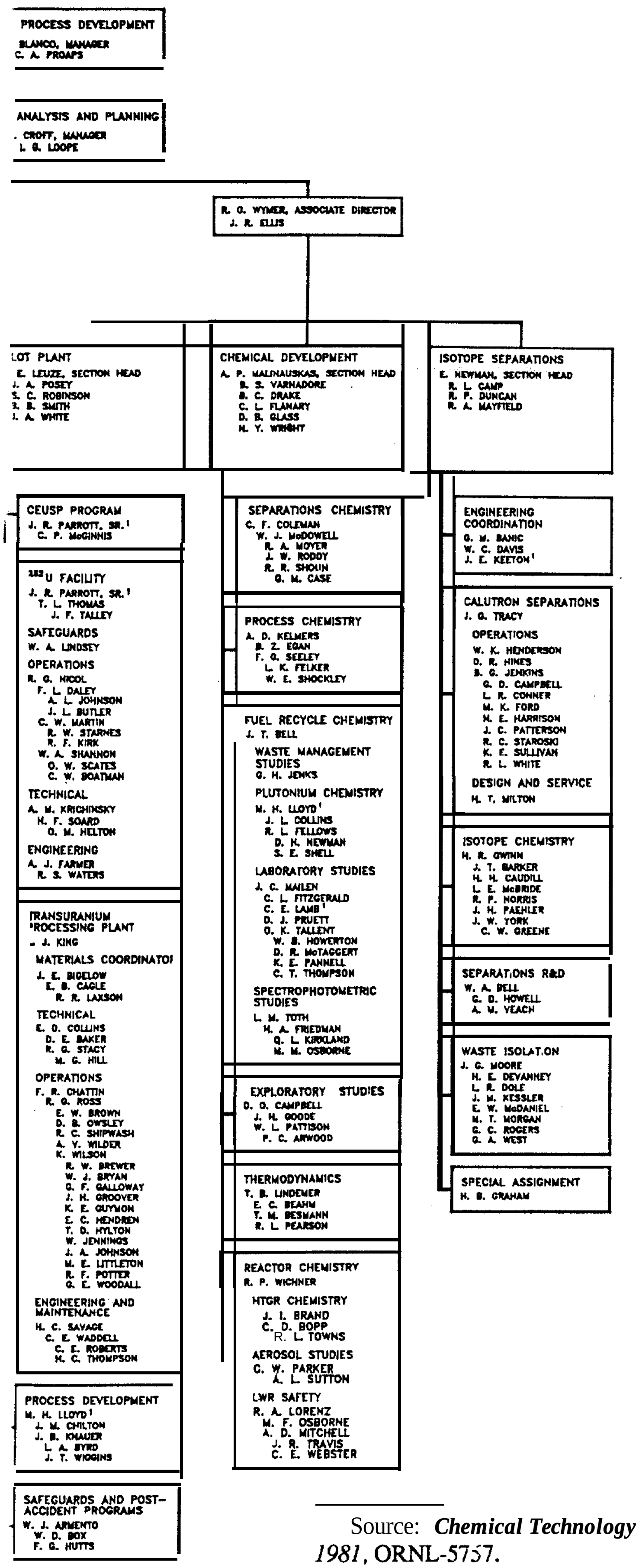
NOTE: THE FUEL RECYCLE SECTION BECAME A SEPARATE DIVISION IN MARCH 1981

1. DUAL CAPACITY
2. ON LOAN FROM FINANCE AND MATERIALS
3. ON LOAN FROM INFORMATION DIVISION
4. UNDER SUBCONTRACT WITH THE UNIVERSITY OF TENNESSEE
5. ON LOAN FROM CHEMISTRY DIVISION
6. ON LOAN FROM ENGINEERING DIVISION
7. ON LEAVE OF ABSENCE
8. PART–TIME EMPLOYEE

ORNL DWG 92A-657

APPENDIX 8.4

ORGANIZATION CHART FOR JULY 1, 1981



Source: ***Chemical Technology Division Progress Report for the Period April 1, 1979 to March 31, 1981***, ORNL-5757.

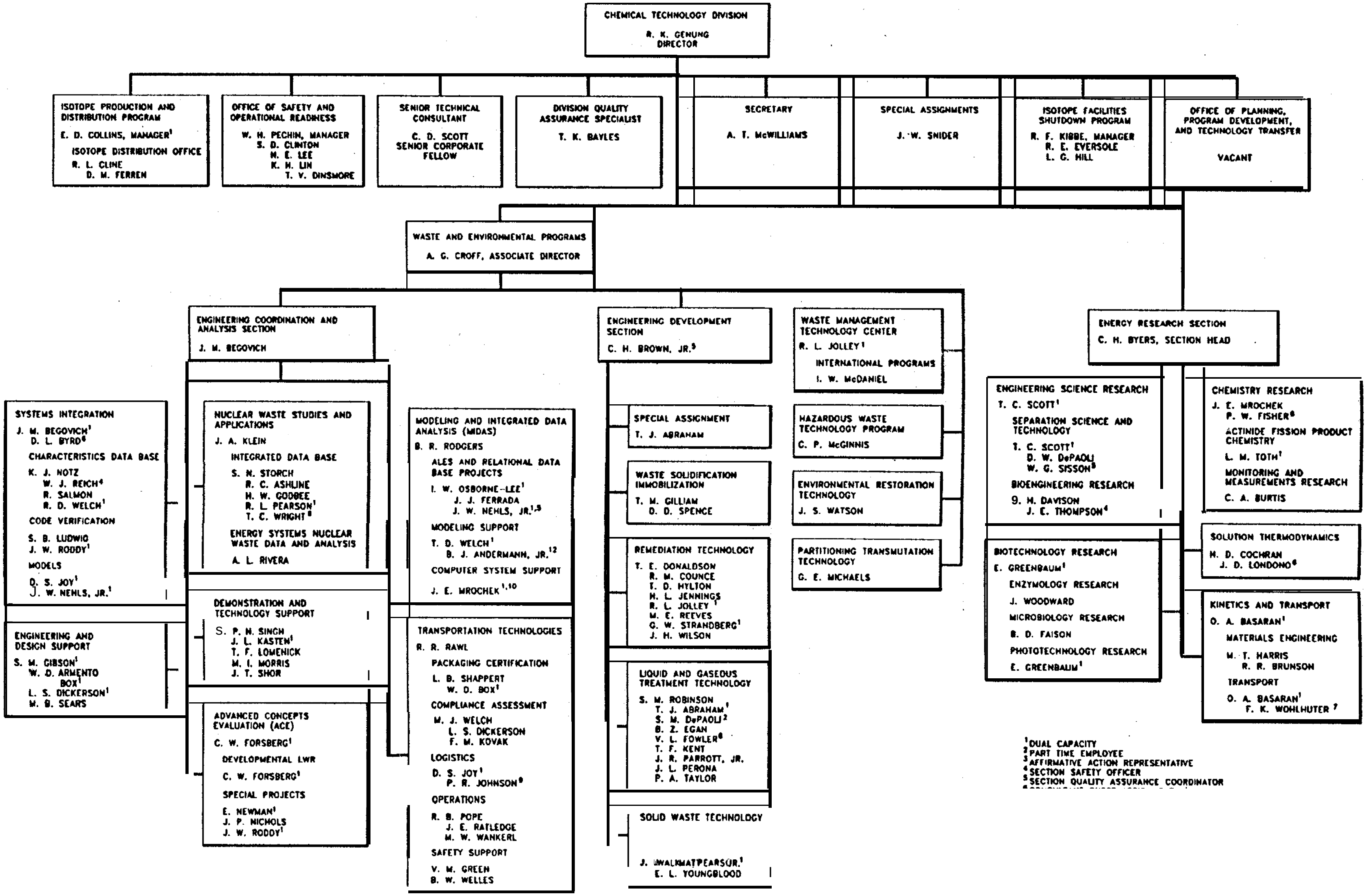
APPENDIX B.5

ORGANIZATION CHART FOR AUGUST 1991

PAGE 1 OF 2
EFFECTIVE DATE: AUGUST 1991

REPORTS TO ASSOCIATE DIRECTOR
ADVANCED ENERGY SYSTEMS

ORNL DWG 92A-659A

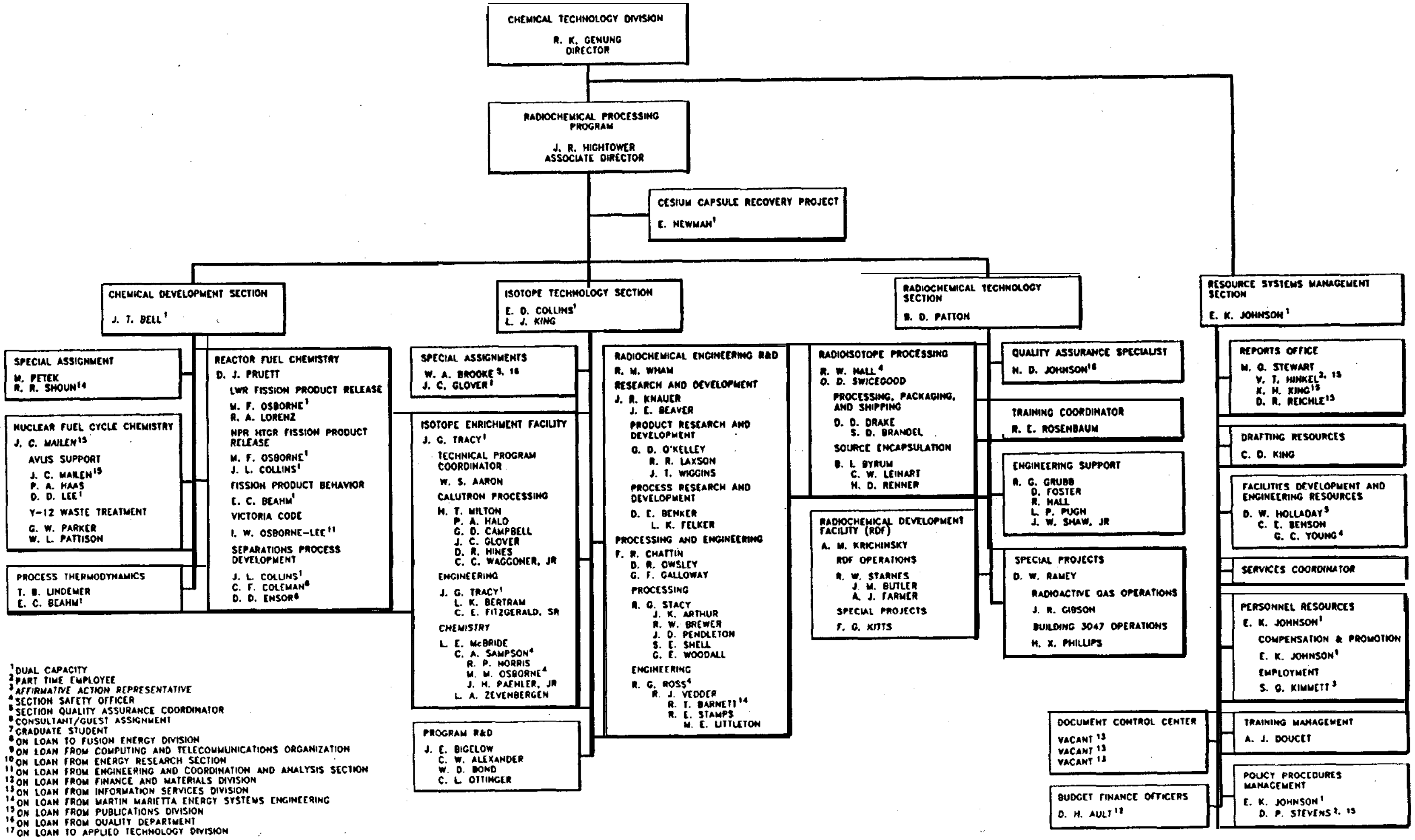


[1] DUAL CAPACITY
[2] PART TIME EMPLOYEE
[3] AFFIRMATIVE ACTION REPRESENTATIVE
[4] SECTION SAFETY OFFICER
[5] SECTION QUALITY ASSURANCE COORDINATOR

PAGE 2 OF 2
EFFECTIVE DATE: AUGUST 1991

ORNL DWG 92A-6599

REPORTS TO ASSOCIATE DIRECTOR
ADVANCED ENERGY SYSTEMS



CHEMICAL TECHNOLOGY DIVISION
R. K. GENUNG
DIRECTOR

RADIOCHEMICAL PROCESSING PROGRAM
J. R. HIGHTOWER
ASSOCIATE DIRECTOR

CESIUM CAPSULE RECOVERY PROJECT
E. NEWMAN[1]

CHEMICAL DEVELOPMENT SECTION
J. T. BELL[1]

SPECIAL ASSIGNMENT
M. PETEK
R. R. SHOUN[14]

NUCLEAR FUEL CYCLE CHEMISTRY
J. C. MAILEN[15]
- AVLIS SUPPORT
  - J. C. MAILEN[15]
  - P. A. HAAS
  - D. D. LEE[1]
- Y-12 WASTE TREATMENT
  - G. W. PARKER
  - W. L. PATTISON

PROCESS THERMODYNAMICS
T. B. LINDEMER
E. C. BEAHM[1]

REACTOR FUEL CHEMISTRY
D. J. PRUETT
- LWR FISSION PRODUCT RELEASE
  - M. F. OSBORNE[1]
  - R. A. LORENZ
- NPR HTGR FISSION PRODUCT RELEASE
  - M. F. OSBORNE[1]
  - J. L. COLLINS[1]
- FISSION PRODUCT BEHAVIOR
  - E. C. BEAHM[1]
- VICTORIA CODE
  - I. W. OSBORNE-LEE[11]
- SEPARATIONS PROCESS DEVELOPMENT
  - J. L. COLLINS[1]
  - C. F. COLEMAN[6]
  - D. D. ENSOR[6]

ISOTOPE TECHNOLOGY SECTION
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L. J. KING

SPECIAL ASSIGNMENTS
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J. C. GLOVER[1]

ISOTOPE ENRICHMENT FACILITY
J. G. TRACY[1]
- TECHNICAL PROGRAM COORDINATOR
  - W. S. AARON
- CALUTRON PROCESSING
  - H. T. MILTON
    - P. A. HALO
    - G. D. CAMPBELL
    - J. C. GLOVER
    - D. R. HINES
    - C. C. WAGGONER, JR
- ENGINEERING
  - J. G. TRACY[1]
    - L. K. BERTRAM
    - C. E. FITZGERALD, SR
- CHEMISTRY
  - L. E. McBRIDE
    - C. A. SAMPSON[4]
      - R. P. NORRIS
      - M. M. OSBORNE[4]
      - J. H. PAEHLER, JR
    - L. A. ZEVENBERGEN

PROGRAM R&D
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- C. W. ALEXANDER
- W. D. BOND
- C. L. OTTINGER

RADIOCHEMICAL ENGINEERING R&D
R. M. WHAM
- RESEARCH AND DEVELOPMENT
  - J. R. KNAUER
    - J. E. BEAVER
    - PRODUCT RESEARCH AND DEVELOPMENT
      - G. D. O'KELLEY
        - R. R. LAXSON
        - J. T. WIGGINS
    - PROCESS RESEARCH AND DEVELOPMENT
      - D. E. BENKER
        - L. K. FELKER
- PROCESSING AND ENGINEERING
  - F. R. CHATTIN
    - D. R. OWSLEY
    - G. F. GALLOWAY
    - PROCESSING
      - R. G. STACY
        - J. K. ARTHUR
        - R. W. BREWER
        - J. D. PENDLETON
        - S. E. SHELL
        - G. E. WOODALL
    - ENGINEERING
      - R. G. ROSS[4]
        - R. J. VEDDER
          - R. T. BARNETT[14]
          - R. E. STAMPS
            - M. E. LITTLETON

RADIOCHEMICAL TECHNOLOGY SECTION
B. D. PATTON

RADIOISOTOPE PROCESSING
R. W. HALL[4]
O. D. SWICEGOOD
- PROCESSING, PACKAGING, AND SHIPPING
  - D. D. DRAKE
    - S. D. BRANDEL
- SOURCE ENCAPSULATION
  - B. L. BYRUM
    - C. W. LEINART
    - H. D. RENNER

RADIOCHEMICAL DEVELOPMENT FACILITY (RDF)
A. M. KRICHINSKY
- RDF OPERATIONS
  - R. W. STARNES
    - J. M. BUTLER
    - A. J. FARMER
- SPECIAL PROJECTS
  - F. G. KITTS

QUALITY ASSURANCE SPECIALIST
N. D. JOHNSON[16]

TRAINING COORDINATOR
R. E. ROSENBAUM

ENGINEERING SUPPORT
R. G. GRUBB
- D. FOSTER
- R. HALL
- L. P. PUGH
- J. W. SHAW, JR

SPECIAL PROJECTS
D. W. RAMEY
- RADIOACTIVE GAS OPERATIONS
  - J. R. GIBSON
- BUILDING 3047 OPERATIONS
  - H. X. PHILLIPS

RESOURCE SYSTEMS MANAGEMENT SECTION
E. K. JOHNSON[1]

REPORTS OFFICE
M. G. STEWART
- V. T. HINKEL[2, 15]
- K. H. KING[15]
- D. R. REICHLE[15]

DRAFTING RESOURCES
C. D. KING

FACILITIES DEVELOPMENT AND ENGINEERING RESOURCES
D. W. HOLLADAY[5]
- C. E. BENSON
  - G. C. YOUNG[4]

SERVICES COORDINATOR

PERSONNEL RESOURCES
E. K. JOHNSON[1]
- COMPENSATION & PROMOTION
  - E. K. JOHNSON[1]
- EMPLOYMENT
  - S. G. KIMMETT[3]

TRAINING MANAGEMENT
A. J. DOUCET

POLICY PROCEDURES MANAGEMENT
E. K. JOHNSON[1]
- D. P. STEVENS[2, 15]

DOCUMENT CONTROL CENTER
VACANT[13]
VACANT[13]
VACANT[13]

BUDGET FINANCE OFFICERS
D. H. AULT[12]

[1] DUAL CAPACITY
[2] PART TIME EMPLOYEE
[3] *AFFIRMATIVE ACTION REPRESENTATIVE*
[4] SECTION SAFETY OFFICER
[5] SECTION QUALITY ASSURANCE COORDINATOR
[6] CONSULTANT/GUEST ASSIGNMENT
[7] GRADUATE STUDENT
[8] ON LOAN TO FUSION ENERGY DIVISION
[9] ON LOAN FROM COMPUTING AND TELECOMMUNICATIONS ORGANIZATION
[10] ON LOAN FROM ENERGY RESEARCH SECTION
[11] ON LOAN FROM ENGINEERING AND COORDINATION AND ANALYSIS SECTION
[12] ON LOAN FROM FINANCE AND MATERIALS DIVISION
[13] ON LOAN FROM INFORMATION SERVICES DIVISION
[14] ON LOAN FROM MARTIN MARIETTA ENERGY SYSTEMS ENGINEERING
[15] ON LOAN FROM PUBLICATIONS DIVISION
[16] ON LOAN FROM QUALITY DEPARTMENT
[17] ON LOAN TO APPLIED TECHNOLOGY DIVISION

APPENDIX C

C.l A COMPILATION OF CHEM TECH LEADERS

C.2 EVOLUTION OF CHEM TECH PROGRAMS

C.3 AN ENUMERATION OF CHEM TECH FACILITIES

APPENDIX C.l

A COMPILATION OF CHEM TECH LEADERS

Many of the technical accomplishments of Chem Tech during the past four decades have been achieved because of the effective use of collective groups or research teams. This fact is a tribute not only to the research teams and individual members, but also to the technical knowledge and good management skills of the Chem Tech leaders. This was specifically recognized by Dr. Herman Postma, Director of ORNL from 1974 to 1989, who stated "Chem Tech managed to develop some of the very best people that ORNL ever had in terms of what they did in science, engineering, and management . . . and they exported these people throughout the laboratory."[1] The following listing of Chem Tech leaders was developed from examination of technical progress reports, organization charts, and Chem Tech records.

1. Division Directors

F. L. Steahly	1950–1953
F. L Culler	1953-1965
D. E. Ferguson	1965-l-1983
R. G. Wymer	1983-1988
R. K. Genung	1988–present

2. Assistant/Associate Directors

F. R. Bruce	1954-1959
D. E. Ferguson	1963-1964
J. C. Bresee	1963-1964
K. B. Brown	1967-1976
R. G. Wymer	1973–1983
C. D. Scott	1976-1984
L. E. McNeese	1977-1978
A. G. Croff	1988–present
J. R. Hightower	1988–present

3. Long-Range Planning Committee

In 1953, soon after he became Director of Chem Tech, Floyd Culler initiated the Long-Range Planning Committee. The purpose of the committee was to determine and evaluate future needs in the atomic energy area and to help prepare or guide Chem Tech to assume major programmatic roles in fulfilling or resolving those needs. The committee functioned well in that capacity. It was discontinued in 1974.

R. J. Klotzbach (Chair 1953–1955)	1953-1955
E. D. Arnold	1953-1971
D. E. Ferguson	1953-1955
A. T. Gresky (Chair 19561965)	1953-1965
A. C. Jealous	1955-1956
C. E. Guthrie	1956–1967
J. W. Ullmann	1956-1967
W. B. Albrecht	1958–1960
J. P. Hammond	1958–1959
J. T. Roberts (Chair 1966-1968)	1961-1973
P. B. Camargo	1965
R. Salmon	1966-1973
0. L. Culberson	1967-1973
S. C. Jacobs	19681970
J. P. Nichols (Chair 1969-1973)	1969-1973

M. J. Bell	1969-1970
H. F. Soard	1969-1973
D. S. Joy	1970-1973
F. G. Welfare	1970-1971
V. A. DeCarlo	1971-1973
R. D. Cheverton	1972-1973
H. C. Claibome	1972-1973
W. Davis, Jr.	1972-1973
M. E. LaVerne	1972-1973
K. H. Lin	19721973

4. Pilot Plant Section Heads

The name of the Pilot Plant Section was changed to Process Development in 1987 and to the current name, Isotope Technology, in 1990.

D. G. Reid	1950-1951
H. K. Jackson	1951-1957
R. B. Lindauer	1957-1960
J. C. Bresee	1960-1963
R. E. Brooksbank	1963-1979
R. E. Leuze	1979-1987
E. D. Collins	1987–present

5. Process Design Section Heads

In 1973, the name of the Process Design Section was changed to Engineering Coordination and Analysis Section.

F. L. Culler	1950-1953
H. E. Goeller	1953-1973
J. P. Nichols	1973-1975
D. E. Ferguson	1975–1977 (Acting)
R. W. Glass	1977-1982
V. C. A. Vaughen	1982-1989
J. M. Begovich	1989–present

6. Laboratory Section Heads

In 1954, the name of the Laboratory Section was changed to Chemical Development Section. In 1955, the Chemical Development Section was split into two sections, Section A and Section B.

F. R. Bruce	1950-1955

6.1 Chemical Development A Section Heads

In 1973, Chemical Development Section A became Chemical Development.

D. E. Ferguson	1955-1963
R. G. Wymer	1963-1973
A. P. Malinauskas	1973-1983
J. R. Hightower	1983-1988
J. T. Bell	1988–present

6.2 Chemical Development B Section Heads

In 1974, the Chemical Development B Section was dissolved, with most personnel joining the Chemistry Division.

R. E. Blanco	1955-1972
C. D. Scott	1972-1973
L. M. Ferris	1973-1974

6.3 Chemical Development C Section Head

In 1948, the Raw Materials Section, under the direction of K. B. Brown, was established within the Y-12 Research Division. This division became the ORNL Materials Chemistry Division. In 1956, the Raw Materials Section became Chemical Development Section C of the Chemical Technology Division. The Section was dissolved in 1967, with personnel joining Sections A and B.

K. B. Brown	1956–1967

7. Unit Operations Section Heads

Originally titled the Semi-Works Section, the section name was soon changed to Unit Operations. The section name was changed again in 1973 to Experimental Engineering. In 1976, the section was split to form the Advanced Technology and Experimental Engineering Sections, and the former, in 1984, became the Energy Research Section.

J. 0. Davis	1950-1952
W. K. Eister	1952-1956
J. C. Bresee	1956–1960
M. E. Whatley	1960-1973

8. Experimental Engineering Section

C. D. Scott	1973-1976
R. E. Leuze	19761979
W. W.Pitt	1979-1989
C. H. Brown	1989–present

9. Advanced Technology Section

J.R Hightower	1976-1984

10. Energy Research Section

C. D. Scott	1984-1990
C. H. Byers	1990–present

11. Isotopes Separations Section

The Isotopes Separation Section was created in June of 1975 and existed until 1983, when it was functionally transferred to the Operations Division of ORNL. In 1988, the Operations Division was eliminated and the entire Isotope Program was transferred to Chem Tech and became the Isotope Section. In 1990, the section became known as Radiochemical Technology.

E. Newman	1975-1983
B. D. Patton	1988–present

12. Fuel Recycle Section

This section enjoyed only a brief history within the division, being formed in 1977 and then becoming a new ORNL division in 1981.

W. D. Burch	1977-1981

13. Technical Support Section

In 1981, a new section called Technical Support was created. It combined in one section all of the rather diverse support functions needed within a large and complex organization, which the division had become. The name of the section was changed to Resource Systems Management Section in 1988.

S. K. Whatley	1981-1984
E. K. Johnson	1984–present

14. Reference for Appendix C.1

1. Herman Postma, personal communication, August 30, 1991.

APPENDIX C.2

EVOLUTION OF CHEM TECH PROGRAMS

The programmatic efforts of this large and complex organization known as the Chemical Technology Division have, of necessity, been many and varied over its 42-year history. The following are program titles drawn from annual reports of the division at five-year intervals. They illustrate an evolving, changing, and vibrant organization.

1950

First Filtered Exhaust Gas Systems — precipitators, glass wool, asbestos
Enriched Uranium-Fission Process (Hexone)
Purex Process
RaLa Process (with Isotopes Division)
Redox Process
Thorex Process
Tri-Butyl Phosphate (TBP) Metal Recovery Process (U + Pu)
Dry Fluoride Process
OREX
First High-Level-Waste Disposal Evaporator (with Operations Division)
Fuel Dissolution Processes with Xe-Kr Recovery for ICPP
Installation of Stainless Steel and High-Level-Waste Tanks

1955

Training of Foreign Scientists (France, Sweden, Germany, India, Belgium, Japan)
Thorex Process
Recovery of Uranium from Wartime Wastes (Metal Recovery Project)
Homogeneous Reactor (HR) Studies
HOPE Project
Slurex Process for Weldon Springs
Metallex Process
Pilot Plant for Organic (EDTH)-Mercury Amalgam Process for ^{6}Li
Fluorox Process
Monex Process
Excer Process
Manhattan Process (Dissolution of Zr Fuel)
Initiation of TRU — Califomium Recovery Program
Establishment of Long-Range Planning Group for Power Fuels

1960

Head-End (Chop Leach) and Solvent Extraction Processing for Power Fuel
Power Reactor Fuel Processing Pilot Plant
Uranium Ore Recovery for U.S. Ores (with Materials Chemistry Division)
- Di-2-Ethylhexyl Phosphoric Acid Process
- Ion Exchange
- Amine Processes

Fused Salt-Fluoride Volatility Process
Production of Multi-Kg Quantities of ^{233}U (Thorex Pilot Plant)
Molten Salt Reactor (MSR) Fuel Processing
Homogeneous Reactor Program (HRP)

- HRP Fuel Processing
- HRP Thoria Blanket Development

Waste Treatment and Disposal

- Operation of Salt-Bed High-Level Waste Test with Health Physics

Fuel Cycle Development
Amex Process
Transuranium (TRU) Studies

- Construction of TRU Facility

Fission Product Recovery
Solvent Extraction Technology
Extraction Reagent Performance
Ion Exchange Technology
Chemical Applications of Nuclear Explosions
Processing of Gas-Cooled Reactor Fuel

1965

Equipment Development for Remote Processing of Power Fuels
Power Reactor Fuel Processing
Fluoride Volatility Processing
Waste Treatment and Disposal
TRU Element Processing and Recycle (with Metals & Ceramics Division)
Development of Thorium Fuel Cycle
Sol-Gel Process for the Thorium Fuel Cycle

- Graphite-Coated Spheres (with Metals & Ceramics Division)

Sol-Gel Process for the Uranium Refueling Cycle
Separations Chemistry Research
Recovery of Fission Products by Solvent Extraction
Mechanisms of Separations Processes
Chemical Applications of Nuclear Explosions
Preparation and Properties of A&tide-Element Oxides
MSR Processing
Biochemical Processes
Metallex Processes for Thorium and Uranium
Processing of Graphite Gas-Cooled Reactor Fuel
Production of Hydrocarbons from Metal Carbides
Studies of Radiolytic Hydrogen Production
Ion Exchange Process for Reduction of Radioisotopes from Low-Level Waste
(Plant in ORNL Tank Farm)

1970

Combustion and Leaching of Graphite-Uranium Rocket Fuel
MSR Processing by Fluoride Volatility Processing
Development of Aqueous Processes for Fast Reactor Fuels
Development of Methods for Reprocessing High-Temperature Gas-Cooled Reactor (HTGR) Fuels
Waste Treatment and Disposal
TRU Element Processing
Studies of Actinide Burning for High-Level Waste
Development of the Thorium Fuel Cycle
238Pu Studies for Prolifention Protection
Sol-Gel Processes for the Uranium Fuel Cycle
Conversion Studies in the Liquid-Metal Fast Breeder Reactor (LMFBR) Fuel Cycle

Preparation of $^{233}UO_2$ for Light-Water Breeder Reactors (LWBR)
Separations Chemistry Research
Chemical Applications of Nuclear Explosives
Biochemical Technology
- Zonal Gradient Centrifuge (with Biology Division)

Preparation and Properties of Actinide Oxides
Nuclear-Powered Agro-Industrial Complexes

1975

Development of Aqueous Processes for LMFBR Fuels
HTGR Fuel Recycle Development Program
$^{14}CO_2$ Removal
MSR Program
Waste Management
TRU Element Processing
Separations Chemistry Research
Biomedical Technology
- Centrifugal Fast Analyzer
- High-Resolution Chromatography

Environmental Studies
Biochemical Engineering
Coal Technology
Controlled Thermonuclear Program
Purification of Multigram Quantities of t-RNA (with Biology Division)
Nuclear Regulatory Assistance
Iodine Studies
Actinide Oxides, Nitrides, and Carbides
Resin Loading Process for Making Microspheres

1980

Consolidated Fuel Reprocessing Program (CFRP)
HTGR Studies
Nuclear Waste Management
Cement and Concrete Technology
Partitioning and Transmutation
FUSRAP Stabilization Technology Development
Fossil Energy-Coal Conversion Process Development
Continuous Chromatography-Annular Chromatograph
Sol-el Studies for Non-Nuclear Applications
Integrated Data Base (IDB) of Radiological Data
Three Mile Island Support

1985

CFRP, Fuel Recycle Section of CTD became ORNL Division in 198 1
Fast Flux Test Facility
Waste Immobilization Technologies
Support of New Hydrofracture Facility
Develop Pumpable Grouts for Hanford
Integrated Data Base (IDB) of Radiological Data
Characteristics Data Base (CDB) of Spent Fuel and High-Level Waste
Environmental Control Technology
Basic Energy Science Research
Multi-Phase Studies in External Fields
Solvent-Solute Interactions in Separations with Supercritical Solvents
Kinetics of Nucleation and Growth of Microcrystalline Systems
Biophotolysis and Enzyme Kinetics
Biotechnology Programs

- Advanced Bioreactor Systems and Microbial Immobilization
- Development of Bioprocessing Systems for Wastewater Treatment
- Development of Bioanalytical Techniques for Monitoring Physiologic Effects of Environmental Pollutants

TRU Processing
NRC Programs
Fission Product Release
Core Melt
Iodine
Consolidated Edison Uranium Solidification (CEUSP)
Miscellaneous Programs

- Fusion Energy
- Three-Mile Island Support
- FUSRAP
- Molten Salt Reactor Decommissioning
- Process Monitoring for Coal Conversion

1990

Radiochemical Processing Programs

- Fission Product Transport Studies
- Process Development for AVLIS Feed Material
- Chemistry of Radioactive Waste Isolation
- High-Temperature Thermodynamics
- Chemical Development for Waste Treatment
- Transuranium Elements for Research
- ^{252}Cf Neutron Sources
- Enrichment of Stable Isotopes
- Isotope Production and Distribution
- ^{85}Kr Loading and Shipping

Waste Management and Environmental Programs

- Waste Management Technology Center
- Waste R&D Program Management
- Environmental Restoration
- Guidance Manual for Technology Demonstrations

Engineering Coordination and Analysis Programs

- Developmental Light-Water Reactors

- Partitioning-Transmutation Concepts
- Modeling and Integrated Data Analysis
- Expert System for Liquid Low-Level Waste
- Aerosol Models for VICTORIA
- Decontamination of Concrete
- Integrated Data Base
- National Profile on Commercially Generated Mixed Waste
- Performance Assessment of Solid Waste Storage Area (SWSA) 6
- Office of Civilian Radiactive Waste Management (OCRWM) Transportation Operations
- Groundwater Treatment Demonstrations
- Control of Metal Emissions from Mixed-Waste Incinerators
- Removal of Toxic Metals from Contaminated Scrap

Engineering Development Programs

- Stack and Vent Survey
- Hazardous and Mixed-Waste Separation
- Sanitary Landfill Analysis
- Bioremediation of polychlorinated biphenyls (PCB)
- Bioremediation of trichloroethylene (TCE)-Contaminated Groundwater
- Destruction of PCBs in Mixed Wastes
- Bioremediation Demonstration on Kwajalein Island
- Soil Remediation Demonstration at Portsmouth Gaseous Diffusion Plant
- Evaluation of Catalytic Incineration
- Ultrahigh-Pressure Water Cleaning Demonstration
- Waste Stabilization and Treatability Studies—Fernald
- Process Waste-Water Treatment Studies
- In-Tank Evaporation
- Low-Level Liquid Waste Treatment
- Development of High-Temperature Membranes for Gas Separation
- Fedeml Facilities Agreement Program Management

Energy Research Programs

- Chemical Kinetics of Enzyme-Catalyzed Reactions
- Development of Bioelectronic Components and Biomaterials
- Fundamental Investigations of Superconductivity
- Multicomponent Separations by Continuous Chromatography
- Fundamental Studies of Homogeneous Nucleation and Particle Growth
- Chemical and Physical Principles in Multiphase Separations
- The Emulsion-Phase Contactor
- Chemistry of Actinides and Fission Products
- Bioprocessing for Energy Applications
- Advanced Bioreactor Concepts
- Bioconversion of Coal
- Isotope Separations from Dilute, Supercritical Solutions

APPENDIX C.3

AN ENUMERATION OF CHEM TECH FACILITIES

From the beginning, the work of Chem Tech and predecessor groups has taken place in many different areas and buildings. Table C.3.1 presents some interesting information concerning major buildings significantly involved in Chem Tech's work and mission. Aerial views (about 1960 and 1965) of Oak Ridge National Laboratory with Chem Tech facilities identified are shown in Figs. C.3.1 and C.3.2.

Table C.3.1. Selected Chemical Technology Division facilities completion and cost information

Year completed	Building No.[a]	Title	Original cost ($K)	Figure
1943	3019(205)	Separations Bldg.	681	c.3.3-C.3.5
1943	3550(706-A)	Chemistry Lab.	804	C.3.6, C.3.7
1948	3503(706-HD,-HB)	High-Radiation-Level Chem. Lab.	566	C.3.8
1951	3019	Addition to Sep. Bldg.	706	
1951	3505	Metal Recovery Plant	487	c.3.9
1951	3508	High Alpha Lab.	313	C.3.10
1951	4500N	Central Research Bldg.[b]	4966	C.3.11
1951	450115	High-Level Radiochemical Lab.[b]	3070	C.3.12
1952	3592	Unit Operation Volatility Lab.	34	
1958	4507	High-Radiation-Level Chemical Development Lab.	281	
1962	4500s	Central Research Bldg. Add.[b]	7495	
1965	7920	Transuranium Processing Plant	8000	c.3.13

[a] Where applicable, former building designations are presented within the parentheses.

[b] Shared with other ORNL divisions and tenants.

Principal Source: W. E. Thompson, ***History of the Oak Ridge National Laboratory 1943–1963***, **ORNL** Central File Number 63-8-75 (Aug. 23, 1963).

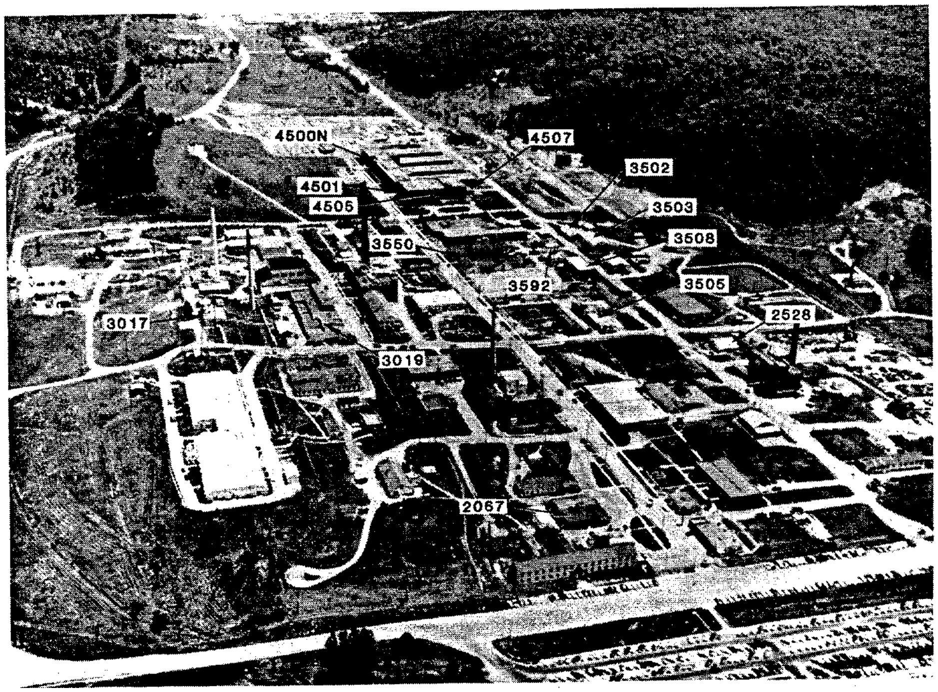

Fig. C.3.1. An aerial view (about 1960) of Oak Ridge National Laboratory, X-10 site, looking eastward. Chem tech facilities are identified.

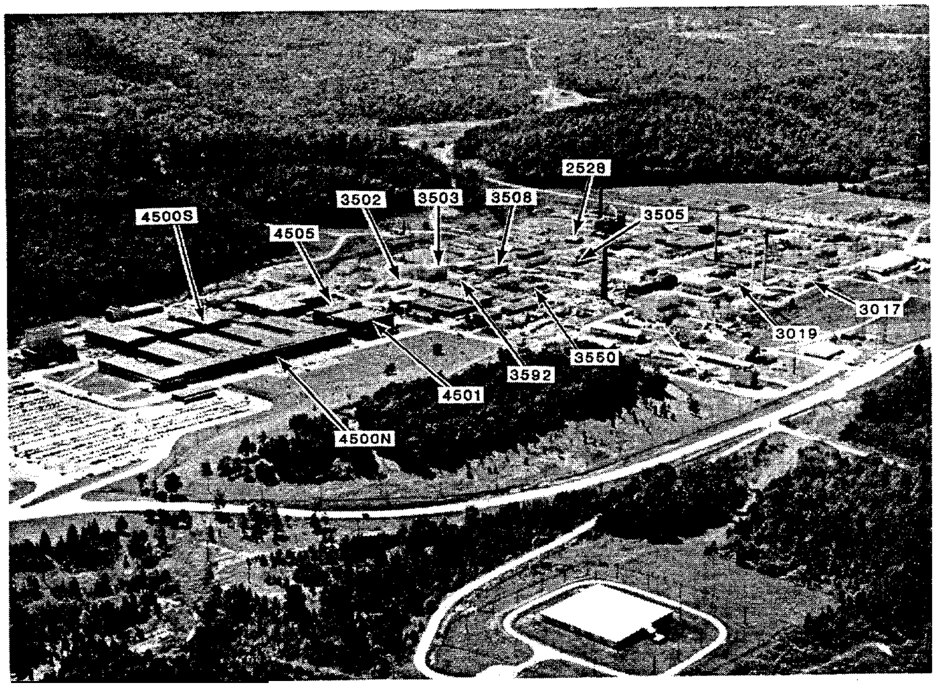

Fig. C.3.2. An aerial view (about 1965) of Oak Ridge National Laboratory, X-10 site, looking approximately south-southwest. Chem tech facilities are Identified.

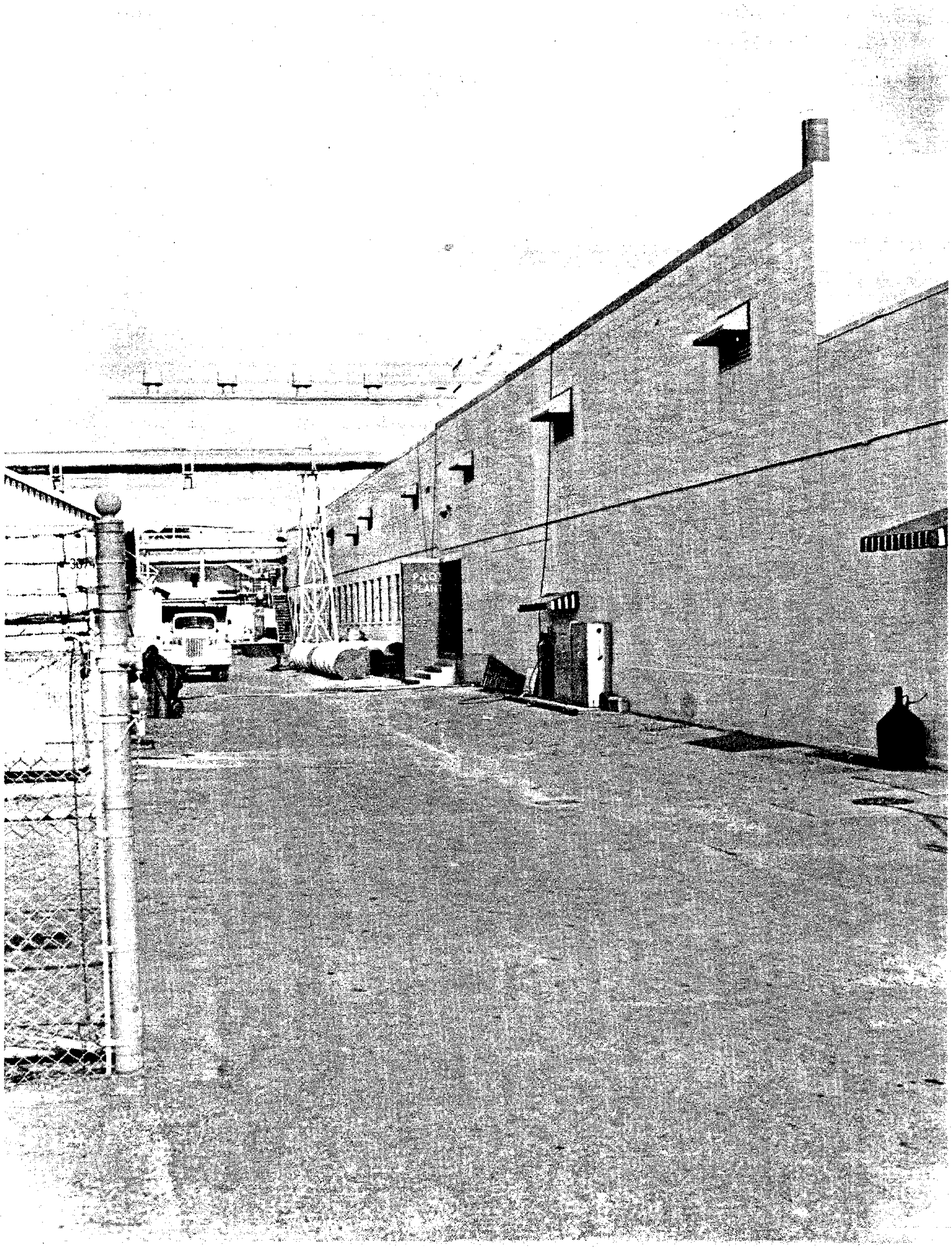

Fig. C.3.3. North side of Building 3019 showing Graphite Reactor in background. See also Figs. 1.1-1.3, 2.1, and 2.2.

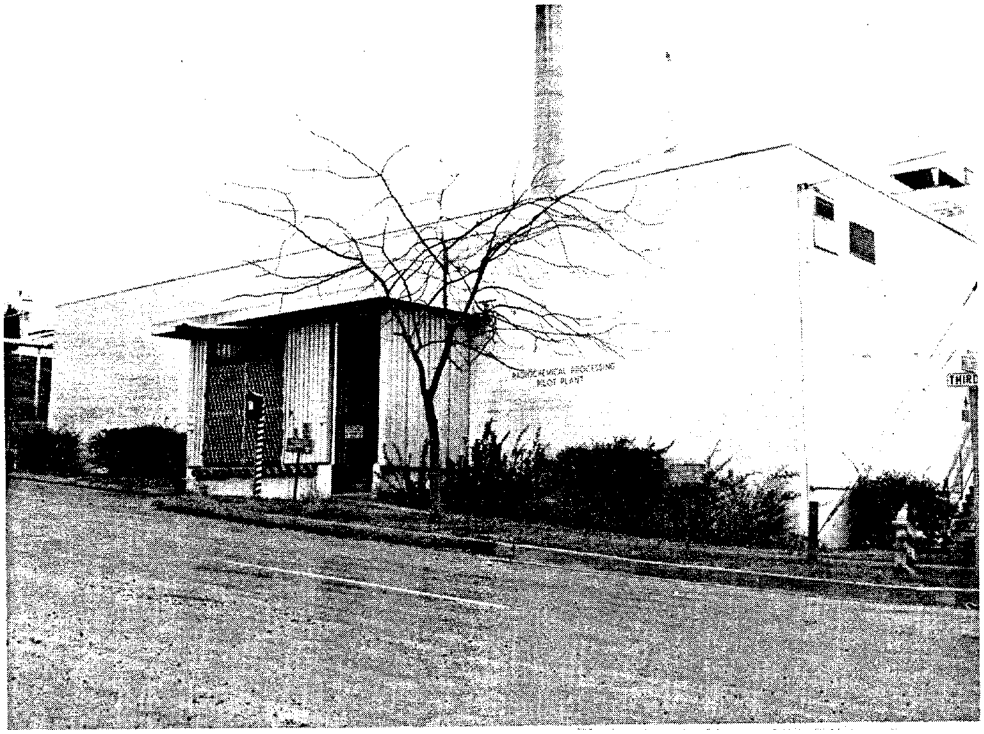

Fig. C.3.4. West end of Building 3019.

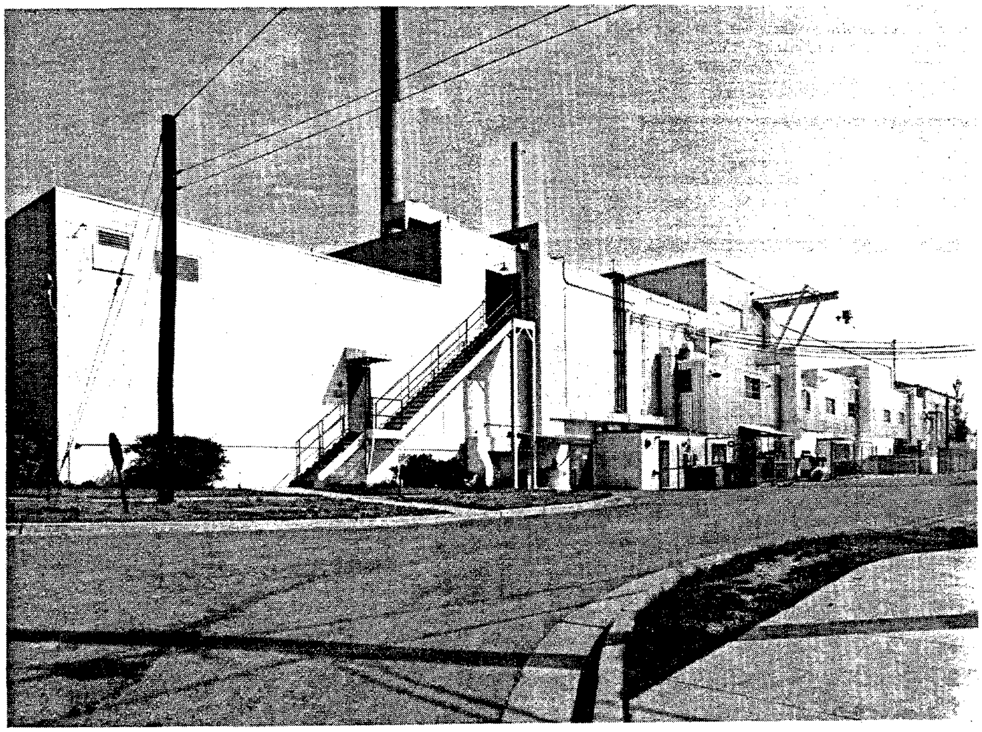

Fig. C.3.5. South side of Building 3019 showing the Graphite Reactor to the extreme right.

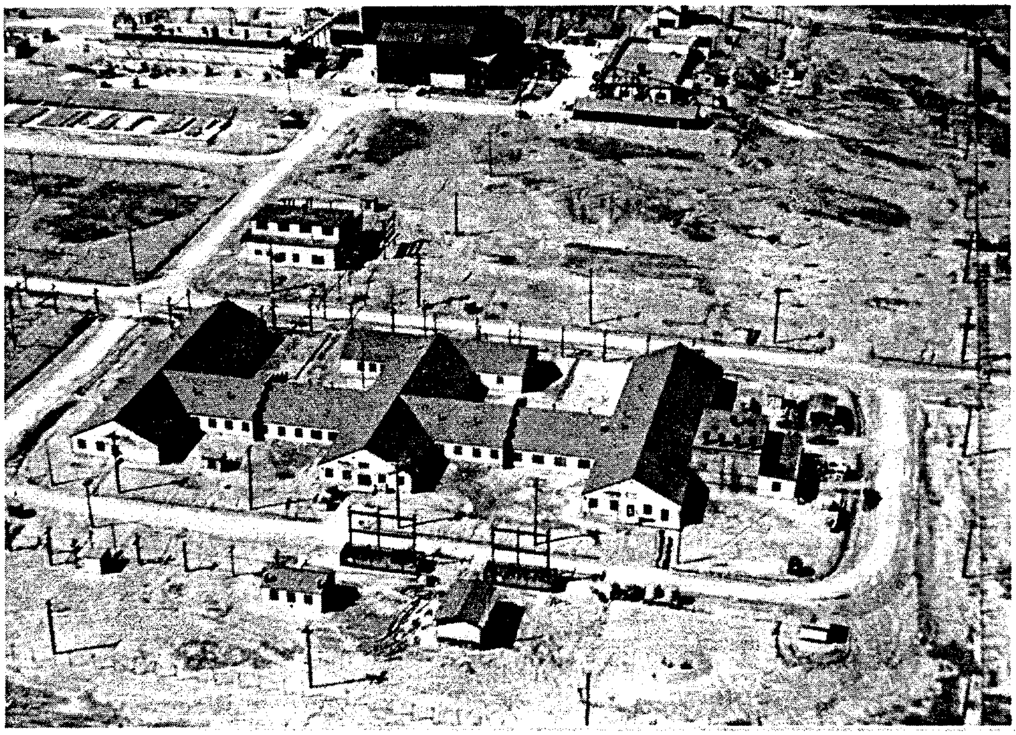

Fig. C.3.6. Aerial view of Main Laboratory, Building 706-A, looking northward at the Clinton Engineer Works In March 1944. The heavily shielded construction at the extreme right of the building was used for high-level radioactive experimentation. That portion was razed during the 1960s. Building 706-A Is now designated Building 3550.

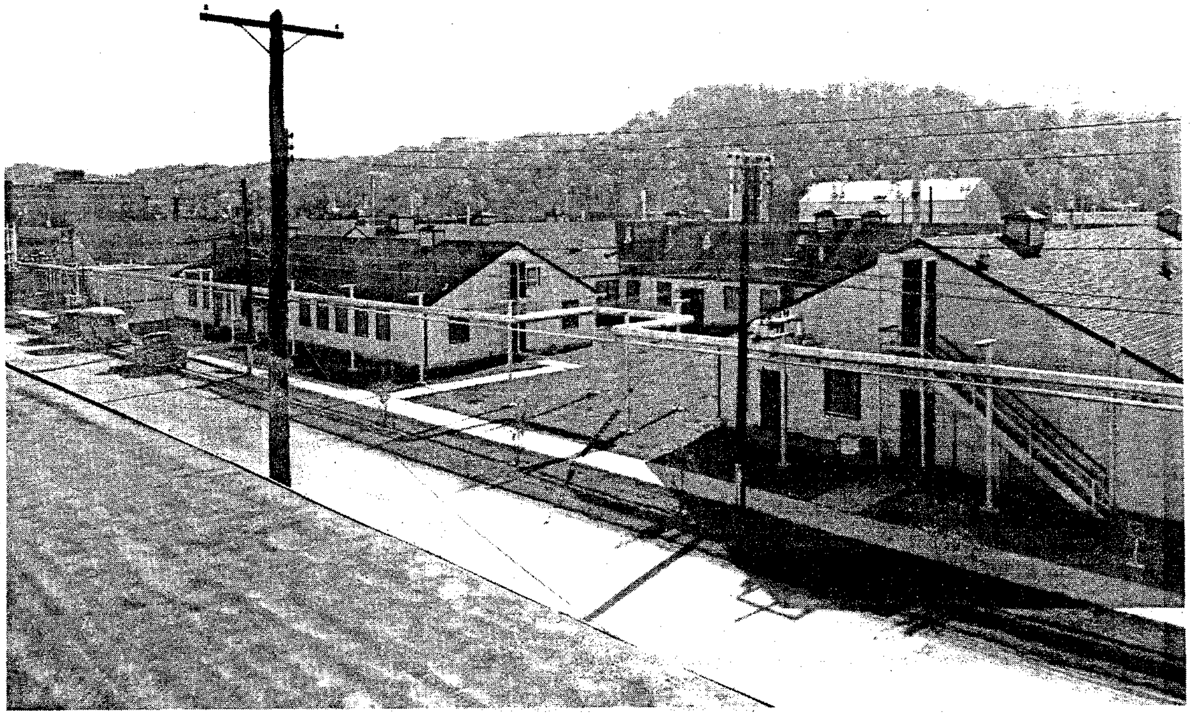

Fig. C.3.7. Building 3550 about 1965 looking southeast. Building 3503 is in the background.

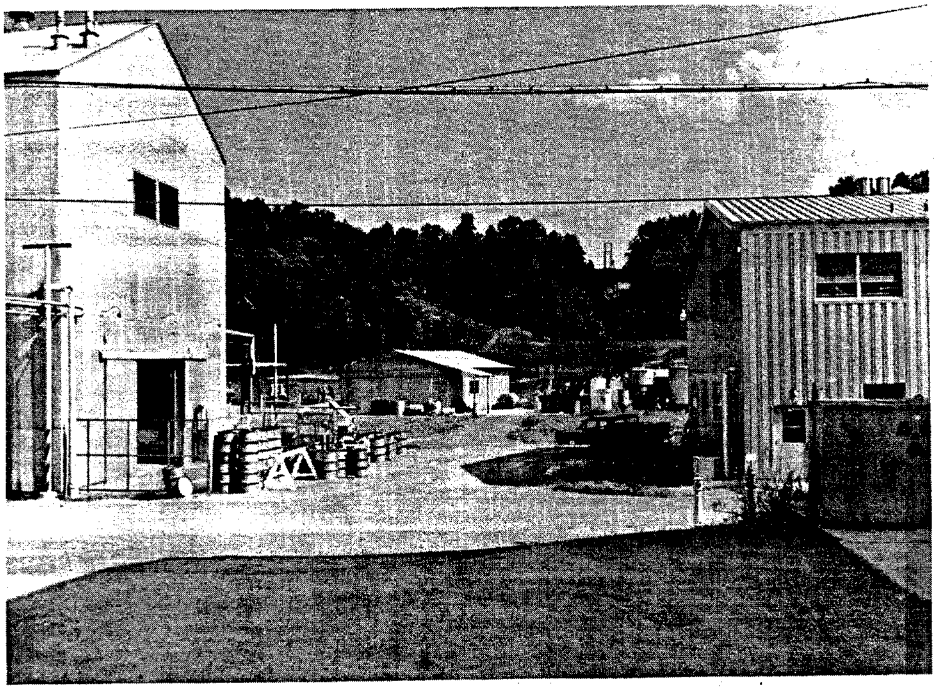

Fig. C.3.8. Part of Building 3503 is shown on the left in this photograph taken about 1980. A Health Physics facility, Building 3504, is on the right. See also Fig. C.3.7.

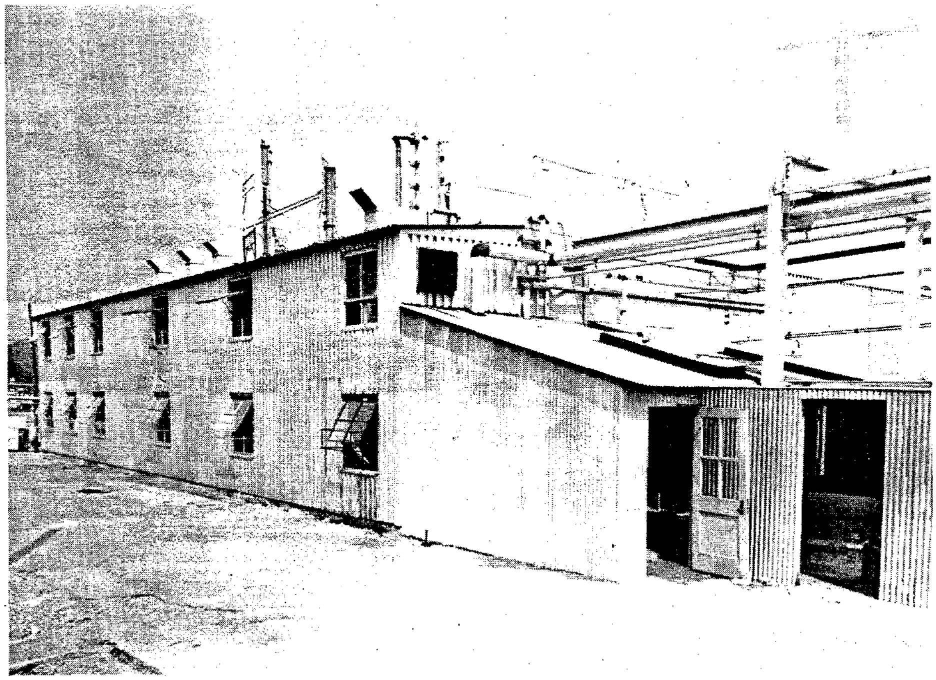

Fig. C.3.9. Building 3505, Metal Recovery Plant.

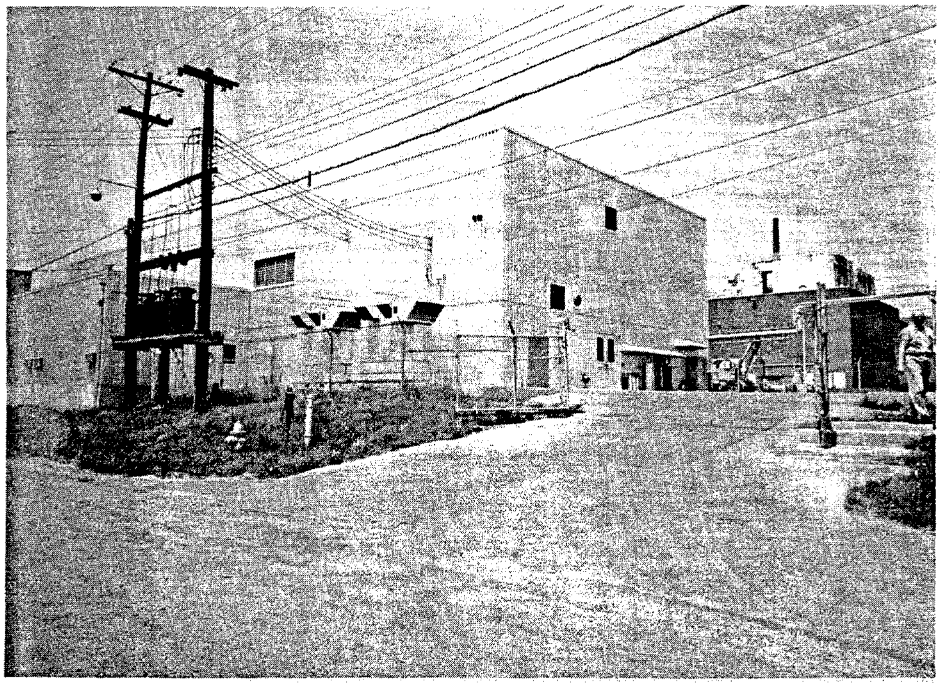

Fig. C.3.10. Building 3508, High Alpha Laboratory.

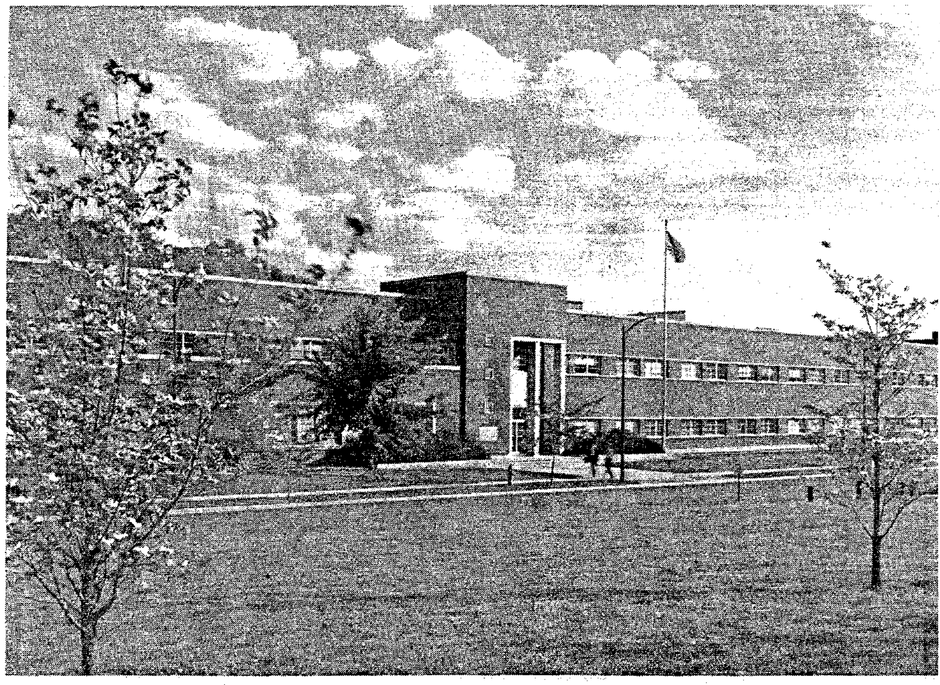

Fig. C.3.11. Building 4500N, Central Research Building.

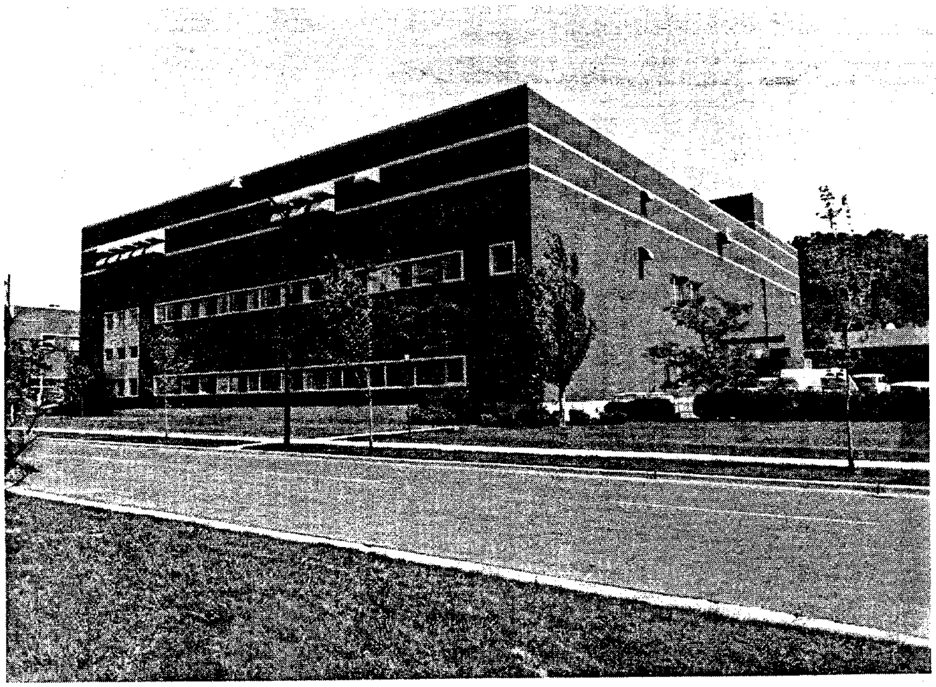

Fig. C.3.12. Building 4501/5, High-Level Radiochemical Laboratory.

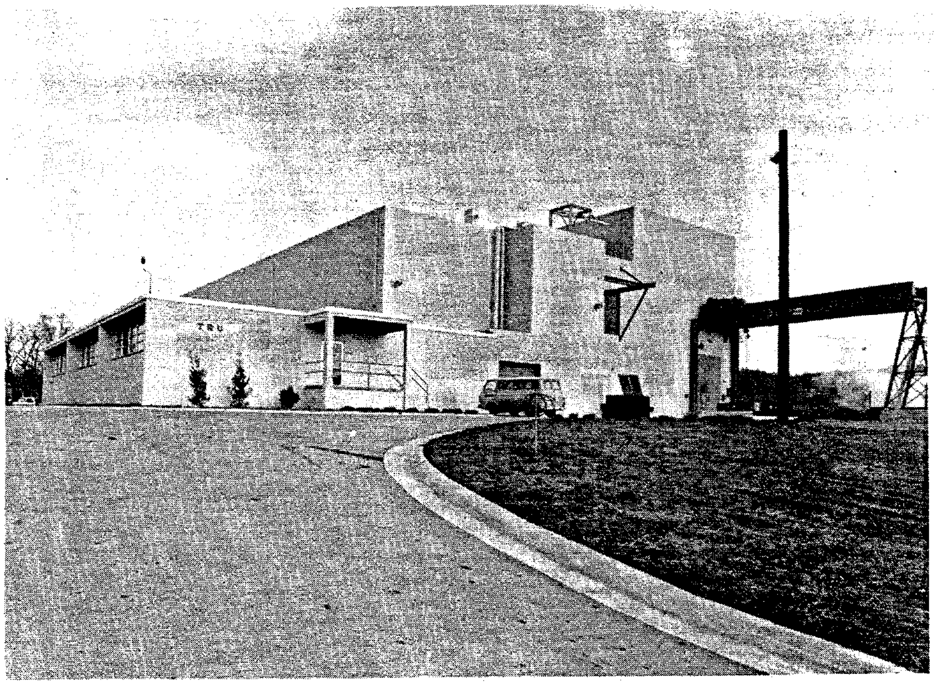

Fig. C.3.13. Building 7920, Transuranium Processing Plant.

APPENDIX D

HONORS AND AWARDS RECEIVED BY CHEMICAL TECHNOLOGY DIVISION PERSONNEL

LIST OF ACRONYMS FOR APPENDIX D

AAAS	American Association for the Advancement of Science,
AACC	American Association for Clinical Chemistry
AAES	American Association of Engineering Societies
AAWA	American Water Works Association
ACerS	American Ceramic Society
AChS	American Chemical Society
AIC	American Institute of Chemists
AIChE	American Institute of Chemical Engineers
ANS	American Nuclear Society
APS	American Physical Society
ASCET	American Society of Certified Engineering Technicians
ASES	American Solar Energy Society
ASME	American Society of Mechanical Engineers
CEUSP	Consolidated Edison Uranium Solidification Program
DOE	United States Department of Energy
ISES	International Solar Energy Society
NAE	National Academy of Engineering
NRC	National Research Council
NSF	National Science Foundation
PSI	Professional Secretaries International
STC*	Society for Technical Communication
STC/ETC	Society for Technical Communication, East Tennessee Chapter
SWE	Society of Women Engineers
TSPE	Tennessee Society of Professional Engineers
UCC	Union Carbide Corporation

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*Society for Technical Communication award ranking: first place, Distinction; second place, Excellence: third place, Merit: fourth place, Achievement.

Table D.l. Honors and Awards received by Chemnical Technology Division personnel

Name	Award	Sponsor
Beahm, Edward C.	Best Paper Award (2)	ANS, Materials Science and Technology Division
	Significant Implications for Energy Technology	DOE, Materials Science Research Competition
	Materials Science Research Competition Award	DOE
Begovich, John	IR-100 Award	Research and Development
	Publications Award	Energy Systems
Bell, Jimmy T.	Silver Acorn Award	Energy Systems
Berry, Jan B.	Waste Minimization Award	DOE
Besmann, Theodore M.	Materials Science Research Competition Award	DOE
	Best Paper Award for Nuclear Division	ACerS
	Membership Chairman, Program and National Public Information Committees	ACerS
Bigelow, John E.	Best Paper Award	ANS, Remote Systems Technology Division
	Community Service Award	UCC
Blanco, Raymond E.	Alumni Achievement Award	North Dakota State University
	Fellow	ANS
	Chairman	ANS, Nuclear Fuel Recycle Division
Blomeke, John	Outstanding Work in Chemical Technology in Fuel Recycle	ANS
Bond, Walter D.	Award of Excellence	STC/ETC
	IR- 100 Award	Industrial Research and Development
Bopp, C. Daniel	Fellow	AIC
Box, W. Donald	Silver Acorn Award	Energy Systems
Brooksbank, Donna T.	Award of Excellence (2)	STC/ETC
	Award of Achievement	STC, International Technical Publications Competition
Brooksbank, Robert E.	Professional Engineering License	TSPE
Brown, Deborah S.	Certified Professional Secretary	PSI
Brown, Janet D.	Certified Professional Secretary	PSI

Table D.l (continued)

Name	Award	Sponsor
Brown, Keith B.	Technical Achievement Award	Mining World
	Certificate of Merit	ANS
	Kirkpatrick Chemical Engineering Achievement Award	Chemical Engineering
	1984 Citation Classics Award (2)	Citation Index Physical, Chemical and Earth Sciences: Engineering,Technology and Applied Sciences
Brunson, Ron R.	Award 1990	STC/ETC
Burtis, Carl A.	President, Board of Directors, and National Lectureship Award	AACC
	Award of Distinction	STC/ETC
	Award of Distinction	STC, International Technical Publications Competition
	Silver Acorn Award	Energy Systems
	IR-100 Award	Industrial Research and Development
Campbell, David 0.	Advancements in Nuclear Technology (2)	ANS
	Silver Acorn Award	Energy Systems
	Glenn T. Seaborg Award for Actinide Separations	Advisory Board; Actinide Separations Conference
Case, Gerry N.	IR-100 Award .	Industrial Research and Development
Clinton, Sam D.	Silver Acorn Award	Energy Systems
Cochran, Henry D.	Silver Acorn Award	Energy Sysytems
Coleman. Charles F.	Kirkpatrick Chemical Engineering Achievement Award	Chemical Engineering
	Silver Acorn Award	Energy Systems
	1984 Citation Classics Award (2)	Citation Index Physical, Chemical and Earth Sciences: Engineering, Technology and Applied Sciences
Collins, Emory D.	Chairman	AIChE, Knoxville-Oak Ridge Chapter
Collins, J. L.	Literary Award for 1987 and 1988	Materials Science and Technology
	Silver Acorn Award	Energy Systems

Table D.l (continued)

Name	Award	Sponsor
Collins, J. L.	Awards of Distinction and Achievement	STC/ETC
	1989 Technical Achievement (Team)	Energy Systems
	1990 Special Achievement Award	Energy Systems
	199 1 President's Award for Performance Improvement	Energy Systems
Crouse, D. J.	IR-100´Award	Industrial Research and Development
	Kirkpatrick Chemical Engineering Achievement Award	Chemical Engineering
	1984 Citation Classics Award (2)	Citation Index Physical, Chemical and Earth Sciences: Engineering, Technology and Applied Sciences
Culberson, Oran L.	Fellow	AIChE
	Silver Acorn Award	Energy Systems
Davis, Wallace R.	Fellow	AAAS
Dawson, Martha M.	Certified Professional Secretary	PSI
Dinsmote, S. R.	Silver Acorn Award	Energy Systems
Dodson, Karen E.	Silver Acorn Award	Energy Systems
Egan, B. Z.	Awards of Distinction (2), Excellence, and Achievement	STC/ETC
Emmett, Mary J.	Certified Professional Secretary	PSI
Felker, Leslie K.	Award of Achievement	STC/ETC
Ferguson, Don E.	Fellow, Board of Directors, and Award for Outstanding Work in Chemical Technology in Fuel Recycle	ANS
	Award of Excellence	STC/ETC
	25th Anniversary Exceptional Service Award	ANS
	Engineer of Distinction	Tennessee Technological Univ.
Forsberg, Charles W.	Award of Achievement	STC, International Technical Publications Competition
	Award of Excellence (2)	STC/ETC
Frederick, Edward J.	Certificate of Appreciation	DOE
	Silver Acorn Award	Energy Systems

Table D.l (continued)

Name	Award	Sponsor
Friedman, Horace A.	Silver Acorn Award	Energy Systems
Gambili, Wallace R.	Editorial Excellence Award of Merit	Chemical Engineering
	Silver Acorn Award	Energy Systems
Genung, Richard K.	IR-100 Award	Industrial Research and Development
Gibson, S. M.	Newsletter Editor	AIChE, Nuclear Engineering Division
Godbee, Herschel W.	Certificate of Appreciation	DOE
	Silver Acorn Award	Energy Systems
	M. Sacid (Sarge) Ozker Award	ASME
	Chairman, Standards Working Group 16.6	ANS
Graves, D. A.	Award of Distinction	STC/ETC
Greenbaum, Elias	Chairman	ASES, Biotechnology and Chemical Sciences Division
	Delegate, U.N. Conference on New and Renewable Sources of Energy, Nairobi, Kenya	ISES
	Awards of Distinctjon, Excellence, Merit (2), and Achievement (2)	STC/ETC
	Award of Achievement	STC, International Technical Publications Competition
	Fellow	APS
	Secretary-Treasurer	APS, Division of Biological Physics
	Delegate. U.S.-Japan Seminar on Artificial Photosynthesis, Okazaki, Japan	NSF
	Grant. U.S.-Japan Program of Cooperation in Photoconversion and Photosynthesis	NSF
	Silver Acorn Award	Energy Systems
	Significant Event Award	Energy Systems
	Technical Achievement Award	Energy Systems
	Fellow	AAAS
	Chairman	APS, Division of Biological Physics

Table D.l (continued)

Name	Award	Sponsor
Greenbaum, Elias	Associate Editor, Biophysical Journal	Biophysical Society
	Co-Chairman, U.S.-Israel Bi-national Workshop on Photosynthesis and Energy Related Biological Science	DOE and Israel Ministry of Science and Technology
Grimes, Warren R.	Fellow, 25th Anniversary Exceptional Service Award and Outstanding Contributions in the Field of Reactor Chemistry	ANS
Haas, P. A.	1990 Award of Distinction	STC/ETC
	IR-100 Award	Industrial Research and Development
	Fellow	AIChE
	Golden Acorn Award	Energy Systems
	Inventor Award	Energy Systems
Harrington, Francis E.	IR-100 Award	Industrial Research and Development
	Silver Acorn Award	Energy Systems
Haws, Claude C.	IR-100 Award	Industrial Research and Development
Hightower, J. R.	Outstanding Advisory Committee Member	Pellissippi State Technical Community College
	Engineer of the Year	AIChE, Knoxville-Oak Ridge Chapter
Holladay, David W.	Silver Acorn Award	Energy Systems
Horton, Robert W.	Professional Engineering License	TSPE
Hurst, F. J.	IR-100 Award	Industrial Research and Development
	Kirkpatrick Chemical Engineering Achievement Award	Chemical Engineering
Irvine, Alvin R	Silver Acorn Award	Energy Systems
Jolley, Robert L.	Distinguished Alumni Award	Friends University, Kansas
	Chairman, Vice-Chairman, Secretary, and Program Chairman	AChS, Division of Environmental Chemistry
	1986 Distinguished Service Award	AChS, Division of Environmental Chemistry
	Councilor	AChS

Table D.l (continued)

Name	Award	Sponsor
Jolley, Robert L.	Member	AChS, Committees on Environmental Improvement and Meetings and Expositions
	Community Service Award	u c c
	Silver Acorn Award	Energy Systems
	Awards of Distinction (2), Achievement, and Merit	STC/ETC
	Award of Distinction, Books	STC, International Technical Publications Competition
	Member	AAWA, Health Effects Research Committee
Joy, Dave S.	Silver Acorn Award	Energy Systems
Kappelmann, Frederick A.	IR-100 Award	Industrial Research and Development
	Silver Acorn Award	Energy Systems
Kelmers, A. Donald	Award of Achievement	STC/ETC
	Silver Acorn Award	Energy Systems
King, Lester J.	Professional Engineering License	TSPE
King, Vaneeta M.	Certified Professional Secretary	PSI
Knight, John R.	Best Paper Award	ANS, Nuclear Criticality Safety Division
Lee, Douglas D.	IR-100 Award	Industrial Research and Development
Lee, Norman E.	Silver Acorn Award	Energy Systems
	Awards of Distinction (2), Merit, and Achievement	STC/ETC
	Award of Distinction	STC, International Technical Publications Competition
	Award for Technical Support	Energy Systems
	Fellow	AIC
Leuze, Rex E.	Fellow	AIC
	IR- 100 Award	Industrial Research and Development
	Silver Acorn Award	Energy Systems
Lewis, Barbara A.	Margaret Oakley Dayhoff Memorial Award	Biophysical Society
Lewis. Susan E.	Publications Award	Energy Systems

Table D.l (continued)

Name	Award	Sponsor
Light, Brenda B.	Certified Professional Secretary	PSI
Lindemer, Terrence B.	Fellow	ACerS
	Chairman, Vice-Chairman, Trustee, and Best Paper Award	ACerS, Nuclear Division
	Publications Award	Energy Systems
	Silver Acorn Award	Energy Systems
Lloyd, Milton HI	R-100 Award	Industrial Research and Development
	Silver Acorn Award	Energy Systems
Lorenz, R A.	Technical Achievement (Cont.).	Energy Systems
	1989 Technical Achievement (Team)	Energy Systems
	Assistance to President's Commission (TMI-2)	DOE
	Award of Distinction	STC/ETC
	Literary Award for 1987 and 1988	ANS, Material Science Division
Lowrie, Robert S.	Silver Acorn Award	Energy Systems
Mailen, J. C.	Fellow	AIChE
	Golden Acorn Award	Energy Systems
Malinauskas, Anthony P.	Special Award, Advancements in Nuclear Technology	ANS
	Ernest 0. Lawrence Memorial Award	DOE
McBride, J. P.	Award of Excellence (2) Award of Excellence	STC/ETC STC, International Technical Publications Competition
McDaniel, Earl W.	Fist Prize. Optical Microscopy, Unique Techniques	ACerS, Ceramographic Exhibit
	Awards of Excellence (2) and Achievement	STC/ETC
McDowell, William J.	IR-100 Award	Industrial Research and Development
McGinnis, C. Phil	Certified Professional Engineer	TSPE
	Editor	AIChE, Nuclear Engineering Division
	Chairman and Engineer of the Year	AIChE, Knoxville-Oak Ridge Chapter
	Award of Achievement, CEUSP Operation	Energy Systems

Table D.l (continued)

Name	Award	Sponsor
McGinnis, C. Phil	Operational Performance Award	Energy Systems
	Awards of Distinction and Merit	STC/ETC
McTaggart, Donald R.	Silver Acorn Award	Energy Systems
Moore, John G.	1984 Citation Classics Award (2)	Citation Index Physical, Chemical, and Earth Sciences; Engineering, Technology and Applied Sciences
Mrochek, John E.	IR-100 Award (2)	Industrial Research and Development
	Silver Acorn Award	Energy Systems
	Inventor Award	Energy Systems
Notz, Karl J.	IR-100 Award	Industrial Research and Development
	Silver Acorn Award	Energy Systems
Osborne, M. F.	Literary Award for 1987 and 1988	Materials Science & Technology
	1989 Technical Achievement (Team)	Energy Systems
	Awards for Distinction and Achievement	STC/ETC
	President's Award, Performance Improvement Project	Energy Systems
Osborne-Lee, Irvin W.	Executive Board Member	National Organization of Black Chemists and Chemical Engineers
	Chairman	AIChE, Minority Affairs Committee
	Chairman	AIChE, Committee for New Technology and Development
Owen, Sue C.	Certified Professional Secretary	PSI
Parrott, John R.	Professional Engineering License	TSPE
Pattison, William L.	Silver Acorn Award	Energy Systems
Pearson, Ray L.	Publications Award	Energy Systems
Peishel, Frank L.	Professional Engineering License	TSPE
Perona, Joseph J.	Chairman	AIChE, Energy Transport Research Committee
	Fellow	AIChE

Table D.1 (continued)

Name	Award	Sponsor
Pitt, W. W., Jr.	IR-100 Award (2)	Industrial Research and Development
	Silver Acorn Award	Energy Systems
	Fellow	AIChE
	Professional Engineering License	TSPE
Pruett, David J	Chairman	ACS, Nuclear Chemistry and Technology Divisioh
	Silver Acorn Award	Energy Systems
Reeves, M. E.	Award of Distinction	STC, 1990 Technical Publications Competition
Robinson, S. M.	Young Engineer of the Year	TSPE
	Director of Executive Committee	Tau Beta Pi, Great Smoky Mountains Alumni Chapter
	Outstanding Young Woman of America for 1988	Outstanding Young Women of America
Roddy, J. William	Award of Excellence (2)	STC/ETC
Rodgers, B. R.	Chainnan, Director, Program Chairman	AIChE, Fuels and Petrochemicals Division
	Director	AIChE
	Silver Acorn Award	Energy Systems
Ross, Robert G.	Silver Acorn Award	Energy Systems
Ryon, Allen D.	Kirkpatrick Chemical Engineering Achievement Award	Chemical Engineering
	IR- 100 Award	Industrial Research and Development
Salmon, Royes	Silver Acorn Award	Energy Systems
Scheitlin, F. M.	Award of Distinction	STC, 1990 International Technical Publications Competition
	Silver Acorn Award	Energy Systems
Scott, C. D.	Award of Distinction	STC, 1990 International Technical Publications Competition
	IR- 100 Award (4)	Industrial Research and Development
	Outstanding Engineering Alumnus Award	The University of Tennessee

Table D.l (continued)

Name	**Award**	**Sponsor**
Scott, C. D.	Senior Corporate Fellow	Energy Systems
	Engineer of the Year	AIChE, Knoxville-Oak Ridge Chapter
	Fellow	AIChE
	Ernest 0. Lawrence Memorial Award	DOE
	Outstanding Contribution Award	AACC
	National Award for Advanced Analytical Concepts	AACC
	Award of Excellence	STC/ETC
	Corporate Fellow	u c c
	Silver Acorn Award	Energy Systems
	Member of Advisory Committee and Committee of Fundamental Research for Emerging'and Critical Engineering Systems	NSF
	Golden Acorn Award	Energy Systems
	Member	NAE
	Inventor of the Year	Energy Systems
	Publications Award	Energy Systems
	Director	AIChE
	Fellow	AAAS
	Missouri Honor Award for Distinguished Service in Engineering	University of Missouri
Scott, T. C.	Award of Achievement	STC/ETC
	Award of Distinction	STC, 1990 International Technical Publications Competition
Seeley, F. G.	Silver Acorn Award	Energy Systems
Shell, S. E	Silver Acorn Award	Energy Systems
Shepherd, Deborah E.	Certified Professional Secretary	PSI
Shockley, William E.	Technician of the Year	ASCET
	Silver Acorn Award	Energy Systems
Shor, Joel T.	Silver Acorn Award	Energy Systems
Singh, S. P. N.	Silver Acorn Award	Energy Systems

Table D.l (continued)

Name	Award	Sponsor
Sisson, Warren G.	IR-100 Award	Industrial Research and Development
Spence, Roger D.	IR-100 Award (2)	Industrial Research and Development
	Award of Achievement	STC/ETC
Stewart, Martha G.	Award of Distinction	STC/ETC
	Award of Excellence (2)	STC/ETC
	Award of Merit (2)	STC/ETC
	Award of Achievement (2)	STC/ETC
	Award of Achievement (2)	STC, International Technical Publications Competition
Strandberg, Gerald W.	Publications Award	Energy Systems
Tallent, 0. K.	Silver Acorn Award	Energy Systems
Toth, L. M	Silver Acorn Award	Energy Systems
Villiers-Fisher, John F.	Golden Acorn Award	Energy Systems
Watson, Jack S.	Member	AIChE, National Research Committee
	Silver Acorn Award	Energy Systems
Weaver, Deborah J.	Certified Professional Secretary	PSI
Weeren. Herman 0.	First Prize. Optical Microscopy, Unique Techniques	ACerS, Ceramographic Exhibit
	Award of Excellence	STC/ETC
Wham, R. M.	Secretary, Treasurer, Director	AIChE, Knoxville-Oak Ridge Chapter
Whatley, Susan K.	President, Vice-President, and Distinguished New Engineers Award	SWE
	Women of Achievement Award	The University of Tennessee
	Member	AAES, Management Committee, Engineering Affairs Council
	Governor	AAES
Woodward, C. A.	Award of Distinction	STC, 1990 International Technical Publications Competition
Woodward, Jonathan	Librarian	AChS, Division of Microbial and Biochemical Technology
	Award of Merit	STC/ETC

Table D.l (continued)

Name	Award	Sponsor
Woodward, Jonathan	Significant Event Award	Energy Systems
	Advisory Editor	Journal of Biomass, Energy Society of China
Wymer, Raymond G.	Fellow	AIC
	Fellow and Award for Outstanding Work in Chemical Technology in Fuel Recycle	ANS
	President	AIC, Tennessee Institute of Chemists
	Member	NRC, Subcommittee on Nuclear and Radiochemistry
	Awards of Excellence (2) and Achievement	STC/ETC
	Silver Acorn Award	Energy Systems
	Robert E. Wilson Award	AIChE, Tennessee Chapter

Table D.2. IR-100 Awards”

Date	Technical Achievement	Inventors
1971	ORNL Ultraviolet Analyzer	C. D. Scott, N. G. Anderson, W. W. Pitt, Jr., and W. F. Johnson
1977	Portable Centrifugal Fast Analyzer	C. D. Scott, J. E. Mrocheck, R. K. Genung. W. F. Johnson, M. L. Bauer, C. A. Burtis, and D. G. Lakomy
1978	Pressurized Continuous Annular Chromatograph (CAC)	C. D. Scott, R. M. Canon, W. G. Sisson, and R. D. Spence
1979	Tapered Fluidized-Bed Bioreactor	C. D. Scott, D. D. Lee, and C. W. Hancher
1979	Gel-Sphere-Pat Nuclear Fuel Fabrication Process	Team of engineers and scientists in the Chemical Technology and Metals and Ceramics Divisions
1980	Portable Fluorescence Spotter	D. D. Schuresko, G. K. Schulze, R. G. Phillips, J. E. Mrocheck, M. S. Blair, W. A. Walker, W. W. Pitt, Jr., M. L. Bauer, and R. W. Wood
1980	DEPA-TOP0 Processed Uranium	Fred J. Hurst and David J. Crouse
1981	Perals Spectrometer	W. J. McDowell and G. N. Case
1985	Metal Oxide Varistor (ORNL SG-2)	R. J. Lauf and W. D. Bond

aThe JR-100 award, sponsored by the magazine ***Industrial Research and Development,*** is a prestigious award for significant technical product achievement. Each year, the 100 most significant technical products are selected and given this award. The Chemical Technology Division has won nine of these awards.

APPENDIX E

PATENTS RECEIVED BY CHEMICAL TECHNOLOGY DIVISION PERSONNEL (1950-1991)

Table E.l. Patents Received by Chemical Technology Division Personnel (1950-1991)

Year	Patent No.	Patentee	Subject
1951	2546,933	Steahly, F. L. Stoughton, R. W. Schuler, F. R.	Method of Dissolving **Thorium** Values
1957	2,815,322	Higgins, I. R.	Counter-Current Liquid-Solid Mass Transfer Method and Apparatus
1958	2,849,283	Stoughton, R. W. Steahly, F. L.	Separation of Uranyl Nitrate by Extraction
	2,852,419	Peterson, M. D. Overholt, D. C. Acken, M. F.	Process of Decontaminating Material Contaminated with Radioactivity
	2,859,094	Schmitt, J. M. Blake, C. A., Jr. Brown, K. B. Coleman, C. F.	Uranium Extraction Process using Synergistic Reagents
	2,863,718	Overholt, D. C. Tober, F. W.	Plutonium Concentration and Decontamination Method
	2,864,668	Baldwin, W. H. Higgins, C. E.	Uranium Extraction Process
	2,865,737	Blanco, R. E. Morrison, B. H.	Method of Purifying Uranium Metal
1959	2,877,131	Overholt, D. C. Peterson, M. D.	Method and Coating Composition for Protecting and Decontaminating **Surfaces**
	2,877,250	Brown, K. B. Crouse, D. J., Jr. Moore, J. G.	Recovery of Uranium Values
	2,895,798	Blanco, R. E.	Barium Recovery Process
	2,909,406	Meservey, A. B. Rainey, R. H. to Ruthenium	Process for Decontaminating Thorium and Uranium with Respect
	2,917,406	McBride, J. P.	Method of Increasing the Dispersibility of Slurry Particles
1960	2,931,706	Gresky, A. T. Nurmi, E. 0. Foster, D. L. Wischow, R. P. Savolainen, J. E.	Preparation of Dibasic Aluminum Nitrate
	2,937,925	Blake, C. A., Jr. Brown, K. B. Homer, D. E.	Solvent Extraction Process for Uranium from Chloride Solutions
	2,945,740	Gresky, A. T.	Ruthenium Decontamination Method

Table E.1 (continued)

Year	Patent No.	Patentee	Subject
1961	2,968,183	Hannaford, B . A. Rosenberg, R. Segaser, C. L. Terry, C. L.	Sampling System
	2,981,643	Baybarz, R. D.	Process for Descaling and Decontaminating Metals
	2,990,244	Brown, K. B. Crouse, D. J., Jr.	Extraction of Thorium and Uranium Values from Acid Leach Liquors
	2,992,249	Boyd, G. E. Russel. E. R. Taylor, M. D.	Ion Exchange Adsorption Process for Plutonium Separation
	2,992,886	Gens, T. A.	Method for Dissolving Zirconium-Uranium Compositions
	3 $09,062	Brooksbank, W. A.,Jr. Leddicotte, G. W. Strain, J. E. Hendon, H. H., Jr.	Absorption Analyzer
	3,009,768	Adams, J. B. Bresee, J. C. Ferris, L. M. Scott, C. D.	Continuous Process for Preparing Uranium Hexafluoride from Uranium Tetrafluoride and Oxygen
	3,023,085	McBride, J. P.	Method of Combining Hydrogen and Oxygen
1962	3,035,895	McCorkle, K. H. Kleinsteuber, A. T. Schilling, C. E. Dean, 0. C.	Preparation of High-Density, Compactible Thorium Oxide Particles
1962	3,039,847	Campbell, D. 0. Cathers, G. I.	Separation of Metal Values from Nuclear Reactor Poison
	3,043,653	Gens, T. A.	Recovery of Uranium from Zirconium-Uranium Nuclear Fuels
	3,048,474	Morse, L. E.	Catalytic Recombination of Radiolytic Gases in Thorium Oxide Slurries
	3,049,400	Rainey, R. H. Moore, J. G.	Solvent Extraction Process for the Separation of Uranium and Thorium from Protactinium and Fission Products
	3,052,361	Whatley, M. E. Woods, W. M.	Liquid Cyclone Contactor
	3,052,513	Crouse, D. J., Jr.	Stripping of Uranium from Organic Extractants
	3,065,044	Blanco, R. E. Higgins, I. R.	Recovery of Aluminum from Fission Products

Table E.l (continued)

Year	Patent No.	Patentee	Subject
1963	3,079,225	Baybarz, R. D. Lloyd, M. H.	Process for Separating Americium and Curium from Rare Earth Elements
	3,086,926	Helton, D. M. Savolainen, J. E.	Method of Dissolving Refractory Alloys
	3,101,058	Carr, W. H., Jr. Dobyns, E. R.	Diaphragm Pumping System
	3,105,052	Haws, C. C., Jr.	Preparation of Refractory Oxide Microspheres
	3,116,106	McNees, R. A., Jr. Taylor, A. J.	Preparation of High-Density Thorium Oxide Spheres
1964	3,117,372	McNees, R. A. Potter, R. A.	Stabilized Rare Earth Oxides for a Control Rod and Method of Preparation
	3,120,493	Clark, W. E. Godbee, H. W.	Suppression of Ruthenium Volatilization in Evaporation and Calcination of Radioactive Waste Solutions
	3,122,414	Homer, D. E. Wischow. R. P.	Process for Recovery of Strontium Values from Fission Product Waste Solutions
1964	3,148,941	Gens, T. A.	Dissolution of Uranium-Molybdenum Reactor Fuel Elements
1965	3,171,815	Kelly, J. L. Dean, 0. C. Ferguson, D. E.	Method for Preparation of Thorium Dicarbide and Thorium-Uranium Dicarbide Particles
	3,178,258	Cathers, G. I. Jolley, R. L.	Separation of Plutonium Hexafluoride from Uranium Hexafluoride by Selective Sorption
	3,179,503	Homer, D. E. Crouse, D. J., Jr. Brown, K. B.	Extraction of Cesium from Aqueous Solutions Using Phenols
	3,202,475	Gens, T. A.	Method for Collecting Zirconium Tetrachloride’
	3,211,526	Crouse, D. J., Jr.	Recovery of Sulfuric Acid from an Aqueous Solution Containing Metal Values by Extraction with Tertiary Amines
	3,218,123	Davis, W., Jr. Yee, W. C.	Recovery of Strontium Values from Sulfate-Containing Waste Solutions
	3,219,408	Bradley, M. J. Ferris, L. M.	Chemical Disintegntion and Recovery of Uranium from Sintered Graphite-Uranium Compacts
	3,222,289	Clark, W. E. Gens, T. A.	Dissolution of Zirconium in Titanium Equipment

Table E.l (continued)

Year	Patent No.	Patentee	Subject
1966	3,228,886	Lloyd, M. H.	Method of Preparing Compactible Thorium Oxide-Plutonium Oxide Particles
	3,230,036	Kappelmann, F. A. Weaver, B. S.	Method for Separating Americium and Curium from the Lanthanide Rare Earths and Yttrium
	3,238,014	Gens, T. A.	Recovery of Uranium and Plutonium Values from Aqueous Solutions of Ammonium Fluoride
	3,243,257	Coleman, C. F.	Recovery of Uranium and Zirconium from Aqueous Fluoride Solutions
	3,262,760	McCorkle, K. H. Morse, L. E. Schilling, C. E.	Method of Preparing High-Density Compactible Uranium Dioxide Particles
	3,265,627	Clark, W. E. Fitzgerald, C. L. Davis, G. D.	Addition of Lithium Values in Conversion of Fission-Product Wastes to a Glass-Like Solid for Disposal
	3,273,973	Bennett, M. R. Ullmann, J. W. Thoma, R. E., Jr.	Method for Processing Aluminum-Containing Nuclear Fuels
	3,275,422	Cathers, G. I. Mailen, J. C.	Continuous-Gas-Phase Volatility Process
	3,276,850	Rainey, R. H.	Method of Selectively Reducing Plutonium Values
	3,278,278	Flanary, J. R. Goode, J. H. Wall, G. C.	Pyrohydrolysis of Carbide-Type Nuclear Fuels
	3,278,387	McNeese, L. E. Scott, C. D.	Fuel Recycle System in a Molten Salt Reactor
	3,288,717	Morse, L. E.	Method for Preparation of Urania Sols
	3,290,122	Clinton, S. D. Haas, P. A. Hirth, L. J. Kleinsteuber, A. T.	Process for Preparing Oxide Gel Microspheres from Sols
1967	3,298,957	Gens, T. A.	Method for Dissolving Zirconium and Forming Stabilized Alkaline Solutions Thereof
	3,298,961	Davis, G. D. Frederick, E. J. Godbee, H. W. Holmes, J. M.	Concentration and Containment of Radioactivity from Radioactive Waste Solutions in Asphalt
	3,300,852	DeBruin, H. J.	Method for Bonding Beryllium Oxide to Graphite

Table E.l (continued)

Year	Patent No.	Patentee	Subject
1967	3,303,004	Cathers, G. I. Bennett, M. R.	Method for Dissolving Stainless Steel Members
	3,310,386	Lloyd, M. H.	Preparation of Plutonium Oxide Sol and Calcined Microspheres
	3,312,526	Hanson, C. K. Horton, R. M. Wadsworth, M. E.	Method and Catalyst for Combining Hydrogen and Oxygen in Thorium Oxide Slurries
	3,320,179	Gens, T. A.	Openly Porous Refractory Nuclear Fuel Microspheres and Method of Preparation
	3,328,132	Bennett, M. R. Cathers, G. I.	Method of Separating UF_6 from Bromine Fluorides
	3,331,898	Haas, P. A. Clinton, S. D.	Method for Preparing Metal Oxide Microspheres
	3,335,095	Moore, J. G.	Preparation of Actinide Sols by Amine Extraction
	3,352,950	Helton, D. M. Wymer, R. G.	Electrical Potential Method for Dispersion of Metal Oxide Microspheres
	3,356,776	Meservey, A. B. Sease, J. D. Fitts, R. B.	Method of Fabricating Ceramic Nuclear Fuel Product
	3,359,064	Crouse, D. J., Jr. Seeley, F. G.	Process for Separating Beryllium Values from Aqueous Solutions
1968	3,361,676	McBride, J. P. Pattison, W. L.	Urania Sol Forming Method in the Presence of Formic Acid and a Palladium-on-Thoria catalyst
	3,362,791	Ryon, A. D.	Apparatus for Separation of Immiscible Liquid Pairs Including Remote Interface Control
	3,367,881	Morse, L. E.	Extraction Method for Preparing UO_2 Microspheres
	3,374,157	Box, W. D.	Electrolyte for the Electrodeposition of Technetium
1969	3,451,790	Katz, S. Cathers, G. I.	Method of Separating Neptunium and Uranium Values
	3,461,076	Lloyd, M. H. Haire, R. G.	Process for Preparing Plutonia Aquasols
	3,463,738	Fitzgerald, C. L. Godbee, H. W. McCorkle, K. H., Jr.	Conversion and Containment of Radioactive Organic Liquids into Solid Form

Table E.l (continued)

Year	Patent No.	Patentee	Subject
1969	3,463,814	Blanco, R. E. Clark, W. E. Yee, W. C.	Chemical Cycle for Evaporative Water Desalination Plant
	3,472,633	McNeese, L. E. Watson, J. S. Whatley, M. E.	Liquid-Liquid Removal of Protactinium from Spent Molten Salt Mixtures Containing Uranium Tetrafluoride
1970	3,513,101	Meservey, A. B.	Preparation of Stable Sols of Thoria-Uranium Trioxide
	3,518,063	Seeley, F. G. Crouse, D. J., Jr.	Purification of Beryllium by Liquid-Liquid Extraction
1971	3,575,875	Pattison, W. L. McBride, J. P.	Method for Preparing Uranium-Containing Aquasols Employing a Platinum Oxide Catalyst
	3,577,485	Beatty, R. L. Leitnaker, J. M. Notz. K. J., Jr.	Method for Preparation of Carbonitride Nuclear Fuel Materials
	3,580,705	Coleman, C. F. Weaver, B. S.	Selective Stripping of Plutonium from Organic Extracts
	3,600,323	Tallent, 0. K.	Method for Preparing Stable Urania-Plutonia Sols
	3,617,585	Haas, P. A. Clinton, S. D.	Method for Preparing Oxide Gel Microspheres from Sols
	3,627,479	Yee, W. C.	Chemical-Electra-Chemical Cycle for Desalination of Water
	3,629,133	McBride, J. P. McCorkle, K. H. Pattison, W. L.	Production of Predominantly Crystalline Sols or Urania
	3,629,138	Thomas, I. L.	Method for Exchanging Counterions in Actinide Oxide Sols
1972	3640,888	Baybarz, R. D. Peterson, J. R.	Califomium-252 Neutron Source and Method of Making Same
	3,672,846	McNeese, L. E. Ferguson, D. E.	Method for Reprocessing Spent Molten Salt Reactor Fuels
	3,675,746	Irvine, A. R.	Impact Energy Absorber
	3,677,719	Whatley, M. E.	Method for Reprocessing Molten Fluoride Salt Reactor Fuels
	3,683,975	Harrington, F. E. Sease, J. D.	Method of Vibratory Loading Nuclear Fuel Elements
	3,716,616	Lin, K. H.	Process for Decontamination of Neutron-Irradiated Beryllium

Table E.l (continued)

Year	Patent No.	Patentee	Subject
1972	3,708,392	Campbell, D. 0.	Isotope Enrichment Process for Lanthanide and Actinide Elements
1973	3,711,591	Hurst, F. J. Crouse, D. J.	Reductive Stripping Process for the Recovery of Uranium from Wet-Process Phosphoric Acid
	3,714,056	Tallent, 0. K.	Method for Preparing Stable Urania-Plutonia Sols
	3,714,322	Bell, M. J. Whatley, M. E.	Method for Preparing High Purity ^{233}Uranium
	3,715,317	Lloyd, M. H. Tallent, 0. K. Leuze, R. E.	Solvent Extraction Process for Producing Low-Nitrate and Large-Crystal-Size PuO_2 Sols
	3,717,582	Bradley, R. A. Lindemer, T. B.	Method for Reducing the Oxygen in Certain Actinide Oxides to Less than Stoichiometric Levels
	3,725,293	Haas, P. A.	Conversion of Fuel-Metal Nitrate Solutions to Oxides
	3,735,736	Yee, W. C. Stout, P. R.	Method for Growing Edible Aquatic Animals on a Large Scale
	3,742,720	Ferguson, D. E. Haas, P. A. Leuze, R. E.	Quantitative Recovery of Krypton from Gas Mixtures Mainly Comprising Carbon Dioxide
	3,744,975	Mailen, J. C.	Rotor for Multistation Photometric Analyzer
	3,752,876	Cathers, G. I. Shockley, W. E.	Removal of Organic and Inorganic Iodine from a Gaseous Atmosphere
	3,758,670	McBride, J. P. McCorkle, K. H. Pattison, W. L.	Production of Predominantly Crystalline Sols
	3,761,564	Mailen, J. C. Ferris, L. M.	Separation of Califomium from Other Actinides
	3,763,292	Stradley, J. G. Sease, J. D.	Manufacture of Bonded-Particle Nuclear Fuel Composites
	3,763,374	Tiffany, T. 0. Mailen, J. C. Thacker, L. H.	Dynamic Multistation Photometer-Fluorometer
	3,764,552	Godbee, H. W. Lovelace, R. C.	Method for Storing Radioactive Combustible Waste
	3,778,348	Harrington, F. E. Sease, J. D.	Nuclear Fuel Element with Axially Aligned Fuel Pellets and Fuel Microspheres Therein

Table E.l (continued)

Year	Patent No.	Patentee	Subject
1974	3,785,161	Singh, S. P. N. Hart, W. C.	Separation of Components of Vaporous Fluids
	3,792,154	Cathers, G. I. Shipman, C. J.	Removal of Iodine from Nitric Acid Solutions
	3,793,433	Seeley, F. G. Baldwin, W. H.	Extraction of Lithium from Neutral Brines Using a Beta Diketone and Trioctylphosphine Oxide
	3,794,715	Lloyd, M. H.	Solvent Extraction Process for Producing Low-Nitrate and Large-Crystal-Size PuO_2 Sols
	3,795,451	Mailen, J. C.	Rotor for Fast Analyzer of Rotary Cuvette Type
	3,798,123	Lindemer, T. B.	Nuclear Fuel for High-Temperature Gas-Cooled Reactors
	3,798,459	Anderson, N. G. Burtis, C. A. Johnson, W. F. Mailen, J. C. Scott, C. D.	Compact Dynamic Multistation Photometer Utilizing Disposable Cuvette Rotor
	3,800,023	Haas, P. A.	Loading a Cation Exchange Resin with Uranyl Ions
	3,800,161	Scott, C. D. Collins, E. L.	Portable Dynamic Multistation Photometer-Fluorometer
	3,803,295	Cathers, G. I. Shipman, C. J.	Method for Removing Iodine from Nitric Acid
	3,804,533	Scott, C. D.	Rotor for Fluorometric Measurements in Rotary Fast Analyzer
	3,806,581	Grimes, W. R. Shaffer, J. H. Doss, F. A.	Removal of Fluoride from Chloride or Bromide Melts
	3,821,356	Baldwin, W. H.	Production of High Purity Halides
	3,825,649	Gresky, A. T. McDuffee, W. T., Jr. Wischow, R. P. Savolainen, J. B.	Process for Separation of Protactinium, Thorium and Uranium from Neutron-Irradiated Thorium
	3,827,989	Scott, C. D.	Impregnated Chemical Separation Particles
	3,835,214	Hurst, F. J. Crouse, D. J., Jr.	Oxidative Stripping Process for the Recovery of Uranium from Wet-Process Phosphoric Acid
	3,835,040	Mahlman, H. A. Sisson, W. G.	Cross-Flow Filtration Process

Table E.l (continued)

Year	Patent No.	Patentee	Subject
1974	3,835,122	Parkinson, W. W., Jr. Kelly, M. J. Sturm, B. J. Martin, W. J.	Novel Polystyrene Product Having Rapid Post-Irradiation Decay of Conductivity and Process of Making Same
	3,847,550	Scott, C. D. Pitt, W. W., Jr.	Differential Chromatographic Method
	3,851,179	Irvine, A. R.	Shipping Cask Neutron and Heat Shield
	3,852,407	Schmitt, J. M. Crouse, D. J.. Jr. Howerton, W. B.	Method for Removing Alkyl Iodides from Air by Mercuric Nitrate Solution
	3,853,979	McNeese, L. E. Ferris, L. M. Smith, F. J.	Method for Removing Rare Earths from Spent Molten Metallic Fluoride Salt Mixtures
	3,854,508	Burtis, C. A. Johnson, W. F. Walker, W. A.	Automated Sample-Reagent Loader
1975	3,860,691	Gens, T. A.	Actinide Mononitride Microspheres and Process
	3,864,089	Mailen, J. C. Tiffany, T. 0. Scott, C. D. Pitt, W. W., Jr. Johnson, W. F.	Multiple-Sample Rotor Assembly for Blood Fraction Preparation
	3,878,041	Leitnaker, J. M. Lindemer, T. B.	Oxynitride Fuel Kernel for Gas-Cooled Reactor Fuel Particles
	3,880,619	Richardson, D. M. Bamberger, C. E.	Solid Sot-bent for Trapping Iodine
	3,890,101	Tiffany, T. 0. Walker, W. A. Johnson, W. F.	Collection Ring for Use in Multiple-Sample Blood Fractionation Centrifugal Rotors
	3,899,296	Mailen, J. C. Johnson, W. F.	Whole Blood Analysis Rotor for a Multistation Dynamic Photometer
	3,901,658	Burtis, C. A. Johnson, W. F.	Whole Blood Analysis Rotor Assembly Having Removable Cellular Sedimentation Bowl
	3,908,123	Veach, A. M. Bell, W. A.. Jr.	Extraction Electrode Geometry for a Calutron
	3,914,388	Cathers, G. I. Shipman, C. J.	Volatilization of Iodine from Nitric Acid Using Peroxide
	3,919,406	Grimes, W. R. Bamberger, C. E.	Thermochemical Production of Hydrogen

Table E.l (continued)

Year	Patent No.	Patentee	Subject
1975	3,920,577	Godbee, H. W. Cathers, G. I. Blanco, R. E.	Iodine Retention During Evaporative Volume Reduction
	3,927,192	Bamberger, C. E. Richardson, D. M.	Chemical Cycle for Thermochemical Production of Hydrogen from Water
	3,929,979	Bamberger, C. E. Richardson, D. M. Grimes, W. R.	Process for Generating Hydrogen
1976	3,969,218	Scott, C. D.	Elution Electrophoresis
	3,979,498	Campbell, D. 0.	Recovery of Cesium and Palladium from Nuclear Reactor Fuel Processing Waste
	3,995,009	Notz, K. J., Jr.	Process for Loading Weak-Acid Ion Exchange Resin with Uranium
1977	4,004,993	Homer, D. E. Mailen, J. D. Posey, F. A.	Electrolytic Trapping of Iodine from Process Gas Streams
	4,005,178	Bennett, M. R. Bamberger, C. E. Kelmers, A. D.	Method for Converting UF_5 to UF_4 in a Molten Fluoride Salt
	4,012,209	Coleman, C. F. McDowell, W. J.	Liquid Film Target Impingement Scrubber
	4,017,417	Clark, W. E. Thompson, C. T.	Immobilization of Iodine in Concrete
	4,025,602	Campbell, D. 0. Buxton, S. R.	Recovery of Transplutonium Elements from Nuclear Reactor Waste
	4,032,407	Scott, C. D. Hancher, C. W.	Tapered Bed Bioreactor
	4,035,156	Shumate, S. E., II	Filter Type Rotor for Multistation Photometer
	4,046,690	Rodgers, B. R. Edwards, M. S.	Filtering Coal-Derived Oil Through a Filter Media Precoated with Particles Partially Solubilized by Said Oil
	4,051,202	Arnold, W. D., Jr.	Method for Separating Mono-and-Di-Octylphenyl Phosphoric Acid Esters
1978	4,069,293	Tallent, 0. K.	Method for Dissolving Plutonium Dioxide
	4,070,438	Notz, K. J. Rainey, R. H. Greene, C. W. Shockley, W. E.	Method for Loading Resin Beds

Table E.1 (continued)

Year	Patent No.	Patentee	Subject
1978	4,077,838	Beatty, R. L. Lindemer, T. B. Long, E. L., Jr.	Pyrolytic Carbon-Coated Nuclear Fuel
	4,083,242	Cochran, H. D., Jr.	Method of Measuring the Mass Flow Rate of a Substance Entering a Cocurrent Fluid Stream
	4,131,527	Friedman, H. A. Toth. L. M. Bell, J. T.	Method for Selectively Reducing Plutonium Values by a Photochemical Process
1979	4,132,639	Katz, S. Rodgers, B. R.	Method for Improving the Sedimentation and Filterability of Coal-Derived Liquids Compact Gate Valve
	4,134,960	Vondra, B. L. Tallent, 0. K. Mailen, J. C.	Method for Dissolving Plutonium Oxide with HI and Separating Plutonium
	4,162,298	Holladny, D. W. Haag, G. L.	Method of Immobilizing Carbon Dioxide from Gas Streams
1980	4,200,801	Schuresko. D. D.	Portable Spotter for Fluorescent Contaminants on Surfaces
	4,225,455	Haas, P. A.	Process for Decomposing Nitrates in Aqueous Solution
1981	4,252,777	Seeley, F. G. McDowell, W. J.	Recovery of Aluminum and Other Metal Values from Fly Ash
	4,254,088	Seeley, F. G. McDowell, W. J.	Salt-Soda Sinter Process for Recovering Aluminum from Fly Ash
	4,263,406	Bostick, W. D. Denton, M. S. Dinsmore, S. R.	Apparatus for Continuously Referenced Analysis of Reactive Components in Solution
	4,290,967	Campbell, D. 0. Buxton, S. R.	Process for Recovery of Palladium from Nuclear Fuel Reprocessing Wastes
1982	4,316,776	Arwood, P. C. Bell, J. T. Mailen, J. C. Tallent, 0. K.	Method of Removing Pu(IV) Polymer from Nuclear Fuel Reclaiming Liquid
	4,336,044	Barker, R. E. Ryon, A. D. Scott, C. D.	Process for Separating Coal Synthesized Methane from Unreacted Intermediate and Contaminant Gases
	4,339,309	Howerton, W. B. Pruett, D. J.	Method for Removing Fluoride Contamination from Nitric Acid
	4,343,478	Morgan, J. G. Rennich, M. J. Whatley, M. E.	Face Seal Assembly Rotating Drum

Table E.1 (continued)

Year	Patent No.	Patentee	Subject
1982	4,357,420	Bostick, W. D. Denton, M. S. Dinsmore, S. R.	Bioluminescence Methods for Enzymatic Determinations
	4,358,426	Crouse, D. J., Jr. Mailen, J. C. Tallent, 0. K.	Method for Cleaning Solution Used in Nuclear Fuel Reprocessing
1983	4,376,792	Angelini, P. Lackey, W. J., Jr. Stinton, D. P. Blanco, R. E. Bond, W. D. Arnold, W. D., Jr.	Method for Primary Containment of Cesium Wastes
	4,382,885	Haas, P. A.	Method for Producing Nuclear Fuel
	4,394,269	Mailen, J. C. Pannell, K. E. Tallent, 0. K.	Method for Cleaning Solution Used in Nuclear Fuel Reprocessing
	4,397,778	Lloyd, M. H.	Coprocessed Nuclear Fuels Containing (U,Pu) Values as Oxides, Carbides or Carbonitrides
	4,409,157	Haas, P. A. Stines, W. B.	Method for Improved Decomposition of Metal Nitrate Solutions
	4,413,058	Donaldson, T. L. Arcuri, D. J.	Continuous Production of Ethanol by Use of Flocculent Zymomonas Mobilis
	4,415,536	Haas, P. A. Ryon, A. D.	Apparatus for Contacting Particulate Material with Processing Liquid
1984	4,431,609	Scheitlin, F. M.	Removal of Radium from Acidic Solutions Containing Same by Adsorption on Coal Fly Ash
	4,432,945	Hurst, F. J. Brown, G. M. Posey, F. A.	Removing Oxygen from a Solvent Extractant in a Uranium Recovery Process
	4,442,211	Greenbaum, E.	Method for Producing Hydrogen and Oxygen by Use of Algae
	4,443,413	McTaggart, D. R. Pruett, D. J.	Separation of Uranium from Technetium in Recovery of Spent Nuclear Fuel
	4,446,105	Dinsmore, S. R. Mrochek, J. E.	System for Analyzing Coal Liquefaction Products
	4,459,245	Haas, P. A. Ryon, A. D. Vavruska, J. S.	Method and Apparatus for Controlled Size Distribution of Gel Microspheres Formed From Aqueous Dispersions
	RE. 31,686	Hurst, F. J. Crouse, D. J., Jr.	Reductive Stripping Process for the Recovery of Uranium from Wet-Process Phosphoric Acid

Table E.l (continued)

Year	Patent No.	Patentee	Subject
1984	4,476,105	Greenbaum, E.	Process for Photosynthetically Splitting Water
1985	4,459,338	Angelini, P. DeVore, C. Lackey, W. J., Jr. Blanco, R. E. Stinton, D. P.	Method for Deposition of Silicon Carbide Layers on Substrates and Product
	4,500,493	Hurst, F. J.	Reductive Stripping Process for Uranium Recovery from Organic Extracts
	4,502,987	Lloyd, M. H. Collins, J. L. Shell, S. E.	Method of Controlling Crystallite Size in Nuclear-Reactor Fuels
	4,509,856	Lee, N. E.	Rotor for Centrifugal Fast Analyzers
	4,528,165	Friedman, H. A.	Separation of Uranium from Technetium in Recovery of Spent Nuclear Fuel
1986	4,562,748	Mrochek, J. E. Dinsmore, S. R. Chandler, E. W.	Disc Valve for Sampling Erosive Process Streams
1987	4,657,646	Greenbaum. E.	Method of Producing Metallized Chloroplasts and Use Thereof in the Photochemical Production of Hydrogen and Oxygen
	4,663,093	Haas, P. A. Fowler, V. L. Lloyd, M. H.	Preparation of Nuclear Fuel Spheres by Flotation-Internal Gelation
	4,666,654	Forsberg, C. W.	Boiling Water Neutronic Reactor Incorporating a Process Inherent Safety Design
	4,683,042	Scott, C. D.	Method and Apparatus for Continuous Annular Electrochromatography
	4,740,472	Burtis, C. A. Johnson, W. F. Walker, W. A.	Method and Apparatus for Automated Processing and Aliquoting of Whole Blood Samples for Analysis in a Centrifugal Fast Analyzer
1988	4,767,929	Scott, T. C. Wham, R. M.	Surface Area Generation and Droplet Size Control in Solvent Extraction Systems Utilizing High Intensity Electric Fields
	4,789,436	Greenbaum, E.	Method and Apparatus for Nondestructive In Vivo Measurement of Photosynthesis
1989	4,800,183	Quinby, T. C.	Method for Producing Refractory Nitrides
	4,835,106	Johnson, W. F. Burtis, C. A. Walker, W. A.	Rotor for Processing Liquids Using Movable Capillary Tubes
	4,840,904	Woodward, J.	Recovery and Reuse of Cellulase Catalyst in an Enzymatic Cellulose Hydrolysis Process

Table E.l (continued)

Year	Patent No.	Patentee	Subject
1989	4,846,964	Scott, C. D. Strandberg, G. W.	Fluidized-Bed Bioreactor Process for the Microbial Solubilization of Coal
	4,847,205	Burtis, C. A. Johnson, W. F.	Device and Method for Automated Separation of a Sample of Whole Blood into Aliquots
	SIR H659	Haas, P. A.	Process for Electrolytically Preparing Uranium Metal
	SIR H660	Tallent, 0. K. Dodson, K. E. McDaniel, E. W.	Method and Composition for Immobilization of Waste in Cement-Based Materials
	4,941,959	Scott, T. C.	Magnetic/Electric Field Solvent Extraction
1990	4,914,024	Strandberg, G. W. Lewis, S. N.	Microbial Solubilization of Coal
	4,916,092	Tiegs, T. N. Lindemer, T. B.	Ceramic Composites Reinforced with Modified Silicon Carbide Whiskers
	SIR H800	Beahm, E. C. Shockley, W. E.	Method for Gettering Organic, Inorganic, and Elemental Iodine in Aqueous Solutions
	4,941,959	Scott, T. C.	Electric Field-Driven, Magnetically Stabilized Fen-o-Emulsion Phase Contactor
	SIR H857	Haas, P. A.	Electrolytic Process for Preparing Uranium Metal
	4,978,647	Scott, C. D. Woodward, C. A. Byers, C. H.	Gel Bead Composition for Metal Adsorption
1991	4,994,416	Tiegs, T. N. Lindemer, T. B.	Ceramic Composites Reinforced with Modified Silicon Carbide Whiskers and Method for Modifying the Whiskers
	4,995,985	Scott, C. D. Woodward, C. A. Byers, C. H.	Gel Bead Composition for Metal Adsorption
	5,100,781	Greenbaum, E.	Measurement of Gas Production by Algal Clones

List of Drawings and Photos

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