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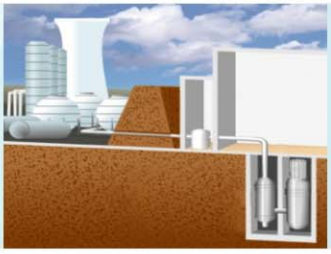

NUCLEAR ENERGY RESEARCH INITIATIVE

2004 Annual Report

Foreword

The Nuclear Energy Research Initiative (NERI) began in Fiscal Year (FY) 1999 as the core of a new, restructured Federal effort to develop advanced nuclear energy concepts and technologies. NERI was developed based on recommendations made in the November 1997 report by the President's Committee of Advisors on Science and Technology, *Federal Energy Research and Development for the Challenges of the Twenty-First Century.*

The NERI program has focused on preserving, advancing, and encouraging innovative nuclear science and technology research and development (R&D) within the Nation's universities, laboratories, and industry. It supports the *National Energy Policy* by conducting research that addresses the potential long-term barriers to both maintaining and expanding nuclear generation of electricity in this country. Working in tandem with other nuclear energy programs, such as the Generation IV Nuclear Energy Systems Initiative (Generation IV), the Advanced Fuel Cycle Initiative (AFCI), the Nuclear Hydrogen Initiative (NHI), and the Nuclear Power 2010 (NP 2010) program, the NERI program assists in responding to the Nation's need for economical and environmentally conscious sources of energy.

Since its inception, NERI has been realizing its goal of both developing advanced nuclear energy systems and providing state-of-the-art research concerning nuclear science and technology. The research effort conducted by the Nation's universities, laboratories, and industry partners has helped to maintain and improve the nuclear research infrastructure in this country. University involvement in NERI-funded research has been particularly important in renewing student interest in pursuing degrees in nuclear engineering and related sciences and enabling educational institutions across the country to stay at the forefront of nuclear science research.

Because of these past successes and the need to address new issues, in 2004, the Office of Nuclear Energy, Science, and Technology (NE) decided to focus the creativity and innovative thinking of the Nation’s universities on nuclear energy research associated with the Department’s high-priority nuclear technology research objectives that are defined by the agency’s major R&D programs—Generation IV, AFCI, and NHI. While all new NERI projects will be led by Principal Investigators at the Nation's universities, National laboratories and industry partners may still participate as collaborators.

This annual report summarizes the results of the 12 NERI projects initiated in FY 2001 and completed in FY 2004, the research progress of the NERI projects initiated in FY 2002, and the research description and work scope of the newly initiated FY 2005 NERI projects. The final summaries for the NERI projects initiated in FY 1999 and FY 2000 can be found in the NERI 2002 and 2003 Annual Reports, respectively. This report disseminates the results of NERI-sponsored research to the wide R&D community to spur yet more innovation, assuring a bright future for nuclear energy in the United States and the world.

William D. Magwood IV, Director

Office of Nuclear Energy, Science and Technology

U.S. Department of Energy

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NUCLEAR ENERGY RESEARCH INITIATIVE

1. Introduction

The United States Department of Energy (DOE) created the Nuclear Energy Research Initiative (NERI) in Fiscal Year (FY) 1999 in response to recommendations provided by the President's Committee of Advisors on Science and Technology. The purpose of NERI is to sponsor research and development (R&D) in the nuclear energy sciences to address the principal barriers to the future use of nuclear energy in the United States. NERI is helping to preserve the nuclear science and engineering infrastructure within the Nation's universities, laboratories, and industry, and is advancing the development of nuclear energy technology, enabling the United States to maintain a competitive position in nuclear science and technology. Research under this initiative also addresses issues associated with the maintenance of existing U.S. nuclear plants. The NERI program is managed and funded by DOE's Office of Nuclear Energy, Science and Technology.

The *Nuclear Energy Research Initiative 2004 Annual Report* serves to inform interested parties of progress made in NERI on a programmatic level as well as research progress made on individual NERI projects. Section 2 of this report provides background on the creation and implementation of NERI and on the focus areas for NERI research. Section 3 provides a discussion on NERI's mission, goals and objectives, and work scope. Section 4 highlights the major accomplishments of the NERI projects and provides brief summaries of the NERI research efforts that were completed in 2004. Section 5 provides a discussion on the impact NERI has had on U.S. university nuclear programs.

Sections 6 through 8 provide project status reports by research area for each of the fiscal year (FY) 2001 and 2002 projects that were active in FY 2004. Research objectives, progress made over the last year, and activities planned for the next year are described for each project. Sections 9 through 11 present each of the newly awarded 2005 NERI projects in their corresponding focus area. Each abstract includes a brief description of the project, its proposed work scope, and its participants.

Project numbers are designated by the FY in which the award was made. At the end of the document, there is an Index of NERI Projects grouped by FY and sequentially ordered by project number.

2. Background

In January 1997, the President tasked his Committee of Advisors on Science and Technology (PCAST) to review the current national energy R&D portfolio and to provide a strategy to ensure that the United States has a program to address the Nation's energy and environmental needs for the next century.

In its November 1997 Report to the President, *Federal Energy Research and Development for the Challenges of the Twenty-First Century*, the PCAST panel on Energy Research and Development determined that it was important to establish nuclear energy as a viable and expandable option and that a properly focused R&D effort to address the potential long-term barriers to the expanded use of nuclear power (e.g., nuclear waste, proliferation, safety, and economics) was appropriate. The PCAST panel further recommended that DOE reinvigorate its nuclear energy R&D activities with a new nuclear energy research initiative to address these potential barriers. DOE would fund research through this new initiative, based on a competitive selection of proposals from the national laboratories, universities, and industry.

DOE endorsed the PCAST recommendations and received Congressional appropriations in FY 1999, allowing NERI to sponsor innovative scientific and engineering R&D to address the key issues affecting the future use of nuclear energy and preserve the Nation's nuclear science and technology leadership.

I-NERI

In 1999, another PCAST report, *Powerful Partnerships: The Federal Role in International Cooperation on Energy Innovation*, recommended creating an international component to NERI to promote "bilateral and multilateral research focused on advanced technologies for improving the cost, safety, waste management, and proliferation-resistance of nuclear fission energy systems." In FY 2001, the Department launched the new International Nuclear Energy Research Initiative (I-NERI) for bilateral and multilateral nuclear energy research. In 2002, appropriated funding supported bilateral, cost-shared research work under the I-NERI program with South Korea, France, and a third collaboration involving Argonne National Laboratory and a consortium of ten international participants represented by the U.S. Nuclear Regulatory Commission (NRC) and the European Organization for Economic Cooperation and Development (OECD). Three new agreements with the European Union, Brazil, and Canada were established in 2003 and a fourth with Japan was established in 2004. Also, in FY 2004, new sets of collaborative projects were awarded to France, Korea, and Canada. Similar international agreements with the Republic of South Africa and the United Kingdom are being considered.

The Department is using the bilateral I-NERI agreements it has implemented with other nations to continue international cost-shared research and development (R&D) in the Generation IV Nuclear Energy Systems Initiative, Advanced Fuel Cycle Initiative, and Nuclear Hydrogen Initiative. This approach to executing international, cost-shared research, allows the Department to use all nuclear energy R&D programs as a basis for international, cost-shared R&D thereby significantly increasing the amount of research achievable. I-NERI allows DOE to leverage Federal investment and international resources through cost-sharing arrangements with each participating country on a wide range of nuclear technology topics. I-NERI will further enhance the influence of the United States and DOE in international policy discussions on the future direction of nuclear energy. Similar to NERI, I-NERI features competitive, researcher-initiated R&D that is selected through an independent peer-review process by international experts from the United States and its partners. A separate report covering the research effort conducted in the I-NERI program will be published in 2005.

NERI's Focus

In order to determine the initial focus of the NERI research areas, DOE convened a workshop of nuclear community stakeholders in April 1998, representing national laboratories, universities, and industry. As a result of this NERI workshop, DOE focused its initial scientific and engineering R&D in the following areas:

- Proliferation-resistant reactors and fuel technology
- New reactor designs to achieve improved performance, higher efficiency, and reduced cost, including low-output power reactors for use where large reactors are not attractive
- Advanced nuclear fuels

- New technologies for management of nuclear waste
- Fundamental nuclear science

To encourage innovative R&D, a unique process for selecting new NERI projects has been employed since the program's inception. In response to the NERI solicitations, principal investigators (PIs) select research topics of interest and define the scope and extent of the R&D in their proposals. DOE employs an independent, expert peer review process to judge the scientific and technical merit of the R&D proposals. DOE reviews those proposals judged to have the highest scientific and technical merit to ensure their conformance with DOE policy and programmatic requirements. After the proposals are judged and the projects are reviewed by DOE, the award-selections are recommended to DOE's Source Selection Official.

Since the initiation of NERI, a number of events have influenced the focus of NERI research activities.

- In 1998, DOE established the independent Nuclear Energy Research Advisory Committee (NERAC). This committee provides advice to the Secretary and to the Director, Office of Nuclear Energy, Science and Technology (NE), on DOE's civilian nuclear technology program. In June 2000, NERAC issued the *Long-Term Nuclear Technology Research and Development Plan.* This plan identifies the research and technology development that is necessary over the next 10-20 years to help ensure the long-term viability of nuclear energy as an electricity generation option in the United States. NERAC also established a task force to identify R&D needs related to non-proliferation issues associated with nuclear power production. Their recommendations for appropriate research in this area were provided to DOE in a January 2001 report titled, *Technical Opportunities to Increase the Proliferation Resistance of Global Civilian Nuclear Power Systems (TOPS).*
- The *National Energy Policy*, issued in May 2001 by the Vice President's National Energy Policy Development Group, supports the expansion of nuclear energy as one of its major initiatives for meeting the growing energy requirements of the United States. The *National Energy Policy* provides the core element in the planning for DOE's nuclear energy research programs addressing, among other areas, the research and development of advanced reactor and fuel cycle concepts, hydrogen production from nuclear energy, and the associated enabling sciences and technologies.
- In April 2003, DOE issued the *A Technology Roadmap for Generation IV Nuclear Energy Systems.* In coordination with the ten-country-member Generation IV International Forum, six reactor system concepts were selected for further development. These include the Very-High-Temperature Reactor System, the Gas-Cooled Fast Reactor System, the Supercritical Water-Cooled Reactor System, the Lead-Cooled Fast Reactor System, the Sodium-Cooled Fast Reactor System, and the Molten Salt Reactor System.
- Because of the initiation of new nuclear R&D programs such as the Generation IV Nuclear Energy Systems Initiative, the Advanced Fuel Cycle Initiative, and the Nuclear Hydrogen Initiative, NERI projects selected in FY 2002 were more focused on providing a supportive role to these important programs. In 2004, the focus of NERI research changed to exclusively fund research and development supporting these three initiatives. In addition, only U.S. universities were now allowed to serve as the PIs for these new projects. National laboratories and private companies can still participate as collaborating organizations. This new focus directly supports NERI's goal to preserve, improve, and advance the Nation's nuclear science and engineering infrastructure and will further assist these nuclear energy initiatives in accomplishing their goals and objectives.

NUCLEAR ENERGY RESEARCH INITIATIVE

3. NERI Program Description

NERI's Mission

The importance of nuclear power to the world's future energy supply requires that DOE apply its unique resources, specialized expertise, and national leadership to address key issues affecting the future of nuclear energy. NERI is a national, research-oriented initiative focused on innovation and competitiveness. The Initiative is helping DOE foster innovative ideas in such areas as advanced nuclear energy systems, hydrogen production from nuclear power, advanced nuclear fuels and fuel cycles, and fundamental science. This research enhances the ability of nuclear energy to help meet the Nation's future energy needs and environmental goals. To achieve these long-range goals, NERI has the following objectives:

- To address and help overcome the potential technical and scientific obstacles to the long-term, future use of nuclear energy in the United States, including those involving non-proliferation, economics, and nuclear waste disposition.
- To advance the state of U.S. nuclear technology so that it can maintain a competitive position in overseas markets and a future domestic market.
- To promote and maintain a nuclear science and engineering infrastructure to meet future technical challenges.

NERI's Work Scope

In FY 2004, NERI continued its R&D focus to address research requirements introduced in the *National Energy Policy*. Work during this year was the continuation of research projects initiated in FY 2001 and FY 2002. These research projects fell under the four research categories described below.

Advanced Nuclear Energy Systems. This program element includes the investigation and preliminary development of advanced concepts for reactor and power conversion systems. These systems offer the prospect of improved performance and operation, design simplification, enhanced safety, and reduced overall cost. Projects involve innovative reactors, system and component designs, alternative power conversion cycles for terrestrial applications, new research in advanced digital instrumentation and control and automation technologies, and other important design features and characteristics.

Hydrogen Production from Nuclear Power. This program element includes R&D to identify and evaluate new and innovative concepts for producing hydrogen using nuclear reactors. This research supports the Nuclear Hydrogen Initiative (NHI) by investigating hydrogen generation processes compatible with advanced reactor systems and integrating parameters needed to develop systems that are efficient and cost effective. Projects in this section have been integrated into either the Advanced Nuclear Energy Systems area or the Fundamental Science area, depending on their technical focus.

Advanced Nuclear Fuels/Fuel Cycles. This element includes R&D to provide measurable improvements in the understanding and performance of nuclear fuel and fuel cycles with respect to safety, waste production, proliferation resistance, and economics, in order to enhance the long-term viability of nuclear energy systems. This research supports the Generation IV concepts by improving the performance of fuels, developing fuels capable of withstanding higher temperature and more highly corrosive environments, and developing advanced proliferation-resistant fuels capable of high burn-up.

Fundamental Science. This element includes R&D in the fields of materials science and fundamental chemistry. Fundamental science research funded by NERI applies to and supports the preceding program elements in advanced nuclear engineering technology. Material sciences applications include research and development on materials for use in advanced nuclear reactor systems, structures, and components (including fuel cladding) that may perform in high-radiation fields, high temperatures and pressures, and/or in highly corrosive environments (i.e., lead-bismuth). Chemical science research may focus on developing and improving primary and secondary coolant chemistry in advanced reactors. Another research subject includes investigating nuclear isomers that could prove beneficial in civilian applications.

Safety, non-proliferation, and waste management are considerations intrinsic to all four research topics, especially for the advanced nuclear energy systems and advanced

fuels/fuel cycles. Thus, they become selection criteria across all four focus areas, and do not in themselves constitute focus areas.

In 2004, NE decided to have new NERI projects conduct applied nuclear energy research related to NE's research and development (R&D) programs. These programs are the:

- Generation IV Nuclear Energy Systems Initiative (Generation IV)
- Advanced Fuel Cycle Initiative (AFCI)
- Nuclear Hydrogen Initiative (NHI)

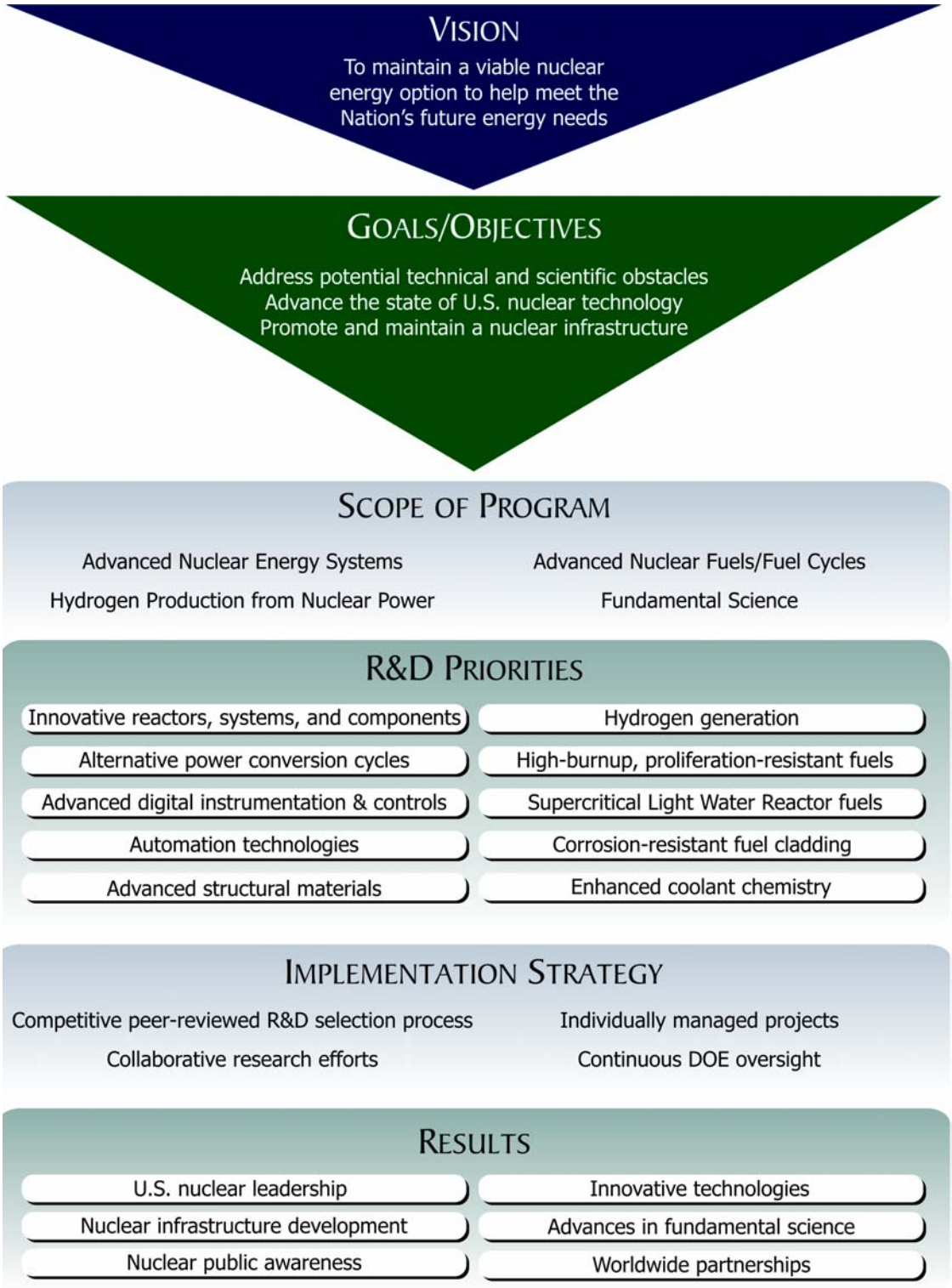
In addition, all new NERI projects will be led entirely by U.S. universities with national laboratories and industry providing a supporting role. The goal of this fresh focus is to concentrate more on highly-needed applied engineering research and to better integrate the educational institutions with the research efforts and initiatives of the Department. The Department and the universities will act as partners in the success of the new, more focused program. To facilitate this transition, NE hosted the *Advanced Reactor, Fuel Cycle, and Energy Products Workshop for Universities* in March 2004 in order to indoctrinate universities into the new NERI program. Over 150 university staff members attended the affair. A similar workshop is being planned for FY 2005.

Applied research in the three R&D program areas will continue to contribute to the revitalization of nuclear technology in our nation. The three initiatives will develop proficient and efficient technologies for future electrical power production, hydrogen production, and other energy conversion systems.

Generation IV is developing and demonstrating advanced nuclear energy systems that will meet future needs for safe, sustainable, environmentally-responsible, economic, proliferation-resistant, and physically secure energy. AFCI is developing advanced fuels and spent fuel treatment and transmutation technologies in order to enable a transition from the current once-through nuclear fuel cycle to a future sustainable, proliferation-resistant, closed nuclear fuel cycle. NHI is working to demonstrate the economic, commercial-scale production of hydrogen from nuclear energy as a step towards a future economy fueled by domestically produced, emission-free hydrogen. All three initiatives clearly support R&D efforts necessary to position nuclear energy as a more viable fuel source that will help the United States sustain its growing energy needs.

The selection of new NERI research projects is based on their relevance to these specific initiatives, and their achievements will play an integral role in the success of the overall NE mission. Chapters 9 through 11 explain the work scope associated with these NERI research areas in more detail and present brief abstracts of the FY 2005 projects that have been awarded under each initiative.

The following graphic summarizes the key features of the NERI program.


VISION
To maintain a viable nuclear energy option to help meet the Nation's future energy needs
GOALS/OBJECTIVES
Address potential technical and scientific obstacles
Advance the state of U.S. nuclear technology
Promote and maintain a nuclear infrastructure
SCOPE OF PROGRAM
Advanced Nuclear Energy Systems
Advanced Nuclear Fuels/Fuel Cycles
Hydrogen Production from Nuclear Power
Fundamental Science
R&D PRIORITIES
Innovative reactors, systems, and components
Hydrogen generation
Alternative power conversion cycles
High-burnup, proliferation-resistant fuels
Advanced digital instrumentation & controls
Supercritical Light Water Reactor fuels
Automation technologies
Corrosion-resistant fuel cladding
Advanced structural materials
Enhanced coolant chemistry
IMPLEMENTATION STRATEGY
Competitive peer-reviewed R&D selection process
Individually managed projects
Collaborative research efforts
Continuous DOE oversight
RESULTS
U.S. nuclear leadership
Innovative technologies
Nuclear infrastructure development
Advances in fundamental science
Nuclear public awareness
Worldwide partnerships

4. NERI's Accomplishments

This section discusses the program's progress in attracting research proposals, awarding annual R&D funding, and facilitating the successful completion of the NERI-funded projects.

Project Awards

In FY 1999, DOE's NERI program received 308 R&D proposals from U.S. universities, national laboratories, and industry in response to its first solicitation. The initial FY 1999 procurement was completed with the awarding and issuing of grants and laboratory work authorizations for 46 R&D projects. The proposed research included participants from 45 organizations. Thirty-two of the projects involved collaborations of multiple organizations. Eleven foreign R&D organizations also participated in NERI collaborative projects. The duration of these annually funded projects was one to three years, with the majority lasting three years. The total cost of these 46 research projects for the three-year period was approximately $52 million. A summary of these projects was included in the *NERI 2002 Annual Report.*

In FY 2004, scientific and technical development advanced through the continuation of research projects that began in FY 2001 and FY 2002. In FY 2001, 13 NERI R&D projects were awarded, involving 23 U.S. and 5 foreign R&D organizations. In FY 2002, 24 projects were awarded, involving 32 U.S. and 5 foreign R&D organizations. In FY 2003 and 2004, no new NERI projects were awarded. In FY 2005, 35 new projects were awarded, involving 37 U.S. and 2 foreign R&D organizations.

Funding for NERI projects in FY 1999 through FY 2004 was appropriated annually by Congress in the Energy and Water Development Appropriations Act. Table 1 provides a history of NERI funding and its distribution.

Figure 1 illustrates the cumulative total of research projects awarded for FY 2001 and FY 2002 in the three major R&D areas. Through 2004, over $110 million had been awarded to fund NERI research projects, with a total of $47.9 million being allocated to the recently completed FY 2001 projects and the ongoing FY 2002 projects. Figure 2 shows the distribution of the $47.9 million among the national laboratories, U.S. universities, and industry.

DOE has not funded foreign participants in existing projects as part of the NERI program. Their participation has been supported by the foreign organizations interested in the research being conducted. Although the PIs have been responsible for soliciting such support,

Year	Total Funding	New Funding	Continuing Funding
FY 1999	$19 million	$17.5 million for 46 new awards	N/A
FY 2000	$21.5 million	$2.7 million for 10 new awards	$18.8 million for projects begun in FY 1999
FY 2001	$27.1 million	$5.7 million for 13 new awards	$20.8 million for projects begun in FY 1999 and FY 2000
FY 2002	$22 million	$10 million for 24 new awards	$9.3 million for projects begun in FY 2000 and FY 2001
FY 2003	$17.5 million	No new awards funded	$15.8 million for projects begun in FY 2001 and FY 2002
FY 2004	$11 million	No new awards funded	$10.2 million for projects begun in FY 2002

Table 1. NERI funding and distribution history.

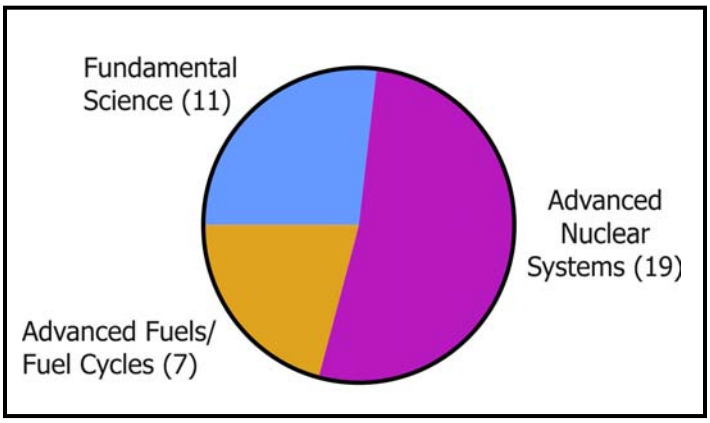


Figure 1. Division of FY 2001 and FY 2002 NERI projects by R&D area.

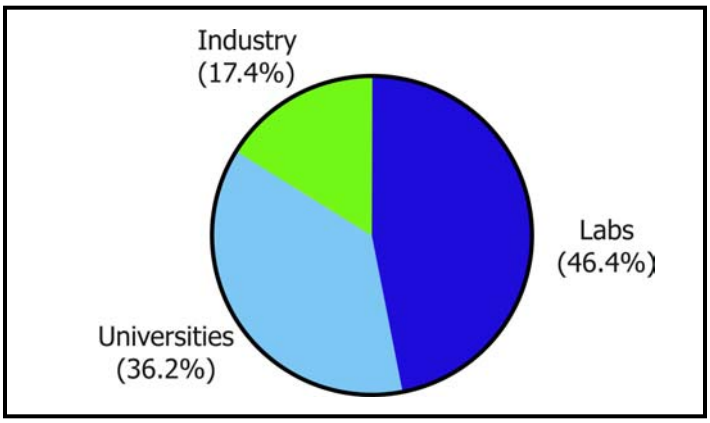


Figure 2. Distribution of NERI research funds in FY 2004.

foreign participation in NERI projects is contingent upon DOE approval.

In FY 2005, 35 new project awards were given to 25 different universities. NERI funding totaling over $21 million will be provided for this new research over the next three years. In FY 2005, over $10.4 million in funding will be provided; $2.5 million from congressional appropriations for NERI and the remainder from FY 2004 and 2005 Generation IV, AFCI, and NHI programs. Figure 3 shows a breakdown of these projects by R&D program area.

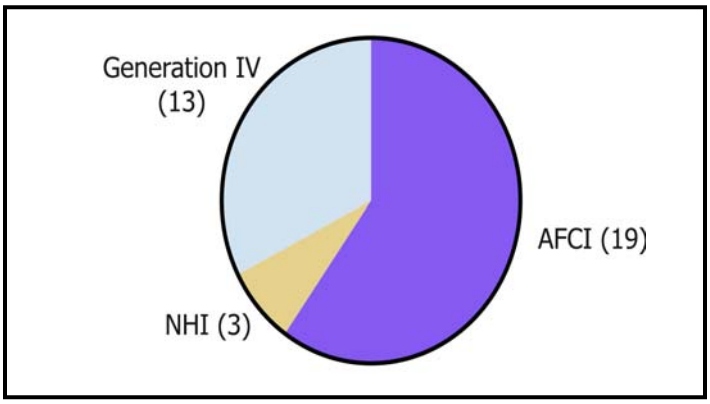


Figure 3. Division of new NERI projects by R&D program area.

NERI Participants

NERI research participants in FY 2004 included 17 U.S. universities, 10 national laboratories, 14 private businesses, and 6 foreign organizations. The participating organizations are provided in the following tables.

U.S. Universities

Georgia Institute of Technology
Iowa State University
Kansas State University
Massachusetts Institute of Technology
North Carolina State University
Oregon State University
Penn State University
Purdue University
Texas A&M University
The Ohio State University
University of Arizona
University of California-Berkeley
University of Florida
University of Michigan
University of Nevada, Las Vegas
University of South Carolina
University of Wisconsin

U.S. Department of Energy Laboratories

Ames Laboratory
Argonne National Laboratory
Brookhaven National Laboratory
Idaho National Engineering & Environmental Laboratory
Lawrence Livermore National Laboratory
Los Alamos National Laboratory
Oak Ridge National Laboratory
Pacific Northwest National Laboratory
Sandia National Laboratories
Westinghouse Savannah River Technology Center

Industrial Organizations

Burns & Roe Enterprises
Duke Engineering Services (now Framatome ANP, Inc.)
Entergy Nuclear, Inc.
Florida Power & Light
Framatome ANP, Inc.
Gamma Engineering Corporation
General Electric Global Research Center
General Atomics
Pacific Southern Electric & Gas
Panlyon Technologies
PBMR (Pty.) Limited
SRI International
Westinghouse Electric Company LLC
(n,p) Energy, Inc.

International Collaborators

Atomic Energy of Canada Limited (Canada)
Commissariat a l'Energie Atomique (France)
Japan Nuclear Cycle Development Institute (Japan)
University of Rome (Italy)
University of Tokyo (Japan)
VTT Manufacturing Technology (Finland)

FY 2001 Project Results

This year marked the scheduled completion of the FY 2001 NERI projects. Based on noted accomplishments, it is clear that NERI's stated goals and objectives are being met. Collaborative efforts between the public and private sectors have resulted in significant enhancements in the U.S. nuclear science and engineering infrastructure, especially in the areas of human and physical resources and capabilities. These efforts, coupled with those of I-NERI, the Generation IV, AFCI, and NHI, have helped to revive the Nation's leadership role in international nuclear R&D. The technology advances will allow the United States to maintain a competitive position in overseas energy markets and a future domestic market.

By accomplishing their research objectives, these NERI projects have addressed and helped overcome a number of potential technical and scientific obstacles to the long-term, future use of nuclear energy in the United States. A brief overview of each of the 12 completed FY 2001 projects is presented below. More detailed information on these projects is contained in the project summaries located in Chapters 7 through 9.

Feasibility Study of Supercritical Light-Water-Cooled Reactors for Electric Power Production

The purpose of this NERI project was to assess the feasibility of the U.S. supercritical water-cooled reactor (SCWR) design and explore alternatives to this design. The project was organized into three main tasks: fuel-cycle neutronic analysis and reactor core design, fuel cladding and structural material corrosion and stress corrosion cracking studies, and plant engineering and reactor safety analysis. Researchers investigated metallic and oxide fertile fuels in a fast-spectrum SCWR and completed neutronic evaluations for two different SCWR fuel assembly designs. Additionally, researchers built and operated a high-temperature autoclave containing a constant extension rate mechanical test device, testing a variety of austenitic and ferritic-martensitic alloys. This project also focused on plant engineering and reactor safety analysis. Researchers defined the boundaries of this analysis, performed simplified calculations to provide an initial characterization of the design, adapted the analyses code for the analysis of supercritical water, implemented new correlations in computer code, analyzed temperature profiles for various core geometries, and concluded that the design should use a geometric configuration. They concluded that the SCWR, due to its very large enthalpy rise along the core, is sensitive to small variations in the flow-to-power ratio; thus, the SCWR core design remains a major feasibility issue. Researchers performed an evaluation of an innovative safety concept for the SCWR, developed a process for examining the SCWR pressure vessel structural consequences of thermal transients, examined hot nozzle isolation and recommended a design for a thermal sleeve, developed a conceptual design of a fuel assembly for the SCWR, and identified the design's main feasibility issues. (Project 01-001)

High-Performance Fuel Design for Next Generation PWRs (Annular Fuel Project)

Massachusetts Institute of Technology led a team that examined the potential for using high-performance, advanced fuel for pressurized water reactors (PWRs). This fuel would accommodate a substantial increase in core power density while simultaneously providing comparable (or larger) thermal margins than current PWRs. This team completed thermal hydraulic and mechanical design analysis, researched reactor core physics and fuel management, completed fuel fabrication studies and economic analyses and optimization, and evaluated fuel performance. The fuel fabrication studies focused on two major fuel fabrication routes, the vibration packing (VIPAC) technique and traditional pellet sintering. Researchers concluded the following: (1) annular fuel with reactor coolant flowing internal and external to the fuel rods can accommodate a 50-percent power uprate in terms of departure from nucleate boiling ratio and a proportional flow increase; (2) higher fuel enrichments are required to obtain the 50 percent increase in power; (3) fabrication of annular fuel elements can be performed by commercial techniques, at a reasonable cost, and should not be an inhibiting factor in the commercial introduction of annular fuel to improve the performance of existing light water reactors; (4) Gen II and Gen III+ plants can increase the power output by 50 percent using annular fuel, which improves the rate of return on invested capital (note: Gen II plants would require new reactor internals, steam generators, and reactor coolant pumps); and (5) the computer performance modeling code, FRAPCON, was modified and confirmed that high burnups are achievable with annular fuel. (Project 01-005)

Miniature, Scintillation-Based, In-Core, Self-Powered Flux and Temperature Probe for HTGRs

The objective of this project was to develop a miniature, scintillation-based, in-core, self-powered neutron flux and temperature probe. The probe is intended to be generally

applicable to any reactor technology, but is specifically designed for the higher temperatures of high-temperature gas reactors. Researchers produced and demonstrated all of the required hardware, completed reactor testing of the developed system, and measured the relationship between the fluorescent light intensity versus reactor power. The project succeeded in performing basic radiation thermometry measurements using a light guide and optical spectrum measurement. (Project 01-039)

Generation IV Nuclear Energy System Construction Cost Reductions Using Virtual Environments

This project demonstrated the feasibility and effectiveness of using full-scale, virtual reality simulation in designing Generation IV nuclear plants using Immersive Projection Display (IPD) technology. Researchers performed five tasks: (1) constructed and tested a full-scale virtual mockup of a selected plant area; (2) studied the effectiveness of the IPD technology to evaluate design and construction in the selected area; (3) developed methods to perform a maintenance task within the virtual mockup; (4) studied training and maintenance using the virtual mockup; and (5) evaluated the lessons learned during the previous activities as they apply to Generation IV designs. Accomplishments included the creation of two mockups: one of a room in the auxiliary building of an AP 600 reactor (chosen for its design complexity) and the other of the entire AP 1000 containment building (chosen to evaluate the capacity of the IPD technology). The design and construction team at Westinghouse considered the spatial correlation and visualization to be realistic. Using the first mockup, the team performed equipment installation sequences 30 percent quicker than using normal construction techniques. The virtual mockups were successfully used to demonstrate training and maintenance applications. In addition, researchers created two mockups of the Pebble Bed Modular Reactor cavity. This last task demonstrated that the IPD technology is compatible with design technologies being used to develop Generation IV reactor designs. (Project 01-069)

On-Line NDE for Advanced Reactor Designs

The key objective of this effort was to develop an on-line structural health monitoring system that will replace or augment current outage-based maintenance practices while ensuring the current level of safety. Specifically, researchers worked to determine the most appropriate, economically valuable inspections; optimize the design parameters to simplify inspections; develop the concept of a built-in structural integrity monitoring system to be integrated into Generation IV reactor designs; evaluate and characterize the performance of conceptual sensor systems by utilizing physics-based simulation models and enhance these models; and select sensor types and materials, find their compatibility with hazardous environments, and examine their possible degradation. Researchers identified several prototypical Generation IV reactor components that can receive the maximum benefit from on-line monitoring with the appropriate non-destructive evaluation (NDE) methodologies and generated conceptual designs of on-line NDE methods. Though the project is complete, researchers believe that the electromagnetic acoustic transducer ultrasonic guided wave is a critically needed technology that merits further investigation. (Project 01-076)

Random Grain Boundary Network Connectivity as a Predictive Tool for Intergranular Stress-Corrosion Cracking

Intergranular stress corrosion cracking (IGSCC) is one of the most pervasive degradation modes in current light water reactor systems and is likely to be a limiting factor in advanced systems as well. In structural materials, IGSCC arising from the combined action of a tensile stress, a "susceptible" material, and an "aggressive" environment has been recognized for many years and the mechanisms widely investigated. Recent work has demonstrated that by sequential thermomechanical processing, properties such as corrosion, IGSCC, and creep of materials can be dramatically improved. This project proposed an explanation for these observations: that the effect of grain boundary engineering is to break the connectivity of the random grain boundary network through the introduction of low energy, degradation-resistant twins and twin variants. The team aimed to verify the mechanism by which sequential thermomechanical processing ameliorates IGSCC of alloys relevant to nuclear reactor applications and to prescribe processing parameters that can be used in the manufacture of IGSCC-resistant structures. Researchers developed boundary tracking algorithms, prepared test microstructures, and completed stress corrosion cracking testing. The major accomplishments of this project were determining that random boundary network connectivity is a major driver of IGSCC in austenitic alloys, developing a predictive tool for ranking IGSCC performance of these alloys, and establishing thermo-mechanical processing parameters to be applied in the manufacture of IGSCC-resistant materials. (Project 01-084)

Supercritical Water Nuclear Steam Supply System: Innovations in Materials, Neutronics, and Thermal-Hydraulics

This project investigated three problems preventing the practical application of a supercritical light water reactor (SCWR). Resolving these problems could improve the efficiency of a supercritical water reactor by 33 percent. Innovations in materials, neutronics, and thermal-hydraulics of SCWRs were the focus of this project. Researchers completed work on surface characterization and analysis of an assortment of candidate materials (stainless steel, Zircaloy-2, and Inconel alloys) under both the optimally heat-treated condition and plasma surface-treated conditions. Testing was performed under representative SCWR conditions to determine surface-related degradation such as corrosion, oxidation, and wear. Stainless steel exhibited the best corrosion resistance by far. Researchers also performed neutronics analyses of alternative fuel cycles which focused on reactor coolant density effects at supercritical conditions to develop a mixed spectrum SCWR. The high coolant density of the outer zone ensured a thermal neutron spectrum, while the reduced coolant density and tight fuel lattice pitch of the inner zone produced a fast neutron spectrum. Lastly, researchers performed a thermal-hydraulics study focusing on heat transfer and stability issues in natural circulation test loops. Stable flow behavior was documented in the Argonne test loop, although the computational model developed at the University of Wisconsin predicted instabilities. Discrepancies between the two were attributed to both the modeling of friction factor distribution in the loop due to lack of experimental data and an assumption of an infinitely large reservoir. To bridge the gap between the experiments and numerical results, researchers developed a linear stability analysis to better predict instability behavior. (Project 01-091)

Testing of Passive Safety System Performance for Higher Power Advanced Reactors

The primary objective of this project was to assess the AP 1000 passive safety system core cooling performance under high decay power conditions for a spectrum of breaks located at a variety of locations in the APEX test facility. In addition, researchers compared predictions of the integral system data to the measured results using a reactor system's thermal hydraulic computer code and provided insights into new passive safety system concepts that could be used for Generation IV higher power reactors. Researchers successfully completed 11 integral system tests and published several reports on their experimental results. The United States Nuclear Regulatory Commission (NRC) granted a Final Design Approval (FDA) to Westinghouse for the AP 1000 advanced reactor design based in part on the APEX-1000 test data. (Project 01-094)

Reactor Physics and Criticality Benchmark Evaluations for Advanced Nuclear Fuel

This project's objectives were to design, perform, and analyze critical benchmark experiments to validate reactor physics methods and models for fuel enrichments greater than 5-weight percent ^{235}U. Benchmarks included critical boron concentration, burnable absorber worth, and relative pin powers. The project was performed at Sandia National Laboratories using modified unirradiated Pathfinder fuel stored at Pennsylvania State University. Phase I included submitting a license application to the NRC for transporting the fuel in WE-1 shipping containers, which required significant technical effort. The primary focus of Phase 2 was analyzing the proposed experiments using current reactor physics computer codes, fabricating new fuel rods using fuel from the Pathfinder elements, and placing the fuel inside aluminum cladding. The experimental facility was assembled and operating procedures were written. Phase 3 included a fuel cycle analysis, which is complete and will be published in *Nuclear Technology*. This project has received a 12-month, no-cost extension through August 31, 2005, to complete the Phase 3 benchmark experiments. (Project 01-124)

Fundamental Understanding of Crack Growth in Structural Components of Generation IV Supercritical Light Water Reactors

In this project, researchers aimed to (1) increase their understanding of the fundamentals of crack growth in stainless steels and nickel-based alloys at supercritical temperatures (SCT); (2) assess the influence of SCTs on the electrochemistry of different types of corrosion processes; (3) measure a material's susceptibility to stress corrosion cracking and other forms of environmentally assisted degradation at SCTs; (4) use these measurements to interpret rate-limiting processes for corrosion during a reactor's lifetime; and (5) use computer analysis (FRASTA) to develop insights into crack nucleation times and crack growth rates. Researchers designed and built a test system for electrochemical and fracture mechanics studies in supercritical water. Candidate structural materials were ranked for their susceptibility to environmentally assisted degradation, and researchers found a correlation between

measurable oxide film properties and the susceptibility for degradation in austenitic steels. Researchers found the chromium (Cr) concentration to decrease in the outer surface layer of the film although the Cr concentration of the film exceeded that of the base metal. Additionally, researchers explored the potential use of the Controlled Distance Electrochemistry (CDE) technique for building in-situ sensors for monitoring water chemistry. (Project 01-130)

New Design Equations for Swelling and Irradiation Creep in Generation IV Reactors

The objectives of this project were to develop physical models for radiation-induced microstructural changes in structural materials to be used in Generation IV nuclear reactors and to derive from these physical models the constitutive laws for void swelling, irradiation creep, stress-induced swelling, and changes in mechanical properties. Researchers tested and validated both the microscopic models and the macroscopic constitutive laws, then incorporated the detailed microstructural models they developed into a computer code reproduces the observed voice swelling curves for neutron-irradiated steels. As a result of their research, researchers proposed a new constitutive equation for void swelling that captures the essential features of the detailed models and can readily serve as a design equation for nuclear reactor design studies. (Project 01-137)

Development and Validation of Temperature-Dependent Thermal Neutron Scattering Laws for Applications and Safety Implications in Generation IV Nuclear Reactor Designs

The purpose of this project was to (1) critically review neutron scattering laws for various moderators, (2) select experimental data sensitive to neutron spectra to generate benchmarks, (3) update scattering models by introducing new developments in thermalization theory and condensed matter physics, (4) benchmark the results against experimental data, and (5) perform a validation experiment for neutron slowing down as a function of temperature in graphite. Accomplishments included generating thermal-neutron scattering cross sections for graphite, beryllium, ZrH_2, ThH_2, and BeO and also completing the set-up for the graphite validation experiment at Oak Ridge Electron Linear Accelerator (ORELA). Because ORELA has recently been plagued by maintenance problems and is inoperable, the project has been extended until the validation experiment can benchmark the thermal scattering libraries for graphite as a function of temperature. (Project 01-140)

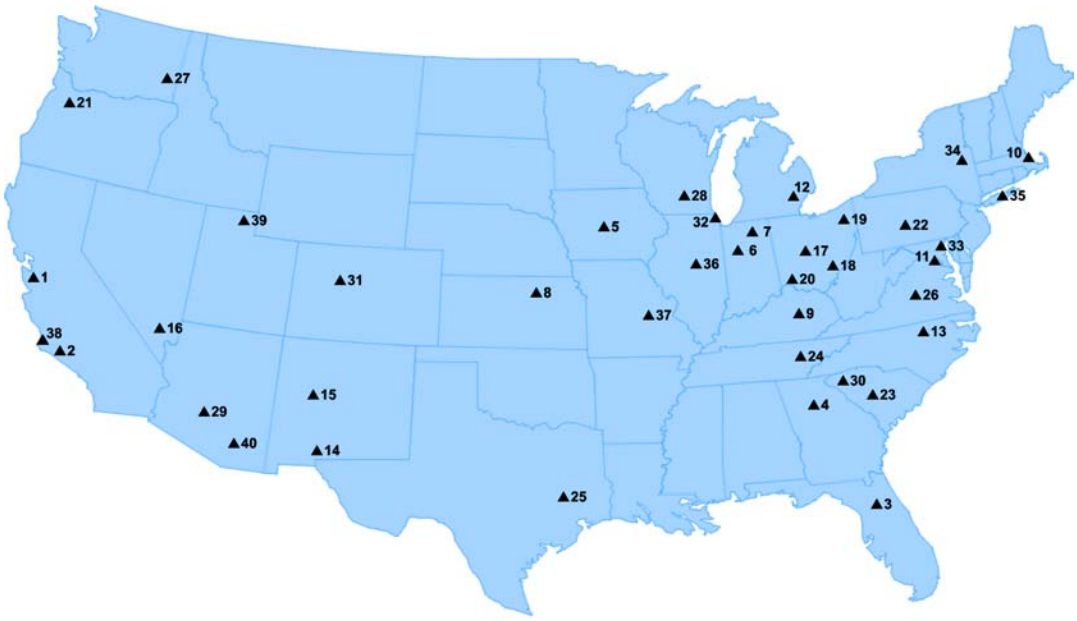
5. NERI's Impact on U.S. University Nuclear-Related Research

Nuclear Programs

One of NERI's long-term goals is to improve the nation's nuclear science and engineering infrastructure in order to maintain the country's leading position in nuclear energy research. One way of achieving this long-range goal is to focus on the classrooms and research laboratories of colleges and universities around the United States. By cultivating research partnerships among U.S. universities and DOE, NERI is helping educational institutions across the country stay at the forefront of science education, advancing the important work of existing DOE's Office of Nuclear Energy, Science and Technology R&D programs, and benefiting future nuclear industry and national laboratory endeavors by training the next generation of nuclear scientists.

University Involvement

DOE believes that funding creative research ideas at the nation's universities and colleges, as well as at national laboratories and industry, helps solve important issues that the private sector is unable to fund alone due to the high-risk nature of the research and/or the extended period before a return on investment is realized.



UNIVERSITY KEY

1. University of California, Berkeley
2. University of California, Los Angeles
3. University of Florida
4. Georgia Institute of Technology
5. Iowa State University
6. Purdue University
7. University of Notre Dame
8. Kansas State University
9. University of Kentucky
10. Massachusetts Institute of Technology
11. University of Maryland, College Park
12. University of Michigan
13. North Carolina State University
14. New Mexico State University
15. University of New Mexico
16. University of Nevada, Las Vegas
17. Ohio State University
18. Ohio University
19. University of Akron
20. University of Cincinnati
21. Oregon State University
22. Pennsylvania State University
23. University of South Carolina
24. University of Tennessee
25. Texas A&M University
26. University of Virginia
27. Washington State University
28. University of Wisconsin
29. Arizona State University
30. Clemson University
31. Colorado School of Mines
32. Illinois Institute of Technology
33. Johns Hopkins University
34. Rensselaer Polytechnic Institute
35. State University of New York
36. University of Illinois
37. University of Missouri-Rolla
38. University of California, Santa Barbara
39. Utah State University
40. University of Arizona

Figure 4. Locations of universities and colleges participating in NERI projects.

Participants in NERI's initial planning workshop recommended that NERI be viewed as a "seed program," where new nuclear-related technological and scientific concepts could be investigated. Based on this philosophy, NERI has provided universities and colleges with a competitive, peer-reviewed research program that allows faculty and students an opportunity to conduct innovative research in nuclear engineering and related areas. Since 1999, 28 U.S. universities have received research awards as lead investigators and/or collaborators. Of the 93 FY 1999 through FY 2002 projects conducted in the NERI program, 75 percent involved U.S. colleges and universities. The FY 2005 NERI projects will add an additional 12 new U.S. universities to this list, raising the number of universities that are involved or have been involved in NERI research to 40. Figure 4 provides a map and complete listing of these universities and colleges.

Student Participation

One great success of NERI and other DOE programs is that nuclear-related educational opportunities at the universities have significantly increased. Universities have benefited from available research dollars which have served as incentives for new student recruitment. As a result of this involvement, student interest in nuclear engineering has been revitalized. In 1998, only 500 students were enrolled in U.S. universities seeking degrees in nuclear engineering; today, 1,500 students are enrolled in nuclear engineering programs. Since the inception of NERI in 1999, 406 of these students have directly contributed to NERI-sponsored research.

This student participation includes all levels—undergraduates and students performing masters and doctorate level work. Figure 5 shows the distribution of participation between undergraduate, graduate, and doctorate students that have worked on NERI projects since the program's inception. In addition, numerous post-doctoral fellows at universities have been involved in NERI research projects.

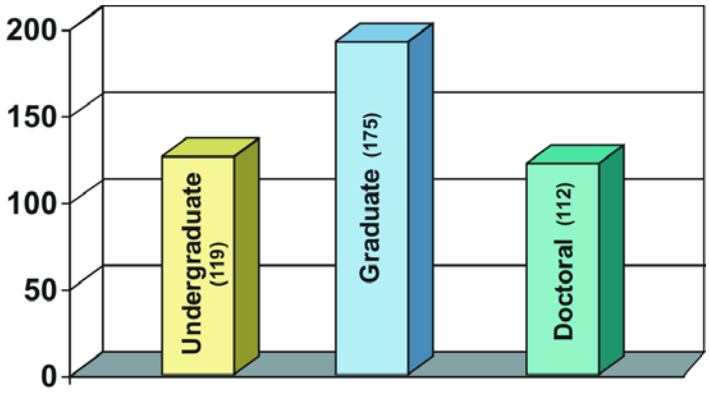


Figure 5. NERI student participation profile through 2004.

In FY 2004 alone, 55 undergraduate, 49 graduate, and 54 doctoral students participated in NERI R&D. Over the past few years, graduates of these programs have had higher than normal grade point averages, showing that these programs are training highly qualified individuals who will sustain the future growth of the nuclear power industry.

NERI has provided U.S. universities and colleges an opportunity to work closely with industry and DOE national laboratories and it has introduced these researchers to other nuclear-energy-related government programs. In addition to research on the Advanced Fuel Cycle Initiative, the Generation IV Nuclear Energy Systems Initiative, the Nuclear Hydrogen Initiative, and the Nuclear Power 2010 program, NERI's activities are coordinated with other relevant energy research programs in the DOE Office of Science, the DOE Office of Energy Efficiency and Renewable Energy, and the U.S. Nuclear Regulatory Commission. Furthermore, the Department leverages NERI program resources by encouraging no-cost collaboration with international research organizations and nuclear technology agencies. In this way, universities are also given the opportunity to gain experience with international research interests and capabilities.

6. Advanced Nuclear Energy Systems: Project Summaries

This research area includes 18 research projects, of which 6 were awarded in FY 2001 and 12 in FY 2002. It includes the investigation and preliminary development of advanced concepts for reactor and power conversion systems. These systems offer the prospect of improved performance and operation, design simplification, enhanced safety, and reduced overall cost. Projects may involve innovative reactor, system, or component designs; alternative power conversion cycles for terrestrial applications; new research in advanced digital instrumentation and control and automation technologies; hydrogen production from nuclear reactors; and identification and evaluation of alternative methods, analyses, and technologies to reduce the costs of constructing, operating, and maintaining future nuclear power plants.

Additionally, this area includes research projects to improve the intrinsic proliferation-resistant qualities of advanced reactors and fuel systems. Possible technology opportunities and subjects of investigation include alternative proliferation-resistant reactor concepts, systems that minimize the generation of weapons-usable nuclear materials (e.g., Pu-239) and waste by-products, and systems that increase energy extraction from the utilization of plutonium and other actinide isotopes generated in the fuel.

Projects under this research area specifically address the characteristics, feasibility, safety features, proliferation-resistance, and economic competitiveness of advanced reactor systems. Research focuses on advancements in light water reactor technology to achieve higher performance, as well as on development of other higher temperature advanced reactor designs for higher efficiencies.

Other research in this area focuses on developing compact or modular reactor designs suitable for transport to remote locations and alternative energy production or co-generation reactor applications. Desirable features include long-lived reactor cores that minimize or avoid the need for refueling, and concepts that maximize fuel burn-up or employ advanced energy conversion technology.

Finally, this area includes research and development to identify and evaluate innovative concepts for producing hydrogen using nuclear reactors. This research includes investigating hydrogen generation processes that are compatible with advanced reactor systems and developing integrated systems that are efficient and cost effective.

NUCLEAR ENERGY RESEARCH INITIATIVE

Directory of Advanced Nuclear Energy Systems Project Summaries

NUCLEAR ENERGY RESEARCH INITIATIVE

Feasibility Study of Supercritical Light-Water-Cooled Fast Reactors for Actinide Burning and Electric Power Production

PI: Philip E. MacDonald, Idaho National Engineering and Environmental Laboratory (INEEL)

Collaborators: Jacopo Buongiorno, James Sterbentz, Robert Witt, and Cliff Davis, INEEL; Gary Was, University of Michigan; Luca Oriani, Vefa Kucukboyaci, Lawrence Conway, Bin Liu and N. Jonsson, Westinghouse Electric Corporation

Project Number: 01-001

Project Start Date: August 2001

Project End Date: Completed September 2004

Research Objectives

The supercritical water-cooled reactor (SCWR) is one of the six reactor technologies selected for research and development under the Generation IV program. SCWRs are promising advanced nuclear systems because of their high thermal efficiency (about 45 percent vs. 33 percent for current light water reactors, LWRs) and considerable plant simplification. SCWRs are basically LWRs operating at higher pressure and temperature with a direct once-through cycle. Operation above the critical pressure eliminates coolant boiling, so the coolant remains single-phase throughout the system. Thus, the need for pressurizers, steam generators, steam separators, and dryers is eliminated. The goal of developing the SCWR is to generate low-cost electricity. The SCWR is based on two proven technologies: LWRs, which are the most commonly deployed power generating nuclear reactors in the world, and supercritical fossil-fired boilers, a large number of which are also in use around the world.

The purpose of this NERI project was to assess and determine the feasibility of the reference U.S. Generation IV SCWR design and to explore alternatives to this design. The project was organized into three main tasks:

- Fuel-cycle neutronic analysis and reactor core design,
- Fuel cladding and structural material corrosion and stress corrosion cracking studies, and
- Plant engineering and reactor safety analysis.

Research Results

Task 1. Fuel-cycle Neutronic Analysis and Reactor Core Design. Researchers investigated metallic and oxide fertile fuels in a fast-spectrum SCWR during Year 1 to evaluate the void and Doppler reactivity coefficients, actinide burn rate, and reactivity swing throughout the irradiation cycle. They also assessed a variety of other core arrangements and moderator types for a thermal-spectrum SCWR during the three years of this project. These designs included solid moderator studies and analysis of an alternative thermal-spectrum SCWR design based on vertical power channels, hexagonal fuel assemblies, and water moderation between the fuel assemblies.

Researchers completed neutronic evaluations for two SCWR fuel assembly designs during Year 3 of this project. The first evaluation was for a 25x25 fuel assembly that used MA956 oxide dispersion steel for the fuel rod cladding, water rod duct, and assembly duct materials. The second was for a 21x21 fuel assembly that used silicon carbide (SiC) for the fuel rod cladding, water rod duct, and assembly duct materials.

The 25x25 fuel assembly with MA956 cladding and duct material contained a 6x6 square array of water rods interspersed uniformly within the UO_2 fuel pin array to increase neutron moderation and assembly reactivity. The assembly exhibited many desirable neutronic characteristics, which included sufficient reactivity to achieve burnups of at least 31.0 GWD/MTU, a strongly negative Doppler coefficient, and a negative void worth for both the coolant and water rods. The assembly also exhibited some characteristics that might complicate the design, such as a wide-spread in the radial enrichments (3.2-12.4 wt% U^{235}) required to flatten the radial power profile and relatively lower reactivity than in LWRs because of the high parasitic neutron absorption of the MA956 cladding and duct material. In addition, the assembly axial power profile exhibited a strong sensitivity to small changes in axial enrichment

that could lead to power oscillations under normal operation if not properly controlled with burnable poisons and control rods. The sensitivity is believed to be primarily due to the interplay between the non-uniform axial water density profiles that affect neutron moderation and the time-dependent axial burnup of the fissile heavy metal.

The 21x21 fuel assembly with SiC for the fuel pin cladding and fuel assembly duct material exhibited better neutronic characteristics than the assembly with MA956 steel structures. For example, this assembly showed a significant increase in core reactivity due to the relatively low parasitic neutron absorption of SiC. This low parasitic neutron absorption in turn translated into significantly higher burnup (41.0 GWD/MTU) when compared to the discharge burnup for the steel assembly (31.0 GWD/MTU). In addition, the SiC assembly exhibited a strong negative Doppler coefficient (-2.5 pcm/°C), and negative void worth for both the coolant and water rods. However, the assembly did require a relatively wide spread in the fuel rod radial enrichment (3.2–12.4 wt% U^{235}) to flatten the radial power profile at beginning-of-life conditions. Under these conditions, it would also require at least a three-zone axial enrichment to flatten and center the unrodded axial power profile about the core midplane at beginning-of-life. As with the MA956 assembly, the SiC fuel assembly axial power profile exhibited a strong sensitivity to small changes in axial enrichments (and therefore burnup).

Task 2: Fuel Cladding and Structural Material Corrosion and Stress Corrosion Cracking. Researchers found the existing data base on the corrosion and stress-corrosion cracking of austenitic stainless steel and nickel-based alloys in supercritical water to be very sparse. Therefore, the focus of this work has been testing the corrosion and stress corrosion cracking properties of candidate fuel cladding and structural materials. During Year 1, researchers built and tested at the University of Michigan a high-temperature autoclave with carefully controlled chemistry and containing a constant extension rate mechanical test device.

During Years 2 and 3, researchers tested a variety of austenitic and ferritic martensitic alloys in this autoclave. Austenitic alloys were tested in deaerated water (dissolved oxygen of the order of few ppb) at temperatures between 400 and 550°C. The researchers determined susceptibility to intergranular stress-corrosion cracking (SCC) by examining the fracture surface and the gage surface and by analyzing cross-sections of the tensile bars. All of the alloys showed varying degrees of susceptibility. These measurements provided a complete description of the cracking behavior. Alloy 625 was the most susceptible, displaying the highest degree of intergranular fracture and some of the deepest cracks, along with a very high crack density. Type 304L stainless steel was the next most susceptible material, showing the deepest intergranular cracks. Alloys 690 and 316L were the least susceptible austenitic alloys from all measures considered: crack density, crack depth, and crack length.

The degree of intergranular SCC of the austenitic alloys increased with temperature. As the temperature increased, crack density decreased but the crack length and depth increased. This resulted in a net increase in the intergranular cracking severity, as measured by the crack length per unit area.

There was also a very strong temperature dependence to the oxidation behavior of the austenitic alloys. The oxidation rate, as measured either by the weight gain or oxide thickness, increased faster with increasing temperature. By 550°C, the austenitic alloy oxide thickness approached 10 μm within a few hundred hours. The predominant feature among all of the austenitic alloy oxides was a two-layer structure consisting of a predominantly iron-rich outer layer and a predominantly chromium-rich inner layer. X-ray diffraction showed that the outer layer was magnetite, Fe_3O_4. The outer oxide on Alloy 690 was probably NiO.

The ferritic-martensitic alloys did not display any evidence of intergranular SCC, as determined by fracture surface and gage surface analysis. They all displayed strain softening and ductile rupture. However, the oxidation rates of the ferritic-martensitic alloys were very high compared to the austenitic alloys. At 500°C, the ferritic-martensitic oxidation rates were a factor of 10 greater than those of the austenitic alloys at the same temperature. These alloys also displayed a two-layer structure, in which the outer layer was identified as magnetite, Fe_3O_4. The oxygen-to-metal (O/M) ratio of the inner layer was closer to hematite, but researchers did not verify the structure of the inner oxide layer. The addition of 100 ppb oxygen to the water at 500°C reduced the total oxide thickness by about 10 percent and slightly increased the O/M ratio. These results were consistent with the objective of combined water chemistry control.

Task 3. Plant Engineering and Reactor Safety Analysis. *SCWR Core Thermal Hydraulic Design Assessment.* The first step in performing the core thermal-hydraulic assessment was identifying the design limits to establish the acceptability of the core design. Once the researchers defined the boundaries of the analysis, they

performed simplified calculations to provide an initial characterization of the design. Based on the results of the simplified analysis, researchers concluded that the SCWR, due to its very large enthalpy rise along the core, is sensitive to small deviations from nominal conditions, especially variations in the flow-to-power ratio. Thus, even small effects due to various hot channel factors (coolant flow channel tolerances, operational variations, etc.) might have a large impact on the peak cladding temperature of some fuel rods. Researchers considered this a major feasibility issue for the SCWR, and decided to perform detailed subchannel analysis of the SCWR core to assess this issue.

The Westinghouse-VIPRE subchannel analyses code was adapted for the analysis of supercritical water, and new correlations that were considered adequate for SCWR analyses were incorporated into the code. Based on the results of sensitivity studies, information was available for a preliminary thermal-hydraulic optimization of the SCWR core design. Researchers then analyzed temperature profiles for various core geometries, with two different objectives: (1) to identify an optimal geometry that minimizes temperature differences between core channels and (2) to confirm and characterize the sensitivity of the temperature profile to the local flow and power ratios. The need to maintain uniform conditions at the exit of the core is dictated by the fact that safety limits need to be verified for the limiting fuel rod, while overall plant performance depends on the average core exit conditions. Thus, a uniform temperature distribution minimizes the "wasted" design margin.

Results of the optimization studies suggested that it is possible to obtain a better temperature profile (hence, lower hot channel factors) by employing a more complex assembly configuration. Based on the results of this study, researchers found that the design should use a geometric configuration with 10 mm outside diameter fuel rods for the coolant channels facing the water rods and at the assembly periphery, 9.5 mm outside diameter fuel rods for the assembly corners, and 10.2 mm outside diameter fuel rods for all other positions. While this study showed a path to obtain an acceptable thermal hydraulic design, it also provided the designer with an important design issue: the flow is extremely sensitive to small variations in the channel flow area. Therefore, rod bowing and even the tolerances in rod dimensions could be crucial in terms of the temperature peaking. This sensitivity, attributed to the large channel enthalpy rise coupled with a region of low-density coolant and high exit velocities, renders the design uncertain.

Based on the results of this study, it appears that the reference SCWR design is not feasible. Although additional design and analysis might allow the recovery of some margin, it is unlikely that an SCWR assembly and core design can be developed that provides acceptable performance (i.e., low enough hot channel exit temperature). Therefore, the SCWR core design remains a major feasibility issue for which a solution has yet to be achieved at this stage of the program.

An Evaluation of an Innovative Safety Concept for the SCWR. Preliminary investigations of the safety characteristics of an SCWR performed by the Idaho National Engineering and Environmental Laboratory and the Westinghouse Electric Company resulted in the development of a novel safety concept for this Generation IV reactor. Previous analyses have shown that the SCWR can meet transient thermal limits for events initiated by loss of main feedwater only if a large-capacity auxiliary feedwater system is actuated rapidly. However, the required rapid initiation of auxiliary feedwater was judged to pose significant technical and economic challenges. Consequently, Westinghouse developed an innovative conceptual design that uses a passive circulation system to mitigate the effects of losing main feedwater. This safety concept utilizes two relatively small, feedwater tanks that store water for reactor cooling during normal operation and provide sufficient cooling capacity to mitigate the effects of losing main feedwater. Main coolant pumps similar to those utilized in advanced light water reactors provide the head required to circulate the flow in the reactor. Although the proposed concept takes advantage of the SCWR once-through, direct cycle concept during normal operation, it allows the establishment of a recirculation path in the system following containment isolation, with an isolation condenser that provides long-term decay heat removal.

Researchers evaluated the safety characteristics of the design for loss-of-flow transients using the RELAP5-3D computer code and obtained acceptable results that confirmed the potential of the design. Specifically, they were able to achieve acceptable short-term results following a loss of flow by adjusting the coastdown characteristics of the main coolant pumps. Additionally, they achieved satisfactory long-term decay heat removal following a loss of flow with 300 or more tubes in the isolation condenser.

Researchers also performed preliminary evaluations of loss-of-coolant accidents. The analysis of an accident initiated by a large cold leg break showed that significantly lower cladding temperatures were obtained after the blowdown peak in the proposed design than in a simple,

once-through design due to the recirculation loop and the added coolant inventory provided by the feedwater tanks. The milder evolution of the accident allowed for a significantly simpler emergency core cooling system design.

Structural Response of SCWR Reactor Pressure Vessel to Thermal Transients. Researchers developed a process for examining the SCWR pressure vessel for structural consequences of thermal transients. This is important because hydraulic transients are often much shorter than the reactor pressure vessel thermal diffusion time. For the case involving a sudden 30°C (54°F) drop in main feedwater temperature, the peak Von Mises stress is about 30 percent higher (65 vs. 50 ksi) than that experienced under static conditions at nominal system pressure. Given the low number of anticipated cycles and the relatively small temperature change, the ASME Code indicates the transient is of no consequence with respect to low-cycle fatigue.

Design of a Thermal Sleeve for the SCWR Hot Leg. The reference SCWR operates at a substantially higher reactor coolant outlet temperature and pressure than existing LWRs. Therefore, the 500°C coolant outlet water must be isolated from the reactor pressure vessel in order to use conventional vessel materials. Researchers examined hot leg nozzle isolation and recommended a design for a thermal sleeve. The recommended thermal sleeve consists of a one-inch thick steel structural insert surrounded by a generous two-inch radial water gap between the thermal sleeve and the reactor pressure vessel outlet nozzle. The outlet nozzles were positioned above the inlet nozzles so that the isolating cold leg flow filled the annular water gap from below. In this way, forced convection augmented natural convection in the gap, and peak vessel temperatures were both low and insensitive to water mass flow rate.

Fuel Assembly Conceptual Design. Researchers developed a conceptual design of a fuel assembly for the SCWR to identify any feasibility issues. There were three main difficulties in the SCWR fuel assembly design: the fuel rod spacer, the assembly top structure, and thermal expansion of the fuel assembly components. The preliminary fuel assembly design addressed all three of these aspects.

NUCLEAR ENERGY RESEARCH INITIATIVE

Miniature, Scintillation-Based, In-Core, Self-Powered Flux and Temperature Probe for HTGRs

PI: David E. Holcomb, Oak Ridge National Laboratory

Collaborators: Don W. Miller, The Ohio State University

Project Number: 01-039

Project Start Date: September 2001

Project End Date: Completed December 2004

Research Objectives

The objective of this project was to develop a miniature, scintillation-based, in-core, self-powered neutron flux and temperature probe. The probe is intended to be generally applicable to any reactor technology, but is specifically designed for the higher temperatures of high-temperature gas reactors (HTGRs). The scintillation assembly consists of a uranium layer placed against a thick film scintillator layer. The fission fragments resulting from neutron interactions in the uranium produce light in the scintillator, which is guided out of the detector using a hollow core optical fiber. Both the converter layer and the scintillator can be segmented. A scintillator of one wavelength is co-located with a lightly ^{235}U-enriched uranium layer, while a scintillator with a different characteristic wavelength is placed against a higher enriched uranium layer (e.g., 2 percent and 4 percent). The scintillators produce different wavelength light which allows independent readout of the scintillation. Neutron flux is indicated by the total amount of light produced by either scintillator; the ratio of light at the different waveband intensities serves as a burn-up monitor. Multi-wavelength pyrometry measures the temperature.

Research Results

The project has now achieved a basic level of success. Researchers measured a scintillation signal that varies directly with reactor power using the developed uranium deposit (including the use of 19 percent ^{235}U enriched uranium), scintillator layer, light guide, reactor test chamber, and photon measurement electronics assembly. The project also succeeded in performing basic radiation thermometry measurements using a light guide and optical spectrum measurement. As the project team did not achieve success until the final measurement sets, they were unable to implement segmented scintillators or uranium deposits, an objective beyond the originally proposed scope.

This project produced all of the required hardware and demonstrated satisfactory performance. The components developed under this project include the uranium-coated nickel disks, scintillation deposits overlaying the uranium, the titanium hollow-core light guide, the silvered glass hollow-core light guide, the coupling between the light guides, the optical fiber and lens system for transmitting light to the photodetector, and the photodetector, data acquisition system, and associated software. Researchers implemented the light guiding scheme and demonstrated its performance in a near-core environment. They also completed integrated system reactor testing and refinement. Project staff developed and demonstrated the scintillation deposition technique and implemented it for

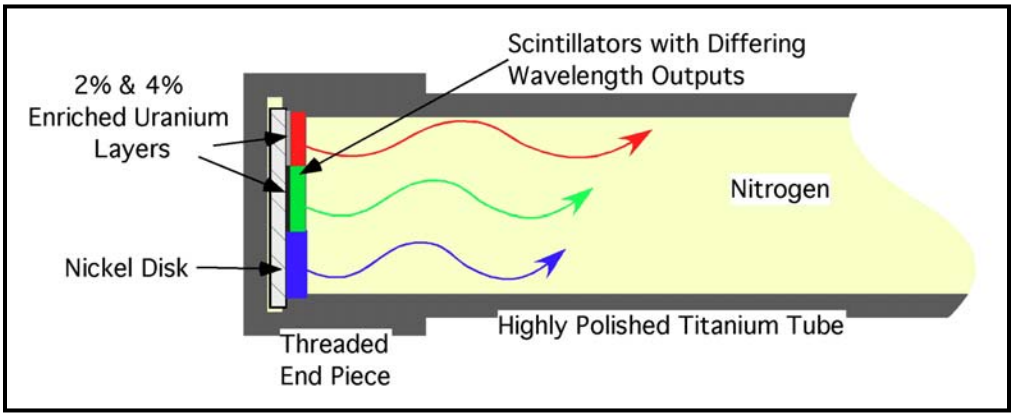


Figure 1. Conceptual layout of scintillation-based neutron flux and temperature probe.

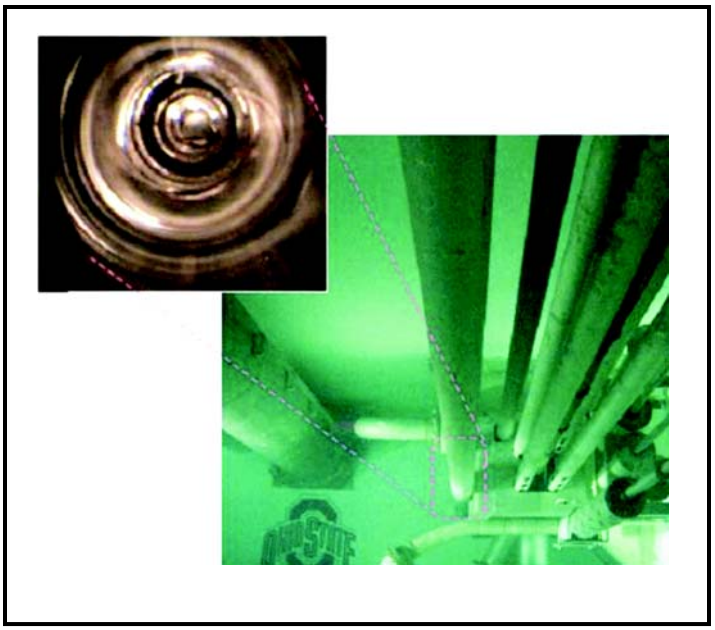


Figure 2. Scintillator disk located at the distal end of a titanium light guide, near the core of The Ohio State University Research Reactor operating at 1 kW.

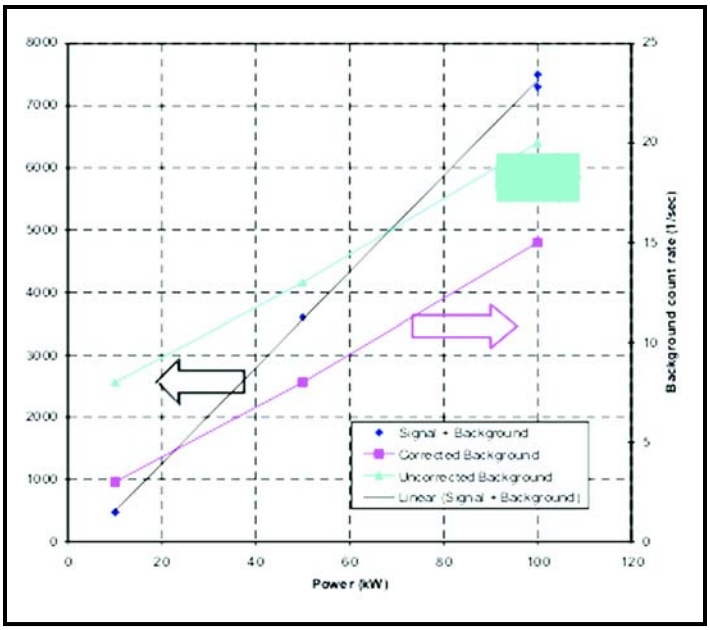


Figure 3. Reactor power versus measured photon count rate.

three different phosphors for in-core deployment. They also developed an electrochemical technique for depositing uranium films onto nickel disks[1] and created both a silvered glass version of the light guide tube and a polished titanium light guide tube for in-core use. Researchers also produced both a wavelength-dependent light measurement system and a pulse-based photon measurement system coupled to the proximal end of the light guide tubes. Finally, they completed a high-temperature, near-core test environment chamber and a gamma dose chamber.

The recent research activity was focused around reactor testing of the developed system. Specifically, project staff measured the relationship between the measured fluorescent light intensity versus reactor power.

1. Maya, B. D. Gonzalez, M. J. Lance, and D. E. Holcomb, Electrodeposition of uranium dioxide films, Journal of Radioanalytical and Nuclear Chemistry, 261(3), 605-607 (2004)

NUCLEAR ENERGY RESEARCH INITIATIVE

Generation IV Nuclear Energy System Construction Cost Reductions Using Virtual Environments

PIs: Timothy Shaw, Anthony Baratta; Pennsylvania State University

Collaborators: Panlyon Technologies, Westinghouse Electric Co., Burns and Roe

Project Number: 01-069

Project Start Date: August 2001

Project End Date: Completed November 2004

Research Objectives

The objective of this project was to demonstrate the feasibility and effectiveness of using full-scale virtual reality simulation in designing future nuclear power plants. Specifically, this project tested the suitability of using Immersive Projection Display (IPD) technology for evaluating the potential cost reductions that can be realized in Generation IV Nuclear Energy Systems installation and construction sequences. The intent was to determine whether this type of information technology can be used to improve arrangements and reduce construction and maintenance costs, as has been done by building full-scale physical mockups.

Development, testing, and evaluation of the virtual environment technology were divided into five tasks. In the first task, researchers created and reviewed a full-scale, virtual mockup of a selected space within an advanced nuclear power plant design for use as an experimental testbed. During the second task, researchers used this testbed to study the effectiveness of the technology to evaluate the design and construction of the selected space. In the third task, researchers developed the methodology and the required tools to perform a prototypical maintenance task using the virtual mockup. Training and performance of maintenance activities using the virtual mockups were studied during the fourth task. Finally, the team investigated the lessons learned during the first four tasks as they apply to a Generation IV design.

Research Results

The first year of the project focused on the development of the full-scale virtual mockup. Three-dimensional CAD models prepared by Westinghouse were successfully converted to a format that could be displayed in Penn State's CAVE immersive virtual environment system, which allowed the models to be projected as full-scale, stereoscopic 3-D images. Two mockups were created: Room 12306 in the auxiliary building of Westinghouse's Advanced Passive (AP) 600 reactor and the containment building of the AP 1000. Room 12306 was chosen as a testbed for the

Figure 1: Full-scale virtual mockup of Limerick training mockup.

virtual mockup due to its relative complexity. The AP 1000 containment mockup was created to evaluate the number of models that could be loaded in a presentation. After the virtual mockups were completed, design and construction personnel reviewed the spaces to determine whether the technology provided sufficient realism. The virtual mockup scored well in categories related to spatial correlation and visualization.

The second year of research focused on evaluating the full-scale virtual reality mockup of Room 12306 to review the installation sequences and determine the benefits of modular construction. Since most of the next-generation nuclear power plant designs are expected to utilize some form of modular construction to speed construction and improve quality, studying the potential methods for developing and analyzing these techniques was an important part of this project.

Researchers performed experiments to test the use of immersive virtual reality for developing and reviewing construction sequences for a next-generation nuclear power plant. These experiments involved creating and reviewing 3-D geometry linked to a construction schedule, called 4-D models, in the CAVE immersive environment. The experiments resulted in a 30 percent reduction in construction time for the room-sized test bed, thus demonstrating that the virtual reality technology could be used to assist construction planners in developing and optimizing installation sequences for equipment in future nuclear power plants, resulting in shorter, more efficient plant construction schedules and increased confidence in the construction schedule.

The third year of research focused on studying the use of virtual mockups for nuclear training and maintenance applications. Researchers divided this activity into two smaller parts. The first part entailed developing new capabilities so that the immersive virtual reality system could be used to simulate a maintenance activity in a nuclear power plant. A radiation dose model was developed to simulate the functions of the survey meters used by radiation protection personnel and the electronic dosimeters worn by radiation workers. Virtual radiation sources of known energy and known source strength can be placed anywhere in the virtual environment. Interaction between the user and the virtual environment was also studied. In the second part of the training and maintenance activities, researchers evaluated the virtual mockup for training activities similar to those typically performed on physical mockups. Three training activities were observed to determine how nuclear utilities use physical mockups in their personnel training programs. For example, the training programs at Perry Nuclear Power Plant, Limerick Generating Station, and Susquehanna Steam Electric Station use physical mockups for training personnel for many skills, including equipment familiarization and reinforcement of proper work practices. Burns and Roe modeled two training programs, one from Limerick and one from Susquehanna, to create virtual mockups. Figure 1 depicts the Limerick mockup. Researchers tested the virtual mockups to determine how effectively they reproduced the tasks performed on the actual physical mockups.

In addition to the training mockups, the researchers created a full-scale virtual mockup of the AP 1000 steam generator compartment to enable Westinghouse to evaluate access and staging requirements for steam generator sludge lancing, a common PWR maintenance task. Westinghouse and Mitsubishi Heavy Industries personnel who reviewed the steam generator mockup noted the accurate portrayal of scale and commented on the

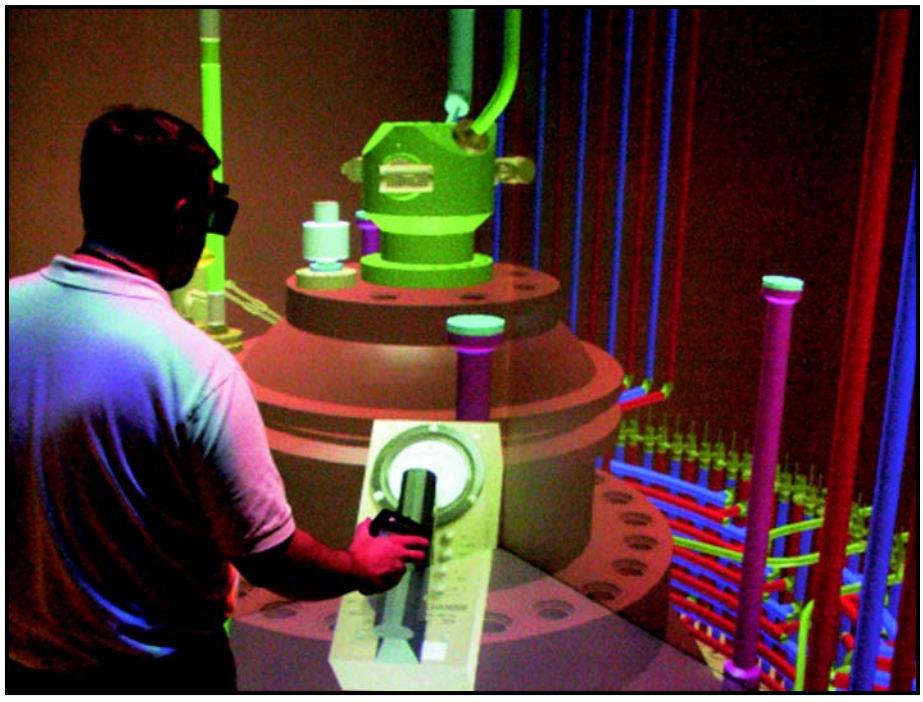


Figure 2: PBMR reactor cavity virtual mockup.

mockup's potential value for familiarization training and work, and for planning for the next generation of nuclear power plants.

In the final task, researchers evaluated virtual mockups of a Generation IV reactor design, the Pebble Bed Modular Reactor (PBMR). Because the designers of the PBMR used the Unigraphics NX CAD suite to create 3-D CAD models of the plant, Pennsylvania State and PBMR evaluated a different data translation process which resulted in the creation of two virtual mockups of the PBMR reactor cavity. One virtual mockup of the complete reactor cavity was created from a single CAD model, while the second mockup was created with fewer parts to present the general layout of the cavity with increased graphical rendering performance. Figure 2 depicts the PBMR reactor cavity mockup. The creation of this virtual mockup demonstrates that it is possible to continue this work with the data being produced for the next generation of nuclear reactors.

NUCLEAR ENERGY RESEARCH INITIATIVE

On-Line NDE for Advanced Reactor Designs

PI: Norio Nakagawa, Ames Laboratory - USDOE

Collaborators: Warren Junker, Westinghouse Electric Company LLC

Project Number: 01-076

Project Start Date: September 2001

Project End Date: Completed August 2004

Research Objectives

The extended refueling interval of Generation IV nuclear reactors creates new maintenance challenges. Current commercial reactors achieve high levels of availability and reliability by employing outage-based maintenance (i.e., performing methodical, periodic, off-line inspections; preventive maintenance; and component repair/replacement during planned refueling periods). Compared to the traditional 18- to 24-month refueling cycle, Generation IV reactors are expected to use extended refueling intervals of four years and beyond. New approaches are required to perform the necessary maintenance activities during power operation. The key objective of this effort was to develop an on-line structural health monitoring system that will replace or augment current outage-based maintenance practices, while ensuring the current level of safety.

The project's specific objectives were to:

1. Determine which inspections are the most appropriate through a comprehensive review of each component of the selected Generation IV reactor design, based on regulatory requirements and commercial reactor experience.
2. Determine which inspections provide the greatest economic benefit when implemented as in-situ monitoring, considering the inspection requirements and reduced off-line inspection opportunities.
3. Optimize mechanical design parameters to simplify inspections.
4. Develop the concept of a built-in structural integrity monitoring system using electromagnetic, ultrasonic, or radiation detectors that can be integrated into the design for Generation IV nuclear power systems.
5. Evaluate and characterize the performance of conceptual sensor systems by utilizing physics-based simulation models.
6. Enhance the capabilities of the simulation models to meet the challenges posed by unique power system environments.
7. Select sensor types and materials, find their compatibility with hazardous environments, and examine their possible degradation.

In summary, the objective of this project was to propose a shift in maintenance practices for future power systems from outage-based maintenance to on-line inspection and monitoring.

Among the various advanced reactor designs considered, the International Reactor Innovative and Secure (IRIS) was selected for investigation because of its significant design maturity. IRIS is currently under development by an international partnership of industry, research organizations, academia, and power producers and is led by the Westinghouse Electric Company.

This project, now complete, identified several prototypical Generation IV reactor components of the IRIS design that can receive the maximum benefit from on-line monitoring with the appropriate non-destructive evaluation (NDE) methodologies. The project team also generated a conceptual design for on-line NDE methods.

Research Results

Task 1. The goal of this task was to identify target applications of on-line NDE sensors and instrumentation, and also identify suitable NDE methodologies for each monitoring need. The task accomplished these goals, as illustrated in Figure 1, which summarizes the identified in-vessel components of IRIS and the potential areas for on-line NDE. Specific components identified for potential on-line NDE applications include: (1) upper head penetration welds, e.g., for instrumentation tubes; (2) pump attachments; (3) steam generator tubes and tube attachments;

and (4) primary coolant flow measurement. They have also identified applicable NDE modalities:

- Coolant flow monitoring by nitrogen-16 (^{16}N) radioactivity measurement
- Steam generator tubing magnetite deposit detection by eddy currents
- Steam generator and the other structural monitoring by ultrasonic guided waves generated by electromagnetic acoustic transducer (EMAT)

In addition, this task made several efforts for the duration of the project period to verify the plausibility of prototype designs for on-line sensor systems and monitoring methodologies. For example, researchers conducted validation experiments to support model-based sensitivity analyses for the ^{16}N on-line coolant flow monitoring and to confirm the EC magnetite detection predictions using deposited magnetite film specimens.

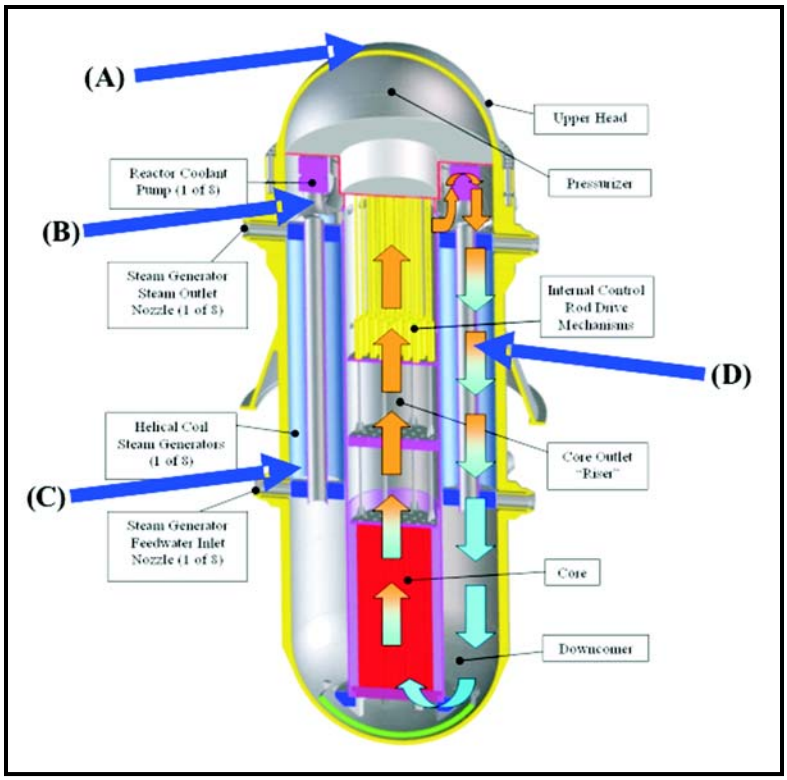


Figure 1. Components inside integral reactor pressure vessel and potential areas for on-line NDE: (A) upper head penetration welds, e.g., instrumentation tubes; (B) pump attachment; (C) steam generator tubes and tube attachment; and (D) primary coolant flow.

Task 2. In this activity, researchers examined the use of radiation detectors for in-vessel component monitoring. Specifically, they examined the reactor coolant flow monitoring method via ^{16}N radiation in detail through calculations and measurements. They calculated ^{16}N activity concentrations and evaluated the impact of background radioactivity in IRIS, determined optimal detector types and location, evaluated a candidate detector for ^{16}N gamma-ray detection, and addressed the effect of ambient temperature conditions of approximately 290°C on detector operation. They concluded that:

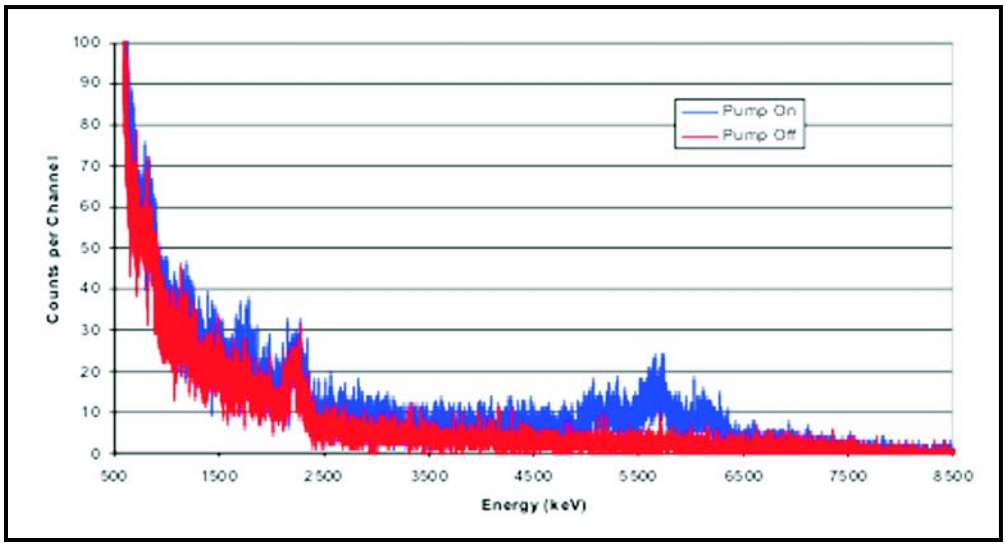


Figure 2. Experimental ^{16}N gamma-ray spectra detected by a Bismuth Germanate detector when circulation pump is on or off. The 6128.6-keV ^{16}N gamma ray plus the single- and double-escape peaks are clearly visible in the pump-on case.

- The total photon flux incident on the detector is less than 6 x 10^3 cm^{-2} sec^{-1}, and most gamma-ray detectors, such as bismuth germanium oxide (BGO) detectors, can handle the resulting count rate.
- A medium-sized detector (such as a BGO 2.54 cm diameter x 2.54 cm) would provide a 300 cps count rate for ^{16}N photons.
- The signal/background ratio of ^{16}N peaks is high because of the high energy of the ^{16}N photons (less than 6 MeV).
- The radiation field at the detector location at the port entrance is approximately 12 mrem/hour; thus, operation of the detector and electronics (preamplifier or phototube) will not be limited by this field.
- Dose rate levels at other points on the wall surface in the steam generator region are low (less than 1 mrem/hour).

- Based on the initial ^{16}N measurements, it can be concluded that even low-resolution gamma-ray detectors (such as BGO) will provide adequate discrimination of ^{16}N gamma rays from the reactor background (Figure 2).

Although ambient temperature effects on the operation of gamma spectrometers such as BGO will be challenging, researchers believe that valuable performance monitoring data should be obtainable through ^{16}N flow-rate measurements.

Task 3. Eddy current sensors are known to work well for detecting magnetite deposits in SG tubing, as well as for stress corrosion cracking detection. Researchers expect on-line extension of this NDE modality to work equally well. Eddy current coils are sufficiently robust to survive the reactor environment. Previously, researchers showed by computation that EC deposit signal characteristics peak at approximately 20 kHz with the optimal phase separation at 27 kHz for encircling coils, and at 28 kHz with the optimal phase separation at 15.5 kHz for side-mount coils. This year, researchers verified the signal behavior by using an encircling coil and a specimen fabricated by deposition. Figure 3 shows the data plots with peaks at essentially the same location. The scale differs because of different coil windings and deposit densities.

Task 4. In this project, on-line ultrasonic applications required shear-horizontal (S/H) guided-wave modes, while the reactor environment precludes the use of the standard alternating-polarity EMAT. As a solution, researchers conceived a novel 3-phase AC EMAT design based on a linear induction motor analogy and using coils only for robustness. Model estimations indicated that 10–20 percent of the total eddy current may couple to S/H modes, which set the maximum efficiency. Transmission/reflection coefficients were calculated, and the results used to determine some of the EMAT design parameters. In conclusion, the project team developed an all-coil S/H-wave EMAT concept that is able to survive the reactor environment, and they have shown, by model calculations, that the sensor may transmit and receive S/H waves at a few percent efficiency.

Researchers completed all the projected developments, except for the transport-equation-based radiography simulation code including the charged-particle sectors. The transport code development became less important after the project initiation because the IRIS core design changed and included an additional shielding/reflector surrounding the core, which precluded the use of very fast prompt gammas and neutrons. In addition, Monte Carlo codes are available that have yielded adequate results for the photon sector. Although the ^{16}N gammas are somewhat hard, they are not so energetic for the charged-particle sectors to modify the present calculations, presumably by a few percent at most.

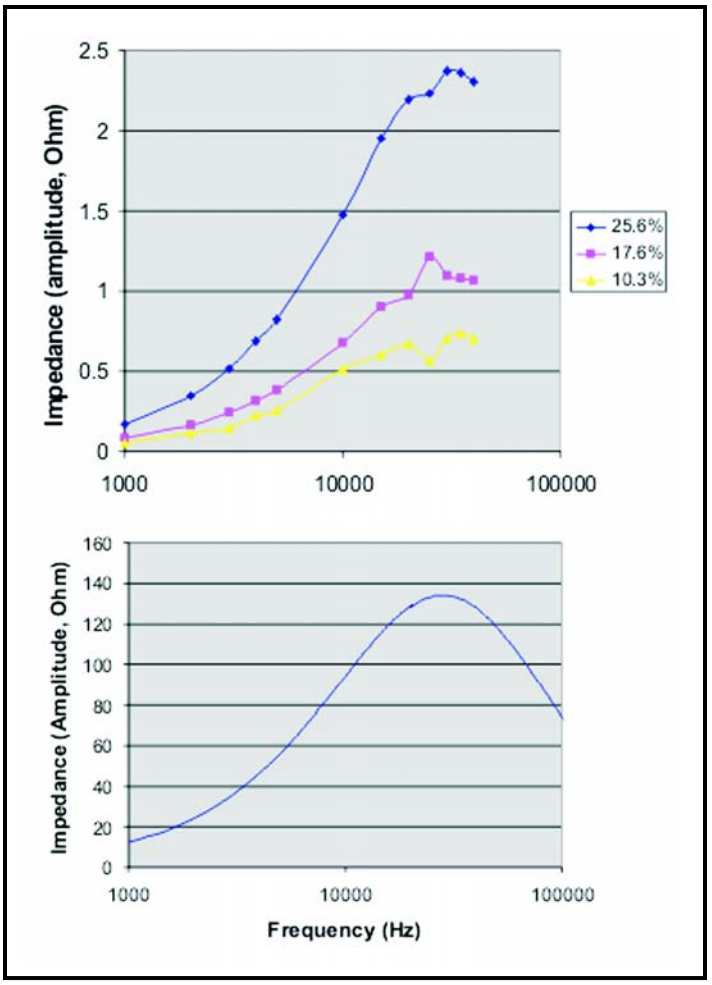


Figure 3. Experimental vs. computational EC deposit signals that peak at the 20-30 kHz range.

The project team believes that the ^{16}N flow monitoring and EC magnetite detection are viable and therefore worthy of further commitment toward actual implementation. The EMAT design requires more laboratory-based verification and possibly further conceptual developments before proceeding forward. Nonetheless, the EMAT UT appears to be a critically needed technology that merits further investigation.

NUCLEAR ENERGY RESEARCH INITIATIVE

Supercritical Water Nuclear Steam Supply System: Innovations in Materials, Neutronics, and Thermal-Hydraulics

PI: Mark Anderson, Michael Corradini, Kumar Sridharan, Paul Wilson, University of Wisconsin

Collaborators: Argonne National Laboratory

Project Number: 01-091

Project Start Date: September 2001

Project End Date: Completed September 2004

Research Objectives

In the 1990s, researchers considered supercritical light-water reactors (SCWR) in conceptual designs. A nuclear reactor cooled by supercritical water would have a much higher thermal efficiency with a once-through direct power cycle and could be based on standardized water reactor components (light water or heavy water). Theoretically, efficiency could be improved by more than 33 percent over that of other water reactors and could be simplified with higher reliability than, for example, a boiling water reactor without steam separators or dryers.

To make such a system technologically feasible, advances are required in high-temperature materials with improved corrosion and wear resistance (cladding and pressure structural boundaries), in neutronics to improve fuel-cycle versatility with these advanced materials, as well as in neutronics and thermal-hydraulics to insure efficient heat removal and passive safety and stability. The research objectives of this project were:

- To improve material compatibility under supercritical conditions by employing innovative, plasma-based, surface modification techniques, including ion implantation, energetic-ion-induced near-surface alloying, and surface homogenization. These techniques were applied to cladding and structural materials with proven bulk properties in order to mitigate surface-initiated degradation phenomena of corrosion, oxidation, and wear under supercritical thermal-hydraulic conditions.
- To identify ranges of alternative fuel cycles, including variations in enrichment, refueling schedules, recycling, and conversion/breeding through performance of neutronics analyses. These analyses, using quantitative metrics, focused on coolant density effects at supercritical conditions to verify passive safety from the standpoint of fuel burnup, flexibility, proliferation resistance, and sustainable development.
- To evaluate heat transfer and flow stability issues associated with coolant density changes for natural and forced circulation of supercritical water by performing thermal-hydraulic studies. Scaled simulated experiments were designed and performed to provide heat transfer and stability data used in developing predictive tools.

Research Results

Task 1. Materials Science and Corrosion in Supercritical Water Corrosion Loop. Researchers completed work on surface characterization and analysis of various candidate materials in two conditions: as received (optimally heat treated condition) and plasma surface-treated. These materials were subjected to corrosion tests in subcritical and supercritical water environments for 7 days at 300, 400, and 500°C. In addition, testing was performed for 3 days at 400°C, 5 days at 400°C, and 6 hours at 500°C. Over 50 samples of various alloys with different plasma treatments were characterized to identify and quantify trends in corrosion mechanisms and propensity for corrosion at the three temperatures. This characterization was performed using scanning electron microscopy (with its associated EDS capabilities) and Auger electron spectroscopy.

The three alloys investigated exhibited quite different corrosion behavior, as noted by high magnification SEM imaging of cross-section and plan-view of the samples, as well as Auger analysis. Stainless steel exhibited homoge-

neous corrosion in the form of a thin oxide film on the surface, although at high magnifications, nanometer-size scale pits were observed. This alloy exhibited the best corrosion resistance by far. Research goals were to quantify the pit and particulate size, density distribution, and oxide film thickness; to assess oxide morphology; and to analyze the effects of erosion, where observed.

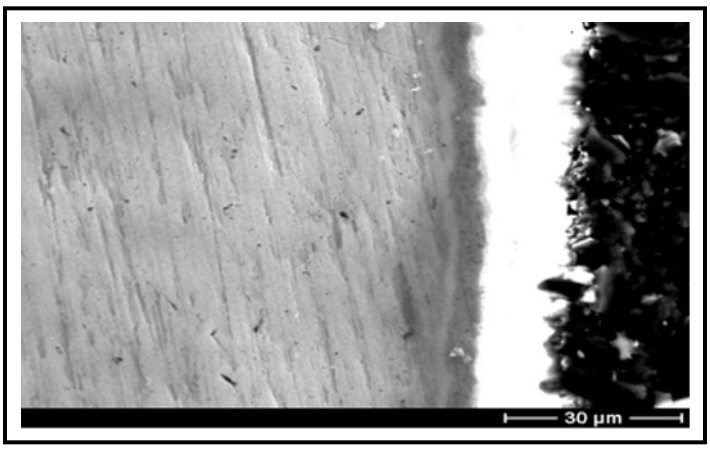


Figure 1. Growth of the oxide corrosion product layer of N^+ ion implanted Zircaloy-2 after exposure to supercritical water at 500°C for 7 days. An oxygen diffusion zone is observed in the underlying alloy. Erosion of the oxide due to the flow of supercritical water was also observed.

Figure 1 shows the oxide film on a N^+ ion implanted Zircaloy-2 sample after being exposed to supercritical water at 500°C for 7 days.

Inconel alloys exhibited an initial tendency to pit in the region of niobium-rich precipitates followed soon after by the formation of oxide particles. Plasma surface treatments affected the density, size, and morphology of these oxide particulates. Figure 2 (a through c) shows oxidation of Inconel 718 samples after exposure to subcritical water at 300°C for 7 days. Examples are shown for the effect of Xe^+ and O^+ ion implantation on the oxidation of Inconel 718.

A consideration in quantifying the thickness of the oxide film corrosion product was the variation in thickness observed in some samples due to the effect of erosion from the flowing supercritical water.

Task 2. Neutronics Analysis of Coolant Density and Cladding Materials on Fuel Cycle. A pitch-to-diameter (P/D) ratio feasibility study has shown that reasonable operating windows exist to accommodate both sufficient moderation and sufficient cooling in the thermal zone of the Mixed Spectrum Supercritical Water Reactor (MS2). In particular, a hexagonal pitch fuel assembly satisfied reactor physics requirements with P/D ratios greater than 1.409, 1.445, and 1.429 for stainless steel, Inconel-600 and -690, respectively. At the same time, with estimated maximum allowable cladding temperatures of

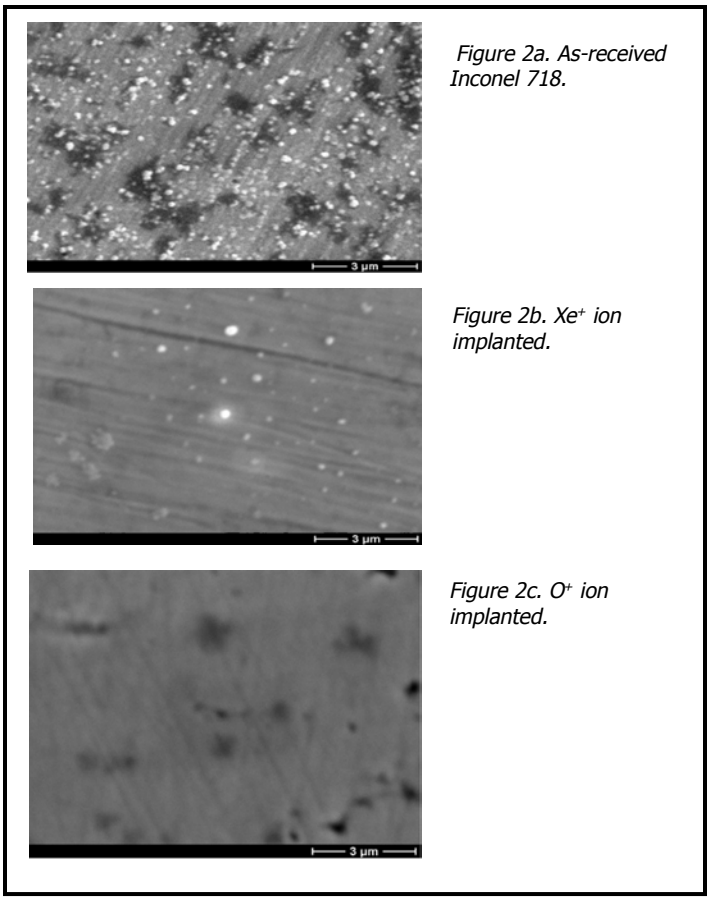


Figure 2a. As-received Inconel 718.

Figure 2b. Xe^+ ion implanted.

Figure 2c. O^+ ion implanted.

Figure 2. Nucleation of oxide on Inconel 718 after exposure to supercritical water at 300°C for 7 days: (a) as-received, (b) Xe^+ ion implanted, and (c) O^+ ion implanted.

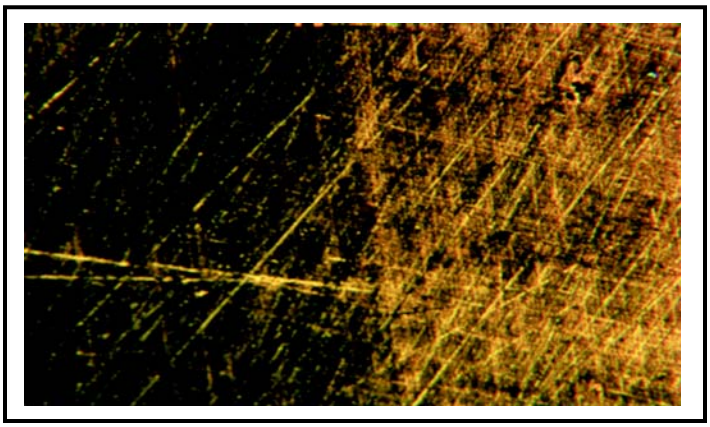


Figure 3. Synergistic effects of erosion and corrosion on C^+ ion implanted Inconel 718 alloy after exposure to supercritical water at 500°C for 7 days. Left side of the sample was the non-reflective oxide deposit (thicker) and the right side showed a reflective metallic surface (thinner). SEM examination showed that the oxide particulates on the region with a metallic appearance were considerably less dense.

873 K and 973 K for stainless steels and Ni-based alloys, respectively, sufficient cladding cooling was achieved for P/D ratios less than 1.492 and 1.563, respectively. The Ni-based alloys offered the most promising alternative from the standpoint of reactor physics and thermal-hydraulics.

Based on the study for the feasible P/D ratio windows, a mixed spectrum supercritical water reactor was developed. The high coolant density of the outer zone ensured a thermal spectrum, while the reduced coolant density and tight lattice pitch of the inner zone resulted in a fast spectrum. The core characteristics such as operating conditions, power density, thermal efficiency, etc., were very similar to other supercritical water reactors. The MS2 core had 25 void fuel assemblies in the inner zone to enhance the neutron leakage and 48 shielding assemblies between the inner and outer zones.

Regarding safety parameters, the moderator temperature reactivity coefficient of the startup MS2 core maintained a negative value from the beginning to the end of the cycle (-125 to -99 pcm/K). Also, the control rods scram worth is 1.6 percent or more negative than the required reactivity for reactor shutdown. However, additional safety analyses are required to confirm that the control rod scram worth is greater than the minimum shutdown margin. From fuel cycle analysis, it is observed that the MS2 concept is capable of stabilizing Pu, Np, and Am (the PNA). During the PNA multi-recycling, the fissile content in the PNA vector degraded from 64.2 to 57.7 percent for 5 recycles, but the desired cycle lengths were ensured by increasing the uranium enrichment of the inner fuel from 6.0 to 6.5 percent. As expected, PNA is burned and bred in inner and outer zones, respectively. The net mass balance of PNA was 47 kg in cycle 5, which is negligible in comparison to the total PNA mass of the fresh fuel in the inner zone (approximately 3,600 kg). Thus, the PNA content was easily stabilized around 15 percent. However, the discharge mass of Cm and higher actinides increased up to 9.9 kg in cycle 5, which would affect the decay heat and radiotoxicity level of the repository for several decades.

Task 3. Natural Circulation Heat Transfer and Flow Stability Studies. In recent years, a growing interest has been generated in investigating the flow instability phenomenon in natural circulation loops under supercritical conditions. This phenomenon is relevant to some of the innovative designs proposed for the Generation IV reactor concept. Supercritical fluid is expected to be susceptible to various kinds of instabilities, similar to those occurring in a two-phase fluid, due to broad variations in its thermodynamic and physical properties near the pseudocritical point. Yet its single-phase characteristics could result in interesting and possibly different stability behavior. A few experiments have been conducted in a rectangular supercritical carbon-dioxide ($SCCO_2$) natural circulation loop at Argonne National Laboratories (ANL) in order to verify the stability margin, as suggested by some previous investigators (Chatoorgoon 2001). A computational model based on the SPORTS formulation has been developed at UW Madison, which provided a good basic simulation tool for the steady state and stability analysis of one-dimensional natural circulation flow. Although the model predicted development of instabilities for the $SCCO_2$ loop, which had good agreement with some previous work, the experiments conducted at the ANL the $SCCO_2$ loop exhibited stable behavior under similar conditions. The discrepancies between the two were attributed to two major factors: (1) modeling of friction factor distribution in the $SCCO_2$ due to the lack of experimental data, and (2) an assumption of a presence of an infinitely large reservoir in the loop that maintains constant inlet conditions to the heater at all times. In order to bridge the gap between the experiments and the transient numerical results, a linear stability analysis was developed to better predict the instability behavior.

NUCLEAR ENERGY RESEARCH INITIATIVE

Testing of Passive Safety System Performance for Higher Power Advanced Reactors

PI: José N. Reyes, Jr., Brian G. Woods, Oregon State University (OSU)

Collaborators: Westinghouse Electric LLC

Project Number: 01-094

Project Start Date: August 2001

Project End Date: Completed July 2004

Research Objectives

The primary objective of this project was to assess the AP1000 passive safety system core cooling performance under high decay power conditions for a spectrum of breaks located at a variety of locations in the APEX test facility at Oregon State University. Researchers compared predictions of the integral system data to the measured results using a reactor systems thermal hydraulic computer code. In addition, the researchers provided insights into new passive safety system concepts that could be used for Generation IV higher power reactors.

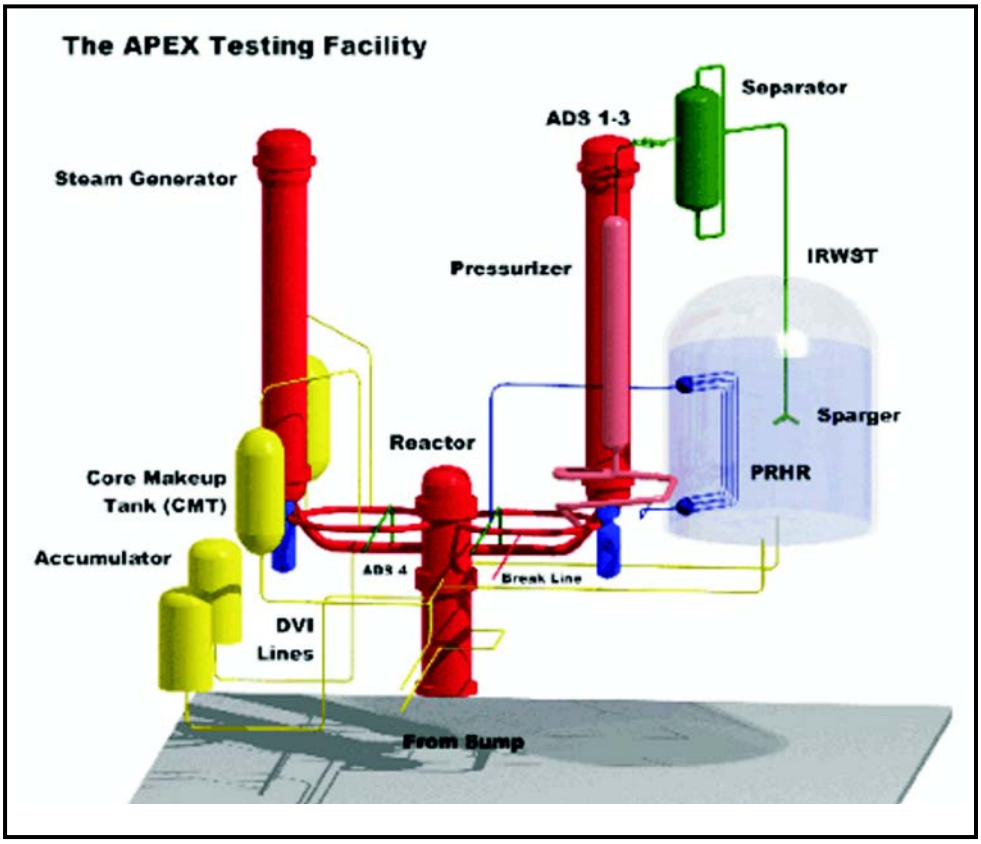


Figure 1. APEX-1000 integral system test facility at OSU.

Research Results

Over the duration of this project, researchers met their primary objective of performing testing to assess the performance of the AP 1000 passive safety systems. They successfully conducted eleven integral system tests, and published test summary and acceptance reports for each. In addition, they documented the results of this project in a scaling analysis report, facility design report, and a final report.

Figure 1 shows the APEX-1000 integral system test facility as modified for this project. It is a one-quarter height scale test facility that includes a complete primary system and all of the passive safety systems and their corresponding safety actuation logic. DOE, Westinghouse, and the U.S. Nuclear Regulatory Commission (NRC) have audited the test facility to assess its adequacy for use in plant certification.

Table 1 shows the eleven integral system tests that have been successfully conducted in the APEX-1000 facility. The tests consisted of Design Basis Accident scenarios specifically selected to examine the important operational characteristics of the AP 1000 passive safety systems, including two blowdown transition tests and two Automatic Depressurization System (ADS) characterization tests. Researchers have transmitted all of the data obtained to Westinghouse for their review and analysis as part of the AP 1000 plant certification process.

Table 1. Completed APEX-1000 integral system tests.

Test ID	Description	Date
OSU-AP1000-01 (DBA-01)	AP1000 DEDVI with 3 of 4 ADS 4	27-Feb-2003
OSU-AP1000-02 (TR-01-D)	AP1000 100 psia Steady-State Entrainment	18-Mar-2003
OSU-AP1000-03 (DBA-02)	AP1000 DEDVI with 3 of 4 ADS 4	1-May-2003
OSU-AP1000-04 (DBA-03)	AP100 DEDVI with 3 of 4 ADS 4	15-May-2003
OSU-AP1000-05 (DBA-04)	AP1000 2" CL4 Break with 3 of 4 ADS4 and Degraded Containment Sump	1-Jul-2003
OSU-AP1000-06 (TR-02)	AP1000 Full Pressure ADS 4 Blowdown w/ 3 of 4 ADS4	1-Jul-2003
OSU-AP1000-07 (ADS-01-D)	ADS 4 parametric study at IRWST water level for 18 inches & 0 inches at Low Decay Power	10-Dec-2003
OSU-AP1000-08 (ADS-02-D)	ADS4 Characterization	6-Feb-2004
OSU-AP1000-09 (DBA-03R-D)	AP1000 Doubled-ended DVI w/ 3/4 ADS 4	15-Apr-2004
OSU-AP1000-10 (DBA-05-D)	Inadvertent ADS Actuation	13-May-2004
OSU-AP1000-11 (DBA-06-D)	AP1000 1/2" CL4 Break w/ 3 of 4 ADS4	3-Jun-2004

Researchers completed testing and analyses in July 2004. On September 13, 2004, the NRC granted a Final Design Approval (FDA) to Westinghouse for the AP 1000 advanced reactor design, based in part on the APEX-1000 test data.

NUCLEAR ENERGY RESEARCH INITIATIVE

Engineering and Physics Optimization of Breed and Burn Fast Reactor Systems

PI: Michael Driscoll, Massachusetts Institute of Technology

Collaborators: Argonne National Laboratory-West, Idaho National Engineering & Environmental Laboratory

Project Number: 02-005

Project Start Date: September 2002

Project End Date: September 2005

Research Objectives

The goal of this project is to develop a practical implementation of breed and burn operation in hard spectrum fast reactors. In the present context, "breed and burn" (B&B) refers to cores in which reload fuel has a significantly lower enrichment than that required to sustain criticality, for example, ≤5 $^{w}/_{o}$ U^{235}. In the ideal case, recycling is not required (but neither is it precluded) to achieve natural uranium utilization that is significantly higher than current light water reactor (LWR) units. High utilization leads to competitive fuel cycle economics and the practicality of commercializing fast reactors without deploying new reprocessing facilities.

Although the general breed and burn concept dates back to the late 1950s, interest in this concept has been sporadic because of the unresolved challenges this concept presents, such as the need for a large, low-leakage core with minimal non-heavy-metal fuel diluents, as well as the need for high power density and burnup to accelerate and sustain fissile buildup in order to quickly achieve an equilibrium fuel cycle.

In contrast to the contemporary effort in Russia on a lead-cooled reactor of this type, this project centers on gas-cooled reactor technology, due in part to extensive technical synergism with the high-temperature thermal reactor designed for hydrogen production, a high priority Generation IV Nuclear Energy Systems Initiative design option.

Research Progress

The second year of this project's efforts was devoted to completing the selection of options open to consideration. Table 1 summarizes the final design features that were selected. Most of the plant-related issues follow directly from the requirements of providing competitive economics and assuring ultra-reliable shutdown and emergency core cooling within the stringent constraints imposed by the physics performance of a B&B reactor.

Table 1. Reference design features of breed and burn GCFR concept.

Core	Design	Comments:
Fuel:	UC or UN-15	UO_2 not viable neutronically
		Reaction of UN and UC with CO_2 precludes its use as coolant
Clad:	ODS	ODS may be able to resist creep adequately up to ≈ 700ºC
Configuration:	Tube-in-Duct; Vented; Orificed	Lower fuel T at increased fuel fraction; venting eliminates ΔP across clad
Coolant:	He@10 MPa, Indirect Cycle; Core ΔTc = 380°C, Exit T≈600°C	He is inert chemically, used in thermal HTGRs
Thermal-Hydraulics:	AXIAL Peaking Factor = 1.45 Radial Peaking Factor = 1.77, Power Density 130 W/cc	Orificing reduces circulator power by factor of ≈ 2
Burnup:	150 MWd/kg Over 18 EFPY	ρ > 0 @ ≈ 20 MWd/kg ρ peaks @ ≈ 80 MWd/kg
Plant		
Power Cycle:	Rankine 2400 MW_{th}	Allows ≈ 380°C ΔTc across core, which reduces coolant flow rate, hence circulator power
Reactor Vessel:	Prestressed Cast Iron Vessel (PCIV) or Prestressed Concrete Reactor Vessel (PCRV)	PCIV is modular, more T resistant than concrete, accommodates large core, envelopes IHX and shutdown loops PCRV is proven in GCR service (but at lower P)
Shutdown Cooling System: (combined shutdown & emergency)	• 3 x 50% capable forced convection loops • Water-boiler heat sink	PRA-guided design supports this selection (basically same No. loops as GCFRs of the 1970's) Natural convection alone suffices if P≥12 atm (5 if CO_2 injected)
Containment:	PWR type sized to keep post-LOCA pressure ≤ 5 atm	Combined with CO_2 injection this permits decay heat removal solely by natural convection

Because of fuel neutronics constraints, the only practical options for fuel are a metal clad uranium nitride (UN^{15}) or uranium carbide (UC) fuel. These options dictate the use of helium as the coolant to avoid fuel-coolant chemical reactions. Because a high fuel volume fraction is required, researchers chose an indirect Rankine cycle—as in the General Atomics (GA) gas-cooled fast reactor (GCFR) of the 1970s—which allows a high core coolant temperature rise and a lower mass flow rate, pressure drop, and circulator power than other options.

Researchers demonstrated a neutronically feasible core design using pin-type UN^{15} fuel. Figure 1 shows the reactivity history of a core having 46 $^{w}/_{o}$ fuel which reloads 5 $^{w}/_{o}$ U^{235} enriched fuel into the core with an average startup enrichment of 8.2 $^{w}/_{o}$, in which one-sixth is refueled during each shutdown.

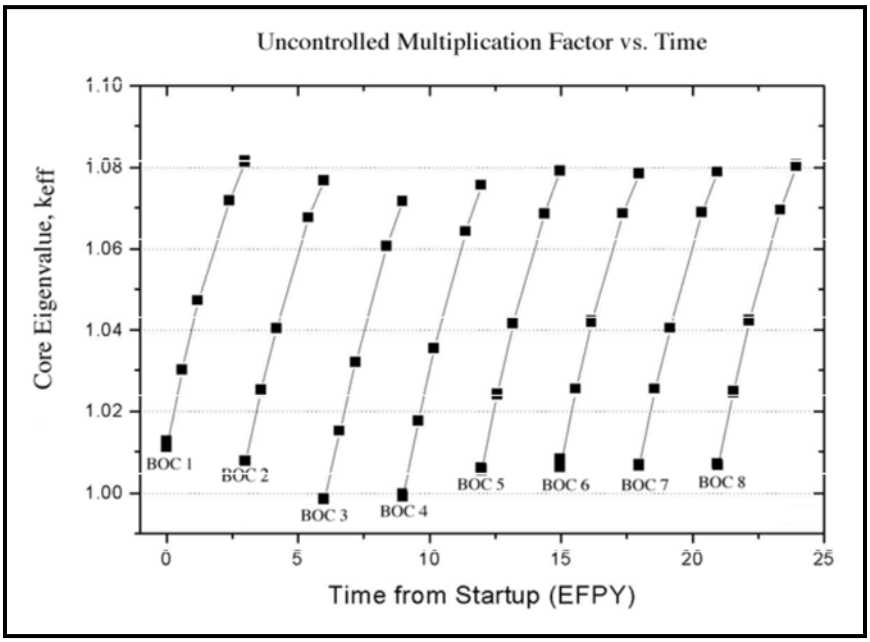


Figure 1. Reactivity history of successive reloads for baseline B&B GCFR core.

Fuel venting, as in the GA GCFR of the 1970s, has considerably ameliorated fuel endurance challenges principally associated with high-temperature clad creep. However, the need for further optimization, including heat transfer enhancement in the upper half of the core, is revealed by first-round assembly steady state and transient calculations.

Planned Activities

In Year 3, researchers will complete performance assessment of the breed and burn concept. The principal emphasis will be on qualifying UC fuel for B&B service in place of UN^{15}, because of the uncertain, but likely high, cost of the separated ^{15}N isotope (which is only 0.368 percent of total nitrogen). UC is also an attractive option because it has been used more often as a fast reactor fuel. The attendant neutronic penalty will require compensatory tradeoffs between achieving the highest practicable fuel volume fraction, and probable re-insertion of refabricated fuel for a second burnup campaign.

Researchers will carry out more detailed analysis of the vented tube-in-duct fuel assembly under both steady-state and transient/accident scenarios. This type of assembly appears to be a key means of enabling the high fuel fraction, power density, and burnup goals essential to B&B service.

The decision late in Year 2 to adopt a Rankine power cycle, in lieu of the supercritical CO_2 Brayton power conversion system under evaluation at MIT for Generation IV applications, removes a wide variety of balance-of-plant issues from the agenda. There remains, however, the issue of providing ultra-high reliability shutdown decay heat removal—the challenge of all GCFR concepts.

The final goal of this project is to assess the economic viability of the B&B GCFR in a future, sustainable nuclear economy.

NUCLEAR ENERGY RESEARCH INITIATIVE

Evaluation of Integral Pressurizers for Generation IV PWR Concepts

PI: David Felde, Oak Ridge National Laboratory

Collaborators: Westinghouse Electric Company

Project Number: 02-018

Project Start Date: September 2002

Project End Date: September 2005

Research Objectives

The objective of this project is to build a knowledge base that will enable designers to better appraise the pros and cons of possible pressurizer designs for integral primary system reactors (IPSR). As part of this process, collaborators will map the performance of integral pressurizers as a function of their design parameters (gas content, vapor volume, and interface with the primary system). Based on a detailed analysis of pressurizer performance, researchers expect to propose new design solutions that will simplify the reactor system and allow it to withstand more severe accident scenarios.

Research Progress

A Computational Fluid Dynamics (CFD) model (Figure 1) was developed for use with the FLUENT computer code, which is a widely used code deemed appropriate for studying mixing phenomena, flow fields, and heat transfer problems in complex geometries, as well as transport of species. A number of analyses were performed with this model.

- Researchers performed steady state calculations of the flow field in the pump region with all reactor coolant pumps in operation, as well as for the case with one pump off, in order to determine flow velocities at the entrance of the pump annular region and to evaluate asymmetries that may be produced by turning off one pump. These effects are important for calculating the mass exchange at the interface between the primary system and pressurizer.
- They performed calculations of the coupled pump and pressurizer regions to determine the degree of flow mixing with and without gravity effects. Because of the geometry size, only a relatively coarse grid model was built. Results obtained for the heat flux through the separating (divider) plate indicated high heat flux values, particularly underneath the plate. Literature data that indicated lower heat transfer coefficients for similar geometries and differences with the model results are being investigated. They also initiated a more detailed assessment of the heat transfer mechanism through the plate.
- They determined the heat transfer coefficient on both sides of the divider plate (over and under). In order to study flow in the pump annular region under the plate, they developed a fine mesh model (structural mesh)

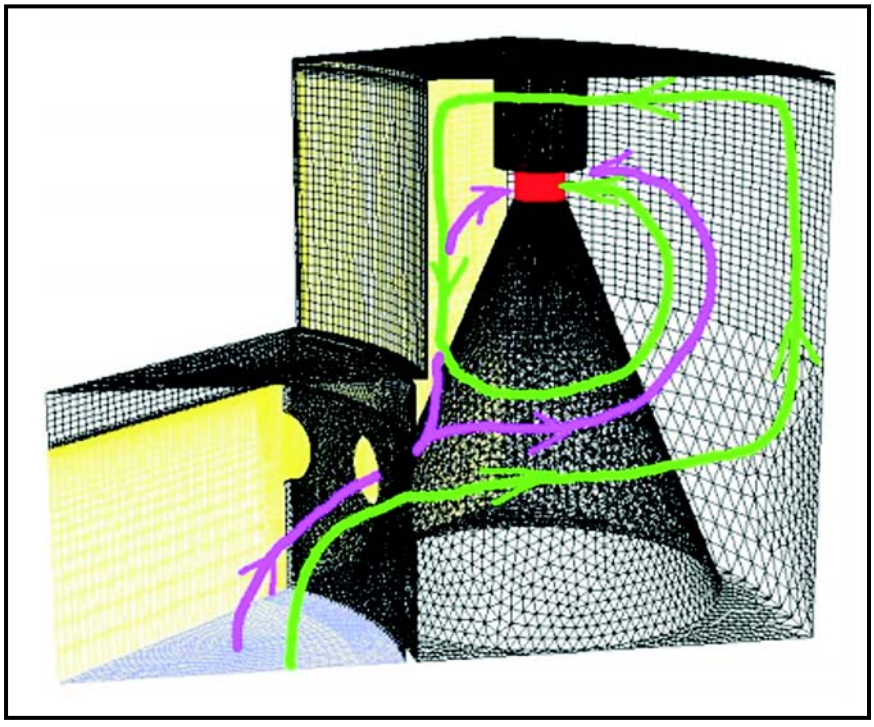


Figure 1. Pressurizer CFD model of pump plenum.

accounting for the specifics in the near-wall region. Next, they performed sensitivity studies on the applied turbulence model and calculated large turbulent intensity flows in the pump annular region, leading to high heat transfer coefficients. These results will serve as boundary conditions when examining the flow in the pressurizer liquid volume (over the plate).

Pressurizer Testing. Two major areas of interest were selected for pressurizer testing: mixing in the liquid volume and evaluating pressurizer dynamics.

- *Mixing Test.* The primary goal of the mixing test is to determine the degree of mixing in the pressurizer volume. It is equally important for both pressurizer types (gas and steam) to have small radial and axial temperature gradients in the liquid part. The gradients are created because of the thermal contact with the primary coolant. In the case of the steam pressurizer, cooling occurs, while in the case of the gas pressurizer, heating is taking place. A test facility is being designed by ORNL with scaling assistance by Westinghouse to further explore and validate these phenomena.
- *Pressurizer Dynamics Test.* The International Reactor Innovative and Secure (IRIS) design incorporates a new type of internal pressurizer having specific proportions with a shallow liquid phase and large free water and dome surface areas. Researchers expect this gas type pressurizer design to provide an improved transient response. However, as available transient response data is for the classical "tube" type pressurizer, the new geometry will require additional testing.

Westinghouse performed the scaling of the experimental facilities. Non-dimensional groups were determined for both the real pressurizer and test facility. A renormalization of the scaling groups is under way that takes into account the planned tests to be performed. Westinghouse is also developing a testing program in accordance with the project milestones as well as with their own plans for testing components of the IRIS reactor.

Planned Activities

Researchers plan to complete the test facility design and fabrication, followed by execution of the test program in the project's third year. They continue modeling two-phase flows, particularly the boiling caused by the pressurizer heater. They will develop a 2-D model to study the heat transfer caused by condensing the vapor produced by the heater in the sub-cooled surrounding water. CFD calculations to model the boiling process in the pressurizer liquid space is a challenging task that will require extensive computational efforts. In the longer term, researchers will perform parametric studies of position and size of the surge holes. Analysis of various flow conditions in the riser is necessary in order to define their impact on the magnitude of the parasitic flow.

NUCLEAR ENERGY RESEARCH INITIATIVE

Nuclear-Energy-Assisted Plasma Technology for Producing Hydrogen

PI: Peter Kong, Idaho National Engineering and Environmental Laboratory

Collaborators: None

Project Number: 02-030

Project Start Date: October 2002

Project End Date: September 2004

Research Objectives

The objective of the project was research and development of a nuclear-energy-assisted plasma process to produce hydrogen carrier materials for use in a hydrogen-based transportation system. The focal point was to develop a plasma electrolysis technology to convert sodium metaborate into sodium borohydride using nuclear-energy-generated electricity to operate the plasma reactor. Plasma electrolysis to convert sodium metaborate to sodium borohydride is a new idea.

Sodium borohydride is a safe and concentrated hydrogen carrier compound that can store an impressive amount of hydrogen. One liter of 44-weight percent sodium borohydride solution at 1 atmosphere can release about 130 grams of hydrogen, more than other sources of hydrogen. For example, cryogenic liquefied hydrogen has a hydrogen density of 70 gm/lit, and hydrogen gas pressurized to 6,000 psi has a density of only 36 gm/lit. Rare-earth-nickel alloys can store hydrogen up to a density slightly higher than liquid hydrogen but still quite a bit less than sodium borohydride. However, this alloy is very expensive and more difficult to handle than a liquid. The borohydride solution is much easier and safer to manage than pure hydrogen in liquid or high-pressure gas form, and the current gasoline distribution infrastructure for automobiles can be easily converted to dispense "sodium borohydride fuel" for hydrogen vehicles.

No known technology currently exists to economically produce sodium borohydride from sodium borate. A successful nuclear-power-assisted plasma technology to convert sodium borate to sodium borohydride fuel suitable for transportation will have a long-term positive economic benefit to the nuclear power industry. During peak operation, nuclear power reactors will generate electricity to meet commercial demand, and during off-peak operation, the nuclear reactor will supply electricity and nuclear process heat to produce sodium borohydride. Producing sodium borohydride during off-peak hours will in turn increase the demand for the nuclear industry.

Research Progress

This research project tested a new technology concept, nuclear-energy-assisted plasma electrolysis, to produce hydrogen carrier materials. The estimated equivalence between gasoline and sodium borohydride on a BTU basis is 10.46 pounds of $NaBH_4$ to one gallon (7 pounds) of gasoline. The energy density of sodium borohydride makes it an attractive alternative fuel for the transportation industry. However, there is no economical technology available for large-scale production of this material for transportation application.

The focal point of this research was to develop the most viable plasma technology to convert sodium borate to sodium borohydride using nuclear-energy-generated electricity and/or heat to run the process. The original concept, plasma electrolysis, has shown no feasibility for sodium borate conversion to sodium borohydride using water as the reactant. The Gibbs reaction free energy study confirmed that the originally proposed reaction,

$$NaBO_2 + 2H_2O \longrightarrow NaBH_4 + 2O_2,$$

was not thermodynamically favorable. Thermodynamic studies indicated that two alternative technologies might have conceptual feasibility. The first was a high-temperature gas phase plasma reaction using hydrocarbon and/or water as the reactant to produce sodium borohydride. In bench-scale investigations, this high-temperature plasma concept had shown positive feasibility to convert sodium borate to borohydride in a single step. The second alternative was a

low-temperature solid-state reaction using water, hydrogen, or hydrogen/water as reactants to produce sodium borohydride. Bench-scale investigation of this concept also demonstrated the feasibility of converting sodium borate to borohydride in a single step. Since the temperature for the reaction in the second technology concept is below 1,000°C, the concept has the potential to directly apply the heated helium gas from the gas-cooled nuclear reactor to heat the reaction.

Plasma Electrolysis. Besides unfavorable thermodynamics for the original reaction to produce sodium borohydride, two other significant factors led to the failure of plasma electrolysis. The first factor was lack of a stable cathode for hydrogen generation suppression. Tellurium (Te) was identified to have hydrogen suppression properties during electrolysis; however, the metal was unstable and disintegrated during the early phase of electrolysis. Figure 1 shows the disintegration of a Te cathode under electrolysis. The cathode showed no sign of hydrogen evolution. The cathode disintegration was much more severe under a plasma operation because the voltage was substantially higher than in a conventional electrolysis. Later, the cathode failure modes were identified as chemical dissolution to form ions and physical weakening of the structure. Addition of Zn, Al, Ni, or Sn would not significantly enhance the structural stability of the Te cathode.

A second factor was plasma stability. Figure 2 shows the plasma instability in the electrolysis cells. The plasma electrolysis process began with a nice arc forming at the cathode tip; however, the arc changed into multiple micro arc discharges and then constantly migrated up and down the cathode assembly. Even heavy Teflon insulation around

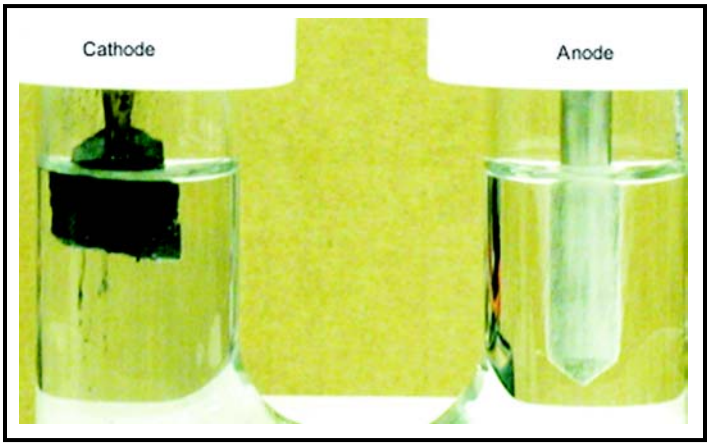


Figure 1. Test of tellurium as cathode for hydrogen generation suppression.

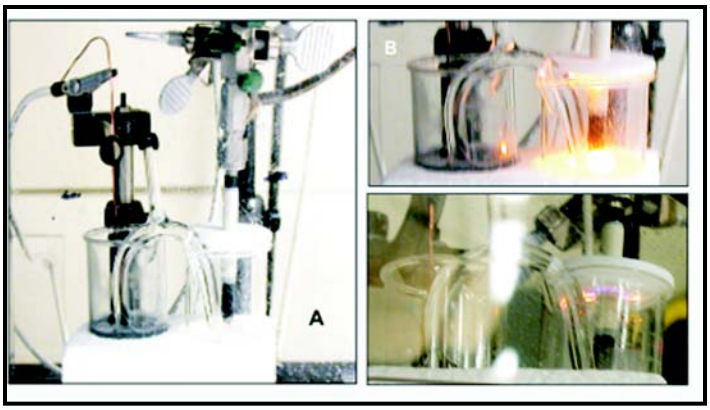


Figure 2. Salt bridge connected half-cells for plasma electrolysis experiments.

the cathode assembly would not stop the multiple micro arcs from being discharged to the wall of the half-cell. Researchers found this phenomenon puzzling since the wall of the half cell is floating and thus no discharges should go to the wall. Because of this, no electrolysis could occur. These technical hurdles could not be overcome to make the technology work. Since the plasma electrolysis concept did not show feasibility, researchers investigated a thermal plasma approach instead.

Thermal Plasma Synthesis. The Gibbs free energy of reaction (ΔG_r) was used to predict the thermodynamic feasibility of sodium borate conversion to sodium borohydride. Seven reactions were considered. Figure 3 shows the plot of ΔG_r versus temperature for the candidate reactions. The reactions are listed in descending magnitude of ΔG_r. A negative ΔG_r indicates the reaction can proceed spontaneously to products. From the plot, reactions (4)–(7) are clearly feasible at high temperatures. The predictive reactions used carbon-based materials for the

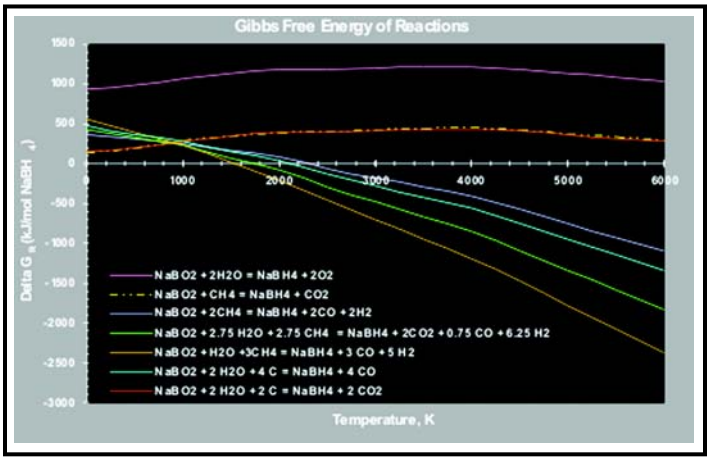


Figure 3. Gibbs free energy of reaction change for sodium borohydride synthesis.

borate reduction and the hydrogen source for hydrogenation was either methane or water.

(1) $NaBO2 + 2H2O \rightarrow NaBH4 + 2O2$
(2) $NaBO2 + CH4 \rightarrow NaBH4 + CO2$
(3) $NaBO2 + 2H2O + 2C \rightarrow NaBH4 + 2CO2$
(4) $NaBO_2 + 2CH_4 \rightarrow NaBH_4 + 2CO + 2H_2$
(5) $NaBO_2 + 2H_2O + 4C \rightarrow NaBH_4 + 4CO$
(6) $NaBO_2 + 2.75H_2O + 2.75CH_4 \rightarrow NaBH_4 + 2CO_2 + 0.75CO + 6.25H_2$
(7) $NaBO_2 + H_2O + 3CH_4 \rightarrow NaBH_4 + 3CO + 5H_2$

Researchers tested one of the predictive reactions in a thermal plasma reactor using borate solution and methane. Several runs had shown evidence of synthesizing sodium borohydride from the sodium metaborate in a single step. The product was not pure and contained borate and free carbon. However, X-ray diffraction and NMR analyses did confirm the presence of a low amount of the borohydride. Figure 4 shows the synthesis reaction in progress. The graphite reaction tube turned white hot during the reaction. At that time, the reaction conditions to maximize the borohydride conversion had not yet been optimized and a significant amount of work remained to achieve that goal.

Researchers also carried out a thermodynamic study to assess additional plasma reactions to convert sodium borate to sodium borohydride using other reductants with or without carbon. The study revealed some very surprising results. Some of the reactions investigated showed distinct low-temperature (<2,000 K) feasibility with water, methane, and hydrogen providing the hydrogen for the conversion. In these reactions, reactive metals are the main driving force to reduce the borate to borohydride. In turn, a solid-state combustion process oxidizes the metals to a very stable state. It is this process of stable oxide formation that made the borohydride synthesis possible. Carbon can also be used in combination with

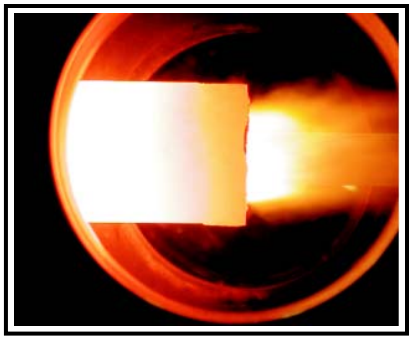


Figure 4. Thermal plasma synthesis of sodium borohydride.

Figure 5 shows the X-ray diffraction analysis for the thermal plasma reaction product. Besides sodium borohydride, impurities included sodium borate, sodium borohydroxide, sodium carbonate, and carbon. Sodium carbonate may have been formed by the water-gas shift reaction between CO and excess H_2O in the reaction. The sodium boron hydroxide formation mechanism was unclear. Researchers will need to conduct more analysis to understand the thermal plasma reaction to synthesize sodium borohydride in a single step.

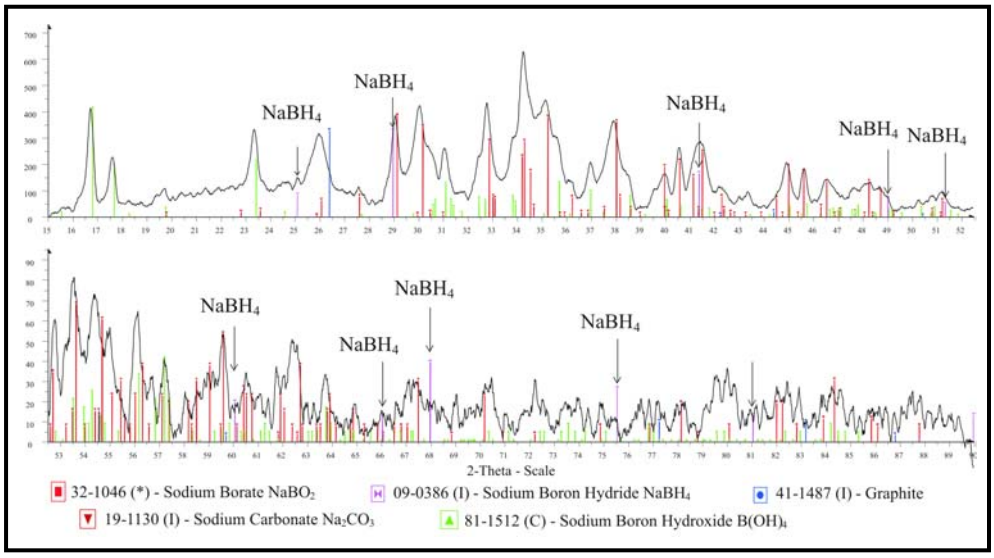


Figure 5. X-ray diffraction trace for the reaction product.

Figure 6 shows the NMR analysis for the product. The formation of $NaBH_4$ is clear. However, the sodium borohydride is unstable in water and will react with water to reform $NaBO_2$. Unreacted and reformed borate contributed to the $NaBO_2$ signal in the analysis.

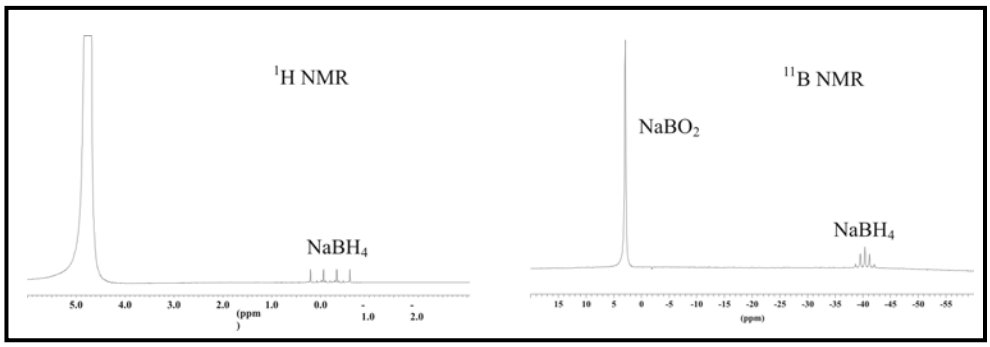


Figure 6. NRM confirmation of sodium borohydride synthesis in thermal plasma.

these reactive metals to synthesize the borohydride. Figure 7 shows the Gibbs free energy of reaction for synthesis of sodium borohydride from sodium borate. These reactions are favored at both high and low temperatures. Particularly, the Gibbs free energy of reactions is very negative at temperatures below 1,200 K. These reactions are shown below.

(1) $NaBO_2 + 4H_2O + 4Al \rightarrow NaBH_4 + 2Al_2O_3 + 2H_2$

(2) $NaBO_2 + H_2O + 2Al + H_2 \rightarrow NaBH_4 + Al_2O_3$

(3) $2NaBO_2 + 2H_2 + 2Al + CH_4 \rightarrow 2NaBH_4 + Al_2O_3 + CO$

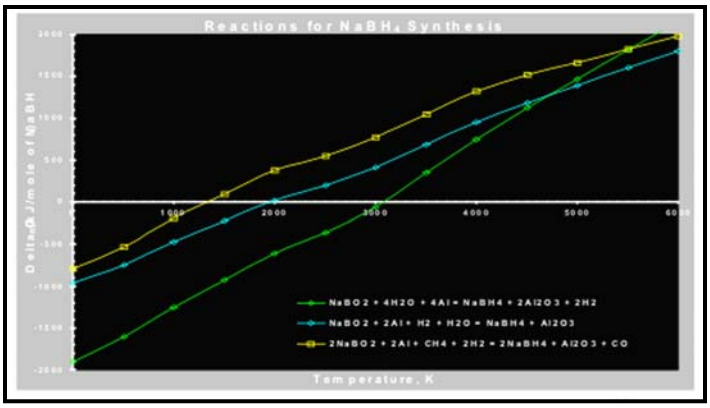


Figure 7. Low-temperature reaction for sodium borate conversion to sodium borohydride.

This figure indicates that the high-temperature helium gas from a gas-cooled nuclear reactor could supply the necessary heat to run the reactions directly. As predicted by reaction (1), water can be used to hydrogenate the reaction to form the borohydride. To test these thermodynamic predications, researchers performed the aluminum reaction in a sealed chamber at about 800–850°C using a thermal plasma heat source. The super heated argon plasma gas was used to simulate the high-temperature helium gas from the gas-cooled nuclear reactor. The sealed chamber was placed downstream of the plasma jet to heat the system to 800–850°C rapidly for 15 minutes to initiate the reaction. The reaction product was analyzed with X-ray diffraction and NMR.

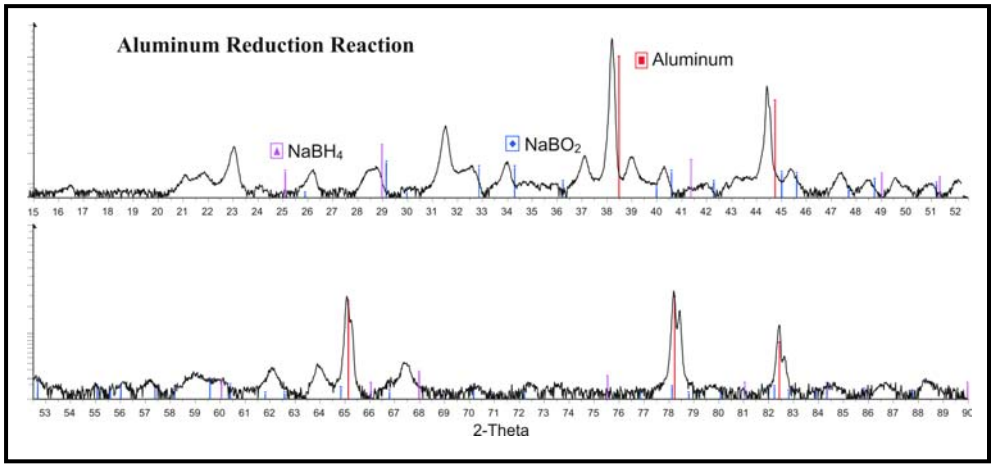


Figure 8. X-ray diffraction analysis for the Al and borate reaction.

In Figure 8, the X-ray analysis shows the reaction was not yet complete. Because of the short heating time, a significant amount of Al and borate remained unreacted with sodium borohydride and alumina formed in the reaction. The NMR analysis confirmed the formation of sodium borohydride in the low-temperature reaction. A simplified flow diagram is presented in Figure 9 for using a gas-cooled nuclear reactor to convert the borate to borohydride.

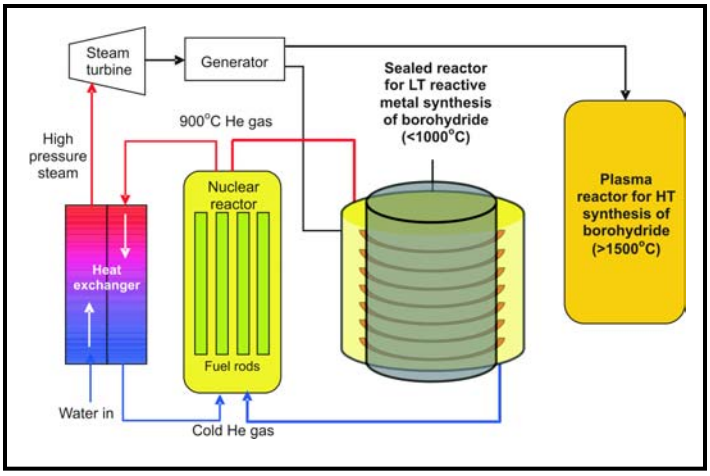


Figure 9. New concept using the gas-cooled reactor to produce hydrogen carrier materials.

Planned Activities

This NERI project has been completed.

NUCLEAR ENERGY RESEARCH INITIATIVE

Coupling of High-Temperature, Lead-Cooled, Closed Fuel Cycle, Fast Reactors to Advanced Energy Converters

PI: James J. Sienicki, Argonne National Laboratory

Collaborators: None

Project Number: 02-065

Project Start Date: September 2002

Project End Date: September 2005

Research Objectives

The objectives of the project are to develop a high-temperature, modular nuclear plant concept that combines the benefits of lead-cooled fast reactors (LFR) with the advantages of a gas turbine Brayton cycle advanced power conversion secondary system utilizing supercritical carbon dioxide as the working fluid. Benefits of such a reactor design include:

- the sustainability and closed fuel cycle benefits of a fast neutron spectrum core;
- the passive safety of molten lead primary coolant;
- the autonomous operational benefits of a fast spectrum core enhanced by lead coolant;
- the economic advantages of modular construction, factory fabrication, full-transportability, and simplification; and
- natural circulation heat transport at power levels in excess of 100 percent.

The advantages of a Brayton cycle advanced power conversion system include:

- significantly improved cycle efficiency (45 percent at 565°C turbine inlet temperature) relative to the Rankine water/steam cycle;
- reduced plant footprint due to fewer, simpler, and smaller-sized secondary side components; and
- possibly reduced capital costs, operating costs, and plant staffing requirements by radically simplifying the plant and eliminating costly Rankine cycle components.

A coupled, autonomous, LFR, supercritical, CO_2 Brayton cycle plant is well-suited for developing nations that do not have a nuclear infrastructure or a base of nuclear expertise. These nations will benefit from the deployment of proliferation-resistant, low power, autonomous, passively safe, modular plants that reduce operator workload and requirements. The plants are designed for extended long-term operation, after which the entire core cartridge is removed for reprocessing. Domestic power producers can also benefit from these plants as they require minimal electric grid modification, can be sited closer to cities, and provide capital and operating cost savings, greater efficiency, plant simplification, and reduced staffing.

Research Progress

Researchers studied the feasibility of coupling the 400-MWt Secure Transportable Autonomous Reactor-Liquid Metal (STAR-LM) natural circulation LFR to a supercritical carbon dioxide (S-CO_2) Brayton cycle. They have shown that such a coupling is indeed feasible and that the S-CO_2 temperatures achieved would result in a Brayton cycle efficiency of 45 percent. To study the feasibility and to determine the efficiency, researchers developed an analysis computer code for the plant's conceptual design. This computer code models the steady state behavior of both the autonomous natural circulation LFR and the S-CO_2 Brayton cycle secondary side. With this code, researchers calculated temperatures, pressures, and velocities for the lead and S-CO_2 around the primary and secondary circuits. They also performed an analysis to determine optimal designs for the S-CO_2 turbine and compressor (i.e., number of stages and stage dimensions) that maximize the cycle

efficiency. The researchers optimized the conceptual design of the lead-to-S-CO_2 heat exchangers so they can fit into the available volume within the reactor vessel and heat the S-CO_2 to high enough temperatures to provide the desired cycle efficiency. In addition, they developed optimal conceptual designs for compact CO_2-to-CO_2 recuperators that preheat the carbon dioxide delivered to the in-reactor heat exchangers to further enhance the cycle efficiency.

They demonstrated that the reactor can achieve autonomous load following through a core clamping and restraint approach that provides favorable reactivity feedback with coolant and fuel temperatures. This allows the reactor to adjust itself to changes in load demand with nearly unvarying core outlet temperature and without the operation of control rods. Researchers also developed new core designs with improved discharge burnup and power peaking. They performed computational fluid dynamics analyses of multidimensional temperatures and velocities to quantify the effects of coolant crossflow, intermixing, and entrance effects in the large hydraulic diameter, open-lattice core. They used engineering mechanics calculations to determine turbine blade stress limitations on turbine design and incorporated the resulting stress models into the conceptual design code.

Researchers also studied the transient operation of the coupled LFR-S-CO_2 Brayton cycle plant during normal conditions as well as its behavior during accidents. They investigated alternative strategies to control the S-CO_2 Brayton cycle, consistent with the lead-cooled fast reactor autonomous load following behavior, such that heat removal from the in-reactor heat exchangers matches the load demand from the electrical grid. One such strategy involves the use of an inventory control scheme whereby CO_2 is added to or removed from the Brayton cycle at load levels between 50 and 100 percent. At load levels below 50 percent, control is accomplished with a combination of the turbine inlet valve and in-reactor heat exchanger bypass. Figure 1 presents a schematic of the nuclear power plant including normal, shutdown, and emergency heat removal paths as well as alternative control mechanisms.

In addition, researchers initiated the development of a

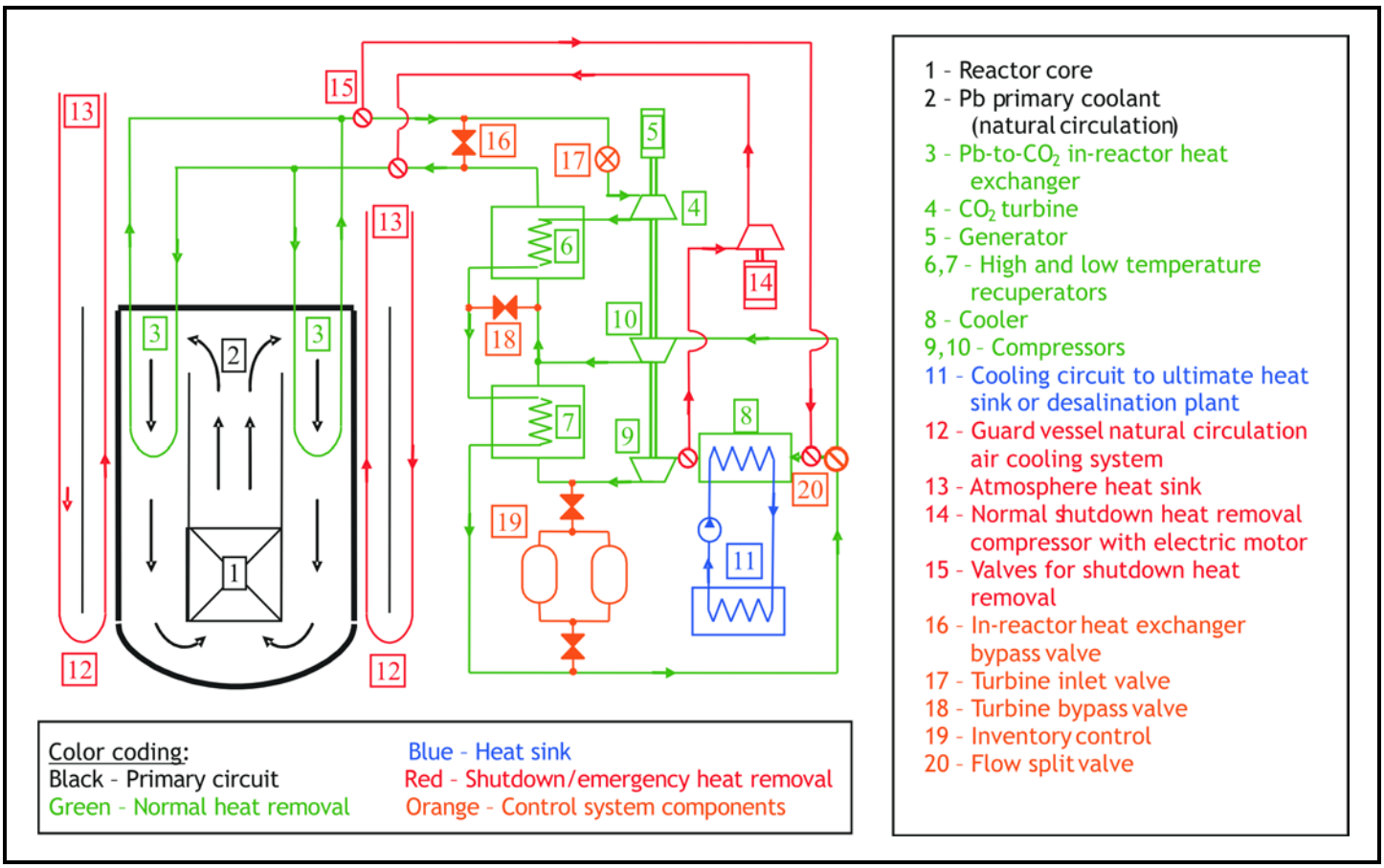


Figure 1. Schematic of coupled STAR-LM-S-CO_2 Brayton cycle nuclear power plant showing heat transfer paths and S-CO_2 Brayton cycle control mechanisms.

new and unique plant dynamics analysis computer code to calculate the behavior of the coupled LFR-S-CO_2 Brayton cycle plant during operational transients and postulated accidents. They have completed the portion of the code that calculates reactor system transient thermal hydraulics and neutron kinetics and have made much progress on the portion that describes the Brayton cycle transient thermal hydraulics. Researchers used the code to analyze a postulated accident involving loss of the normal heat removal path through the in-reactor heat exchangers without scram. The results showed that the reactor autonomously shuts itself down to decay heat levels due to passive reactivity feedback and that the system temperatures remain within acceptable accident limitations. Researchers also analyzed the consequences of an accident involving the postulated rupture of a heat exchanger tube and blowdown of CO_2 into the lead coolant. In addition, researchers examined the stability of lead natural circulation.

Researchers also completed an investigation of the feasibility of coupling STAR-LM to an alternative advanced energy converter that utilizes two-phase liquid metal magnetohydrodynamic (MHD) power conversion. They showed that a further significant increase in plant efficiency might be achieved through the application of MHD power conversion but that its realization would require significant technological challenges to be overcome. In addition, they conducted an assessment of experimental data needs for the coupled plant, encompassing both the S-CO_2 Brayton cycle as well as the MHD power conversion designs.

Planned Activities

Researchers will complete plant dynamics code development and analyze a set of operational transients and postulated accidents. They will also complete stability analyses for two-phase liquid metal flow, evaluate mechanical design issues, and carry out a limited analysis of the economic benefits of the S-CO_2 Brayton cycle.

Nuclear Energy Research Initiative

Experimental Verification of Magnetic Insulation of Direct Energy Conversion Fission Reactors

PI: Donald King, Sandia National Laboratories

Collaborators: Texas A&M University, General Atomics

Project Number: 02-068

Project Start Date: September 2002

Project End Date: September 2005

Research Objectives

To operate successfully, a direct energy conversion (DEC) device must direct fission fragments and electrons to the appropriate device electrodes and withstand large internal voltage gradients created during the conversion process. The objective of this project is to design and fabricate prototype DEC devices that can direct high energy, positively charged particles, and electrons to different electrodes in the device and withstand voltage gradients of one-half to several million volts. When finished, this project will have fabricated and demonstrated the operation of two prototype DEC devices, the fission electric cell and the magnetic collimator; developed the theory of direct energy conversion; and established models and analytical tools.

In particular, this project will demonstrate that high-energy ions representing fission fragments and electrons can be reliably guided to the DEC collectors by an applied external magnetic field from a central solenoid. For the magnetic collimator, this project will demonstrate that electrons can be suppressed so that they do not reach the collector stages. It will also demonstrate that the scaled prototypes can withstand and be operated at high voltages required for high DEC efficiencies.

Research Progress

In the project's second year, researchers focused on completing the Fission Electric Cell (FEC) and Fission Fragment Magnetic Collimator (FFMC) prototype designs. In addition, they commenced fabrication of the two prototypes and began preparing the test facilities. They developed the scaled FEC and FFMC prototype layouts and characteristics through detailed computational analysis and completed the electric field and magnetic field strength calculations for the FEC prototype. They calculated the simulated electron trajectories for this prototype and will attempt to limit field strengths to less than 50 kV/cm in non-critical prototype parts.

For the FEC (Figure 1), researchers procured the necessary materials, including the niobium raw stock and Kovar, and began fabricating the prototype. Niobium is the material of choice for the cathode and anode centers, where the electric fields can approach 4,000 KV/cm, while Kovar is the material that will balance the diode's cathode and anode. They also released quotes for fabrication of the

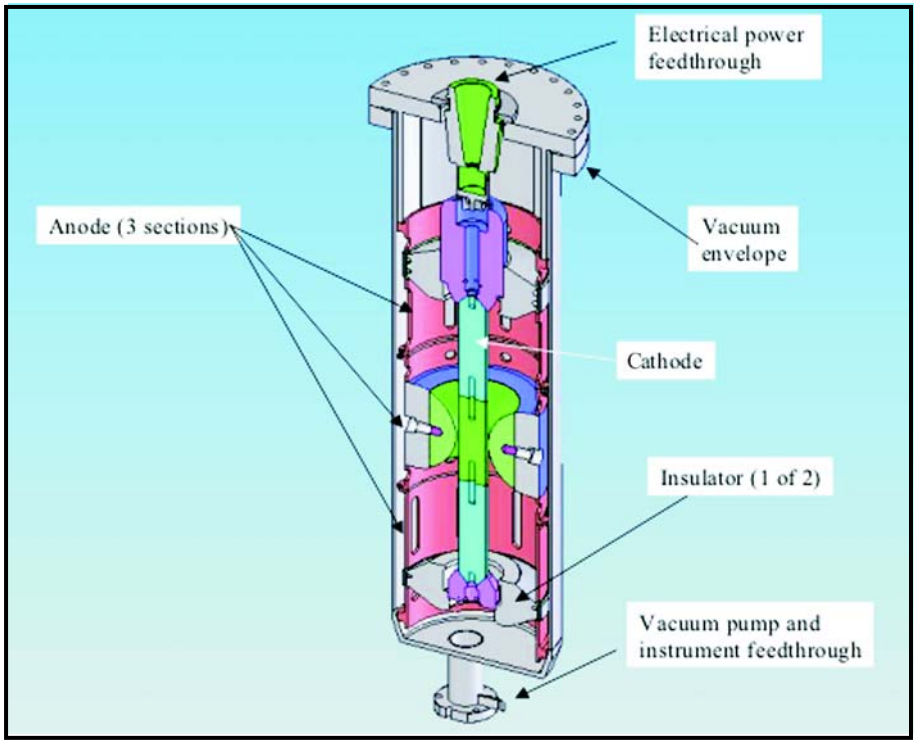


Figure 1. Final FEC prototype layout.

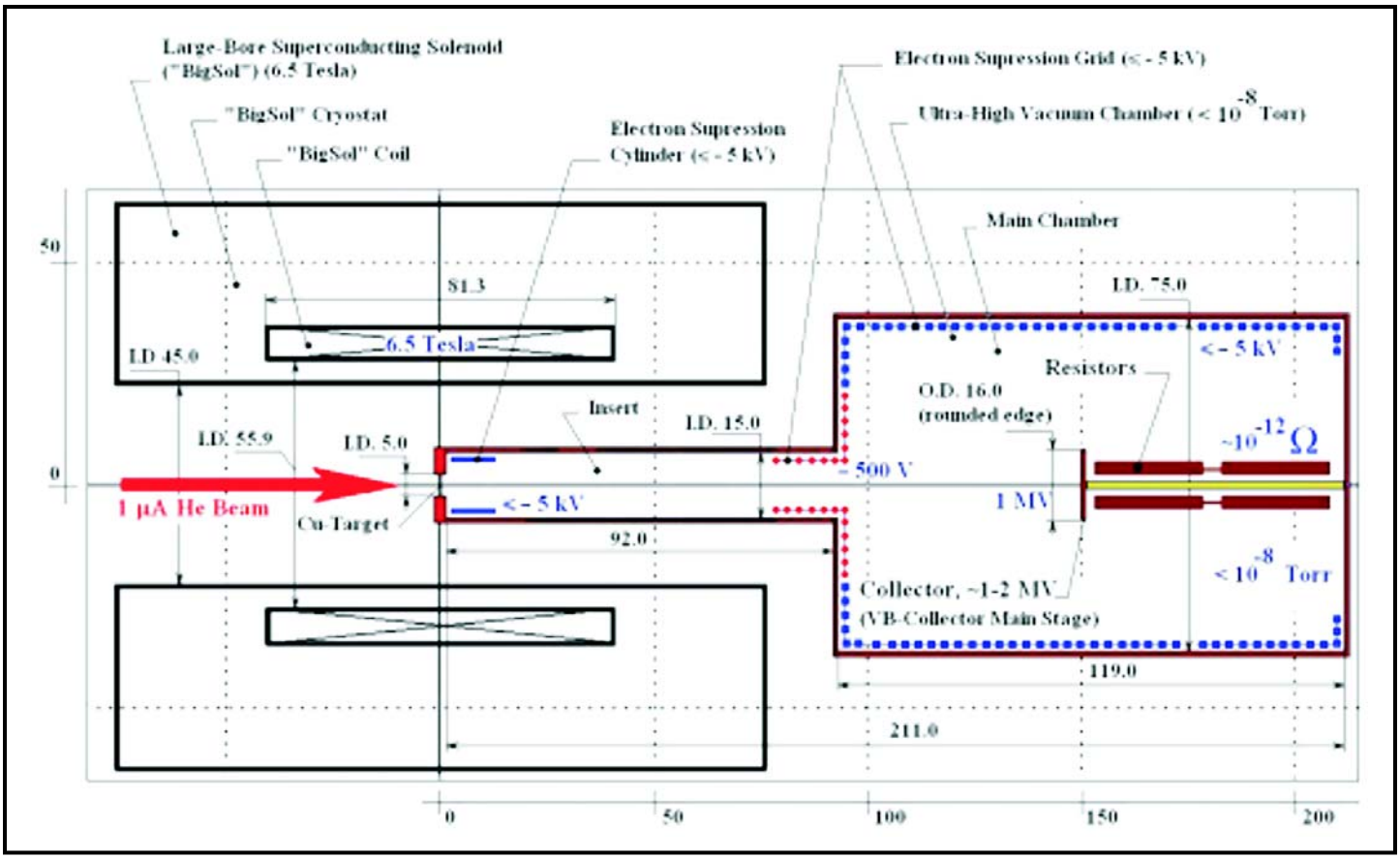


Figure 2. Component layout and main design parameters of the FFMC prototype.

diode and test fixture and began preparing the test facilities for the FEC. The 5-tessla gyrotron magnet was shipped from General Atomics to Sandia, lab space for testing was procured, and the facility infrastructure was prepared. The gyrotron magnet was started and characterized after approval of site documents required for operation. General Atomics initiated the design and fabrication of the instrumentation and power systems to control the FEC during testing. They also prepared environmental, safety, and health (ES&H) procedures for DOE operational readiness reviews and approvals, which are in process. The Cu target and ultra-high vacuum chamber assembly will be bolted to the cyclotron solenoid that is located in the beam line.

Researchers completed the final design and performance requirements for the FFMC prototype (Figure 2). They also completed the ion beam and target selection, finished selected prototype layout, and modeled its performance with a single-stage DEC collector. From the calculated results, researchers anticipate successfully demonstrating the feasibility of the FEC and FFMC concepts through prototype testing.

For the FFMCR, researchers completed the vacuum system and initiated the procurement of components. They fabricated and tested the Cu target and started to fabricate the DEC collector and the ultra-high vacuum chamber.

Planned Activities

Researchers will complete fabrication of the fission electric cell and magnetic collimator prototype. They will also complete test facility preparations and modifications and will install the prototypes. The project team will then test the prototypes to demonstrate their abilities to control high energy ions and electrons in high electric fields through the use of magnetic fields. They will test the FFMC prototype at the Texas A&M cyclotron and will complete the testing in phase 3 of the project.

NUCLEAR ENERGY RESEARCH INITIATIVE

Innovative, Low-Cost Approaches to Automating QA/QC of Fuel Particle Production Using On-line Nondestructive Methods for Higher Reliability

PI: Ronald L Hockey, Pacific Northwest National Laboratory (PNNL)

Collaborators: John Saurwein, General Atomics; Joseph Gray, Iowa State University; Richard A. Lowden and James H. Miller, Oak Ridge National Laboratory

Project Number: 02-103

Project Start Date: September 2002

Project End Date: September 2005

Research Objectives

Particle fuels, being considered for use in both near-term and Generation IV nuclear systems, consist of sub-millimeter diameter uranium oxide spheres (shown in Figure 1) uniformly coated to prevent the release of fission products into the reactor. About 15 billion of these spheres are needed to fuel a single reactor. This project is exploring, adapting, developing, and demonstrating innovative nondestructive test methods to cost effectively assure the quality of each of the fuel particles.

Current quality control methods are manual and, in many cases, involve destruction of the test specimen. Replacing these methods with nondestructive techniques, which are automated for higher speed, will make fuel production and reactor operation economically feasible, considering the requirement for extremely large fuel particle throughput rates. The primary task for achieving this objective is to establish standard signatures for the most problematic types of defects, as well as for acceptable particles, using several nondestructive examination methods.

Following is the technical scope of this project:

- To establish a full set of characterized test standards representing "good" and defective particles, so when a particle or a group of particles pass a sensor, researchers can measure a multiple attribute-dependent signature and use it for qualification or process control decisions.
- To evaluate, develop, and demonstrate nondestructive evaluation (NDE) QC capability on unfueled and fueled coated particles to reduce cost and improve reliability of manufacturing particle-based fuels.
- To develop and demonstrate a multiple attribute "Quality Index."
- To specify design parameters for commercial equipment for producing a large-scale NDE fuel manufacturing inspection system.

Research Progress

Particle Defect Library. A particle defect library containing over 50 different particle-coating runs, producing 16 different variations on the standard tri-isotropic (TRISO)-coated particle was completed. Researchers selected variations on the standard coating formula on the basis of producing particles with specific properties leading to larger failure rates and fission product release under

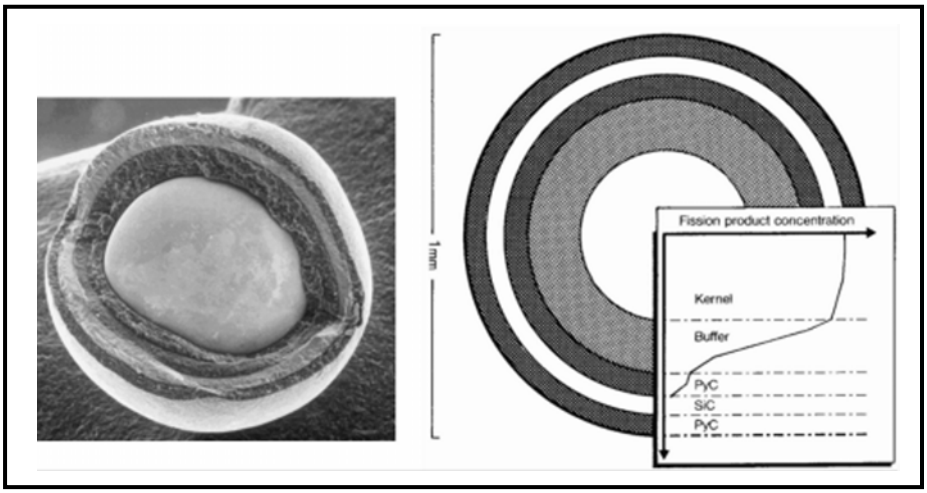


Figure 1. Coating cutaway view of a fuel particle (left), and the typical radial distribution of fission products (right).

reactor operating conditions. Table 1 summarizes part of the surrogate defect particle library coated at ORNL for this project. Researchers are evaluating a limited number of depleted uranium dioxide (DUO_2) and natural uranium oxycarbide (NUCO) kernels, coated at ORNL, to establish how kernel chemistry may affect the NDE measured properties of the fully coated particle for both surrogate and fueled kernel particles. Particles coated in FY 2004 will become available for testing in FY 2005.

Coating Condition	Coating Run	Description
Buffer only	1,2	Vary buffer thickness about standard
Buffer & IPyC	3,4	Vary buffer thickness with standard IPyC
All TRISO coatings	6	Thin buffer
PyC only	7,8,9,10	Vary density & anisotropy
"Standard" TRISO	5	Standard coating formula
All TRISO coatings	12,13	Vary SiC thickness, missing and 50% thin.
SiC only	14	SiC on kernel for independent effects.
Metal Inclusions	11	Added metal to coater, inducing inclusions.

Table 1. Coating conditions evaluated in particle defect library.

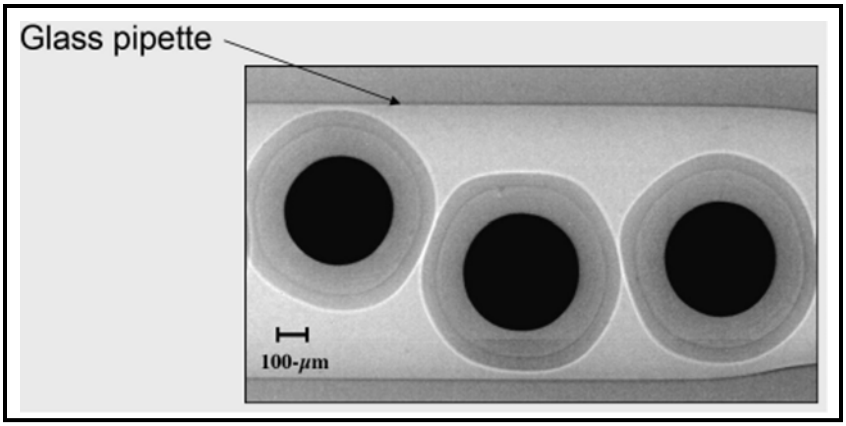


Figure 2. Cropped radiographic image obtained in analyzing and preserving the identity of about 20 particles stacked end-to-end in a thin-walled glass pipette.

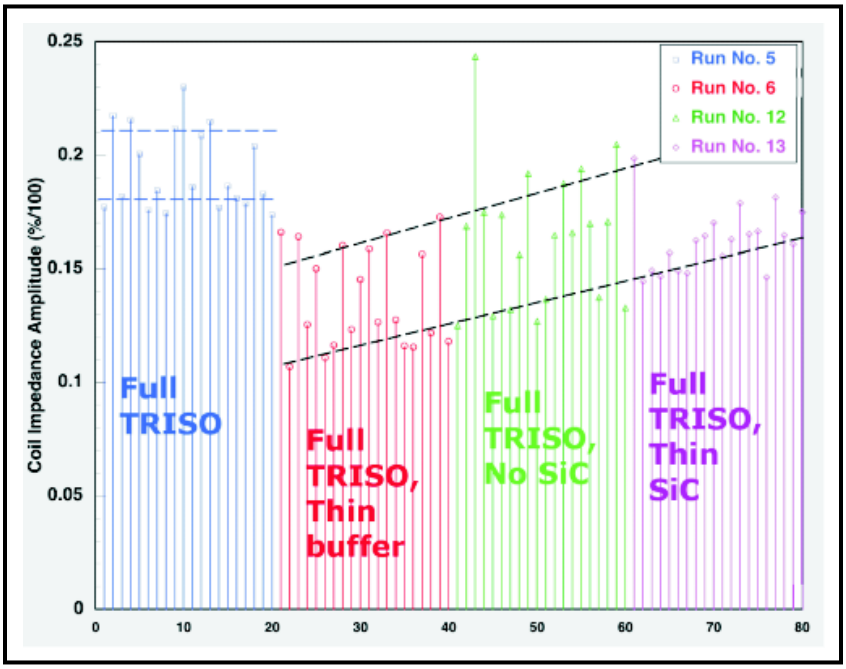


Figure. 3. Induction coil measurements from randomly selected particles of four different coating conditions.

X-Ray Characterization. Researchers utilized a radiographic method developed at Iowa State University to characterize particles in the defect library. The library of characterized defective particles is being used to develop, evaluate, and calibrate high-speed NDE inspection methods. In Figure 2, a radiographic image shows how particles were stacked and tracked in a thin-walled pipette. A radiograph provides a high (2.4 μm) resolution digital image of the structural density distribution within each particle from which attributes such as kernel size, coating layer thickness, and ovality can be derived. This method preserves the identity of each particle by confining them to a unique sequence throughout the radiographic process. High-resolution computed tomography provides an additional tool for viewing multi-dimensional defect structure found to occur between coating layers in several particles.

In-Line Sensor Development. Quality control and assurance are highly dependant on the ability to monitor the dimensional parameters for a large fraction of all coated fuel particles. Achieving this objective requires automated, on-line measurements at high throughput rates. Two high-speed electrical measurement methods showing promise for meeting these requirements were further developed and found to correspond with a combination of dimensional properties within fully and partially coated particles.

In the electrical measurement methods, particles are coupled to (1) an induction coil through its magnetic field and (2) a capacitive sensor through its electric field. Figure 3 shows induction coil measurements on a random selection of particles with four different coating conditions applied to surrogate kernels (ZrO_2), centered about two different diameters. When kernel size is held fixed within a narrow range, a thin (or "missing") buffer coating layer could be detected. Changes in other coating layers

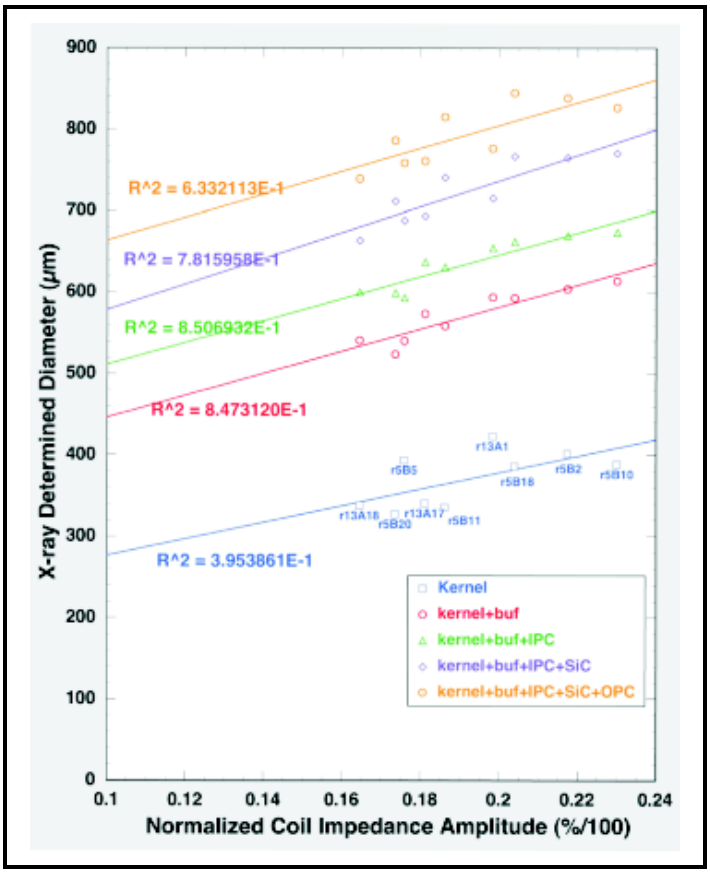


Figure 4. Radiographically determined coating layer diameter and induction coil impedance measurements.

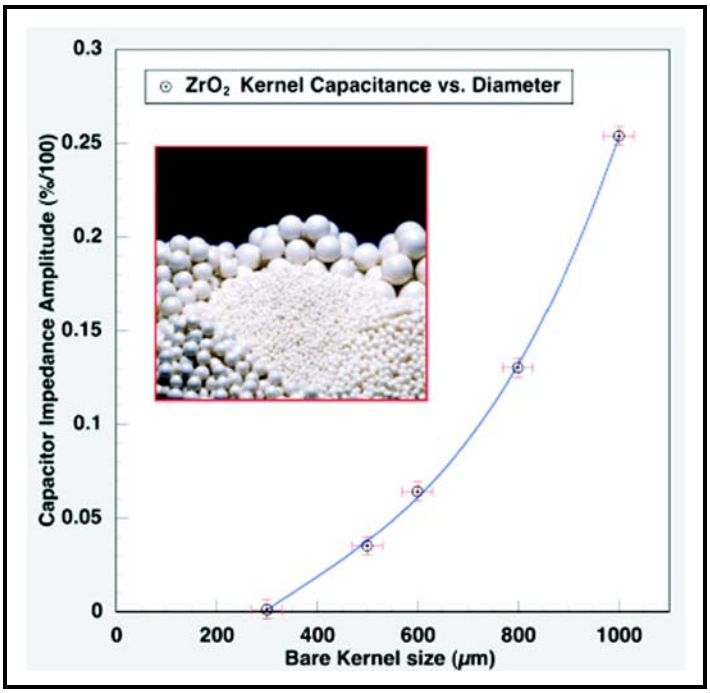


Figure 5. Capacitive sensor impedance amplitude vs. kernel diameter.

were also detectable when the kernel size was known or held within a narrowly defined range of diameters.

Figure 4 displays radiographically determined coating layer diameter measurements plotted against induction coil impedance. The R^2 values derived from linear regression between these two independent NDE measurements show that the two inner pyrocarbon layers—buffer and the higher density inner pyrocarbon—have a higher degree of correlation than other layers within the fully coated particle.

Characterizing the SiC layer in the TRISO fuel particle is crucial because it provides containment for fission products. In an attempt to independently measure the amount of nonconductive SiC material in the TRISO fuel particles, researchers evaluated a capacitive sensor on surrogate kernels. The electric field generated by the capacitive sensor responds to the dielectric constant and volume of material passing the sensor. Initial measurements, acquired from five different sizes of uncoated kernels, are shown in Figure 5. Subsequent results show that the capacitive method is sensitive to the thickness of the SiC layer. Remaining tests are scheduled to calibrate the capacitive sensor on several fully coated particles that vary only the thickness of the SiC layer. Recent work shows it is feasible to unite the capacitive sensor with the inductive sensor, making it possible to obtain a composite signature of the thickness and material property effects for each coating layer.

If the kernel diameter is controlled (using a screening process or capacitive sensor) before entering the coating process, then combining both the inductive and capacitive sensors into a single on-line inspection probe will enable a quality assurance measure for each particle in less than a few milliseconds. Deviations in any one coating parameter from its accepted value would be detected, even when identification of the deviant parameter is unnecessary.

Completion of Digital Image Analysis Evaluation. An analysis for using automated digital image analyzers is complete. The evaluation included several different optical imaging systems in combination with software designed to automatically analyze composite images. Existing commercial equipment was found to impose trade-offs among resolution, inspection speed, and cost. This evaluation found that real-time inspection at 0.69-micron pixel resolution is feasible using commercially available technology; and even better resolution is possible with further development.

On-Process and Model Coater Developed. A model coater was developed for simulating the fluidized bed coating process. An ultrasonic attenuation measurement system was developed and evaluated (at room temperature)

for real-time monitoring and control of the coating process. Preliminary measurements, using glass beads in the 500 to 1,000-micron range, exhibited a nearly linear relationship between glass-bead diameter and ultrasonic attenuation.

Planned Activities

Remaining activities include:

- Demonstrating key defect detection using on-line electrical methods.
- Assessing NDE methods for kernel chemistry effects of surrogate and DUO_2 particles.
- Showing NDE method process control capability.
- Implementing a measurement quality index.
- Specifying design parameters for the developed NDE inspection system.

In the final year of this project, researchers will demonstrate a laboratory-scale TRISO-coated particle fuel inspection system. The system will rely on multiple sensors that electromagnetically couple to all components of a particle. Particles having defects likely to diminish fission product retention will be used in demonstrating detection efficiency. The particle defect library will be expanded with radiographically characterized DUO_2 and NUCO coated kernels for demonstrating how kernel chemistry may affect the NDE sensor response to defective coating conditions.

Researchers will also further evaluate the gas-coupled ultrasonic attenuation method and determine its ability to monitor and control the coating process. A quality index, currently under development, will relate the NDE measurement data to acceptable process control parameters. Design specifications for the NDE inspection methods will appear in the final project report.

NUCLEAR ENERGY RESEARCH INITIATIVE

Model-Based Transient Control and Component Degradation Monitoring in Generation IV Nuclear Power Plants

PI: James Paul Holloway, University of Michigan

Collaborators: Westinghouse Electric Company, Sandia National Laboratories

Project Number: 02-113

Project Start Date: September 2002

Project End Date: September 2005

Research Objectives

The overall objective of this research project is to develop and demonstrate advanced algorithms for the safe, efficient control of operational transients in nuclear power plants and for the continuous, on-line monitoring of the system's status to warn of system degradation and incipient failures. Specifically, this research is developing two new capabilities for the nuclear engineering community:

- A method to develop robust, nonlinear, model-based, predictive control algorithms that combine plant sensor measurements with a predictive physical model of key plant systems to provide optimal closed loop performance of complex nonlinear systems.
- A method for monitoring plant system degradation based on comparing plant sensor readings with a physical model of key plant systems and exploiting component transition probabilities to provide probability distributions for degraded states.

Because identifying the system's state is an important requirement for both of these activities, researchers on this project will also seek an optimized approach for acquiring sensor data, that is, an approach that will allow them to extract the best information about the plant's state. Methods for integrating sensor data with physical models of plant systems will allow nuclear plant engineers to design optimal maintenance and control strategies into the new generation of nuclear plants. These methods will also provide nuclear plant operators with tools to operate their plant safely and efficiently within the complex energy market of the 21st century.

Research Progress

A key part of the component degradation monitor is an adaptive Kalman filter to identify when faults have occurred and to identify the faulted components. Researchers have developed an adaptive Kalman filtering algorithm for the diagnosis of faults or degradation of multiple components in nuclear power plants. The diagnostic algorithm uses measurement residuals, i.e., the difference between a measurement and its prediction. This residual generates a noise input to the uncertain component state in an adaptive Kalman filtering algorithm so that various postulated component transitions or degradation may be statistically represented. Researchers begin the fault detection process by monitoring the residual sequence with a standard Kalman filter utilizing a system model. If the model was no longer representative of the true system behavior, as indicated by a significant value of the measurement residual, then researchers determined how far the model deviated. The excess measurement residual was used to determine suitable perturbations to the system to model uncertainty. The first algorithm for this purpose assumed independence of all state uncertainties and assigned all of the measurement residuals to uncertainty in the degraded component state. This year, researchers also started a parallel degradation monitoring program focused on implementing a hierarchical Bayesian method and using Markov Chain Monte Carlo methods.

Researchers previously laid out a model-based predictive control methodology that relies on performing an optimization over a predicted future for the reactor plant system. This methodology requires an estimate of the state of the real reactor to provide the initial state for the predictive model. This year the focus of control algorithm development has concentrated on the state estimation problem. The current approach to state estimation requires only a dynamic model that can advance an initial state to a final state. Thus, researchers assume only that they can compute the flow of the dynamics, but do not predict the vector-field that generates the flow. They also did not make assumptions about stochastic processes that might or

might not be disturbing the system. However, they introduced a regularization parameter to balance the relative certainty in the measurements versus the model. No linearization of the model is required, so the system model can be treated as a black box.

The state estimation algorithm works by minimizing a function that includes two terms: (1) the difference between the measured data and the data generated by a model integrated over a finite history, and (2) a regularization term that favors estimates that are close to those predicted by the model, discouraging rapid changes in the estimation. The minimization is accomplished only approximately during any one time step by using a fixed number of iterations of a pattern search, initially centered on the state that was predicted by the model.

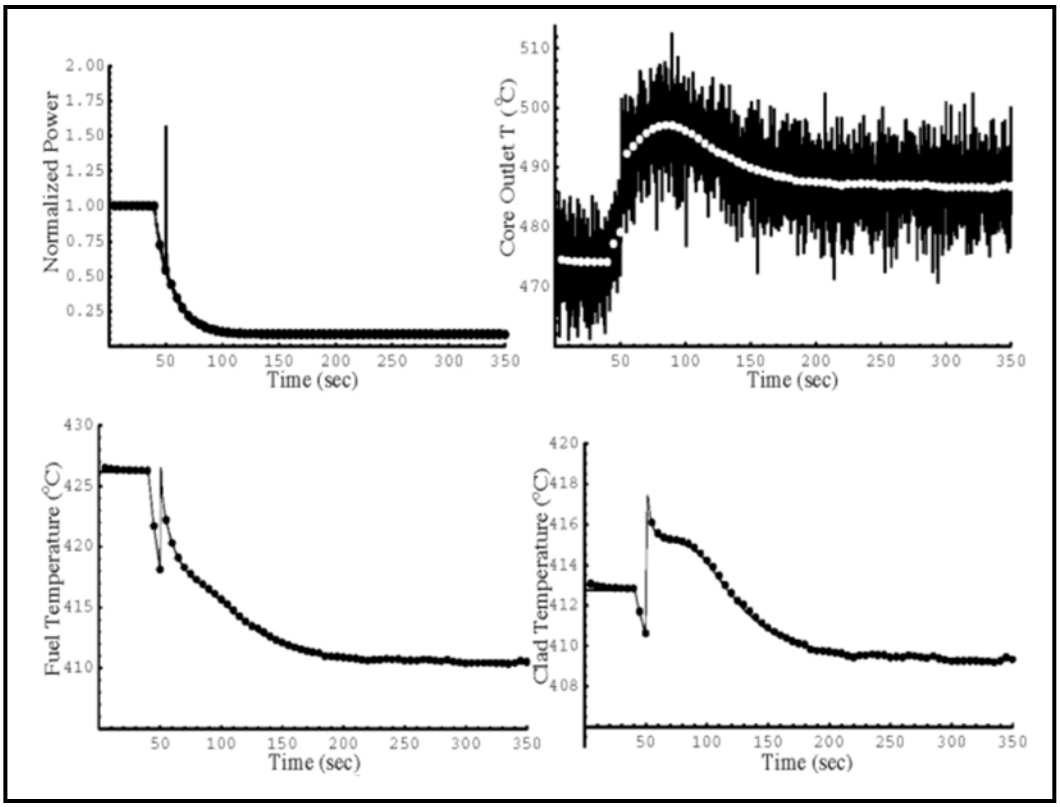


Figure 1. Reactor power, core outlet, fuel and clad temperature state estimates (dots) compared to true or measured system values (lines). Note that measured values include significant noise but state estimates are smooth.

As a test of this state reconstruction method, researchers performed several state estimations of a reactor including fuel temperature feedback and separate core and coolant expansion feedback contributions. The system has a 10-dimensional state space, of which two were measured: core power and core outlet temperature. Figure 1 shows the reconstructed dynamics of the plant for one test transient. In this transient, the reactor ran at steady state for 40 seconds, at which time a pump failed and began to coast down. The power dropped due to temperature feedback, but at 50 seconds a step reactivity insertion momentarily raised the power. Fuel and coolant temperature feedback effects very quickly overcame this rapid rise so that the power continued to fall. State estimates were made every 5 seconds and are shown in Figure 1 for the power and core outlet temperature for which there are noise measurements, as well as for the fuel and clad temperature, which were not measured. The measured data were well filtered by the technique, and the unmeasured data values were well recovered.

Researchers completed a detailed RELAP-based nuclear steam supply system and balance-of-plant model for safety analysis and for control systems studies, with considerable input from Westinghouse's International Reactor Innovative and Secure (IRIS) partners. This detailed simulator can be used as a high-fidelity plant model, but the extensive detail included in the model makes it problematic for control studies because its computational time is 5–7 times slower than real time on high-performance personal computers. To complement this tool, a low order, fast-running simulator has now also been developed. To enable development of this plant model within the budget of this program, Westinghouse has cooperated with the Polytechnic of Milan to develop an IRIS plant simulator using the Modelica language. Aside from providing a much needed control systems analysis tool, this fast-running simulator (which runs 10 times faster than real time) can be used as a basis for developing the low-order algorithms used in innovative control systems.

In parallel with the development and initial validation of the low-order plant model, Westinghouse has completed the conceptual design of the IRIS control systems architecture, providing a complete identification of the main control signals and actuators. This activity was initiated during the

first year, during which a preliminary control system based on existing light water reactor (LWR) experience was implemented. Preliminary studies provided an improved understanding of the IRIS control response and were used during the second year to complete a plant-specific control systems structure.

Planned Activities

In degradation monitoring, the adaptive Kalman filter algorithm will undergo continued development. Researchers have outlined a new means to adapt the filter that can consistently represent the uncertainties associated with component degradation and system state, rather than assigning the entire deviation in measurement residuals to component degradation. Thus, they postulate that the component degradation occurs right before the combined state is updated, at a time step which explicitly accounts for the effect of the component degradation in the state transition matrix. In the next year of this project, the project team will test this approach. In addition, this project will continue to explore the Bayesian model of system component degradation. There is some work remaining to generalize the theoretical development so it can be applied to a broad range of degradation issues and to validate and verify the algorithm. However, researchers feel the approach shows great promise for a broad range of problems; in particular, it will permit the inclusion of multiple sources of failure in a system-level reliability analysis.

The two main parts of the model-based predictive control methodology—state space-based predictive control and fully nonlinear state space identification—are now developed and have been tested separately. These will be combined into a full model-based predictive control algorithm. A key issue for this algorithm development is the computational complexity of the two optimizations that must occur at each control time-step. Researchers believe that the use of pattern searches will enable several approaches to proceed without achieving a full optimization within any one time-step.

Researchers have also tracked developments in the IRIS control system in order to understand a set of control problems to apply model-based predictive control in the final year of this project. To complete the validation and optimization, the full-function transient analysis model will be completed and its fidelity verified by analyzing selected transients and comparing the results with those obtained by other (generally slower but more detailed) codes. Then, the verified model will be used to perform a wide range of transient analyses to validate the control system's correct operation.

NUCLEAR ENERGY RESEARCH INITIATIVE

Centralized Hydrogen Production from Nuclear Power: Infrastructure Analysis and Test-Case Design Study

PI: William A. Summers, Savannah River National Laboratory

Collaborators: General Atomics, University of South Carolina, Entergy Nuclear, Inc., Argonne National Laboratory

Project Number: 02-160

Project Start Date: September 2002

Project End Date: September 2005

Research Objectives

Hydrogen production from nuclear power can provide a significant new source of energy for both the transportation and industrial sectors. The objectives of this project are to identify, characterize, and evaluate the critical technical and economic issues associated with nuclear hydrogen production, including the integration of the centralized nuclear hydrogen production plant into an overall hydrogen infrastructure. This work complements other design studies related to nuclear hydrogen production by expanding their analyses to encompass factors beyond the production plant, including consideration of on-site hydrogen storage and handling, hydrogen transmission and distribution, integration with end-user processes, and overall energy system economics. The work is divided into two phases with the following objectives:

Phase A - Plant Definition and Economic Analysis: Define the physical characteristics, infrastructure requirements, and economics for an Nth-of-a-Kind centralized thermochemical hydrogen production plant driven by a helium-cooled modular nuclear reactor.

Phase B - Application-specific Pre-conceptual Design: Prepare a pre-conceptual design, including site selection and cost estimate, for a test-case nuclear hydrogen plant providing hydrogen export to a local chemical plant end-user.

Research Progress

During Phase A, researchers were able to identify and analyze the major requirements for nuclear hydrogen production; to define the physical characteristics and major considerations for the nuclear reactor, the thermochemical hydrogen production process, and the related hydrogen infrastructure; and to perform an economic analysis of hydrogen production costs, infrastructure costs, end-user economics, commercial hydrogen cost comparison, distributed electrolysis comparison, and tradeoff studies and sensitivity analysis. The infrastructure issues included systems for hydrogen storage and delivery, as well as requirements and considerations related to potential hydrogen end-users. Researchers also investigated externalities, such as the effects of carbon taxes or credits. The results of these analyses were documented in a topical report at the completion of Phase A.

The general concept of a "nuclear hydrogen future" is shown in Figure 1. Hydrogen is produced in a centralized nuclear hydrogen production facility, temporarily stored if

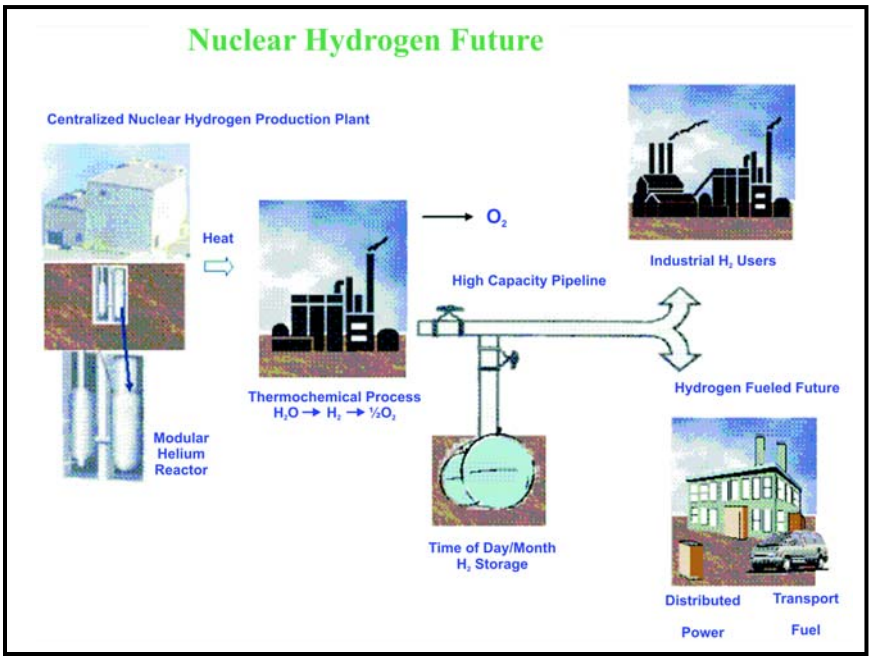


Figure 1. Schematic of a nuclear hydrogen future.

necessary for short periods at the production site, transported by pipeline, and distributed to industrial and retail end-users. Through the use of a thermochemical water-splitting process, the only consumables are nuclear fuel and water. Oxygen can be recovered and sold as a by-product, thereby reducing the net cost of hydrogen production.

The baseline design approach for the nuclear hydrogen production plant was the use of a high-temperature gas-cooled nuclear reactor combined with a thermochemical water-splitting process using the sulfur-iodine (S-I) cycle. High-temperature gas reactors (HTGRs) are the most developed of the next generation nuclear reactors (known as Generation IV). They are expected to be the first reactor type to be used for the production of hydrogen via water-splitting because of their design maturity and their capability to supply the high-temperature heat requirements of thermochemical hydrogen production processes. Figure 2 shows an artist's rendition of a nuclear hydrogen production plant.

The nuclear reactor is based on the Modular Helium Reactor (MHR) under development by General Atomics, Inc. It is a helium-cooled reactor using a graphite moderator and coated-particle tri-isotropic (TRISO) fuel, and employing an annular core design with prismatic blocks and a passive safety system. When utilized for electricity production, the MHR is designed to operate with a thermal power of 600 MW_{th} and a helium coolant outlet temperature of 850°C. A typical nuclear hydrogen reactor plant would employ four reactors for a total plant rating of 2,400 MW_{th}. For hydrogen production, the MHR can be configured to operate with an outlet helium temperature up to 1,000°C. A secondary helium cooling loop, heated to 925–975°C in an intermediate heat exchanger (IHX), then delivers heat to the thermochemical process for the production of hydrogen. During this project, researchers analyzed the impact of a higher operating temperature requirement on the nuclear reactor design, including challenges posed for the reactor, nuclear fuel, and IHX. They identified various design options to address these issues.

Researchers selected the S-I thermochemical cycle as the baseline hydrogen production process. The S-I process consists of three chemical reactions, coupled in two process loops. The process involves thermal decomposition of sulfuric acid and hydrogen iodide, followed by regeneration of these reagents using the exothermic Bunsen reaction. Process heat supplied at temperatures greater than 800°C concentrates and decomposes sulfuric acid, while heat at lower temperatures provides for HI decomposition. Researchers prepared a conceptual design of the S-I process based on previous work performed by General Atomics, incorporating updates to the flowsheet and the latest process and hardware information. They performed process modeling using these flowsheets and the AspenPLUS® computer model to calculate the overall thermal efficiency for the nuclear hydrogen plant. The results indicated an efficiency range of 42–56 percent based on the higher heating value of the hydrogen product, depending on the operating temperature and the degree of optimization in the process design. The modeling results indicate that outlet helium temperatures from the MHR less than previously assumed (below 950°C) may be viable. Researchers continue to work on refining the process design and the efficiency estimates.

An important aspect of this study was assessing the integration of a nuclear plant with a hydrogen production process, considering the broader hydrogen infrastructure needs and end-user hydrogen requirements. Researchers analyzed various options for storing the hydrogen, including compressed storage, converting to a liquid form by inclusion of a hydrogen liquefier at the nuclear production plant, and the use of pipelines for storage. They also compared various hydrogen transport options for delivery to the end-user, including tube trailers, liquid hydrogen trucks, and pipelines.

Researchers performed an economic analysis for a mature, Nth-of-a-Kind nuclear hydrogen production plant coupled to an S-I thermochemical hydrogen production process. They determined the plant costs from updated estimates of

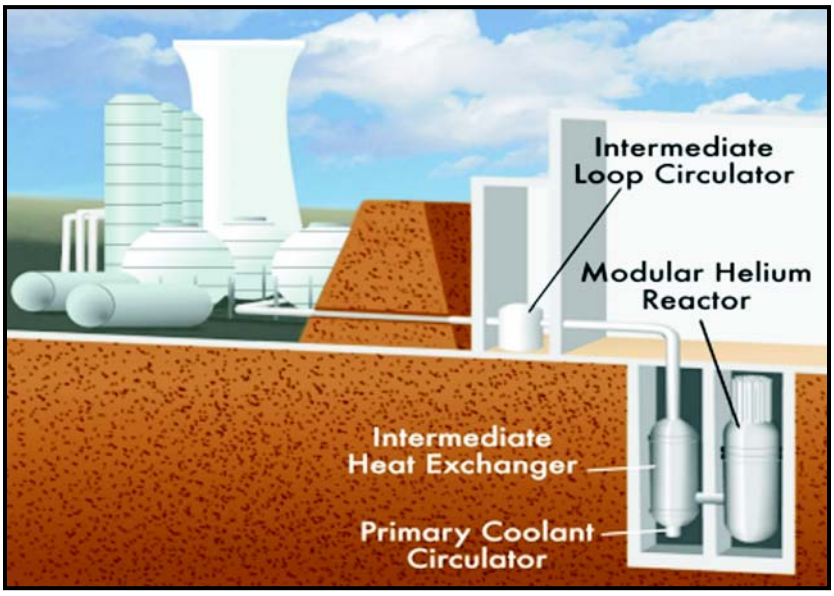


Figure 2. Artist's rendition of a nuclear hydrogen plant.

capital and operating costs, estimating the total capital investment to be $1,577 per kilowatt (thermal), based on the higher heating value of the hydrogen product. They calculated the cost of hydrogen production at the plant gate for various economic scenarios and for a range of estimated capital and O&M costs. The Base Case hydrogen production costs with and without by-product oxygen credit were $1.35 and $1.65 per kilogram, respectively. Cost estimates were made for the infrastructure issues necessary to deliver the hydrogen to a large industrial plant, including on-site hydrogen storage, pipeline transport, and end-user processing. The delivered cost of nuclear hydrogen to a large chemical plant through a 20- mile long regional pipeline was determined to be $1.57 per kilogram, assuming a by-product oxygen credit. The two main types of end users for nuclear-produced hydrogen include large industrial users (e.g., oil refineries and ammonia plants) and hydrogen energy consumers (e.g., hydrogen vehicles, distributed power, etc.). The industrial users are a large and growing current market, whereas the hydrogen energy users' market will evolve over time with the hydrogen economy.

In order to establish the competitiveness of nuclear hydrogen, researchers analyzed the technology and economics of conventional hydrogen plants which use steam reforming of natural gas. They determined the cost of hydrogen production for a variety of industrial applications, and compared it to the cost of importing nuclear hydrogen, for cases both with and without CO_2 emission restrictions.

Figure 3 compares the cost of imported nuclear hydrogen and on-site hydrogen produced from steam reforming of natural gas for a 200 TPD hydrogen plant. All costs are levelized over a 30-year period and take into account natural gas price real escalation, assumed to be 2 percent per year. The first year natural gas cost is shown on the abscissa.

The breakeven (Year 1) natural gas price where nuclear hydrogen costs equal the costs based on natural gas steam reforming is $6.36/MMBTU without CO_2 removal, and $4.37/MMBTU considering the additional cost of CO_2 removal and sequestration. Since recent natural gas prices have already exceeded these values, one can conclude that future nuclear hydrogen costs for the Base Case will be economically competitive with conventional natural gas hydrogen production, even without CO_2 restrictions.

Finally, researchers performed an economic analysis for the scenario of a centralized nuclear hydrogen plant delivering hydrogen through a pipeline system to distributed users in a future hydrogen economy. The sample case included the distribution of hydrogen from a centralized production plant to distributed consumers equivalent to today's neighborhood gas stations. The estimated cost resulting from the distribution system, including the cost of main transmission pipelines, smaller distribution pipelines, hydrogen compressors, station compressors, station dispensers, and station storage, amounts to approximately $2.00 per kilogram of hydrogen. When the nuclear hydrogen production cost is added to the distribution costs, the cost of hydrogen at refueling stations ranges from $3.37 to $3.66 per kilogram of hydrogen, with and without a by-product oxygen credit, respectively. This is considerably higher than the delivered cost of nuclear hydrogen to industrial users due to the more extensive delivery costs and the forecourt costs for hydrogen distribution to end users.

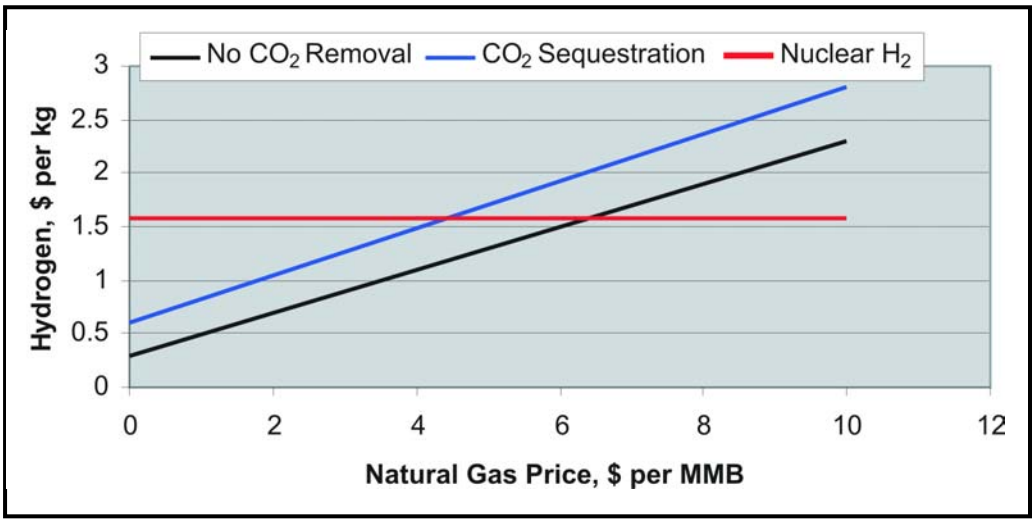


Figure 3. Nuclear hydrogen cost comparison with steam reforming.

The overall conclusion reached by the researchers based on the Phase A work was that nuclear hydrogen production is a technically feasible and economically viable option for addressing future national energy needs. No significant technical or economic barriers were identified. Potential end users for nuclear hydrogen include large industrial hydrogen facilities, such as ammonia plants and oil refineries, and future hydrogen economy users, such as fuel cell

automobiles. The industrial hydrogen users constitute a large near-term market with hydrogen supply requirements that match well with the characteristics of a nuclear hydrogen plant. Constructing nuclear hydrogen plants to serve industrial customers and then extending the hydrogen supply system to serve automotive and other users, may be a means of addressing the perplexing "chicken-and-egg" problem that challenges the establishment of a hydrogen economy.

Planned Activities

During the final year of this project, researchers will complete Phase B, an application-specific pre-conceptual design of a nuclear hydrogen production plant. This will take the analysis to the next level and attempt to further characterize the nuclear hydrogen production option. Phase B will provide for a more detailed evaluation of nuclear hydrogen production, including plant design and site selection issues, integration with an existing industrial hydrogen end-user, sizing and characterization of hydrogen storage and pipeline transmission systems, identification and evaluation of other hydrogen infrastructure issues, and a detailed economic assessment. Researchers will perform an in-depth study of plant site selection and estimate the cost of constructing the plant.

NUCLEAR ENERGY RESEARCH INITIATIVE

Near-Core and In-Core Neutron Radiation Monitors for Real-Time Neutron Flux Monitoring and Reactor Power Level Measurements

PI: Douglas McGregor, Kansas State University

Collaborators: None

Project Number: 02-174

Project Start Date: September 2002

Project End Date: September 2005

Research Objectives

There is a need for advanced neutron radiation detectors for performing "near-core" reactor measurements that are tiny enough to be inserted directly into a nuclear reactor without significantly disturbing the neutron flux. These detectors must be capable of withstanding intense radiation fields, adept at both pulse-mode and current-mode operation, and able to discriminate neutron signals from background gamma ray signals. In this NERI research project, miniaturized fission chambers are being developed and deployed in a Training, Research, Isotopes, General Atomics (TRIGA) research reactor. These unique, miniaturized, neutron detectors will be used for three specific purposes: 1) as reactor power level monitors, 2) as power transient monitors, and 3) for real-time monitoring of the thermal neutron flux profile in a core. The third application has the unique benefit of providing information that, with mathematical inversion techniques, can be used to infer the three-dimensional distribution of fission neutron production in the core. The project has two specific tasks which address detector hardware and software. Researchers will first focus on detector development, characterization, and deployment, followed by data reduction and development of computer codes for determining real-time reactor core power levels.

Detector development involves the design, construction, and deployment of radiation-hardened, thermally resistant, inexpensive, gas-filled fission chambers. These are referred to as "micro-pocket fission detectors" or MPFDs. Electronics attached to the devices will allow for both pulse-mode and current-mode read-out. Each detector pack consists of a triad of devices, including a ^{233}Th-based fast neutron detector, a ^{235}U-based thermal neutron detector, and an uncoated device for gamma ray background measurement. Seventy-five triads, amounting to a total of 225 detectors, will be distributed through a plane in Kansas State University's (KSU's) TRIGA reactor core. Researchers are developing a back-projection algorithm that uses information extracted from the detector triad arrays to determine the power density within the fuel elements of the nuclear core.

Research Progress

An MPFD device consists of a small parallel-plate gas detector lined with a neutron reactive material such as ^{235}U or ^{232}Th. The micro-pocket is made out of aluminum oxide, a material that can withstand the high-radiation and high-temperature environments in a nuclear reactor core. Early generation MPFDs have been fabricated and tested in neutron beam ports and in the core center of a 250-kW TRIGA nuclear reactor. First and second generation MPFDs were 1 mm thick and 3 mm in diameter.

Prototype uranium-coated MPFDs manufactured with aluminum oxide substrates yielded spectacular results. The observed fission product spectra collected by the MPFDs in a neutron beam of approximately 10^6 cm^{-2}-s^{-1} neutrons and 10^2 R/h gamma rays correlated well with modeled results [1, 2]. Further, with the neutron beam blocked by a 2-mm-thick sheet of cadmium, absolutely no radiation counts were registered from the gamma rays, thereby proving that the devices are truly insensitive to gamma ray background. The insensitivity to gamma ray background is a fortunate consequence of their small size and low atomic mass composition. Hence, the detectors can be operated in a nuclear core with confidence that the signals observed are exclusively from neutron interactions and not gamma. Further tests in the core center of a TRIGA Mark II nuclear reactor showed that the detectors could be operated in

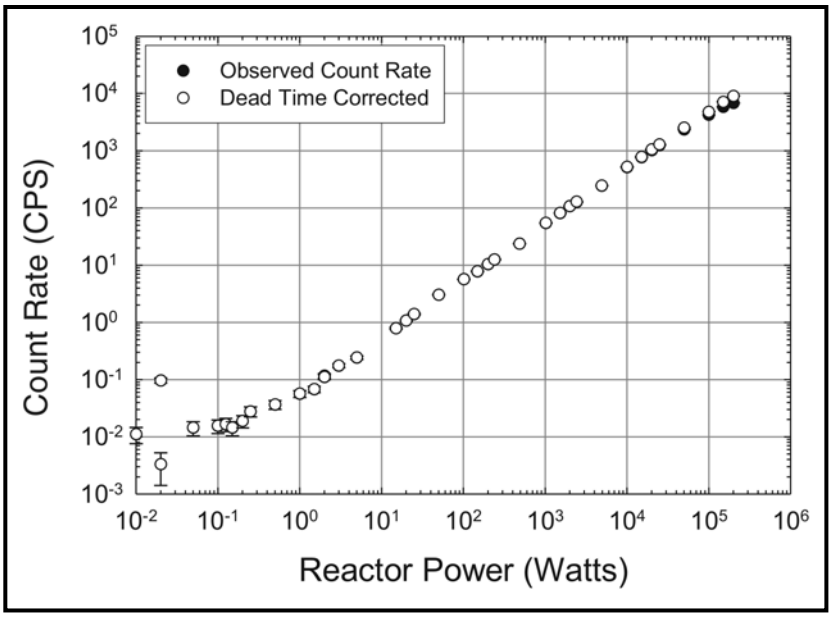


Figure 1. MPFD count rate as a function of reactor power.

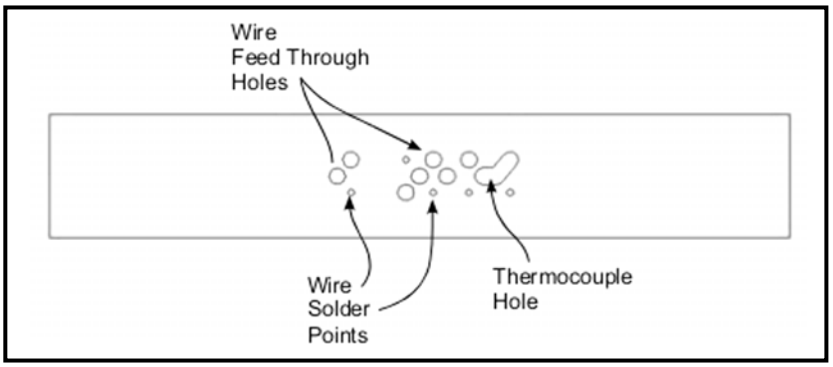


Figure 2. Aluminum oxide base (front view) showing the locations for feed through wires, wire attachment locations, and a temperature monitor (thermocouple).

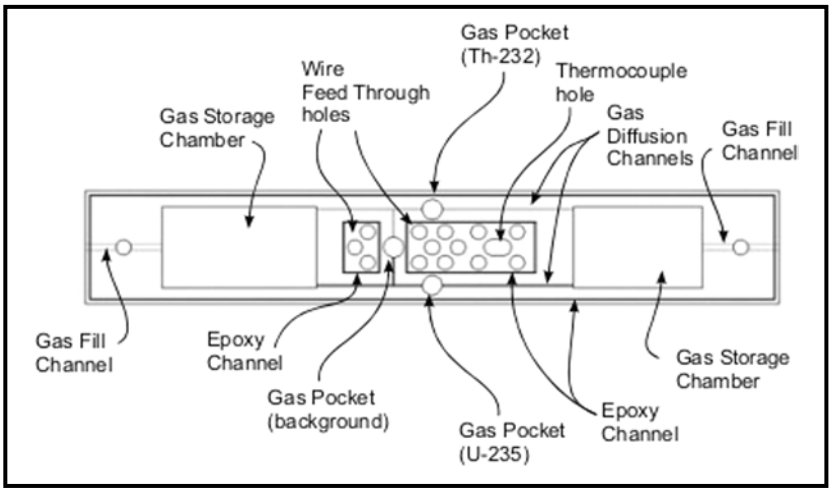


Figure 3. Aluminum oxide cavity (front view line drawing) piece showing the locations for feed-through wires, gas storage, diffusion channels, detector pockets, and the thermocouple. Additional epoxy channels were added to aid in bonding the parts together.

pulse mode up to full power of 225 kW and a neutron flux of $5x10^{12}$ n-cm^{-2}-s^{-1}, with minor dead time losses (see Figure 1). The device was placed in the center of the KSU TRIGA nuclear reactor core, thereby experiencing thermal neutron fluxes ranging from 10^3 to $5x10^{12}$ n-cm^{-2}-s^{-1}. The pulse mode operation remained linear with no apparent dead time losses until a thermal neutron flux of 10^{11} n-cm^{-2}-s^{-1} was reached. Still, the dead time losses were correctable at a flux of $5x10^{12}$ n-cm^{-2}-s^{-1}, being only 25 percent maximum.

New (third) generation MPFDs are smaller in size, being 1 mm thick and 1 mm in diameter, and also comprise a triad of three detectors per unit. They consist of two base plates and one cavity plate. Each plate is designed with three gas pockets and numerous feed-through holes to attach conducting wires to the cathodes and anodes. The three cavities are operated simultaneously so researchers can observe the thermal neutron flux, the fast neutron flux, and the gamma ray background. These three different sensitivities are achieved by coating one device in the triad with enriched ^{235}U (thermal neutrons), another with ^{232}Th (fast neutrons), and leaving the third uncoated (sensitive only to gamma rays). The uncoated device can be used much like a compensated ion chamber in which the gamma ray background can be subtracted from the other two neutron sensitive MPFDs. Figures 2 and 3 show the basic components of the third generation MPFDs, and Figure 4 shows a photograph of the MPFD triad parts.

Researchers fabricated the devices by first evaporating Ti/Au layers as the cathodes and anodes. Connections are made possible to the contacts with micro-drilled access holes. The neutron reactive coatings are then plated onto the base pieces and a sealant is used to hold the ceramics together and trap a fill gas in the cavity. Either argon or P-10 (10 percent methane/90 percent argon) is used as the charge-detecting medium. The cavity widths and diameters are both only 1 mm, and additional reservoir cavities have been added to prolong the detection gas lifetime. Such small devices can be analytically treated as point detectors, thereby greatly simplifying the back-projection calculations.

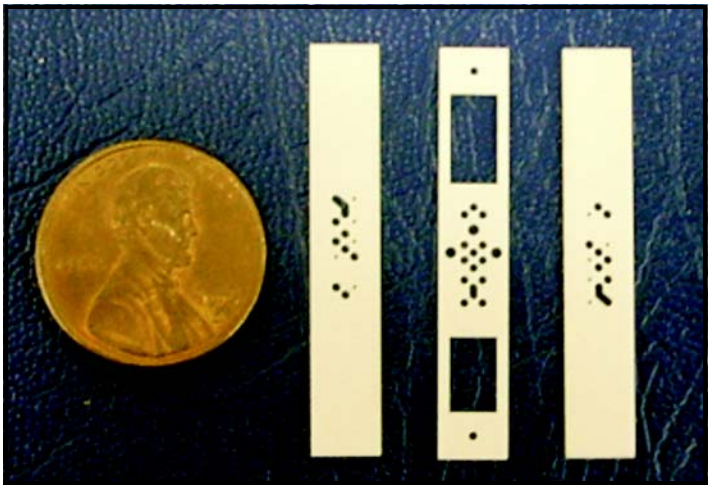


Figure 4. Alumina substrates for final (third) generation of MPFDs, showing the cavity and base pieces that form the detector.

As researchers already observed, argon-filled chambers have an inherently low probability of gamma ray interactions, allowing for natural discrimination of gamma-ray background noise. In addition, radiation cannot destroy the detecting medium because it is gas-filled, which is a clear advantage over liquid-filled or solid detectors. Being a small chamber, less voltage is needed to sweep the electrons and ions across the chamber, thus lowering the power requirement. This will also limit the potential of detecting non-neutron-induced events and will prolong the life of the gas. Finally, third generation MPFDs are significantly smaller than previous generations, and the neutron reactive material is plated under much more stringent conditions to reduce the amount of material deposited. Hence, it is expected that the devices can be made to operate in pulse mode in a neutron flux exceeding 10^{13} n-cm^{-2}-s^{-1}.

In addition to the new low-cost MPFD designs, an automated plating station has been built that allows for sequential plating of ^{235}U and ^{232}Th on a single device without cross contamination. The plating system holds ten cavity pieces per run, is fully automated with feedback sensors, and can be used to apply accurate films of neutron reactive materials. Film mass for the devices is measured with a high-resolution silicon surface barrier detector for activity and compared to a ^{235}U check standard.

Researchers have developed a preamplifier, amplifier, and discriminator for the pulse-mode circuitry. This allows for custom tuning of the system for their specific needs. These systems are simple and economical, built using only a few readily available chips that total less than $10 per detector. Ongoing work includes defining and modeling other portions of the circuitry and developing the circuit board designs.

The researchers are developing mathematical models that can relate the power density profiles in a reactor's fuel rods to the flux densities at the detector locations. Key to this formulation is constructing an appropriate response function that relates the flux at any position in the core to the fast neutrons born at an arbitrary axial depth in any of the core fuel rods. Recent developments show that, with only five triads per string of detectors inserted into a core location and with strings placed in a plane across the reactor, response functions yield promising results, even including statistical fluctuations. The system should permit operators to observe the reactor power density across the detection plane in real time. Current results indicate that the detector array strings must be placed near the fuel rod or bundle that is to be monitored. The goal of the project is to refine this system and demonstrate its ability to perform real-time power density monitoring. At present, researchers are developing new response functions that will allow for faster data unfolding when the entire detector array is positioned in the reactor.

Planned Activities

During the third research year, researchers will deploy 75 triads of MPFDs (coated with ^{232}Th and ^{235}U) into the flux probe holes of KSU's TRIGA Mark II nuclear reactor to observe real-time neutron flux measurements. These triads will be distributed at five triads per string, with 15 strings forming a plane across the reactor core. The signals can be read in both pulse and current modes, and the information accumulated will be used to calculate the power density in the fuel. Real-time core mapping will be performed. Researchers will use the data acquired from the MPFDs for inverse power density distribution calculations. The completion of the third-year goals will demonstrate that MPFDs can be used to simultaneously give information on the nuclear reactor power levels at various locations inside the reactor core.

[1] M.F. Ohmes, D. S. McGregor, J. K. Shultis, P. M. Whaley, A.S.M. Sabbir Ahmed, C. C. Bolinger, T. C. Pinsent, "Development of Micro-Pocket Fission Detectors (MPFD) for Near-Core and In-Core Neutron Flux Monitoring," Proc. SPIE, Vol. 5198 (2003) 234-242.

[2] D. S. McGregor, J. K. Shultis, M. F. Ohmes, A.S.M. Sabbir Ahmed, R. Ortiz , and K. Hoffert, "Micro-Pocket Fission Detectors (MPFD) for Near-Core and In-Core Neutron Flux Monitoring," ANS 4th International Topical Meeting NPIC & HMIT, Columbus, Ohio, September 19-22, 2004.

NUCLEAR ENERGY RESEARCH INITIATIVE

Development of a Supercritical Carbon Dioxide Brayton Cycle: Improving Pebble Bed Reactor Efficiency and Testing Material Compatibility

PI: Chang Oh, Idaho National Engineering and Environmental Laboratory (INEEL)

Collaborators: Thomas Lillo, William Windes, Terry Totemeier, Richard Moore, INEEL

Project Number: 02-190

Project Start Date: October 2003

Project End Date: September 2004

Research Objectives

The very high temperature gas reactor (VHTGR) is a promising technology for high-efficiency electricity generation and high-temperature process heat applications. The U.S. needs to make the VHTGR intrinsically safe and proliferation-resistant; however, certain technical and economic issues must be overcome before this reactor design can be built in the U.S. A nuclear power cost goal of 3.3 cents/kWh is desirable in order to compete with fossil combined-cycle, gas turbine power generation. This goal requires a 30 percent reduction in power cost for state-of-the-art nuclear plants, a large cost differential that can only be overcome by technology improvements that lead to a combination of better efficiency and more compatible reactor materials.

The objectives of this research were (1) to develop a supercritical carbon dioxide Brayton cycle in the secondary power conversion side that can be applied to some Generation-IV reactors such as the high temperature gas reactor (HTGR) and supercritical water reactor, (2) to improve the plant net efficiency by using the carbon dioxide Brayton cycle, and (3) to test material compatibility at high temperatures and pressures. The reduced volumetric flow rate of carbon dioxide compared to helium, due to its higher density, will reduce compression work, which eventually will increase turbine output and enhance the plant's efficiency.

Research Progress

Advanced gas reactor technology has been identified as one of the best passively safe, thermally efficient, proliferation resistant, modular reactor systems capable of electricity and hydrogen production, as well as efficiently burning spent fuel. Internationally, the HTGR concept has become a top priority for implementation because of its inherent safety and near-term implementation feasibility. The U.S. Department of Energy (DOE) indicates that the Next Generation Nuclear Plant (NGNP) will be built at the INEEL site by 2017. In order to resolve technical issues, this project consists of three major tasks. All tasks have been performed on schedule and budget, and their technical accomplishments are summarized below.

Task 1. Development of CO_2 Brayton Cycle. The proposed supercritical carbon dioxide Brayton cycle deals with high pressures and temperatures. At these conditions, an ideal gas law isentropic compression and isentropic expansion cannot be applied because of real gas effects in helium and carbon dioxide associated with non-ideal compressibility and expansion. Therefore, researchers needed to develop analytical equations for polytropic expansion and compression through a sequence of turbines and compressors, respectively. This is just for scoping analyses of parametric studies aimed at investigating the effect of the overall plant efficiency. For the detailed computation of the balance of plant (BOP) efficiency calculations, a CO_2 database is required to make accurate calculations.

Researchers compared a number of equations of state and carbon dioxide databases and determined that the National Institute of Standards and Technology (NIST) CO_2 database is the most accurate, having properties consistent with those referenced in Perry's Handbook. They also investigated and compared the Aspen Plus and HYSYS computer codes for BOP process optimization. Both codes agree well with conditions defined in a simple reference design, with results deviating less than 0.5 percent. However, Aspen Plus is limited to isentropic expansions without multiple turbine performance curve capabilities. Therefore, the HYSYS code was used for the calculations.

Researchers also utilized a numerical model that was originally developed at the Massachusetts Institute of Technology using the Visual Basic (V-B) computer language and later revised by INEEL by incorporating the NIST CO_2 database and a pressure drop equation. The results from HYSYS were compared with those from the V-B model and found to agree very well for the 3-shaft baseline case. The CO_2 Brayton cycle gives 51 percent plant efficiency, which is an improvement over the 47 percent for the helium Brayton cycle using the same BOP layout. The improvement in plant efficiency is attributed to the reduced volumetric flow of carbon dioxide compared to that of helium, which results in less work for the compressors.

Task 2. Improvement of HTGR Net Efficiency. The objective of this task was to determine the overall plant cycle efficiency by the integration and optimization of each of the components used in the power conversion side. In order to perform this task in an efficient manner, researchers used their original reference design with a three-shaft turbo-machinery arrangement and helium as the working fluid to conduct a parametric study with the HYSYS code and to optimize the cycle. In parallel, they also used the V-B model with an appropriate friction pressure drop equation for a pebble bed reactor. This term is important in determining the net power plant efficiency and is not directly calculated by the HYSYS model. A similar pressure drop equation can be input for a prismatic NGNP core. After the helium Brayton Cycle loop was optimized, a number of HYSYS simulations with supercritical CO_2 were completed.

A number of important parameters were investigated for this study including: various temperature differences across the reactor; reactor inlet temperatures; inlet cooling temperatures to compressors; effectiveness factor of the intermediate heat exchanger (IHX) and recuperator; and efficiencies of the compressors, turbines, and other components. Results for a 250-MW thermal helium Brayton cycle using a 92-percent effectiveness factor for the intermediate heat exchanger and 90-percent polytropic efficiency for the turbines and compressors indicate that the maximum cycle efficiency peaks at 45 percent, corresponding to a relatively low reactor outlet temperature of 850°C and a reactor inlet temperature of 520°C. As the reactor outlet temperature is allowed to increase to 1,000°C, the maximum efficiency increases to 51.5 percent with the corresponding reactor inlet temperature increasing from 520°C to 640°C.

The effect of compressor efficiency on the overall efficiency of the Brayton cycle was obtained by varying the compressor polytropic efficiency from 90 to 94 percent and using a constant reactor outlet temperature of 900°C for all three compressor efficiencies. The results indicate that the cycle efficiency ranges from 48.2 percent to 50.2 percent, with the maximum efficiencies all occurring at a reactor inlet temperature of 550°C.

A practical way of reducing the compressor work is to keep the specific volume of the gas as small as possible during the polytropic compression. This is achieved by maintaining the temperature of the gas as low as possible because specific volume is proportional to temperature. By dividing the compression process into stages and cooling the gas between stages, the total work done during the compression process is reduced. By reducing the compressor inlet temperature to 5°C, the overall cycle efficiency increases by 0.65 percent.

The project team also investigated the sensitivity of the effectiveness of the IHX on the overall cycle efficiency. If the IHX effectiveness is improved from 90 percent to 92 percent at a core outlet temperature of 950°C and a core inlet temperature of 400°C, for example, there is an initial improvement in the overall Brayton efficiency of 0.65 percent. The IHX effectiveness has less impact on the overall cycle efficiency compared to improvements in compressor efficiency.

In order to validate the HYSYS and V-B models, a simple one-shaft Brayton cycle layout was used, incorporating a GTHTR-300 reference design. As shown in Figure 1, the GTHTR-300 is a direct cycle plant that consists of three subsystem modules including a reactor with a prismatic core, a gas turbine generator module with one turbine, one compressor, a generator on a single shaft in a horizontal arrangement, and a heat exchanger module with one recuperator and one precooler.

The model assumed values of 92.8-percent turbine polytropic efficiency, 90.5-percent compressor polytropic efficiency, 95-percent recuperator effectiveness, and 28°C compressor inlet temperature, along with the same conditions of other parameters in both the V-B and HYSYS models. The V-B model gives lower net cycle efficiencies than those of HYSYS, except at the maximum efficiency point. The one difference between the HYSYS and V-B models lies in a different helium property database. The V-B model uses the NIST database while HYSYS uses an equation of state to define the helium properties. A CO_2 pressure-enthalpy diagram was used to check the accuracy of HYSYS simulation. The results calculated by HYSYS agree very well with those calculated using the CO_2 pressure-enthalpy diagram.

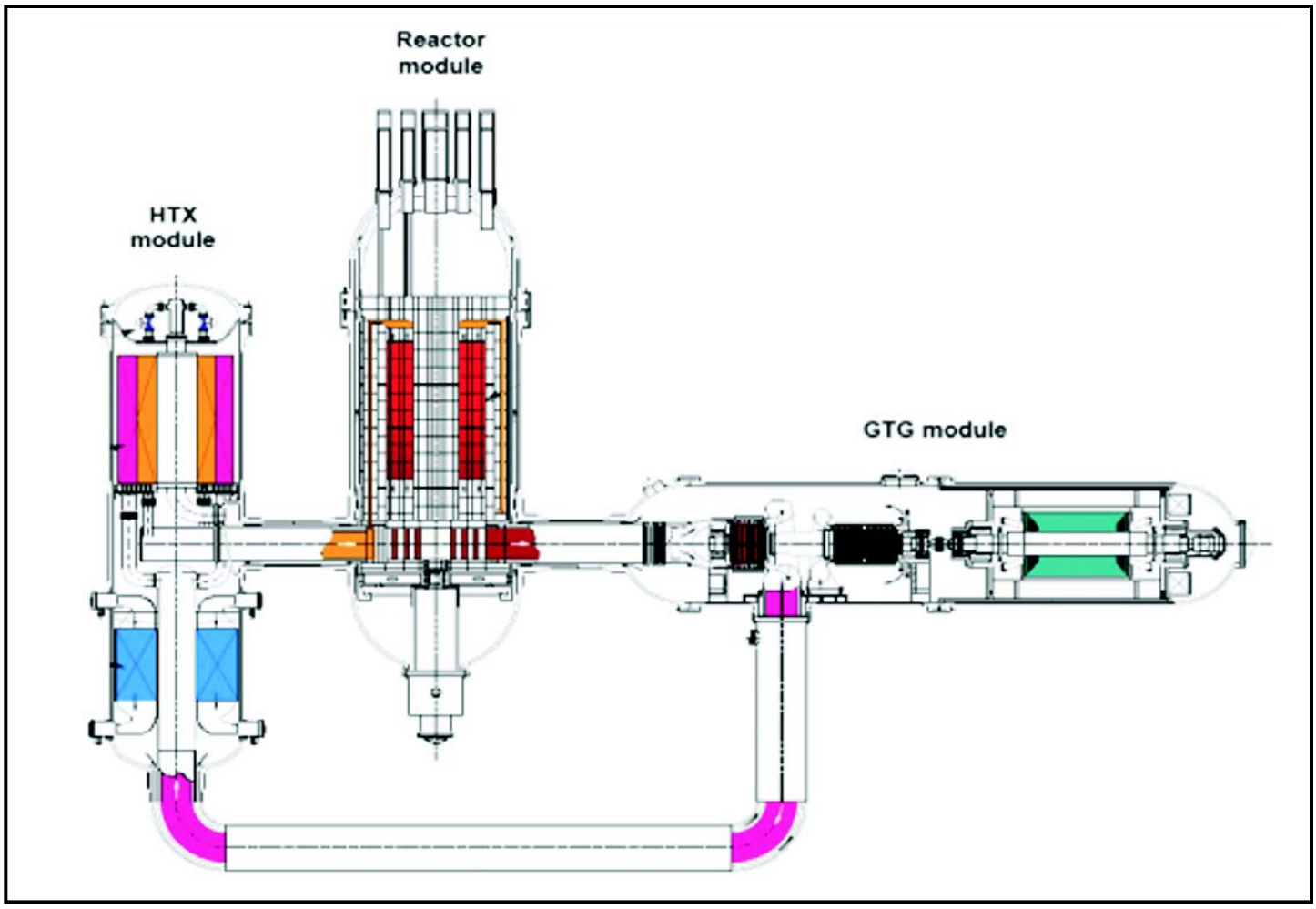


Figure 1. GTHTR-300.

A parametric investigation was made with a supercritical CO_2 cycle using a 250-MW, three-shaft reference design. Three different CO_2 secondary system pressures of 6.3 MPa, 13 MPa, and 20 MPa resulted in cycle efficiencies of 48 percent, 49.3 percent, and 50.3 percent, respectively, which are not significantly different from each other. However, over the lifetime of a power plant, a 2.3 percent increase in the plant efficiency can result in substantial savings.

Researchers investigated the following working fluids: helium for both direct and indirect cycles, nitrogen for an indirect cycle, and carbon dioxide for an indirect cycle. The difference in efficiency between the helium direct cycle and the indirect cycle was 50.9 percent vs. 48.7 percent, respectively. Nitrogen gave a cycle efficiency of 45.5 percent, while CO_2 gave 50.7 percent. The total area ratio (defined as total heat transfer area of working fluid divided by total heat transfer area of a helium indirect cycle) gives 0.65 for the helium direct cycle, unity for the helium indirect cycle, 1.32 for the nitrogen indirect cycle, and 1.18 for the CO_2 indirect cycle at 20 MPa.

Conclusions from the investigation are that among the three working fluids studied, CO_2 has the highest cycle efficiency due to less compression, and would also require the smallest size turbo-machinery. A helium direct cycle eliminates the intermediate heat exchanger and consequently requires the smallest heat transfer area of the fluids considered, due to its higher heat capacity and thermal conductivity.

Theoretically, a combination of reheat and intercooling increases the cycle efficiency in a closed loop. Preliminary multiple reheat with a number of intercooling passes were investigated using a molten salt (Flibe) in the primary side and helium in the power conversion loop. This configuration yields 56-percent cycle efficiency. Further investigation of a molten salt/CO_2 cycle is being considered.

Task 3: Material Compatibility. Research on the creep behavior and corrosion resistance of Inconel MA 754 in supercritical carbon dioxide continued during FY 2004. The creep behavior of both coarse-grained as well as fine-grained Inconel MA 754 was documented. Creep

microstructures were documented and fracture analysis was carried out on failed creep samples. The results showed that MA 754 did, indeed, exhibit superior creep resistance at temperatures approaching 1,000°C. The material, however, displayed lower creep rates in a direction perpendicular to the extrusion direction, indicating that creep rate is a function of direction in the material. Also, it exhibited low creep ductility, less than 1 percent, at elevated temperatures. Fine-grained MA 754 demonstrated lower creep resistance but higher creep ductility. A manuscript documenting the results was prepared and submitted to a peer-reviewed journal for publication.

Final modifications were made to a supercritical carbon dioxide loop to document the corrosion behavior of MA 754 in flowing supercritical CO_2 at 1,000°C and 1,500 psi. Experiments were carried out at exposure times ranging from 47 to 335 hours. The time-averaged corrosion rate was found to decrease as the exposure time increased. The corrosion rate from the 335-hour exposure test equated to less than 1 mm/year. Microscopic examination indicated that a corrosion layer formed and grew slowly, which protected the base material from further corrosion.

Overall, it was concluded that MA 754 possessed high creep resistance at 1,000°C, better than other high-temperature metallic alloys, and has acceptable corrosion resistance to supercritical carbon dioxide. MA 754 seems acceptable for application in components of a supercritical CO_2 Brayton cycle.

Highlights of the annual activities are summarized below:

- Completed development of CO_2 Brayton cycle, including a number of baseline calculations.
- Completed parametric studies to determine the sensitivity of overall cycle efficiency to key parameters.
- Validated that compressor exit conditions of a supercritical CO_2 Brayton cycle, calculated using HYSYS, agreed very well with those determined from a CO_2 pressure-enthalpy diagram through sequential compression steps.
- Completed an evaluation of three working fluids for the indirect power conversion study. Results indicated that supercritical CO_2 has the highest cycle efficiency due to less compression work because of its higher density, resulting in the smallest turbomachinery components.
- Determined that a helium direct cycle, which eliminates an intermediate heat exchanger, consequently requires the smallest heat transfer area due to the higher heat capacity and thermal conductivity compared to other fluids.
- For the final selection of the best working fluid or fluid mixture, trade-off studies need to be performed for efficiency, capital cost, maintenance cost, and the stability of fluids through compressor, potential leakage from PCS, and other relevant issues. Researchers will include some of these issues in their FY 2005 efforts.
- Published two technical papers and had a third accepted by a peer review journal.
- Characterized the creep deformation of coarse-grain MA 754, including high-temperature mechanical properties, and determined the mechanical and creep properties of fine-grained MA 754. A manuscript describing the results was accepted for publication in a peer-reviewed journal.
- Completed corrosion testing of MA 754 in supercritical CO_2.
- Characterized the creep microstructures of MA 754.

Planned Activities

- Continue comparing other cycle configurations, including a combined cycle, recompression, and multiple reheat cycle. The cycle will be optimized using HYSYS.
- Investigate an intermediate flow loop heat transfer study for NGNP hydrogen, if time is allowed.
- Complete corrosion testing of fine-grain MA 754 and compare the corrosion behavior to coarse-grained MA 754.
- Initiate and complete corrosion testing of alternate alloys. Researchers plan to document the corrosion behavior of I-617, an alloy receiving considerable attention for use in various high-temperature areas of the NGNP.
- Complete the summary report on MA 754, including recommendations on the suitability of MA 754 for use in a supercritical Brayton cycle.

NUCLEAR ENERGY RESEARCH INITIATIVE

Hydrogen Production Plant Using the Modular Helium Reactor

PI: Arkal Shenoy, General Atomics

Collaborators: Idaho National Laboratory, Texas A&M University

Project Number: 02-196

Project Start Date: September 2002

Project End Date: September 2005

Research Objectives

The primary objectives of this project are to develop a conceptual design of a modular helium reactor (MHR) for the production of hydrogen and to provide DOE, utilities, and energy-policy planners the information needed to make decisions regarding additional research and development for producing hydrogen from nuclear energy. To achieve these objectives, the work will be structured to satisfy five major goals:

- Develop functions and requirements for a hydrogen production plant coupled to an MHR using a systems engineering approach
- Develop a conceptual MHR system design for supplying process heat and/or electricity to an H_2 production system
- Develop conceptual designs for a thermochemical H_2 production system and a high-temperature electrolysis (HTE) H_2 production system that receives process heat from an MHR
- Develop a conceptual design for thermochemical- and HTE-based H_2 production plants that integrate the reactor and hydrogen production systems
- Complete assessments of the plant design with respect to performance, safety, economics, and licensing.

Research Progress

Work performed during the second year of the project is summarized below.

Plant Functions and Requirements. During the second year, researchers made minor modifications to the H_2-MHR Functions and Requirements Document based on reviewer comments. In addition, they began developing functions and requirements for an HTE-based hydrogen-MHR plant, which is receiving increased interest as a possible alternative to the sulfur-iodine (S-I) process for hydrogen production. The HTE-based plant would use solid oxide electrolyzers (SOEs) operating at high temperature to produce hydrogen with high efficiency. For this concept, most of the heat produced by the MHR is converted to electricity using a Brayton-cycle power conversion system, and the remaining heat is used to generate the steam that is supplied to the SOE. Researchers developed a flow schematic and a function flow block diagram to define the higher level functions for the HTE-based H_2-MHR plant.

Core Nuclear/Thermal Hydraulic H_2 Design. The following table shows design points adopted for the H_2-MHR conceptual design:

Core thermal power	600 MWt
Coolant inlet temperature	590°C
Core-average coolant outlet temperature	950°C
Fuel design basis	SiC-TRISO fuel
Peak SI process temperature	900°C

Previous studies indicate that it should be possible to increase the coolant outlet temperature from 850°C to 950°C without a proportional increase in fuel temperature. Researchers will accomplish this through modest design modifications to reduce the bypass flow fraction and optimize the physics design, thereby reducing power peaking factors.

Increasing the coolant inlet temperature from 490°C to 590°C may require some design modifications, including alternative inlet flow configurations that place more thermal resistance between the location of the inlet flow path and the reactor vessel (see Figure 1). Preliminary assessment of these modifications indicates that routing the inlet flow through the permanent side reflector can lower vessel temperatures by about 50°C during normal operation.

Researchers also studied the effects of using a small flow stream of colder helium (at 140°C) for vessel cooling during normal operation in a manner similar to the

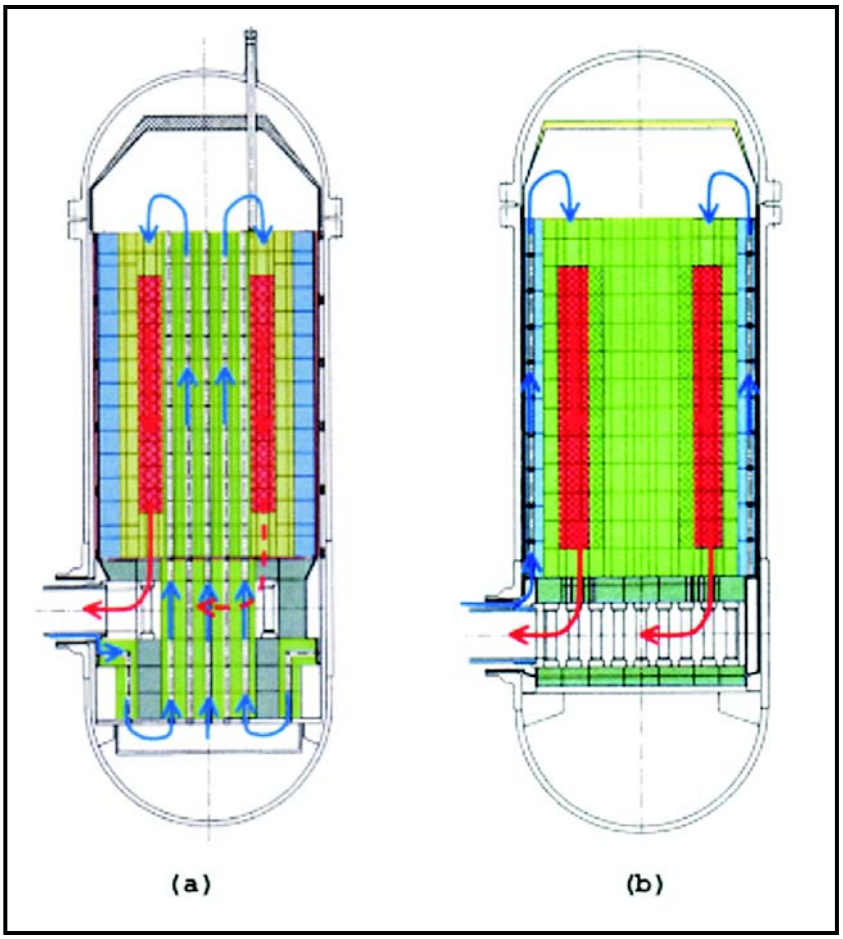


Figure 1. GT-MHR reactor vessel configured with alternative inlet flow paths. (a) Flow routed through central reflector. (b) Flow routed through permanent side reflector.

Gas Turbine High Temperature Reactor (GTHTR) -300 design being developed by Japan Atomic Energy Research Institute (JAERI). Results were consistent with the JAERI results and showed a significant reduction in vessel temperatures during normal operation. Additional studies are needed to determine if a flow configuration of this type could be implemented for the H_2-MHR without impacting passive safety.

Replacement of silicone carbide Tri-Isotropic (SiC-TRISO) fuel with zirconium carbide (ZrC)-TRISO fuel is a potential option for increasing the margins on fuel performance at higher temperatures. Studies have shown that zirconium adds additional neutron poison, but its effects can be compensated for by reducing the amount of fixed B_4C burnable poison.

Fuel Performance/Fission Product Transport. Investigators compared the General Atomics (GA) MHR and JAERI GTHTR-300 fuel designs. The GA two-particle (fissile and fertile) system has an effective enrichment that is nearly the same as the JAERI single particle system. However, the GA design provides more flexibility with regard to zoning enrichment, which helps to reduce power-peaking factors. This capability, along with the lower thermal resistance of the GA block design relative to the JAERI pin-in-block design, are the main reasons why the peak fuel temperatures during irradiation are lower for the GA reactor design (1,250°C vs. 1,350°C). The GA and JAERI particle designs have about the same internal pressure from fission gases at the end of irradiation. However, because the JAERI particle design has no carbide phase in the kernel to getter excess oxygen, a significant carbon monoxide partial pressure develops that actually exceeds the partial pressure from fission gases. Because of this, the JAERI design places much more importance on performance of the pyrocarbon layers to provide the compressive forces needed to compensate for internal pressure. The high CO pressure can also cause increased failure by the kernel migration mechanism (amoeba effect), and can contribute to corrosion of the SiC layer during accident conditions.

S-I Process Flow Sheet. Researchers performed studies to determine the effect of increasing the pressure during sulfuric acid decomposition to as high as 7.5 MPa, in order to better match the pressure in the secondary helium loop and to simplify the design of process heat exchangers. The higher pressure shifts the equilibrium such that a greater fraction of the sulfuric acid solution requires recycling. However, the impacts on efficiency are not significant since the heat is retained in the recycled sulfuric acid. The higher pressure results in increased mass flow rate through the decomposer, but the volumetric flow rate is reduced. In general, these studies have shown that there is no major detriment to operating the sulfuric acid decomposer at higher pressure. The benefits include lower differential pressures across the heat exchanger walls, which results in lower stresses and longer component lifetimes, and the ability to use thinner-walled heat exchangers, which enhances heat transfer.

Availability Assessment. Researchers developed and refined a SAPHIRE model to evaluate the reliability and availability of the hydrogen production system. They began the SAPHIRE modeling process by developing a master fault tree for the S-I process. This fault tree includes three

transfer gates that link to the individual fault trees for each of the chemical reaction sections in the S-I process (i.e., the Bunsen reaction section, the H_2SO_4 decomposition section, and the Hydrogen-Iodine [H-I] decomposition section). They also used a house event with the transfer gate to provide the fault tree model, which was capable of analyzing each of the sections in the process flowsheets separately or together. This master fault tree, when linked to the individual fault trees for each of the three separate chemical reactions, provides the basis for evaluating and improving overall plant reliability and assessing plant availability based on component failure rates and mission times.

Researchers prepared and presented the following papers at technical conferences:

- "Hydrogen Generation Using the Modular Helium Reactor," 12th International Conference on Nuclear Engineering (April 25-29, 2004, Arlington, VA).
- "Assessment of MHR-Based Hydrogen Energy Systems," 15th World Hydrogen Energy Conference (June 27 - July 2, 2004, Yokohama, Japan).
- "MHR-Based Hydrogen Production Systems," 2004 International Congress on Advances in Nuclear Power Plants (ICAPP-04, June 13-17, 2004, Pittsburgh, PA, embedded with the 2004 American Nuclear Society Annual Meeting).
- "Thermal Hydraulic Design of a Modular Helium Reactor Core Operating at 1,000°C Coolant Outlet Temperature," The 6th International Conference on Nuclear Thermal Hydraulics, Operations and Safety (NUTHOS-6), Nara, Japan, October 4-8, 2004.

Planned Activities

Researchers will continue to work with the H_2-MHR conceptual design in the areas defined by the work breakdown structure for the project. Specific tasks planned for Year 3 include additional work to optimize the reactor system, S-I process design, and the HTE process design. The project's final report will consist of conceptual design descriptions for the S-I-based plant and the HTE-based plant.

NUCLEAR ENERGY RESEARCH INITIATIVE

Nuclear Reactor Power Monitoring Using Silicon Carbide Semiconductor Radiation Detectors

PI: Don W. Miller and Thomas E. Blue, The Ohio State University

Collaborators: Westinghouse Science and Technology Center, General Atomics

Project Number: 02-207

Project Start Date: October 2002

Project End Date: September 2005

Research Objectives

The objective of this research program is to investigate the use of silicon carbide (SiC)-based sensor arrays as ex-core neutron monitors in advanced reactor designs. Specifically, the International Reactor Innovative and Secure (IRIS) and the prismatic-core, gas turbine, modular, helium-cooled reactor (GT-MHR) designs are being modeled. This investigation has two primary tasks: (1) to analytically define the ideal location for SiC-based sensor arrays in the IRIS and GT-MHR nuclear power plants and use the predicted neutron flux levels and environmental conditions at these locations to estimate the performance of the SiC sensor arrays and (2) to design, construct, and evaluate a high-speed pulse counting system, which will be used in the SiC neutron sensor channels to optimize sensor performance in the power monitoring system.

Research Progress

Task 1. In placing SiC detectors within a reactor, there is a conflict between achieving the high count rate necessary to generate a scram signal in an appropriately small time-frame, and the risk of degrading the detector through radiation exposure. In order to predict the detector count rate and to quantify the radiation exposure in terms that are technically precise and relevant, researchers calculated the neutron and gamma-ray radial flux distributions in the Reactor Cavity (RC) and in the Reactor Cavity Cooling System (RCCS) resulting from fission events within GT-MHR.

Researchers from The Ohio State University (OSU) modeled the GT-MHR reactor in 3-dimensions using the radiation transport code MCNP, accounting for radial and axial variations of the core power distribution. They modeled various regions of the core as physically distinct (e.g., the fueled region of the core as distinct from the various reflector regions) and treated the fueled region as a homogeneous mixture. They calculated the neutron and gamma-ray flux distributions as a function of axial position for the following three radial locations:

- Within the central reflector, for detector capsules located at radial distances R=117 and R=153 cm from the central reflector centerline
- In the RC at R=505 cm
- In the RCCS at R=610 cm

OSU researchers used this data to predict the detector count rate and the $1\ MeV$ equivalent neutron flux in SiC ($\phi^{Total}_{eq,1MeV,SiC}$) at the respective detector locations.

$\phi^{Total}_{eq,1MeV,SiC}$ for SiC was calculated using the general expression given in Eq. (1):

$$\phi^{Total}_{eq,1MeV,mat} = \int_0^{\infty} \frac{F_{D,mat}(E)}{F_{D,1MeV,mat}} \phi(E) dE \quad (1)$$

where $F_{D,mat}(E)$ is the damage function for the material of interest (SiC) evaluated at energy E; $F_{D,1,MeV,mat}$ is the damage function for the material of interest (SiC) evaluated at energy $E=1\ MeV$; and $\phi\ (E)$ is the energy-dependent neutron flux density function. Table 1 presents $\phi^{Total}_{eq,1MeV,SiC}$ averaged over all axial layers of the core for the GT-MHR with radius as a parameter.

Researchers characterized the radiation field in Position 4 in the semi-conductor device characterization vessel in

Detector Radial Location (cm)	117	153	505	610
$\phi^{Total}_{eq,1MeV,SiC}$ (n/cm^2-s)	3.50E+11	3.75E+12	9.68E+07	1.95E+07

Table 1. 1 MeV equivalent neutron flux $\phi^{Total}_{eq,1MeV,SiC}$ in SiC at various locations within the GT-MHR.

Beam Port 1 of OSU's Research Reactor (OSURR) in terms of $\phi^{Total}_{eq,1MeV,SiC}$ in order to correlate observed degradation of the SiC detectors in the OSURR with the degradation that can be expected for various possible detector locations in the GT-MHR. The values of ϕ (E) that were used in the calculations were determined using foil activation data and the SAND-II neutron energy spectrum deconvolution code. The results of the calculations are $\phi^{Total}_{eq,1MeV,SiC}$ = 7.20 x 10^{11} neutrons $cm^{-2}s^{-1}$ for operation at nominal full power of 500 kW.

By comparing the value of $\phi^{Total}_{eq,1MeV,SiC}$ for the GT-MHR with the value for the OSU research reactor, the investigators concluded that SiC devices can be adequately tested for the radiation damage that would be incurred over a GT-MHR refueling cycle. This conclusion is valid for detectors located in the RC and RCCS, but researchers believe that this would not follow for detectors located in-core in the GT-MHR.

Researchers also used calculations of the neutron radial flux distributions in the RC, the RCCS, and the central reflector of the GT-MHR to predict the maximum triton count rate at the respective detector locations. The maximum triton count rate for capsules at Z=520 cm for full power operation (P=600 MWt) is presented in Table 2 for radial locations of 117, 153, 530, and 610 cm for a detector with a 500 μm diameter and a 1 μm thick LiF radiator with 90 percent ^{6}Li enrichment.

Detector Radial Location (cm)	117	153	530	610
Maximum Triton Count Rate	2.4 E+5 T/s	1.7 E+06 T/s	5 T/s	40 T/s

Table 2. Maximum triton count rate at various locations within the GT-MHR at Z=520 cm for full power operation (P=600 MWt).

Researchers predicted that the depletion of ^{6}Li for one cycle would be about 0.9 percent, if the detector is positioned at R=153 cm and Z=320 cm, and would be 0.1 percent if the detector is positioned at R=117 cm and Z=320 cm.

Task 2. The team continued developing the simulation model of the counting channel from the output of the SiC detector to the channel's end with the addition of a distributed parameter model of the cable, using the program PSpice and the inclusion of Matlab models of a voltage-sensitive preamplifier and discriminator. From the model, they concluded the following:

- The proposed electronic channel should be able to monitor the neutron flux with a count rate of 4.0 E+7 counts with a 9 percent dead time loss.
- The pulse shape is not so greatly altered by the use of mineral-insulated cable that a large increase in the counting system dead time should result.
- With a voltage sensitive preamplifier of the type that they specified, the speed of the channel is limited by the discriminator dead time.

Researchers purchased equipment, constructed the counting channel, and measured detector pulses. One such pulse is shown in Figure 1. In general, the measured pulses compared well with the simulations.

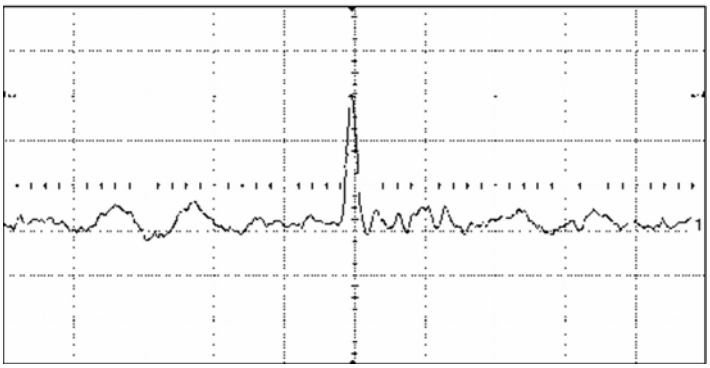


Figure 1. The preamplifier output with a voltage scale of 50 mV/div and a time scale of 5 ns/div.

Planned Activities

Task 1. OSU researchers will predict $\phi^{Total}_{eq,1MeV,SiC}$ in the IRIS. Then they will measure the degradation of SiC detectors in the OSURR and translate the observed degradation to the degradation that can be expected for various detector locations in the GTMHR and IRIS, using their knowledge of $\phi^{Total}_{eq,1MeV,SiC}$ in the OSURR.

Task 2. Researchers will include the discriminator in the pulse-counting system and work toward ensuring that the entire detector channel functions properly.

7. Advanced Nuclear Fuels/Fuel Cycles: Project Summaries

This research area includes 7 NERI research projects, of which 1 was awarded in FY 2001 and 6 in FY 2002. It includes research and development to provide measurable improvements in the understanding and performance of nuclear fuel and fuel cycles with respect to safety, waste production, proliferation-resistance, and economics, in order to enhance the long-term viability of nuclear energy systems. This effort includes enhanced performance of fuels for advanced systems, development of fuels capable of withstanding the conditions in the supercritical light water reactor (LWR) regime, and development of advanced proliferation-resistant fuels capable of high burn-up such as those needed in support of the Generation IV concepts.

The scope of this long-term R&D encompasses a variety of thermal and fast spectrum power reactor fuel forms, including ceramic, metal, hybrid (e.g., cermet or cercet), and liquid, as well as fuel types such as oxides, nitrides, carbides, and metallics. Enabling technologies such as advanced cladding, water chemistry, and alternative moderators and coolants are also considered. The fuel cycle research includes consideration of advanced enrichment technologies for fuel and burnable absorbers and considers the impact of fuel cycle options on the proliferation of nuclear weapons materials. R&D topics also include development of higher density low-enriched uranium (LEU) (<20 percent U-235) fuels for research and development reactors.

Currently, selected projects include innovative concepts for the following:

- Material preparation and production of nuclear fuels.
- Inherently safe fuel designs and core response.
- Study of life-limiting phenomena for high burn-up or long-life fuels.
- High-temperature fuel and material performance.
- Critical safety data and reactor physics data for advanced fuel compositions and enrichments above five percent.
- Innovation in fuel design, composition, or other attributes that maximize energy production, optimize fissile material utilization, or reduce production costs.

NUCLEAR ENERGY RESEARCH INITIATIVE

Directory of Advanced Nuclear Fuels/Fuel Cycles Project Summaries

NUCLEAR ENERGY RESEARCH INITIATIVE

High-Performance Fuel Design for Next Generation PWRs (Annular Fuel Project)

PI: Mujid S. Kazimi, Massachusetts Institute of Technology

Collaborators: Gamma Engineering Corporation, Westinghouse Electric Corporation, Duke Engineering & Services (now Framatome ANP), Atomic Energy of Canada Limited

Project Number: 01-005

Project Start Date: August 2001

Project End Date: July 2005

Research Objectives

The overall objective of this NERI project is to examine the potential for a high-performance advanced fuel for pressurized water reactors (PWRs), which would accommodate a substantial increase of core power density while simultaneously providing comparable (or larger) thermal margins than current PWRs. This advanced fuel will have annular geometry that allows internal and external coolant flow and heat removal (see Figure 1). Following are the detailed tasks of this project:

1. Identify the most promising fuel assembly arrangement for PWRs to achieve a significant increase in power density of at least 30 percent.
2. Optimize the fuel for superior thermal hydraulic and safety performance. Examine the flow distribution, core pressure drop, departure from nucleate boiling ratio (DNBR), and resistance to parallel channel instabilities. Perform safety analyses, such as loss of coolant accident (LOCA) analyses, to confirm safety benefits.

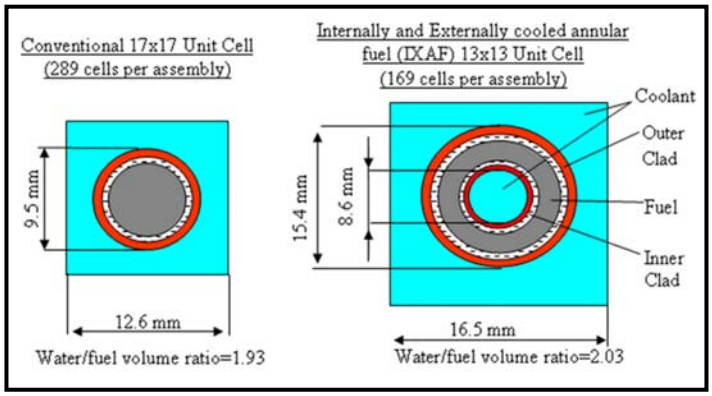


Figure 1. Annular fuel geometry compared to conventional solid PWR fuel.

3. Evaluate the neutronic fuel design to achieve high reactivity, limited burn-up, and a reasonably long refueling cycle to attain good economic features. Confirm the acceptability of the coefficients for reactivity feedback and control.
4. Select fabrication processes to produce annular fuel elements with the required product characteristics, including fissile loading and high integrity cladding, which are capable of eventual scale-up into efficient production processes for economic and reliable fuel element performance.
5. Evaluate the performance of UO_2 fuel forms obtained by production technologies different from current U.S. practices (e.g., vibropacked fuel), and operating under new conditions (such as very low fuel temperature) with regards to fission gas release and fuel dimensional properties during burnup. Develop models for assessing fuel performance as well as for scoping irradiation tests performed at the Massachusetts Institute of Technology (MIT) research reactor.
6. Estimate the electricity cost in cases of using the annular fuel for uprating current Generation II PWRs and in new advanced PWRs.

Research Progress

Task 1. Thermal Hydraulic and Mechanical Design and Safety Analysis. The DNBR analyses using a whole-core VIPRE model of a typical 4-loop Westinghouse plant with annular fuel rods show that 13x13 and 12x12 arrangements yield the highest minimum DNBR (MDNBR). Because thermal expansion and swelling of fuel pellets during operation are expected to be towards the outer cladding,

and the 13x13 array accommodates higher heat flux to the outer channel, the 13x13 array (see Figure 2) was selected as the most promising design. This design allows a 50-percent power uprate, in terms of DNBR limit, if the flow rate is increased proportionally. The extracted power from the same core size thus increases plant output from the current 1,150 MWe to 1,750 MWe. The same power uprate capability was also shown to be possible for the largest currently available reactor coolant pumps, which deliver smaller flow rates than 150 percent if the core inlet temperature is reduced by 10°C. Similar power uprates with annular fuel are found possible for the hexagonal lattice cores typical of Russian VVER reactor designs. At such high power, the peak fuel temperature is still about 1,300°C lower than the solid fuel in today's PWRs. The high increase in flow rate corresponding to the higher power results in a larger pressure drop, which in turn raises vibration and fuel assembly lift-off concerns. However, vibration analyses showed that annular fuel, even at 150-percent flow rates, is more resistant to various modes of vibration than solid fuel, due to its high rigidity. Annular fuel design was also found to be resistant to both excursive (Ledinegg) instability and density wave instability. However, hydraulic lift-off forces are several times higher than for the reference fuel and will require modifications of the fuel assembly and fuel rod holding mechanisms.

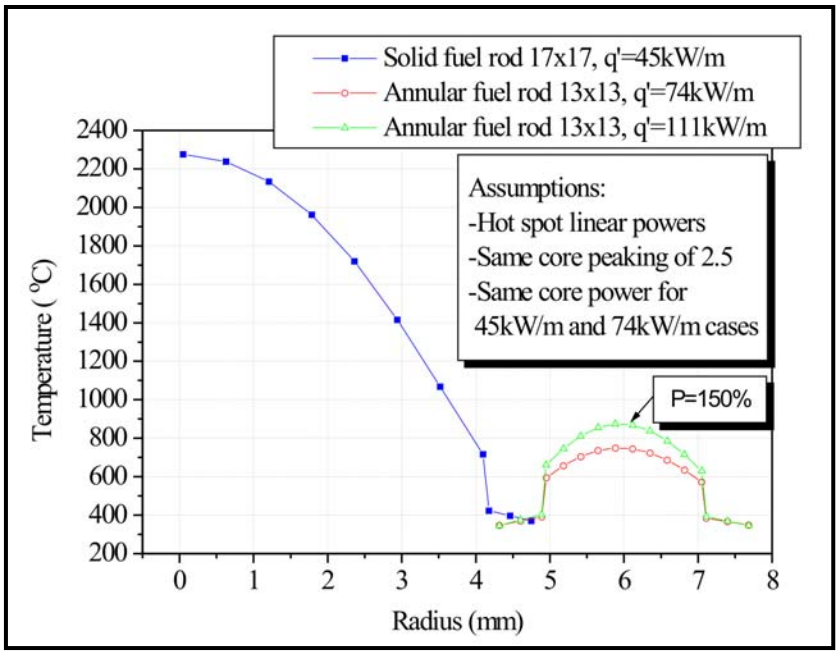


Figure 2. The graphic compares performance of solid and annular fuel.

Researchers developed a RELAP5/MOD3.2 model of a Westinghouse 4-loop plant and used it for analysis of Large Break LOCA, complete Loss of Flow, and Main Steam Line Break accidents. LOCA analysis showed that the peak cladding temperatures of an annular fuel core operating at 150-percent power, with a safety injection system uprated to 150-percent capacity remained well below the safety limit of 1,200ºC. This peak occurred during the fuel heatup period from decay heat, while the early blowdown peak was eliminated due to the very small stored energy of annular fuel. The loss of flow results confirmed the conclusion from earlier steady-state MDNBR analyses at overpower, which showed the capability of annular fuel to operate at 150-percent power. The main steam line break results for annular fuel operating at 150-percent power had MDNBR margins during the accident comparable to those of the solid fuel at 100 percent. In addition, rod ejection analysis results showed that the annular fuel temperatures would remain significantly below those of the solid fuel, indicating substantial margin to the fuel energy deposition limits.

Task 2. Reactor Core Physics and Fuel Management. Using the CASMO/SIMULATE code package, researchers modeled the annular fuel design for both 100-percent power and 150-percent power PWR cores with Internally and Externally Cooled Annular Fuel (IXAF). The design targets included assurance of core operation at a 90-percent capacity factor for an 18-month cycle, maximum core boron concentration of 1,750 ppm, maximum radial pin power peaking of 1.65, and a hot spot factor of 2.5. The generic annular core design philosophy follows the prevailing industry practice of 3-batch cores. The 72 reload assemblies were subdivided into two enrichment levels in groups of 48 and 24, with higher and lower enrichment, respectively. The sub-batch with higher enrichment stays in the core for 3 cycles whereas the lower enriched fuel remains for 2 cycles, but in higher flux regions to achieve comparable burnup.

Without fuel design changes, the IXAF uprated cores would require higher enriched fuel beyond the 5 $^{w}/_{o}$ licensing limit of most commercial nuclear plants, since 50 percent more energy is generated within the fixed cycle length of 18 months. However, if higher density uranium compounds, such as uranium nitride, can be considered and the number of reload assemblies can be increased, the fuel enrichment can be maintained within 5 $^{w}/_{o}$. The fuel burnup increased significantly above conventional cores, reaching a core average discharge burnup of 85 MWd/kg.

The high enrichment fuel affects the core parameters considerably. Because the reactivity worth of control materials is reduced due to neutron spectrum hardening, satisfying the target core boron concentration requires more burnable poison in the fresh fuel. Management of the power distribution through the entire cycle becomes more challenging in order to devise a feasible pattern to ensure acceptable burnable poison residue. Although the reactivity feedback effects are comparable to a solid fuel core, the shutdown margin is reduced for the IXAF cores based on conventional silver-indium-cadmium control rods. Overall, comprehensive reactor physics analyses have shown that IXAF cores for 150-percent power can be designed with steady-state performance, including power distribution and reactivity coefficients, comparable to a solid fuel core. From the point of view of reactor physics, the new IXAF fuel design is also deployable in the next-generation, high-power-density PWRs.

The researchers also explored the potential for burning plutonium in a high power density core with annular fuel. The annular, fertile-free, Pu-loaded, and highly uprated core appears to be feasible, but power uprates in these cores is restricted to less than 50 percent because of relatively high power peaking and a potential for slightly positive moderator temperature coefficient at beginning-of-cycle. A power density increase of 40 percent has an initial Pu loading that is sufficiently low to assure acceptable core power peaking and negative temperature coefficient at all times during the cycle. However, the need for a large amount of enriched burnable poisons makes the design of a fertile-free, Pu-burning core economically unattractive.

Task 3. Fuel Fabrication Studies. This task focused on two major fuel fabrication routes: traditional pellet sintering and vibration packing (VIPAC). Using VIPAC fuel fabrication technology, Atomic Energy of Canada Limited (AECL) produced six annular fuel test specimens for irradiation testing at MIT. In addition, AECL successfully loaded two four-foot lengths of annular fuel using the VIPAC approach, achieving a maximum density of 77 percent of theoretical UO_2 density. This is insufficient to meet the fuel loading requirements, which require an effective uranium oxide density of at least 85 percent of the theoretical value. Higher uranium densities may be achieved by adding a uranium metal powder component to the oxide particle fuel mixture before compaction.

With respect to the traditional press and sinter fabrication route, Westinghouse Nuclear Fuel Company in Columbia, South Carolina, successfully demonstrated pellet fabrication of the 13x13 annular fuels. The pellets are shown in Figure 3. This showed that press-and-sinter fabrication appears to be a very promising technology for commercially manufacturing the annular fuel. A second set of annular pellet fabrication and rod loading trials, using commercial fabrication techniques, is currently underway and is intended to demonstrate improved dimensional control. The new trials will also include loading pellets in foot-long cladding and demonstrating end plug welding.

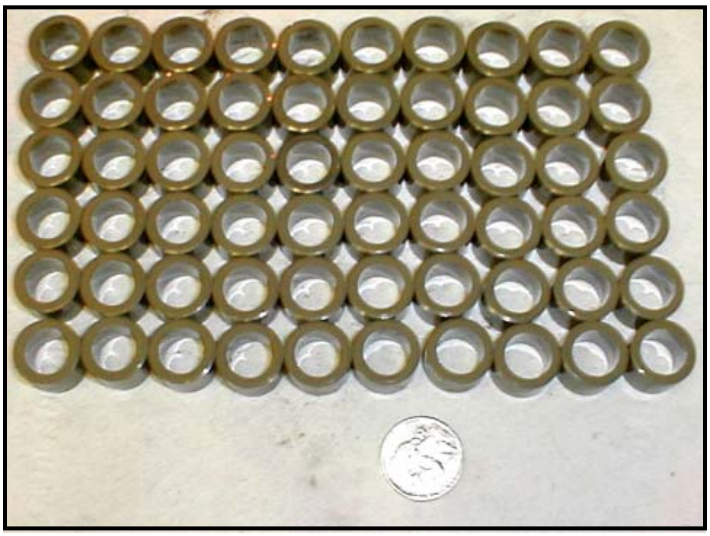


Figure 3. Sintered annular pellets manufactured at Westinghouse.

Annular fuel elements can be fabricated by commercial techniques at reasonable costs and should not be an inhibiting factor in the commercial introduction of annular fuel to improve the performance of existing light water reactors. Economic benefits of this fuel system will more than compensate for the initial small investment required to convert and modify existing fuel manufacturing equipment to handle annular fuel. The use of VIPAC fabrication techniques, which is not in commercial use today, is considered a possible backup to the use of the proven conventional pellet press and sinter techniques for fuel manufacture.

Task 4. Economic Analyses and Optimization. This task was finished in FY 2003 and the results were reported in the *NERI 2003 Annual Report*. The researchers identified manufacturing issues encountered when a central tube is introduced in the annulus of a fuel rod. They also evaluated the fuel cycle and capital cost benefits of annular fuel. The baseline constraints and assumptions used reflect the current permitting and operational constraints at the Westinghouse Nuclear Fuels Plant. Two types of fuel were considered: an annular pellet fuel similar to but bigger than the UO_2-based annular pellets currently made for wet annular burnable absorber (WABA) rods at Columbia and a

particulate fuel using vibration packing of crushed and screened irregular UO_2. For annular pellet fuels, no significant manufacturing issues were identified. For the VIPAC fuel, the major issues were low production rates during fuel rod loading; low final fuel density; maintaining uniform density during handling, shipping, and use; and increased dust loads during manufacture. No significant increases in manufacturing costs were expected for either fuel type.

To analyze the fuel cycle and capital costs, researchers compared three options using annular fuel to two options using standard solid pellet fuel. The discounted rates of returns for the various options were (listed best to worst):

1. 500 MW(e) uprate to an existing Generation II PWR; annular fuel (Option 2): 10.0 percent
2. 1,500 MW(e) Generation III new PWR; annular fuel (Option 3): 4.7 percent
3. 1,500 MW(e) Generation III new PWR; standard fuel (Option 4): 4.3 percent
4. 1,000 MW(e) Generation III new PWR; annular fuel (Option 5): 1.8 percent
5. 1,000 MW(e) Generation III new PWR; standard fuel (Option 1): 1.5 percent

The use of annular fuel always improves the rate of return on invested capital. In the case of a new power plant, the use of annular fuel can be used either to get more capacity from the same-sized nuclear island or to reduce the size of the nuclear island. For the currently operating Generation II power plants, annular fuel provides a means to significantly upgrade the power output (50 percent) using the same nuclear island with new reactor internals, steam generators and circulation pumps, and a new incremental balance of plant to handle the increased power production. Since the balance of plant is built while the power plant remains operational, the Generation II PWR is out of service for only the time it takes to replace the reactor internals, steam generators, and circulation pumps. Based on current steam generator replacement times of about 90 days, the downtime is estimated to be about six months.

Task 5. Fuel Performance Evaluation. Part of this task is to design and conduct the fuel irradiation experiment and the other is to develop a fuel performance model. MIT researchers completed manufacturing and testing of all the components for the in-core test capsules. Two of the VIPAC annular fuel samples manufactured by AECL were instrumented with thermocouples and assembled into irradiation capsules. Then researchers seal welded and pressure tested the capsules. After melting the lead bismuth eutectic heat transfer material in the clad-to-capsule gap and completing an X-ray inspection to ensure that the gap was completely filled in the fuel region, the capsules were approved for irradiation. Researchers also completed fit, handling, and flow tests in the reactor core tank. An initial startup irradiation test with one fueled capsule showed that the temperatures were within safety limits. The two assembled capsules were irradiated in the MITR-II research reactor to a burnup of 6.8 and 6 MWd/kg-U from March to September 2004. Linear heat generation rates in the fuel samples were near those that would be experienced in a peak rod at 150 percent of current PWR power density, with clad temperatures up to 240°C. Temperature asymmetries were observed between the inner and outer clad surfaces for both capsules, with considerable variation over the course of the run. These differences are attributed to the fact that the lead-bismuth heat transfer medium is in the solid phase near the aluminum capsule wall, with the thermal resistance of the solid/solid gap varying with the thermal history of the capsule. Fission product gas activity in the irradiation capsule cover gas was monitored throughout the irradiation. Buildup of activity was consistent with routine release through pre-existing micro-pores in the clad welds—no major clad defects developed during the irradiation. In early September, the irradiation was ended and the fuel samples were transferred from the core to the fuel storage ring in the reactor for decay before removal for post irradiation examination (PIE). In addition to continued preparation for PIE at MIT, plans for more extensive examination at Argonne National Laboratory West (ANL-W) were initiated.

For fuel performance modeling, the FRAPCON-3 code has been modified and applied to the pellet annular fuel. The results confirmed expectations of low fission gas release from the annular fuel due to low fuel temperature, which makes it possible to achieve high burnups. For high burnup at 150-percent power, fission gas release increased to values larger than those for solid fuel at 100-percent power and low burnups, but remained at an acceptable level of 6 percent. The cladding strain and oxide thickness were within the design limits, even for high burnup and at high power density. On the other hand, the model predicts an imbalance between the inner and outer gap conductances, which would negatively affect the minimum DNBR in the outer channels. A number of approaches have been explored to resolve the issue of gap conductance asymmetry; with the most promising identified to be sputtering on the outer fuel surface of ZrO_2 particles. This technique

reduces the outer gap conductance, due to the formation of an effective gap and a smaller thermal conductivity of ZrO_2, at the time of outer gap closure when the inner gap is still open. After the inner gap closure, the ZrO_2 at the outer surface is crushed due to large contact pressure, and the outer gap thermal conductance increases to closely match that of the inner gap. Researchers found the 50-percent coverage of the outer fuel surface with ZrO_2 particles, in combination with typical cladding and fuel roughness of 3 μm each, to yield excellent gap conductance symmetry throughout the entire irradiation period.

Planned Activities

This project continues under a no-cost extension to finish activities under Tasks 3 and 5. Respectively, these activities will focus on:

- Completing demonstration testing of the most promising fuel fabrication technology—the fabrication of long annular fuel rods by the press-and-sinter pellet fuel route.
- Conducting post-irradiation examination of the two irradiated fuel capsule samples including fission gas release, burnup confirmation, gamma scan, and analysis of results. The irradiated capsules will be transferred to a reactor floor hot box from the fuel storage ring in the reactor core tank. Gamma activity will be counted using a highly collimated Ge detector. Spectra from the gas plenum region will be used to estimate fission gas release; spectra from the fuel region will be used to calculate actual burnup numbers. This testing will be entirely non-destructive since fuel clad integrity is required to allow shipping of the fuel to ANL-W for additional PIE or to define the Idaho National Laboratory (INL) for disposal. If funding is available, the PIE at ANL-W will include confirmation of the burnup and fission gas release measurements. Neutron radiography will be used to investigate post-irradiation fuel morphology and thus possible fuel sintering/densification, void formation, and fuel-cladding gap formation.

NUCLEAR ENERGY RESEARCH INITIATIVE

An Innovative Transport Theory Method for Efficient Design, Analysis, and Monitoring of Generation IV Reactor Cores

PI: Farzad Rahnema, Georgia Institute of Technology

Collaborators: Idaho National Engineering and Environmental Laboratory, Penn State University

Project Number: 02-081

Project Start Date: September 2002

Project End Date: September 2005

Research Objectives

The objective of this research project is to develop a next-generation, high-order variational transport method for core-level neutronics calculations in advanced and Generation IV light water reactors (LWRs) and pebble bed reactors (PBRs). Researchers will implement the method into a computer code and apply realistic models of large-scale reactor cores in order to demonstrate the practical feasibility of the method. The research team expects that the new approach will achieve a significantly higher degree of accuracy than current state-of-the-art methods, especially for heterogeneous reactors and smaller Generation IV designs. The products of the project will be:

- A computer code system based on the new transport method for the following advanced reactor types and geometries:

Reactor Type	Geometry	Abbreviation
LWR	*1-D Slab*	*1- D(z)*
LWR	*2-D Cartesian*	*2- D(x, y)*
PBR	*2-D Cylindrical*	*2- D(r,θ)**

**Revised from the original goal of D(r,z).*

- A computer code system for automated calculations of PBR models in the asymptotic fuel loading pattern, developed by integrating the transport method into the Idaho National Engineering and Environmental Laboratory (INEEL) PBR fuel cycle code PEBBED.
- A theoretical method for 3-D Cartesian geometry advanced reactors and Generation IV LWR models.
- A suite of benchmark problems for the PBR and heterogeneous LWR cores with detailed reference transport results.

Research Progress

The main focus of the second-year work was to develop and complete the *2-D(x,y)* coarse mesh transport code, implement the *1-D(z)* code into PEBBED, and carry out some initial work on the *2-D*(r,θ) computer code.

Researchers coded a non-variational *2-D(x,y)* coarse-mesh iteration technique into REXTRAN2, which is essentially a straightforward extension of the *1-D(z)* method in REXTRAN. This iteration technique required much more computational time in large problems (e.g., a two-group, 40 direction, 10x10 coarse-mesh) than was initially expected. The original numerical method required the computation of the expansion coefficients, which are the dot product of incident fluxes and response function boundary conditions, as well as the reconstruction of the coarse-mesh boundary fluxes during each inner iteration. In *1-D(z)*, this procedure was extremely fast in real time, since the coarse-mesh boundary fluxes were only a function of energy group and polar direction. In *2-D(x,y)*, the fluxes have two additional degrees of freedom—namely, the spatial and azimuthal dimensions. Consequently, the number of computations grew geometrically.

Researchers reformulated the numerical, non-variational, inner iteration technique in terms of only the expansion coefficients and partial currents, thereby avoiding the explicit calculation of fluxes until the inner iterations have converged. No approximations were required to accomplish this reformulation. The computational speed-up is of the order of 10, which makes this coarse-mesh transport method competitive with conventional nodal diffusion methods in terms of computational cost. Researchers coded the reformulated non-variational iterations along with the variational technique.

The project team halted the implementation of the *1-D(z)* coarse mesh code into PEBBED. The coarse mesh code under consideration provides a one-dimensional solution of the transport equation in the z-direction. However, they found that in the pebble bed reactor, transport effects are essentially nonexistent in the z-direction in the core region. On the other hand, in the reflector region, transport effects can be significant near the control rods. Since the control rods cannot be modeled in the *1-D(z)* geometry, the coarse-mesh transport module was not implemented in the PEBBED code. The proper two-dimensional geometry for modeling the control rods is (r,θ), which resulted in changing the geometry for accomplishing objective 2.

The researchers proceeded with reformulating tasks involving the computer code for the PBR reactor. The preferred method for implementation consists of embedding within the diffusion theory solver an efficient transport theory solution technique for the zones that exhibit transport behavior. Researchers have determined theoretically that the response function equations can be transformed into a formulation that allows a derivation of Analytical Coarse Mesh Finite Difference (A-CMFD) equations used in PEBBED. In the project's third year, they will derive the actual coefficients in the resulting A-CMFD equations for the nodes of interest.

Researchers developed a preliminary set of two-dimensional (r,θ) benchmark cases using the TWODANT code (Figure 1). They also developed cross sections for the four energy group TWODANT simulations using the COMBINE spectral code. These calculations used cross sections generated at operating temperatures of approximately 900 K for the core and 600 K for the reflector.

The second-year accomplishments of this project are listed below:

- Researchers wrote and tested the final version of a computer code system for *1-D(z)* advanced and Generation IV LWR models using the new method.
- They developed the final version of a *2-D(x,y)* (fine-mesh) computer code to compute response functions.
- They developed a set of *1-D(z)* benchmark problems typical of LWR and PBR configurations. The reference transport results were also obtained.
- They obtained fine mesh reference results for the *2-D (x,y)* benchmark problems.
- They developed a final version of the *2-D (x,y)* computer code for the coarse-mesh solution of advanced and Generation-IV LWR models using the new methods.

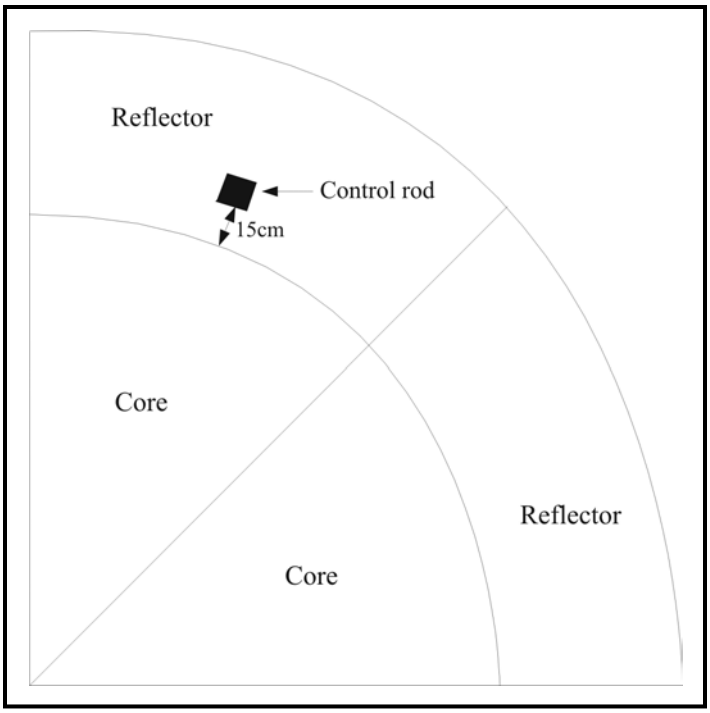


Figure 1. Two-dimensional PBR benchmark configuration.

- They wrote one journal paper:
  - Y.Y. Azmy, J.C. Gehin and R. Orsi: "DORT Solutions to the Two-Dimensional C5G7 MOX Benchmark Problem," *Progress in Nuclear Energy*, **45**, 2-4, pp. 215-232 (2004).

Planned Activities

The main focus of the third year is to complete the *2-D* code in (x,y), couple the transport and diffusion methods in (r,θ) geometry within PEBBED, and develop the *2-D* benchmark problems. The first part of the year will involve deriving the applicable equations while minimizing disruptions within the PEBBED code. Researchers will then incorporate the method (equations) into PEBBED in the second part of the year.

Additional third year activities include:

- Develop new *2-D(x,y)* BWR benchmarks.
- Test the *2-D(x,y)* coarse mesh transport code.
- Revise the new code, if needed, based on the test results above.
- Identify response functions reduction techniques in *2-D(r,θ)*.
- Develop the theoretical three-dimensional Cartesian geometry transport method.

NUCLEAR ENERGY RESEARCH INITIATIVE

Advanced Extraction Methods for Actinide/Lanthanide Separations

PI: Michael Scott, University of Florida

Collaborators: George Vandegrift, Argonne National Laboratory

Project Number: 02-098

Project Start Date: September 2002

Project End Date: September 2005

Research Objectives

Separation chemistry has always played a crucial role in preparing fuels for use in nuclear reactors. It is now assuming a central position in strategies for cleaning up decommissioned nuclear facilities and disposing of high-level radioactive waste. Although the United States does not currently allow spent fuel to be recycled, it is practiced around the world and separation chemistry has taken an active role in developing nuclear waste reprocessing technologies.

Nuclear fuel reprocessing is based on the dissolution of irradiated material in a nitric acid solution. The reprocessing operations produce high- and medium-level liquid wastes (HLLW and MLLW, respectively) containing different radioactive elements such as beta, gamma, and alpha emitters. In order to simplify the conditioning of such wastes, it would be highly desirable to separate the different radioactive components with respect to their lifetime. Separation would decrease the volume of high level waste intended for disposal in deep geological repositories, such as Yucca Mountain, and could instead be disposed in subsurface repositories, which are easier to manage. Once isolated, these nuclides could be turned into short-lived or non-radioactive elements, thereby eliminating the radiological hazards and waste disposal problems. The objective of this research project is to produce an advanced extraction process for separating the chemically similar actinides and lanthanides found in acidic nitrate nuclear waste streams.

The separation of Americium (Am) ions from chemically similar Lanthanum (Ln) ions is perhaps one of the most difficult problems encountered during the processing of nuclear waste. In the 3+ oxidation state, the Am (III) and Ln (III) metal ions have an identical charge and roughly the same ionic radius. They differ strictly in the relative energies of their f- and d-orbitals. In order to separate these metal ions, ligands will need to be developed that take advantage of this small but important distinction. At Argonne National Laboratory, researchers will test the binding attributes of various ligand systems developed at the University of Florida with Am (III), in addition to several other actinides including Pu (IV) and U (III). The information gained from these studies will be used to develop new and more sophisticated ligands. Further refinement should produce ligands adept at selectively sequestering actinide ions from acidic nuclear waste streams.

Research Progress

The extraction of uranium and plutonium from nitric acid solution can be performed quantitatively with the monofunctional organophosphorous compound tributyl phosphate (TBP). Commercially, this process has found wide use in the PUREX (*p*lutonium *ur*anium *ex*traction) reprocessing method. The TRUEX (*tr*ans*ur*anium *ex*traction) process is further used to coextract the trivalent lanthanide and actinide ions from the HLLW that is generated during the PUREX extraction. This method uses CMPO [(*N*, *N*-diisobutylcarbamoylmethyl) octylphenylphosphineoxide] **(1)** intermixed with TBP as a synergistic agent. However, researchers believe the final separation of trivalent actinides from trivalent lanthanides will be a challenging task.

In a TRUEX nitric acid solution, the Am (III) ion is coordinated by three CMPO molecules and three nitrate anions. Integrating this data with results of previous work on calix[4]arene systems, researchers have developed a C_3-symmetric tris-CMPO ligand system using a triphenoxymethane platform as a base, compound **2-5**. The triphenoxymethane ligand systems have many advantages for preparing complex ligand systems. The compounds are very easy to prepare and the steric and solubility properties can be tuned through an extreme range by

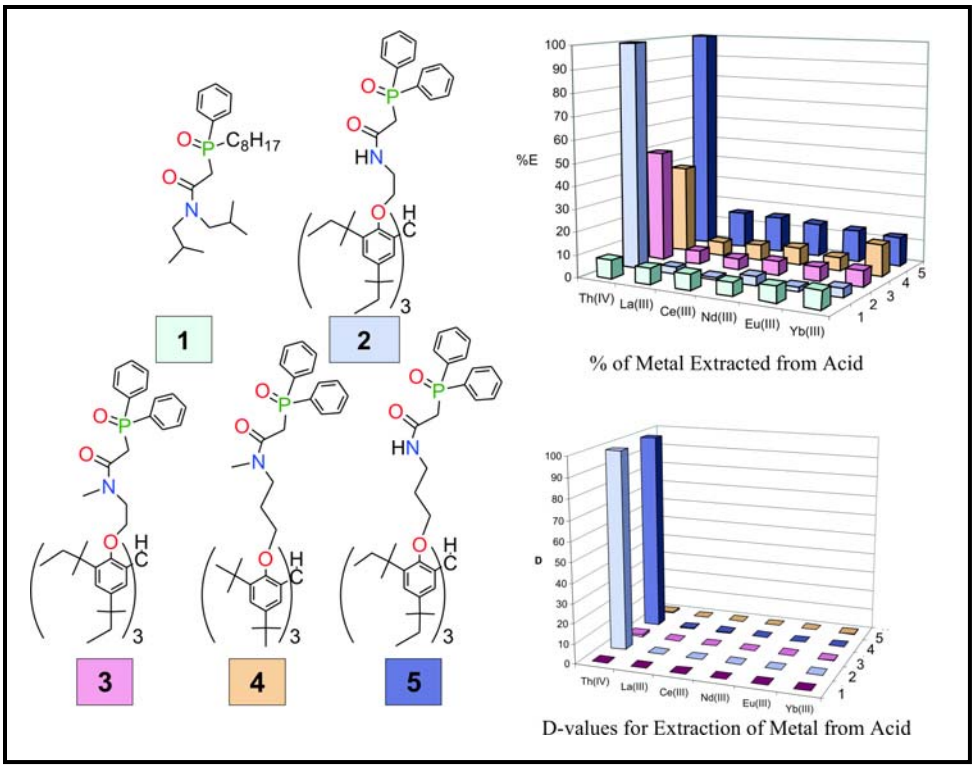
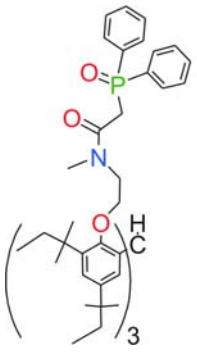


Figure 1. Extraction properties of the CMPO ligands developed at University of Florida.

including different alkoxy and alkyl groups such as methyoxy, ethoxy, *t*-butoxy, methyl, octyl, *t*-pentyl, or even *t*-pentyl at the *ortho*- and *para*-positions of the aryl rings. The new extractants also exhibit much higher selectivity for the actinide thorium over all of the lanthanides in comparison to other multi-CMPO ligands as well as CMPO itself. They show considerable promise for actinide recovery from high-level liquid waste and for general clean-up operations. The design and synthesis of that tris-CMPO ligand system has focused on combining high distribution coefficients for the selected metal ions, high ion selectivity, and great stability toward hydrolysis in acidic media.

In view of the interesting trends witnessed for these ligands, researchers sent several grams of each compound to Argonne for analysis with actinides. The ligands **2**, **3**, **4**, and **5** are all extremely selective for the Am (IV) ions such as plutonium in both dichloromethane and octanol, particularly in comparison to the simple CMPO ligand **1** that is currently used for commercial processes. Although the D value is low, ligand **4** does show an enhanced affinity (2.2 times) for Am (III) or Eu (III) as outlined in Table 1.

Inspired by the success of the DIAMEX industrial process, the researchers made an effort during year 2 to construct a ligand containing three arms, each with two amide donors and an oxygen donor. The ligand is perfectly suited to present nine donor groups to a metal center; hence, it should be particularly adept at binding large actinides. Researchers isolated large quantities of two different varieties of the ligand and examined their binding properties. They then compared them to a simple monomeric species, such as ligand **16**. Ligand **17** with three arms exhibited an enhanced selectivity for the smaller lanthanides, as illustrated in Figure 2. With the incorporation of three soft ether donors into the arms, the ligand should demonstrate a proclivity to bind actinides. To further facilitate the coordination of softer actinides, researchers replaced the central oxygen atoms in each of the three arms with sulfur donors. Both the oxygen and sulfur ligands have been sent to Argonne for further study.



Solvent - dichloromethane

Anion	D value	Extraction %
Pu(IV)	33.1	97.1
UO_2^{2+}	4.85	82.9
Am(III)	0.50	33.2
Eu(III)	0.23	18.8

2.2

Solvent - octanol

Anion	D value	Extraction %
Pu(IV)	33.1	85.9
UO_2^{2+}	4.85	60.6
Am(III)	0.06	5.6
Eu(III)	0.02	2.4

*Table 1. Actinide extraction properties of Ligand **4** in Dichloromethane (left) and Octanol (right) {$D = \Sigma [M_{org}]/ \Sigma [M_{aq}]$}.*

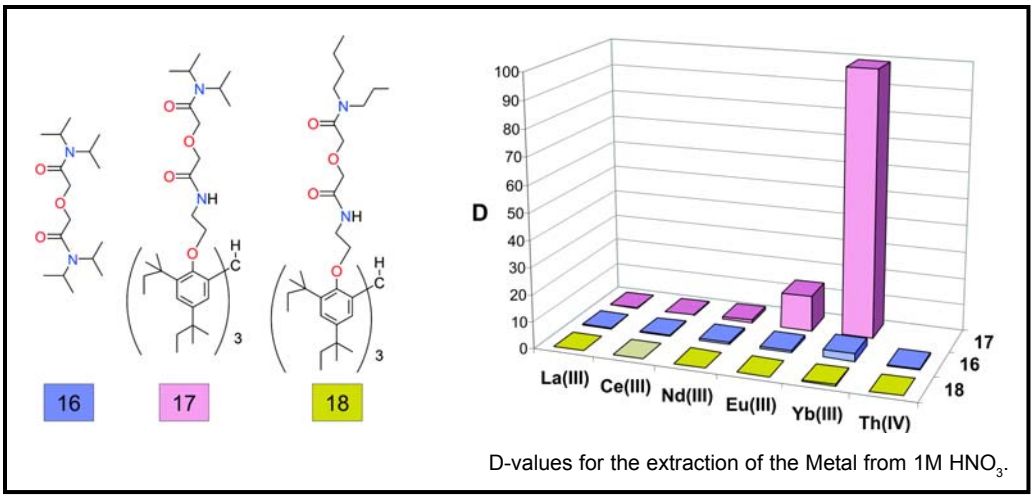


Figure 2. Extraction properties of the diamide ligands developed at University of Florida.

Planned Activities

The ligand systems developed at the University of Florida show promise as improved extractants for both tetravalent and trivalent actinide recoveries from acidic waste streams. The selectivity of the standard extractant for tetravalent and trivalent actinides, including CMPO and diamides, were markedly improved by the attachment of three arms containing these functionalities onto a triphenoxymethane platform. The results clearly demonstrate a cooperative action of these three arms within a single molecule, confirmed by composition and structure of the extracted complexes. The use of such an extractant permits the extraction of metal ions in highly acidic environments through the ability of the compound to buffer the effect of high acid concentration. Stability toward hydrolysis and ease of synthesis and purification are additional favorable properties. A variety of 3-fold symmetric ligands with functionalized arms are not only easily available in large quantities, but also amenable to nearly unlimited chemical modifications. Researchers at the University of Florida continue to pursue ligand refinement and testing while Argonne National Laboratory will continue to study the actinide binding properties. Based on the results from the experiments at Argonne, the Florida group will refine the ligands to enhance the binding properties. The continued cooperation between the two organizations should produce an advanced extraction process for separating the chemically similar actinides and lanthanides found in acidic nitrate nuclear waste streams.

NUCLEAR ENERGY RESEARCH INITIATIVE

Improving the Integrity of Coated Fuel Particles: Measurement of Constituent Properties of SiC and ZrC, Effects of Irradiation, and Modeling

PI: Lance L. Snead, Oak Ridge National Laboratory (ORNL)

Collaborators: Idaho National Engineering & Environmental Laboratory

Project Number: 02-131

Project Start Date: September 2001

Project End Date: September 2005

Research Objectives

Maintaining the silicon-carbide (SiC) layer integrity in tri-coated-isotropic (TRISO)-coated, gas-reactor fuel particles is critical to the performance, burn-up, and intrinsic efficiency of high-temperature, gas-cooled reactors. While there has been significant developmental work on manufacturing fuel particles, the materials properties under irradiation and their effect on the complex, in-service stress states are not adequately understood. Furthermore, although zirconium carbide (ZrC) has been proposed as a higher temperature replacement for SiC, there is virtually no experimental data on the effects of irradiation on thermomechanical properties.

This project will develop and apply techniques for measuring physical properties required to accurately model the TRISO fuel particle. In particular, researchers will measure information on the statistical distribution of strengths, elastic modulus, and creep of both SiC and ZrC, along with irradiation-induced changes in pyrolytic carbon interlayers. They will use this information in updated models for predicting the failure of the TRISO pressure vessels during processing and under irradiation, and will also generate baseline and post-irradiation properties data of ZrC for use as TRISO pressure vessels. Information generated under this program will be included with earlier published data on TRISO fuel and compiled into the *TRISO Fuels Coating Materials Handbook.*

Research Progress

In the first quarter of FY 2004, researchers successfully completed processing all the coated specimens required for unirradiated and irradiated property evaluation. These specimens included TRISO-coated, non-fuel surrogate particles as the coated materials most relevant to the fuel particles; TRISO-coated, non-fuel rods with graphite cores for internally pressurized hoop strength evaluation; and buffer-PyC/IPyC/SiC-coated thin rods with CVD-SiC fiber cores for characterizing interfacial strength property. Researchers evaluated the statistical strength properties of subsets of the produced samples and examined their microstructure. They concluded that the coatings produced in the processing task are qualitatively relevant to those for fueled TRISO particles and qualify as suitable materials for the irradiation task. The team will compile the results in the *TRISO Fuels Coating Materials Handbook*, to be delivered at the end of the project period.

In another task, the project team developed three fracture strength testing methods for miniature tubular and hemispherical shell specimens. The first two methods are based on the internal pressurization using elastomeric inserts for hemispherical and tubular shell specimens. The third method is a crush testing technique, or a diametrical loading technique, for tubular specimens. Using these methods, researchers obtained fracture strengths for the SiC specimens from surrogate fuel particles and rods, particularly the baseline fracture strength data for the tubular SiC specimens which will be subjected to neutron irradiation. The tubular specimens for this baseline test had nominal dimensions of about 1 mm inner diameter, 0.1 mm wall-thickness, and 5.8 mm length. In the statistics of the SiC fracture strength data, the mean value was 263 MPa with a standard deviation of 54 MPa; Weibull modulus and scale parameter were 5.0 and 286 MPa, respectively.

The research team developed a testing technique for determining interfacial shear strength of cylindrical layered coatings. This information allows calculation of the force transmitted between the pyrolytic carbon coatings and the SiC shell at a fuel particle. In this technique, inner layers are pushed out by a flat-bottomed indenter while outer

layers are rigidly held in a fixture. Researchers calculated the interfacial bonding and sliding stresses using the recorded load–displacement relationship. They successfully demonstrated strength characterization for the IPyC/SiC interfaces in TRISO-coated SiC-fiber samples. Also, they successfully developed an experimental method for determining the elastic modulus and hardness of very small specimen volumes by applying a nano-indentation technique. Researchers found this technique suitable and will apply it to the characterization of each TRISO layer in coated particles before and after neutron irradiation. Application of this technique will allow systematic measurement of the effect of irradiation on the elastic modulus of irradiated TRISO fuel SiC shells.

Researchers developed two novel neutron irradiation vehicles: one, extremely tiny and fragile coating samples, and the other, strained thin strip ceramic samples. The first design uses a powder-bed rabbit capsule that irradiates tiny samples in a bed of fine silicon carbide powder encapsulated in a silicon carbide-lined molybdenum case. The powder bed contains a number of irregular-shaped fragile samples, accommodates swelling of the samples without damaging them, and provides sufficient temperature homogeneity over the entire capsule interior. A second design is a rabbit capsule for bend stress relaxation (BSR) creep. In the BSR capsule, bend-strained thin (50 microns thick) strip samples of silicon carbide and zirconium carbide are tightly held during irradiation in a curved gap within a fixture made of silicon carbide. Irradiation creep strain rates are estimated based on the extent of stress relaxation following irradiation as a function of neutron fluence. The delay in the operation of the High Flux Isotope Reactor (HFIR) at Oak Ridge National Laboratory and material fabrication caused the significant delay in starting the irradiation program; fortunately, it also resulted in sufficient time to develop these novel irradiation experiments.

Researchers have completed the preparation of samples and parts, QA and QC procedures, and the construction and examination of capsules. The capsules are waiting for insertion into HFIR at Operation Cycle 403. Moreover, monolithic SiC and zone refined ZrC specimens began irradiation separately in the METS (Mapping Elevated Temperature Swelling) core capsules. The METS capsules are made up of three HFIR in-core vehicles which are irradiated in the approximately 2, 4 and 10 dpa ranges in a range of temperatures from 600–1,500°C. The two lower fluence capsules have finished irradiation.

Planned Activities

This program has been extended for another year (FY 2005) in order to finish the study on neutron irradiation effects, which had experienced a major delay due to the unexpected delay in operation of the high flux isotope reactor (HFIR). Therefore, researchers plan to irradiate surrogate fuel and creep capsules in HFIR and complete post-irradiation examination in FY 2005. Irradiation of the creep and the surrogate fuel capsules will be completed in April–May 2005. The final determination of irradiation creep compliance of SiC will be done in July 2005.

Researchers will use both the conventional crush test and the newly developed internal-pressurization method to evaluate the statistical strength of the irradiated SiC coatings. They will determine the post-irradiation shear strength of the IPyC/SiC interface by the push-in/push-out technique that has also been developed in this program. Other post-irradiation properties, such as the baseline properties of the TRISO constituents and zone-refined ZrC, will be determined in parallel. These post-irradiation testing activities are scheduled to finish in August 2005. The generated post-irradiation property data will be compiled into the *TRISO Fuels Coating Materials Handbook*, which will be made available at the end of the project period.

NUCLEAR ENERGY RESEARCH INITIATIVE

Enhanced Thermal Conductivity Oxide Fuels

PI: Alvin Solomon, Purdue University

Collaborators: Shripad Revankar, Purdue University; Kevin McCoy, AREVA, Framatome-ANP; Argonne National Laboratory-West (ANL-W)

Project Number: 02-180

Project Start Date: October 2002

Project End Date: September 2005

Research Objectives

The objective of this project is to develop the methodology to produce enhanced conductivity oxide (ECO) fuels by incorporating a high conductivity solid phase that is stable, compatible with UO_2, does not impose a strong neutronic penalty, and is commercially realizable. Improved thermal conductivity has many advantages: it would lower stored energy in the fuel, reduce radiation-induced degradation like fuel cracking, swelling, and gas release, and allow possible fuel uprating. In addition, properly chosen compounds can yield a more stable waste form.

The goal of the originally proposed research was to develop a process to impregnate sintered oxide fuels of near commercial density with large-scale open porosity and a continuous high conductivity phase. This high conductivity phase would be stable at high temperatures, resistant to neutron and fission fragment damage, and compatible with oxide fuels. It would also exhibit minimum neutronic effects and provide the maximum improvement in thermal conductivity. Commercially available organic precursors of silicon carbide (SiC) were selected for this approach, called Polymer Impregnation and Pyrolysis, or PIP.

Although SiC has many of the desired features mentioned previously, it is known to react with UO_2 at temperatures exceeding 1,370°C, forming gaseous and solid species. If the gases reach their low equilibrium pressures, the reaction stops. This occurs up to 1,900°C. Also, the impregnation process requires several cycles to fill the porosity. Therefore, researchers will investigate other approaches and high conductivity materials, namely beryllium oxide (BeO) and carbon, as dispersions in UO_2.

Researchers are examining various forms of ECO fuel both theoretically and experimentally in terms of compatibility with UO_2, and will model the forms to measure their thermal properties. Researchers will also produce pellets using enriched material for subsequent irradiation.

Research Progress

Researchers achieved the first processing task: producing the required UO_2 fuel of high density and open porosity through a new method, the "slug/bisque process," in which the powder was pre-slugged at high pressure, granulated, and sieved to a desired size range. The granules were then "smoothed" by a "self-milling" process and presintered at low temperatures to retain their spherical shape after a lower pressure pelletization. Researchers carried out sintering at typical temperatures to achieve densities of 85 to 95 percent. Polymer impregnation was done under vacuum, followed by pressurization at several atmospheres and pyrolysis at 1,300°C to avoid reaction with UO_2. Researchers determined that each impregnation cycle filled approximately 30 percent of the remaining open porosity with SiC. However, porosity began to close after four impregnations, so that even after eight impregnations only 70 to 80 percent of the original open porosity volume fraction was filled with SiC, starting with UO_2 originally at 90 percent theoretical density (TD). An example of the SiC phase in UO_2 is shown in Figure 1. Finite element modeling studies showed that this still produced a 30 to 50 percent increase in thermal conductivity.

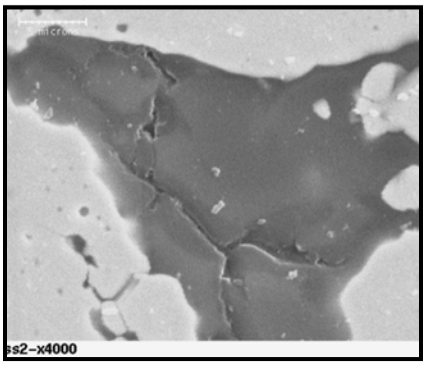


Figure 1. SiC phase (dark) within the open porosity of UO_2 produced by Polymer Impregnation and Pyrolysis.

Researchers performed compatibility experiments with high purity β-SiC and UO_2 pellets and confirmed the onset of detectable reactions at 1,400°C, which provided the temperature limit for crystallization of the SiC phase and led to a very fine grain structure.

Although the PIP processing was successful, it required several cycles. To reduce the processing time, researchers used SiC "filler particles" between the original UO_2 granules. This enabled them to achieve the same density of SiC with fewer impregnations.

The finite element thermal modeling showed that if dispersions of fully dense high conductivity phases could be incorporated in UO_2, similar improvement in thermal conductivity could be obtained as with the PIP process, with perhaps only one sintering cycle. Towards that end, researchers tried SiC whiskers dispersed in UO_2 powder, chopped graphite fibers, and large SiC particles. With the SiC whiskers, they were unsuccessful in achieving the desired fuel density due to the well-known phenomenon of constrained sintering with whisker networks. Moreover, a supply of inexpensive high purity SiC whiskers is not known. On the other hand, very inexpensive high conductivity graphite fibers are readily available, but when researchers tried to sinter them with UO_2 at temperatures below 1,400°C (in order to avoid carbothermic reduction of UO_2) they again were not able to produce the desired fuel density. Only the SiC particles distributed in UO_2 granules led to acceptable densities.

An important variant of such dispersions was the use of beryllium oxide (BeO) as the dispersed particles. BeO is the highest conductivity oxide and is compatible with UO_2 up to approximately 2,100°C, where it forms a eutectic. Unlike SiC, BeO sinters readily at 1,600°C at a rate comparable to that of UO_2. Researchers call this process "co-sintering." Under appropriate conditions, BeO and UO_2 powders exhibit compatible shrinkage, leading to densities as high as 95 percent TD. Pellets of pure BeO and of co-sintered UO_2/BeO are shown in Figure 2 (left). Researchers demonstrated that the high conductivity BeO phase forms a minor continuous phase within a continuous UO_2 phase. This was confirmed by dissolving the UO_2, leaving only the BeO phase free standing as seen in Figure 2 (right). The skeleton is surprisingly robust and can be easily handled with tweezers. The microstructures of these specimens are shown in Figure 3.

The success and ease of the UO_2/BeO co-sintering process has led to a more detailed examination of the toxicity issue in a commercial fuel processing facility. Pellet grinding is the most questionable manufacturing process, but the existing controls for UO_2, as specified in 10CFR850, may also be adequate for BeO.

Researchers initiated thermo-mechanical and mechanical tests on the PIP-derived SiC specimens. The geometry and thermal conductivity of these specimens are such that the Biot Modulus is less than 20, indicating that thermal conductivity influences the thermal stresses generated by a quench test. UO_2 disks at 85°C readily crack when quenched in ice water, whereas ECO specimens did not crack at temperatures up to 240°C when quenched in ice water. They are presently performing finite element thermal modeling to determine the stresses as a function of quench time.

Figure 2. Photograph of co-sintered UO_2 - BeO (black) & BeO (white) pellets (left). BeO 'skeleton' obtained by chemical dissolution of UO_2 from a co-sintered pellet (right). The skeleton was about 85 percent dense and absence of UO_2 was confirmed via EDS & XRD analysis.

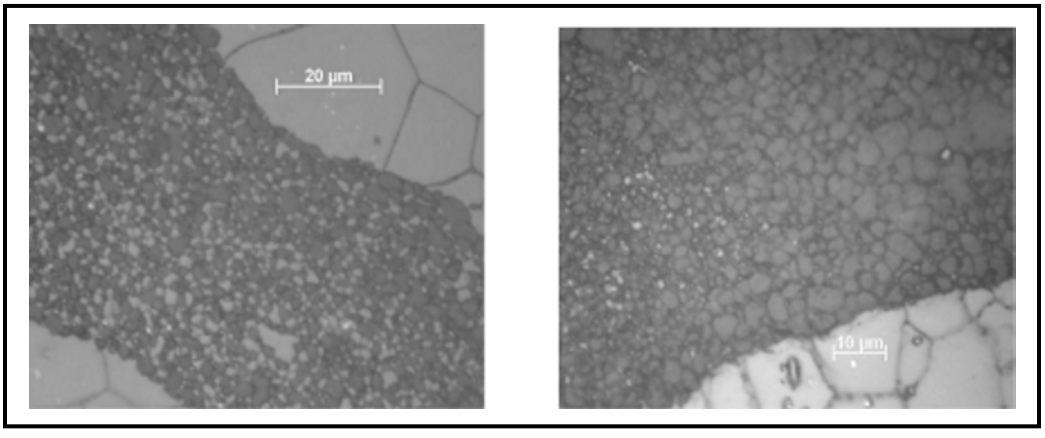


Figure 3. Polished, thermally etched section of a co-sintered UO_2 – BeO pellet fabricated from green granules (left), and from bisque granules (right). Light regions correspond to the UO_2 phase, while the grey regions correspond to the BeO phase. Reduction in the amount of dispersed UO_2 phase within the BeO phase from the green to bisque granules is apparent.

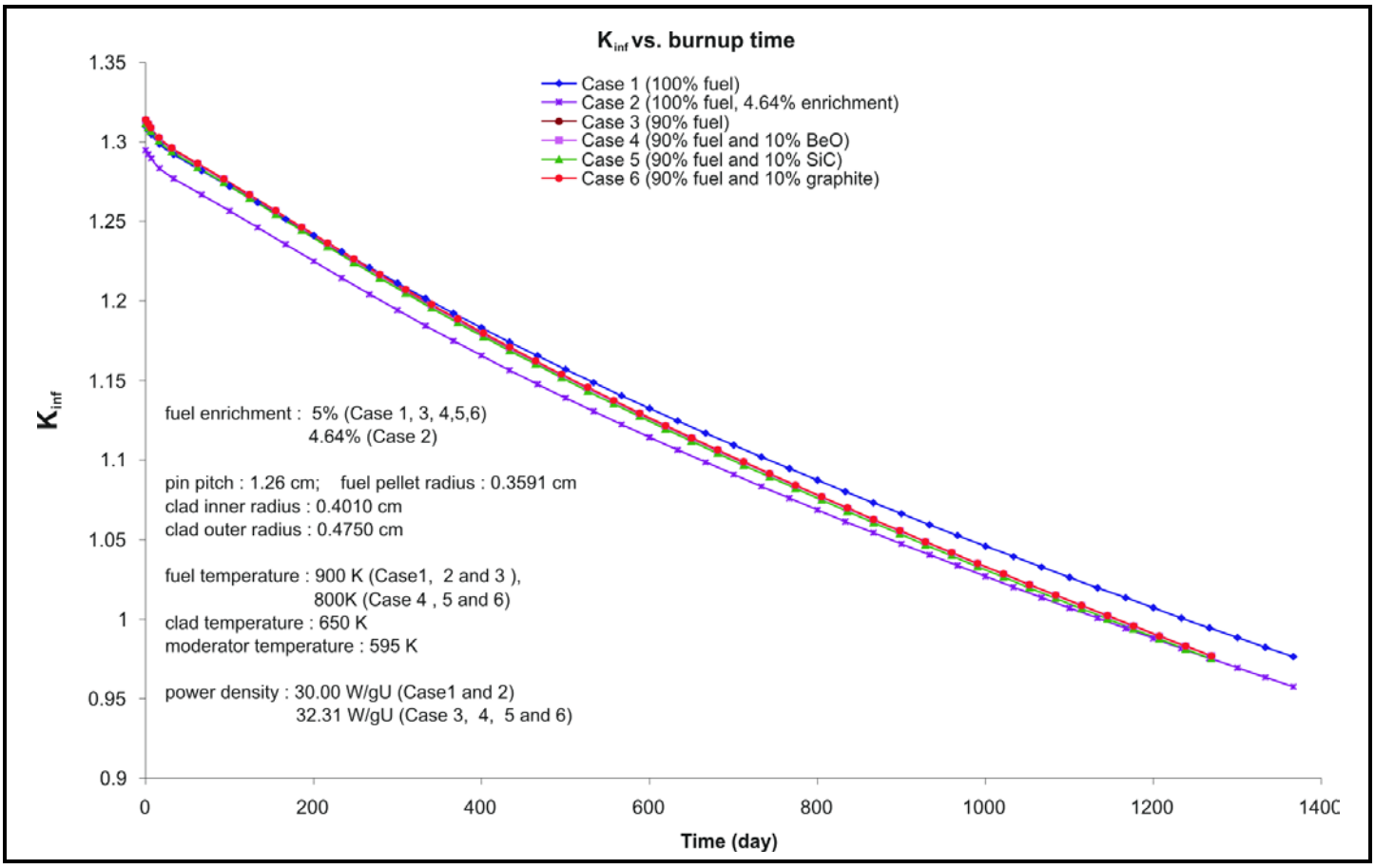


Figure 4. Differences in reactivity for various ECO fuels vs. UO_2 fuels of two enrichments.

Researchers performed neutronic calculations comparing the various high conductivity phases (SiC, BeO, and C) in the same amounts, assuming a 100°C reduction in fuel temperature. The reactivity swing for the three materials in UO_2 of the same enrichment is identical and only slightly different from typical 95 percent TD UO_2 fuel, as shown in Figure 4.

Planned Activities

Thermal property measurements of SiC-containing specimens are currently underway at the Idaho National Laboratory (formerly ANL-W). So far, they have not renewed their procedures for BeO handling and have not accepted shipment of these specimens. Although they do not anticipate problems in eventually accepting the material, researchers are still looking at alternative sites. The research team is also building their own thermal conductivity apparatus which will allow measurements at low temperatures under steady state conditions.

Once they have obtained actual conductivity measurements, researchers plan to address certain performance issues with these fuels, including: the effect on fuel temperatures, fission gas release, LOCA initialization, economics, and fission product degradation of the high conductivity phase. They also plan to perform thermo-mechanical quench tests on the various ECO fuel forms, as well as diametral compression tests to determine their fracture strength.

Currently, the researchers are searching for a source of reactor-grade, enriched UO_2. They would prefer to dilute higher assay material so that the sintering characteristics are well known, but this will require a modification of the Purdue license. Alternatively, they might use some archival enriched material stored at Purdue in the form of helical spring specimens, some of which may be green. They have discussed possible irradiations with Idaho National Laboratory.

NUCLEAR ENERGY RESEARCH INITIATIVE

Use of Solid Hydride Fuel for Improved Long-Life LWR Core Designs

PI: Ehud Greenspan, University of California, Berkeley (UCB)

Collaborators: Massachusetts Institute of Technology, Westinghouse Electric Company, University of Tokyo

Project Number: 02-189

Project Start Date: September 2002

Project End Date: September 2005

Research Objectives

The general objective of this project is to assess the feasibility of improving the performance of light water reactors (LWRs) by using solid hydrides as one of the core constituents. Following is a list of potential improvements:

- Increasing the core life, discharge burnup, total energy generated per fuel load, and the capacity factor of LWRs. Expected outcomes are reduced fuel cycle cost and the cost of electricity (COE), increased fuel utilization, reduced amount and toxicity of high-level waste, and improved proliferation resistance.
- Reducing the volume of the core and pressure vessel for a given power LWR, or increasing the power level corresponding to a given core volume, thus reducing the COE.
- Increasing the capability of LWRs to recycle commercial plutonium and to dispose of military plutonium.
- Increasing the capability of LWRs to utilize thorium.
- Improving the safety of LWRs, due to the inherent negative temperature reactivity effect of hydride fuel, and the safety of boiling water reactors (BWR) by reducing the negative void reactivity coefficient.
- Reducing the heterogeneity of BWR cores and their negative void coefficient to simplify fuel assembly design and the reactor control system, thereby improving BWR economics.

Research Progress

The main thrust of the second year of this project was completing the development of a methodology for identifying an optimal oxide or a hydride fueled pressurized water reactor (PWR) core design that gives the lowest COE, followed by performance of a comprehensive parametric study. Researchers revised the database used for the analysis, primarily affecting the fuel rod gap and clad thickness dependence on the fuel rod diameter and, in the case of hydride fuel, also on the fuel discharge burnup. They significantly upgraded the neutronic analysis methodology to enable estimating the discharge burnup as well as the burnup-dependent soluble boron concentration and reactivity coefficients based on SAS2H unit cell calculations. The methodology accounts for the following: the non-linearity in k_{∞} versus burnup, the average burnup-dependent fuel composition in multi-batch PWR cores, and the burnup-dependent hydride fuel rod gap thickness. Researchers then performed a neutronic analysis for oxide and for several types of hydride fuels, each for three enrichment levels.

The thermal-hydraulic analysis methodology was upgraded primarily by replacing the coolant velocity constraint of 8 m/s with a detailed vibration analysis. The following vibration and wear mechanisms were considered: vortex-induced vibration, fluid-elastic instability, turbulence-induced vibration in cross and axial flow, fretting wear, and sliding (or adhesive) wear. They then repeated the thermal-hydraulic parametric study and completed the fuel rod mechanical integrity analysis for oxide and for U-$ZrH_{1.6}$ fuels.

A methodology was developed and applied to assess the maximum attainable power that assures reactor safety in case of a number of accidents, including a large break loss of coolant accident, a complete loss of flow accident, and an overpower transient due to a control rod bank withdrawal at full power.

Researchers completed the economic analysis model and used it to search for that combination of design variables that offers the minimum cost of electricity. They

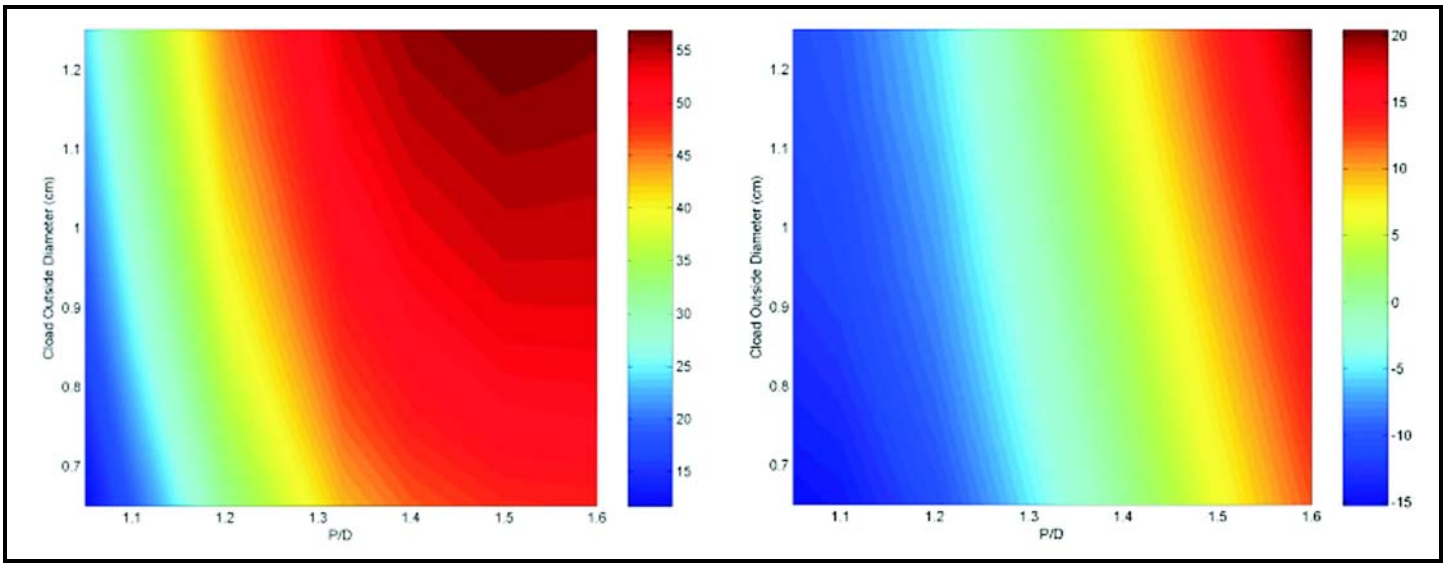


Figure 1. Attainable burnup (left) and coolant temperature coefficient of reactivity (right) for U-ThH_2-$ZrH_{1.6}$ fuel having 55 volume percent ThH_2 and using 12.5 percent enriched uranium.

used a wide range of core design variables for this study: (1) fuel rod outer diameter, D— in the range from 0.65 to 1.5 cm; (2) lattice pitch-to-diameter ratio, P/D – 1.05 to 1.6; (3) fuel type – UO_2, U-$ZrH_{1.6}$, U-ThH_2-$ZrH_{1.6}$, U-ErH_3-$ZrH_{1.6}$, as well as Pu-bearing hydride fuels; (4) uranium enrichment – 5, 7.5, or 10 percent for UO_2 and from 5 to 20 percent for hydride fuel; (5) coolant pressure drop across the core–either 29 psia or 60 psia; and (6) type of fuel rod support–grid spacers or wire-wraps. The design constraints considered include minimum excess reactivity, negative Doppler coefficient, negative moderator temperature coefficient, negative void coefficient, MDNBR, peak and average fuel temperature, coolant inlet and outlet temperatures, coolant pressure drop, fission-gas pressure, clad strain, clad water-side corrosion, and constraints imposed by vibration and wear corresponding to the above mentioned mechanisms.

Figure 1 illustrates the final results of the neutronic analysis. It shows the attainable burnup and the coolant temperature coefficient of reactivity for a wide range of lattice geometries. These results correspond to U-ThH_2-$ZrH_{1.6}$ fuel having 55 volume percent ThH_2 and using 12.5–percent enriched uranium. Only lattice geometries (defined by their D and P/D) having negative moderator temperature coefficient of reactivity are acceptable. The results account for the effect of soluble boron.

Figure 2 illustrates the final results of the thermal-hydraulic analysis combined with the vibration and wear analysis. It shows the power that is attainable from UO_2

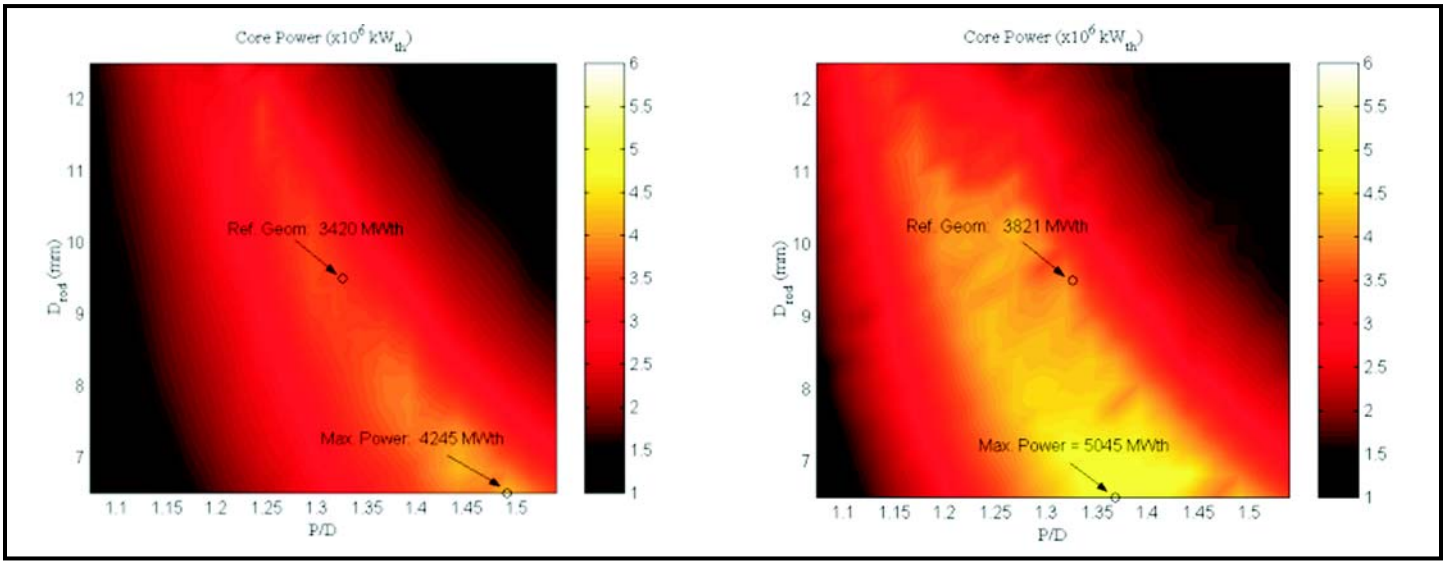


Figure 2. Attainable power from cores having different combinations of D and P/D in a square array of UO_2 fuel with a coolant pressure drop of 29 psia (left) or 60 psia (right).

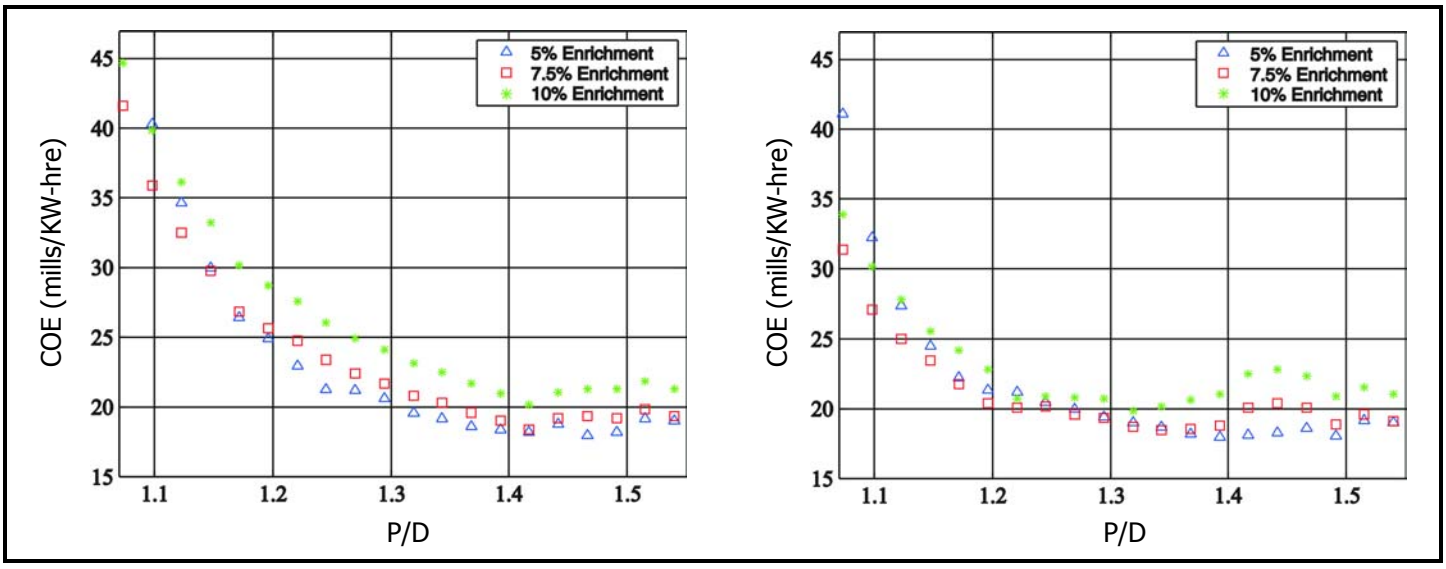


Figure 3. Minimum cost of electricity (COE) versus P/D for a major backfit scenario using UO_2 fuel and coolant pressure drop of either 29 psia (left) or 60 psia (right).

fueled cores having different combinations of D and P/D for either a coolant pressure drop of 29 psia (left) or 60 psia (right).

Figure 3 shows the results of the parametric economic analysis to determine the minimum cost of UO_2 fuel for each P/D ratio. For a 29 psia coolant pressure drop, the lowest minimum COE of 18 mills/KWH was obtained for 5–percent enriched uranium at P/D = 1.47 and D = 7.13 mm (not shown in Figure 3). This geometry is significantly different from that adopted by the industry for the reference design (D = 0.95 cm; P/D = 1.326), for which the calculated cost was approximately 20 mills/KWH. The corresponding core power is 8 percent higher than that of the reference design. For 60 psia, the lowest minimum COE was 17.9 mills/KWH, obtained for 5–percent enriched fuel at the peak power geometry of P/D = 1.39 and D = 6.5 mm.

Researchers also found that using a hexagonal array of D = 0.65 cm oxide fuel rods having P/D = 1.42 with wire wraps, while maintaining a coolant pressure drop of 29 psia, it was possible to increase the power of a given volume core by at least 30 percent while also reducing the COE. This was due to the decreased pressure loss for wires versus grids. The beneficial effect of wire wrap in raising the critical heat flux has not yet been integrated into the analysis.

An additional project thrust during the second year was developing the methodology for BWR core analysis. Researchers established a database for BWR cores, successfully developed a 3-D fuel assembly model for the MOCUP code, and successfully modeled the BWR core with the VIPRE thermal-hydraulic analysis code.

Planned Activities

In the third year of the project, researchers will complete the comparison of PWR economics using hydride fuel versus oxide fuel. A number of hydride fuels will be considered, including U-ThH_2-$ZrH_{1.6}$, U-ErH_3-$ZrH_{1.6}$, PuH_2-U-$ZrH_{1.6}$, PuH_2-ThH_2, and PuH_2-ThH_2-$ZrH_{1.6}$. They will also expand the economic analysis model to account for thorium- and plutonium-containing fuels, and will assess the environmental and repository implications of using hydride fuel. The FRAPCON code used for the mechanical integrity analysis will be replaced by the FALCON code, which researchers found to be more reliable for the purposes of this study. Researchers also plan to continue studying the feasibility of designing PWR cores using a hexagonal array of fuel rods that are supported by wire wraps instead of grid plates. Preliminary analysis indicated that this design approach is very promising.

The major thrust in the third year is to repeat the comparison between hydride and oxide fuel for BWR cores following a similar general approach to that developed for the analysis of PWR cores, but using different computational tools and models.

The final task planned includes an overall assessment of the feasibility of hydride fuel to improve the performance of either PWRs and/or BWRs, recommendations for future R&D, and preparation of a summary report for this project.

NUCLEAR ENERGY RESEARCH INITIATIVE

Development of Advanced Methods for Pebble-Bed Reactor Neutronics: Design, Analysis, and Fuel Cycle Optimization

PI: Abderrafi M. Ougouag, Idaho National Engineering and Environmental Laboratory

Collaborators: Georgia Institute of Technology, Pennsylvania State University, PBMR (Pty.) Ltd., and University of Arizona

Project Number: 02-195

Project Start Date: October 2002

Project End Date: September 2005

Research Objectives

This project is a comprehensive effort to develop analysis methods for pebble-bed reactors (PBRs). These methods are based on modern analytical nodal methods for solving the neutron diffusion equation. They also include efficient techniques for node homogenization. The nodal methods and node homogenization techniques will be incorporated into (or tightly interfaced with) the Idaho National Engineering and Environmental Laboratory's (INEEL) PEBBED code for pebble bed reactor neutronics and fuel cycle analysis. The most novel aspect of this effort is that researchers are developing these analytical nodal methods for three-dimensional cylindrical geometry, a development that had not previously been accomplished because of mathematical obstacles encountered in the traditional approach. In prior work, the investigators found a way around these obstacles. The initial objective of this project was to implement that advance.

Additional enhancements to PEBBED include techniques to account for the dependency of reaction cross sections on thermohydraulic feedback and a genetic algorithm to facilitate design optimization.

Research Progress

In earlier work, researchers at INEEL developed the PEBBED code. As originally written, the code applied traditional finite-difference techniques to solve the neutron diffusion equation, but it was organized in a modular form. This modular form allows researchers to incorporate upgrades in any physics area as improvements become available. It was always intended that the finite-difference diffusion solver would be replaced or augmented by the new nodal method developed at INEEL.

Early in the project, the investigators decided to apply the nodal solution through the Coarse-Mesh Finite-Difference (CMFD) approach, which takes advantage of the existing finite-difference framework in PEBBED. Initially, an iterative scheme was applied, following methods established for Cartesian geometry. However, difficulties in achieving convergence with this Cartesian geometry-based iterative method led to the selection of a much more efficient direct CMFD technique. This method has been successfully installed in PEBBED in the radial and axial directions.

In the azimuthal direction, the original solution involved the computation of a series of hyperbolic Bessel functions. Evaluation of this series consumed so much computation time that it offset the advantages of a nodal solution. However, investigators discovered a much more efficient approach to the nodal solution for the azimuthal variable and will implement it in the PEBBED nodal formulation.

In order to optimize a PBR, researchers must compare various core dimensions, pebble types, and pebble recirculation patterns. The PBR fuel management problem is different from that in light water reactors (LWRs) because the fuel moves, individual fuel elements are small, and fueling is essentially continuous. Little research has been performed previously in this area.

Genetic Algorithms (GAs) are a stochastic method recently applied to LWR fuel cycle optimization. As the name implies, these algorithms mimic the process of biological evolution, in which adaptive traits are favored in future generations. Under this NERI project, researchers have developed such an optimization feature and incorporated it into PEBBED to perform design studies. They tested this feature by applying it to studies of the PBR version of the next generation nuclear plant (NGNP), a

very-high-temperature reactor proposed for siting at the INEEL. Application of the algorithm facilitated the design of passively safe PBR cores of much higher power output than previously thought possible (i.e., up to 700 megawatts of thermal power).

The approach, which was initially conceived for cross section homogenization, was a straightforward application of perturbation techniques developed by the Georgia Institute of Technology. However, as research progressed, researchers found discrepancies between results produced by the continuous-energy Monte Carlo code, MCNP, and the multigroup techniques needed for nodal methods. These discrepancies led to the discovery that fundamental deficiencies pervade all cross section processing codes in their treatment of low-lying resonances important in graphite-moderated reactors at high burnup, although not in LWRs at low burnup. Also, research under this project and elsewhere revealed the importance of the double heterogeneity of PBR fuel (discrete fuel particles and discrete pebbles), and the inadequacy of available methods to account for it. Specifically, the Dancoff factor was never computed correctly in doubly heterogeneous systems. This factor quantifies the effect of neighboring fuel regions on the escape probability for a neutron from a given fuel region. The investigators and colleagues embarked on projects to improve the treatment of the low-lying resonances and to calculate the Dancoff factor rigorously in random dual heterogeneous media.

When generating PBR cross-section libraries, one must correctly model the spectrum dependence, which depends on temperature as well as on the material compositions of the surrounding pebbles. Therefore, thermohydraulic feedback effects must be taken into account. Pennsylvania State University has provided a suitable method to the INEEL in two software packages. Furthermore, in collaboration with Purdue University, the THERMIX thermohydraulics package has been coupled to PEBBED for accurate calculation of temperature distributions.

Planned Activities

In the final year of the project, three principal tasks will be pursued. The first is to complete the coding, verification, and validation of the cylindrical geometry nodal solver in PEBBED. The second is to develop a cylindrical geometry nodal depletion code that is compatible with the solver within the PEBBED code and to incorporate that new coding. The third is to apply the enhanced PEBBED code to two optimization applications: fuel cycle planning and nonproliferation. These tasks will be pursued in collaboration with Georgia Institute of Technology and Pennsylvania State University.

8. Fundamental Nuclear Sciences: Project Summaries

This research area includes 11 NERI projects, of which 5 were awarded in FY 2001 and 6 in FY 2002. It addresses the long-term R&D goals of developing new technologies for nuclear energy applications, of educating young scientists and engineers and training a technical workforce, and of contributing to the broader scientific and technological enterprise.

Today's U.S. reactors, which are based largely on technology from the 1970s, operate under close supervision in a conservative regulatory environment. Although the knowledge base is adequate for these purposes, improvements in the Nation's knowledge base and reduction of the inherent uncertainties concerning nuclear reactors could bring cost savings to current reactor operations and reduce the costs of future reactors. They could also enable innovative designs that reduce the need for excessively conservative and costly safety and reliability factors, and significantly extend safe operating lifetimes. Future reactor technologies are likely to involve higher operating temperatures, advanced fuels, higher fuel burn-up, longer plant lifetimes, better materials for cladding and containment vessels, and alternative coolants. To implement such features, substantial research must be carried out in fundamental science and engineering to supplement applied research on promising design concepts. Such fundamental research need not and should not be directed to any specific design. Although motivated in part by the need for new nuclear reactor system designs, the research would also have a far-reaching impact elsewhere in engineering and technology.

The five broad topics identified in the Nuclear Energy Research Advisory Committee (NERAC) *Long-Term Nuclear Technology R&D Plan* related to fundamental nuclear sciences include the following:

- Environmental effects on materials, in particular the effects of the radiation, chemical, and thermal environments, and aging.
- Thermal fluids, including multiphase fluid dynamics and fluid structure interactions.
- The mechanical behavior of materials, including fracture mechanics, creep, and fatigue.
- Advanced material processes and diagnostics.
- Reactor physics.

Projects currently being pursued under this element include R&D in fundamental science in the fields of material science, chemical science, computational science, nuclear physics, or other applicable basic research fields. Selected research subjects include irradiation, chemistry, and corrosion effects on nuclear plant materials; advanced new materials research; innovative computational models; and the investigation of nuclear isomers that could prove beneficial in civilian applications.

NUCLEAR ENERGY RESEARCH INITIATIVE

Directory of Fundamental Nuclear Sciences Project Summaries

NUCLEAR ENERGY RESEARCH INITIATIVE

Random Grain Boundary Network Connectivity as a Predictive Tool for Intergranular Stress-Corrosion Cracking

PI: Mukul Kumar, Lawrence Livermore National Laboratory

Collaborators: University of Michigan, GE Global Research Center

Project Number: 01-084

Project Start Date: October 2001

Project End Date: Completed September 2004

Research Objectives

Intergranular stress corrosion cracking (IGSCC) is one of the most pervasive degradation modes in current light water reactor systems (LWR) and is likely to be a limiting factor in advanced systems as well. In structural materials, IGSCC arising from the combined action of a tensile stress, a "susceptible" material, and an "aggressive" environment has been recognized for many years and the mechanisms widely investigated. Recent work has demonstrated that by sequential thermomechanical processing, properties such as corrosion, IGSCC, and creep of materials can be dramatically improved. The improvements have been correlated with the fraction of so-called "special" grain boundaries in the microstructure.

A multi-institutional team comprised of researchers from Lawrence Livermore National Laboratory (LLNL), University of Michigan (UM), and General Electric Corporate Research & Development (GECRD) proposed an alternative explanation for these observations: *that the effect of grain boundary engineering is to break the connectivity of the random grain boundary network through the introduction of low energy, degradation-resistant twins, and twin variants.* The team carried out a collaborative science and technology research project aimed at verifying the mechanism by which sequential thermomechanical processing ameliorates IGSCC of alloys relevant to nuclear reactor applications and to prescribe processing parameters that can be used in the manufacture of IGSCC-resistant structures. Specific objectives were:

- To develop a quantitative method for characterizing the connectivity of random grain boundary networks.
- To prove a direct correlation between the random grain boundary network connectivity and susceptibility to IGSCC.
- To develop a tool to predict the relative IGSCC performance of off-the-shelf materials with well-characterized IGSCC performance.

To accomplish this work, the team developed algorithms to quantify the interconnectivity characteristics of the random grain boundary network and measured the interconnectivity of a series of materials where the interconnectivity was systematically altered. Researchers then performed property measurements on the altered materials and compared their performance ranking with the boundary network measurements in order to correlate actual crack paths with the measurements of the random grain boundary network.

With this data, the team evaluated and improved the methods for describing the random grain boundary network. They tested the characterization method by evaluating the interconnectivity of the random grain boundary network in a series of as-received materials, ranking their expected performance, and comparing that result with property measurements.

Research Results

The major accomplishments of this project were determining that random boundary network connectivity is a major driver of IGSCC in austenitic alloys, developing a predictive tool for ranking IGSCC performance of these alloys, and establishing thermo-mechanical processing parameters to be applied in the manufacture of IGSCC-resistant materials.

Task 1. Development of Boundary Tracking Algorithms. The research team developed tools for characterizing and quantifying random grain boundary networks. Microstructural changes were quantified by using election backscatter diffraction (EBSD) orientation

mapping and then applying the algorithms to the datasets. In the experimental study of connectivity, researchers first calculated misorientations and identified grain boundaries, and then they isolated random (special) boundaries and identified "clusters" of like-type boundaries by graph-searching. Figure 1 shows qualitative results for increasing the number of processing cycles.

Grain boundary engineering has been found to cause remarkable changes in the microstructural topology, reducing the scale of random boundary networks and increasing those composed of special boundaries.

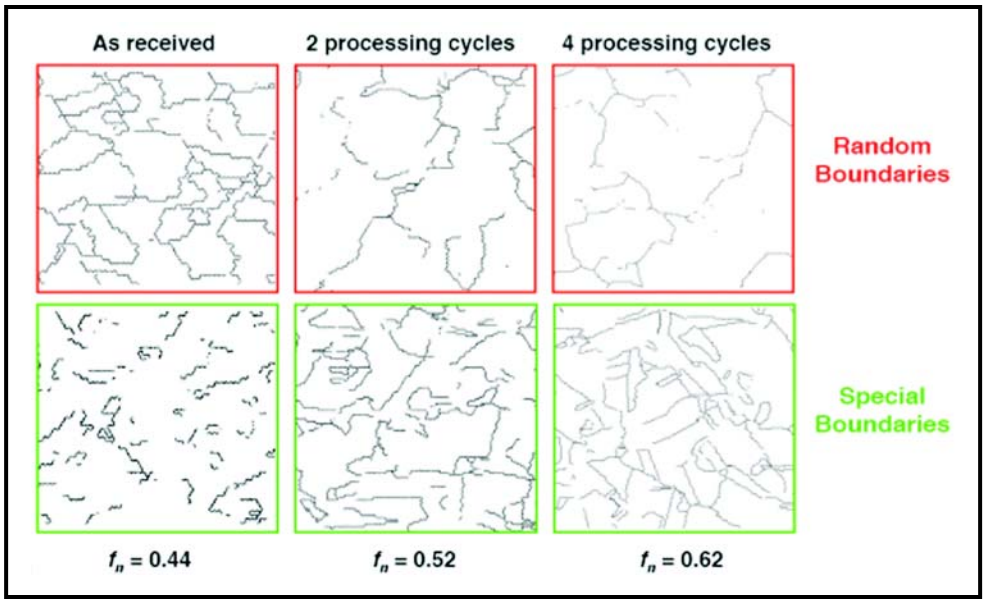


Figure 1. Qualitative results, a topological inversion.

Task 2. Preparation of Test Microstructures. The objective of this task was to identify optimum processing techniques in order to obtain alloy microstructures with different fractions of special boundaries and random boundary network connectivity. Researchers reviewed candidate commercial heats of stainless steel and Inconel Alloy 600 from among those archived at General Electric, and identified type 304 stainless steel (SS304 [heat AJ9139]) and Alloy 600 (heat 3110439) as optimal for this program. The first experimental test material, SS304, was subjected to thermomechanical processing, with each processing cycle consisting of rolling at room temperature to a reduction of 20 percent and annealing at 1,000°C, followed by water quenching. This was repeated up to four times with an annealing time of 10 minutes in each cycle. Researchers analyzed the specimens first in the as-received state and again after two and four processing cycles. They observed that the processed condition had a lower special fraction than the reference condition, and that the grain size of the as-received condition (26 μm) was larger than that on the CSLE sample (18 μm). The team performed another heat treatment after the fourth cycle for an hour at 1,100°C to get a grain size comparable to the as-received condition. This treatment only had a marginal effect on the other microstructural parameters. They observed that the alloy condition maintained its low special fraction (0.49) with a grain size increase to 36 μm. The researchers decided that this condition would be used for testing against the as-received condition. In addition, pieces of stock material in the as-received and heat-treated (1 hr at 1,100°C) conditions were forged by 20 percent at 140°C and subjected to similar testing.

The microstructural observations for SS304 from these experiments show that after four cycles of thermomechanical processing and additional heat treatment, samples have a lower fraction of special boundaries as well as a more connected network of random grain boundaries. In addition, some samples were processed to boost the special fraction, and the experiments provided further data on the connectivity of grain boundary networks.

Researchers processed Inconel 600 in a similar manner and developed comparisons between the as-received and the processed microstructures. It is evident, both visually and from the data, that the connectivity of the random boundary networks is considerably broken in the processed condition.

Task 3. Stress Corrosion Cracking Testing. These research activities, which were carried out at UM and GECRD, focused on determining whether modified grain boundary character distributions resulting from thermomechanical processing influenced the cracking behavior of SS304 or Alloy 600 under different water chemistry conditions. Researchers performed these experiments using fracture mechanics specimens (0.5T or 1T CT specimens) in sophisticated equipment, which involved precision control and monitoring of water chemistry and corrosion potential, digital servo-control loading systems under computer control, high pressure/high temperature autoclave systems with digital temperature controllers, continuous high-resolution dc potential drop monitoring of crack length, and computer data acquisition and test control.

Researchers at UM conducted constant extension rate testing (CERT) of SS304 alloy samples in both the as-received and thermo-mechanically processed conditions. In addition to these conditions, an as-received/processed pair of samples was forged by 20 percent at 140°C in order to have similar conditions to those tested at GE. The previous NERI annual report described the SS304 experimental results in detail. During the past year, researchers conducted similar SCC testing on Alloy 600 samples. Results are presented and compared to the previous SS304 experimental data.

For the CERT experiment at UM, researchers strained all four SS304 tensile specimens simultaneously at an initial strain rate of 3×10^{-7} s^{-1} in an aggressive environment consisting of 288°C non-deaerated water (~8,000 ppb dissolved oxygen), and a conductivity of 0.5 μS/cm. Only three of the SS304 samples reached 20-percent strain and one sample failed at 8-percent strain, as shown in Figure 2. Inspection in the SEM of the EBSD scanned areas revealed that no significant cracking took place in the non-deformed samples in spite of the more aggressive environment. This observation is consistent with previous work on non-irradiated SS304 samples in similar experimental conditions.

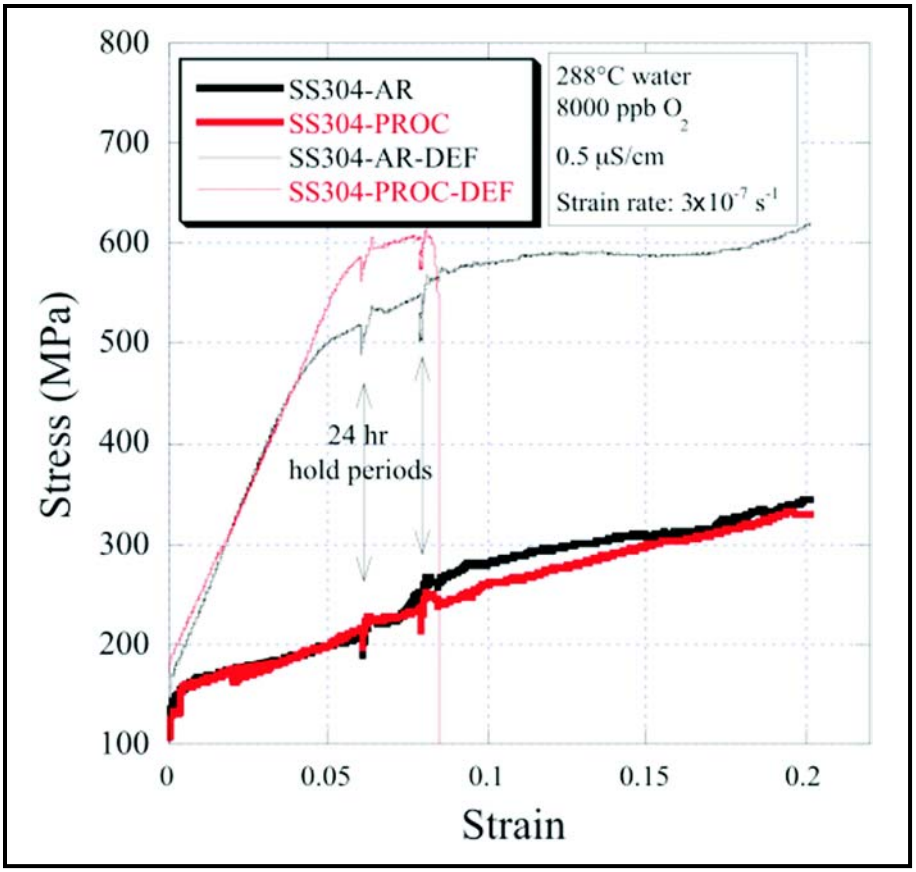


Figure 2. Stress-strain curves for the four SS304 samples tested.

On the contrary, the samples forged by 20 percent prior to CERT testing were found to develop cracks on the surface. Assuming that this cracking was initiated at the beginning of the CERT experiment, the estimated crack growth rate for the as-received forged sample was:

$CGR \cong 4.52 \times 10^{-7}$ mm s^{-1}.

After processing, the low-CSL, low connectivity forged SS304 sample failed at approximately 8-percent strain. Approximately seven cracks were counted on the surface of this sample, mostly in the vicinity of the fracture surface. Cracks at this location suggested that the failure of this sample might have initiated intergranularly. However, the fracture surface showed a transgranular fracture mode, although high magnification images gave some indication of intergranular cracking, mostly near the sample surface. Regardless of the failure mechanism, the low-CSL, low connectivity forged sample failed before its as-received counterpart. In the same manner, assuming that cracking was initiated at the beginning of the CERT experiment, and considering that this sample lasted for 120 hours, a crack growth rate can be estimated at:

$CGR \cong 5.09 \times 10^{-6}$ mm s^{-1},

which is approximately an order of magnitude higher than the as-received sample.

Four Alloy 600 tensile bars were tested simultaneously in an environment consisting of 360°C deaerated, hydrogenated high-purity water simulating PWR conditions. Oxygen levels were reduced below 5 percent by bubbling pure hydrogen and adding LiOH and H_3BO_3 for solution concentrations of 0.001M and 0.01M, respectively. The test specimens had been solution annealed to increase the grain boundary special fraction in both as-received samples and samples deformed by cold working. As for the SS304 experiments, researchers applied an initial strain rate of 3×10^{-7} s^{-1}.

All four experiments resulted in the test specimens rupturing, with the solution-annealed samples failing much more quickly. In the unprocessed state, the as-received and cold worked samples achieved strain of 46 percent and 9.2 percent, respectively, before rupturing

(Figure 3). Processing to increase the special fraction resulted in maximum strain values of 24 percent and 6.8 percent. From this data, researchers concluded that processing Alloy 600 to increase the special fraction results in considerably higher susceptibility to intergranular cracking. These results are considered highly unusual and are, as yet, unexplained.

Additional testing at GE compared the SCC growth rate response of the different SS304 microstructures (with fatigue pre-cracking) under different water chemistry conditions. The data were obtained in high purity water representative of typical boiling water reactor and pressurized water reactor conditions. Results were promising, indicating that a higher fraction of special grain boundaries in SS304 retards SCC growth. The observed benefit was consistently increased at low stress intensity factor, which correlates with small crack sizes in plant components. Similarly, a more dramatic benefit is expected on smooth surfaces during the crack nucleation process.

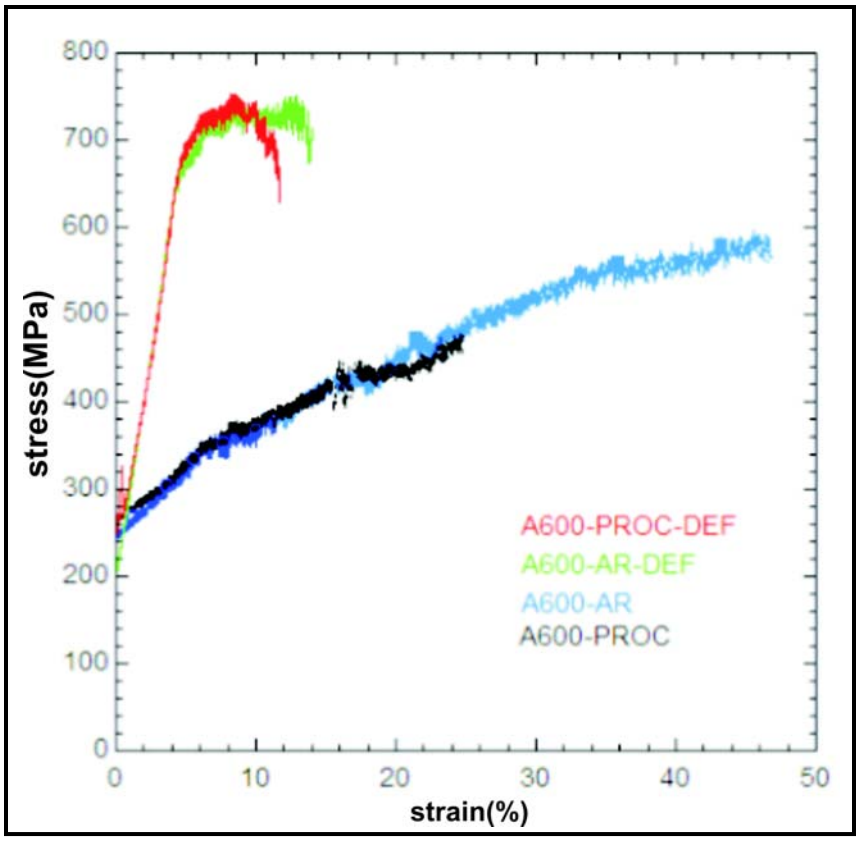


Figure 3. Constitutive response of Alloy 600 in 360°C primary water at a strain rate of 3 x 10^{-7} s^{-1}.

Researchers continued to refine their analytical models. As grain boundary networks are not described by standard network theory, they developed Monte Carlo simulations to explore issues of connectivity and percolation, providing valuable insight into the material microstructural design. These simulation techniques have generated grain boundaries networks that accurately reproduce experimental data.

Overall contributions of this project include an improved understanding of the influence of random grain boundary network connectivity on IGSCC. This knowledge will enable greater predictability of the material's useful lifetime before failure and makes it possible to predict the growth of pre-existing cracks in components. The outcome of the project can be used both in identifying a mitigation strategy for IGSCC in current LWR conditions and enabling the development of economically and operationally competitive water-cooled advanced reactor systems.

NUCLEAR ENERGY RESEARCH INITIATIVE

Reactor Physics and Criticality Benchmark Evaluations for Advanced Nuclear Fuel

PI: William J. Anderson, Framatome ANP, Inc.

Collaborators: Sandia National Laboratories, Oak Ridge National Laboratory, University of Florida (UF)

Project Number: 01-124

Project Start Date: September 2001

Project End Date: August 2005

Research Objectives

The objective of this project is to design, perform, and analyze critical benchmark experiments for validating reactor physics methods and models for fuel enrichments greater than 5-weight percent ^{235}U. These experiments also provide additional information for application to the criticality-safety bases for commercial fuel facilities handling greater than 5-weight percent ^{235}U fuel. Because these experiments are designed not only as criticality benchmarks but also as reactor physics benchmarks, they include measurements of critical boron concentration, burnable absorber worth, and relative pin powers.

Research Progress

Phase 1 of this project focused primarily on designing the experiments using available fuel; preparing the necessary plans, procedures, and authorization basis for performing the experiments; and preparing for the transportation, receipt, and storage of the unirradiated Pathfinder fuel previously stored at the Pennsylvania State University (PSU).

In Phase 1, Framatome ANP, Inc., Oak Ridge National Laboratory (ORNL), and Sandia National Laboratories (SNL) designed the proposed experiments and prepared the safety authorization to perform these experiments at the SNL facility. The proposed experiments were documented in the project's Annual Report issued September 2002.

Work in Phase 1 also included the development and submittal of a license application for a shipping package in support of the transportation of fuel from PSU to SNL. On February 10, 2003, the NRC issued the WE-1 Shipping Container Safety Evaluation Report and Certificate of Compliance. The WE-1 is the only known shipping package capable of housing this fuel and licensed to transport uranium dioxide fuel with enrichments up to 7.5-weight percent ^{235}U. This major accomplishment required significant technical effort in the form of criticality benchmarking for fuels in the 7.5-weight percent enrichment range and structural design and analysis.

Phase 2 was the manufacturing and application phase of this project. Preparation for transportation, receipt, and storage of the Pathfinder fuel continued into Phase 2. However, the primary focus was on the actual experiments. These tasks included analyzing the proposed experiments using current industry computer codes, shipping the fuel from PSU to SNL, and fabricating fuel rods using the deconstructed Pathfinder fuel for use in the experiments. A code-to-code comparison report analyzing the proposed experiments was issued in February 2003 and was included in the Quarterly Status Report for Quarter 6.

Phase 3, which received a 12-month, no-cost extension through August 31, 2005, involved transporting the remaining nine shipments of fuel from PSU to SNL, downloading the fuel from the Pathfinder elements, reinserting the fuel into aluminum cladding for use in the experiments, and installing hardware for the experiments.

The first experiment fuel element was assembled on July 2, 2004. Assembly of the fuel elements continued until August 26, 2004, when element number 2200 was assembled. A special welding process was developed to weld the end caps onto these elements. The welding operations are complete and the elements are ready for the first core.

The procurement of all experiment hardware has been completed. The new hardware and the control system were mated with the existing critical assembly. Figure 1 shows a view of the critical assembly taken during testing of the hardware. As shown in the figure, the two safety elements and the control element (with drive attached) protrude above the upper guide plate. Ancillary hardware used to measure the drop time of the safety elements is

located near those elements. The two detectors of the plant protection system reside in the dry wells on either side of the core. The upper end of the neutron source extends above the guide plate in the central position in the core.

In Phase 3, all of the reactor operator training was completed, and the operating procedures from previous critical experiments were updated for the current experiments. A new procedure for preparation and monitoring of the borated moderator was also developed.

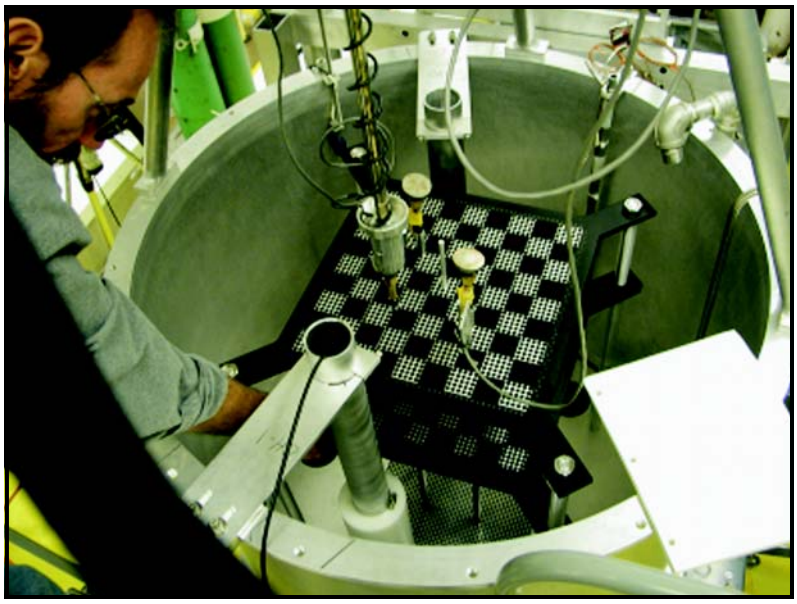


Figure 1. The core of the critical experiment during hardware testing.

An Operational Readiness Review (ORR) of critical experiment operations must be completed prior to commencement of the critical experiments. The purpose of the ORR is to assess the readiness of SNL to begin operations in accordance with the Authorization Basis approved by SNL line management and the Department of Energy. Significant delays in the ORR process have placed the completion of the ORR and the operation of the experiments in direct conflict with other high-priority programs in the Sandia Pulsed Reactor Facility (SPRF). This has resulted in the experiments being placed on hold while the other programs are completed. This action will result in a significant delay in the completion of the experiments.

The status of this issue will continue to be tracked. The project is moving forward, watching for an operational window in which to run the experiments in the SPRF.

Researchers have identified a procedure to quantify the isotopics, impurities, and their uncertainties in the Pathfinder fuel. A sampling of excess fuel pellets will be shipped from SNL to ORNL for analysis at the Radioactive Materials Analytical Laboratory using a high-resolution, inductively coupled plasma mass spectroscopy process. An article detailing the experiment configurations and the sensitivity and uncertainty analysis used to demonstrate their applicability to commercial fuels with higher enrichments has been accepted for publication in *Nuclear Technology.*

This project also involves evaluating typical fuel-processing operations to determine the limits and restrictions required for fabricating higher enriched fuel. During Phase 3, the fuel cycle analysis was completed. The University of Florida submitted a paper for publication in *Nuclear Technology* documenting the economic analysis of the feasibility of using greater than 5 $^{w}/_{o}$ enriched ^{235}U commercial fuel, integrating all aspects of the nuclear fuel cycle. The results of this study indicate that the incentive to increase enrichments depends upon the cycle length used and the current rate of interest. According to the university analysis, the commercial nuclear industry could potentially save millions of dollars per year in reactors operating on a 24-month cycle by increasing their fuel enrichments up to 6.5 $^{w}/_{o}$ ^{235}U. The study concluded that nuclear power plants could be modified economically to handle the higher enrichment.

Planned Activities

The remaining activities planned through the end of this project include:

- Perform isotopic assessment of fuel samples from the Pathfinder fuel.
- Perform the critical experiments, when a window of operation arises.
- Analyze the experiments using industry methodology.
- Document the experiments.
- Evaluate and document the experiments for inclusion in the *International Handbook of Evaluated Criticality Safety Benchmark Experiments.*

NUCLEAR ENERGY RESEARCH INITIATIVE

Fundamental Understanding of Crack Growth in Structural Components of Generation IV Supercritical Light Water Reactors

PI: Iouri Balachov, SRI International

Collaborators: VTT Manufacturing Technology

Project Number: 01-130

Project Start Date: August 2001

Project End Date: Completed July 2004

Research Objectives

The objectives of this project were to:

- Increase the understanding of the fundamentals of crack growth in structural components of Generation-IV supercritical light water reactors (SCWRs) made of stainless steels and nickel base alloys at supercritical temperatures.
- Provide the tools for assessing the influence of the operating conditions in power plants with supercritical coolant temperatures on the electrochemistry of different types of corrosion processes taking place in the coolant circuits of supercritical power plants.
- Measure material-specific parameters describing the material's susceptibility to stress corrosion cracking and other forms of environmentally assisted degradation of structural materials at supercritical coolant conditions.
- Use these measurements to interpret the rate-limiting processes in the corrosion phenomena and as input data for lifetime analysis.
- Use the SRI-developed FRASTA (fracture surface topography analysis) technique to obtain information on crack nucleation times and crack growth rates via analysis of conjugate fracture surfaces. Identify candidate remedial actions by which the susceptibility to stress corrosion cracking can be decreased.

In this project, researchers used a unique combination of two advanced techniques for studying material reliability. Using Controlled Distance Electrochemistry (CDE) in relatively short experiments, they determined a measurable material parameter that describes the transport of ions or ionic defects in the oxide films. They correlated this with the susceptibility to cracking using FRASTA to reconstruct the evolution of crack initiation and growth.

Research Results

During the first year, researchers built and tested a computer-controlled supercritical water loop system. They performed material studies in a reactor vessel capable of sustaining supercritical temperatures up to 500°C and pressure up to 5,000 psi. The CDE arrangement included contact electric resistance (CER), electrochemical impedance spectroscopy (EIS), and contact electric impedance (CEI) configurations. Researchers installed the CDE arrangement in a replaceable top head of the reactor with supercritical water.

Years two and three of the project proceeded as two parallel activities: (1) electrochemical studies of properties of the oxide films and (2) fracture surface analysis for candidate structural materials. The objectives of the first activity were to:

- Characterize the formation, reduction kinetics, and the properties of metal oxide films using the CER technique.
- Measure the solid contact impedance spectra of oxide films using CEI.
- Characterize the oxidation and reduction kinetics and mechanisms of metals as well as the properties of metal oxide films by using Thin-Layer Electrochemical (TLEC) impedance measurements.
- Derive material-specific parameters which describe the susceptibility of metals to stress corrosion cracking in supercritical coolant conditions for the materials in which crack growth is controlled by the phenomena in oxide films within the crack.
- Identify candidate remedial actions such as changes in water chemistry, material chemical composition, and metallurgical parameters (the degree of cold work in particular) that can decrease the susceptibility to stress corrosion cracking.

The objectives of the second activity were to:

- Expose loaded specimens to a supercritical aqueous environment simultaneously with electrochemical studies of the oxide films on the same materials.
- Examine fracture surfaces of specimens by using the FRASTA technique.
- Identify crack nucleation sites and times for specimens made of different candidate materials. To use FRASTA to determine crack front formation and movement, including formation of discontinuities ahead of the crack and their possible coalescence later while the crack is advancing.
- Estimate crack growth rates for different candidate materials under a set of conditions using FRASTA for examination of conjugate fracture surfaces.
- Correlate electrochemical information on material-environment interactions with crack nucleation and growth data from FRASTA to delineate the fundamentals of uniform and localized degradation of structural materials in Generation-IV SCWRs and to estimate lifetimes of candidate materials for structural components under a variety of normal and offset operating conditions.

Following were the major accomplishments of this project:

- Designed and built a test system for electrochemical and fracture mechanics studies in supercritical water. The system includes an electrochemical part and a fracture mechanics part. The electrochemical part includes a CDE arrangement for in-situ studies of oxide films at supercritical temperatures. The fracture mechanics part includes fracture mechanics specimens for stress corrosion cracking studies and the loading system.
- Obtained systematic experimental data on the charge and mass transfer properties of the oxide films for a representative set of candidate structural materials. CDE measurements in three configurations (CER, CEI, and TLEC) provided in-situ information on the rate of transport of electronic and ionic charge carriers in the oxide film and at the film/supercritical water interface.
- Discovered that kinetic parameters of the charge and mass transfer processes in the oxide films and at the film/supercritical water interface determined from CDE measurements were in good agreement with independent results obtained at VTT. Defined a set of kinetic parameters that describe transport processes. This set included interfacial rate constants for the generation and consumption of ionic current carriers, the diffusion coefficient of ionic point defects (vacancies or interstitials), the diffusion coefficient of electronic current carriers, the field strength in the oxide, the thickness of the layer in which point defect transport occurs, and the ability to polarize the oxide/supercritical water interface completed work on deriving kinetic data for oxide films formed on candidate structural materials from CDE measurements.
- Ranked the first group of candidate structural materials (austenitic stainless steels SS304, SS316, and SS347) against their susceptibility to environmentally assisted degradation. Stress corrosion cracking (SCC) was not observed for nickel alloys, or martensitic stainless steel alloys T91 and HT9. The ranking was based on the in-situ data on film stability parameters (or film transport properties) measured with a CDE arrangement designed at SRI.
- Researchers compared these observations and the results of CDE measurements at SRI. They believe that the observations clearly indicate a correlation between measurable oxide film properties and the susceptibility of austenitic steels to environmentally assisted degradation. Experimental proof of the existence of such correlation was one of the major goals of the completed work. Researchers believe their experimentally proven ability of the economical CDE technique to supply in-situ data for ranking candidate structural materials for SCWR is a major accomplishment of this project.
- Examined the morphology and thickness of the oxide films with SEM and measured the chemical composition of the films with the EDX technique. Direct measurements of film thickness were in good agreement with in-situ film thickness measurements using the CDE technique. They found oxide films to be enriched in Cr (relative to the base metal), and found the Cr concentration to decrease in the outer surface layer of the film.
- Researchers also estimated the potential use of the CDE technique for building in-situ sensors for monitoring water chemistry in the heat transport circuit of Generation-IV SCWRs. Two types of sensors are envisioned: (1) a monitor to measure oxide film stability and, accordingly, the susceptibility of the in-vessel

structural materials to various forms of environmentally assisted degradation, and (2) a water conductivity sensor working in a wide temperature range—from room temperature to supercritical temperatures.

- Found that multiple data on the crack front movement obtained from FRASTA confirmed that the cracks in austenitic stainless steels advance by formation of discontinuities ahead of the crack front, followed by their coalescence. The occluded environment formed by discontinuities becomes filled with supercritical water, most likely creating an aggressive medium. Formation of the thick deposits in the areas of discontinuities confirmed this hypothesis.
- Obtained experimental data during the reporting period to allow estimation of the crack growth rates for austenitic stainless steels in supercritical water. This group included SS304 as a "baseline" material and SS316 and SS347 as candidate materials. Our results indicated that SS347 is expected to have an order-of-magnitude lower crack growth rate than SS304 and that the rate for SS316 is expected to be several times lower than that for SS304.
- Demonstrated a combined effect of film repassivation rate and stability of the oxide film on SCC behavior and life-time of structural materials for a representative set of oxidizing conditions. Researchers suggested an approach for quantitative estimation of materials lifetime depending on material and environment combination.

NUCLEAR ENERGY RESEARCH INITIATIVE

New Design Equations for Swelling and Irradiation Creep in Generation IV Reactors

PI: Wilhelm G. Wolfer, Lawrence Livermore National Laboratory (LLNL)

Collaborators: Pacific Northwest National Laboratory (PNNL), University of California - Berkeley

Project Number: 01-137

Project Start Date: October 2001

Project End Date: Completed September 2004

Research Objectives

The objectives of this research project were to develop physical models for radiation-induced microstructural changes in structural materials to be used in Generation IV nuclear reactors and to derive from these the constitutive laws for void swelling, irradiation creep, stress-induced swelling, and changes in mechanical properties. The approach was based on modeling the cause-effect relationships from the fundamental atomistic processes to the macroscopic constitutive laws.

Researchers tested and validated both the microscopic models and the macroscopic constitutive laws by analyzing data on electron microscopy, density, irradiation creep, and post-irradiation tensile strength and measuring diameter changes of the pressurized tubes. Validating both microstructure models and macroscopic constitutive laws provided a more stringent test of the underlying science for radiation effects in structural materials for nuclear reactors.

Research Results

Swelling. Both the methodical data analysis for void swelling in metals and the theoretical predictions with the void nucleation code, which this project developed, revealed that the rate of swelling follows a sigmoidal dose dependence. The rate of swelling begins very small, then accelerates and eventually reaches a constant value. This behavior can be presented by an inverted Fermi-Dirac function:

$$\dot{S} = \frac{\dot{S}_0}{1+\exp[(D_0 - D)/w]} \qquad (1)$$

Here, $\dot{S}_0$ is the final constant swelling rate; D is the irradiation dose in dpa; D_0 is the incubation dose; and w is the transient parameter that determines the abruptness of the onset of swelling. The larger the value of w, the more gradual the transition from low to steady state swelling rate. Integration of equation (1) gives the new void-swelling equation:

$$S = \dot{S}_0 \left\{ D - w\ell n\left(\frac{1+e^{D_o/w}}{1+e^{(D_o-D)/w}} \right) \right\} \qquad (2)$$

Researchers noted that when $D=D_0$, $\dot{S}=\dot{S}_0/2$, meaning the swelling rate was half of the final rate. This final rate was about 1 percent/dpa for austenitic stainless and 0.2 percent for ferritic steels. It was nearly independent of dose rate, temperature, or stress within the range of temperatures where void swelling occurs, namely between 35–55 percent of the melting temperature. Figure 1 shows the quality of fitting swelling data from solution annealed 304 and from 20 percent cold-worked 316 stain-

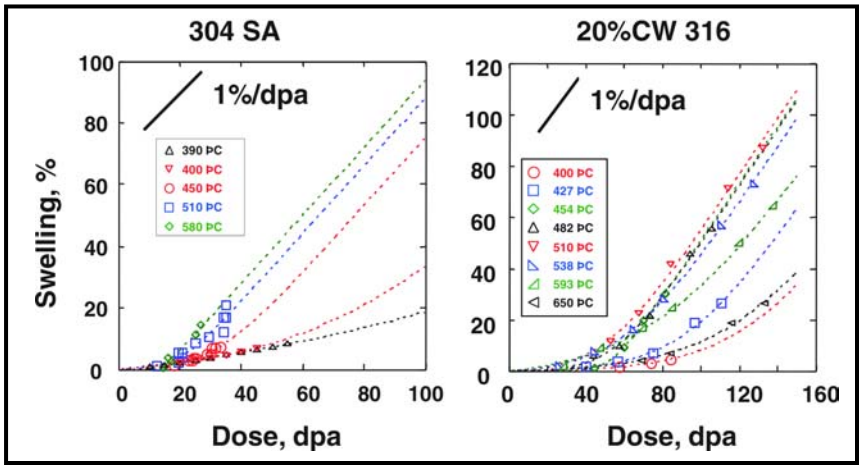


Figure 1. Swelling data for neutron irradiated solution-annealed 304 and for 20 percent cold-worked 316 stainless steels fitted to equation (2).

less steel to the new swelling equation.

In the new swelling equation, only the incubation dose, D_0, and the transient parameter, w, are treated as functions of the irradiation temperature, dose rate, level of cold-work, alloy composition, and radiation-induced segregation.

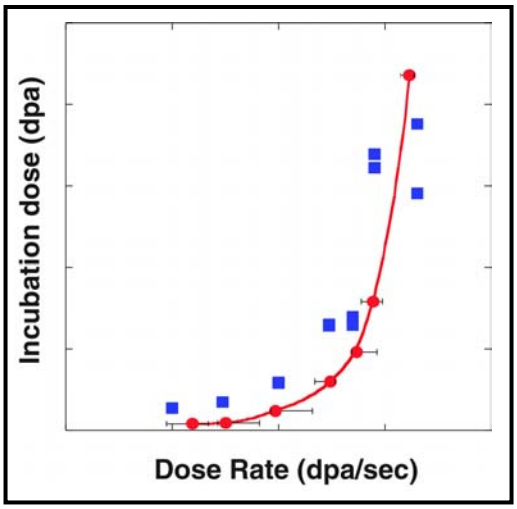


Figure 2. Computed vs. measured incubation dose for void swelling in pure ternary austenitic steels irradiated at about 400°C and at different dose rates.

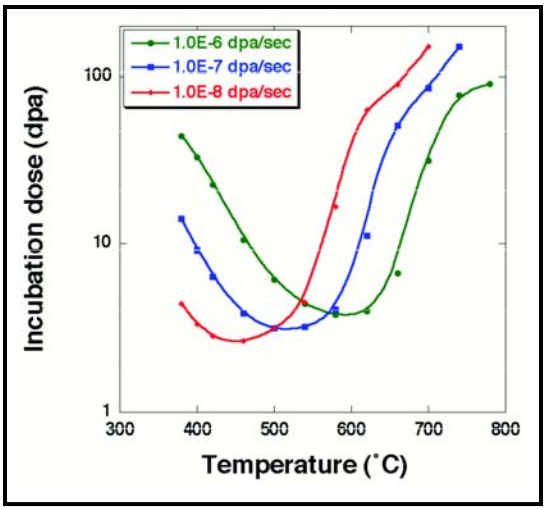


Figure 3. Incubation dose for void swelling in pure ternary austenitic steels as a function of irradiation temperature and dose rate.

The scientific underpinning for the new swelling equation is contained in a void nucleation and swelling code which follows the evolution of the size distribution of vacancy clusters and voids in parallel with the evolution of dislocations. Researchers made no distinction between single self-interstitials and mobile interstitial clusters. They treated all as an effective population of mobile interstitials, possessing an average diffusivity and bias factor per interstitial for voids and dislocations, which prevents introducing a "production bias." The bias factors were combined with the sink strength present at any given dose to determine the net bias for void swelling. Due to the evolution of the net bias with dose, the transient in void swelling was reproduced. Furthermore, steady state swelling emerged as dislocations and voids evolved towards the same sink strength.

Researchers applied the comprehensive void nucleation and swelling code to the experimental results of a simple ternary austenitic alloy irradiated in DOE's Fast Flux Test Facility at Hanford. Figure 2 shows the results of comparing measured incubation dose against the computed values.

Researchers obtained the best agreement when they chose 1.4 eV as the activation energy for vacancy migration. The strong dose rate effect seen in Figure 2 applied only to irradiation temperatures around 400°C. As seen from the theoretical predictions in Figure 3, at intermediate irradiation temperatures the incubation dose was not strongly dependent on dose rate. At high irradiation temperatures, the dose rate dependence was reversed.

Stress-Enhanced Swelling. Through a review of all available data on the effect of stress on void swelling, researchers confirmed their theoretical conclusion that stress affects the incubation dose for void swelling, but not the final rate of swelling. Fitting the data from pressurized tube irradiations to equation (2) revealed that the transient parameter w appears to be independent of stress, while the incubation dose remains constant or decreases with rising hoop stress. The stress has an effect only when the incubation dose is large, as Figure 4 illustrates for 20 percent cold-worked stainless steels.

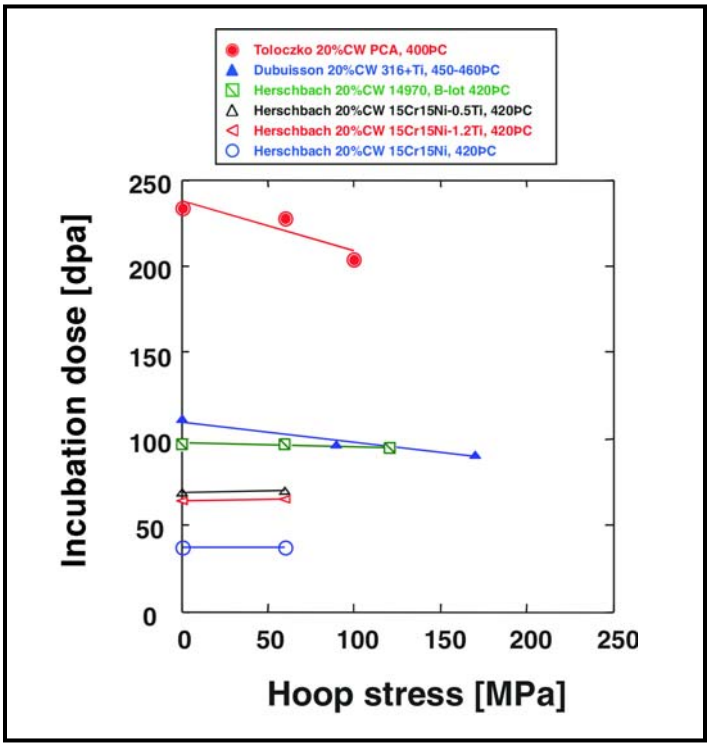


Figure 4. The effect of hoop stress on the incubation dose for void swelling.

Irradiation Creep. Several mechanisms contribute to the portion of irradiation creep that depends on the deviatoric stress, and this dependence should be linear.

However, a comprehensive analysis of the data with a stress dependence of σ^n shows that the stress exponent n varies considerably but is, on average, somewhat larger than unity (Figure 5).

Summary. The detailed microstructural models developed in this project for the formation and evolution of voids, dislocation loops, and network dislocations have been incorporated into a computer code which produces the observed void swelling curves for neutron-irradiated steels. The dependence of void swelling on irradiation temperature, neutron flux or dose rate, initial dislocation density, helium production, and stress is largely confined to the incubation period. Based on predictions from the detailed void nucleation and growth code, researchers proposed a new constitutive equation for void swelling that captures the essential features of the detailed models and can readily serve as a design equation for nuclear reactor design studies.

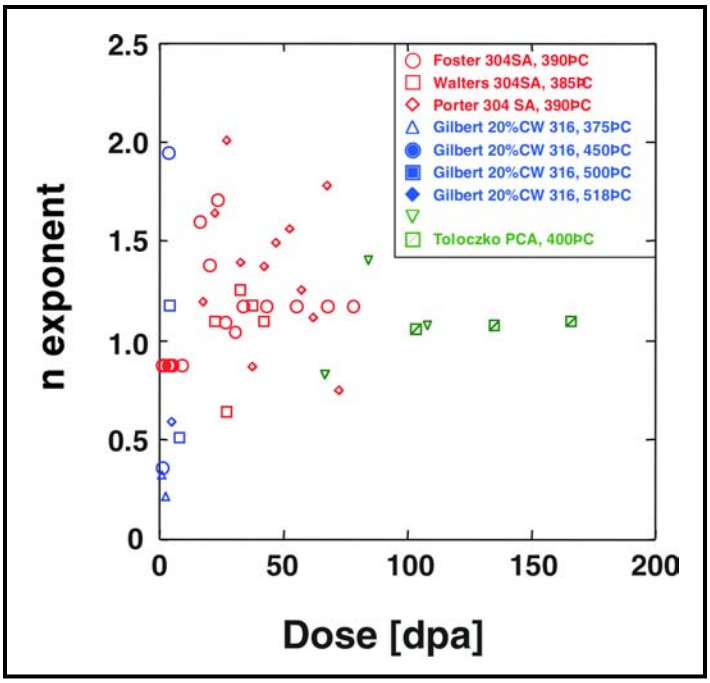


Figure 5. The exponent (n) for the stress dependence of irradiation creep, σ^n.

NUCLEAR ENERGY RESEARCH INITIATIVE

Development and Validation of Temperature-Dependent Thermal Neutron Scattering Laws for Applications and Safety Implications in Generation IV Nuclear Reactor Designs

PI: Ayman I. Hawari, North Carolina State University

Collaborators: Oak Ridge National Laboratory; Instituto Balseiro, Argentina

Project Number: 01-140

Project Start Date: September 2001

Project End Date: May 2005

Research Objectives

The overall objective of this project was to develop an improved model for thermal neutron scattering laws in order to understand and accurately determine the operating and safety characteristics of advanced reactor designs (such as the moderator temperature coefficient of reactivity and the buildup and depletion of transuranic elements). Researchers would accomplish this work through a critical review of the currently used thermal neutron scattering laws for various moderators as a function of temperature in order to generate a database of benchmarks from a well-documented and representative set of experimental data that is sensitive to the neutron spectra. To establish the scattering laws, they would update models and model parameters by introducing new developments in thermalization theory and condensed matter physics into various computational approaches, and benchmark the results against the experimental set. In the case of graphite, researchers would perform a validation experiment by observing neutrons slowing down as a function of temperatures equal to or greater than room temperature. This research would have applications to Generation IV nuclear power systems, advanced research reactors, and power reactors for space applications.

Research Progress

During the third year, researchers successfully used ab-initio methods to calculate the phonon distributions required to generate the thermal-neutron scattering cross sections for graphite, beryllium, zirconium, and thorium compounds (Be, BeO, ZrH_2, and ThH_2). Figure 1 shows a comparison between the ab-initio dispersion relations and

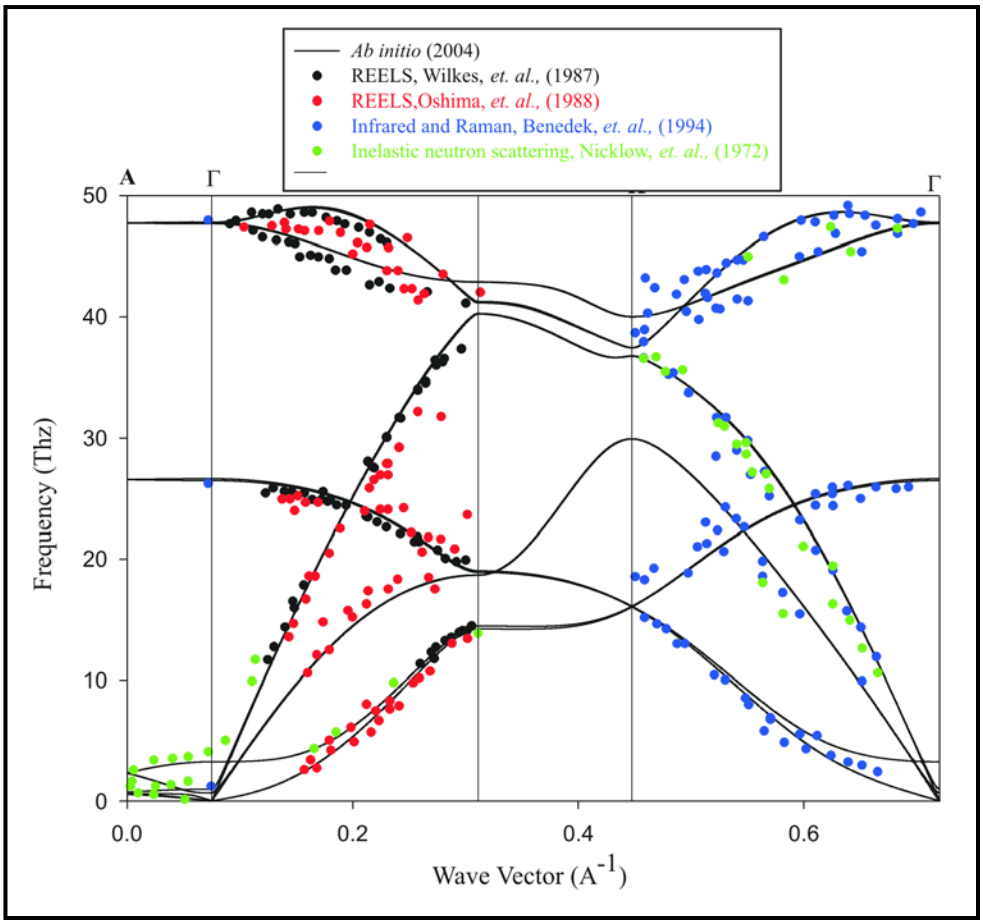


Figure 1. The dispersion relations for graphite as compared to experimental data. The calculation is based on a 6x6x1 supercell model and a 3x3x4 k mesh.

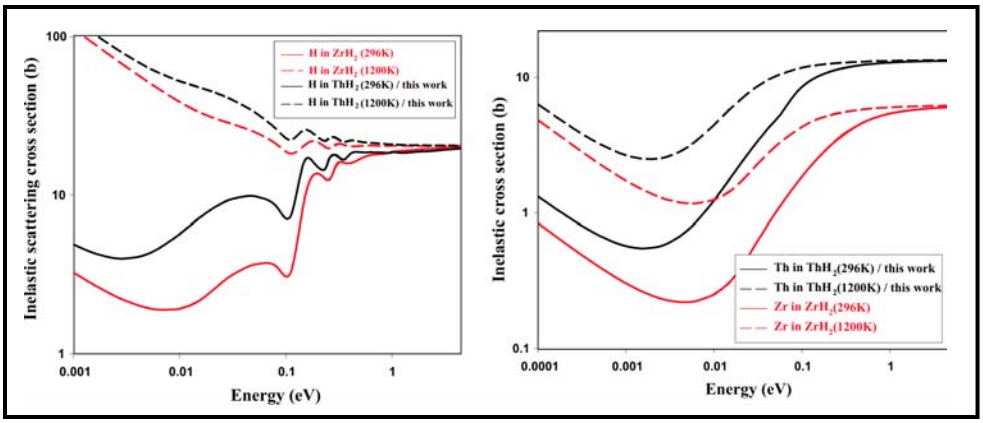


Figure 2. Inelastic scattering cross section of H, Th, and Zr in ThH_2 and ZrH_2.

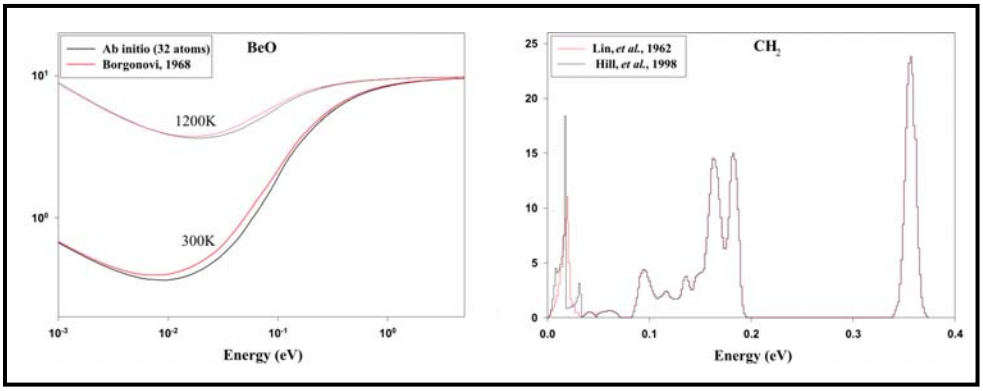


Figure 3. The themal neutron scattering cross sections for BeO (left). The phonon spectrum of polyethylene (right).

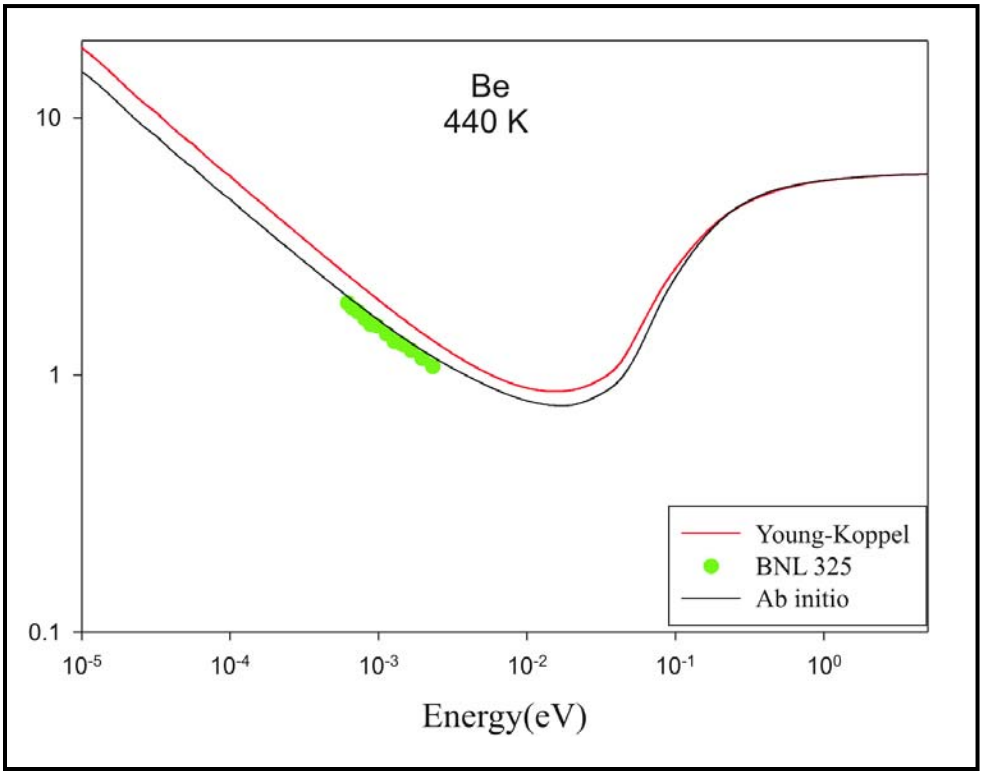


Figure 4. The impact of the one-phonon coherent inelastic scattering component on the inelastic scattering cross section of Be.

an extensive set of experimental data for graphite. As can be seen, the agreement is excellent. The Be results were previously reported during phase 2. Researchers have now completed both their graphite and beryllium models. Figure 2 shows the thermal neutron scattering cross sections for ZrH_2 and ThH_2 as generated by the NJOY/LEAPR code system. Figure 3 illustrates the results for BeO. In this case, the results of the North Carolina State University (NCSU) analysis are in good agreement with those obtained using the Borgonovie model, which is the basis of the common data libraries. In the case of polyethylene, researchers generated the thermal scattering cross sections using a previously unpublished phonon frequency spectrum (Figure 3).

During this phase, researchers continued their calculations of the thermal neutron scattering cross section libraries for Be and graphite using the one-phonon coherent inelastic scattering models from NCSU. Figure 4 shows the results for beryllium. Researchers are currently finalizing the graphite results.

Researchers completed the set-up of the graphite experiment at the Oak Ridge Electron Linear Accelerator (ORELA) facility. All of their equipment was placed in the "electron room." Computers for data acquisition, temperature monitoring, and control were set up in "Lab A," and communication among the detectors, temperature control system, and computers was established using Ethernet and BNC connections.

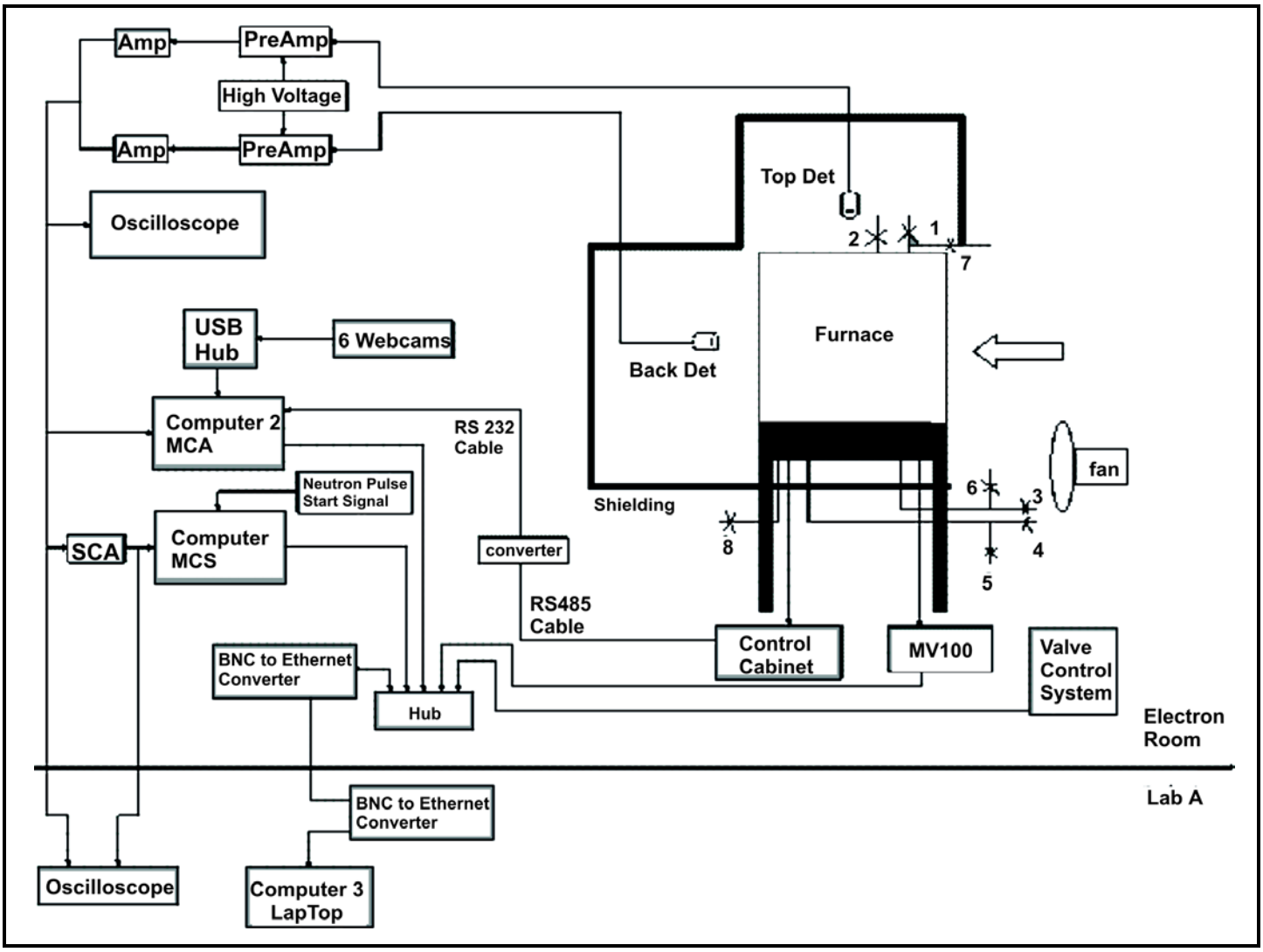


Figure 5. A schematic of the graphite experiment as it currently stands at ORELA.

Figure 5 shows an overall diagram of the entire experimental system as it currently stands at ORELA. Figure 6 shows a picture of the assembled system in position at the end of the ORELA beam line, including the borated polyethylene structure which surrounds the graphite heating system.

Planned Activities

The project has been extended to complete the ORELA experiment due to major maintenance problems at the accelerator facility. Once it is back on-line, researchers will execute the experiment to benchmark the thermal neutron scattering libraries of graphite as a function of temperature.

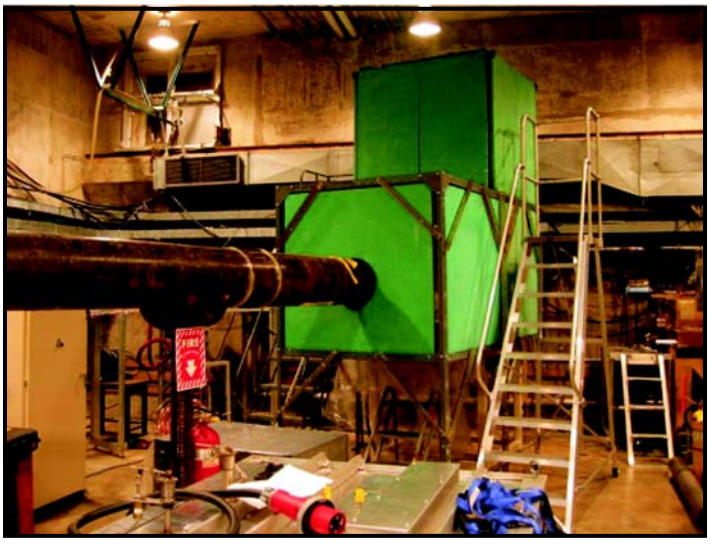


Figure 6. The assembled experimental system showing the polyethylene shielding structure surrounding the graphite heating system.

NUCLEAR ENERGY RESEARCH INITIATIVE

The Oxidation of Zircaloy Fuel Cladding in Water-Cooled Nuclear Reactors

PIs: Digby D. Macdonald and Mirna Urquidi-Macdonald, Pennsylvania State University

Collaborators: None

Project Number: 02-042

Project Start Date: September 2002

Project End Date: September 2005

Research Objectives

This work seeks to develop a comprehensive model that can be used to predict performance and assess the risk of failure of Zircaloy fuel cladding in commercial boiling water reactors (BWR) and pressurized water reactors (PWR) under high burn-up conditions.

Research Progress

Task 1. Modified Boiling Crevice Model. Under this task, researchers have modified the Boiling Crevice Model to describe the evolution of the environment in crud pores and, therefore, in contact with the cladding Zircaloy surface. The model considers heat transfer conditions both of low superheat (nucleate boiling in PWRs) and high superheat (sustained boiling in BWRs). Boiling is believed to be the mechanism for the concentration of Li^+ and $B(OH)_4^-$ in crud pores on the fuel cladding and, ultimately, for the precipitation of $LiB(OH)_4$, which results in the Axial Offset Anomaly. Therefore, researchers have derived the volume-averaged concentration for $LiB(OH)_4$ in the pore as follows:

$$C^* = \theta S_0 C_0 \exp(-t/\tau) + \theta S_0 C_{\lim}(1-\exp(-t/\tau)),$$

where S is the saturation; θ is the porosity; subscript 0 means the mouth of the pore; $C_{\lim}$ is the limiting concentration in the pores; and $\tau = \frac{\theta S_0 C_{\lim}}{C_0 B}$.

Task 2. Development of Point Defect Model. The objective of this task is to develop a Point Defect Model (PDM) for the growth of bilayer passive films on zirconium and Zircaloys under PWR and BWR coolant conditions, where hydride and oxide barrier layers form, respectively. Both of these alloys are covered by a porous outer layer. Starting with equations describing the total steady-state current density, open circuit potential, and coulombic efficiency, the researchers derived formulas for the thickness of the hydride barrier layer under PWR coolant conditions and for the oxide barrier layer under BWR conditions. They then derived an equation for porosity in the outer layer under PWR and BWR conditions by assuming the Gibbs energy change for the reaction $ZrO_2^{+}+H_2O \rightarrow ZrO_2+2H^+$ in the pore interior to be 0.

Task 3. Incorporation of the PDM into the Mixed Potential Model. This task incorporates the modified PDM into the mixed potential model (MPM), in order to describe the oxidation of Zircaloy under open circuit conditions. The task has now been largely completed, at least from the theoretical viewpoint, as researchers have derived an expression for the open circuit potential of the alloy in contact with the appropriate coolant. However, they are awaiting the acquisition of model parameter values for Zircaloy, including values for the kinetic parameters for the reduction of oxygen and the evolution of hydrogen. The researchers are measuring values in Task 6, but classify this task as having been completed, pending a final insertion of parameter.

Task 4. Radiochemistry/Electrochemical Modeling. The objective of this task was to develop robust radiolysis codes for calculating the concentrations of electroactive radiolysis products in the primary coolant circuits of commercial BWRs and PWRs, particularly in the vicinity of the fuel, in order to provide the link between reactor operating conditions and Zircaloy oxidation. The present codes are now sufficiently sophisticated to provide accurate simulations of the electrochemical conditions that exist within the primary coolant circuits of both BWRs and PWRs, thereby achieving the goals of this task.

Task 5. Modeling Growth Stresses. This task involves modeling the evolution of growth stresses within the growing oxide film, taking into account creep of the substrate. An accurate model for this process is required in order to estimate the time of mechanical oxide rupture (i.e., transition in growth kinetics or the onset of nodular

attack). Growth stresses arise because oxygen vacancies at the metal/barrier layer interface generate excess volume for which other processes at this interface, such as annihilation of cation vacancies, do not compensate. Thus, the rate of generation of excess volume, and therefore the rate of generation of stress, which is related to the cube root of the excess volume, can be correlated with the fluxes of the vacancies across the barrier layer. However, stress relaxation also occurs via plastic deformation of the substrate, which must be included to obtain an accurate description of the mechanical properties of oxide films.

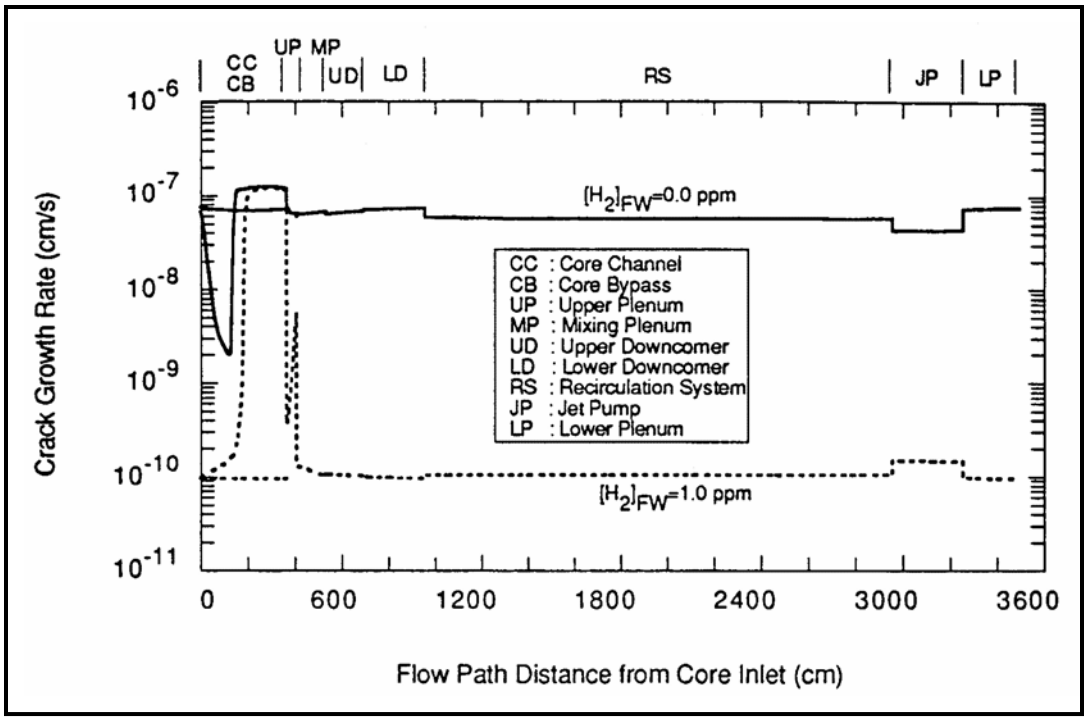


Figure 1. The relationship between the rate of generation of stress and the fluxes of the vacancies across the barrier layer.

Task 6. Experimental Measurement of Model Parameters. The purpose of this task is to ascertain the appropriate oxidation mechanism and the predictions of diagnostic criteria discussed in the original proposal with experimental data. This task also involves the experimental measurement of critically important model parameters, such as the electrochemical kinetic parameters for the redox reactions that occur on the metal surface, diffusivity of oxygen vacancies in the barrier layer, and other parameters, including passivity breakdown potentials, corresponding to the onset of "nodular attack." The measurements are being made using potentiodynamic and impedance techniques as a function of voltage sweep rate and frequency, respectively, and as a function of heat treatment.

Researchers have largely completed work to characterize the zirconium oxide film formed on pure zirconium surfaces under simulated BWR and PWR primary coolant conditions, including polarization scans, impedance measurements, Mott-Schottky analysis, current transients, thickness transients, etc. Based on these measurements, various properties of zirconium oxide films have been determined, including: electronic structure, donor density, effect of potentials on film formation rate and steady state thickness, and dissolution rates. Nodular corrosion is clearly shown on the as-received samples, while only uniform corrosion is shown for samples polished to a 1 μm finish after exposure to simulated BWR core conditions.

The current density transients and the observed changes in film thickness of zirconium passive films have confirmed that the new rate law derived from the PDM can be used to predict the performance of zirconium passive film growth and thinning at high temperatures, as shown by experimental results of Figure 2. The dissolution rate is $c \approx 0.006$ nm/s, measured by stepping the potential from 0.90 V_{she} to 0.60 V_{she} and simultaneously measuring the capacitance of the film as a function of time. Researchers then converted the capacitance data into film thickness using the parallel plate capacitor model and an assumed value for the dielectric constant for ZrO_2. They are also working on extracting important model parameters from the electrochemical transients in the current and film thickness. The independently obtained kinetic parameters will provide a means of assessing the success of the theory.

Planned Activities

Researchers will continue working on model development and experimental measurements over the next phase of the project. They will continue to develop more advanced versions of the models for the passive films on zirconium and Zircaloys under prototypical BWR and PWR core operating conditions and will encode the models so that the properties of the films can be calculated and

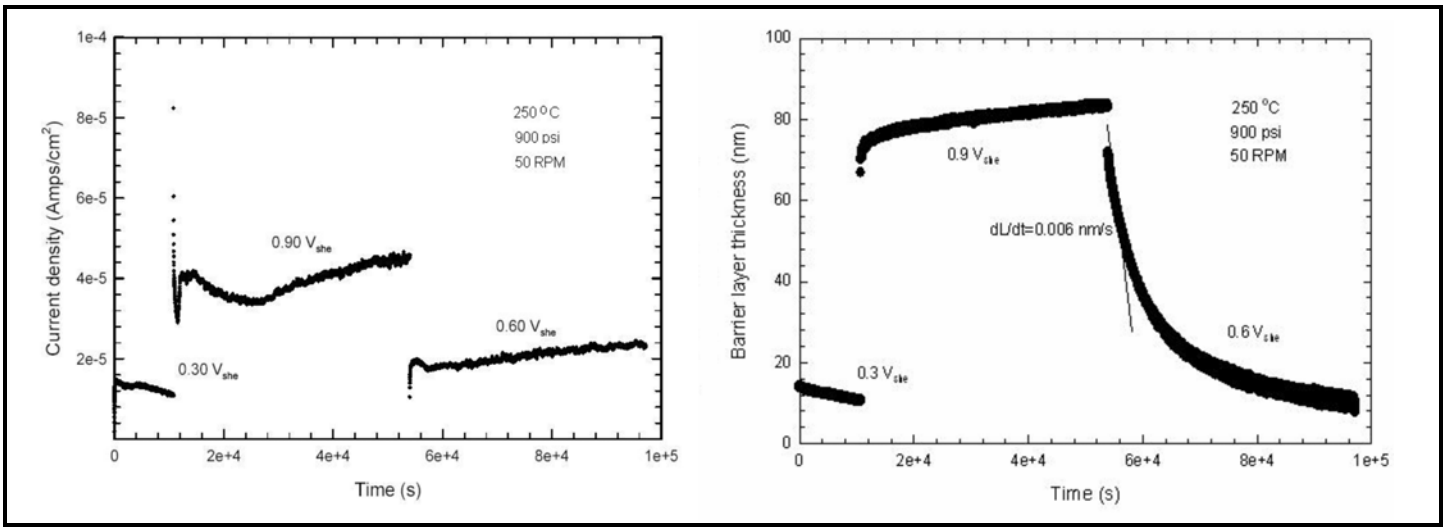


Figure 2. Experimental transients of current density and film thickness for the growth and thinning of the passive film on zirconium in 0.1M $B(OH)_3$ and 0.001M LiOH solution at 250°C and 900 psi upon stepping the potential from 0.30 V_{she} → 0.90 V_{she} → 0.60 V_{she}. (a) current density, (b) thickness calculated from capacitance (f = 1 kHz).

compared with experimental data. They will also optimize the models by accounting for measured impedance data in order to evaluate key model parameters (see Task 6). Researchers have scheduled experimental work to examine the passivity breakdown potentials, corresponding to the onset of "nodular attack," using potentiodynamic and impedance techniques as a function of voltage sweep rate and frequency, respectively, and as a function of heat treatment, i.e., distribution of second-phase particles.

Upon completing the boiling porous deposit model, they will use its output to simulate the environment in CRUD pores and hence in contact with the Zircaloy surface. Researchers will then compare the properties of passive film grown in this actual environment with those obtained in typical bulk concentration of BWR and PWR. Other planned activities are identified in the tasks above.

NUCLEAR ENERGY RESEARCH INITIATIVE

Incorporation of Integral Fuel Burnable Absorbers Boron and Gadolinium into Zirconium Alloy Fuel Clad Material

PI: Kumar Sridharan, University of Wisconsin

Collaborators: Sandia National Laboratories, Westinghouse Science and Technology Department

Project Number: 02-044

Project Start Date: September 2002

Project End Date: September 2004

Research Objectives

Long-lived fuels require the use of higher enrichments of ^{235}U or other fissile materials. However, high levels of fissile material lead to excessive initial reactivity at the beginning of life. To counteract this excessive activity, integral fuel burnable absorbers (IFBA) are added to some rods in the fuel assembly. The two commonly used IFBA elements are gadolinium, which is added as gadolinium-oxide to the UO_2 powder, and boron, which is applied as a zirconium-diboride coating on the UO_2 pellets using plasma spraying or chemical vapor deposition techniques. The incorporation of IFBA into the fuel has to be performed in a nuclear-regulated facility that is physically separated from the main plant. These operations tend to be very costly because of their small volume and can add 20 to 30 percent to the fuel manufacturing cost. Other manufacturing issues that impact cost and performance associated with the present approach to IFBA additions are maintaining the correct levels of dosing, reducing the fuel melting point due to gadolinium-oxide additions, generating internal pressure in the cladding by transmutation gases, and parasitic neutron absorption at the fuel's end-of-life.

The goal of this research was to develop an alternative approach that allows for the incorporation of boron or gadolinium into the outer surface of the fuel cladding, rather than as an additive to the fuel pellets. This would allow for the addition of IFBA in a non-nuclear-regulated environment, thereby obviating the additional handling and processing of the fuel pellets. This approach would also overcome the other drawbacks associated with the present method of IFBA addition. This could represent significant cost savings and potentially lead to greater reproducibility and control of the burnable fuel in the early stages of reactor operation.

Research Progress

This research was performed collaboratively among the University of Wisconsin, Sandia National Laboratories, and Westinghouse Corporation. The surface alloying for this research was performed using the ion beam surface treatment (IBEST) process developed at Sandia National Laboratories. IBEST involves the delivery of intense, energetic ion beam pulses onto the surface of a material, near-surface melting, and rapid solidification. The non-equilibrium nature of such processing allows for surface alloying in excess of the thermodynamically dictated solubility limits, an effect that is particularly relevant to this research due to the negligible solubility of boron and gadolinium in zirconium. The University of Wisconsin performed the near-surface materials characterization, assisted Sandia National Laboratories in process optimization, coordinated the overall program, and promoted educational activities. Westinghouse performed the steam/water autoclave tests on the surface-treated samples and conducted a detailed process manufacturability evaluation.

Two zirconium alloys used for fuel cladding, Zirlo and Zircaloy-4, were sputter deposited with either gadolinium or boron and subjected to the IBEST process. The thickness of the sputter-deposited films investigated in this study ranged from 0.5 μm to 1.5 μm. Researchers performed studies on the surface alloying of gadolinium by bombarding it with neon ions. Material characterization studies performed using scanning and transmission electron microscopy, X-ray diffraction, and Rutherford Back-scattered Spectroscopy showed that gadolinium could be successfully alloyed into the zirconium alloys as a single-phase solid solution, using neon ions in the energy range of 1-2 kJ/cm^2. At higher energies, ablation of the gadolinium film by either sputtering or evaporation outpaced the kinetics of melting and liquid phase diffusion necessary for alloying. Numerical

simulations supported these experimental observations. Although complete alloying was successfully achieved, the alloyed surface did not perform satisfactorily in high-temperature autoclave tests. For example, sample tests in steam autoclaves at 800°F showed that the weight gain due to oxidation nearly doubled for the Gd-alloyed sample compared to the control zirconium alloy sample after exposure for just one day.

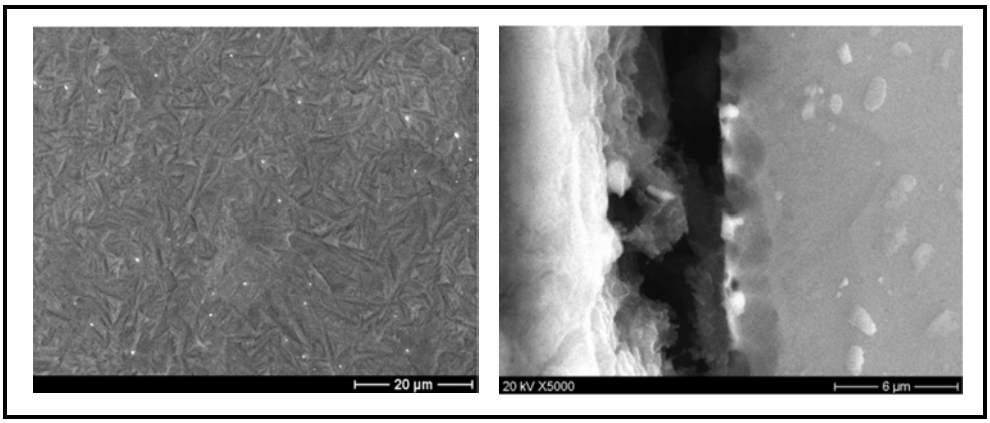


Figure 1. Surface of a zirconium alloy deposited with a 0.75µm film of boron and IBEST treated with an ion energy range of 4-6 kJ/cm², (a) surface view and (b) cross-sectional view where arrows indicate the boron alloyed layer.

Researchers believe that the studies conducted on boron surface alloying by the IBEST process were successful and have pointed to several useful avenues for future research. Unlike gadolinium, optimum boron alloying in zirconium alloys was achieved using nitrogen ions and at a higher energy range of 4-6 kJ/cm². This is because boron is much more resistant to ablation than gadolinium and requires more energetic ions to induce surface melting and mixing. Figure 1 shows the SEM images of the surface and cross-section of a zirconium alloy, which has been completely alloyed with an ion energy range of 4-6 kJ/cm².

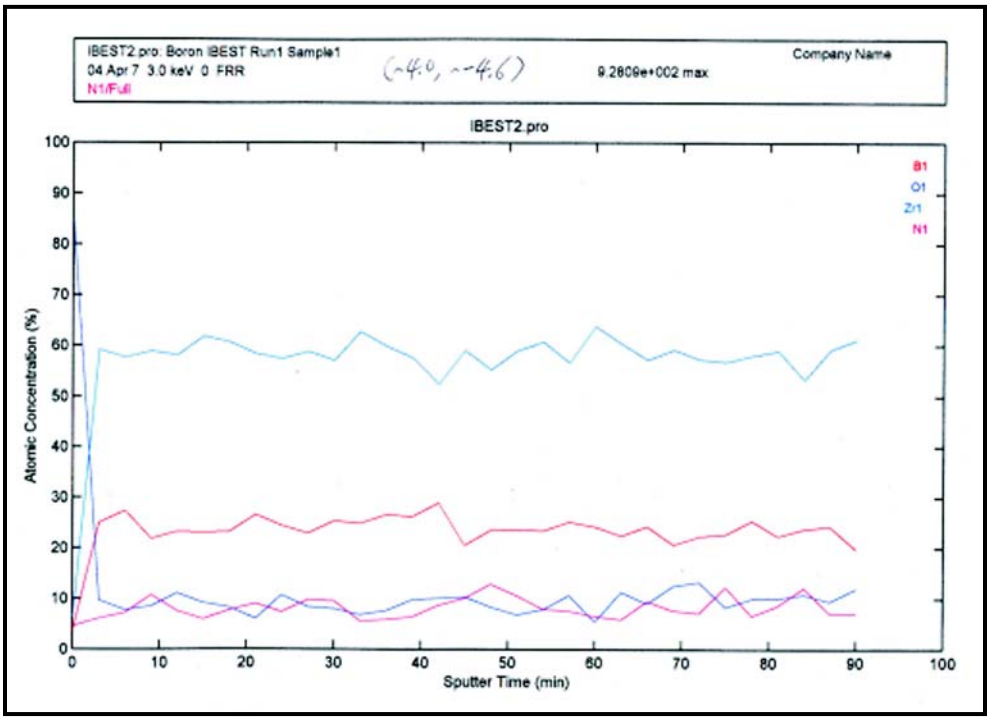


Figure 2. Auger composition vs. depth (sputter time) analysis showed uniform coexistence of boron and zirconium (at about 25 atomic percent boron).

Figure 2 shows the results of Auger Electron Spectroscopy (AES) of a 0.75 µm boron film that was alloyed into a zirconium alloy by the IBEST treatment. Sputtering for up to 5 hours in the AES instrument yielded a uniform boron composition of about 25 atomic percent. Laser profiling of the sputter crater showed that this sputtering time in AES corresponded to about 3.75 µm, while cross-sectional scanning electron microscopy shown in Figure 1 indicates the depth of the boron-alloyed layer to be about 4 µm.

Autoclave testing of the boron-alloyed samples was performed in steam autoclaves at Westinghouse at 800°F with weight gain and surface examinations made daily. Examination of the surface of the samples after 3 days of exposure showed no visible signs of corrosion (Figure 3) in the boron alloyed sample. The samples appeared similar to

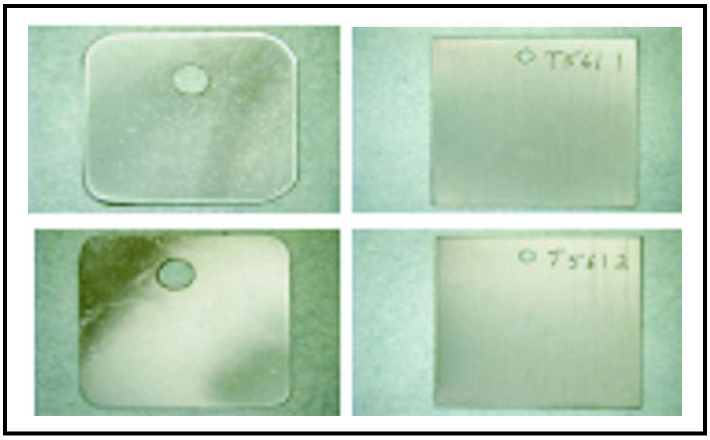


Figure 3. Surface of zirconium samples after autoclave testing at 800°F for 3 days (a) boron IBEST treated sample after sputter deposition with 0.75 µm boron layer and (b) control zirconium samples.

the zirconium alloy control samples. The cross-sectional examination of the boron IBEST-treated samples and the control samples after autoclave testing also showed that both samples had a similar build-up of the oxide corrosion product (Figure 4).

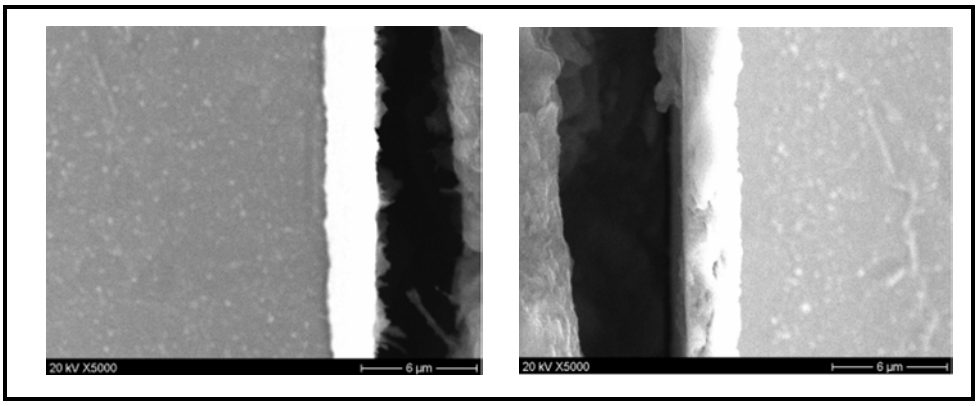


Figure 4. Cross-sectional SEM micrograph showing a nearly identical build-up of the oxide corrosion product on zirconium-alloy samples after autoclave testing for (a) control sample and (b) boron IBEST treated samples after sputter deposition with 0.75 µm boron layer.

Figure 5 shows a numerical simulation of melt thickness and duration for a 1 µm boron film on zirconium alloy after IBEST bombardment with 6 kJ/cm^2 nitrogen beam. The simulation showed that the entire 1 µm boron film and 2 µm of underlying zirconium will melt over a duration of approximately 4,000 nanoseconds.

With regard to alloying and oxidation performance, similar successful trends as those observed for 0.75 µm thick boron film were also demonstrated for an IBEST-treated 1 µm boron sputtered film. Detailed cost analysis performed by Westinghouse based on operations at their commercial plants showed that with this type of surface modification, the cost per cladding tube would be about $34 compared to the $11 present cost for an untreated cladding tube.

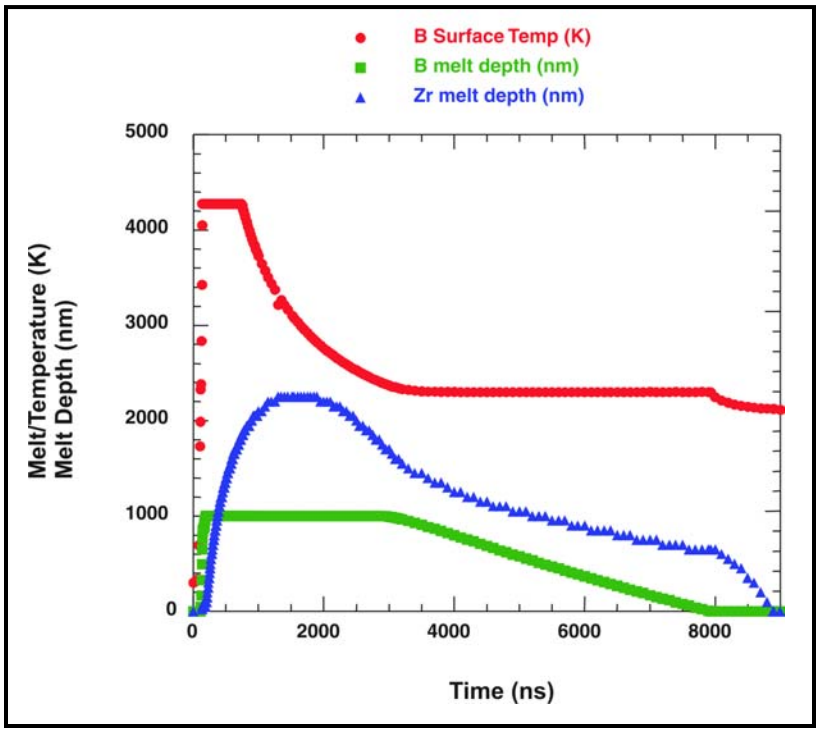


Figure 5. Numerical simulation of melting, liquid diffusion, and rapid solidification for a 1 µm boron film sputter deposited on zirconium alloy.

Planned Activities:

This NERI project has been completed.

NUCLEAR ENERGY RESEARCH INITIATIVE

Neutron and Beta/Gamma Radiolysis of Supercritical Water

PI: David M. Bartels, Argonne National Laboratory

Collaborators: University of Wisconsin, Notre Dame Radiation Laboratory

Project Number: 02-060

Project Start Date: August 2002

Project End Date: September 2005

Research Objectives

Current pressurized water reactors run at roughly 300°C and 100 atmospheres pressure. Advanced water-cooled designs under consideration in the Generation IV Initiative would operate at 500°C and 250 atmospheres, which is well beyond the critical point of water. This would improve the thermodynamic efficiency by about 30 percent, and provide considerable capital cost savings due to plant simplification. However, two questions regarding the effects of high temperatures and pressures remain unanswered:

- What changes occur in the radiation-induced chemistry of water as the temperature and pressure are raised beyond the critical point?
- How will this affect the limiting corrosion processes in the materials of the primary cooling loop?

The cooling water in a water-cooled reactor undergoes radiolytic decomposition induced by gamma, fast-electron, and neutron radiation in the reactor cores. Unless mitigating steps are taken, oxidizing species produced by the coolant radiolysis can promote intergranular stress-corrosion cracking and irradiation-assisted stress-corrosion cracking of iron-, nickel-, and zirconium-based alloys. These mechanisms will alter the corrosion rates of these alloys in reactors. One commonly used remedial measure to limit corrosion by oxidizing species is to add hydrogen in sufficient quantity to chemically reduce the transient radiolytic primary oxidizing species (OH, H_2O_2, HO_2/O_2^-), thereby preventing the formation of oxidizing products (H_2O_2 and O_2). It is still unclear to researchers whether this will be effective at the higher temperatures proposed for future reactors. While an earlier NERI project investigated some of the most important radiation chemistry in supercritical water (SCW), there is no information on the effect of neutron radiolysis, which is the main source of the troublesome oxidizing species.

The objective of this project is to discover most of the fundamental information necessary to develop a predictive model of radiation-induced chemistry in a supercritical water reactor core. Electron pulse radiolysis coupled with transient absorption spectroscopy is the method of choice for measuring kinetics of radiation-induced species, and also product yields for fast electron and gamma radiation. Measurements of second order free radical reaction rates are being carried out in high-temperature water using the Argonne Chemistry Division electron linear accelerator. The University of Wisconsin Nuclear Reactor Facility is a very convenient source of neutron radiation that can be exploited for radiolysis experiments at temperatures up to 500°C. The combined capabilities will make it possible to create a quantitative model for water radiolysis in both current PWR systems and supercritical water cooled plants in the future.

Research Progress

The beginning of this year marked the start of experiments using the flow loop at the University of Wisconsin Reactor. One of the first experiments, shown in Figure 1, yielded the most qualitatively interesting and important results so far obtained. In this experiment, N_2-sparged water was heated to a high temperature, with pressure held constant at 250 bar, and flowed through the irradiation zone for approximately 20-30 seconds. The water was then cooled and analyzed for dissolved gases using a mass spectrometer. Hydrogen (H_2) was the main product detected in this experiment. The figure shows that the yield of hydrogen gas remains relatively constant up to about 300°C, then increases substantially at higher temperature up to 450°C. Researchers find this result to be remarkable because the density of the water in the irradiation zone decreased over the whole range of temperatures, especially near the critical region (350–400°C). Because the energy

absorbed is proportional to density, the actual energy-normalized yield of H_2 from radiolysis increased by over 20 times in the transition from PWR conditions (near 300°C) to SCWR conditions (450°C). Researchers expected to measure a stoichiometric amount of oxygen (O_2) in this experiment as well, but apparently oxygen is largely consumed by corrosion reactions on the tubing walls before it exits the reactor. Nevertheless, the yield of oxidizing peroxide and oxygen must increase 20 times as well. The major question is whether this net radiolysis of water can be controlled in supercritical reactors in the same manner as PWRs, by adding a small amount of H_2 at the inlet. Experiments are underway to test this idea.

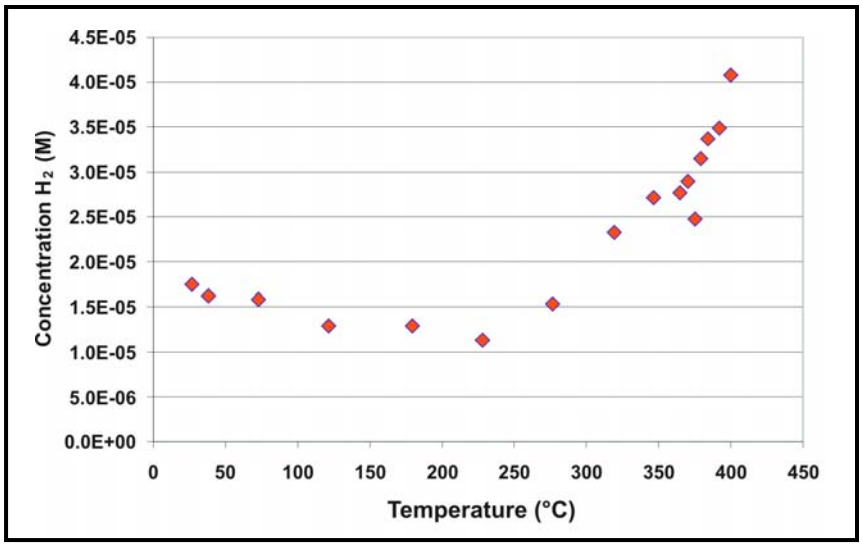


Figure 1. The concentration of H_2 measured in the N_2 saturated solution as a function of temperature at 250 bar pressure.

Experiments carried out at Argonne during this year concentrated on measuring key second order recombination reactions so that researchers can construct a comprehensive model of the chemistry. The measurement of OH radical second order recombination rates, shown in Figure 2, represents an experimental tour-de-force for these optical transient absorption measurements. The absorption of OH radical at 250 nm is weak, and deep UV lamps used for analyzing lights are also weak. Measurements require a large amount of signal averaging, and the conditions must be kept stable for long periods. The sapphire windows also display a dose-dependent absorption of the same magnitude; thus, for every experiment, researchers must empty the cell and measure the background absorption. The spectrum also changes as a function of temperature, so a new extinction coefficient must be measured under each condition. Ultimately, researchers concluded that the OH radical recombination rate barely changes between 200 and 375°C. But in the supercritical regime, where the density change is large, the rate constant decreases markedly; it becomes too slow, and the absorption is too weak to measure above 400°C.

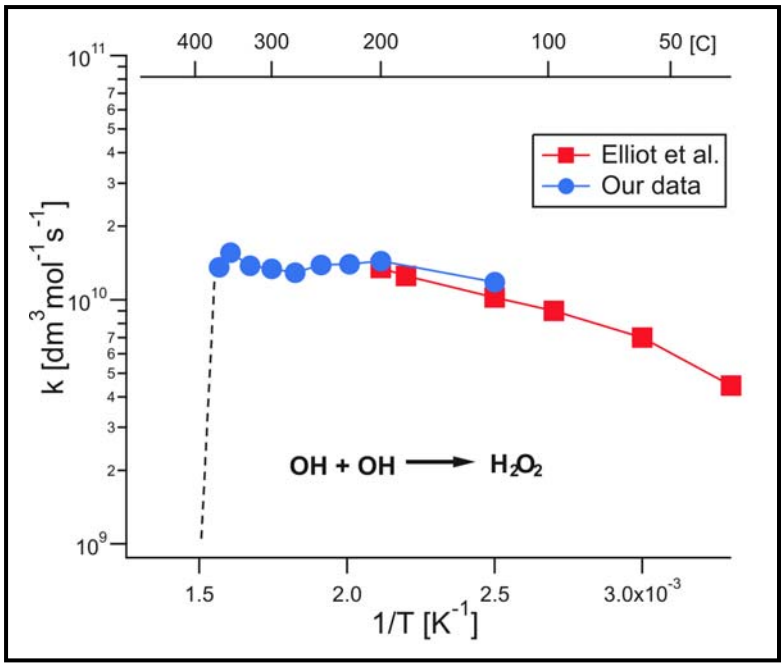


Figure 2. Arrhenius plot for the second order recombination of OH radicals at 250 bar pressure.

Planned Activities

Researchers will continue experiments at Wisconsin's reactor flow loop, measuring yields with good precision over a wide temperature range. They will continue to make modifications of the shielding to optimize the neutron/gamma ratio to make these results transferable. Notre Dame Radiation Laboratory will carry out complementary studies using just beta/gamma radiolysis. In both locations, researchers will test the suppression of net radiolysis by addition of H_2.

Studies at Argonne will continue to focus on the very difficult measurement of second order free radical recombination rates. In addition, researchers will measure the very critical reaction rate of H_2 with OH radicals in supercritical water. A new, gold-coated cell will be constructed to overcome corrosion problems. An additional experimental capability will be constructed at Notre Dame Radiation Laboratory to detect hydrogen atom reactions by pulsed EPR.

NUCLEAR ENERGY RESEARCH INITIATIVE

Innovative Approach to Establish Root Causes for Cracking in Aggressive Reactor Environments

PI: Stephen M. Bruemmer, Pacific Northwest National Laboratory (PNNL)

Collaborators: General Electric Global Research Center

Project Number: 02-075

Project Start Date: September 2002

Project End Date: September 2005

Research Objectives

This research project focuses on characterizing critical iron and nickel-based stainless steel alloys by testing them under well-controlled conditions to minimize in-service complexities. Researchers will use quantitative assessment of crack-growth rates to isolate the effects of key variables and high-resolution analytical transmission electron microscopy to provide mechanistic insights into crack-tip corrosion/oxidation reactions and crack-tip structures at near-atomic dimensions. Reactions at buried interfaces that are not accessible by conventional approaches are being systematically interrogated for the first time. Novel mechanistic "fingerprinting" of crack-tip structures tied to thermodynamic and kinetic modeling of crack-tip processes will be used to isolate causes of environmental cracking. Comparisons will be made with failed components removed from light water reactor (LWR) service (funded separately by industry collaborators).

This research strategy capitalizes on unique national laboratory, industry, and university capabilities to generate basic materials and corrosion science results with immediate impact on next generation nuclear power systems. Project activities will be integrated with existing NERI projects and with U.S. and international projects dealing with current LWR degradation issues. This leveraged approach will facilitate the revolutionary advances envisioned in NERI by creating a multi-faceted effort combining the basic and applied science necessary to drive mechanistic understanding and promote development of next generation materials that meet advanced reactor performance goals.

Research Progress

Single-Variable Stress Corrosion Cracking (SCC) Experimentation. Conducting experimental research under well-controlled material, environmental, and stress conditions is an essential first step towards elucidating environmental cracking mechanisms. The selection of these conditions is critical to producing degradation where crack initiation and advance processes are best understood. Material, environmental, and electrochemical conditions are being controlled to promote degradation by slip dissolution, hydrogen-induced cracking, or internal oxidation. This enables researchers to produce unique samples with known cracking mechanisms for crack-tip characterizations in the subsequent task. A key limitation in extracting useful information from failed service components of operating reactors is that material and environmental conditions are uncontrolled. As a result, off-normal conditions often contribute to the cracking process and complicate interpretation of basic environmental degradation mechanisms.

Researchers completed SCC experimentation in several areas, including crack-growth rate measurements on specially tailored stainless steels and nickel-base alloys in high-temperature water environments. They observed a remarkable increase in crack-growth rate (exceeding a factor of 10) in high silicon stainless steels, particularly in low-potential, hydrogenated water environments. The influence of grain boundary carbides on intergranular SCC was also isolated, revealing a decrease in crack-growth rates by approximately five times below that for precipitate-free boundaries. These tests continue to provide unique samples for crack-tip characterizations that complemented contributions from other collaborators who supplied cracked service components from operating commercial nuclear power plants.

Crack-Tip Characterization. The goal of this task is to generate a library of high-resolution, crack-tip "fingerprints" of different known mechanisms of environmental cracking. These fingerprints can then be used to verify model predictions and to compare with in-service cracking. Analytical transmission electron microscopy (ATEM) examination of crack tips has proved particularly effective in discriminating different mechanisms of degradation in Ni-base alloy steam-generator tubes and stainless steel components removed from high-temperature water service.

Crack and crack-tip corrosion structures were documented at near-atomic dimensions for environment-induced cracks in stainless steel crack-growth samples, alloy 182 weldments from PWR service, and alloy 600 and corrosion-resistant alloy 690 in laboratory lead-doped solutions. For the first time, crack-tip characteristics were established under well-controlled laboratory test conditions. The crack tip shown in Figure 1 was produced by constant-stress-intensity crack-growth tests in high-purity, high-temperature water and was backfilled with epoxy to protect cracks under load and maintain the dynamic crack-tip openings. Contrary to predictions, crack openings are found to be a few nanometers and establish stable oxide films on the crack walls. Although the basic crack propagation is intergranular, transgranular attack is observed at matrix deformation bands. Work was also initiated on additional alloy 182 welds from PWR outlet nozzles. Highlights include orientation-imaging microscopy discovering cracking along low sigma grain boundaries and ATEM crack-tip exams linking dissolution of sulfide inclusions to degradation. Researchers also continued investigating Pb-induced cracking in Ni-base alloys. Unique results were produced that elucidate transgranular SCC and the complex, precursor, deformation-induced dislocation structures, as shown in Figure 2.

Modeling Crack-Tip Corrosion Reactions. The final key area of research during the second year was modeling the crack-tip corrosion reactions. Predictions of solution thermodynamics and corrosion-product phase stabilities focused on high-temperature water system with Pb as an impurity. Important aspects of Pb migration down cracks and enrichment at crack tips were explored. Kinetic calculations for transport of Pb to crack tips were shown to account for measured Pb enrichments greater than 10 wt% only for bulk solution concentrations of less than 0.1 ppm, narrow crack openings (approximately 10-nm wide), and expected service SCC crack-growth rates.

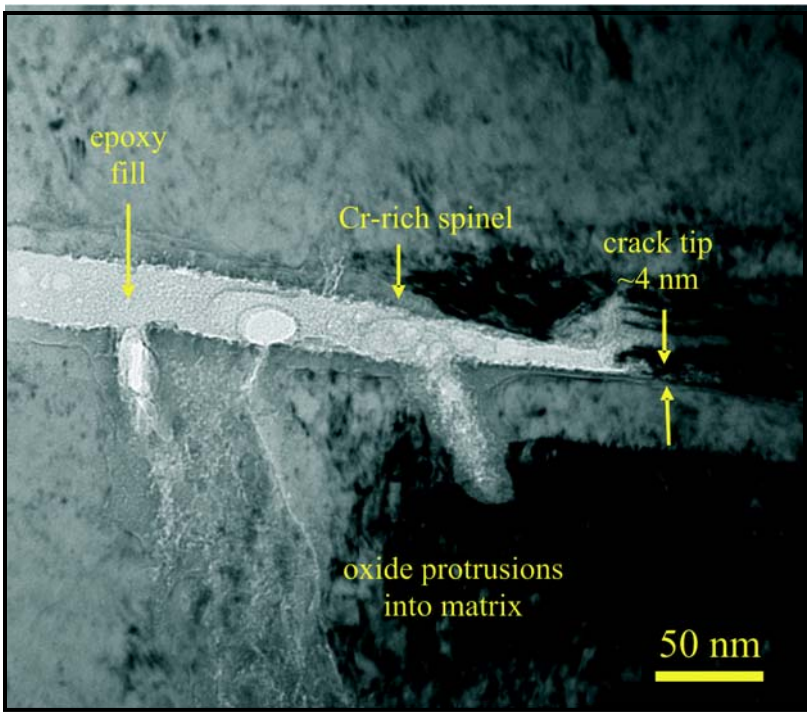


Figure 1. Crack tip in a 304SS crack-growth sample after test in oxidizing water at 288°C. Intergranular cracks decrease in width from >100 nm to ~4 nm at the tip.

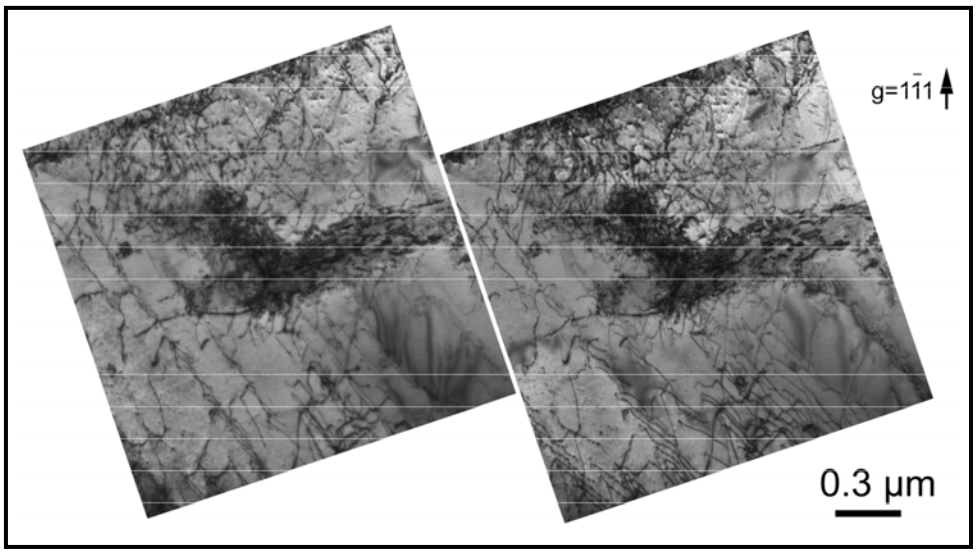


Figure 2. TEM brightfield stereo pair illustrating the crack-tip dislocation structure in an alloy 690 sample after exposure to Pb-doped caustic environment.

Planned Activities

Single-Variable SCC Experimentation. Crack-growth SCC tests will continue under well-controlled material, environmental, and stress conditions. Researchers will isolate material issues including bulk composition, grain boundary structure, composition, and matrix strength on cracking response, and environmental issues such as solution composition and electrochemical potential. Clear examples of cracking will be produced where the mechanism of crack growth is well established, i.e., slip-oxidation, hydrogen-induced cracking, and internal oxidation. Much of this work in the final year will be on advanced replacement materials such as alloy 690.

Crack-Tip Characterization. Through high-resolution ATEM characterizations, researchers will continue to generate a library of crack-tip "fingerprints" of different known mechanisms of environmental cracking for model verification and comparisons with service failures. Researchers will use well-controlled crack-growth samples to establish SCC crack-tip and crack-wall signatures in high-temperature water environments. They will investigate the structure, composition, and kinetics of crack-wall film growth. Also, they will complete their research on solution impurity effects on crack advance and corrosion-product characteristics by using static test samples that focus on isolating the influence of sulfate and lead. Research on hot cracking and SCC in nickel-base alloy weldments will expand with the intent to document processes controlling cracking mechanisms in current and advanced weld metals.

Modeling Crack-Tip Corrosion Reactions. Mechanistic modeling continues, relating measured SCC response and crack-tip corrosion products to predictions using activity, solubility, and kinetics of corroding species to rationalize stable and metastable phases observed at crack tips. Researchers will pursue three modeling subtasks: (1) thermodynamically favored phases, (2) kinetically allowed phases, and (3) SCC crack growth.

NUCLEAR ENERGY RESEARCH INITIATIVE

Design of Radiation-Tolerant Structural Alloys for Generation IV Nuclear Energy Systems

PI: Todd Allen, University of Wisconsin-Madison

Collaborators: Argonne National Laboratory, University of Michigan, Pacific Northwest National Laboratory, Japan Nuclear Cycle Development Institute

Project Number: 02-110

Project Start Date: September 2002

Project End Date: September 2005

Research Objectives

This project focuses on improving the radiation tolerance of both austenitic and ferritic-martensitic (F-M) alloys projected for use in Generation IV reactor systems. The expected materials limitations of Generation IV components include higher temperature creep strength, dimensional stability, and corrosion/stress corrosion compatibility. The research focuses on three main concepts for achieving increased radiation tolerance in these alloys: 1) grain boundary engineering, 2) the addition of oversized solutes to the matrix, and 3) microstructural/nanostructural control by matrix alteration. Each approach is aimed at mitigating specific irradiation effects. Grain boundary engineering will increase the fraction of special boundaries, reducing radiation-induced segregation (RIS) to those boundaries and strengthening grain boundaries against creep from sliding and deformation. Oversized solute additions will promote recombination, reducing RIS and altering the dislocation microstructure. Matrix precipitate additions provide a high density of interfaces to promote recombination and trap helium, while also strengthening the matrix against high-temperature creep.

Research Progress

Grain Boundary Engineering. Grain boundary engineering involves a series of thermo-mechanical treatments designed to convert a fraction of the high-energy boundaries to low-energy boundaries (identified as Coincident Site Lattice [CSL] boundaries), thus reducing cracking susceptibility and improving creep strength. This project developed the first treatment to enhance the fraction of CSL boundaries in an advanced, ferritic-martensitic steel—T91. The challenge was to enhance the grain boundaries without disturbing the original microstructure. Since the microstructure is critical to achieving high-temperature properties, the heat treatment process requires strict control. In the first year of this project, a treatment was developed in which the CSL boundary fraction of T91 was enhanced over the as-received case without changing other critical features of the microstructure, such as grain size, carbide size and location, density, and hardness. Improved creep strength should result from the preservation of all the microstructural features decisive for creep strength in T91 and from increasing the fraction of low angle boundaries. In the second year of the project, creep testing of the CSL-enhanced alloys was initiated. These samples had a creep rate approximately one-third that of untreated samples at 500°C and 225 MPa. In addition to T91, CSL enhancement techniques were developed for alloy 800H, a higher temperature alloy seen as a candidate for high-temperature, gas-cooled reactors. Researchers will continue creep testing of CSL-enhanced 800H in the last year of this project.

Oversized Solute Additions. The second task was to continue studying the ability of oversized solute additions to reduce radiation damage. The addition of oversized solutes is expected to restrict the development of deleterious microstructural features through interaction of the oversized solutes and radiation-induced point defects. Researchers irradiated hafnium- and zirconium-containing alloys with protons, nickel ions, and reactor neutrons. The reactor-irradiated samples were exposed in the Fast Flux Test Facility (FFTF) at DOE's Pacific Northwest National Laboratory and the BOR-60 reactor in Russia. For hafnium-containing alloys irradiated with high-energy nickel ions, the addition of hafnium reduced or delayed the formation of voids significantly, compared to alloys without the additions. Using a step-cooling heat treatment to take the hafnium out of solution and into precipitates reduced the effectiveness of hafnium in preventing void swelling. A similar step-cooling heat treatment to a 316 alloy without

hafnium produced the smallest swelling. Figure 1 shows a comparison of void swelling behavior for various base materials.

Although adding zirconium to a high-purity stainless-steel alloy reduced swelling at low dose, the swelling reduction did not persist after high-dose irradiation in the FFTF. Changes to grain boundary and void surface composition due to radiation-induced segregation were measured in the Zr-loaded alloy and found to be significantly larger than observed in alloys without Zr. A similar effect was previously noted for Hf-loaded alloys. Initial modeling efforts have not yet isolated a reason for the larger than expected segregation; researchers will continue to focus on this in Year 3 of the project. To better understand the effect of oversized alloys on grain boundary segregation, researchers developed a model that includes the trapping of point defects and will continue to refine it against the expanding database.

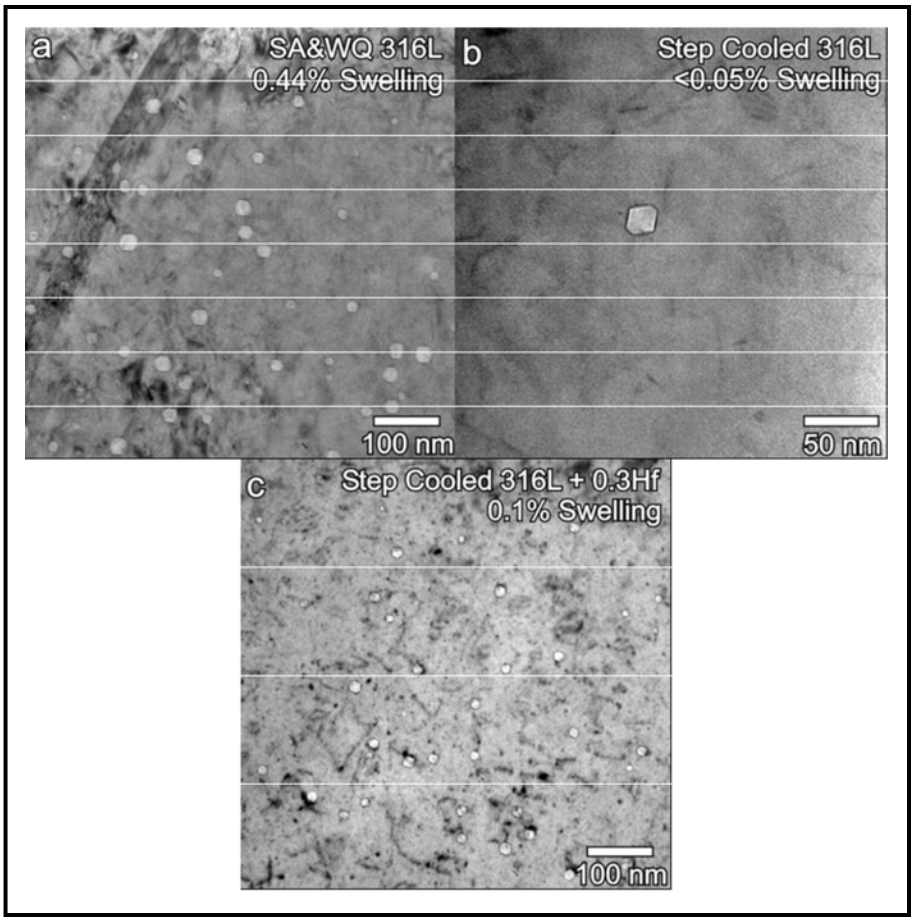


Figure 1. Void swelling behavior at 20 dpa is compared among (a) solution-annealed 316L base material, (b) step-cooled 316L base material, and (c) step-cooled 316L + 1.2Hf. The step-cooled 316L base material has the lowest swelling of the three conditions shown.

Radiation Stability Following Matrix Alteration. The final major task was to examine the radiation stability of advanced alloys. Proton and heavy ion irradiation are being performed to evaluate the radiation response of Alloy 800H, HCM12A, and a 9Cr oxide dispersion strengthened (ODS) martensitic steel. HCM12A is a nominal 12Cr steel developed for higher temperature application than HT9, the fast reactor alloy. Prior to this project, limited information was available on the response of this alloy to radiation. HCM12A was irradiated with 3.2 MeV protons at 400°C to a dose of 10 dpa and with 5 MeV nickel ions at 500°C to 50 dpa. The hardness of HCM12A increases with dose, saturating after approximately 5 dpa, with an approximate 70 percent increase of hardness. Using hardness-to-yield strength correlations, this corresponds roughly to a 520-550 MPa yield strength increase. Dislocation loops and voids do not form in significant quantities under these irradiation conditions, so the increase in hardness appears to be attributable to precipitate formation. Chromium is enriched at boundaries and iron is depleted in the unirradiated state. The higher chromium concentration at the boundaries is likely due to chromium carbides. Irradiation causes a further increase in chromium concentration and a decrease in iron concentration at the boundaries. Researchers believe this could be due to coarsening of existing carbide particles, nucleation of new carbide particles, or radiation-induced segregation of chromium to the boundary away from carbides. Nothing in these first irradiations indicated adverse properties in this alloy as compared to HT9.

Planned Activities

In Year 3 of this project, the grain boundary engineering task will focus on creep testing of the CSL-enhanced T91 and 800H alloys to verify that the CSL enhancements did improve the creep resistance. Radiation testing of CSL-enhanced T91 will take place in the final year. Researchers will perform additional irradiation, examination, and modeling of the microstructural development of alloys with oversized solute additions. They will perform studies of oxide stability in 9Cr ODS at 600°C and 700°C and will examine the microstructure of HCM12A irradiated with protons at 500°C to get data on the effect of temperature on microstructural development under radiation.

NUCLEAR ENERGY RESEARCH INITIATIVE

Enhanced Control of PWR Primary Coolant Water Chemistry Using Selective Separation Systems for Recovery and Recycle of Enriched Boric Acid

PI: Ken Czerwinski, University of Nevada-Las Vegas (UNLV)

Collaborators: University of California-Berkeley, Los Alamos National Laboratory, Pacific Southern Electric & Gas Company, Florida Power & Light, (n, p) Energy, Inc.

Project Number: 02-146

Project Start Date: October 2002

Project End Date: September 2005

Research Objectives

The fundamental science of operating existing and advanced pressurized water reactors (PWRs) shows that increasing nuclear fuel enrichment will produce more energy. However, operating within the nuclear reactor safety requirements at higher enrichments requires an increased concentration of natural boric acid used as a chemical flux shim. The key objective of this research project was to develop systems for efficiently and selectively recovering special purpose isotopes that are added to the primary coolant water in light water reactors (LWRs) for neutron flux control and water chemistry. Typically, ^{10}B-enriched boric acid, $B(OH)_3$, is favored over natural boron for neutron flux control because it allows the use of a lower concentration of lithium hydroxide (LiOH) for operational pH control. This helps to decrease the extent of corrosion cracking resulting from the LiOH. However, the expense of producing and using enriched isotopes such as ^{10}B and ^{7}Li requires a cost-effective means to recover and reuse them.

The goal of this project is to develop and field test polymeric sequestering systems designed to efficiently and selectively recover enriched boric acid/lithium hydroxide from the primary coolant water of LWRs. These advanced separation materials will reduce the cost of operating existing and advanced LWR systems by improving the chemical control of the primary reactor coolant. Researchers on this project characterized contaminants present in the coolant system according to their potential for interfering with the selective recovery of ^{10}B and ^{7}Li, in an effort to assess the impact of actinides and activation products on target isotope separation. They developed counter measures to mitigate the interference of non-target radionuclides. Cost benefits will result from greater energy production per reactor unit, reduced operational radiation exposure, and protection from accelerated corrosion of critical core components.

Research Progress

UC Berkeley (UCB), Department of Nuclear Engineering, and UNLV performed work on polymer extraction modeling. They introduced ion pairing to backbone cation sites into the model based on limitations of using a solely covalent approach. A large discrepancy was found for low boron concentrations due to the focus of ion pairing in the model (Figure 1). Researchers added a model correction to account for pH variation in the microenvironment near the polymer, correcting for the observed differences. At this point, computer modeling of the boron/water/polymer system is essentially complete. The model, where polyborate species pair to cation sites by order of charge-to-volume ratio, is sufficiently complex to demonstrate a significant contribution to the binding behavior from ion pairing of polyborate anions to cationic amine sites on the polymer. The model can incorporate additional input such as ionic strength.

Researchers investigated the solid state and solution behavior of simple borate salt ligand complexes as well as the synthesis of terephthalamide borate ester complexes. They used potentiometric and spectrophotometric titrations and ^{11}B NMR spectroscopy to study the structure, solution thermodynamics, and kinetics of borate complexes. These properties are currently being investigated at various conditions in order to find an optimized ligand system. Boric acid readily forms what are basically covalent bonds

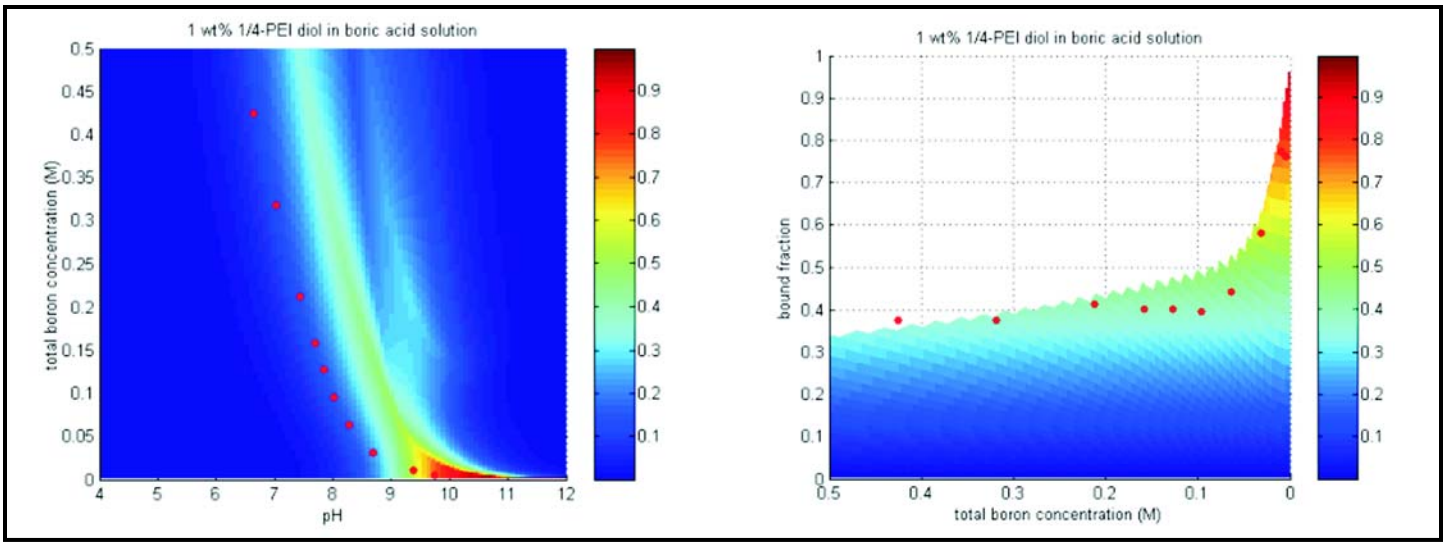


Figure 1. Results using Simple Ion Pairing Model: This model describes most of the behavior of the system. Researchers calculated the surface. The red dots represent experimental data. The color axis represents the fraction of total boron bound by the polymer.

with alcohol functionalities through the displacement of the boron hydroxyl group. Based on the equilibrium constants reported, reagents that have two sets of phenolic hydroxyl groups generally form much more stable borate complexes than those of alcohols. The spectroscopic results confirm that boric acid/borate combines with polyhydroxyl compounds to form complexes with 1:1 and 1:2 stoichiometries (Figure 2). The UCB-Nuclear Engineering team used this information in their modeling efforts.

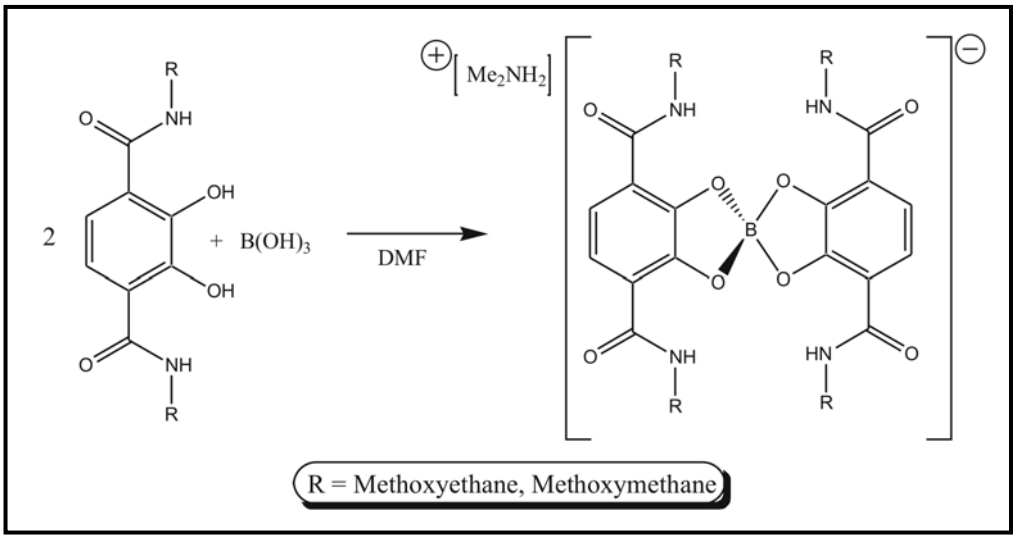


Figure 2. 1:2 Boron to TAM ligand complexes.

Based on previous reactor studies, the Los Alamos National Laboratory (LANL) team is developing ligands to avoid the co-extraction of radionuclides with the boron and is supporting the power plant studies of (n,p) Energy, Inc., by supplying them with polymers for testing and helping with protocol and procedures. They prepared functionalized polymers from 25 percent to 100 percent and characterized them for structure, binding, and selectivity. Some unique boron-binding results occurred that are not readily explainable, such as the possibility of neutral trigonal boric acid esters binding in addition to tetrahedral borate esters. Researchers used ^{11}B NMR to further examine ester formation and found the 1,2,3-triol-PEI has larger peak areas for the borate esters than

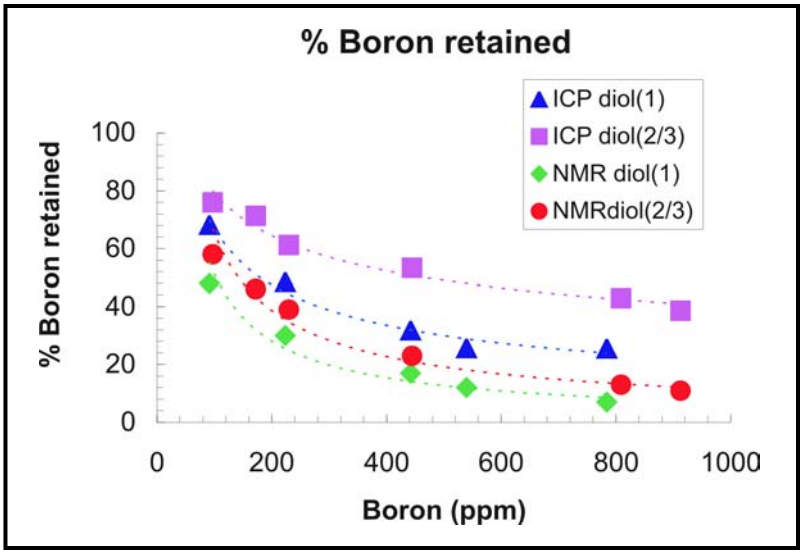


Figure 3. Percentage of total boron retained as measured by extraction experiments and as measured by ^{11}B NMR. Extraction data: diol fully functionalized (▲), diol 2/3 functionalized (■). ^{11}B NMR data: diol fully functionalized (◆), diol 2/3 functionalized (●).

the 1,2-diol-PEI, indicating that overall 1,2,3-triol is a better boron chelator (Figure 3). In the most recent power plant studies, researchers observed that antimony (^{125}Sb) and cobalt (Co) isotopes could cause severe interference with the boron recovery process. Preparation of an Sb-specific ligand is under development, which will selectively remove Sb and Co from the reactant coolant so that the recovered enriched boric acid is not radioactively contaminated and can be readily recycled.

The (n,p) Energy team traveled to Florida Power and Light's (FPL) Turkey Point plant and requested the use of their site for field testing. FPL gave preliminary approval, but requested a description of tests, support required from FPL, and a test plan.

Planned Activities

The following work is planned for the final phase of the project:

- To further develop the reactor contamination model, use experimental data to evaluate species interference with B and Li, develop methods to remove interferences, and test the methods in commercial reactors.
- To look at the extraction of boric acid and Li with UF membranes and focus on temperature effects.
- To evaluate the modeling results with data from commercial reactors to assess the utility of the extraction polymers for treating reactor coolant water.

9. Generation IV Nuclear Energy Systems Initiative: Project Abstracts

This new program element includes 13 research projects, all of which were awarded in FY 2005. The following abstracts present each project's participants, its objectives, and a brief description of the proposed work scope.

The Generation IV Nuclear Energy Systems Initiative (Generation IV) is focused on developing new reactor systems to be deployed during the next 20 years. Planned NERI research efforts for FY 2005–FY 2007 related to this initiative will focus on the following technologies and research areas:

- Next Generation Nuclear Plant (NGNP): The major aim of NGNP projects is to build and demonstrate advanced high-temperature reactor technology that is able to produce both hydrogen and electricity at an economic rate. The NGNP is in the first phase of project development and will lead to the actual design, construction, and future operation of an Advanced Nuclear Reactor-Hydrogen Cogeneration Demonstration unit. New research projects will focus on the validation of reactor physics and core design analyses tools, development and validation of reactor thermal-hydraulic and mechanical design analyses tools, materials research, power-conversion unit assessments, and safety and risk analysis. The scope of these projects also includes project design, system design and analysis methodology, and fuel development and qualification. The longer-term NGNP research objectives are: (1) to demonstrate a full-scale unit in the 2015 timeframe; (2) to demonstrate the Brayton Cycle for the full-scale unit; (3) to test the exceptional safety capabilities of advanced very-high-temperature reactors; (3) to obtain a Combined Operating License (COL) for the NGNP using the NRC's proposed performance-based, risk-informed licensing process (i.e., 10 CFR Part 53, now in development); and (4) to support the development of other technologies involving hydrogen applications.
- Supercritical Water-Cooled Reactor: Projects associated with this technology will concentrate on showing the technical feasibility of a light water reactor operating above the critical pressure of water, thus producing low-cost electricity. The focus will be on five main areas: the design of safety systems and containment to handle abnormal events; the evaluation of dynamic power/flow instabilities; corrosion and stress-corrosion cracking testing of materials for the core and vessel internals; the investigation of basic thermal phenomena for the reactor; and core neutronic, mechanical, and thermal design.
- Lead-Alloy Liquid-Metal-Cooled Fast Reactor (LFR): The objective of these projects is to produce a small nuclear energy system suitable for deploying in remote locations and in developing countries. Research and development efforts are aimed at defining and selecting the reference system, preparing a defendable safety case for the system, and licensing, perhaps through testing a demonstration reactor system. Research scope encompasses several milestone goals, such as concept development and completing a paper discussing the implementation of these concepts. Specifically, plans include establishing a design for ensuing concept development and issuing economic requirements and proliferation-resistance principles, creating a design and data requirements document, revising the safety and licensing approach, establishing a fuel qualification strategy and resolving materials and corrosion issues, submitting a concept paper on implementation of fuel cycle centers, and ascertaining the achievability of autonomous load following concepts.
- Gas-Cooled Fast Reactor (GFR): The objective of these projects is to develop a safe and sustainable GFR reactor that has a closed fuel cycle, is highly efficient (the Brayton power conversion cycle), and is capable of producing electrical power and/or hydrogen. Research for this reactor concept includes defining design features (fuel, coolant, unit power), designing safety systems for decay heat removal, identifying/testing fuel and core materials capable of high-temperature operation including developing fuels with high fission product confinement and reasonable burnup and fluence, and developing fabrication techniques. FY 2003 accomplishments included identifying possible passive/semi-passive safety systems, initially testing CO_2 compatible materials, and identifying candidate high-temperature fuels. Research will involve material testing and defining the physical properties of these materials, fuel performance modeling, preparation for fuel irradiation,

ion and neutron irradiation of fuel matrix materials, thermal-hydraulic and physics analysis, and optimization of candidate safety systems.

- Design and Evaluation Methods: Analytical methods, modeling techniques, computer codes, and databases must be developed for Generation IV plants. In addition, methodologies must be developed to evaluate system performance against Generation IV goals. Examples include evaluating the economic feasibility of these plants and ensuring proliferation resistance (PR) and physical protection (PP) of the public. Major activities for FY 2005–2007 include improving the NGNP analytical capabilities and validate these capabilities, defining fast reactor modeling requirements for the GFR and LFR designs, and improving the Generation IV evaluation methodologies and selecting preferred options (e.g., preferred methods for evaluating PR and PP measures; improving economic models for comparing economies of scale and non-power applications). Other activities include (1) identifying phenomena and parameters to be included in thermal/hydraulic and safety analysis codes and (2) improving Monte Carlo and deterministic methods for neutronic, fuel depletion, and material damage analyses.
- Materials: Projects in this area include the selection, development, and qualification of structural materials necessary to design and construct reactors within the Generation IV Program and the coordination with similar research being conducted for the Advanced Fuel Cycle Initiative and the Nuclear Hydrogen Initiative. Research plans include both material crosscutting and reactor-specific material ambitions. Material requirements for the NGNP, GFR, SCWR, and LFR reactor systems, as well as for their energy conversion systems, are addressed under this research area. Specific cross-cutting activities applicable to all designs will be performed including (1) designing a test facility, then initiating low flux and high flux, high-temperature irradiations; (2) establishing a database of candidate materials for high temperature and radiation service, then performing studies to identify mechanical properties of these materials; (3) preparing documents for alloys 316FR and 617 to initiate ASME codification; (4) performing microstructural analysis and modeling for materials of interest; and (5) developing a high-temperature design methodology for materials/applications of interest.
- Energy Conversion: Projects in this area will focus on both the supercritical carbon dioxide Brayton cycle and the high-temperature helium Brayton cycle. Specifically, objectives associated with the supercritical carbon dioxide Brayton cycle include completing system design and economic assessment; performing an experiment to validate key features; and engineering seals, materials, heat exchangers, and ducting. A demonstration experiment and simulation model are also planned to determine plant characteristics, performance, and the supercritical cycle dynamic response. For the high-temperature helium Brayton cycle, project goals include the engineering analysis of inter-stage heating (IH) and cooling (IC) configurations, design analysis of heat exchangers and turbo-machinery, and planning of a small-scale demonstration experiment.

NUCLEAR ENERGY RESEARCH INITIATIVE

Directory of Generation IV Nuclear Energy Systems Initiative Project Abstracts

NUCLEAR ENERGY RESEARCH INITIATIVE

In-Situ X-ray Spectroscopic Studies of the Fundamental Chemistry of Pb and Pb-Bi Corrosion Processes at High Temperatures: Development and Assessment of Composite Corrosion-Resistant Materials

PI: Carlo U. Segre, Illinois Institute of Technology

Collaborators: None

Project Number: 05-028

Project Start Date: March 2005

Project End Date: February 2008

Project Description

This project is in support of the development of fast reactor designs for transmutation and small, proliferation-resistant power reactors using lead (Pb) and lead-bismuth eutectic (LBE) liquid metal coolants. Use of these reactors for direct production of hydrogen becomes possible if temperatures of 800°C can be achieved, but current experience is limited to 550°C due to accelerated corrosion and embrittlement observed at higher temperatures.

The project will characterize the corrosion tolerance of various materials for use in advanced liquid metal reactors by probing their surface interactions with Pb and LBE liquid metal coolant at temperatures up to 1,000°C. A thin film of coolant, on the order of a few atomic layers, will be deposited on the surface before heating, enabling researchers to study the solid-liquid interface between the candidate composite materials and liquid metal coolants. The coolant edges will be probed with X-ray Absorption Spectroscopy (XAS) as a function of temperature and time to determine how they react with the solid underneath. Researchers will utilize a unique resource, the undulator beamline available from the Materials Research Collaborative Access Team, to support this project. Real time *in-situ* methods for corrosion characterization enable researchers to directly observe the fundamental chemical mechanisms that lead to corrosion.

Promising candidate materials as well as materials whose surfaces have been modified by applying the novel process of Ionized Plasma Deposition (IPD) will be studied. The IPD process has recently been developed and promises to revolutionize the development of composite coatings and to create surface treatments not possible by simple deposition techniques. IPD can impregnate a substrate material with a coating designed to have a specific composition, thickness, penetration, and nanostructure.

Work Scope

- Develop techniques for deposition of thin coolant layers onto a substrate, heat treatment procedures, and XAS measurement methodology.
- Study coolant reaction with steel substrates and candidate materials such as molybdenum, tantalum, zirconium, and silicon-carbide, and correlate data with conventional dip tests.
- Apply IPD to prepare surface-modified steel samples and perform further *in-situ* and long-term static tests using the best candidate materials.

NUCLEAR ENERGY RESEARCH INITIATIVE

Optimized, Competitive Supercritical-CO_2 Cycle GFR for Generation IV Service

PI: Michael J. Driscoll, Massachusetts Institute of Technology

Collaborators: None

Project Number: 05-044

Project Start Date: March 2005

Project End Date: February 2008

Project Description

This project will develop an integrated overall plant design for a gas-cooled fast reactor (GFR) based on the compact and highly efficient, direct supercritical carbon dioxide (CO_2) Brayton cycle. This plant will be capable of performing a full spectrum of desired Generation IV objectives, including economical electric power generation, high uranium utilization, the ability to burn transuranics and/or minor actinides, and production of hydrogen through the high-temperature electrolysis of steam. There are tradeoffs associated with achieving attractive economics and assuring a high degree of safety for the final plant design. Researchers will use probabilistic techniques to optimize these tradeoffs and develop a design that is both economical and safe. In addition, researchers will use existing Nuclear Regulatory Commission (NRC) deterministic prescriptions and proposed risk-informed licensing requirements to evaluate prospective plant designs.

Most worldwide research on this type of gas reactor is based on a helium cycle that operates at temperatures approaching 900°C, which poses severe core and component material challenges. This S-CO_2 cycle can achieve high thermodynamic efficiency (approximately 44 percent to 51 percent) at modest temperatures of 550 to 650°C. Therefore, creative synthesis of proven technology should suffice in many instances. The focal point of the major tasks of this project will be designing the reactor core and the decay heat removal systems. Overall, this project will provide a sufficient basis for assessing this type of GFR among the candidate Generation IV designs being evaluated for final selection.

Work Scope

Task 1. Core Design and Performance Assessment.

- Optimize features of vented fuel concept using tube-in-duct assemblies.
- Develop pin type core design as a benchmark for comparisons and as a fallback option.
- Confirm the burning capability of TRU and minor actinides.

Task 2. PRA Guided Design of Safety Systems.

- Develop decay heat removal design for accident, normal shutdown, and refueling.
- Develop improved emergency power systems, such as microturbines or fuel cells.
- Develop both active and passive means of shutdown assurance to preclude an anticipatory transient without scram (ATWS).

Task 3. Overall Plant Design and Economic Assessment.

- Design power cycles for core exit temperatures ranging from 550 to 700°C.
- Demonstrate integration with high-temperature electrolysis of steam for H_2 production.
- Estimate busbar costs of electricity relative to other reactor options.

NUCLEAR ENERGY RESEARCH INITIATIVE

Development of High-Temperature Ferritic Alloys and Performance Prediction Methods for Advanced Fission Energy Systems

PI: G. R. Odette, University of California- Santa Barbara

Collaborators: None

Project Number: 05-074

Project Start Date: March 2005

Project End Date: February 2008

Project Description

This project will focus on developing and evaluating a high-temperature alloy class, known as "nanostructured ferritic alloys" (NFAs). The effort will build on previous reactor materials research and ongoing fusion and fission materials programs. These alloys possess remarkable creep strength, excellent ductility, good fracture toughness potential, and may mitigate many radiation damage problems. The study will consider the combined effects of high service temperatures (up to 900°C or more) and high dose irradiations on NFAs. At these temperatures, material instabilities and degradation present great challenges. This study will focus on commercial-type alloys such as MA957. However, model alloy experiments will be continued to help researchers obtain a better understanding of NFAs and to optimize NFA processing paths and their balance of performance sustaining properties.

Work Scope

- Develop a comprehensive mechanical property and microstructural database on MA957 and other commercial NFAs. Develop semi-empirical constitutive models, including high-temperature irradiation effects, and assess the applicability of the master fracture toughness-temperature curve-shifts method to these alloys.
- Continue to establish a fundamental nano-microstructural and property database to support processing models that will help optimize properties and minimize costs. Conduct thermal aging studies and post irradiation examinations.
- Investigate advanced joining methods (e.g., solid-state welding and diffusion bonding) for MA957 that will have the least disruption on the beneficial NFA micro-nanostructures.
- Develop a semi-empirical model of the high-temperature thermal and irradiation stability of the nm-scale precipitates in NFAs and compare the predictions to the results of thermal aging and irradiation experiments.
- Conduct other activities, including collaboration on additional irradiation studies and design of an advanced reactor surveillance-component monitoring program.

NUCLEAR ENERGY RESEARCH INITIATIVE

Development and Analysis of Advanced High-Temperature Technology for Nuclear Heat Transport and Power Conversion

PI: Per F. Peterson, University of California, Berkeley

Collaborators: None

Project Number: 05-079

Project Start Date: May 2005

Project End Date: April 2008

Project Description

This three-year project will research advanced high-temperature heat transport and power conversion technology, in support of both the Nuclear Hydrogen Initiative and Generation IV Nuclear Energy System Initiative. It will focus on fundamental and applied questions about high-temperature heat transport using different combinations of gases and liquid molten salts. Selection of the working fluid is a major design issue for the Next Generation Nuclear Plant (NGNP), which will employ a 50-MWt, high-temperature, intermediate loop to demonstrate production of nuclear hydrogen.

The project makes contributions in four interrelated areas. First, researchers will develop detailed reference designs for the NGNP intermediate heat exchanger system, evaluating different heat transport fluids (high-pressure helium, intermediate-pressure helium, and molten flouride salt for hydrogen production). Second, they will conduct integral experiments to study transient high-temperature heat transport, generating experimental data for model development and code verification. Third, they will evaluate improvements to high-temperature, multi-reheat, helium Brayton cycle technology which will increase efficiency 5 to 10 percent and roughly double the power density. Fourth, researchers will conduct additional analysis work to further initial designs of the molten salt-cooled advanced high-temperature reactor (AHTR), which is a potential high-temperature heat source for electricity and power production.

Work Scope

- Develop intermediate heat exchanger design parameters and conduct a detailed design analysis comparing the various candidate fluids (high and intermediate pressure helium and molten fluoride salts). Evaluate both metallic and ceramic variants of each fluid and generate a detailed analysis for safety, stored energy, and reliability.
- Design, construct, and perform scaled, transient, heat transport experiments to characterize the proposed fluids.
- Develop detailed design and analyses of multiple-reheat helium Brayton power conversion systems for the AHTR and for application to modular Lead Fast Reactor.
- Develop intermediate heat transfer loop designs for AHTR based on results of the preceding tasks.

NUCLEAR ENERGY RESEARCH INITIATIVE

Development of Risk-Based and Technology-Independent Safety Criteria for Generation IV Systems

PI: William E. Kastenberg, University of California at Berkeley

Collaborators: None

Project Number: 05-080

Project Start Date: March 2005

Project End Date: February 2008

Project Description

This project will develop quantitative safety goals for Generation IV nuclear energy systems. These safety goals will be risk-based and technology-independent. The foundations for a new approach to risk analysis will be developed, along with a new operational definition of risk. This project will further the current state-of-the-art by developing quantitative safety goals for both Generation IV reactors and for the overall Generation IV nuclear fuel cycle. The risk analysis approach being developed will quantify performance measures, characterize uncertainty, and address a more comprehensive view of safety as it relates to the overall system.

Appropriate safety criteria are necessary to manage risk in a prudent and cost-effective manner. This study is also important for government agencies responsible for managing, reviewing, and approving advanced reactor systems because they are charged with assuring the health and safety of the public.

Work Scope

- Formulate a trial set of quantitative safety goals, considering the risks to the general public and those residing near nuclear plants and analyzing NRC's safety goals for commercial reactors, DOE's safety goals for defense facilities, safety goals for other hazardous industries, and radioactive exposure information. Risk will be formulated in absolute, incremental, and relative terms.
- Create an operational definition of risk, based on recent advances in Network Theory, which accounts for the complexity, uncertainty, and ambiguity of Generation IV systems. Create a nodal model for a nuclear energy system and numerically simulate loss of bulk materials, particulate and gaseous release, and waste materials to characterize risk.
- Develop a benefit/cost risk management scheme to evaluate design options of Generation IV systems and formulate top-level safety goals for allocation to nuclear systems, sub-systems, and components.

NUCLEAR ENERGY RESEARCH INITIATIVE

Development of Modeling Capabilities for the Analysis of Supercritical Water-Cooled Reactor Thermal-Hydraulics and Dynamics

PI: Michael Podowski, Rensselaer Polytechnic Institute

Collaborators: The Royal Institute of Technology, Sweden

Project Number: 05-086

Project Start Date: March 2005

Project End Date: February 2008

Project Description

The supercritical water-cooled reactor (SWCR) has several advantages over existing light water reactor (LWR) systems. For example, the SWCR uses a direct cycle with a single-phase working fluid and has high thermal efficiency. The objective of this project, undertaken in collaboration with the Royal Institute of Technology in Stockholm, Sweden, is to develop improved analytical and computational models, supported by new databases and validated scalable correlations, and to simulate SWCR behavior during normal operation and transients. Existing analytical techniques developed for current reactor technology are outdated and may cause inaccuracies in predicated system response. Advanced, next generation codes under development will be applied to supercritical water reactors. Researchers will also study the effect of coupling between hydrodynamic/thermal phenomena and reactor neutronics. The results of the experiments will be used to model SCWR thermal-hydraulics and dynamics.

Work Scope

- Develop an accurate and calibrated experimental database for supercritical water reactors.
- Develop and validate new scalable correlations and mechanistic models of flow and heat transfer.
- Implement, test, and validate the new models for application in existing reactor system analysis codes (RELAP) and computational fluid dynamics codes. The major computational tool will be NPHASE.
- Perform numerical simulations of supercritical water system dynamics using the NPHASE code.

NUCLEAR ENERGY RESEARCH INITIATIVE

Novel Processing of Unique Ceramic-Based Nuclear Materials and Fuels

PI: John D. Metzger, State University of New York

Collaborators: Brookhaven National Laboratory

Project Number: 05-110

Project Start Date: March 2005

Project End Date: February 2008

Project Description

This project will develop high-temperature refractory ceramic fuels, in-core materials, and control elements applicable to gas-cooled fast reactors. Refractory-based ceramics such as carbides, borides, and nitrides display a number of unique properties, including extremely high melting points, hardness, high thermal and electrical conductivity, and solid state phase stability. These unique properties make them potential candidates for a variety of high-temperature nuclear reactor components.

This research effort will demonstrate the feasibility of a novel processing approach for fabricating mixed-carbide refractory composite materials. The process is based upon the pyrolysis of a mixture of preceramic polymers and submicron/nano-sized metal particles of uranium, zirconium, niobium, and hafnium, carried out in both conventional and microwave ovens under an inert atmosphere. The materials produced will include metal ceramic carbides, mixed metal carbides, and unique metal silicocarbides. The resulting materials can be graded and will have a controlled microstructure (at both micron and nano levels), fiber-reinforced configurations, and a wide range of compositional control. This processing technique involves much lower energetic requirements compared to hot isostatic sintering. It also provides the capability of fabricating net-shaped components and does not suffer from maximum component size limitations, such as that of chemical vapor deposition.

Upon completion of this project, processes for fabricating a variety of unique high-temperature ceramic materials and fuels will be developed.

Work Scope

- Develop and implement a process to test and characterize materials produced for physical and microstructural composition, mechanical properties, and resistance to oxidation and thermal shock.
- Develop global and local models to predict the pyrolysis and reaction process; then, integrate the models for comparison to experimental measurements.
- Carry out a nuclear transport analysis with a ceramic-based fuel form to establish nuclear characteristics and potential fuel element configurations in order to determine a reactor core design and operational conditions.

NUCLEAR ENERGY RESEARCH INITIATIVE

Real-Time Corrosion Monitoring in Lead and Lead-Bismuth Systems

PI: James Stubbins, University of Illinois at Urbana-Champaign

Collaborators: None

Project Number: 05-114

Project Start Date: March 2005

Project End Date: February 2008

Project Description

This project addresses major corrosion issues associated with using lead and lead-bismuth liquid metals as working fluids in advanced nuclear systems, such as the Generation IV lead-cooled fast reactor concept or accelerator-based applications. The project's key approach to mitigating corrosion is to develop a persistent oxide film on the surface of internal structural components. The researchers will use Impedance Spectroscopy (IS) corrosion monitoring techniques to measure the kinetics and thermodynamics of the formation of oxides films on reactor structural materials. If properly formed and maintained, these films can provide a useful barrier to inhibit the corrosive attack of liquid metal. Researchers will study material alloying and surface treatment approaches that will form these protective films. The IS technology can measure the oxide film formation in real time and is sufficiently compact that it can be deployed at numerous locations in an operating system to monitor local corrosion processes directly.

The goals of this project are to develop advanced real-time corrosion monitoring methods, study the thermodynamics and kinetics of scale formation on selected alloys, and apply this understanding to develop new corrosion-resistant alloy compositions or surface treatment techniques. A central objective is to further develop impedance spectroscopy for characterizing and monitoring corrosion in liquid metal systems.

Work Scope

The research test plan is formulated to accomplish the three major goals:

- Develop the IS technique for lead and lead-bismuths systems, which requires significantly extending the scanning frequency range.
- Use the enhanced IS techniques to measure the thermodynamics and kinetics of oxide scale formation for a variety of materials and oxygen pressures at temperatures ranging from 400 to 700°C.
- Develop improved surface treatments and alloy compositions for enhanced corrosion resistance.

NUCLEAR ENERGY RESEARCH INITIATIVE

The Effect of Hydrogen and Helium on Irradiation Performance of Iron and Ferritic Alloys

PI: James Stubbins, University of Illinois at Urbana-Champaign

Collaborators: None

Project Number: 05-116

Project Start Date: March 2005

Project End Date: February 2008

Project Description

The goals of this research project are to develop a fundamental understanding of irradiation-induced defects on ferritic alloys, to understand the roles of hydrogen and helium in damage evolution, to extend this knowledge to increasingly complex iron alloy materials that are of interest for use in advanced reactor and accelerator-based applications, and to develop more irradiation-resistant alloys. Researchers will accomplish these goals through a combination of experiments and modeling.

Ferritic alloys are the prime choice for structural and component applications in several advanced reactor and accelerator-based system concepts. This class of alloy has relatively high irradiation resistance and retains good structural properties in temperatures ranging from 400 to 700°C. However, as the existing irradiation performance data was developed for the fast breeder program, the database represents fast fission neutron spectra. This research project will address irradiation damage and effects in advanced reactor and accelerator-driven systems concepts. The project will encompass substantial new developments in experimental analysis and high-speed computational modeling of the formation and evolution of defects in ferritic metals and alloys during radiation exposure. This work will also focus on the influence and possible synergistic effects of hydrogen and helium during defect production, clustering, and extended damage structures during irradiation. The experimental program will examine the evolution of point defects and defect structures as a function of radiation exposure, irradiation temperature, and H/He production in increasingly complex materials, ranging from the simplest single crystal iron, to polycrystalline iron and iron alloys, through advanced ferritic/martensitic steels.

Work Scope

- Perform microstructural and microchemical analyses for a set of irradiation exposure conditions. Materials include single crystal iron and increasingly complex binary, ternary, and commercial alloy systems, using ion irradiation to accurately control hydrogen and helium levels and irradiation dose.
- Perform large-scale molecular dynamics and kinetic lattice Monte Carlo simulations to understand the effects of irradiation on material properties and damage processes.

NUCLEAR ENERGY RESEARCH INITIATIVE

Alloys for 1,000°C Service in the Next Generation Nuclear Plant

PI: Gary S. Was, J. Wayne Jones, Tresa Pollock, University of Michigan

Collaborators: Special Metals Inc., Idaho National Laboratory, Oak Ridge National Laboratory

Project Number: 05-143

Project Start Date: March 2005

Project End Date: February 2008

Project Description

The objective of this project is to define strategies for improving alloys used for structural components in high-temperature helium reactors, such as the intermediate heat exchangers and primary-to-secondary piping. Specifically, the project will investigate oxidation/carburization from helium impurities, microstructural stability, and impact on creep behavior at temperatures between 900 and 1,000°C. The aim is to better understand the synergisms among these critical processes and to provide data for long-term prediction of properties.

The design of the very high temperature reactor that has been selected by the Department of Energy for the Next Generation Nuclear Plant project calls for outlet gas temperatures of 1,000°C. These are extremely challenging conditions for the operation of metallic components that will be required in the intermediate heat exchanger and primary-to-secondary piping. Inconel 617, an advanced nickel-based alloy, has been identified as the leading candidate for such applications. However, material properties in a high-temperature, impure helium environment are not sufficiently understood to qualify the alloy for service. Therefore, this study will also investigate alloy and microstructure modifications to enhance material properties.

Work Scope

- Study surface oxidation and carburization reactions over a range of conditions in a system designed to precisely control levels of specific impurities (hydrogen, water, carbon dioxide, carbon monoxide, and methane) in a helium environment. Degradation of surface microstructure as a function of impurity, exposure times, and temperatures will be studied in detail.
- Investigate the influence of impurity-induced surface degradation along with evolution of carbide and grain structure during creep deformation. The synergistic effects of these degradation mechanisms will be used to refine existing models for the prediction of creep behavior.
- Investigate alloying additions that are likely to improve carbide stability and/or influence diffusion and modification of the grain boundary structure through grain boundary engineering, since it is likely that alloy improvements will be required to reach the goal of 1,000°C operating temperature.

NUCLEAR ENERGY RESEARCH INITIATIVE

Heat Exchanger Studies for Supercritical CO_2 Power Conversion System

PI: Akira Tokuhiro, University of Missouri-Rolla

Collaborators: Argonne National Laboratory

Project Number: 05-146

Project Start Date: March 2005

Project End Date: February 2008

Project Description

Advanced power conversion technology, consisting of a gas turbine Brayton Cycle utilizing supercritical carbon dioxide as the working fluid, is being considered for application in some of the Generation IV nuclear energy systems. This design is expected to significantly reduce plant cost, size, and complexity, and to increase efficiency. An added benefit is that this design can operate at high temperatures suitable for cogeneration of hydrogen. While such benefits derive from the unique thermo-physical properties of supercritical CO_2, these same properties also present technical challenges to the regenerative heat exchanger design. To maximize the usefulness of this design, a compact heat exchanger would be required. Basic heat exchanger performance data are needed in order to develop an improved design for application to the supercritical CO_2 power conversion system.

The objectives of this project are to establish heat exchanger performance under design conditions, estimate performance for beyond design-basis accidents, and compare different heat exchanger design options.

Work Scope

The project will conduct the following activities:

- Design, construct, and operate an experimental facility for performance testing of compact heat exchangers for the supercritical CO_2 Brayton cycle recuperator/cooler application.
- Obtain heat transfer and pressure drop data to evaluate performance of selected compact heat exchanger designs (e.g., Printed Circuit Heat Exchanger).
- Develop fluid flow and heat transfer simulation models and tools to support the evaluation of heat exchanger designs.

NUCLEAR ENERGY RESEARCH INITIATIVE

Candidate Materials Evaluation for the Supercritical Water-Cooled Reactor

PI: Todd Allen, University of Wisconsin-Madison

Collaborators: University of Michigan

Project Number: 05-151

Project Start Date: March 2005

Project End Date: February 2008

Project Description

The supercritical water-cooled reactor (SCWR) system is being evaluated as a Generation IV concept because it builds on currently proven light water technology to provide high thermal efficiency and plant simplification. Developing, testing, and selecting suitable materials for cladding and internal components are central to designing such a reactor, as supercritical water presents unique challenges to the long-term performance of engineering materials.

The objective of this research is to investigate degradation of materials in the supercritical water environment. In particular, corrosion and stress corrosion cracking (SCC) have been identified as critical problems because the temperature and the oxidative nature of supercritical water may accelerate the processes. In addition, radiation can influence corrosion and SCC both by altering the material's microstructure and by accelerating corrosion and SCC, due to the generation of oxygen and other free radicals via radiolysis. Existing data on corrosion and stress corrosion cracking of materials in supercritical water are sparse and data on the behavior of irradiated alloys are non-existent.

Work Scope

- Study representative alloys (ferritic-martensitic steels, austenitic stainless steels, and nickel-based alloys) for their corrosion and stress corrosion cracking resistance in supercritical water. Conduct corrosion and SCC tests at various temperatures, exposure times, and water chemistries.
- Modify the near-surface chemistry, microstructure, and stress state of the alloys prior to corrosion testing by applying emerging plasma surface modification and grain boundary engineering technologies.
- Examine the effect of irradiation on corrosion and SCC of alloys in the as-received and modified/engineered conditions by irradiating samples using high-energy protons and then exposing them to supercritical water.

All of these tests will be performed in close coordination with and as a complement to the Generation IV testing programs on radiolysis corrosion/SCC of neutron irradiated materials in supercritical water.

NUCLEAR ENERGY RESEARCH INITIATIVE

Validation and Enhancement of Computational Fluid Dynamics and Heat Transfer Predictive Capabilities for Generation IV Reactor Systems

PI: Robert E. Spall, Utah State University

Collaborators: Fluent, Inc; Idaho National Laboratory

Project Number: 05-160

Project Start Date: March 2005

Project End Date: February 2008

Project Description

Currently, two primary approaches exist for computational fluid dynamics (CFD) modeling of reactor systems. Thermal/hydraulic analysis codes, such as RELAP, model the entire plant using coarse nodes but cannot predict small-scale flow details. Traditional CFD codes, such as FLUENT, are most adept at detailed flow and temperature predictions over specific regions. However, there are many unanswered questions regarding the ability of traditional codes to accurately model and predict complex flow patterns inherent in nuclear reactors, particularly turbulence. Turbulence is modeled either through direct numerical simulation (which is not practical for engineering design), large eddy simulation (which combines direct and empirical subgrid scale calculations), or Reynolds-averaged Navier-Stokes (RANS) equations (which is viable for complex geometries). Because no single model can handle all geometries, research is needed to validate, modify, and improve CFD predictive capabilities for the Generation IV reactors.

This project will validate and improve CFD predictive methods for Generation IV nuclear reactor systems. Researchers will assess the ability of large eddy simulation and RANS closure models, which are available in the FLUENT code, to predict flows for specific, fundamental geometries inherent in Next Generation Nuclear Plant reactors. Based on the results of the assessment, researchers will modify the closure models to improve predictive capabilities and obtain experimental data for relevant geometries to support code validation.

Work Scope

- Design and conduct experiments for parallel jets, crossflow, and crossflow cylinder array configurations.
- Perform a systematic validation study solving RANS equations in FLUENT using the above configurations, and determine necessary modifications to the turbulence model.
- Perform a systematic validation study using large eddy simulations to verify subgrid scale models in FLUENT and determine necessary modifications.

10. Advanced Fuel Cycles Initiative: Project Abstracts

This new program element includes 19 research projects, all of which were awarded in FY 2005. The following abstracts present each project's participants, its goals and objectives, and a description of the proposed work scope.

The Advanced Fuel Cycle Initiative (AFCI) is a wide-ranging research and development program whose mission is to develop and exhibit technologies that facilitate the conversion to an environmentally, socially, politically, and economically acceptable advanced fuel cycle. The chief goals are to develop fuel systems for Generation IV reactors and to create enabling fuel technologies, such as fuel, cladding, waste forms, separations, and disposal technology to decrease spent fuel volume; separate long-lived, highly radiotoxic elements; and recover valuable energy from spent fuel. The technologies will support both existing and forthcoming nuclear energy systems, including Generation IV systems. Planned NERI research efforts in FY 2005–FY 2007 associated with the AFCI include projects related to separations, fuels, transmutation, and systems analysis.

- Separations: Separations research is comprised of areas such as the development of aqueous and hybrid aqueous-pyrochemical separations technologies; advancement of spent fuel treatment processes; improvement of temporary or permanent waste storage forms; treatment of spent fuel from the experimental breeder reactor (EBR-II) in preparation for disposal; and conceptual planning of future spent fuel treatment plants and advanced processing technologies. Planned projects will include selecting a process for LWR spent fuel reprocessing, including isotope separation and recovery, and fabrication of waste/storage modules (forms) for storage in a repository; demonstrating the one-group transuranic extraction process; starting the accelerated EBR-II blanket treatment using advanced processing technology; demonstrating internationally advanced LWR spent fuel processing; demonstrating americium/curium separations processes and selecting a reference process; and choosing reference storage or disposal forms for uranium, plutonium/neptunium, americium/curium, and cesium/strontium.
- Fuels: This research area includes projects associated with advanced fuel development for LWRs, Generation IV reactors, and dedicated transmuters; remote fuel fabrication evaluation; development and selection of advanced clad materials; safety analyses of different advanced fuel types; and fabrication, characterization, performance testing, post irradiation examinations (PIE), and modeling of reference and transmutation fuels for various reactors (LWR, VHTR, fast spectrum systems). NERI research projects for the next three years will focus on continuation of tristructural isotropic (TRISO) fuel development including shakedown testing, performance testing, and fission product transport; and Advanced Test Reactor (ATR at INL) irradiation and post irradiation examination (PIE) of mixed-oxide and inert matrix fuel, as well as nitride, metal, and dispersion fuels.
- Transmutation: Transmutation is a process by which long-lived radioactive species are converted to short-lived nuclides via neutron capture or fission. Transmutation science and engineering supports AFCI, specifically in accelerator-driven systems, transmuter materials and coolants, and transmutation physics. The primary purpose of this research area is to develop an engineering basis for the transmutation of plutonium, minor actinides, and long-lived fission products. During the next three years the program will focus on (1) continued measurements and evaluations of higher actinides to reduce transmutation uncertainty, (2) new irradiations at the PHENIX reactor in France, (3) examination of irradiated materials from the Fast Flux Test Facility (FFTF) and the European PSI facility, (4) development of an intense fast neutron source for materials testing, (5) development of alloys and surface treatment to increase Lead-Bismuth Eutectic (LBE) corrosion resistance, and (6) international collaboration to advance Accelerator Driven Systems technology.
- Systems Analysis: Broad systems studies, transmutation system studies and integrated model development, and fuel cycle proliferation resistance, economics, and safety assessment are all tasks within the domain of this research area. The research objectives of NERI projects under this area are to provide analysis on fuel cycle infrastructure needs, supply recommendations on fuel types and reactor systems from the perspective of the overall fuel cycle, and perform analyses to assist DOE in determining the need for a second repository.

NUCLEAR ENERGY RESEARCH INITIATIVE

Directory of Advanced Fuel Cycle Initiative Project Abstracts

NUCLEAR ENERGY RESEARCH INITIATIVE

Determination of Basic Structure-Property Relations for Processing and Modeling in Advanced Nuclear Fuels: Microstructure Evolution and Mechanical Properties

PI: Pedro D. Peralta, Arizona State University

Collaborators: Los Alamos National Laboratory

Project Number: 05-001

Project Start Date: March 2005

Project End Date: February 2008

Project Description

Fuel development under the Advanced Fuel Cycle Initiative (AFCI) will rely heavily on modeling physical mechanisms to predict performance rather than conducting an experimental characterization or using empirical relationships derived from experimental data. This project will investigate a complementary strategy for fuel development. Through performing experimental work on relevant surrogate materials, particularly in the case of inert matrix fuels where less experimental characterization is currently available, researchers will study the structure-property relationship of nitrides and oxides in solid solutions using surrogate elements to simulate the behavior of inert matrix fuels emphasizing zirconium-based materials. The goal is to provide insight into processing fuel that has better performance and greater structural reliability during manufacturing and service and to develop structure-property relations that can be used as input for fuel performance models.

Researchers will explore three key aspects of these materials. They will characterize the microstructure by measuring global texture evolution and local crystallographic variations, determine mechanical properties (including fracture toughness, compression strength, and hardness) as functions of load and temperature, and develop structure-property relations to describe mechanical behavior of the materials based on experimental data. Using crystallographic information in evaluating fuel performance and incorporating statistical variations of microstructural variables into simplified models of mechanical behavior of fuels are new aspects of this research. Work with actual fuels will be carried out in parallel in collaboration with Los Alamos National Laboratory.

Work Scope

- Develop processing procedures for pellet fabrication and characterize their properties.
- Establish appropriate surrogates for plutonium, fabricate samples, and optimize the process.
- Translate techniques from surrogate processing to actual fuel; fabricate and characterize.
- Establish models for high-temperature behavior, characterize properties, and perform testing and analysis.

NUCLEAR ENERGY RESEARCH INITIATIVE

The Application of Self-Propagating High-Temperature Synthesis (SHS) to the Fabrication of Actinide Bearing Nitride and Other Ceramic Nuclear Fuels

PI: John J. Moore, Colorado School of Mines

Collaborators: Idaho National Laboratory

Project Number: 05-013

Project Start Date: March 2005

Project End Date: February 2008

Project Description

This project will use an exothermic combustion synthesis method, termed self-propagating high-temperature synthesis (SHS), to produce high-quality, reproducible nitride fuels and possibly other ceramic-type nuclear fuels (cercers and cermets, etc.) in conjunction with the fabrication of transmutation fuels. This method of fuel fabrication has potential application in the design of other nuclear energy systems, including advanced power reactors and space nuclear propulsion reactors. The method, which is based on the concept of high reaction and cooling rates, is applicable to a wide variety of ceramic and intermetallic materials including nitrides, zirconides, oxides, carbides, oxycarbides, carbonitrides, and chalcogenides, which have been synthesized using SHS techniques for a range of applications. SHS offers a number of advantages that are suitable for remote processing of radioactive materials, such as one-step synthesis and consolidation, simplicity, and ease of containment, equating to reduced cost and high product throughput. Careful control of operational parameters offers the possibility of tailoring material properties to specific requirements.

The major research objective of this project is to determine the fundamental SHS processing parameters by first using manganese as a surrogate for americium to produce dense Zr-Mn-N ceramic compounds. These fundamental principles will then be transferred to the production of dense Zr-Am-N ceramic materials. Because of the high vapor pressures of Am and americium nitride (AmN), there is concern about producing nitride ceramic nuclear fuels containing Am. Sintering results in a major retention problem of Am utilizing current processing methods and it adversely affects the synthesis of a consistent product with desirable homogeneity, density, and porosity. Similar difficulties experienced during laboratory-scale process development for producing metal alloys containing Am have led researchers to abandon compact powder sintering methods and investigate other synthesis methods, such as SHS.

A further research objective in this research program will be to generate fundamental SHS processing data to the synthesis of (i) Pu-Am-Zr-N and (ii) U-Pu-Am-N ceramic fuels. In this case, Ce will be used as the surrogate for Pu, Mn as the surrogate for Am, and depleted uranium. The sequence for SHS synthesis of these ceramic fuels will be as follows:

(i) Pu-Am-Zr-N: using Ce as the surrogate for Pu, and Mn as the surrogate for Am
   a. $(Ce_{0.5}Mn_{0.5})N$
   b. 64wt%$[(Ce_{0.5}Mn_{0.5})N]$-36wt%ZrN

(ii) U-Pu-Am-N: using depleted uranium (DU), Ce as the surrogate for Pu, and Mn as the surrogate for Am:
   a. $(DU_{0.5}Mn_{0.5})N$
   b. $(DU_{0.5}Ce_{0.25}Mn_{0.25})N$

Once sufficient fundamental data has been determined for these surrogate systems, the data will be transferred to Idaho National Laboratory for synthesis of Zr-Am-N, Pu-Am-Zr-N, and U-Pu-Am-N ceramic fuels.

Work Scope

- Synthesize porous zirconium-nitride and manganese-nitride compounds using high pressure N_2 gas. The porous nitrides are readily ground into powder for subsequent processing. Fundamental thermochemical property data will be collected throughout and the processes optimized.

- Synthesize dense Zr-Mn-N (ZrAmN) compounds using a one-step SHS consolidation process. Determine fundamental thermochemistry and kinetic data and optimize the process.
- Synthesize porous $(Ce_{0.5}Mn_{0.5})N$, dense 64wt%$[(Ce_{0.5}Mn_{0.5})N]$-36wt%ZrN, $(DU_{0.5}Mn_{0.5})N$, and dense $(DU_{0.5}Ce_{0.25}Mn_{0.25})N$.
- Transfer the technology to synthesize dense Zr-Am-N, Zr-Am-N, Pu-Am-Zr-N, and U-Pu-Am-N ceramic compounds containing uranium and plutonium, and optimize the processes.

NUCLEAR ENERGY RESEARCH INITIATIVE

Minor Actinide Doppler Coefficient Measurement Assessment

PI: Nolan Hertel, Georgia Institute of Technology

Collaborators: Education, Research and Development Association of Georgia Universities

Project Number: 05-024

Project Start Date: March 2005

Project End Date: February 2006

Project Description

This project will assess the viability of measuring the Doppler coefficient of minor actinides. Using a series of calculations, researchers will estimate the change in reactivity resulting from a change in the operating temperature of small (approximately 1 gram) quantities of pure minor actinides. Resultant data will be used to design experiments in order to validate the computer calculations.

Calculations will be performed as a function of five parameters (isotope, sample quantity, operating temperature, critical assembly, and data library), using the radiation transport code MCNPX. The objective is to determine the quantity of minor actinide material needed to effect a 1E-5 change in reactivity due to a 200 degree Kelvin change in the operating temperature of the sample (e.g., from 800 K to 1,000 K). Results from the calculations will be used to develop an experiment that will measure the Doppler coefficient of each isotope. For these experiments, researchers will place actinide sample material in the FLATTOP and COMET critical assemblies at the Los Alamos Critical Experimental Facilities. The isotopes proposed for this study are: Pu-239, Np-237, Pu-238, Am-242m, Am-243, Pu-241, Am-241, and Cm-244.

As part of the Advanced Fuel Cycle Initiative (AFCI), research using advanced fast reactors in combination with accelerator-driven systems for the transmutation of waste has shown the ability to reduce the amount of plutonium and transuranic materials for disposal. The Doppler coefficients of each isotope are needed so researchers can conduct safety assessments of transmutation systems containing fuel with a high minor actinide content.

Work Scope

This project will perform the following activities:

- Determine critical assembly setup and operating temperatures. Perform initial calculations to identify the range of sample quantities for each isotope.
- Perform MCNPX calculations and quantify the amount of each minor actinide required for a 1E-5 change in reactivity.
- Analyze calculation results to determine the possibility of performing experimental validation on the two critical assemblies.
- If results are favorable, design a set of benchmark experiments to measure the Doppler coefficients.

NUCLEAR ENERGY RESEARCH INITIATIVE

Detailed Reaction Kinetics for CFD Modeling of Nuclear Fuel Pellet Coating for High-Temperature Gas-Cooled Reactors

PI: Francine Battaglia, Rodney O. Fox, and Mark S. Gordon, Iowa State University

Collaborators: None

Project Number: 05-030

Project Start Date: March 2005

Project End Date: February 2008

Project Description

High-temperature gas-cooled nuclear reactors are being adapted to produce a safe, effective, and economic source of energy. The fuel source for the reactors is from small uranium pellets coated to trap and retain radioactive fission products from escaping into the environment. Spouting fluidized beds, in which a central jet of gas fluidizes particles so they circulate in a regular pattern, are used to uniformly coat the fuel pellets. However, issues such as agglomeration, attrition, and breakage of particles need to be resolved in order to produce the highest quality particle coating. Further studies are required to improve the chemical vapor deposition (CVD) process to produce quality mixing and reactions for coating. While additional experimental data are necessary, direct measurements can be difficult to obtain. A complementary approach is to use computational fluid dynamics (CFD) to model the CVD process.

The objective of this research is to conduct a state-of-the-art computational study of the chemical vapor deposition process in a spouting bed in order to validate and improve CFD models for the reaction kinetics to coat uranium fuel pellets with carbon and silicon carbide. This project will take a complementary approach using computational methods to model the CVD process and apply it as a tool for reactor design, scale-up, and optimization. The work will validate the computations with experimental data for the multiphase fluid mechanics and species chemistry predictions required to describe the CVD process.

Work Scope

- Use computational chemistry to develop detailed reaction kinetics models of the gas-phase and surface molecule interaction, with a goal of predicting surface coating rates.
- Implement the reaction kinetics using in-situ adaptive tabulation for complex chemistry and couple to the MFIX computer code.
- Implement a polydispersity model in MFIX to account for effects of particle size distribution.

NUCLEAR ENERGY RESEARCH INITIATIVE

On-Line Fuel Failure Monitor for Fuel Testing and Monitoring of Gas-Cooled Very High Temperature Reactors

PI: Ayman I. Hawari, North Carolina State University

Collaborators: None

Project Number: 05-054

Project Start Date: March 2005

Project End Date: February 2008

Project Description

This project will devise an accurate approach to detect failures of tristructural isotropic (TRISO) fuel based on measurements of fission gas activity released into the effluent stream. In the gas-cooled very high temperature reactor (VHTR), the fuel is made up of TRISO microspheres composed of a UO_2 kernel surrounded by a porous pyrolytic graphite buffer, an inner pyrolytic graphite layer, a silicon carbide (SiC) coating, and an outer pyrolytic graphite layer. The layer/coating system that surrounds the UO_2 kernel acts as the containment and main barrier against the environmental release of radioactivity. However, due to hostile in-core conditions (e.g., high temperature, fast neutron flux, etc.), it is anticipated that a certain number of TRISO microspheres will fail during reactor operation. To ensure compliance with radiological and safety requirements, it is essential to detect any fuel failure at the earliest stage possible.

The aim of this project is detection of a single failed TRISO particle per testing capsule. Researchers believe that fuel failure rates on the order of 10^{-5} are detectable and that the detection method will provide insight into the failure mode. Various detection methods will be studied to differentiate the minute fission product signal from background. As part of the fuel development program for the VHTR, researchers will conduct fuel failure experiments at the Advanced Test Reactor of Idaho National Laboratory (INL). Methods and instrumentation developed by this project are expected to be applicable to on-line fuel failure monitoring of VHTRs.

Work Scope

Researchers will study the option of performing on-line measurements using gamma-ray spectrometers that are placed along the path of the coolant flow. They will also consider sampling the main flow and collecting the appropriate fission products in an optimal environment to perform signal-to-noise measurements. In both cases, specialized instruments, measurement techniques, and data processing methods will be required to discern minimum detectable activity limits. The project tasks will include:

- Calculating isotopics of TRISO microspheres and estimating fission product activity released from failure of a single microsphere.
- Examining the response of various detectors and analyzing the resulting spectra.
- Designing, constructing, and testing a prototype gas extraction system coupled to various detectors.

NUCLEAR ENERGY RESEARCH INITIATIVE

Plutonium Chemistry in the UREX+ Separation Processes

PI: Alena Paulenova, Oregon State University; Ken Czerwinski, University of Nevada-Las Vegas

Collaborators: None

Project Number: 05-062

Project Start Date: March 2005

Project End Date: February 2008

Project Description

The transmutation of long-lived radionuclides is being developed as a part of DOE's Advanced Fuel Cycle Initiative to address the disposal of commercial nuclear reactor fuel and to improve the performance of the geologic repository. This project will examine the chemical speciation of plutonium in UREX+ (uranium/tributylphosphate) extraction processes for advanced fuel technology. Researchers will investigate the role of nitric acid and nitrates in plutonium separation and speciation. They will identify the plutonium species formed in aqueous and organic extraction phases and at the interface between phases, and will also determine plutonium distribution in each phase. This will allow a greater understanding of plutonium's extraction chemistry and enable the separation processes to be optimized.

Work Scope

Researchers will analyze the change in speciation using kinetic computer codes and existing thermodynamics to examine the speciation of plutonium in aqueous and organic phases. They will examine the different oxidation states of plutonium to find the relative distribution between the aqueous and organic phases under various conditions, such as different concentrations of nitric acid, total nitrates, or actinide ions. Other parameters that may affect plutonium speciation include pH levels, a range of oxidation-reduction (redox) potential, varying temperatures, and contact time.

The experimental procedures will be based mainly on radiochemical techniques and will focus on evaluating plutonium and uranium distribution as a function of the test conditions. Researchers will perform spectroscopic and radiochemical experiments on the actinides. They will also utilize techniques such as X-ray Absorbance Spectroscopy and Small-Angle Neutron Scattering for determining plutonium and uranium speciation in all separation stages.

NUCLEAR ENERGY RESEARCH INITIATIVE

Development of an Engineered Product Storage Concept for the UREX+1 Combined Transuranic/Lanthanide Product Streams

PI: Sean M. McDeavitt, Purdue University

Collaborators: Argonne National Laboratory

Project Number: 05-066

Project Start Date: March 2005

Project End Date: February 2008

Project Description

This project will develop a storage form for transuranic (TRU) oxides and rare earth fission product (REFP) oxides isolated from spent nuclear fuel using advanced aqueous processing methods. The candidate storage forms include simple powder storage with engineered barriers and manufactured cermet forms that would enable TRU burning in a fast reactor after an intermediate storage period.

TRU and REFP are being extracted from spent nuclear fuel through the aqueous processing methods developed under the Advanced Fuel Cycle Initiative. The goal of this project is to secure these oxides in a proliferation-resistant state until the closed fuel cycle fast reactor infrastructure is in place. The required TRU storage "lifetime" is estimated to be 50 to 60 years before a fast burner technology is available, but the maximum design storage duration is set at approximately 500 years.

The goals of the project are to (1) design and develop the individual processing steps required to take the uranium and transuranic products from an aqueous nitrate solution to a final storage state, and (2) evaluate the impact of phenomena that govern durability of the storage form, material processing, and TRU utilization in fast reactor fuel.

Work Scope

- Develop the processing logic and methods for converting actinide and lanthanide nitrate solutions into oxide powders, recycle the irradiated Zircaloy cladding to be used as a cermet matrix, and fabricate a cermet storage form that may be used as a TRU burning fuel pin in a future fast reactor system.
- Use computational methods to evaluate the TRU burning performance of the cermet storage/fuel pin in a simulated Generation IV reactor system and compare the proposed cermet storage form with alternative approaches such as sealed container storage of TRU oxide powders.

NUCLEAR ENERGY RESEARCH INITIATIVE

Selective Separation of Americium from Lanthanides and Curium by Aqueous Processing with Redox Adjustment

PI: Kenneth L. Nash, Washington State University

Collaborators: Idaho National Laboratory

Project Number: 05-082

Project Start Date: March 2005

Project End Date: February 2008

Project Description

This project investigates application of conventional chemical methods for selective separation of actinide isotopes from lanthanides in mixed oxide (MOX) fuels using aqueous processing with oxidation state adjustment. Increased application of MOX fuels and longer burnup times for conventional fuels result in production of higher concentrations of the transplutonium actinides americium and curium and also several heavier species. The half-lives of the americium isotopes (up to 7,370 years) are significantly longer than those of the other isotopes. With the removal and transmutation of americium (Am), radiation of high-level wastes are reduced to levels approaching uranium mineral within less than 1,000 years, as opposed to the 10^4 to 10^5 year time-frame required for natural decay. The Advanced Fuel Cycle Initiative (AFCI) established a separation factor exceeding 10^4 as a goal for selectively isolating Am from spent fuel. Because the chemistries of trivalent lanthanides and actinides under aqueous processing conditions are so similar, satisfying this objective is extremely challenging. Since trivalent ion-recognition techniques cannot provide an adequate thermodynamic basis for such a separation in an acceptable number of contacts, this project will investigate oxidation state adjustment methods to achieve selective partitioning of Am from lanthanides and Cm using a multifaceted approach employing both conventional and unconventional aqueous separations methods and materials.

The research goal is to develop strategies for adjusting the oxidation state and demonstrating a successful separation based on oxidation of americium. Additional studies will be conducted on the most promising candidate process identified.

Work Scope

- Investigate the applicability of conventional aqueous separation processes to americium.
- Examine methods of facilitating the spontaneous organization of molybdenum to self-assemble around an oxidized americium species. (Using a material naturally present in fission products is advantageous for waste management.)
- Investigate use of room temperature ionic liquids as media for americium oxidation state adjustment.

NUCLEAR ENERGY RESEARCH INITIATIVE

Selective Separation of Trivalent Actinides from Lanthanides by Aqueous Processing with Introduction of Soft Donor Atoms

PI: Kenneth L. Nash, Washington State University

Collaborators: Pacific Northwest National Laboratory

Project Number: 05-083

Project Start Date: March 2005

Project End Date: February 2008

Project Description

This project investigates new chemical methods for selective separation of actinide isotopes from lanthanides in mixed oxide (MOX) fuels by aqueous processing with soft donor atoms. Implementation of a closed loop nuclear fuel cycle requires the utilization of MOX fuels containing plutonium. This results in increased production of transplutonium actinides, most significantly isotopes of americium and curium. Because the presence of these isotopes significantly impacts the long-term radiotoxicity of high-level waste, it is important to develop effective methods for their isolation and transmutation. Transmutation is most efficiently done in the absence of lanthanide fission products, making effective procedures to separate the actinides from the lanthanides important.

As the chemistries of these elements are nearly identical, differing only in the slightly stronger interaction of trivalent actinides with certain ligand donor atoms, separation is a complex task. Current research has led to the development of new reagents and processes with considerable potential for accomplishing separation. However, pilot-scale testing indicates these reagents are susceptible to radiolytic and hydrolytic degradation.

The objective of this research project is to identify the most effective and economic means of controlling radiation damage in actinide/lanthanide extraction systems. Researchers will study the extraction and chemical stability properties of a class of soft-donor extractants adapted from water-soluble analogs, and the application of simple soft-donor anions, like thiocyanate, in tandem with conventional extractant molecules. Researchers will also study the potential of radical scavenger molecules to protect soft-donor extractant molecules from radiolytic degradation.

Work Scope

- Investigate selective extraction of trivalent americium and curium from ammonium thiocyanate solution.
- Adapt the TALSPEAK process to these extractant systems using water-soluble species.
- Explore radiation-protection methods based on removal of radiolytic degradation products from the extractant phase.
- Design ligands for selected new classes of reagents (lipophilic analogs of EDTA and thio polyamide derivatives).

NUCLEAR ENERGY RESEARCH INITIATIVE

Development of Nanostructured Materials with Improved Radiation Tolerance for Advanced Nuclear Systems

PI: Xinghang Zhang and K. Ted Hartwig, Texas A&M University

Collaborators: Los Alamos National Laboratory

Project Number: 05-088

Project Start Date: March 2005

Project End Date: February 2008

Project Description

This project will explore the fundamental mechanisms through which interfaces in nanolayered structures and grain boundaries of bulk nanomaterials are able to attract and rapidly eliminate point defects and unwanted foreign species. Candidate materials that will be studied include both nanostructured multilayer composites synthesized by magnetron sputtering (a bottom-up approach) and structural bulk nanomaterials produced by severe plastic deformation, equal channel angular extrusion (a top-down approach).

This project will have a profound and broad impact on understanding the fundamental science of improving radiation resistance by inducing interfaces and grain boundaries in nanomaterials and by designing and engineering structural nanomaterials for advanced nuclear reactors. Data from this study will be used for estimating defect capture rates and lifetimes of multilayered structures and bulk nanomaterials in conditions that would seriously degrade conventional microstructures. Researchers expect this project to benefit a broad spectrum of DOE programs associated with fission technology research and development.

Work Scope

- Evaluate the thermal stability of nanomaterials via differential scanning calorimetry (DSC), resistivity measurements, and long-term annealing studies.
- Explore the corrosion resistance of multilayer coatings.
- Conduct a detailed examination of ion-irradiated multilayered nanocomposites and bulk nanomaterials via high-resolution transmission electron microscopy (HRTEM), scanning transmission electron microscopy (STEM), and x-ray diffraction (XRD) in order to examine the development of stable defect clusters as a function of proximity to interfaces and grain boundaries and the evolution of interface undulations that develop with IR radiation.
- Correlate microscopy studies on defect accumulation and migration with available simulations on the energetics of point defect migration to and within interfaces and grain boundaries and the stability of layered structures during various perturbations, including radiation-induced mixing.
- Explore the role of the atomic structure of the interface in eliminating damage.

NUCLEAR ENERGY RESEARCH INITIATIVE

Utilization of Minor Actinides as a Fuel Component for Ultra-Long Life VHTR Configurations: Designs, Advantages, and Limitations

PI: Pavel V. Tsvetkov, Texas A&M University

Collaborators: None

Project Number: 05-094

Project Start Date: March 2005

Project End Date: February 2008

Project Description

This project will assess the advantages and limitations of using minor actinides as a fuel component to achieve ultra-long life very high temperature reactor (VHTR) configurations. Researchers will consider and compare the capabilities of pebble-bed and prismatic core designs with advanced actinide fuels to achieve ultra-long operation without refueling. Since both core designs permit flexibility in component configuration, fuel utilization, and fuel management, it is possible to improve fissile properties of minor actinides by neutron spectrum shifting through configuration adjustments. Consequently, small reactivity swings will be sustained in the VHTR configurations with minor actinides over a prolonged irradiation period, yielding high levels of burn-up.

This project will study advanced actinide fuels, which could reduce the long-term radiotoxicity and heat load of high-level waste sent to a geologic repository and enable recovery of the energy contained in spent fuel. The ultra-long core life approach would reduce the technical need for additional repositories and would improve marketability of the Generation IV VHTR by allowing worldwide deployment, including in developing countries. Utilizing minor actinides would facilitate development of new fuel cycles and support fuel supply sustainability.

Work Scope

This work will consist of six tasks:

- Develop whole-core/system models with explicit multi-heterogeneity treatments.
- Develop benchmark test problems to compare with experimental data.
- Validate and verify the VHTR models.
- Analyze uncertainty effects on VHTR performance characteristics.
- Analyze configuration variation capabilities to achieve ultra-long operation without refueling, maximize burn-up levels, and minimize reactivity swings.
- Complete control, dynamics, safety, and proliferation-resistance studies of ultra-long life VHTR configurations with advanced actinide fuels.

Researchers will also evaluate ex-core fuel cycle segments and study the impacts of increasing burn-up levels on the generated radioactivity.

NUCLEAR ENERGY RESEARCH INITIATIVE

Ambient Laboratory Coater for Advanced Gas Reactor Fuel Development

PI: Duane D. Bruns, University of Tennessee

Collaborators: None

Project Number: 05-118

Project Start Date: March 2005

Project End Date: February 2008

Project Description

Researchers will explore the behavior of fluidized bed technology for applying coatings to nuclear fuel particles for the Advanced Gas Reactor. They will evaluate the effects of particle characteristics (size, density, surface sphericity), along with bed size, inlet nozzle diameter, distributor shape, and gas flow rates on the hydrodynamics of fuel coating. They will provide experimental measurements to the Oak Ridge National Laboratory (ORNL) fluidization reactor modeling team to validate numerical models for use in parametric design simulations. The modeling team can then utilize the simulation code to design improved beds that will be constructed and characterized in the laboratory.

This project will also develop online feedback control strategies, study ozone reaction to access mass transfer in limiting regimes with simple reaction kinetics, study step changes in gas inlet temperature to evaluate heat transfer, and evaluate the effect of particle size distribution and segregation on operation. A major challenge will be to account for the effects of hydrodynamics on the chemical vapor deposition rate and to characterize the controlling factors and final product quality of the turbulent gas-solid particle interface in the fluidized bed reactor.

Four- and six-inch ambient laboratory mockup coaters that are designed to accommodate various nozzles and distributors will be fabricated. The bed will be instrumented with appropriate sensors and the resulting measurements synchronized with digital images to develop correlations of bed characteristics as a function of pressure, gas and particle velocity, void fraction, spout height, and spout diameter. Ultimately, this project will establish a flexible laboratory coater, collect high-quality measurements for a range of operating regimes, and develop correlations from the experimental data. The measurements and correlations will be used to enhance simulations, improve operations techniques, and provide for online feedback control.

Work Scope

- Design and fabricate fluidized beds with changeable nozzles, distributors, and other components.
- Develop LabVIEW software for operation, control, data collection/synchronization, and feedback control.
- Develop new measurement techniques and implement maturing technology, such as the ability to use fiber optic probes for void fraction measurement and particle velocity.
- Integrate design of an ozone generator/analyzer; determine catalytic properties of surrogate particles and their reaction to ozone.

NUCLEAR ENERGY RESEARCH INITIATIVE

Uncertainty Analyses of Advanced Fuel Cycles

PI: L.F. Miller, University of Tennessee

Collaborators: Argonne National Laboratory

Project Number: 05-123

Project Start Date: March 2005

Project End Date: February 2008

Project Description

In order for nuclear power to make a significant long-term contribution to the supply of high-quality energy, it is essential to deploy breeder reactors and other advanced nuclear fuel cycles. Using advanced systems with conversion ratios that exceed unity essentially provides an infinite fuel supply. Given that many alternative fuel cycles appear to be attractive, it is important to evaluate their costs and sustainability. The quality of the results should also be evaluated based on understanding the uncertainty associated with these assessments. Results from this study will enable DOE to better understand the implications of fuel cycle options relative to sustainability of energy production and of costs imputed through utilization of advanced fuel cycles.

To perform this work, researchers will conduct a detailed assessment of fuel cycles considered to be viable for relatively near-term implementation and they will thoroughly evaluate the uncertainties of information associated with these fuel cycles. The objective is to understand the issues that influence the sustainability of power generated by nuclear energy. One result of this research is expected to include an information tool that consists of a set of nuclear energy development scenarios that would allow policy-makers to explore different planning options through a data-mining application.

Work Scope

- Design and analyze advanced LWR fuel cycles.
- Evaluate effect of data and technological uncertainties on advanced fuel cycles.
- Assess repository benefits, including the effect of uncertainties, and optimize key resources.
- Conduct dynamic fuel cycle scenario studies considering fast and accelerator-driven reactors and evaluate their optimal use from technological and market perspectives.

NUCLEAR ENERGY RESEARCH INITIATIVE

BWR Assembly Optimization for Minor Actinide Recycling

PI: G. Ivan Maldonado, University of Cincinnati

Collaborators: Oak Ridge National Laboratory, Los Alamos National Laboratory, and Westinghouse Electric Company

Project Number: 05-125

Project Start Date: March 2005

Project End Date: February 2008

Project Description

The primary objective of this project is to apply and extend the latest advancements in light water reactor (LWR) fuel management optimization to design advanced boiling water reactor (BWR) fuel assemblies for the recycling of minor actinides. Two specific objectives are: (1) developing a new methodology for the direct coupling between the pin-by-pin bundle loading control variables and the core-wide (bundle-by-bundle) optimization objectives, and (2) extending this new methodology into a new application that includes control variables, objectives, and constraints designed to maximize targeted minor actinide incineration. The first objective is expected to uncover considerable dormant thermal margin, while the second objective (the addition of minor actinides) is expected to consume some of this uncovered margin. Therefore, a goal of this project is for optimization-led improvements in fuel cycle efficiency to offset potential losses in efficiency associated with the recycling of minor actinides.

This project implements an Advanced Fuel Cycle Systems Analysis program initiative to investigate spent fuel treatment and recycling options for current generation LWRs, and it supports the DOE's technical assessment of a second high-level waste repository.

Work Scope

- Perform within-bundle related developments. Define candidate minor actinides. Also, specify control variables, objectives, and constraints at the bundle level. Generate families of lattices and bundles as a function of relevant parameters (exposure, void fraction, etc.).
- Perform core-wide activities. Define bundle-to-core coupling. Generate families of core loadings and corresponding operational strategies (control rod patterns, core flow). Validate diffusion theory analyses via transport theory benchmarks.
- Complete global integration of the project. Verify proper functioning of models, generate optimized advanced BWR bundles and associated fuel cycle strategies.

NUCLEAR ENERGY RESEARCH INITIATIVE

Optimization of Oxide Compounds for Advanced Inert Matrix Materials

PI: Juan Nino, University of Florida

Collaborators: Imperial College of Science, Technology, and Medicine (London); Idaho National Laboratory

Project Number: 05-134

Project Start Date: March 2005

Project End Date: February 2008

Project Description

This project will model, design, synthesize, and characterize oxide ceramics that have optimized thermophysical properties for potential use as inert matrix fuel (IMF). This will be accomplished through a combined computational and experimental approach. The objective of this research program is to develop oxide compounds and composites that exhibit minimum radiation swelling, high thermal conductivity, and excellent hot water corrosion resistance.

Another potential benefit of the optimized IMF materials compared to the uranium oxide (UO_2) fuel used in current generation reactors is higher thermal conductivity, which will considerably reduce the centerline temperature of the fuel. The resulting lower stored heat in the fuel will minimize the impact of a loss of coolant accident. Also, use of an inert matrix will avoid formation of transuranic fission products by neutron capture, thereby allowing higher fuel burnup. Better corrosion resistance will make the IMF compatible with coolant under cladding breach accidents.

Work Scope

The following activities will be completed for this project:

- Develop the experimental and computational infrastructure.
- Develop and optimize the magnesium oxide (MgO)-pyrochlore composite matrix through the following tasks: (1) perform computer simulation to select pyrochlore compounds with optimum thermal conductivity, irradiation tolerance, and thermal transport properties; (2) synthesize and characterize composites selected in Task 1; (3) fabricate MgO-pyrochlore composites; and (4) refine simulation codes and experimental techniques.
- Develop and optimize a single-phase inert matrix compound through the following tasks: (1) use computer simulation to select candidates for single-phase IMF, (2) synthesize/characterize the thermo-physical properties and microstructure of selected compounds, (3) perform irradiation testing, and (4) refine simulation codes and experimental techniques.

NUCLEAR ENERGY RESEARCH INITIATIVE

Synthesis and Optimization of the Sintering Kinetics of Actinide Nitrides

PI: Darryl Butt, University of Florida

Collaborators: None

Project Number: 05-135

Project Start Date: May 2005

Project End Date: April 2008

Project Description

Nitride-based nuclear fuels, particularly those based on uranium mononitride (UN) and plutonium mononitride (PuN), are candidate fuels for the Advanced Fuel Cycle Initiative. These fuels have a combination of higher uranium loadings with lower enrichments and higher thermal conductivity compared to other fuel forms. It is possible to produce inert matrix fuels from these nitrides because of their compatibility with candidate matrices such as zirconium nitride (ZrN) and refractory metals. Although several processes have been developed for synthesizing powder and monolithic forms of actinide nitrides, the sintering kinetics and mechanisms are not understood. Having a quantitative understanding of the kinetics and mechanisms of densification and grain coarsening is critical to both the outcome economics and product performance of any ceramic fabrication process. Through optimized processing conditions, sintering temperatures and equipment size can be greatly reduced, production rates can be increased, and properties can be improved and more precisely controlled.

Work Scope

- Synthesize actinide and surrogate powders of varying morphologies and particle size. Thoroughly characterize the powders and press them into fuel forms.
- Conduct detailed sintering studies to assess the specific rate equations and kinetics models as a function of time, temperature, grain size, and other processing variables.
- Determine rate limiting and process controlling mechanisms by applying fundamental models, characterizing microstructures, and comparing transport properties including solid state diffusion and gas transport.

An end result of these studies will be a processing model and economic analysis that will enable fuel processing to be done at lower temperatures, shorter times, and with less costly infrastructure.

NUCLEAR ENERGY RESEARCH INITIATIVE

The Development of Models to Optimize Selection of Nuclear Fuel Materials through Atomic-Level Simulation

PI: Simon R. Phillpot, University of Florida

Collaborators: S. B. Sinnott, H. J. Seifert, J. Tulenko, University of Florida

Project Number: 05-136

Project Start Date: March 2005

Project End Date: February 2008

Project Description

This project will develop an advanced fuel performance calculational platform based on the FRAPCON code. Detailed input about the properties of nuclear materials will be provided by databases determined from thermodynamic, atomic-level, and electronic-level calculations and simulations. Currently, all fuel performance codes are based on correlations derived from experimental data. The challenge is that these correlations cannot be extrapolated to operating conditions beyond the experimental points upon which the correlation is based, nor can they be used for materials lacking experimental data.

The objective of this project is to evaluate the possibility of extending performance codes to new materials based on databases developed from thermodynamic, atomistic, and first principles calculations. This project will create a prototype of a new generation of advanced fuel performance codes with inputs from state-of-the-art electronic-structure, atomic, and thermodynamic calculations. Pertinent input parameters will include material composition, temperature, density, closed porosity, surface roughness, fuel grain size, sintering temperature, and fractional cold work of the cladding. Development of this "first principles" based code will allow fuel development programs to reduce costly and time-consuming experimentation by transition to a selection process based on modeling, simulation, and analysis that will require only confirmatory irradiation testing.

Work Scope

This project will consist of two tasks:

- Dissect and analyze FRAPCON to identify physical inputs; rebuild the code into a form that can use materials databases developed from atomic-level, electronic-structure, and thermodynamic calculations; and apply sensitivity analyses to identify properties on which fuel performance is most dependent.
- Develop detailed databases of the U-Pu-O and U-Pu-N ternary systems and identify compositions that can meet key performance criteria.

NUCLEAR ENERGY RESEARCH INITIATIVE

Development of TRU Transmuters for Optimization of the Global Fuel Cycle

PI: John C. Lee, University of Michigan

Collaborators: Argonne National Laboratory

Project Number: 05-142

Project Start Date: March 2005

Project End Date: February 2008

Project Description

This project will develop advanced fuel cycles for the transmutation of transuranic (TRU) elements in irradiated nuclear fuel from light water reactor (LWR) power plants. The research will focus on developing fast-spectrum nuclear reactors that could efficiently transmute long-lived TRUs, thereby significantly reducing the radioactivity of the irradiated fuel. Researchers will evaluate diverse fuel cycles and energy production systems in a systematic, integrated approach to optimize the global fuel cycle. The project will study deployment of fast-spectrum transmuters together with LWRs and other advanced reactors to minimize risks associated with the disposal and storage of irradiated nuclear fuel, including the radiological toxicity, proliferation risk, and radiological dose of the irradiated fuel in underground repositories. Researchers will develop an equilibrium fuel cycle methodology to consistently compare the performance of LWR transmuters with that of other designs. A key objective of this research is to develop a simplified, analytical fuel cycle methodology that could provide physical insights into the overall performance of an integrated nuclear energy fuel cycle economy.

Work Scope

- Develop and optimize low conversion-ratio fast reactor transmuters.
- Apply LWR equilibrium cycle methodology to global power distribution calculations and fuel loading optimization.
- Evaluate diverse fuel cycle options with the DANESS code and enhance the model.
- Develop algorithms to optimize the global fuel cycle and map costs as a function of system parameters.
- Develop a simplified model to provide physical insights and guidance for evaluation of diverse fuel cycles.

NUCLEAR ENERGY RESEARCH INITIATIVE

The Adoption of Advanced Fuel Cycle Technology Under a Single Repository Policy

PI: Paul Wilson, University of Wisconsin-Madison

Collaborators: None

Project Number: 05-157

Project Start Date: March 2005

Project End Date: February 2008

Project Description

This project will develop models to study the impact of a policy that retains a single spent fuel storage repository (Yucca Mountain). An increasing amount of technology-driven analysis is being done to determine how reprocessing and separation technologies can improve the loading of Yucca Mountain while maintaining the same general design concept and licensing basis. However, little policy-driven analysis has been conducted to study the adoption of new reprocessing and separation technologies assuming only a single repository is constructed. Given the current mismatch between the legislative limit for capacity of Yucca Mountain and the expected spent fuel inventory from the existing reactor fleet, a single-repository policy would likely improve the economic attractiveness of advanced fuel cycle technologies. In addition, this project will study the relationship between fuel cycle advances driven primarily by repository performance and those advances necessary for a transition to a sustainable fuel cycle that is both economical and feasible. This project supports the Department of Energy's evaluation of additional geologic repositories for spent nuclear fuel beyond Yucca Mountain, which is one of the near-term goals of the Advanced Fuel Cycle Initiative.

Work Scope

This project consists of four primary tasks:

- Develop economic models to evaluate limited repository space for various scenarios.
- Define technical specifications for waste forms and packages with arbitrary contents, based on long-term repository performance.
- Develop fuel cycle modeling to track and relate waste forms to the nuclear energy produced by the particular reactor that produced it, and couple results to the economic model.
- Integrate these concepts and models into the DANESS code package (a framework for dynamic modeling of a national nuclear energy enterprise) to assess the consequences of this strategy on other long-term nuclear energy goals.

11. Nuclear Hydrogen Initiative: Project Abstracts

This new program element includes three research projects, all of which were awarded in FY 2005. The following abstracts present each project's participants, its goals and objectives, and a brief description of the proposed work.

The mission of the Nuclear Hydrogen Initiative is to exhibit hydrogen production technologies utilizing nuclear energy. The goal is to demonstrate hydrogen production compatible with nuclear energy systems through scaled experiments, and then to couple an engineering-scale demonstration plant with a Generation IV demonstration facility by 2017. Planned NERI research for FY 2005–FY 2007 related to this initiative includes projects that are associated with thermochemical cycles, high-temperature electrolysis, and reactor-hydrogen production process interface. Each of these research areas are discussed below.

- Thermochemical cycles: Specific realms of investigation under this research area are thermochemical cycles for nuclear application, such as sulfur-based cycles, calcium-bromide cycles, and alternative cycles; flowsheet methodology for comparison of thermochemical cycles; and high-temperature interface requirements for heat exchangers and materials. Planned research projects consist of laboratory-scale demonstrations of candidate processes for sulfur-iodine, hybrid sulfur, and calcium-bromide; alternative thermochemical process assessment; and enhancement of membranes to increase process efficiency. Projects will include materials testing, control system design, and cost evaluations.
- High-temperature electrolysis (HTE): This research area seeks to reduce the cost of manufacturing electrolytic cells and components and increase the useful lifetime of these components thereby producing hydrogen at the lowest possible cost. Research activities include cell and stack experiments; modeling of plant and cell dynamics; and design of plants beginning with a laboratory unit and later scaling up to include a pilot-scale experiment followed by demonstration and commercial units. Materials for cells, heat exchangers, and separations are also topics within this research area. Planned NERI projects include completing HTE cell and stack/module testing, completing the design and then assembling components for the integrated laboratory test unit by 2007, completing the design and fabrication of a pilot-scale experiment by 2011, and conducting tests for high-temperature heat exchanger and separations.
- Reactor-hydrogen production process interface: Research in this area includes process-side high-temperature heat exchanger (HX) design (including HX type, operating conditions, efficiency, and material qualification), implications of intermediate heat transfer loop on reactor operation (e.g., corrosion, isolation, connection), and design of support systems. NERI projects over the next three years will explore laboratory-scale HX development for a variety of high-temperature hydrogen production processes, including design, short and long-term materials testing, fabrication, and system support, which includes assessing the process, infrastructure, and facilities requirements for the pilot plant and the balance-of-plant design.

NUCLEAR ENERGY RESEARCH INITIATIVE

Directory of Nuclear Hydrogen Initiative Project Abstracts

NUCLEAR ENERGY RESEARCH INITIATIVE

The Sulfur-Iodine Cycle: Process Analysis and Design Using Comprehensive Phase Equilibrium Measurements and Modeling

PI: Mark C. Thies, Clemson University

Collaborators: University of Virginia, Savannah River National Laboratory, Aspen Technology, Inc.

Project Number: 05-006

Project Start Date: March 2005

Project End Date: February 2008

Project Description

Of the many thermochemical hydrogen cycles that have been proposed for the centralized production of hydrogen from nuclear power, the Sulfur-Iodine (S-I) cycle has been identified as one of the most promising and is of international interest. However, the S-I cycle involves complex, highly non-ideal phase behavior and reactions. Many of the performance projections associated with this technology are currently based on uncertain and incomplete data. Thus, thermodynamic measurements and physical property models for this cycle have been identified as basic research requirements for the successful development of a future hydrogen economy.

This project will be performed by an experienced team of academic, industrial, and DOE national laboratory experts who will assess the true potential of the S-I cycle. Work will focus on three research areas: thermodynamic measurements, physical property modeling, and process modeling. For the first time, researchers will measure comprehensive phase equilibrium data for the process-limiting Hydrogen-Iodine (H-I) dissociation section of the S-I cycle. The team will use a unique, integrated approach that will allow for highly efficient exploration as well as feedback among the three focus areas. That is, researchers will use data from initial property and process modeling to guide the selection of conditions for experimental measurement. Then, as measurements become available, property models will be refined and provided for the process modeling effort. Finally, updated process modeling results will be used to identify additional experiments that are most critical for minimizing the remaining process uncertainties.

Work Scope

- Obtain new, comprehensive phase equilibrium data for the S-I cycle over wide ranges of conditions that have not previously been investigated.
- Develop models for the phase behavior and properties of chemical systems critical to S-I cycle analysis for which no adequate models currently exist.
- Establish and disseminate a comprehensive and reliable process modeling capability for thermochemical cycle analysis.

Specific deliverables will include comprehensive phase equilibrium measurements for the H-I dissociation step of the S-I cycle, property models for all process streams, and an optimized flowsheet model for the S-I cycle.

NUCLEAR ENERGY RESEARCH INITIATIVE

Silicon Carbide Ceramics for Compact Heat Exchangers

PI: Dennis C. Nagle and Cila Herman, Johns Hopkins University Advanced Technology Laboratory (JHU-ATL)

Collaborators: None

Project Number: 05-032

Project Start Date: March 2005

Project End Date: February 2008

Project Description

In support of DOE's need for high-temperature, compact heat exchangers for the next generation of nuclear plants, this project will develop revolutionary, cellulose-derived-carbon (CDC), silicon carbide (SiC) materials and structures that are impervious to hot hydrogen gas. SiC materials offer a 10-fold improvement in thermal conductivity over nickel super-alloy, but existing manufacturing technology does not support the production of acceptable quality block materials. The unique CDC-SiC materials and manufacturing approach that will be used in the project will permit the development of net-shaped SiC structures and assemblies suitable for use in compact heat exchanger components.

In addition, this project team will develop glass-sealing technology that is critical to sealing residual porosity in the silicon carbide in order to make hydrogen-impervious seals. These seals have the potential for sealing between ceramic and metal components. Successful completion of this project will enable the development of a new generation of heat exchangers that are capable of continuous operation at temperatures over 900°C.

Work Scope

Following is the work scope of this project:

- Develop models to define the optimum channel configuration for SiC, including pressure drops and heat transfer rates related to the configuration and geometry.
- Synthesize materials to develop CDC-SiC materials for compact block-design heat exchangers.
- Characterize materials to define micro-structural material and thermal and mechanical properties, and to measure gas permeability.
- Manufacture a heat exchanger prototype.
- Test the heat exchanger to determine heat transfer capability and pressure drops.
- Compare experimental data to model predictions.

NUCLEAR ENERGY RESEARCH INITIATIVE

Molten Salt Heat Transport Loop: Materials Corrosion and Heat Transfer Phenomena

PI: Kumar Sridharan, University of Wisconsin

Collaborators: None

Project Number: 05-154

Project Start Date: March 2005

Project End Date: February 2008

Project Description

The next generation nuclear power plant (NGNP) is likely to be a high-temperature reactor utilizing graphite moderation with TRISO (isotropic coatings of three materials) fuel particles in either a matrix or pebble bed configuration. The NGNP will be designed to produce two energy products: electricity and process heat for hydrogen production. Producing process heat requires a suitable heat transport fluid such as high-pressure inert gas (helium or carbon dioxide) or molten fluoride salt.

Because the interface between the reactor and the hydrogen production system will likely involve long heat transfer paths at elevated temperatures, a heat transport working fluid that has superior heat transfer characteristics would be required. The heat transport fluid should (1) be chemically compatible with the surrounding structural materials, (2) have superior fluid-mechanical and heat transfer proprieties, and (3) have acceptable safety characteristics under normal and abnormal conditions. This project investigates the potential of molten salt as a possible transport fluid and also investigates the corrosion resistance of structural materials that would come into contact with the molten salt.

The objective of this project is to demonstrate that molten fluoride salt can be successfully implemented in a low-pressure intermediate heat exchange loop. This project will focus on researching molten fluoride salts as the process-heat transport medium and the corrosion compatibility of surrounding materials with this medium.

Work Scope

- Design, fabricate, and operate molten fluoride salt capsules in a flow loop under prototypical NGNP conditions using appropriate geometric scaling.
- Plan, design, and execute a series of corrosion compatibility experiments in the heat exchange loop in order to develop a database on the structural materials' behavior for a range of candidate materials that are being considered for the NGNP design.
- Document the observed corrosion effects and heat transport performance as integral and separate-effects data.

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