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Nuclear Energy Research Initiative

2005 Annual Report

DEPARTMENT OF ENERGY
UNITED STATES OF AMERICA

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Foreword

The Nuclear Energy Research Initiative (NERI) is at the core of a Federal effort to develop advanced nuclear energy concepts and technologies. This program focuses 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 and the production of hydrogen in this country. Currently funded NERI projects are directly related to other nuclear energy programs within the Office of Nuclear Energy (NE), including the Generation IV Nuclear Energy Systems Initiative, the Advanced Fuel Cycle Initiative, and the Nuclear Hydrogen Initiative. With its focus on applied nuclear energy research, the NERI Program has been realizing its goal of both developing advanced nuclear energy systems and providing state-of-the-art research concerning nuclear science and technology.

In addition to responding to the Nation's need for current nuclear energy research, NERI has helped to maintain and improve the nuclear research infrastructure in this country. While NERI research in the past has been funded through universities, laboratories, and industry partners, university involvement has been particularly important in promoting and maintaining a nuclear science and engineering infrastructure to meet future technical challenges. To further this mission, in 2004, NE decided to refocus NERI to exclusively fund research led solely by the Nation's universities, with national laboratories and industry partners providing valuable contributions as collaborators. This focus enables educational institutions across the country to stay at the forefront of nuclear science research, renews student interest in pursuing degrees in nuclear engineering and related sciences, and integrates further the Nation's universities with the research efforts and initiatives of the Department of Energy.

The *Nuclear Energy Research Initiative 2005 Annual Report* summarizes the results of the 22 fiscal year (FY) 2002 NERI projects completed in 2005, the research progress of the 35 projects initiated in FY 2005, and the objectives and work scope of the 24 newly awarded FY 2006 research projects. The final summaries for the projects initiated in FY 1999 through FY 2001 can be found in the NERI 2002, 2003, and 2004 Annual Reports, respectively. This report disseminates the results of NERI-sponsored research to the wide R&D community to spur yet more innovation, ensuring a bright future for nuclear energy in the United States and the world.

Dennis R. Spurgeon

Dennis R. Spurgeon, Assistant Secretary
for Nuclear Energy
U.S. Department of Energy

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Nuclear Energy Research Initiative

1. Overview

The Nuclear Energy Research Initiative

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 (PCAST). 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 its use.

NERI, managed and funded by DOE's Office of Nuclear Energy, is a national, research-oriented initiative focused on innovation and competitiveness. The purpose of NERI is to sponsor research and development (R&D) in science and technology that addresses the principal barriers related to the growth of nuclear energy in the United States. This initiative has helped 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. 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, thereby enabling the United States to maintain a competitive position in nuclear science and technology. A link to the program website can be found at http://neri.ne.doe.gov/.

To achieve the Nation's long-range goal of establishing nuclear energy as a viable and expandable option, 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 2005 Annual Report

The *Nuclear Energy Research Initiative 2005 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, the International Nuclear Energy Research Initiative (I-NERI), and the programs' evolving research focus. Section 3 provides a discussion of NERI's current work scope. Section 4 highlights the major accomplishments of the NERI projects and provides brief summaries of the NERI research efforts completed in 2005. Section 5 provides a discussion on the impact NERI has had on U.S. university nuclear programs. Section 6 presents final progress reports for each of the 22 FY 2002 NERI projects. Research objectives, progress made over the last year, and results obtained upon project completion are described for each project. Some of these efforts are still ongoing under approved no-cost project extensions.

Section 7 presents progress reports for each of the 35 ongoing FY 2005 projects and abstracts for the 24 newly awarded FY 2006 NERI projects in their corresponding focus area. These abstracts include a brief description of each project's objectives, its proposed work scope, and its participants.

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

2. Program History

In January 1997, PCAST was tasked by President Clinton 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 the Republic of Korea, France, and a third collaboration involving Argonne National Laboratory and a consortium of 10 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 2005, a new implementing agreement was signed with Japan and new projects were initiated with Brazil, Japan, and the Republic of Korea. Similar international agreements with the Republic of South Africa and the United Kingdom are being considered.

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. A separate 2005 annual report is being published covering the research effort conducted under the I-NERI program during the past fiscal year.

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, several Federal directives have influenced the focus of NERI research activities:

The Long-Term Nuclear Technology Research and Development Plan. 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, 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, the *National Energy Policy* 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.

The Technology Roadmap for Generation IV Nuclear Energy Systems. In September 2002, NERAC issued the *Draft Technology Roadmap for Generation IV Nuclear Energy Systems*. In coordination with the 10 member countries of the Generation IV International Forum (GIF), 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 (Generation IV), the Advanced Fuel Cycle Initiative (AFCI), and the Nuclear Hydrogen Initiative (NHI), 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 are now allowed to serve as the principal investigators 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.

3. 2005 Work Scope

In FY 2005, NERI continued its R&D focus to address research requirements introduced in the *National Energy Policy*. Work during this year included the completion of research projects initiated in FY 2002 under the original NERI work scope and the first year of research on projects begun in FY 2005 under the new NERI focus.

The FY 2002 project research areas included efforts in advacnced nuclear energy systems, advanced nuclear fuel/fuel cycles, and fundamental science. A description of these research areas is provided in the introduction of Section 6.

Safety, non-proliferation, and waste management considerations are intrinsic to all three research topics, especially for the advanced nuclear energy systems and advanced fuels/fuel cycles. Thus, they become selection criteria across all three focus areas, and do not in themselves constitute focus areas.

In addition to the work performed on the FY 2002 projects, work was initiated on the 35 projects awarded in FY 2005. These projects fall in line with NERI's new focus to conduct applied nuclear energy research related to NE's R&D programs, including Generation IV, AFCI, and NHI. These three initiatives will develop proficient and efficient technologies for future electrical power production, hydrogen production, and other energy conversion systems and will continue to contribute to the revitalization of nuclear technology in our nation.

Generation IV is developing and demonstrating advanced nuclear energy systems that will meet future needs for secure, safe, sustainable, economic, clean, and proliferation-resistant energy.

AFCI is developing advanced fuels and spent fuel treatment and transmutation technologies 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. A significant change in the NERI Program is that projects are now led by universities, with national laboratories and industry playing a supporting role. To facilitate this transition, NE hosted the Advanced Reactor, Fuel Cycle, and Energy Products Workshop for Universities in March 2004 and in June 2005 to indoctrinate universities into the new NERI program. Section 7 describes the work scope associated with these NERI research areas in more detail and presents progress reports for the FY 2005 projects, along with brief abstracts of the FY 2006 projects that were recently awarded under each initiative.

The following graphic (Figure 1) summarizes the key features of the NERI program in 2005.

Figure 1. Key features of the NERI Program in 2005.

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 these 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.

Similar progress was made in FYs 2000, 2001, and 2002 with an additional 47 NERI projects awarded to universities, national laboratories, and industry research organizations. The total cost of these 47 research projects for the three-year period was approximately $58 million. These projects involved 20 U.S. universities, 10 national laboratories, 18 privates businesses, and seven foreign R&D organizations. In FY 2003 and 2004, no new NERI projects were awarded; however, research funding continued for the previously awarded FY 2001 and 2002 NERI projects.

In 2005, scientific and technical development advanced through the completion of the remaining 22 research projects that began in FY 2002. Brief summaries of the final results of the FY 2002 projects are presented in Section 6 of this report.

Funding History

Funding for NERI in FY 1999 through FY 2004 was appropriated annually by Congress in the Energy and Water Development Appropriations Act as a separate budget line item. The following chart (Table 1) provides a history of this NERI funding and its distribution.

Figure 2 illustrates the cumulative total of research projects awarded in FY 1999 through FY 2002 in the four major R&D areas. Over $110 million was awarded to fund

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	$21.4 million for projects begun in FY 1999 and FY 2000
FY 2002	$22 million	$10 million for 24 new awards	$12 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	$11 million for projects begun in FY 2002

Table 1. NERI funding and distribution history through FY 2004.

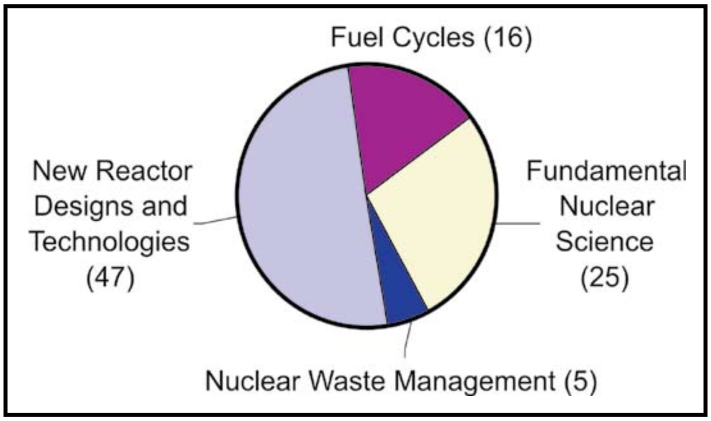


Figure 2. Division of FY 1999 – 2002 NERI projects by R&D area.

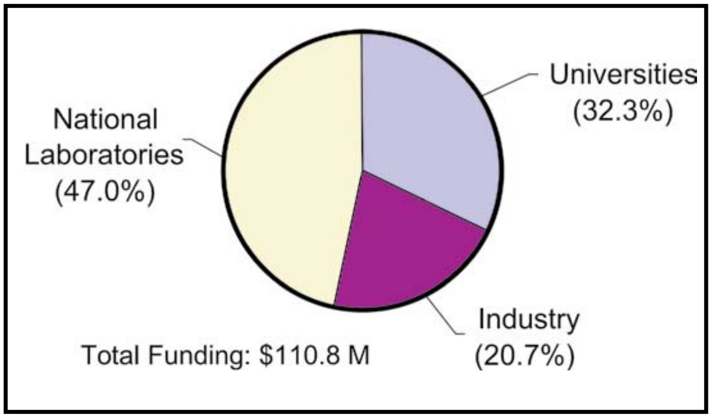


Figure 3. Distribution of NERI research funds in FY 1999 – 2004.

these NERI research projects. Figure 3 shows the total distribution of this funding among the national laboratories, U.S. universities, and industry.

DOE does not fund foreign participants 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, foreign participation in NERI projects is contingent upon DOE approval.

In FY 2005, NERI was refocused to provide more direct support to the goals and objectives of the Generation IV, AFCI, and NHI programs. In addition, greater emphasis was placed on university research program involvement. A total of 35 projects were awarded, involving 28 U.S. universities, seven national laboratories, two U.S. businesses, and four foreign research organizations. The total cost of these research projects for the three-year period was approximately $20.6 million. Over $10.4 million in funding was provided: $2.5M from congressional appropriations for NERI and the remainder from FY 2004 and 2005 contributions made by the Generation IV, AFCI, and NHI programs. Also in 2005, 24 new FY 2006 project awards totaling over $12 million in funding were announced to 17 universities who responded to the FY 2005 NERI solicitation. Progress made over the first year of the FY 2005 projects and abstracts related to the newly awarded FY 2006 projects are presented in Section 7 of this report. Figure 4 shows the distribution of these 59 projects by R&D program area. Figure 5 illustrates the funding distribution by university, national laboratory, and industry participants. The specific contributions made by each of the three program areas to fund these projects include $11.8M from Generation IV, $16.1M from AFCI, and $4.5M from NHI.

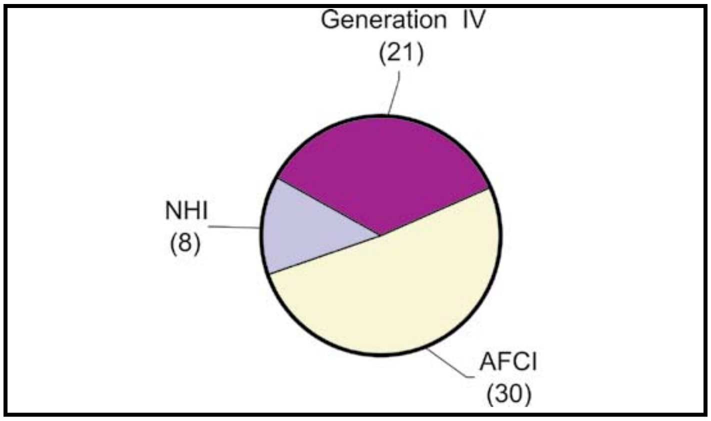


Figure 4. Division of new NERI projects by R&D program area since FY 2005.

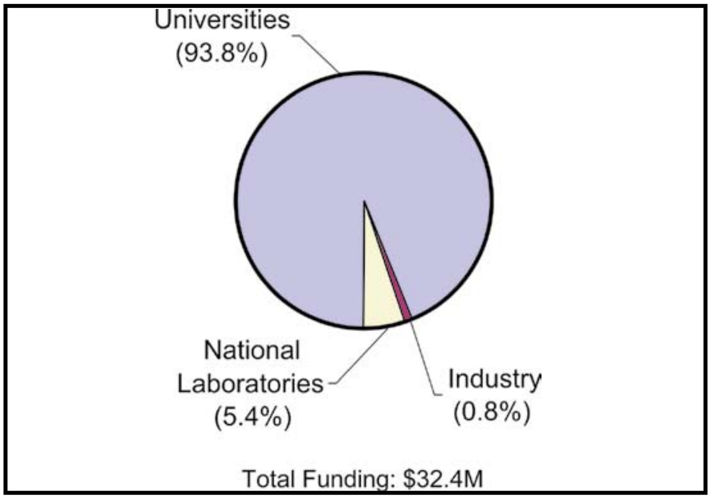


Figure 5. Funding distribution for new NERI projects by participant since FY 2005.

NERI Participants

NERI research participants in 2005 included 34 U.S. universities, 8 national laboratories, 11 private businesses, and 7 foreign organizations. The participating organizations are provided in the following tables.

U.S. Universities

Arizona State University
Boise State University
Colorado School of Mines
Clemson University
Georgia Institute of Technology
Illinois Institute of Technology
Iowa State University
Johns Hopkins University
Kansas State University
Massachusetts Institute of Technology
North Carolina State University
Oregon State University
Pennsylvania State University
Purdue University
Rensselaer Polytechnic Institute
SUNY at Stony Brook
Texas A&M University
The Ohio State University
University of Arizona
University of California-Berkeley
University of California-Santa Barbara
University of Cincinnati
University of Florida
University of Illinois
University of Michigan
University of Missouri-Rolla
University of Nevada-Las Vegas
University of Notre Dame
University of South Carolina
University of Tennessee
University of Virginia
University of Wisconsin-Madison
Utah State University
Washington State University

U.S. DOE National Laboratories

Argonne National Laboratory
Brookhaven National Laboratory
Idaho National Laboratory
Los Alamos National Laboratory
Oak Ridge National Laboratory
Pacific Northwest National Laboratory
Sandia National Laboratories
Westinghouse Savannah River Technology Center

Industrial Organizations

Electric Power Research Institute
Entergy Nuclear, Inc.
Florida Power & Light
Fluent, Inc.
Framatome ANP, Inc.
General Electric Global Research Center
General Atomics
Pacific Southern Electric & Gas
Special Metals, Inc.
Westinghouse Electric Company LLC
(n,p) Energy, Inc.

International Collaborators

Imperial College of Science, Technology, and Medicine (United Kingdom)
Japan Nuclear Cycle Development Institute (Japan)
The Royal Institute of Technology (Sweden)
PBMR, Ltd. (South Africa)
University of Tokyo (Japan)
Belgian Nuclear Research Centre (SCK/CEN)
Hokkaido University (Japan)

FY 2002 Project Completions

This year marked the scheduled completion of the remaining 22 FY 2002 NERI projects. Based on noted accomplishments, it is clear that NERI's stated goals and objectives are being met. These 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, Generation IV, AFCI, and NHI, have helped to revive the Nation's leadership role in international nuclear R&D. The technology advances will support the United States in regaining a competitive position in future energy markets.

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. Table 2 identifies each of the 22 FY 2002 projects completed in 2005. More detailed information on these projects is contained in the project summaries located in Section 6 of this report.

Table 2. FY 2002 NERI projects scheduled for completion.

Project Number	Title	Lead Organization
02-005	Engineering and Physics Optimization of Breed and Burn Fast Reactor Systems	Massachusetts Institute of Technology
02-018	Evaluation of Integral Pressurizers for Generation IV PWR Concepts	Oak Ridge National Laboratory
02-042	Oxidation of Zircaloy Fuel Cladding in Water-Cooled Nuclear Reactors	The Pennsylvania State University
02-060	Neutron and Beta/Gamma Radiolysis of Supercritical Water	Argonne National Laboratory
02-065	Coupling of High-Temperature, Lead-Cooled, Closed Fuel Cycle Fast Reactors to Advanced Energy Converters	Argonne National Laboratory
02-068	Experimental Verification of Magnetic Insulation of Direct Energy Conversion Fission Reactors	Sandia National Laboratories
02-075	Innovative Approach to Establish Root Causes for Cracking in Aggressive Reactor Environments	Pacific Northwest National Laboratory
02-081	An Innovative Transport Theory Method for Efficient Design, Analysis, and Monitoring of Generation IV Reactor Cores	Georgia Institute of Technology
02-098	Advanced Extraction Methods for Actinide/Lanthanide Separations	University of Florida
02-103	Innovative, Low-Cost Approaches to Automating QA/QC of Fuel Particle Production Using On-line Nondestructive Methods for Higher Reliability	Pacific Northwest National Laboratory; Applied Research Associates
02-110	Design of Radiation-Tolerant Structural Alloys for Generation IV Nuclear Energy Systems	University of Madison, Wisconsin
02-113	Model-Based Transient Control and Component Degradation Monitoring in Generation IV Nuclear Power Plants	University of Michigan
02-131	Improving the Integrity of Coated Fuel Particles: Measurement of Constituent Properties of SiC and ZrC, Effects of Irradiation, and Modeling	Oak Ridge National Laboratory
02-146	Enhanced Control of PWR Primary Coolant Water Chemistry Using Selective Separation Systems for Recovery and Recycle of Enriched Boric Acid	University of Nevada, Las Vegas
02-160	Centralized Hydrogen Production from Nuclear Power: Infrastructure Analysis and Test-Case Design Study	Savannah River National Laboratory
02-174	Near-Core and In-Core Neutron Radiatior Monitors for Real-Time Neutron Flux Monitoring and Reactor Power Level Measurements	Kansas State University
02-180	Enhanced Thermal Conductivity Oxide (ECO) Fuels	Purdue University
02-189	Use of Solid Hydride Fuel for Improved Long-Life LWR Core Designs	University of California, Berkeley
02-190	Development of a Supercritical Carbon Dioxide Brayton Cycle: Improving PBR Efficiency and Testing Material Compatibility	Idaho National Laboratory
02-195	Development of Advanced Methods for Pebble-Bed Reactor Neutronics: Design, Analysis, and Fuel Cycle Optimization	Idaho National Laboratory
02-196	Hydrogen Production Plant Using the Modular Helium Reactor	General Atomics
02-207	Nuclear Reactor Power Monitoring Using Silicon Carbide Semiconductor Radiation Detectors	The Ohio State University

5. NERI and U.S. Universities: Advancing the Goals of Nuclear R&D Programs

Focus on Universities

One of NERI's long-term goals is to improve the nation's nuclear science and engineering infrastructure 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 between 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 NE R&D programs, and benefiting future nuclear industry and national laboratory endeavors by training the next generation of nuclear scientists. Funding creative research ideas at the nation's universities and colleges also helps solve important issues 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.

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, 44 U.S. universities have received research awards as lead investigators or collaborators. Of the 93 FY 1999 through FY 2002 projects awarded through NERI, 75 percent involved U.S. colleges and universities. The FY 2005 and FY 2006 NERI projects have required 100 percent university-led research efforts, involving 33 different U.S. universities and colleges.

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 related to the AFCI, Generation IV, and NHI programs, NERI's activities are coordinated with other relevant DOE energy research programs in the Office of Science, the Office of Energy Efficiency and Renewable Energy, and the 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.

Student Participation

One great success of NERI and other DOE programs is the significant increase in nuclear-related educational opportunities at the universities. Universities have benefited from increased research dollars which have served as incentives for new student recruitment. Partly, as a result of this involvement, student interest in nuclear engineering is being revitalized. In 1998, only 500 students were enrolled in U.S. universities seeking degrees in nuclear engineering; today, approximately 1,800 students are enrolled in nuclear engineering programs.

This student participation includes all levels—undergraduates and those students performing masters and doctorate level work. Figure 6 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

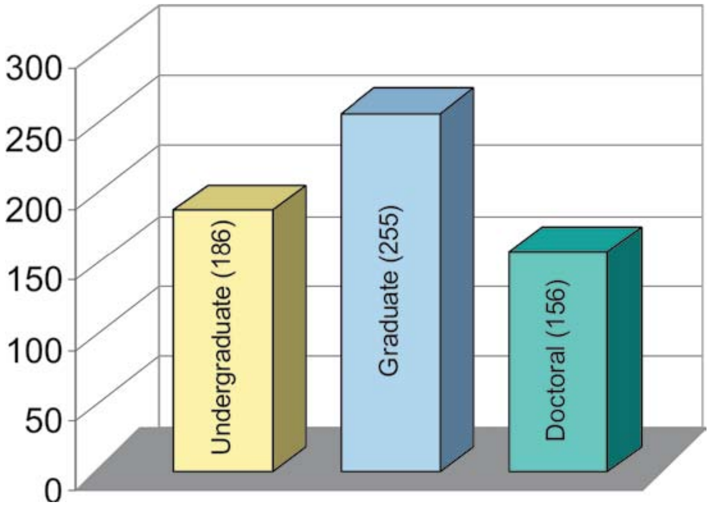


Figure 6. NERI Student Participation Profile – FY 1999 through FY 2005.

research projects. In FY 2005 alone, 109 undergraduate, 130 graduate, and 75 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 contribute to the future growth of the nuclear power industry. Figure 7 provides a map and complete listing of the 44 universities and

UNIVERSITY KEY

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

Figure 7. Locations of universities and colleges participating in NERI projects.

6. FY 2002 NERI Project Summaries

This section presents detailed summaries of the completed FY 2002 NERI projects. Details include the research projects' objectives and their completed accomplishments. In the event that these projects have received a no-cost-extension, planned accomplishments are also described. The following is a description of the three research areas associated with the FY 2002 projects and summaries of the 22 FY 2002 projects by research area.

Advanced Nuclear Energy System Projects

This research area includes 11 research projects focused on 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 development of 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, or systems that increase energy extraction from the utilization of plutonium and other actinide isotopes generated in the fuel.

Advanced Nuclear Fuel/Fuel Cycle Projects

This research area includes six NERI research projects focused on providing 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 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.

Fundamental Nuclear Science Projects

This research area includes five NERI projects focused on addressing the long-term R&D goals of developing new technologies for nuclear energy applications. 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 selected 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 Completed FY 2002 Projects

Advanced Nuclear Energy Systems Projects

Advanced Nuclear Fuels/Fuel Cycles Projects

Fundamental Nuclear Sciences Projects

Advanced Nuclear Energy Systems Projects

NUCLEAR ENERGY RESEARCH INITIATIVE

Engineering and Physics Optimization of Breed and Burn Fast Reactor Systems

PI: Michael Driscoll, Massachusetts Institute of Technology

Collaborators: Idaho National Laboratory, University of Nevada, Las Vegas

Project Number: 02-005

Project Start Date: September 2002

Project End Date: September 2005

Research Objectives

The goal of this project was to develop a practical implementation of breed and burn operation in hard spectrum gas cooled fast reactors (GCFR). "Breed and burn" (B&B) refers to cores in which steady-state reload fuel has a significantly lower enrichment than that required to sustain criticality. Although the general breed and burn concept dates back to the late 1950s, interest has been sporadic because of unresolved challenges, such as the need for a large, low-leakage core with minimal non-heavy-metal fuel diluents and the need for high power density and burnup to accelerate and sustain fissile buildup in order to quickly achieve an equilibrium fuel cycle.

In this project, 5 $^{w}/_{o}$ U^{235} was evaluated as a B&B gas-cooled reactor reload fuel, compared to the startup core which uses 8 $^{w}/_{o}$ average and 10 $^{w}/_{o}$ maximum enrichment. Exceptional neutronic efficiency allows sufficient plutonium to build up to sustain burnup to very high values exceeding 150 Mwd/kg. In the ideal case, recycling is not required to achieve natural uranium utilization that is significantly higher (by a factor of three or more) than current light water reactor (LWR) units. High utilization leads to competitive fuel cycle economics and the practicality of commercializing fast reactors without first deploying fuel reprocessing and recycling facilities.

Research Results

In the final year of this project, the research team devised a successful version of the breed and burn GCFR concept. Table 1 summarizes the final design features. The high volume fraction fuel needed used uranium carbide (UC) fuel, instead of the inordinately expensive uranium nitride (UN^{15}), and motivated researchers to use an innovative vented tube-in-duct fuel assembly design. To keep circulator power tolerable, a large coolant temperature rise was required across the core. This requirement dictated the use of a conventional Rankine cycle. The result was that the balance-of-plant is an entirely conventional design using proven British AGR and US/German HTGR technology. Thus, the project's focus evolved to center on core and fuel design and performance assessment.

In addition, researchers on this project also successfully transferred the shutdown cooling loop design for a helium-cooled cercer core (developed by MIT under an I-NERI contract managed by ANL and CEA) to the B&B application.

During previous years, the project's efforts were devoted to the selection of viable options for consideration. Most of the plant-related issues followed directly from the requirements to provide competitive economics and assure ultra-reliable shutdown and emergency core cooling within the stringent constraints imposed by the physics performance of a B&B reactor. Due to neutronics constraints, researchers concluded that the only practical fuel options were metal clad UN^{15} or UC fuel. Both of these options dictated the use of helium as the coolant to avoid fuel-coolant chemical reactions. The team demonstrated a neutronically feasible core design using pin-type UN^{15} fuel. Figure 1 shows the reactivity history of a core in which one-sixth is refueled during each shutdown. Because UN^{15} is cost-prohibitive due to separation of the ^{15}N isotope, subsequent research focused on determining suitable design parameters for UC fuel.

Because of the high fuel volume fraction required, researchers chose an indirect Rankine cycle. This cycle allows a high core coolant temperature rise and a lower mass flow rate, pressure drop, and circulator power than other options, such as the supercritical CO_2 Brayton power conversion system under evaluation at MIT for Generation IV applications. This design decision, made late in Year 2, removed a wide variety of balance-of-plant issues from the agenda. The remaining issue of providing ultra-high reliability shutdown decay heat removal was resolved in the final year by incorporating design features developed for a helium-cooled cercer core. Fuel venting has considerably ameliorated fuel endurance challenges principally associated with high-temperature clad creep.

In addition, this project showed that spent B&B GFR fuel could be re-used as B&B GFR reload fuel by blending it with natural or depleted uranium after minimal, non-separative AIROX-type processing. It also showed that spent B&B fuel could be used in a PWR after 50/50 blending with 4.4 $^{w}/_{o}$ U-235 UO_2. Laboratory studies confirmed that it was advantageous to use CO_2 in place of air as the oxidizing medium and to use powdered graphite as the reductant. As LWR fuel, the accumulation of Sm-149 fission product acts as a self-provided burnable poison.

Planned Activities

Work on this project was completed in September 2005.

The principal need for future R&D in support of this concept is in the area of analytical and experimental qualification of fuel assembly design and confirmation of clad and fuel materials endurance under core fluence and temperature conditions.

Core	Design	Comments:
Fuel:	UC	UO_2 not viable neutronically Reaction of CO_2 with UC precludes its use as coolant
Clad:	ODS	ODS may be able to resist creep adequately up to ≈ 750°C
Configuration:	Tube-in-Duct; Vented; Orificed	Lower fuel T than pin-type at increased fuel fraction; venting eliminates ΔP across clad
Coolant:	He at 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 Peak clad T=716°C	Orificing reduces circulator power by factor of ≈ 2
Fuel Management:	5 $^{w}/_{o}$ U-235 reloads Startup Core: 8 $^{w}/_{o}$ avg., 10 $^{w}/_{o}$ max.	6 radial zones; 1/6th of core replaced each fueling
Burnup:	150 MWd/kg over 18 EFPY	assembly ρ > 0 @ ≈ 20 MWd/kg core ρ peaks @ ≈ 80 MWd/kg
Plant		
Power Cycle:	Rankine 2,400 MW_{th}	Allows ≈ 380°C ΔTc across core, which reduces coolant flow rate, hence circulator power
Reactor Vessel:	Prestressed Concrete Reactor Vessel (PCRV)	PCRV is proven in GCR and HTGR service (but at lower P)
Shutdown Cooling System: (combined shutdown & emergency)	• 4 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 ≥ 35 atm (15 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 after ~24 hrs

Table 1. Design features of preferred final version of breed and burn GCFR concept.

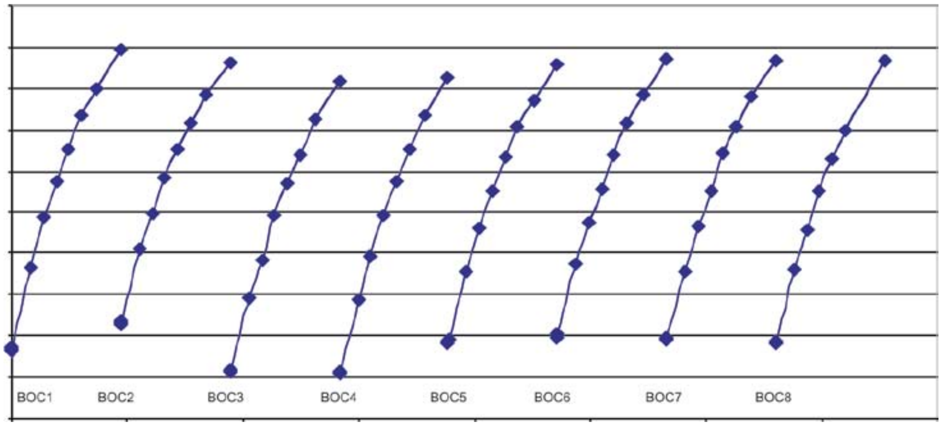


Figure 1. Reactivity history of successive B&B core reloads.

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: March 2006

Research Objectives

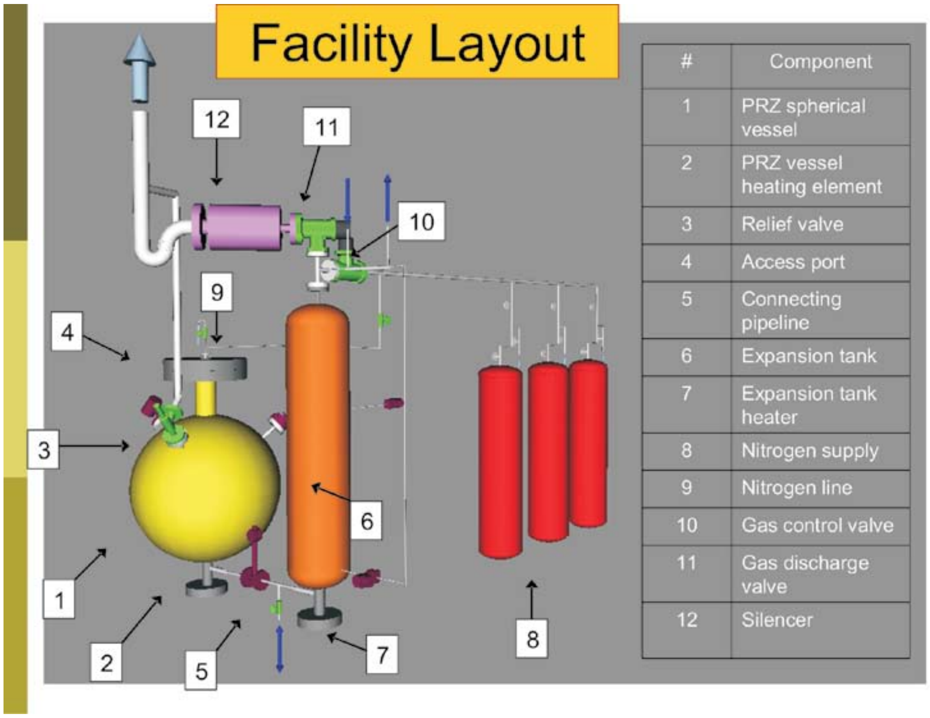
The objective of this project is to build a knowledge base that will enable designers to better evaluate possible pressurizer designs for integral primary system reactors (IPSR). As part of this process, researchers are mapping the performance of integral pressurizers as a function of certain design parameters (gas content, vapor volume, and interface with the primary system). Based on a detailed analysis of pressurizer performance, researchers proposed new design solutions to simplify the reactor system and allow it to withstand more severe accident scenarios. The primary goal of this project is to study the reaction of the pressurizer to dynamic transients using two types of working gases: steam and a steam-nitrogen mixture.

Research Progress

Researchers completed the test facility design during this reporting period. The Pressurizer Dynamic Test Facility (Figure 1), which represents the Generation IV International Reactor Innovative & Secure (IRIS) reactor internal pressurizer volumetrically scaled down by a factor of 314, is designed to conduct dynamic tests on pressurizers with steam or a steam-nitrogen mixture as the working fluids. Some specific features, such as shallow liquid phase, large dome surface area, and large gas-to-liquid ratio, may have a unique effect on the reaction of the pressurizer to dynamic transients.

Researchers have completed fabrication of the pressurizer and expansion vessels along with procurement of the associated instrumentation and controls. The enclosure and mounting of the high-pressure control valves is currently underway at the vendor location. The team has also set up a computer-controlled instrumentation and control system with appropriate logic. The nitrogen and discharge valves, labeled 10 and 11 in Figure 1, will require precise tuning in order to perform their functions of programmed flow control. Target flow rates were programmed and control algorithms for the facility were developed.

Along with the fabrication effort, detailed operating procedures were developed for initialization, heat up, and shutdown of the pressurizer facility. In view of the small size of the expansion tank, it was necessary to develop a



#	Component
1	PRZ spherical vessel
2	PRZ vessel heating element
3	Relief valve
4	Access port
5	Connecting pipeline
6	Expansion tank
7	Expansion tank heater
8	Nitrogen supply
9	Nitrogen line
10	Gas control valve
11	Gas discharge valve
12	Silencer

Figure 1. Pressurizer dynamic test facility.

method for letting out part of the water volume during heat up and an interactive method was developed using the control system.

Two different transients will be studied during testing in the facility: insurge (incoming or feeding) and outsurge (outgoing or bleeding) time variable water flows to and from the pressurizer. The character of these flows is based on preliminary analyses of the IRIS pressurizer, where the magnitude, time dependence, and duration of transients are defined. In general, these insurge and outsurge flows are typical for category II reactor events and are used as pressurizer design parameters.

In support of the experimental phase of the project, Westinghouse developed and tested an input data deck for the RELAP5 code describing the test facility. They have conducted the following preliminary runs:

- Manometric Test. Preliminary runs are performed to check whether the hydraulic part of the input deck is set up properly. The system is set up at t=0 s with a collapsed liquid level in the filling tank higher than the one in the pressurizer. The control valves are kept closed up to t=60 s and then are both opened. The hydraulic resistance of the perforated plate is reduced in order to emphasize the level oscillations in the system.
- Perfect Insurge. Pressurizer inflow is simulated by means of a time-dependent junction, which implements the injection flow profile developed in the previous phase of the project. The levels are measured from the bottom of the cylindrical part of the pressurizer and should envelop all the levels of interest during the insurge events.
- Nitrogen-Driven Insurge. Preliminary inflow runs are performed with an idealized nitrogen flow injection profile into the filling tank. The primary purpose is to compare the implemented nitrogen-injection profile and the corresponding water-injection profiles in order to get some preliminary information regarding the system response to a gas-driven injection for system control.

Planned Activities

Delivery of the facility from the vendor is expected in the near term. A no-cost extension of the original project end date from September 2005 to March 31, 2006 was granted in order to complete the test facility. Connection of utilities and instrumentation, as well as initial system checkout, remain to be completed once the equipment is delivered to the test location. Performance of the testing program will follow (under separate funding).

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: Oregon State University

Project Number: 02-065

Project Start Date: September 2002

Project End Date: September 2005

Research Objectives

The objectives of this project were 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 in which supercritical carbon dioxide (S-CO_2) is the working fluid.

Benefits of this reactor design include:

- Sustainability and closed fuel cycle benefits of a fast neutron spectrum core
- Passive safety of molten lead primary coolant
- Autonomous operational benefits of a fast spectrum core enhanced by lead coolant
- Economic advantages of modular construction, factory fabrication, full-transportability, and simplification
- Natural circulation heat transport at power levels in excess of 100 percent

The advantages of a S-CO_2 Brayton cycle advanced power conversion system include:

- Significantly improved cycle efficiency (45 percent at 578°C turbine inlet temperature) relative to the Rankine water/steam cycle operating at the same reactor core outlet temperature
- A reduced plant footprint due to fewer, simpler, and smaller sized secondary side components
- 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 an extensive nuclear infrastructure or a base of nuclear expertise. These nations will benefit from the deployment of proliferation-resistant, autonomous, passively safe, modular plants of small to medium power rating 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 an S-CO_2 Brayton cycle. They have shown that such a coupling is indeed feasible and that the core outlet and 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 a suite of analysis computer codes for the plant's conceptual design, control strategy development, and safety evaluation. One computer code modeled the coupled steady state behavior of 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 modeled the behavior of turbomachinery for use with S-CO_2 and analyzed optimal designs for the S-CO_2 turbine and compressors (i.e., number of stages and stage dimensions) to maximize the cycle efficiency. The researchers optimized the conceptual design of the lead-to-S-CO_2 heat exchangers so they could 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.

Researchers 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 constant core outlet temperature and without the operation of control rods. Researchers also developed new core designs with improved discharge burnup and reduced power peaking. They performed computational fluid dynamics analyses of multidimensional temperature and velocity fields 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. They then incorporated the resulting stress models into the conceptual design code to design the compressors and turbine. They investigated the consequences of supporting the reactor vessel, along with its massive load of lead and surrounding guard vessel, with a bottom support skirt versus hanging from above on a structural ledge. The method of support affects reactor behavior during postulated earthquakes and impacts vessel thickness. In addition, they developed and analyzed the performance of an internal structural barrier to protect the reactor vessel from the thermo-mechanical effects of changes in the lead temperatures during operational cycles.

During the project's final year, researchers developed a dynamics computer code for the coupled LFR-S-CO_2 Brayton cycle plant which simulates the time dependent and off-design behavior of the Brayton cycle compressors and turbine as well as the compact recuperators and cooler with greater realism and accuracy than other existing dynamics code for analyzing the transient behavior of this class of nuclear power plant. Researchers investigated alternative strategies for controlling the S-CO_2 Brayton cycle and enabling the LFR autonomous load following behavior. They ran the plant dynamics code to simulate normal operation of the plant and to analyze autonomous load following capabilities as well as behavior during several postulated accidents. In one analysis, calculations for a loss-of-heat sink accident without reactor scram demonstrated that reactor power autonomously shut down to decay heat levels due to passive reactivity feedback. It also showed that system temperatures remained within

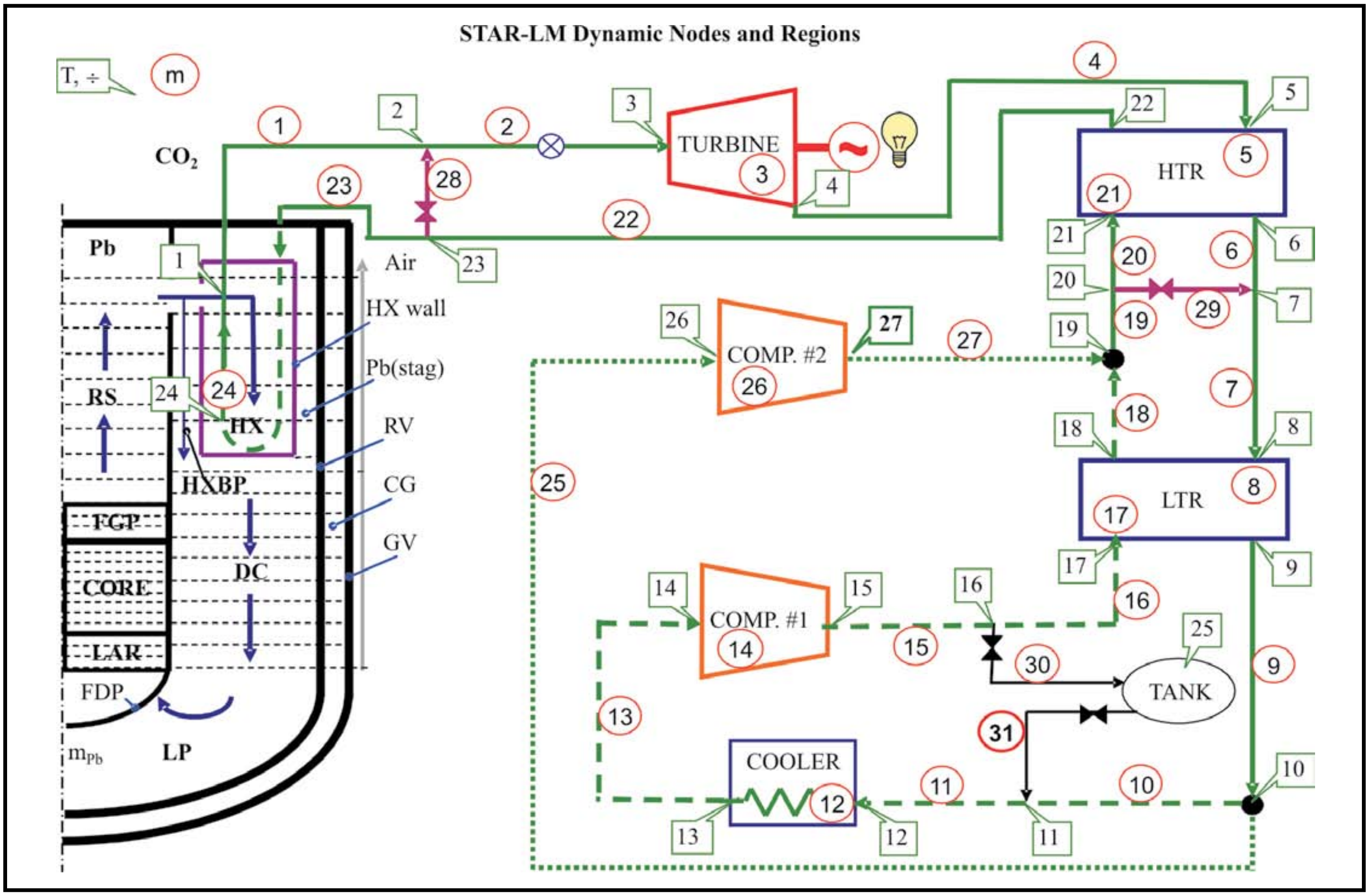


Figure 1. Representation of the STAR-LM-S-CO_2 Brayton cycle nuclear power plant as modeled using the plant dynamics computer code.

acceptable accident limitations. Figure 1 presents a schematic of the nuclear power plant as it was simulated using the plant dynamics computer code, including spatial points and regions where dynamic equations for state variables are solved.

Researchers investigated the ability to increase the STAR-LM reactor power rating in a fixed geometry by injecting noncondensable gas above the core to further enhance the natural circulation flowrate of the lead coolant. They found that the nominal operating power can be increased significantly and that the lead coolant flow and heat transport remain stable under both single-phase and two-phase natural circulation conditions.

Researchers applied an economics model to demonstrate that the higher efficiency achieved with the S-CO_2 Brayton cycle significantly enhances the net present value of the plant relative to a plant in which the LFR is coupled to a Rankine saturated steam cycle. This improved economics resulted from the greater revenue that can be realized from the sale of electricity over the plant's lifetime.

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 significant increase in plant efficiency might be achieved through the application of MHD power conversion, but that its realization would require overcoming significant technological challenges. 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

This project has been completed. Through journal articles and international conference proceedings, researchers will describe the plant dynamics computer code for the coupled LFR-S-CO_2 Brayton cycle plant. They will also explain the use of this code in analyzing the operation of the coupled LFR-S-CO_2 Brayton cycle plant and the plant's performance during postulated accident scenarios as well as other analyses performed during this project.

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 the large internal voltage gradients created during the conversion process. The goal 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 up to several million volts. This project has three specific objectives:
1) to fabricate and demonstrate the operation of two prototype DEC devices—the fission electric cell and the magnetic collimator, 2) to develop the theory of direct energy conversion, and 3) to establish supporting models and analytical tools.

In particular, this project will demonstrate that an applied external magnetic field from a central solenoid can reliably guide high-energy ions representing fission fragments and electrons to the DEC collectors. 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 be operated at the high voltages required for high conversion efficiencies and withstand the harsh conditions imposed.

Research Progress

To accomplish the program objectives, the researchers have down-scaled the fission fragment magnetic collimator reactor (FFMCR) system to the level of a non-fission prototype that allows experimental analysis of electromagnetic components. The final design of the FFMCR prototype is shown in Figure 1. The prototype represents one-half of the actual FFMCR system and is approximately 14 times smaller. The reactor simulates fission fragment motion using a high-energy helium-ion beam scattered by a primary copper-target. A voltage level of 1.0 MV is set as a limit due to safety and performance constraints enforced by smaller dimensions of the FFMCR prototype.

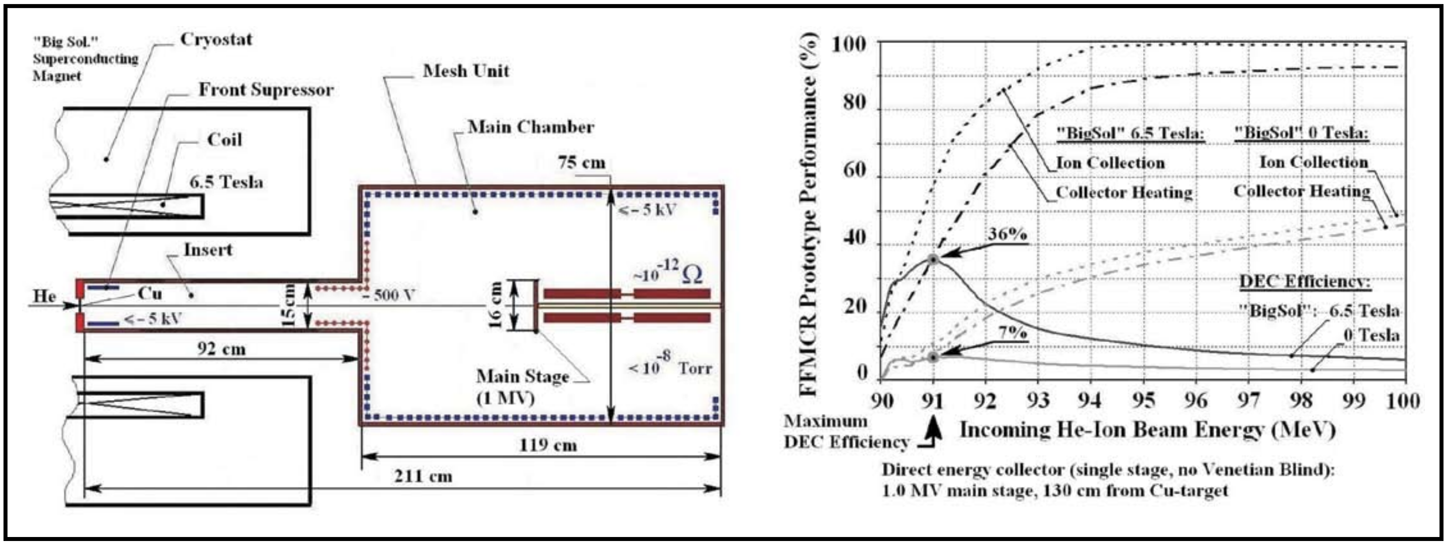


Figure 1. Magnetic collimator reactor prototype design (left) and anticipated performance (right).

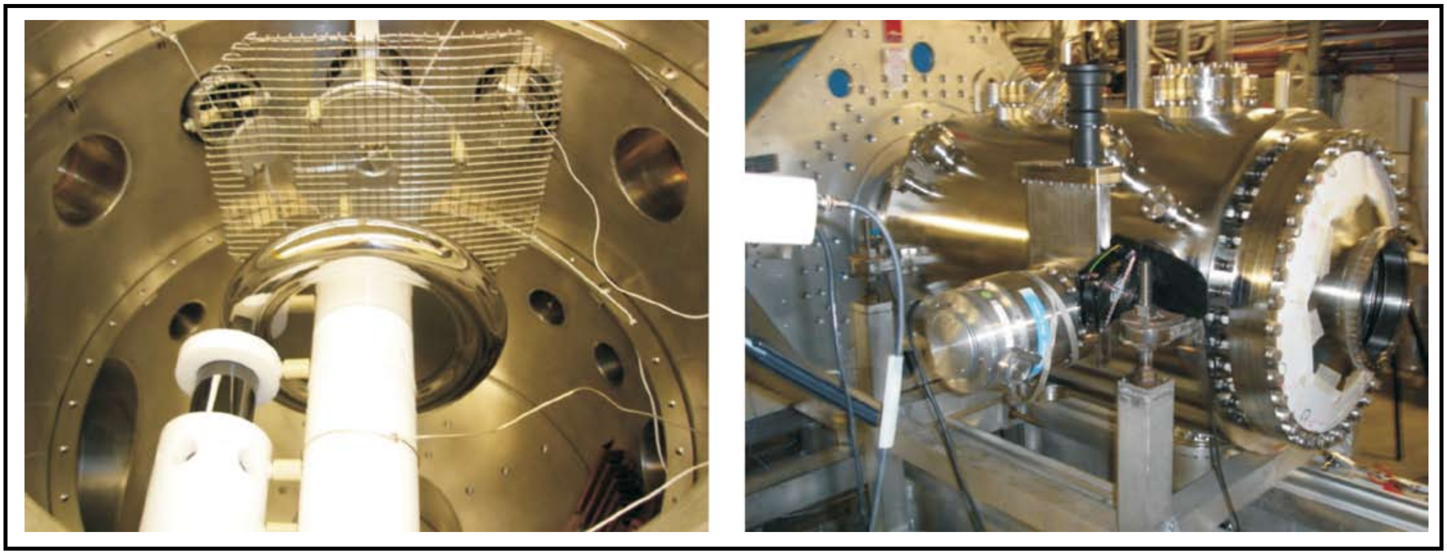


Figure 2. Experimental setups: small-scale prototype (left) and main prototype assembly (right).

The 150-keV linear accelerator system at Texas A&M University has been selected for preliminary testing in order to validate basic performance characteristics. Upon completing these preliminary tests, researchers will conduct their primary experimental studies of the scaled FFMCR prototype at the Texas A&M K500 Superconducting Cyclotron Facility. Figure 2 illustrates both experimental setups. Researchers developed these designs based on results of the previously detailed computational analysis. The experimental results using the 150-keV accelerator system indicate that testing of the main prototype using the K500 Cyclotron should successfully demonstrate the feasibility of the FFMCR concept. Due to a helium leak in the BigSol magnet system, performance of the main experiment has been delayed awaiting completion of required repairs. As a result, DOE has granted a no-cost extension of this project.

Planned Activities

Once the superconducting magnet is repaired, the research team plans to begin experimentation with the 100 MeV beam and an initial current of about 2.5 nA, corresponding to an anode voltage of 50 kV. With the BigSol magnetic intensity exceeding 2 Tesla and an estimated RC time constant of about 120 sec, researchers will measure the actual time dependence of the anode current. They anticipate sharp spikes that will die away with time. After the spikes diminish and the current becomes steady, the current will be increased in 2.5 nA increments to at least 25 nA (500 kV). They will repeat this process for several different incident beam energies on the copper entrance disk. At the lowest incident energy, the team hopes to see a leveling out of the beam current at the median beam energy incident on the anode. They

Figure 3. The prototype assembly with Texas A&M University team.

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: Allan F. Pardini, Pacific Northwest National Laboratory; Ronald L. Hockey, Applied Research Associates

Collaborators: General Atomics, Iowa State University, and Oak Ridge National Laboratory

Project Number: 02-103

Project Start Date: September 2002

Project End Date: January 2006

Research Objectives

Tri-isotropic (TRISO) particle fuels are being considered for use in both near-term and Generation IV nuclear systems. These fuels consist of sub-millimeter diameter uranium oxide spheres uniformly coated to prevent the release of fission products into the reactor (Figure 1). 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 assure the quality of the fuel particles in a cost-effective manner.

Current quality control (QC) methods are manual, can destroy test specimens, and are not economically feasible on the scale needed. Replacing these methods with fast and accurate automated nondestructive evaluation (NDE) techniques 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 develop and demonstrate NDE methods that will detect defective TRISO fuel particles and reject them from the flow stream.

The primary objectives are to:

- Establish a set of test standards representing acceptable fuel particles through a full spectrum of characterized defects
- Evaluate, develop, and demonstrate new NDE QC capability on unfueled and fueled coated particles needed to both reduce cost and provide improved reliability and uniformity of manufacturing for batches of particle based fuels
- Develop the methodology for a multiple attribute "Quality Index," describing both single particle and batch conformity to acceptance specifications
- Identify NDE design parameters for an integrated in-line "advanced QC system" for next generation, large scale fuel manufacturing inspection systems required to support advanced reactor deployment

Research Progress

This NERI project was completed in January 2006. Following were the beneficial outcomes of this work:

- The demonstration of the feasibility of using in-line electrical measurements (inductive/capacitive sensor impedance) for defect detection and TRISO QC.
- The completion of a defect detection and capability assessment of acoustic microscopy, resonant ultrasound spectroscopy, high-resolution computed tomography, and advanced optical measurements of both in-line and/or advanced off-line for NDE-QC measurements on TRISO fuel particles.

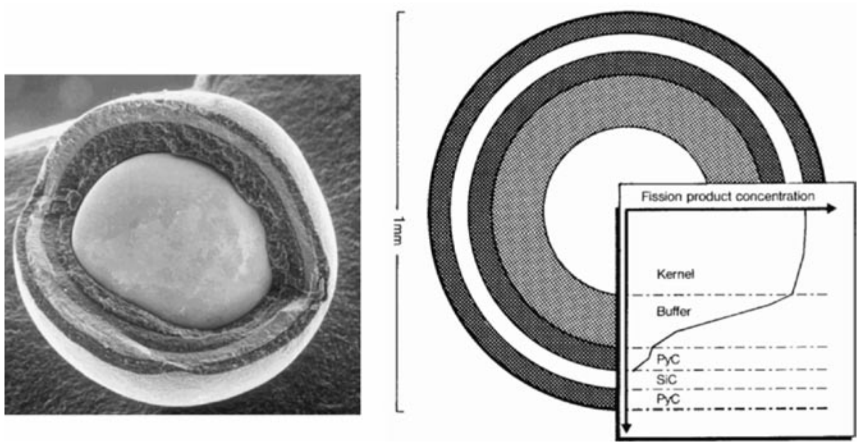


Figure 1. Coating cutaway view of a fuel particle (left) and the typical radial distribution of fission products (right).

- Development of a family of QC tools, evaluated on both surrogate and uranium-bearing fuel particles, which could be transitioned for use on a pilot scale TRISO coater or similar nuclear fuel fabrication line.
- The development of a Quality Index approach for implementing the technology developed under this project.

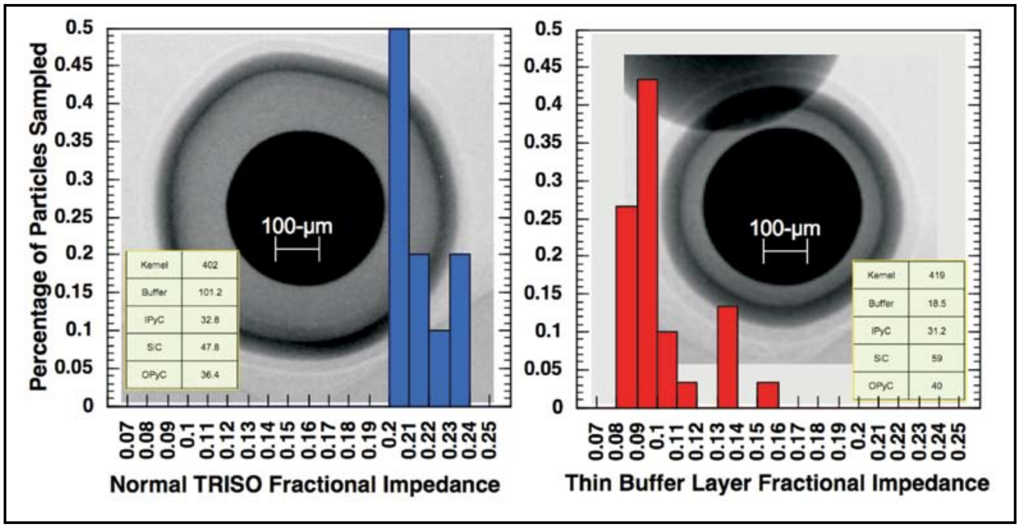


Figure 2. Examples of impedance sensor measurement distributions of a select group of surrogate TRISO particles from coating runs with normal layer dimensions (left) and a thin buffer layer (right).

Researchers made significant progress toward demonstrating the technical feasibility of using electrical (inductive and capacitive impedance) sensor measurements for high-speed defect detection and sorting. The capacitive impedance sensor was significantly improved and is now showing good correlation with coating parameters. The complementary inductive sensor was modified for integration into a single package with the capacitive sensor and the integrated design was completed, fabricated into a prototypical particle delivery system, and successfully demonstrated to show the potential of high-speed electrical measurements for inspecting TRISO particles.

Researchers also evaluated the kernel chemistry effects on the electrical measurement techniques being proposed. They obtained two batches of coated and uncoated depleted uranium-oxide (DUO_2) and natural uranium oxycarbide (NUCO) kernels in 2005 and conducted the required electrical measurements.

Researchers procured and customized special resonant ultrasound spectroscopy (RUS) transducers for use with sub-millimeter particles. The data indicated that the resonance modes of TRISO particles are extremely sensitive to the size and shape of the particles and to internal defects, including thin or missing layers. Even though there is variability in the location, number, and shape of the resonant peaks for nominally identical particles, the defective particles are clearly distinguishable from normal TRISO particles.

Completion of the particle library provided data necessary to fully evaluate the electromagnetic methods for defect detection. All of the particles examined to date were included in the library along with process parameters, radiographic data, and electrical measurements. Utilizing the particle library, further statistical and physical property modeling was performed.

This NERI project addressed several of the "QC" requirement issues for fuel particle production at a financially viable cost by automating as much of the test inspection as possible using high-speed measurement techniques. In particular, the techniques proved adept at detecting particles having unacceptable coating layer thicknesses (see Figure 2). The project provided a structured approach to evaluate the science and technology for 1) in line measurements, 2) on process measurements, and 3) advanced off-line NDE measurements needed to improve quality at a reasonable cost. Portions of the technology developed under this project may also economically improve the QC of other processes involved in manufacturing the final fuel assemblies required for Generation IV reactor designs.

This project has resulted in a total of seven conference proceedings and journal publications and is in the process of filing a patent application.

Planned Activities

This project received a no-cost extension until January 2006. Upon completion, the research team proposes the following recommendations for follow-on work:

- Establish a wider set of fully characterized particles with additional variation and quantity to provide for a more comprehensive statistical analysis
- Fabricate a robust particle feeder system that can supply sufficient particle throughput to a prototype inspection system
- Complete a quality index of attributes that have been directly correlated to nondestructive test methods

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: June 2006

Research Objectives

The overall objective of this research project was to develop and demonstrate advanced algorithms for the safe and efficient control of operational transients in nuclear power plants and for the continuous, on-line status monitoring to warn of system degradation and incipient failure. This resulted in two new capabilities:

- 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

Identifying the system's state is an important requirement for both of these activities; therefore, researchers on this project will also seek an optimized approach for acquiring sensor data 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

Researchers have developed and demonstrated an algorithm for the off-line diagnosis and surveillance of multi-component systems. The integrated fault detection and diagnosis algorithms included a fault detection procedure using X^2-hypothesis testing to detect the onset of a system perturbation. Upon detecting an anomaly, the algorithm runs additional steps to diagnose the possible causes of the perturbation. These include the application of multiple adaptive Kalman filters, each using a unique phenomenological system model that is characteristic of a specific component fault/no-fault scenario. Researchers then screened the resulting set of joint probability density functions (pdf's) for system/component characteristics using hypothesis testing procedures to determine which characteristics in the set of density functions constitute a component fault and which estimates are significant and unique. The probability associated with each of these discrete component degradation scenarios are calculated by performing a Monte Carlo sampling of the salient component characteristic-dependent state transition rates. A marginalization procedure was also developed to combine the information contained in the probabilities of component fault hypotheses with the component characteristic pdf's that define the given hypotheses in order to get a "snapshot" of the marginal density function for each individual component characteristic.

The researchers analyzed simulated binary component faults in a balance-of-plant model of a reactor plant. The algorithms gave the probability of each fault hypothesis, and also marginal pdf's for faulted component states. The results of these marginal pdf's are of greatest interest, as they give the probabilities for component characteristics as computed by the algorithm (see Figure 1). These probabilities will continue to evolve as additional plant measurements are provided to the algorithm. The performance of the integrated detection and diagnosis algorithms, particularly the fault diagnosis algorithm, were found to be effective for detecting and diagnosing even relatively small-magnitude, multiple faults.

The theory and algorithms developed in this work combine system modeling with process measurement and reliability data. This provides a set of practical algorithms for the off-line analysis of system observations for the detection and diagnosis of incipient component faults. The general probabilistic development and associated ideas could also prove useful as a starting point for other diagnostic procedures.

Applicability of the proposed method may be limited by the scarcity of the component degradation data, especially for complex systems comprising high-reliability components in nuclear power plants. This would require judicious choices of component characteristics together with the elicitation of expert opinion for near-term applications. Despite significant approximations inherent in the component transition rates, it is still important to include component reliability data as an integral part of fault detection and diagnosis methods, both for off- and on-line applications.

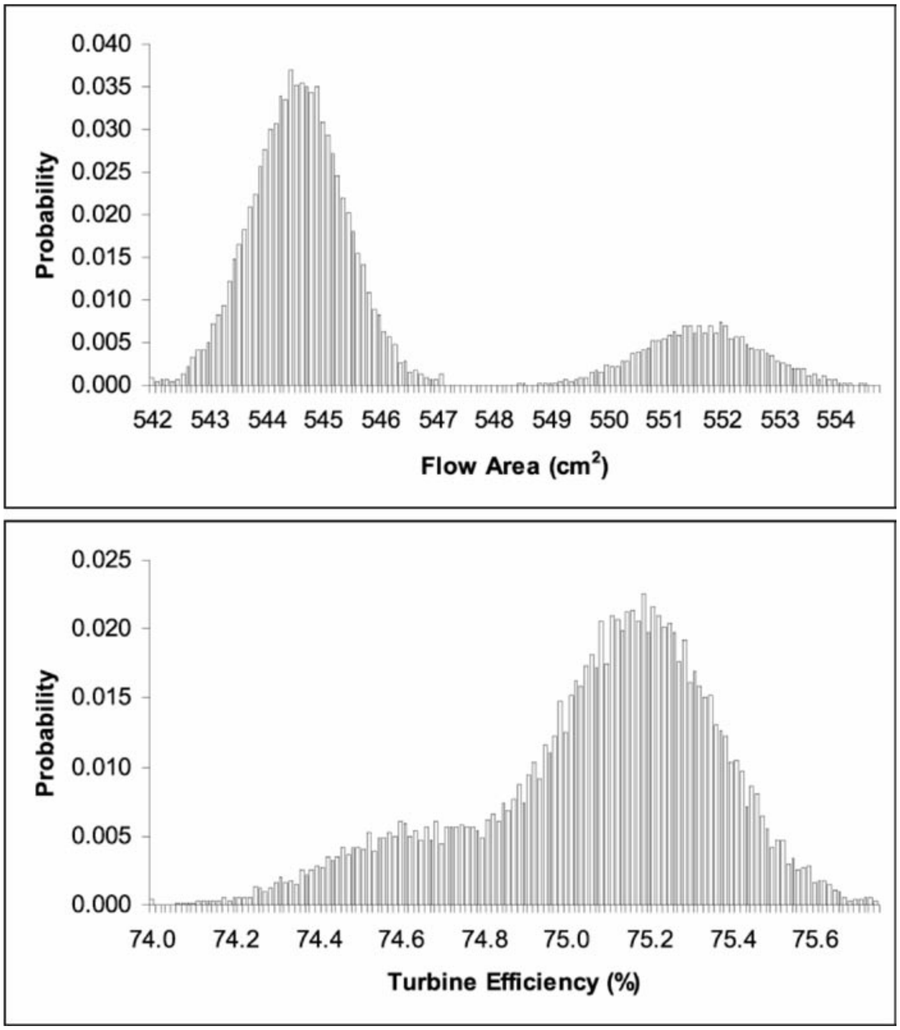


Figure 1. The marginal pdf's predicted for faulted components, effective flow area and low-pressure (LP) turbine efficiency, based on both plant measurements and reliability data. In this system, researchers simulated a binary fault comprising a 5 percent increase (from nominal) in the effective flow area of the main steam valve and a simultaneous 10 percent decrease (from nominal) in the LP turbine efficiency.

A potential monitor for steam generator tube degradation is the acoustic signature from flow-induced vibration of the steam generator tubes. In order to investigate the potential of this monitoring approach, researchers need to develop a coupled physics model of the unsteady fluid forces on steam generator tubes including the tube structural response and feedback. Steps in that direction have been undertaken as part of this project. A Large Eddy Simulation model in FLUENT has been validated for the prediction of unsteady fluid flow and forces by comparison with experimental data for a single isolated tube and a tube bundle. A coupled FLUENT-ABAQUS model was also developed to couple flow and structural models. However, results are not available due to problems with the deforming mesh in FLUENT. This problem has been communicated to and must be resolved by FLUENT Inc.

In model-based predictive control, researchers have demonstrated the efficacy of a very simple and fast state-estimator in avoiding DC offset errors in plant set-point tracking. This demonstration was also made within the context of a nuclear plant model with significant amounts of noise polluting the measured plant core power and outlet temperature signals.

Planned Activities

This project has received a no-cost extension until June 30, 2006. The research team will continue further development of the adaptive filter algorithm for degradation monitoring, perhaps through the deployment of particle filtering algorithms. They will also test the model-based predictive control methodology in a full model-based predictive control algorithm. Finally, as the project end date approaches, they will prepare a final report of results and develop recommendations for future research.

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: February 2006

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 were to identify, characterize, and evaluate the critical technical and economic issues associated with nuclear hydrogen production, including 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 prior analyses to encompass factors beyond the production plant, including on-site hydrogen storage and handling, hydrogen transmission and distribution, integration with end-user processes, and overall energy system economics. This research was divided into two phases with the following objectives:

- Define the physical characteristics, infrastructure requirements, and economics for an N^th^-of-a-kind centralized thermochemical hydrogen production plant driven by a helium-cooled modular nuclear reactor
- 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

The project team was able to identify and analyze the major requirements for nuclear hydrogen production. They defined the physical characteristics and major considerations for the nuclear reactor, the thermochemical hydrogen production process, and the related hydrogen infrastructure. The team also performed 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. External factors, such as the effects of carbon taxes or credits, were also investigated. The results of these analyses were documented in a topical report.[1]

Researchers have completed a site-specific, pre-conceptual design of a nuclear hydrogen production plant. The test-case design provides a more detailed evaluation of nuclear hydrogen production, including preconceptual plant design, heat transfer loop and circulator design, 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 including a total project cost (TPC).

The baseline design approach for the nuclear hydrogen production plant was the use of the high-temperature gas-cooled nuclear reactor combined with a thermochemical water-splitting process using the sulfur-iodine (S-I) cycle. The nuclear reactor was based on the Modular Helium Reactor (MHR) design under development by General Atomics, Inc. (GA). It is a helium-cooled reactor using a graphite moderator and coated-particle (TRISO) fuel. For the hydrogen production process, heat from the nuclear reactor is delivered to the thermochemical process plant through a secondary helium cooling loop heated to 925-975°C in an intermediate heat exchanger (IHX). Researchers developed a conceptual design for the S-I process and performed process modeling using AspenPlus®

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[1] WSRC-TR-2004-00318 Rev. 0, July 31, 2004

to calculate the overall thermal efficiency of the nuclear hydrogen plant. The results of this effort indicated an efficiency range of 42-56 percent on a higher heating value (HHV) basis, depending on the operating temperature and the degree of the optimization in the process design.

An important aspect of this study was assessing the integration of a nuclear plant with the hydrogen production process, considering the broader hydrogen infrastructure needs and end-user hydrogen requirements. The researchers identified two main types of end-users for nuclear produced hydrogen: 1) large industrial facilities and 2) the consumer market (e.g., hydrogen vehicles, distributed power). Researchers analyzed various options for storing hydrogen, including compressed storage, converting to a liquid form by including 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.

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 nuclear hydrogen, for cases both with and without CO_2 emission restrictions. They conducted sufficient analysis to quantify the impact that carbon credits, CO_2 sequestration requirements, or other related factors may have on the economic competitiveness of nuclear hydrogen production, versus conventional natural gas steam reforming production of hydrogen.

The break-even natural gas price where the cost of nuclear hydrogen equals that based on natural gas steam reforming is \$6.36/MMBTU without CO_2 removal. When the additional costs associated with CO_2 removal and sequestration are considered, the break-even price drops to \$4.37/MMBTU. Since recent natural gas prices have already exceeded these values, it is evident that future nuclear hydrogen costs for the Base Case will be economically competitive with conventional natural gas hydrogen production, even without CO_2 restrictions.

Pre-conceptual design work consisted of developing a design for a prototype commercial nuclear hydrogen plant located at the Department of Energy's Savannah River Site near Aiken, South Carolina. The plant design chosen consisted of a single 600 MW_{th} Modular Helium Reactor. Based on this work, the researchers concluded 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 supply system to encompass automotive and other users may be a viable approach for addressing some of the perplexing challenges facing the establishment of a hydrogen economy.

Planned Activities

This project received a no-cost extension until February 28, 2006. The primary task remaining is to compile and document results of the pre-conceptual design in a final report. This report will contain all of the primary information in support of the application of a specific plant design for the integrated MHR nuclear reactor and thermochemical hydrogen production plant and to determine plant layout, process flowsheet efficiencies, site selection, total project cost, and construction schedule. This final report is intended for use as a reference document and a basis for future nuclear hydrogen production plant projects.

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 (KSU)

Collaborators: None

Project Number: 02-174

Project Start Date: September 2002

Project End Date: March 2006

Research Objectives

The objective of this project was to develop neutron radiation detectors that are capable of withstanding intense radiation fields; performing "near-core" reactor measurements; operating in pulse-mode and current-mode; and discriminating neutron signals from background gamma ray signals. These detectors must also be tiny enough to be inserted directly into a nuclear reactor without significantly perturbing the neutron flux. Researchers developed miniaturized fission chambers that will be deployed in a TRIGA (Training, Research, Isotopes, General Atomics) research reactor at Kansas State University.

The unique miniaturized neutron detectors will be used for three specific purposes: 1) reactor power-level monitors, 2) power transient monitors, and 3) 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 primary goals of this project were to develop, characterize and deploy the detectors, and to develop computer codes with associated data reduction for real-time reactor core flux monitoring and power-level determination. Detector development involves the design, construction, and deployment of radiation hardened, thermally resistant, inexpensive gas-filled fission chambers. Electronics attached to the devices will allow for both pulse-mode and current-mode read-out. Further, each detector pack consists of a triad of devices: a ^{232}Th-based fast neutron detector, a ^{235}U-based thermal neutron detector, and an uncoated device for gamma ray background. Seventy-five triads, amounting to a total of 225 individual detectors, will be distributed through a plane in KSU's TRIGA reactor core. Researchers are developing a back-projection algorithm that uses detection information extracted from the detector triad arrays to determine the power density within the fuel elements of the nuclear core.

Research Progress

Micro-Pocket Fission Detectors (MPFD) consist of radiation-resistant ceramic plates that are configured to produce miniaturized fission chambers. These devices are the smallest available, being over 100 times smaller than "sub-miniature" fission chambers. Further, the devices are simple to manufacture relative to other fission chambers. The MPFDs are fabricated from aluminum oxide, a material that can withstand the high-radiation and high-temperature environments in a commercial nuclear reactor core. The detectors are tested and calibrated in KSU's TRIGA Mark II nuclear reactor.

Figure 1 shows a cutaway view of the basic detector concept, which consists of a small ceramic structure with

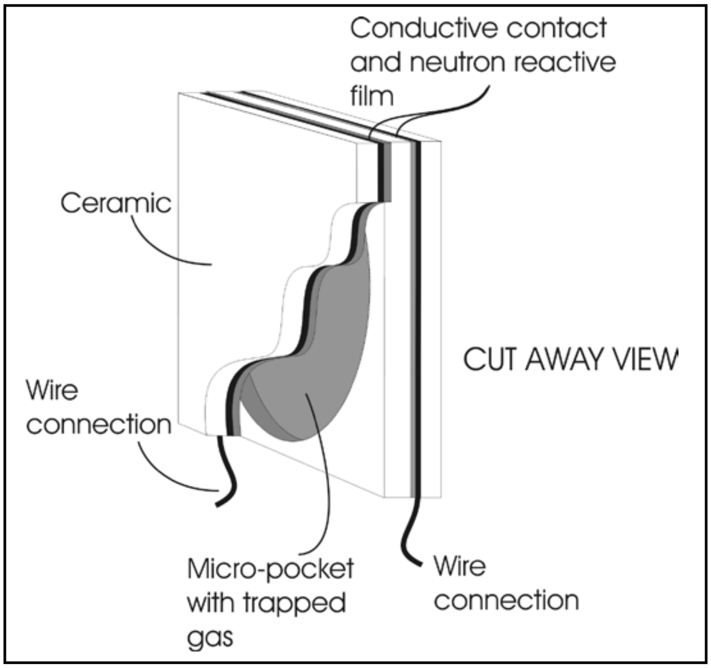


Figure 1. Basic construction of an MPFD.

a miniature gas-filled pocket inside. A conductive layer is deposited on opposite sides of the device, but not the perimeter. Neutron reactive material, such as ^{235}U, ^{232}Th, ^{10}B, or some material form incorporating ^{6}Li, is applied over both sides of the conductive contacts, although the device will operate with only one side coated. The ceramic pieces must be insulators (such as aluminum oxide or oxidized silicon) and must *not* contain neutron-absorbing material. Connecting wires must be sealed well so that no gas leaks out. Additionally, the ceramic pieces must be sealed with high-temperature cement so that the seal integrity is secure within the hostile environment of the reactor's core.

Use of ^{235}U as a conversion material for the MPFDs is of great interest. Pure ^{235}U has microscopic and macroscopic 2200 m/s neutron fission cross-sections of 577 barns and 28 cm^{-1}, respectively. Fission reactions in ^{235}U cause the emission of two fission fragments per fission with energies predominantly ranging from 60 MeV to 110 MeV, which are easily discernable from background gamma rays.

The device is operated with a voltage applied across the gas chamber. Neutron interactions in the reactive coating cause the ejection of energetic fission products, which produce ionization in the form of electron-ion pairs when these energetic ionizing particles enter the gas pocket. The applied voltage causes the positive ions and the electrons to separate and drift apart--electrons are attracted to the anode and positive ions to the cathode. The motion of the charges produces an induced current that is recognized and measured by sensitive external electronics. The result is a measurable pulse indicating a neutron interaction in the detector. Since the charge-detecting medium is a gas, it is improbable that gamma rays interact in the detecting medium; hence, the MPFD naturally discriminates out gamma-ray background noise.

Figure 2 shows the results of an MPFD tested within the center of the TRIGA reactor core, demonstrating that the device exhibits excellent count rate linearity with reactor power. Further, the small size and minute amount of uranium used allowed for pulse-mode operation without appreciable dead-time distortions or problems. Dead-time losses became noticeable at neutron fluxes above 10^{12} cm^{-2}s^{-1}, reaching a maximum of 24 percent near 10^{13}cm^{-2}s^{1}. Thus far, the MPFDs have withstood neutron fluences exceeding 10^{19} cm^{-2} without noticeable performance degradation.

The most recent versions of MPFDs being manufactured at KSU have three gas pockets that are only 1 mm in diameter and 1 mm thick (see Figure 3), allowing for simultaneous detection of fast neutrons, thermal neutrons, and background at a single point location in the reactor. Under these conditions, fission fragments will deposit over 5 MeV of energy in the detector region, easily discernable from gamma ray interactions. Physical vapor deposition is used to apply Pt contacts on the MPFD substrates, which are then annealed to improve metal adhesion and conductivity. Afterward, fissionable material is plated onto the devices with a custom precision electroplating system designed to handle radioactive materials. This system, known as the Automated System for the Consistent Electroplating of Fissionable Isotopes (ASCEFI), is shown in Figure 4.

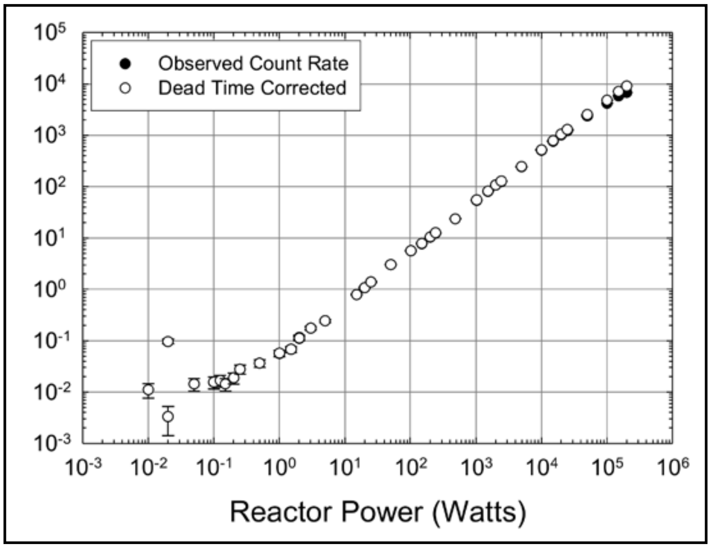


Figure 2. MPFD count rate as a function of TRIGA reactor power. The device was placed in the center of the nuclear core, where the highest thermal flux was 10^{13} n cm^{-2} s^{-1}. The device responded linearly in pulse mode through the entire power range.

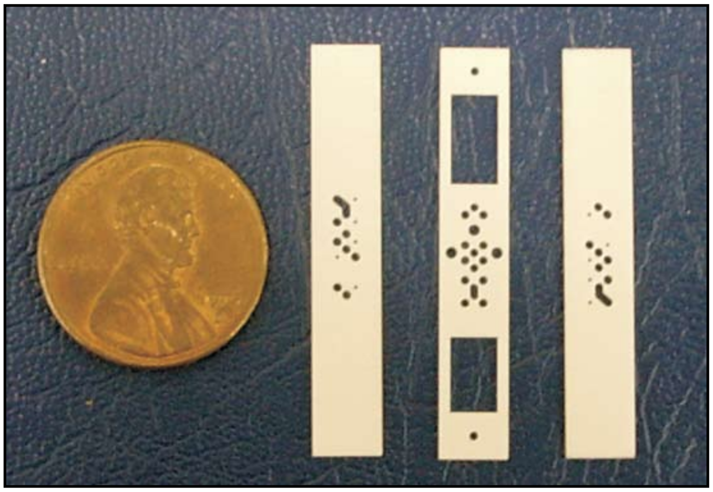


Figure 3. Alumina substrates for final generation of MPFDs.

The detectors are inserted into the KSU TRIGA reactor through the flux probe holes in the grid plate. In order to do so, aluminum flux probes (Figure 5) were designed such that a string of five MPFD triads are inserted into the probe, which is then indexed and inserted into the reactor grid plate.

Figure 4. The ASCEFI system inside a radiological fume hood.

Long-term use of an MPFD can result in appreciable burn-up of the fissionable material. If the devices are to be used for power reactors, the fissionable material mixture can be tailored for optimum results. A simulation program has been developed to determine the expected lifetime of different fissionable coatings for the MPFDs. The program also has the capability to determine an optimized coating made from mixing multiple isotopes in order to extend the life of the detector as long as possible while maintaining a flat response. Using a combination of 65 percent ^{235}U (enriched to 1.357 weight-percent) and 35 percent ^{232}Th will allow the detectors to be operated for 58 years in the KSU TRIGA reactor at full power with less than 1 percent deviation in count rate (Figure 6).

Planned Activities

This project has received a no-cost extension until March 14, 2006. MPFDs have shown exceptional radiation hardness to neutrons, gamma rays, and charged-particle reaction products, with no performance degradation for devices exposed to neutron fluences exceeding 10^{19} cm^{-2}. Further, pulse-mode-operated devices have shown a linear relation to reactor power for neutron fluxes up to 10^{13} cm^{-2} s^{-1}, and smaller MPFDs are expected to operate in pulse mode in higher neutron fluxes.

Data from an MPFD array can be converted into a power density map of the reactor core for real-time analysis. Researchers are currently 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 the construction of an appropriate response function that gives 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. Response functions have been derived and used to illustrate the analysis methods. Thus far, modeled results using predicted sensitivities of the MPFDs indicated that the power density in the fuel can be determined provided that appropriate boundary conditions regarding device placement are met. Good matching to power density profiles can be achieved with as few as five detector triads per detector string.

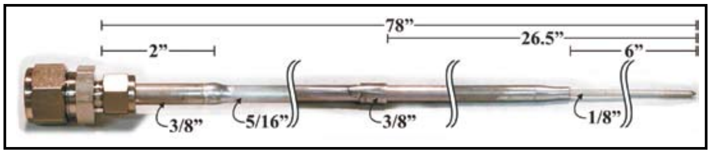


Figure 5. Flux probe design showing (from right to left) the 1/8″ lower grid plate pin welded inside a 5/16″ (0.030″ wall) aluminum tube. A 3/8″ collar is welded 26.5″ from the bottom to provide an indexing guide to the top grid plate. Another 3/8″ collar is added to the end of the tube to support a 3/8″ to 5/8″ Swagelok coupler for connection to a stainless steel tube.

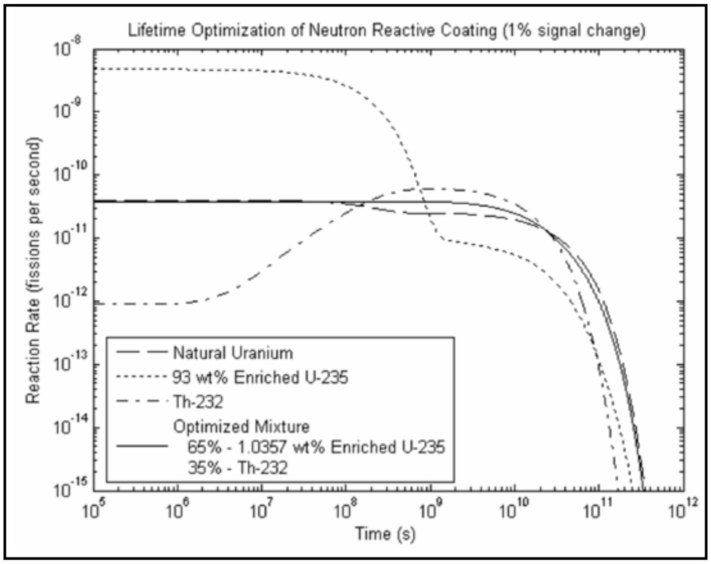


Figure 6. Lifetime optimization using a mixture of enriched uranium and thorium for full power continuous operations in the KSU TRIGA MARK II nuclear reactor.

During the last part of the research project, researchers on this project will deploy arrays of MPFDs (which have ^{232}Th and ^{235}U inside) into the flux probe holes of the KSU TRIGA to observe real-time neutron flux measurements. These array assemblies will form a "sheet" of detectors across the core. Researchers will extract the signals in both pulse and current modes and use this information to calculate the power density in the fuel. Real-time core mapping will be performed. Data acquired from the MPFDs will be used for inverse power density distribution calculations.

NUCLEAR ENERGY RESEARCH INITIATIVE

Development of a Supercritical Carbon Dioxide Brayton Cycle: Improving PBR Efficiency and Testing Material Compatibility

PI: Chang Oh, Idaho National Laboratory

Collaborators: None

Project Number: 02-190

Project Start Date: October 2004

Project End Date: February 2006

Research Objectives

The very high temperature gas reactor (VHTR) is a promising Generation IV nuclear reactor concept for high-efficiency electricity generation and hydrogen production. Certain technical and economic issues must be overcome before this reactor design can be built in the U.S., including making the VHTR intrinsically safe and proliferation resistant. The economic cost of this nuclear plant design is not yet favorable to compete with power generation from combined-cycle, gas turbine plants. The cost gap 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 project were to: 1) develop a supercritical carbon dioxide (SCO_2) Brayton cycle in the secondary power conversion side for application to Generation IV reactors such as the Gas-Cooled Fast Reactor and Supercritical Water Reactor, 2) improve plant net efficiency by using the carbon dioxide Brayton cycle, and 3) test material compatibility at high temperatures and pressures. The higher density of carbon dioxide compared to helium reduces its volumetric flow rate, thereby reducing compression work, which will ultimately result in increased plant efficiency.

Advanced gas reactor technology has been identified as a promising modular reactor system that is passively safe, thermally efficient, and proliferation resistant. This design is capable of both electricity and hydrogen production, as well as the efficient burning of spent fuel. Internationally, the VHTR concept has become the top priority for implementation because of its inherent safety and near-term implementation feasibility. It is also a candidate design for the Next Generation Nuclear Plant (NGNP) expected to be built at the Idaho National Laboratory (INL) site by 2017.

Research Progress

All tasks (with the exception of corrosion testing) have been performed on schedule and budget, and their technical accomplishments are summarized below.

The objective of this task was to develop an SCO_2 Brayton cycle and to improve the overall plant cycle efficiency by increasing the efficiency of secondary side of the VHTR. To accomplish this task, researchers performed a number of HYSYS simulations to investigate different cycle configurations, including 3-shaft Brayton, combined, and reheat cycles. They also extended the workscope to include the intermediate heat exchanger, which links the nuclear plant to the hydrogen production plant. This summary documents analytical results for the combined cycle and intermediate heat exchanger loop. Results of the 3-shaft configuration and reheat cycle in the power conversion system will be included in the final report.

The combined cycle, illustrated in Figure 1, is an indirect cycle consisting of a primary loop, a combined power conversion unit (PCU), and an intermediate heat transport

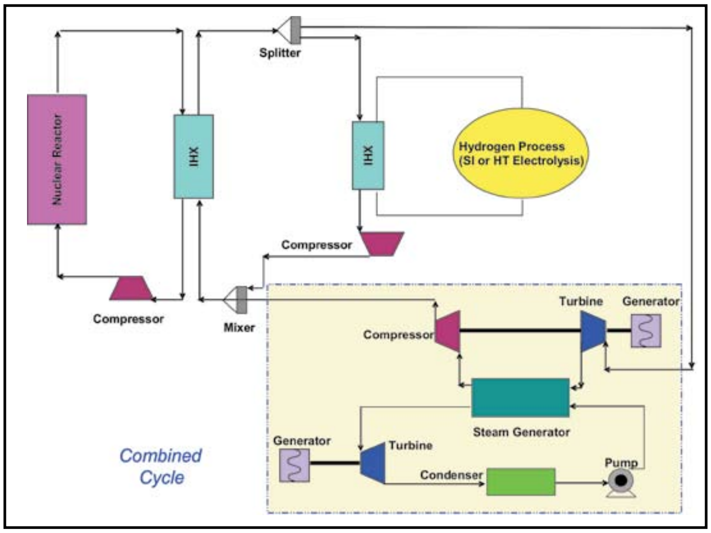


Figure 1. Pressurizer CFD model of pump plenum.

loop for hydrogen production. The primary side of the loop consists of a high-temperature nuclear reactor, intermediate heat exchanger (IHX), and a circulator (compressor). Researchers developed a split intermediate heat transfer loop in order to deliver process heat in parallel to both the hydrogen production system and the PCU (summarized in INL/EXT-05-00453). The PCU configuration, also illustrated in Figure 1, consists of a Brayton top cycle with a gas turbine and compressor, coupled to a Rankine bottoming cycle through a steam generator. The Rankine cycle is a typical configuration consisting of a steam turbine, condenser, and pumps. Researchers simulated the overall cycle using helium as the working fluid in both the primary and intermediate heat transport loops. They also modeled CO_2 and a nitrogen-helium mixture in the intermediate loop and Brayton cycle. Water/steam was used in the Rankine cycle. Table 1 shows a summary of the primary loop operating conditions.

Parameter	Nominal Value
Power, MW	600
Inlet temperature, °C	500
Inlet Pressure, MPa	7.05
Outlet temperature, °C	900
Outlet pressure, MPa	7
IHX pressure drop, MPa	0.05
Mass Flow, kg/s	289
Working fluid	He

Table 1. Summary of primary working conditions for the combined cycle.

The same Rankine bottoming cycle was used for all three working fluids. This cycle gets heat from the Brayton cycle through a steam generator located between the gas turbine and compressor. The steam turbine inlet temperature was set at 575°C to take advantage of the superheat option and keep the turbine outlet quality at 85 percent.

Comparing the working fluids in this cycle, it can be seen that the CO_2 working fluid produces the highest efficiency and smallest component sizes.[1] Helium and the N_2-He mixture produced similar component sizes; however, helium had a slightly higher efficiency.

Researchers commenced a long-term corrosion test of fine-grained, oxide dispersion-strengthened Inconel MA 754 in supercritical CO_2 at 1,000°C in the fourth quarter 2004. This test was scheduled to be completed by mid-July, but a water leak developed after 300 hours of exposure and the experiment had to be stopped. There was substantial degradation of the water-cooled connector that required extensive repairs. The damage appears to have resulted from the cooling water and not the supercritical CO_2. Testing was expected to resume in October 2005.

Researchers completed modification of a second supercritical CO_2 corrosion test system during the fourth quarter and prepared samples for long-term testing (>1,000 hrs) at 1,000°C. This system is capable of simultaneously testing over 200 samples. Therefore, a commercial vendor prepared 240 samples of fine-grained MA754, I-617, and SiC with a variety of surface finishes. This allowed not only the inherent corrosion behavior of the three materials to be determined, but also the effect of surface finish on corrosion.

Researchers also determined the corrosion behavior as a function of temperature, using a temperature gradient along the length of the furnace. This particular system has been demonstrated to be highly reliable in previous supercritical CO_2 corrosion tests at lower temperatures and is not expected to suffer from the issues inherent to the other system outlined above. The final paperwork is being processed and the remaining corrosion test (>1,000 hours) is expected to end in late January 2006 when analysis of the samples will begin.

Researchers characterized a coarse-grained MA-754 sample, exposed for 500 hours at 1,000°C. There were some indications in the preliminary analysis that void

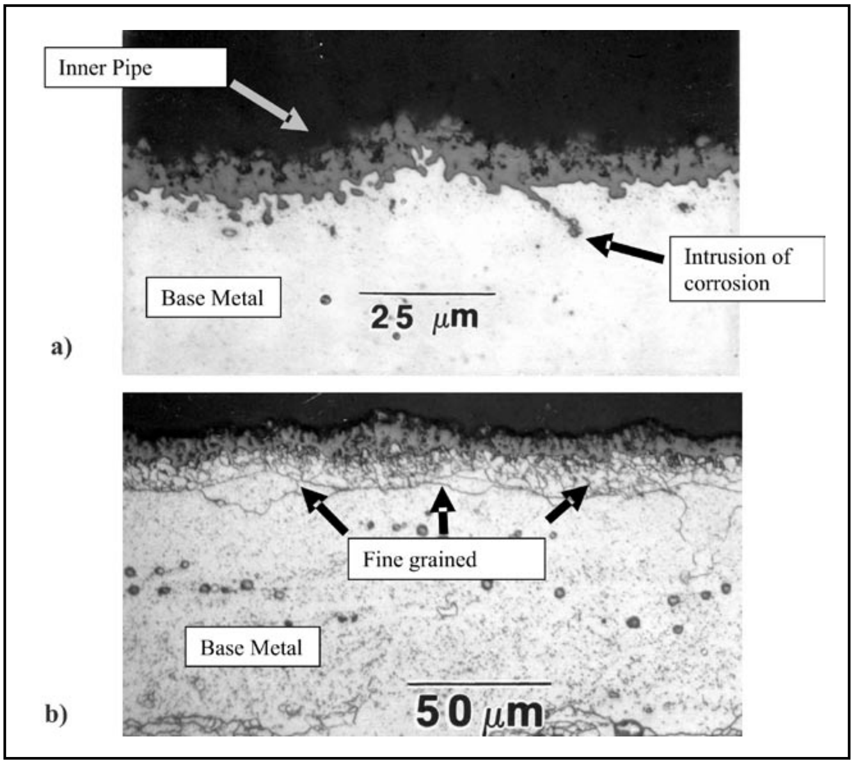


Figure 2. The intrusion of corrosion into the base metal, a), may be due to corrosion along the grain boundaries of the fine grain material near the inner surface of the sample, b).

formation may have occurred at grain boundaries. In the unetched condition, there appeared to be intrusions into the base metal (Figure 2a). The sample was etched to determine if the intrusion followed grain boundaries, which would indicate intergranular corrosion. The etch revealed that the base metal had recrystallized, producing a fine-grained microstructure very near the surface of the base metal (Figure 2b). It is not known at this time why this inner surface recrystallized. It is possible that machining of this inner diameter may have produced significant strain in the near surface that caused recrystallization when exposed to 1,000°C during the corrosion test.

Planned Activities

Completion of this project has been delayed to correct an unforeseen cooling system leak, as previously discussed. DOE granted a no-cost extension until February 2006.

- Complete corrosion testing of alternate alloys: fine grained Inconel (I)-617 in supercritical CO_2 at 1,000°C
- Document the corrosion behavior of I-617, an alloy receiving considerable attention for use in various high-temperature areas of the NGNP.
- Publish the final report in March 2006.

REFERENCE

[1] Oh, C.H and Moore, R.M., "Brayton Cycle for High-Temperature Gas-Cooled Reactors," *Nuclear Technology*, Vol. 149, pp. 324-336, March 2005.

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: March 2006

Research Objectives

The primary goal of this project was to develop a conceptual design of a modular helium reactor (MHR) for producing hydrogen. The plant designs under evaluation consisted of an MHR supplying either process heat or electricity to a hydrogen production plant. An additional goal was to provide DOE, utilities, and energy-policy planners the information needed to make decisions regarding future research and development for producing hydrogen using nuclear energy. To achieve these goals, the work was structured to meet five major objectives:

- Using a systems engineering approach, develop functions and requirements for an H_2 production plant using the MHR
- 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 receive process heat from an MHR
- Develop conceptual designs for an overall facility that integrates the reactor with thermochemical-based and HTE-based H_2 production plants
- Complete assessments of the plant design with respect to performance, safety, economics, and licensing

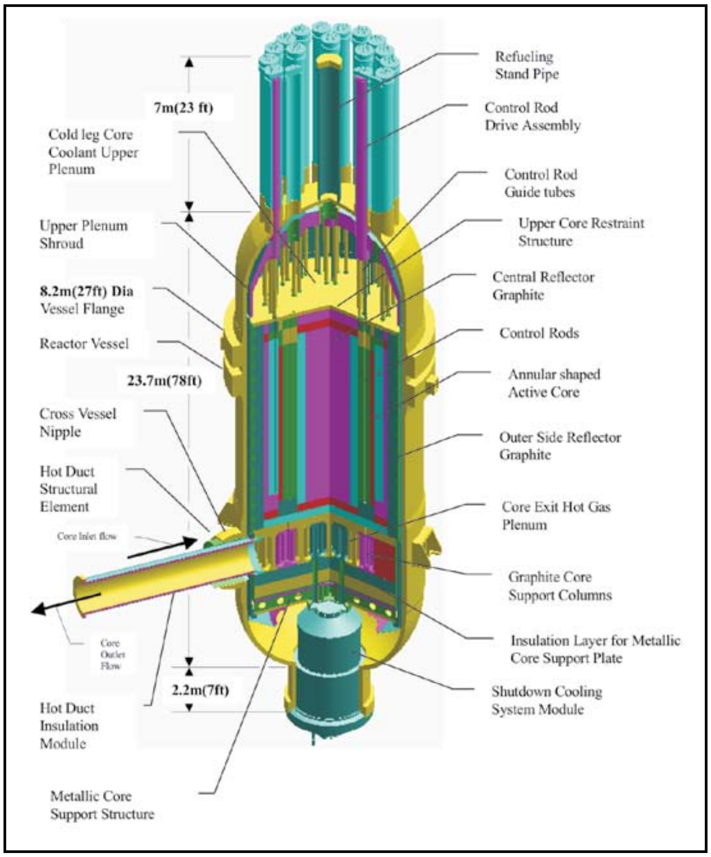


Figure 1. MHR Reactor Internals.

Research Progress

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

Researchers have finalized the nuclear and thermal-hydraulic design of the reactor core. Figure 1 shows the reactor design and internals, which are based on the Gas Turbine, Modular Helium Reactor (GT-MHR) design. Researchers developed a number of modifications to the basic GT-MHR design to optimize its use for hydrogen production. These modifications included: 1) lowering vessel temperatures by changing the inlet-flow configuration from channel boxes located between the core barrel and reactor vessel to flow paths in the permanent side reflector, 2) reducing power peaking factors and fuel temperatures by adopting an optimized fuel-placement refueling scheme, and 3) reducing coolant bypass flow by incorporating additional lateral restraints and sealing keys to close gaps between the fuel blocks.

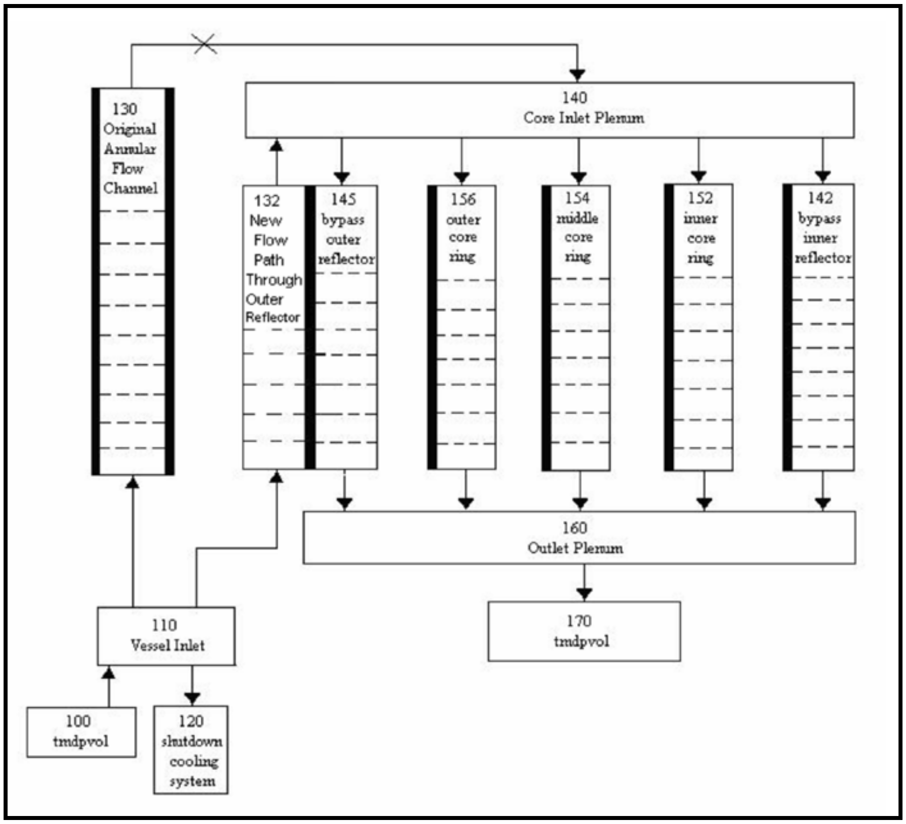


Figure 2. MHR RELAP5-3D/ATHENA model.

Researchers continued work on optimizing the new inlet flow configuration using the RELAP5-3D/ATHENA model of the MHR developed by Idaho National Laboratory (see Figure 2). With the new inlet flow configuration, they were able to reduce the calculated peak vessel temperature during normal operation from 541°C to 452°C, with coolant inlet and outlet temperatures of 590°C and 950°C, respectively. By optimizing the number and size of coolant passages through the permanent outer reflector, they reduced the pressure drop through the core region to approximately 60 kPa. The total pressure drop through the primary circuit is approximately 100 kPa, which corresponds to a primary circulator pumping power requirement of about 8.2 MW.

The Intermediate Heat Exchanger (IHX) design is based on the printed circuit heat exchanger (PCHE) concept developed by Heatric, which consists of metal plates that are diffusion bonded to restore the properties of the base metal. Fluid-flow channels are chemically milled into the plates using a technique that is similar to that used for etching printed electrical circuits. The PCHE concept allows for simultaneous high-temperature and high-pressure operation with relatively thin wall thicknesses between the primary and secondary coolants.

The IHX design was updated and now consists of 40 Heatric-type modules manufactured from a high-temperature alloy (Inconel 617 and Hastelloy-XR are candidate materials). Each module has a heat duty of approximately 15 MW(t), and weighs approximately 5 tonnes with dimensions of 0.6 m × 0.65 m × 1.5 m. In order to minimize the size and weight of the IHX vessel, it is desirable to use a compact arrangement to house the PCHE modules. However, sufficient room must be provided to accommodate differential thermal expansion (using expansion joints) and headers. Preliminary results indicate that it should be possible to design a 600 MW_t IHX with a vessel that is of similar size as the reactor vessel. Within the IHX vessel, the modules are arranged in eight axial layers, with five modules per layer. The IHX vessel is a pressure boundary for the secondary helium coolant and will be designed according to Section III of the ASME Code. The IHX vessel is manufactured using SA533 steel, and insulated to maintain operating temperatures below 350°C and to prevent creep damage.

Researchers completed refinements to the HTE plant process model using Hyprotech's HYSYS process modeling software. These refinements provide more realistic representations of the heat exchangers in the system in terms of pressure drop and effectiveness. The overall efficiency of the process is calculated to be 51.3 percent, based on the lower heating value (LHV) of hydrogen.

In addition to the work described above, the researchers also continued work on the hydrogen production plants. They updated and optimized the Aspen process model for the sulfur-iodine thermochemical water splitting process, refined the SAPHIRE availability models for the SI and HTE process plants, and updated the economic evaluations.

The following papers were prepared and presented at technical conferences throughout 2005:

- "The H_2-MHR: Nuclear Hydrogen Production Using the Modular Helium Reactor," ICAPP-05, Seoul, Korea, May 11-19, 2005.
- "Hydrogen Production Using the Modular Helium Reactor," ICONE-13, Beijing, China, May 16-20, 2005.
- "Conceptual Designs of an MHR Plant Used to Produce Hydrogen," 2005 ANS Annual Meeting, San Diego, CA, June 5-9, 2005.

- "H_2-MHR Conceptual Designs Based on the SI Process and HTE," OECD-NEA Third Information Exchange Meeting on the Nuclear Production of Hydrogen, Japan Atomic Energy Agency, Oarai, Japan, October 5-7, 2005.
- "Conceptual Designs for MHR-Based Hydrogen Production Systems," Global 2005, Tsukuba, Japan, October 9-15, 2005.

Planned Activities

Work has been completed on this project. DOE granted a no-cost extension until March 31, 2006 in order to prepare the final project report.

Nuclear Reactor Power Monitoring Using Silicon Carbide Semiconductor Radiation Detectors

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

Collaborators: Westinghouse, General Atomics

Project Number: 02-207

Project Start Date: October 2002

Project End Date: July 2006

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. The two designs being modeled include the International Reactor Innovative and Secure (IRIS) and the prismatic-core, gas turbine modular helium-cooled reactor (GT-MHR). This project has two primary tasks: 1) to analytically define the best location for SiC-based sensor arrays in the IRIS and GT-MHR nuclear power plants and to estimate the performance of the SiC sensor arrays using the predicted neutron flux levels and environmental conditions at these locations 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 (CR) necessary to generate a scram signal in an appropriately small time-frame and the risk of degrading the detector through radiation exposure. The 1 MeV equivalent neutron flux in SiC ($\phi^{Total}_{eq,1MeV,SiC}$) is an appropriate parameter with which to compare the radiation damage rate in SiC at different reactor locations at low temperature. Researchers from The Ohio State University (OSU) predicted the count rate and equivalent flux for SiC detectors at several locations, including various radial positions in the downcomer and core of the IRIS, and in the Reactor Cavity (RC) and Reactor Cavity Cooling System (RCCS) of the GT-MHR. Equivalent neutron flux, $\phi^{Total}_{eq,1MeV,SiC}$, was calculated using the general expression given in Eq. (1):

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

where FD,mat(E) is the damage function for the material of interest (SiC) evaluated at energy E; $F_{D,1MeV,mat}$ is the damage function for the material of interest (SiC) evaluated at energy E=1 MeV; and ϕ(E) is the energy-dependent neutron flux density function.

Per the design of Westinghouse Electric Corporation collaborators [1], it was assumed in the OSU calculations that the SiC diodes were covered with a thin layer of LiF to enhance their response to neutrons and to create features in the spectra that could be used to separate neutron-induced events from gamma-ray-induced events and noise. These features are a consequence of the 2.73 MeV triton reaction product of the Li-6(n, alpha) H-3 reaction. To minimize the radiation damage in the SiC diodes, the alpha particles are shielded from the SiC by a thin aluminum layer that is placed between the LiF radiator and the SiC diode.

IRIS: In the past year, researchers modeled the IRIS reactor in three dimensions using the radiation transport code MCNP, accounting for radial and axial variations of the core power distribution. They calculated the energy-dependent neutron flux distributions as functions of axial location for four radii in instrumentation tubes in the downcomer region (155 cm, 170 cm, 185 cm, and 200 cm). Then they used the calculated energy-dependent neutron flux to predict $\phi^{Total}_{eq,1MeV,SiC}$ and the maximum triton count rate, as shown in Table 1, for a detector with a 500 μm diameter and a 1 μm thick LiF radiator with 90 percent Li-6 enrichment for full power operation (600 MWt).

GT-MHR: The GT-MHR reactor was also modeled in three dimensions using MCNP, accounting for radial and axial variations of the core power distribution. The various regions of the core were treated as physically distinct. For example, the fueled region of the core was distinct from the various reflector regions; however, it was modeled as a homogeneous mixture. OSU researchers calculated the energy-dependent neutron flux distributions as functions of axial location for the following three radial locations: 1) in the central reflector, for detector capsules located at radial distances R=117 and R=153 cm from the central reflector centerline; 2) in the RC at R= 530 cm; and 3) in the RCCS at R=610 cm. Then they used the calculated energy- dependent neutron flux to predict the equivalent flux for full power operation (600 MWt), averaged over all axial layers, and to predict the maximum triton count rate (for the detector that is described above at Z=520 cm), as shown in Table 1.

IRIS			GT-MHR		
Detector Radial Location (cm)	$\phi^{Total}_{eq,1MeV,SiC}$ (n cm^{-2} s^{-1})	Max Triton CR (cps)	Detector Radial Location (cm)	$\phi^{Total}_{eq,1MeV,SiC}$ (n cm^{-2} s^{-1})	Max Triton CR (cps)
155	8.3E+10	7.5E+04	117	1.2E+11	6.0E+05
170	1.0E+10	7.7E+03	153	1.9E+12	6.4E+05
185	1.8E+09	1.1E+03	530	1.4E+07	<20
200	4.7E+08	2.3E+02	610	3.4E+07	<50

Table 1. 1 MeV equivalent neutron flux ($\phi^{Total}_{eq,1MeV,SiC}$) in SiC and maximum triton count rate at various locations within IRIS and GT-MHR for full power operation (an efficiency of 0.39 is applied to the (n,alpha) reaction rate in the ^{6}LiF radiator to determine the maximum triton count rate)

OSURR: Researchers characterized the radiation field in Position 4 in the semi-conductor device characterization vessel in Beam Port 1 of OSU's Research Reactor (OSURR) in terms of 1 MeV equivalent flux in order to correlate observed degradation of the SiC detectors in the OSURR with expected degradation for various possible detector locations in the GT-MHR. The values of $\phi(E)$ 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 500kW.

By comparing the equivalent flux values for the GT-MHR with the OSURR, the investigators concluded that SiC devices can be adequately tested for the radiation damage that would be incurred over a GT-MHR refueling cycle for detectors located in the RC and RCCS, and IRIS refueling cycle for detectors located at radii 185 cm and 200 cm. However, this conclusion is not valid for detectors located in-core in the GT-MHR, and at radii of 155 cm and 170 in IRIS.

Task 2. The team developed a simulation model of the counting channel from the SiC detector to the channel's end, including a distributed parameter model of the cable, using the program PSpice, and 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.
- Because the pulse shape is not greatly altered by the use of mineral-insulated cable, there should not be a large increase in the counting system dead time.
- With a voltage sensitive preamplifier of the type that they specified, the speed of the channel is limited by the discriminator dead time.

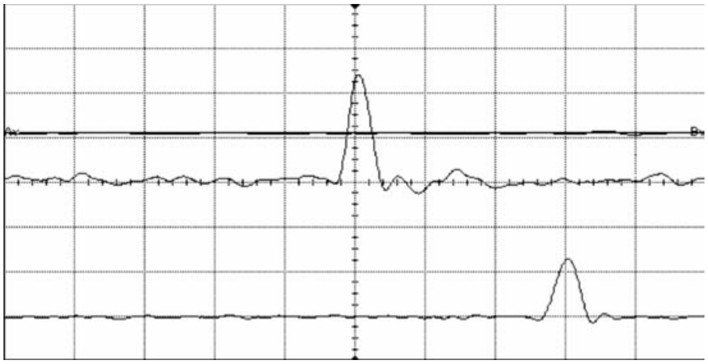


Figure 1. The preamplifier output, discriminator output, and discrimination level with a voltage scale of 100 mV/div and a time scale of 5 ns/div.

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

Planned Activities

The remaining work items to complete this project are to integrate the calculational and experimental results to define the best location for SiC-based sensor arrays in the IRIS and GT-MHR nuclear power plants, to estimate the performance of the SiC sensor arrays using the predicted neutron flux levels and environmental conditions at these locations, and to test the high-speed pulse counting system, which will be used in the SiC neutron sensor channels. Two important planned activities are described below in Tasks 1 and 2.

Task 1. OSU researchers will observe the degradation of SiC detectors in the OSURR and, using their knowledge of equivalent flux in the OSURR, translate that observed degradation into an expected degradation for various detector locations in the GT-MHR and in IRIS.

Task 2. Researchers will test the voltage-sensitive, pulse-counting system at high count rate and work toward ensuring that the entire detector channel functions properly.

Advanced Nuclear Fuel/Fuel Cycles Projects

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 Laboratory, Pennsylvania State University

Project Number: 02-081

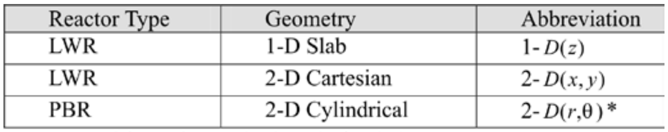
Project Start Date: September 2002

Project End Date: December 2005

Research Objectives

The objective of this research project was to develop a next-generation, high-order transport method for core neutronics calculations in advanced and Generation IV light water reactors (LWRs) and pebble bed reactors (PBRs). Researchers implemented the method into a computer code and applied 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 are:

- 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- $D(r,\theta)$ *

*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 Laboratory (INL) 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 year 3 of the contract has been coupling the transport and diffusion methods in 2-D (r, θ) geometry within PEBBED and validating the 2-D code for Cartesian geometry.

The researchers recognized that incorporating the transport solution based on the response functions method could not be done in a transparent way within the PEBBED code, as currently formulated. Therefore, they concluded that reformulating and recoding PEBBED is an unrealistic goal in the context of this project. Therefore, they explored other possibilities for reformulating the control rod treatment method in a way that integrates efficiently without excessive recoding. This task was divided into two objectives: 1) to determine the theoretical feasibility of such a transparent formulation and 2) to develop the required theoretical basis. The researchers developed a response-matrix-based nodal balance (RMNB) equation. This proposed methodology must be implemented transparently into the current PEBBED coarse-mesh finite difference (CMFD) methodology to preserve the diffusion theory treatment in nodes that do not require transport theory analysis. Also, in certain nodes, a mix of transport and diffusion effects must be taken into account. In order to appropriately deal with such intermediate zones, researchers derived a diffusion-transport hybrid nodal current formulation. They tested this method in 1-D using response functions generated from diffusion theory. The method proved to be very accurate and its coding was verified to be correct. The next step was to test the method with response functions generated from transport theory.

In order to validate the new method with transport generated response functions, researchers developed a

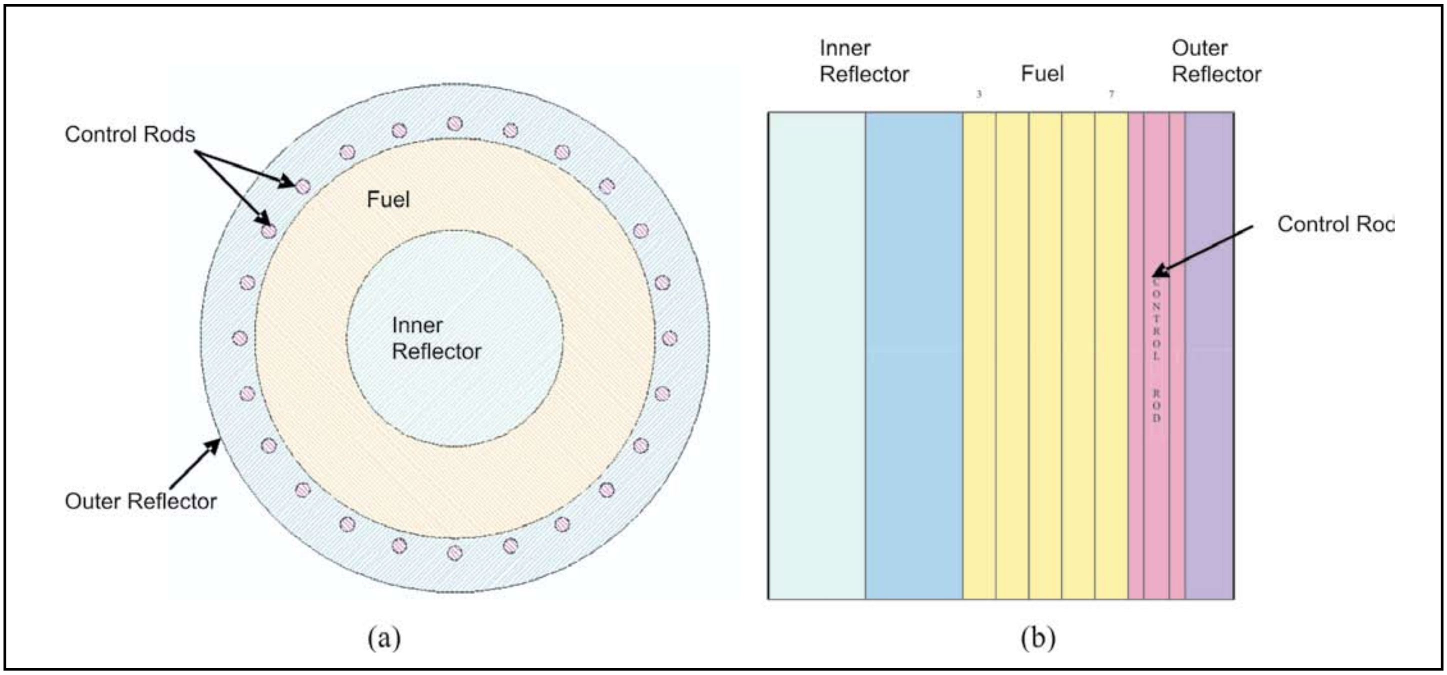


Figure 1. (a) 2-D and (b) 1-D simplified PBMR400 models.

more rigorous benchmark problem. Testing is underway. The benchmark is a 1-D adaptation of a realistic 2-D model of the PBMR400 pebble bed reactor. The specifications for the 2-D benchmark are based on data provided by INL and are representative of an equilibrium core. Figure 1(a) illustrates the geometrical configuration of the 2-D benchmark, consisting of an inner reflector region with a diameter of 2 m, an annular fuel region of 0.85 m thickness, and a 0.5 m thick controlled outer reflector region. There are 24 control rods positioned circumferentially outside the fuel ring. The 1-D benchmark is a cross section of the 2-D configuration taken along a plane perpendicular to the paper and passing through the center of the reactor and a control rod. The geometrical layout and subdivision into nodes are illustrated in Figure 1(b). Reflective and vacuum boundary conditions are imposed on the left and right boundary of the system, respectively.

Researchers tested the 2-D (x,y) code, REXTRAN, on the modified Henry-Worley and the HAFAS benchmark problems, with the goal of solving the C5G7 2-D NEA MOX benchmark problem. The results proved to be very accurate with low-order expansions. The discrete Legendre polynomials used in this coarse mesh code are derived for a uniform meshing scheme. Because modeling complicated geometries is difficult with the DORT code, the team adapted a Monte Carlo method to generate the response functions. The new method allows the use of continuous Legendre polynomials, thus avoiding meshing scheme issues. With this new adaptation, the researchers solved the C5G7 benchmark with excellent accuracy.

The researchers conducted additional 2-D (x,y) benchmark modeling. Figure 2(a) is an extension of the 2-D C5G7 MOX benchmark to a full core PWR. The team constructed a full core model from published data and calculated a reference solution using MCNP. This new benchmark was then successfully solved using the 2-D (x,y) coarse mesh code with great accuracy. The eigenvalue error was -0.02 percent, and the average and maximum pin power errors were 0.9 and 3.3 percent, respectively.

The researchers also constructed an updated version of the HAFAS benchmark problem, replacing the original fuel assembly with a GE9 fuel assembly. The solution to this benchmark is currently underway.

Finally, the team completed their derivation of the theoretical 3-D Cartesian geometry transport method during this fiscal year (see Mosher *et al.*, 2005).

Planned Activities

A no-cost extension was granted until December 31, 2005, to all institutions involved in this project. The main goal of the extension period will be the implementation of the new coupled (r,θ) transport/diffusion methodology in the PEBBED code. Response functions based on transport theory on curved geometries for the 2-D (r,θ) code will be generated. Work will also continue in producing uniform meshing schemes for heterogeneous lattices in 2-D (x,y) geometry, to explore a finer "staircasing" meshing scheme

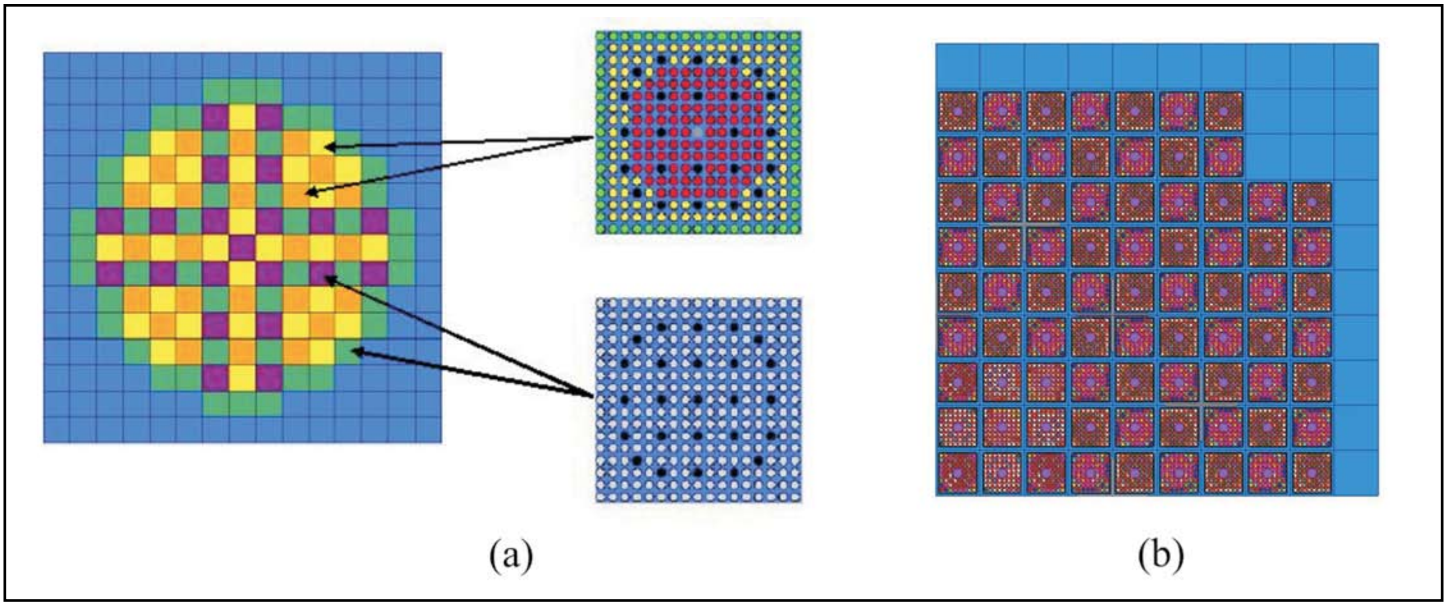


Figure 2. (a) full core PWR and (b) updated HAFAS benchmark.

for use in DORT. The extension period will also be used to finalize the 2-D updated HAFAS benchmark problem, which will then be used to test the coarse mesh method.

Publications

Researchers prepared a journal paper describing the new formulation of the coarse mesh method in one dimension:

- S.W. Mosher and F. Rahnema, "The Incident Flux Response Expansion Method for Heterogeneous Coarse Mesh Transport Problems," *Transport Theory and Statistical Physics*, submitted (2004).

Five conferences papers were also written and presented on various aspects of this project:

- J. Breen and B. Forget, "An updated HAFAS BWR benchmark," ANS Student Conference, Ohio State, April 14-16, (2005).
- B. Forget and F. Rahnema, "Improved Monte Carlo Adaptation of the Heterogeneous Coarse Mesh Transport Method," Monte Carlo 2005, Chattanooga, Tennessee, April 17-21, (2005).
- B. Forget and F. Rahnema, "High Order Spatial Expansion of the Incident Current in the Heterogeneous Coarse Mesh Transport Method," *Transactions of the American Nuclear Society*, 92, 725 (2005).
- S.W. Mosher and F. Rahnema, "A Decoupled Finite Element Heterogeneous Coarse Mesh Transport Method," *Transactions of the American Nuclear Society*, 92, 731 (2005).
- B. Forget and F. Rahnema, "New Eigenvalue Evaluation Technique in the Heterogeneous Coarse Mesh Transport Method," ANS Winter Meeting, Washington, November 13-17 (2005).

NUCLEAR ENERGY RESEARCH INITIATIVE

Advanced Extraction Methods for Actinide/Lanthanide Separations

PI: Michael Scott, University of Florida

Collaborators: 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 has encouraged the development of nuclear waste reprocessing technologies.

Nuclear fuel reprocessing is based on the dissolution of irradiated material in a nitric acid solution. These reprocessing operations produce high- and medium-level liquid wastes (HLLW and MLLW, respectively) containing different radioactive elements. In order to simplify the conditioning of such wastes, it would be highly desirable to separate the different radioactive components according to their lifetimes. Separation would decrease the volume of HLLW intended for disposal in deep geological repositories, such as Yucca Mountain. Instead, these wastes could 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 was 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 states, 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. Researchers tested the binding attributes of various ligand systems 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, 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 *u*ranium *ex*traction) reprocessing method. The TRUEX (*tr*ans*u*ranium *ex*traction) process is further used to co-extract the trivalent lanthanide and actinide ions from the HLLW generated during PUREX processing. 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 for the researchers to prepare. The steric and solubility properties can be tuned through an extreme range by 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 (Figure 1). They show considerable promise for actinide recovery from high-level liquid waste and for general clean-up operations.

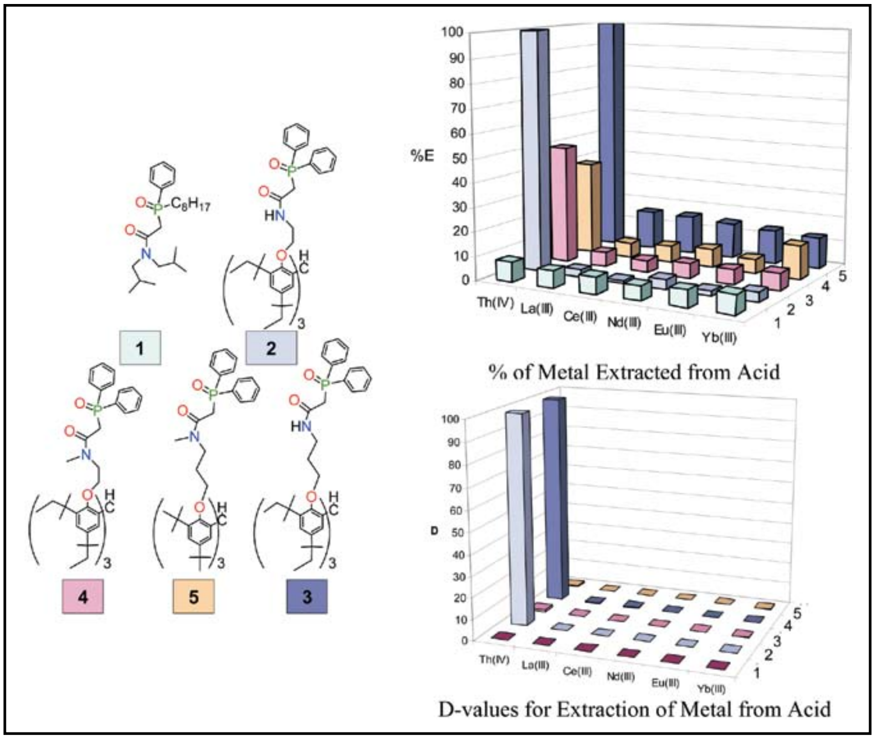


Figure 1. Extraction properties of CMPO ligands 1-5 developed at the University of Florida.

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 ANL for analysis with actinides. Ligands **2**, **3**, **4**, and **5** are all extremely selective for the Am (IV) ions such as plutonium in both dichloromethane and octanol (D values > 40) with only 10 equivalents of ligand in the organic phase, particularly when compared to the simple CMPO ligand **1** that is currently used for commercial processes. For Pu (IV), ligand **2** and **4** are particularly effective for extraction at very low concentrations, and they are among the most selective ligands available for binding of plutonium. Although the D value is low, ligand **4** does show an enhanced affinity (2.2 times) for Am (III) or Eu (III).

The coordination number of trivalent lanthanide hydrated salts in solution vary from 9 to 8 across the series, and in the solid state, the lanthanide aqua complexes adopt nearly rigorous tricapped trigonal prismatic (TTP) geometry. Inspired by the success of the DIAMEX industrial process, the researchers made an effort 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 in idealized TPP geometry; hence, it should be particularly adept at binding lanthanides and actinides. Researchers isolated large quantities of two different varieties of the ligand and examined their binding properties.

Typically, high distributions of trivalent f-elements in organic/acidic extraction systems are reported with approximately ~100,000:1 DGA to metal ion concentration [e.g., 0.2 M N,N′-dimethyl-N,N′-diphenyl-3-oxapenanediamide DMDPhOPDA, 10^{-6} M Ln (III)]. With the tri-DGA ligand (**15**), comparable distribution ratios can be obtained with a 10:1 ligand to metal ratio. To provide some context for the extraction of efficiency of **15** under controlled experimental conditions, the properties of ligand have been evaluated with respect to the performance of the related DGA, **16**. Since **15** contains three arms, the concentration of **16** was increased by a factor of three (Figure 2). This oversimplified comparison of preliminary

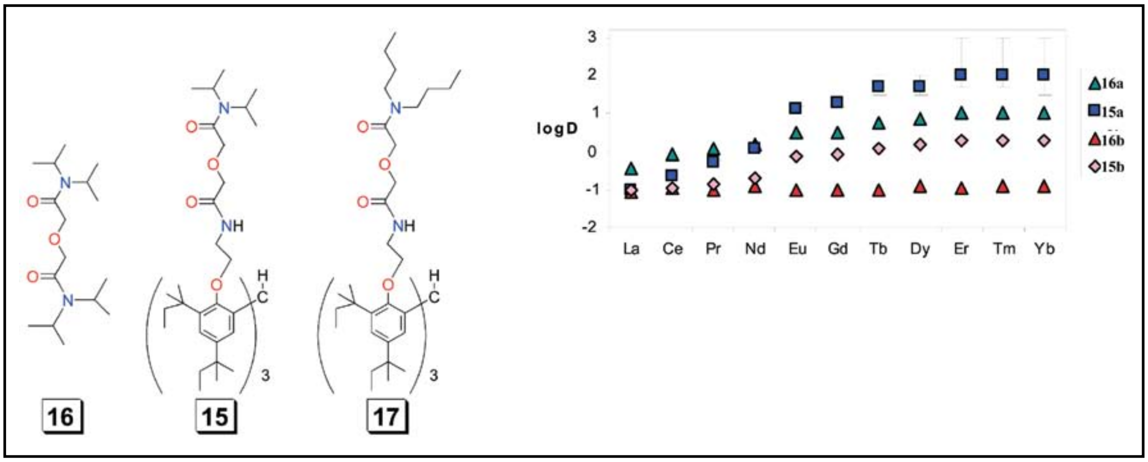


Figure 2. Extraction of 10^{-4} M solutions of Ln(III) in 1 M HNO_3 with solutions containing $3x10^{-3}$ M (16a) or $3x10^{-4}$ M (16b) and 10^{-3} M (15a) and 10^{-4} M (15b). Due to the limitations of the spectrochemical assay, error bars for D are large at high extraction efficiency (>98 percent).

data is used strictly to present the significant changes in the efficiency and selectivity of studied diglycolamide chelates. The delineation of the nature of extracted species requires further systematic investigation and will be published elsewhere.

An experiment with the reference molecule at 3x10^{-3} M (**15a**) showed the typical extraction pattern for diglycolamides, which gradually ascends across the lanthanide series. Once three DGA moieties were attached to the triphenoxymethane platform (**15**), the efficiency of the ligand for the heaviest lanthanides was remarkably improved. Only a ten-fold excess of ligand **15** is allowed for quantitative removal of trivalent erbium, thulium, and ytterbium from 1 M HNO_3 solution. The unique design of the rather flexible, nine oxygen donor cavitand allowed for much more sensitive ion size recognition than in the case of any other DGA chelates. Enhanced affinity of tris-DGA for the heaviest Ln and decreased attraction for the lightest (La, Ce, Pr), resulted in improved separation factor between the elements in the group. For instance, the value of the SF of Yb (III) and La (III) increased from SFYb/La=26 for **16a** to SFYb/La=1138 for **15a**. The tris-DGA ligand, **15**, perfectly satisfies the coordination preference of trivalent lanthanides for tricapped trigonal prismatic geometry, and the preorganization of these three DGA arms onto the triphenoxymethane platform causes significant enhancement in the extraction efficiency especially at low ligand concentrations. Further dilution of the organic phase to a 1:1 metal to ligand ratio maintained a high extraction efficiency for the heaviest lanthanides; over two-thirds of Tm (III) and Yb (III) was transferred into the organic phase. At the same time, with a three times higher concentration of **16b** (3x10^{-4} M), the extraction was negligible and no separation was observed suggesting necessity of the significant excess of ligand **16** to achieve appreciable extraction, and once again confirming the efficiency gain through the DGA preorganization in the compound **15**.

A significant modulation in the extraction pattern was noted when 1-octanol was substituted for dichloromethane as the organic solvent in the extraction with **15** at low concentrations. From a gradually rising affinity toward heavier lanthanides in dichloromethane, the extractant tends to favor the middle lanthanides in 1-octanol, in particular europium (Figure 3).

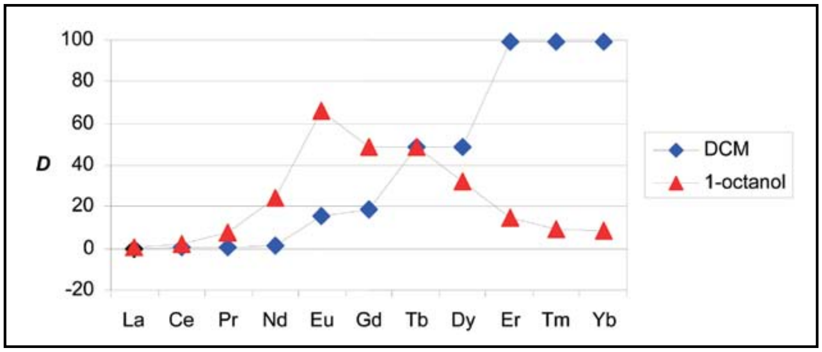


Figure 3. Extraction of 10^{-4} M solutions of Ln (III) in 1 M HNO_3 with dichloromethane (DCM) and 1-octanol containing 15 at 10^{-3} M.

Planned Activities

This NERI project is complete. 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 these ligands allows for the extraction of metal ions in highly acidic environments through the ability of the compound to buffer the effect of high acid concentration. While normally very high concentrations of ligand (10^{6} excess) are commonly used in liquid-liquid extraction schemes, the three-armed structures can obtain similar extraction profiles with only a ten-fold excess of the ligand in the organic phase. Hence, the amount of waste can be dramatically decreased. A variety of three-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. Argonne National Laboratory (ANL) will continue to study the actinide binding properties. Based on the results from the experiments at ANL, 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

Collaborators: Idaho National Laboratory

Project Number: 02-131

Project Start Date: September 2002

Project End Date: September 2006

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 its thermomechanical properties.

This project developed and applied techniques for measuring physical properties required for accurately modeling the TRISO fuel particle. In particular, researchers measured 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 used this information in updated models for predicting the failure of the TRISO pressure vessels during processing and under irradiation, and also generated 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 FY05, researchers commenced the study on neutron irradiation effects which had been carried over from the previous year. In addition, they completed the non-irradiated testing phase of this work. Irradiated 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 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 published results from three fracture strength testing methods for miniature tubular and hemispherical shell specimens. The first two methods were based on internal pressurization using elastomeric inserts for hemispherical and tubular shell specimens. The third method was 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.

Researchers completed irradiation of the neutron irradiation vehicles developed for testing the different specimen materials—one consisted of extremely tiny and fragile coating samples, and the other, strained thin strip ceramic samples. The first design used a powder-bed rabbit capsule that irradiated tiny samples in a bed of fine silicon carbide powder encapsulated in a silicon carbide-lined molybdenum case. The powder bed contained a number of irregular-shaped fragile samples to accommodate swelling of the samples without damaging them, and provided sufficient temperature homogeneity over the entire capsule interior. The second design was a rabbit capsule for bend stress relaxation (BSR) creep. In the BSR capsule, bend-strained thin strip samples (50 microns thick) of silicon carbide and zirconium carbide were tightly held during irradiation in a curved gap within a fixture made of silicon carbide. Irradiation creep strain rates were estimated based on the extent of stress relaxation following irradiation as a function of neutron fluence. The delay in the operation of High Flux Isotope Reactor (HFIR) and material fabrication caused a significant delay in starting the irradiation program; fortunately, it also resulted in sufficient time to develop these novel irradiation experiments. The post irradiation analysis of materials from these irradiation studies is currently underway. Three papers containing results from these capsules have already been submitted to the *Journal of Nuclear Materials*.

Monolithic SiC and zone-refined ZrC specimens irradiated in the METS (Mapping Elevated Temperature Swelling) core capsules are currently undergoing post irradiation examination. The METS capsules are made up of three HFIR in-core vehicles which are irradiated in the approximately 2, 4, and 10 dpa ranges for temperatures from 600-1,500°C. The two lower fluence capsules have finished irradiation.

Planned Activities

This NERI project received a no-cost extension and funds have been saved to carry out FY06 activities. These activities will include using the techniques described above to perform post-irradiation examination of capsules. A wealth of information is expected on the high-temperature of SiC and the state of the PyC/SiC interface following irradiation. A reference handbook will be assembled with thermomechanical property information including this new data, which will be invaluable to fuels designers.

NUCLEAR ENERGY RESEARCH INITIATIVE

Enhanced Thermal Conductivity Oxide Fuels

PI: Alvin Solomon, Purdue University

Collaborators: Framatome, ANP

Project Number: 02-180

Project Start Date: September 2002

Project End Date: September 2005

Research Objectives

The objective of this project was to develop a processing methodology to enhance the normally very low thermal conductivity of oxide nuclear fuels by incorporating a small volume fraction of a high-conductivity solid, *continuous* phase that is 1) stable and compatible with UO_2 and zircaloy, 2) does not impose significant neutronic penalties, and 3) is commercially realizable. The researchers have dubbed these "ECO" fuels. Higher thermal conductivity has the promise of greater operating flexibility, higher fuel ratings, higher burnup, and reduced waste generation.

The original approach proposed was to produce sintered UO_2 fuel with open interconnected porosity, and to impregnate this fuel with an organic precursor for silicon carbide (SiC) using polymer impregnation and pyrolysis (PIP) to form a stable interconnected high thermal conductivity phase of near stoichiometric silicon carbide. Although many other organic precursors exist, SiC has the highest conductivity in dense pure form and is readily available commercially. Such precursors have been commonly employed to form ceramic matrix composites, but not in commercial nuclear fuels.

Research Progress

Researchers measured the thermal properties of ECO fuels by thermal diffusivity coupled with density and specific heat determination. The research was bolstered by theoretical and experimental studies of structural/chemical stability of the two-phase structure, thermal property correlations, and finite element heat transfer codes to optimize the microstructure (fraction of high-conductivity phase, spacing, scale, and pore structure) and to obtain thermal responses of the optimized structures to steady-state and transient reactor conditions using the COPERNIC code and for neutronics, CASMO-3, and SCALE-5.0.

Although this research was directed towards light water reactor fuels, the results and techniques can also be applied to fast reactor fuels, gas-cooled reactor fuels, and inert matrix fuels, and experiments were carried out to demonstrate these possibilities. In addition to using the PIP processing approach, researchers employed a second technique: "co-sintering" uranium dioxide fuels with a high conductivity, *continuous* beryllia phase. They found that this approach was feasible and could be easily incorporated into commercial fuel processing methods. They examined issues associated with beryllia toxicity, which is necessary to address before employing this approach in a commercial setting. Researchers also examined fuel reprocessing and recycling issues for both processing approaches and investigated the costs of incorporating the high-conductivity phase in current PWR's. In the final step, the researchers prepared specimens for irradiation.

The results of analytical thermodynamic studies showed that the reactions between SiC and UO_2 at temperatures approaching 1,400°C and above depend on the release of two gaseous species, carbon monoxide (CO) and silicon oxide (SiO). Calculations show that the equilibrium partial pressures of these two species at 1,500°C are below 1 atm. Compatibility experiments with high-purity β-SiC and sintered high-density UO_2 verified these reactions, and characterized the reaction products and degree of reaction for various times. The reaction products under these conditions was a uranium silicon phase, $USi_{1.67}$ previously reported in the literature, and other proposed solid phases. Scanning electron microscope (SEM) examination after processing PIP specimens at temperatures above 1,400°C showed only interdiffusion of carbon into the UO_2 to form a uranium oxycarbide and a uranium carbide phase, leaving a Si-rich species behind.

Published experiments showed stability between SiC and UO_2 for reasonable times to temperatures of 1,800°C in sealed containers. Therefore, the researchers fabricated a special set of nested Y_2O_3 crucibles to allow evacuation around the specimens and backfilling with pure argon, to restrict the escape of SiO and CO. As the nested crucibles did not completely eliminate the reactions, they restricted all processing of PIP-SiC to 1,300°C for 15 hours, where no reactions were observed.

To perform PIP-SiC impregnations, researchers first developed a new "slug-bisque" granulation technique to form a stable open pore structure of 5-15 volume percent in sintered UO_2, with the UO_2 phase at 98 percent theoretical density to maximize the fuel and fissile atom density. They then investigated many processing techniques to increase the density of the "PIP-SiC" phase and to simplify and/or optimize the processing. Results indicated that the temperature and measured viscosity of the precursor, along with the pressure applied during impregnation, were important factors to maximize the PIP-SiC density, especially as the open porosity was progressively filled. After six to nine impregnations at 60°C and 4 atm, the researchers obtained a maximum SiC density of approximately 80 percent. From microstructural examination, the SiC phase exhibited intimate contact with the UO_2 phase and uniform impregnation throughout the specimen, but showed pockets of porosity that were not completely impregnated, Figure 1(a). They then developed techniques to simulate the SiC phase by impregnating SiC powders with the precursor to approximately the same density. Measurement of the thermal conductivity of this SiC phase produced at 1,300°C was dramatically lower than the SiC phase produced at 1,500°C. Therefore, it was not unexpected that the UO_2/PIP-SiC specimens processed at 1,300°C did not show increased thermal conductivity. Finite element modeling using the *measured* conductivity of the SiC phase in the UO_2/PIP-SiC specimens was reasonably consistent with the measured values.

To show that the PIP-SiC processing technique has merit when the processing temperatures can be higher, researchers also consolidated simulant TRISO microspheres with SiC outer layers using SiC filler particles between the spheres [Figure 1(b)] and UC microspheres for gas-cooled reactor applications [Figure 1(c)], both processed at 1,600°C. The advantages of the PIP-SiC process are 1) the spheres are not damaged during consolidation, 2) a highly conductive, stable matrix can be readily formed in any desired geometry, and 3) they can be safe during core disruptive accidents.

Beryllium oxide (BeO), with the highest thermal conductivity of any oxide, has the major advantages of being stable in contact with UO_2 up to approximately 2,160°C, and also has the same thermal conductivity as SiC in pure, dense, defect-free form. Above that temperature, a eutectic can form, but the kinetics of the reaction were not investigated.

A technique of "co-sintering" UO_2 "green" granules uniformly coated with BeO powder proved to be successful in yielding a continuous BeO phase surrounding the UO_2 granules (Figure 2). In this case, because of the sintering compatibility of the two phases under reducing conditions, both phases are of high density. Figure 2(a) shows a ten volume percent BeO specimen, but five volume percent specimens were also produced. It is important to note from Figure 2 that the "contamination" of the BeO phase by UO_2, resulted in a fine BeO grain size. This is beneficial in delaying or inhibiting the phenomenon of fast neutron-induced cracking in BeO at high doses.

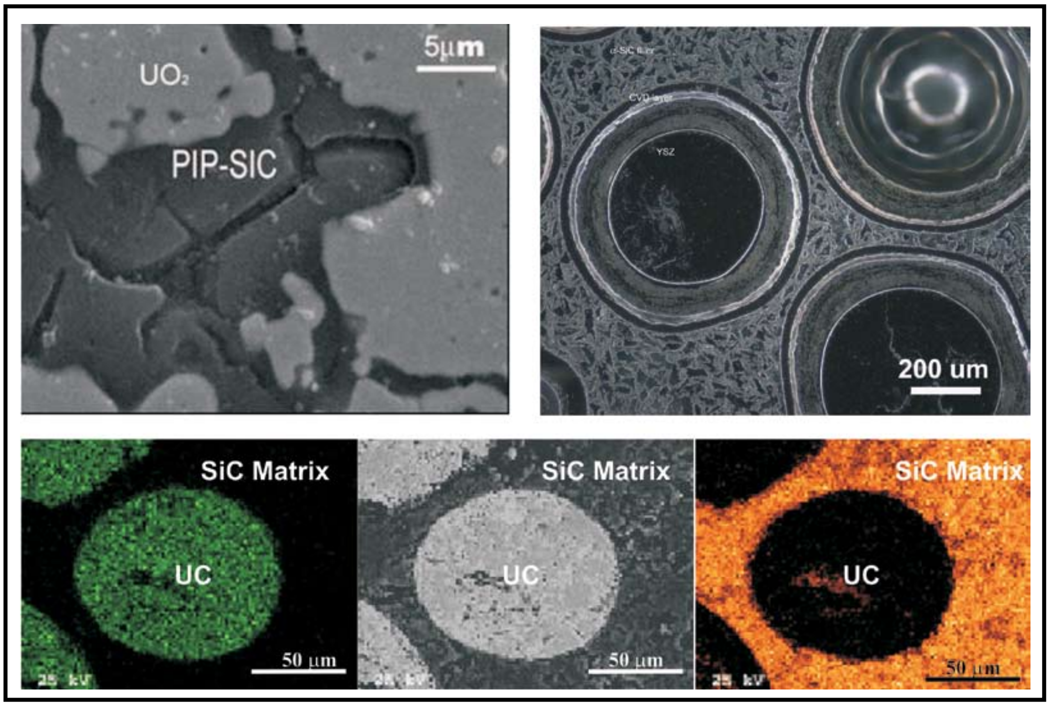


Figure 1. PIP-SiC processing methodology applied to (a) ECO fuels, (b) TRISO fuels, and (c) HTR fuels. SEM of UC microspheres is shown in the center, with scans for U (left) and Si (right).

The measured thermal conductivity of the 10 volume percent specimen, Figure 2(b), shows a ~50 percent increase, which is consistent

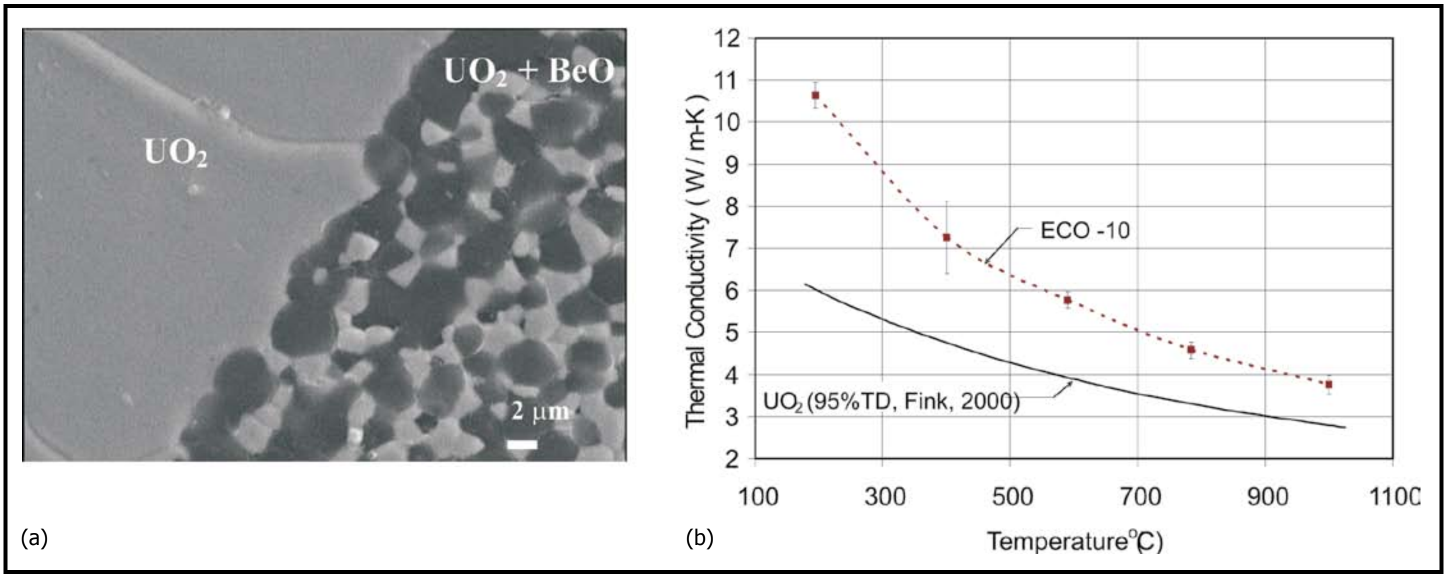


Figure 2. (a) Co-sintered "green" UO$_2$ granules and BeO showing the fine BeO grain size due to UO$_2$ dispersion in the BeO; (b) the measured thermal conductivity of the 10 vol percent BeO ECO fuel vs. standard UO$_2$ fuel from Fink's correlation (J. Nucl. Mat. 279 (2000) 1-18). The ~50 percent increase in thermal conductivity is seen at all temperatures.

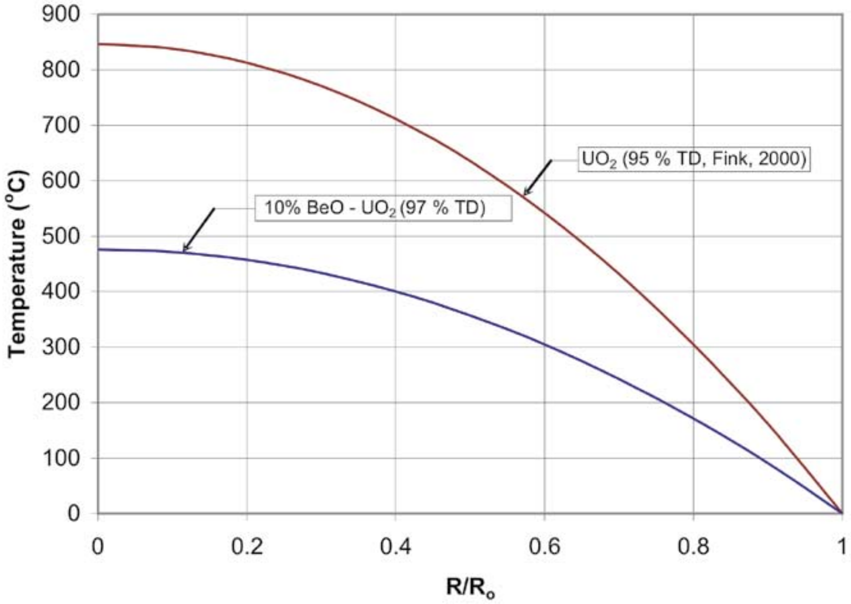


Figure 3. Temperature profiles for UO$_2$ and UO$_2$ /10 vol percent BeO fuel for a PWR hot channel at 17.5 kWm^{-1} and a peaking factor of 1.7. oxide fuel surface temperature is 576°C.

with the literature. This translates to a 380°C reduction in centerline temperature for the hot channel of a PWR at 17.5 kW/ft (Figure 3).

Although researchers did not experimentally examine all possible microstructural variants, the ANSYS finite element code indicated an insensitivity to the "scale" of the microstructure from coarse to fine granules on thermal conductivity. However, optimization would be desirable, especially at small volume fractions of BeO.

In contrast to the PIP-SiC process, the co-sintering process varies little from standard commercial processing of oxide fuels, except for the granule preparation and mixing with a small quantity of BeO powder. However, this led to an examination of the toxicity issue in terms of federal regulations (i.e., 10 CFR850) for handling BeO in DOE facilities. The pellet grinding step was considered the most problematic. Analyses showed that the controls used for UO$_2$ should be adequate for BeO, even when performed dry. Wet centerless grinding would give an additional factor of safety.

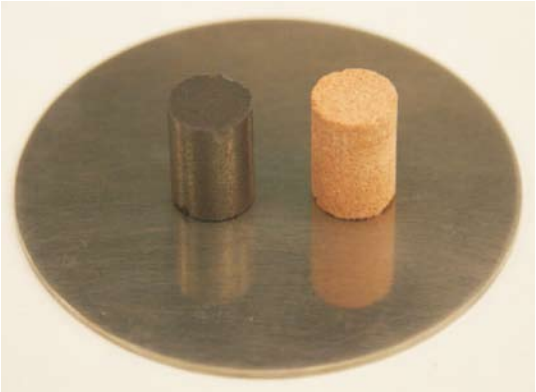


Figure 4. UO$_2$/ 10 vol percent BeO ECO fuel pellet before and after dissolution in boiling HNO$_3$ for 1 hour.

Another issue that is difficult to quantify is the three-dimensional continuity of the BeO phase. This was easily visualized by dissolving the UO$_2$ phase in boiling nitric acid to leave the BeO "skeleton." Figure 4 shows typical UO$_2$/BeO pellets and the BeO skeleton. X-ray analysis showed that essentially no UO$_2$ remained, which demonstrates the possibility of reprocessing the UO$_2$ and possibly the BeO as well.

Based on the success of the thermal conductivity enhancements for the UO$_2$/BeO ECO fuel, a number of performance and cost analyses were carried out using the COPERNIC Code. Assuming the core was either all standard or all ECO fuel of equal enrichment, the ECO fuel with less uranium must go to 4 percent higher burnup to obtain the same total energy extraction per fuel assembly. For the calculations, it was assumed that the fuel temperatures are reduced by 10 percent. (This is reasonable for 5 volume percent BeO, but conservative for the 10 volume percent BeO fuel where the thermal conductivities are 50 percent higher at all temperatures, which reduces average temperatures by about 200°C) Comparing standard UO_2 and 5 volume percent BeO ECO fuel at end-of-life (EOL), the fission gas release is reduced from 17 to 11 percent and the pin plenum pressure is reduced from 160 to 128 bar. A LOCA initialization transient analysis was also performed. When compared to standard UO_2 fuel, the ECO fuel provides an increase in the maximum linear heat generation rate by 4.9 kW/m at BOL, and 3.2 kW/m at EOL (62 GWd/MTU).

The researchers also performed neutronics calculations by comparing results with previous Babcock and Wilcox reactor analyses. In this case, the enrichments of the ECO fuels were increased to give the same core-averaged k_{inf}, and the fuel temperatures were again reduced 100°C. Both SiC and BeO type ECO fuels were considered, but while CASMO-3 was suitable for SiC, the (*n*, 2*n*) reactions in Be necessitated using the TRITON control module of SCALE 5.0 with the 238-energy group cross-section library. The TRITON calculations are performed with an appropriate cross-section library for beryllium, and to establish the amount of depletion of the beryllium isotope for three cycles of operation. Both the finer energy-group structure and the use of ORIGEN-S mean that TRITON is better able to treat (*n*, 2*n*) reactions of beryllium.

Comparison of the results for the two ECO fuel types revealed a striking difference in their neutronic performance. Substitution of UO_2-SiC fuel for UO_2 fuel required an increase in enrichment by 0.16 weight percent to maintain the end-of-cycle value of k_{inf}. In contrast, substitution of UO_2-BeO fuel for UO_2 fuel required an increase in enrichment by only 0.0073 weight percent to maintain the end-of-cycle k_{inf} value. The consumption of ^{9}Be through (*n*, 2*n*) reactions is only 0.15 percent over three cycles of irradiation.

Researchers also estimated the relative costs of ECO versus standard fuel assuming 60 and 56 subassemblies, and the additional enrichments necessary to maintain the same end-of-cycle k_{inf}. A significant factor is that there is less uranium in the ECO fuels so, despite the higher enrichments, the net result for the 60 subassembly core is a predicted *decrease* in uranium cost of about \$139,000 per batch for UO_2-SiC fuel and \$501,000 per batch for UO_2-BeO fuel.

A reduction in the reload batch size from 60 to 56 is expected to impose penalties in terms of increased power peaking, but these may be acceptable with ECO fuel because of its improved thermal conductivity. It was also assumed that an increase in the ^{235}U enrichment by 0.25 weight percent would be required to maintain cycle length. The same increase in enrichment was assumed to apply for all three fuel types. The increase in enrichment was strictly an engineering judgment and was not supported by a specific neutronic analysis or core design. Therefore, it is not appropriate to compare costs between batches with 56 assemblies and those with 60. The *decrease* in uranium cost per batch is \$236,000 for UO_2-SiC fuel and \$506,000 for UO_2-BeO fuel. Using \$115/lb (or \$253/Kg) as the cost of BeO, and assuming 5 volume percent BeO gives an additional cost of \$350,000 for BeO at 95 percent theoretical density.

In addition, the net *savings* from reducing the batch size is estimated to be between \$100,000 and \$200,000. The reduction in uranium cost for UO_2-BeO fuel is sufficient to offset an increase in manufacturing cost of nearly \$20/kg U, whereas the reduction in uranium cost for UO_2-SiC fuel would offset an increase in manufacturing cost in the range of about \$5 to \$10/kg U. However, the economic impact of using ECO fuel extends beyond the fuel cost. The improved thermal performance of the fuel could support less restrictive peaking, operating, and maneuvering limits, and less restrictive limits can contribute to the economic performance of a power plant. Selective use of ECO fuels in critical high-power regions would also be possible.

Planned Activities

The current project is complete, and no further activities are planned.

The next step for developing and demonstrating the performance of ECO fuels would be irradiation experiments. The researchers have considered irradiations in the DOE Advanced Test Reactor (ATR), and have examined techniques to produce specimens with the required diameter for the LWR-2 irradiations. Proton beam irradiations would also be very interesting in terms of materials optimization at lower cost and difficulty than reactor irradiations, but the effects of concurrent fissioning are not present.

The remaining question concerns the radiation stability of BeO at high fast neutron fluences. BeO is an excellent moderator, but in previous experiments conducted nearly 50 years ago with bulk BeO and dispersed UO_2 granules, microcracking was observed near the end-of-life doses—a phenomenon that is not well-understood since it is sometimes also not observed at the same doses for different grain sizes and specimens. ECO fuels differ from those experiments because they contain much lower volume fractions of BeO surrounded and constrained by UO_2, and traversed by fission fragments which appear to be beneficial to delay microcracking in the earlier experiments.

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: January 2006

Research Objectives

The primary objective of this project was to assess the feasibility of improving the performance of light water reactors (LWRs) by using hydride fuels instead of oxide fuels.

This research is expected to result in a number of improvements in LWR economics, safety, and plutonium incineration capability. By increasing the power density of LWR cores, the cost of electricity is reduced. Safety is improved due to the inherent negative temperature reactivity effect of hydride fuel and the reduced negative void reactivity coefficient of BWRs. BWR fuel bundle design is significantly simplified by eliminating water rods, water gaps, and partial length fuel rods and by reducing the number of fuel enrichment levels. By increasing the hydrogen-to-heavy metal atom ratio, the capability of incinerating commercial and military plutonium is greatly enhanced. Hydride fuel also provides new options for utilizing thorium in LWRs.

Research Progress

Following were the objectives for the third year of this research:

- Search for an optimal burnable poison for hydride fuel in PWRs
- Search for maximum power attainable from PWR cores designed with wire-wrapped hexagonal fuel assemblies using either hydride or oxide fuel
- Estimate the minimum cost of electricity (COE) attainable from hydride- versus oxide-fueled PWR cores of the wire wrapped design
- Assess the feasibility of improving the incineration of plutonium in PWR using hydride fuel
- Perform neutronic and thermal-hydraulic analyses of hydride- versus oxide-fueled BWR cores

Optimal burnable poison for hydride fuel in PWR. Researchers investigated the effectiveness of four types of burnable poisons as well as of thorium to make negative all the reactivity coefficients of the hydride-fueled core with the maximum possible power density. The burnable poisons considered were gadolinium, natural erbium, erbium enriched to 100 percent ^{167}Er, and integral fuel burnable absorbers (IFBA). The latter is a boron-containing compound deposited on the surface of fuel pellets. Researchers found that neither gadolinium nor IFBA can provide a negative coolant temperature coefficient of reactivity. Fortunately, erbium and thorium can. However, use of natural erbium penalizes the attainable discharge burnup by approximately 15 percent. Use of erbium enriched to 100 percent ^{167}Er reduces the penalty in the attainable discharge burnup to approximately 7 percent. By using thorium, it is possible to provide negative reactivity coefficients without imposing a penalty on the attainable discharge burnup.

Wire-wrapped PWR core designs. Preliminary investigation indicated that for tight lattices, wire-wrapped fuel bundle designs significantly reduce coolant pressure drop across the core in comparison with grid spacer designs having the same hydrogen-to-heavy metal (H/HM) ratio. As a result, researchers undertook a detailed comparison of U-$ZrH_{1.6}$ and UO_2 PWR core designs using wire wraps in a hexagonal array. This comparison included a steady-state and transient thermal hydraulic analysis, vibration and fretting analysis, as well as economic analysis. It covered a wide range of fuel rod diameters (D) and lattice pitch (P) as well as several uranium-enrichment levels. The neutronic characteristics of the wire wrap designs were inferred from the parametric study performed for a square lattice with grid spacers by assuming that wire wrap and grid spacer designs featuring the same fuel type, fuel rod diameter, and H/HM ratio are neutronically identical. The transient analyses considered LOCA and overpower events.

Researchers found that the optimal wire wrap designs offer significantly higher power density than the grid-supported reference core design, possibly allowing for power uprates of up to approximately 50 percent. This is due to improved vibrations, pressure drop, and critical heat flux performance of the wire wrap designs. As a result, the cost of electricity from a PWR that is uprated to the maximum power offered by optimal wire wrap design is estimated to be nearly 40 percent lower than that of the reference PWR design. The thermal hydraulic and economic performance of U-ZrH$_{1.6}$ and UO$_2$ are very similar, although the hydride fuel shows some cost advantage. For recycling plutonium in LWRs, however, hydride fuel shows a distinct advantage over oxide.

Improved plutonium incineration in PWRs. Researchers compared the plutonium transmutation capability of PWRs loaded with U-PuH$_2$-ZrH$_{1.6}$ (PUZH) fuel against that of uniformly loaded mixed U-Pu oxide (MOX) fuel. They found that use of hydride fuel offers the following improvements in PWR performance over oxides:

- Inventory of Pu required to provide the same cycle length is only 74 percent, implying lower cost of fuel
- Close to two times the discharge burnup, yielding smaller HM inventory to be handled or disposed of per unit energy generated
- Incineration of ~60 percent of the loaded Pu in one pass through the core versus less than 30 percent for homogeneous MOX fuel assemblies resulting in faster reduction of Pu inventory, smaller amount of actinides in the waste streams, and improved economics of Pu incineration
- Smaller fissile Pu fraction in the discharged fuel (44 percent versus 63 percent) implying higher spontaneous neutron yield and higher decay heat per gram of Pu or TRU, making the discharged fuel more proliferation resistant
- Somewhat smaller neutron source strength and decay heat per fuel assembly due to reduced TRU amount, simplifying the handling of discharged fuel assemblies

A very preliminary economic analysis indicated that the cost of a PUZH fuel assembly could be about 80 percent of the cost of a MOX fuel assembly operating at the same power density for the same cycle length. PuH$_2$-ThH$_2$-ZrH$_{1.6}$ hydride fuel was also found very promising for plutonium recycling in PWR. The superior incineration capability of the hydride fuel is due, in part, to the softer neutron spectrum it offers due to its high H/Pu ratio.

Neutronics of hydride-fueled BWRs. The neutronic analysis of hydride-fueled BWRs has proceeded in two parts. First, researchers conducted a simplified one-dimensional parametric study covering a wide range of diameters (from 0.65 to 1.5 cm) and P/D (from 1.05 to 1.6). Second, they performed a 3-D analysis of BWR fuel bundles, accounting for all the heterogeneities, including control rods. The objective of the 1-D parametric study was to determine the attainable discharge burnup from that combination of diameter and P/D that will make all reactivity coefficients sufficiently negative. They developed a procedure to account for the strong axially varying water density and for the four-batch fuel management strategy in processing the results from the 1-D calculations.

The 3-D analysis yielded attainable discharge burnups, pin-by-pin power distributions, axial power shapes, reactivity coefficients, reactivity worth of control elements, and preferable burnable absorber. The reference GE BWR/5 9x9 fuel bundle used for this analysis has two large water rods that occupy seven spaces of the square lattice, reducing the number of fuel rods to 74, eight of which are partial length rods. The average bundle enrichment is 3.90 percent. For the hydride fuel bundles considered, the control blades were eliminated and the water gap thickness between fuel bundles was reduced to 1 mm to enable bundle insertion and removal from the core. The number of rods per bundle was increased from 9x9 to 10x10, while maintaining the same pitch as the reference BWR bundle. All the water rods and partial length fuel rods were eliminated. Of the 100 lattice sites, 96 are occupied by full-length fuel rods and four by guide tubes for cylindrical control rods. The fuel rod diameter was adjusted to maximize the achievable burnup while assuring negative temperature and void coefficients of reactivity, as well as to maximize the achievable power. Following are a number of observations from the 3-D analysis:

- The hydride fuel burnup is significantly higher than that of the oxide fuel even though the ^{235}U inventory per bundle is somewhat lower. The ^{235}U enrichment, though, is higher for hydride fuel.
- The hydride fuel cycle length is shorter by ~20 percent although the HM inventory is lower by more than 30 percent.
- The reactivity margin provided at full power by the insertion of control elements is practically the same— \$16 for the oxide, versus \$16.5 for the hydride fuel design.

- The radial power distribution across the hydride fuel bundle is flatter all along the fuel life, relative to the reference oxide fuel bundle that is very heterogeneous.

Researchers concluded that U-$ZrH_{1.6}$ fuel can significantly simplify the BWR fuel bundle design by eliminating water rods, partial-length fuel rods, and relatively wide water channels between fuel bundles and by using uniform radial enrichment level. On the other hand, the HM inventory of the hydride fuel core is smaller than that of the reference oxide fuel core by 32 percent so that the ^{235}U loading per core is somewhat smaller for hydride fuel. However, the reference cycle length can be achieved by using a somewhat higher enrichment for the hydride fuel.

Thermal-hydraulics of hydride-fueled BWR. The search for the maximum achievable power considered both oxide fuel (as a reference) and hydride fuel, and performed over 400 fuel bundle geometries encompassing rod diameters ranging from $0.6 \le D \le 1.6$ cm and pitch from $1.1 \le P/D \le 1.6$. This search was based on a steady-state, whole core, thermal-hydraulic analysis and accounted for the following constraints: minimum critical power ratio (MCPR), fuel peak temperature, fuel average temperature, clad surface temperature, core pressure drop, fuel rod displacement-to-diameter ratio (used as a measure for vibration induced constraint), and coolant flow rate-to-core power ratio (assuring similar exit quality for similar coolant inlet temperature). The Hench-Gillis correlation was used for the MCPR calculations. Two values were considered for the maximum pressure drop: the reference BWR value of 24.74 psia and a nearly 50 percent higher value of 38 psia assumed attainable in the near future. The inlet and outlet coolant enthalpy, inlet coolant pressure, axial power profile, and peak bundle to average bundle power ratio was assumed to be the same for all of the designs.

Researchers found that the implementation of a hydride fuel core into a BWR/5-size vessel could result in a 45 percent gain in power with respect to the oxide core used as reference. About 19 percent out of the 45 percent are attributed to an increase in the number of fuel rods per given core volume due to the elimination of the water rods and bypass channels. The additional 26 percent power increase is due to the power-based optimization of the lattice parameters, i.e., fuel rod diameter (~0.8 cm), pitch-to-diameter ratio (~ 1.5-1.6), and fuel lattice size (14×14).

Researchers examined in greater detail bundle designs that feature the reference oxide fuel lattice pitch and equal, or slightly larger, fuel rod diameter. A number of 10×10 hydride fuel bundles, including the bundles for which researchers performed a detailed three-dimensional neutronic analysis described above, were analyzed by means of a subchannel analysis performed with the VIPRE-EPRI code. Researchers found that hydride fuel bundles can be designed to deliver nearly 40 percent higher power than equal volume oxide fuel bundles. This remarkable possible increase in the power density justifies embarking upon a thorough assessment of the feasibility of commercializing hydride fuel for BWR.

Planned Activities

This project has been granted a no-cost extension until January 31, 2006. This period will be used for completing the analysis of the promise of hydride fuel in BWR and for summarizing the project. The summary is planned to be 10 to 12 papers, published in a special issue of *Nuclear Engineering and Design*.

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 Laboratory

Collaborators: Georgia Institute of Technology, Pennsylvania State University, Pebble Bed Modular Reactor Company, PBMR (Pty.) Ltd., and University of Arizona

Project Number: 02-195

Project Start Date: October 2002

Project End Date: September 2006

Research Objectives

This project was a comprehensive effort to develop analysis methods for pebble-bed reactors (PBRs) based on modern analytical nodal methods for solving the neutron diffusion equation and efficient techniques for node homogenization. Researchers will incorporate (or tightly interface) the nodal methods and node homogenization techniques into the Idaho National Laboratory’s (INL) 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 feat 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. One objective of this project is to implement these advancements. Additional planned enhancements to PEBBED include incorporating techniques to account for the dependency of reaction cross sections on thermohydraulic feedback and incorporating a genetic algorithm to facilitate design optimization.

Research Progress

As originally written, the PEBBED code applied traditional finite-difference techniques to solve the neutron diffusion equation, organized in a modular form. This modular form allowed 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 INL.

Early in the project, the investigators decided to apply the nodal solution through the Coarse-Mesh Finite-Difference (CMFD) approach, which took advantage of the existing finite-difference framework in PEBBED. Initially, they applied an iterative scheme, 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 both the radial and axial directions.

In the azimuthal direction, the original solution involved the computation of a series of hyperbolic Bessel functions. Evaluating this series consumed so much computational time that it offset the advantages of a nodal solution. 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. These algorithms mimic the process of biological evolution; that is, adaptive traits are favored in future generations. Under this NERI project, researchers developed 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 INL. Application of the algorithm facilitated the design of passively safe PBR cores of up to 700 megawatts thermal, a much higher power output than previously thought possible.

The approach, initially conceived for cross section homogenization, is a straightforward application of perturbation techniques developed by the Georgia Institute of Technology. However, as research progressed, discrepancies were found 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 (i.e., discrete fuel particles and discrete pebbles), and the inadequacy of available methods to account for it. Specifically, the Dancoff factor was never computed correctly for doubly heterogeneous pebble bed 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 developed methods that improve the treatment of these low-lying resonances and that calculate the Dancoff factor rigorously in randomly doubly heterogeneous media.

When generating PBR cross-section libraries, one must correctly model the spectrum dependence, which is a function of the material compositions of the surrounding pebbles. In addition, temperature feedback must also be accounted for. Two thermal-hydraulic software packages are available to this project, one of which (THERMIX) has been coupled to PEBBED for accurate calculation of temperature distributions.

In the most recent year, researchers have completed incorporating a comprehensive methodology for thermal feedback into PEBBED and successfully tested the algorithm. They have concurrently finished developing a comprehensive methodology for quick nuclear data adjustment to account for the effects of depletion and the influence of neutron leakage from neighboring zones. Within the PEBBED code, the previously developed radial-direction nodal solution has been replaced by a new, more robust, and mathematically rigorous formulation that does not suffer convergence problems.

During the past year, this project has resulted in the publication of seven papers at international conferences and the submittal of three journal articles, one of which has been accepted and is in press. The remaining two are still in the review process.

Planned Activities

The project has received a no-cost extension for one year (to September 30, 2006) so researchers can complete the integration of the various elements accumulated during the previous years into a cohesive package that automates most of the interfacing between the various components. In addition, they will finalize the coding and testing of an analytical nodal treatment for the theta direction, thus completing the nodal solver within the PEBBED code. The resulting nodal depletion capability will be tested and used to analyze non-proliferation characteristics of the PBMR-400 reactor design and possibly optimize its fuel cycle.

Fundamental Nuclear Sciences Projects

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 2006

Research Objectives

The primary objective of this project was to develop a comprehensive model to predict performance and assess the risk of failure of Zircaloy fuel cladding in commercial boiling water reactors (BWRs) and pressurized water reactors (PWRs) under high burn-up conditions.

Research Progress

Task 1: Modified Boiling Crevice Model. Researchers have modified the Boiling Crevice Model (BCM) to describe the evolution of the environment in crud pores and in contact with the cladding Zircaloy surface. The model considers heat transfer conditions under both low superheat (nucleate boiling in PWRs) and high superheat (sustained boiling in BWRs) conditions. Boiling is believed to be the mechanism for: 1) concentrating Li^+ and $B(OH)_4^-$ ions in crud pores on the fuel cladding, and; 2) the precipitation of $LiB(OH)_4$, which results in the Axial Offset Anomaly (AOA). Therefore, researchers have derived the concentration factor as:

$$CF \equiv \frac{C_s}{C_0} = \exp\left(\frac{qL}{h\rho\varepsilon D}\right)$$

where C_s is the concentration in contact with the Zircaloy cladding, C_0 is the concentration of bulk solution, q is the heat flux (j/cm^{2}s), L is the thickness of crud deposit, h is the latent heat of vaporization (j/g), ρ is the density (g/cm^3), ε is the porosity, and D is the superficial diffusion coefficient. The concentrations of lithium and borate ions that govern whether a precipitate will form are the bulk concentrations multiplied by the respective concentration factors determined by the above equation. Non-volatile species tend to concentrate in a porous deposit layer overlaying a boiling surface.

Task 2: Development of the Point Defect Model. The objectives of this task are to:

- Develop point defect models (PDM) for the oxidation and corrosion of zirconium alloys based on the coolant conditions that exist in BWR and PWR cores.
- Acquire values for various model parameters for the growth of bi-layer passive films on zirconium and Zircaloys under simulated PWR- and BWR-like coolant conditions, where hydride or oxide barrier layers form. Both of these barrier layers are covered by porous oxide outer layers.

In this task, researchers have modified the point defect model to develop impedance models for the growth of the passive films formed on zirconium and Zircaloys in high temperature hydrogenated and dehydrogenated aqueous solutions, respectively. They used optimization techniques to extract the model parameter values from electrochemical impedance spectroscopy data for zirconium measured in borate buffer solutions (pH 6.94) at 250°C.

Typical experimental and simulated impedance data and extracted parameter values are shown in Figure 1. The impedance models based on the PDM provide a reasonable account of the growth of the bi-layer passive films described above. The extracted model parameter values can be used, for example, in deterministic models to predict the accumulation of general corrosion damage to Zircaloy fuel sheath under BWR and PWR operating environmental conditions.

Task 3: Incorporation of the PDM into the Mixed Potential Model. This task is to incorporate the modified point defect model into the mixed potential model (MPM), in order to calculate the corrosion potential and general corrosion rate and, hence, to describe the oxidation of Zircaloy under open circuit conditions. The researchers have developed the required algorithms and

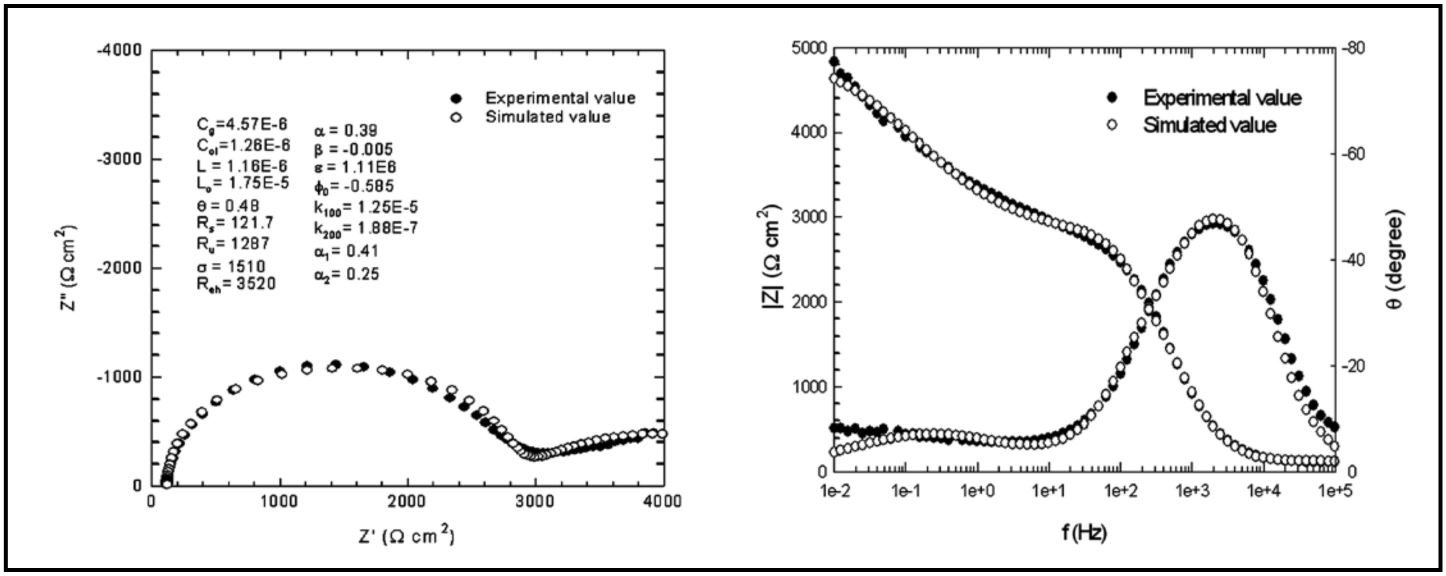


Figure 1. Nyquist and bode plots of impedance data for passive zirconium polarized at 0.6 V_{she} for 24 hours of film formation in 0.1 M $B(OH)_3$ + 0.001 M LiOH solutions at 250°C and 62 bar.

have incorporated the model parameters generated in Task 2, but some other model parameter values, primarily concerned with the reduction of oxygen and the evolution of hydrogen, need to be obtained in order to apply the MPM to zirconium and Zircaloy under the BWR and PWR coolant conditions. The model is currently employing estimated values for the exchange current density and Tafel constants for oxygen reduction and hydrogen evolution, because the experimental acquisition of the required data is well beyond the scope of the current project.

Task 4: Radiochemistry/Electrochemical Modeling. The objective of this task is 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 new codes developed are 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. To the researchers knowledge, they are the only codes in existence that are capable of accurately defining in-core electrochemical conditions as the operating conditions vary during normal operation.

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. The oxidation of a metal is almost always accompanied by the formation of corrosion products that occupy a greater volume than that of the metal replaced (described by the Pilling-Bedworth ratio). Large compressive stresses often exist inside the oxide film formed on a metal surface, whereas significant tensile stresses develop in the substrate under the oxide. If the stresses in the film become sufficiently high, oxide rupture may occur, presumably by buckling, that may then lead to deformation of the substrate and, thus, stress relaxation. In order to estimate the time of mechanical oxide rupture (i.e., transition in growth kinetics or the onset of nodular attack), the researchers are developing models to describe the processes involved. They are currently considering how to best develop a numerical model using an elasto-plastic finite element method to analyze the stress evolution within the thickness of the oxide film, and then to estimate the time of mechanical oxide rupture.

Task 6: Experimental Measurement of Model Parameters. The purpose of this task is to ascertain the appropriate oxidation mechanism for predicting diagnostic criteria with experimental data. This task also involves the experimental measurement of critically important model parameters, such as the electrochemical kinetic parameters for reactions that occur on the metal surface or the diffusivity of oxygen vacancies in the barrier layer. The activities originally scheduled for this task are now being accomplished in Task 2 with the successful development of the optimization procedure for extracting model parameter values from electrochemical impedance spectroscopic data.

Researchers have completed the measurements of critically important model parameters for the passive film formed on the zirconium surfaces under simulated BWR and PWR primary coolant conditions. They have elucidated oxidation/hydriding mechanisms by measuring the current, impedance, and capacitance as a function of formation potential. The kinetics of film growth and dissolution are demonstrated by simultaneously monitoring the changes in film thickness and current density as a function of time when stepping the potential in the positive or negative direction. The data are interpreted in terms of the modified point defect models that recognize the existence of a thick oxide outer layer over a thin barrier layer.

From thermodynamic analysis, researchers postulated that a hydride barrier layer forms under PWR coolant conditions, whereas an oxide barrier layer forms under BWR primary coolant conditions. Thus, the introduction of hydrogen into the solution lowers the corrosion potential of zirconium to the extent that ZrH_2 is predicted to form spontaneously, rather than the ZrO_2. The zirconium passive films formed in the hydrogenated environments have higher corrosion resistance and lower growth rate. The current density transients and the observed changes in film thickness of zirconium passive films have confirmed that the rate law derived from the PDM can be used to predict zirconium passive film growth and thinning at high temperatures under any set of electrochemical conditions that may exist in a reactor core.

Researchers have examined the effects of second-phase particles (SPPs) on the electrochemical and corrosion properties of passive zirconium by using different heat-treated Zircaloy-4 samples, i.e., as-received, β-quenched, and α-annealed. To our knowledge, this is the first *in-situ* study carried out in high-temperature aqueous solutions to examine the effects of SPPs on the electrochemistry of passive zirconium. Researchers have also employed optical microscopy with polarized light and scanning electron microscopy (SEM) to characterize the microstructure and determine the size and density of SPPs. The average size of the second phase particles in the Zircaloy-4 samples is in the sequence of β-quenched < α-annealed < as-received, with the reverse sequence being observed in the areal density. The size of the SPPs is found to be comparable to the passive film thickness. Electrochemical studies show that the size and density of the second phase particles are the determining factors of the electrochemical properties of the passive films. Thus, the β-quenched sample (smallest SPPs) has the highest corrosion resistance, while the α-annealed sample has the lowest. It is also evident that the second phase particles play an important role in determining the electronic character of a passive film, as demonstrated by Mott-Schottky analysis. The second phase particles may cause short circuits in the electrical path across the passive film, which would explain the effect of the size and the density of the SPPs, and hence heat treatment, on the corrosion properties of passive Zircaloy-4.

Planned Activities

DOE granted a no-cost extension on this project until September 14, 2006. Work on model development and experimental measurements will continue over the next reporting period. Other planned activities are identified in the tasks above.

Researchers will continue to develop a more advanced version of the models for the passive films on zirconium and Zircaloys under prototypical BWR- and PWR-like core operating conditions and will encode the models so that the properties of the films can be calculated and compared with experimental data. They will continue to refine the mixed potential model, in order to predict the oxidation and corrosion of Zircaloy under open circuit conditions. They will also continue to develop the numerical model using an elasto-plastic finite element method to analyze the stress evolution with the thickness of the oxide film and, then, to estimate the time of mechanical oxide rupture (see Task 5). They will also improve the models and optimization procedures for evaluating reaction mechanisms and for extracting parameter values from measured impedance data in order to identify the mechanisms of greatest fidelity and predictive power (see Task 6). Researchers will complete the experimental work to examine the passivity of Zircaloy-2, and they will integrate all of the models (PDM, radiolysis model for the coolant, BCM, MPM, etc.) into a comprehensive algorithm for the oxidation of zirconium and Zircaloy fuel sheathing under simulated reactor operating conditions.

NUCLEAR ENERGY RESEARCH INITIATIVE

Neutron and Beta/Gamma Radiolysis of Supercritical Water

PI: David M. Bartels, Argonne National Laboratory

Collaborators: University of Wisconsin, University of Notre Dame

Project Number: 02-060

Project Start Date: August 2002

Project End Date: September 2005

Research Objectives

Current pressurized water reactors operate 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 major unanswered questions remain:

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

The objective of this project was to discover most of the fundamental information necessary for developing a predictive model of radiation-induced chemistry in a supercritical water reactor core.

The coolant in any 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 will promote intergranular stress-corrosion cracking and irradiation-assisted stress-corrosion cracking of iron- and nickel-based alloys. These will alter corrosion rates of iron- and nickel-based alloys and zirconium alloys in reactors. One commonly used remedial measure to limit corrosion by oxidizing species is to add hydrogen in sufficient quantities, which will chemically reduce the transient radiolytic primary oxidizing species (i.e., OH, H_2O_2, and HO_2/O_2-) and stop the formation of oxidizing products (H_2O_2 and O_2). It is still unclear whether this will be effective at the higher temperatures proposed for future reactors. While an earlier NERI project has 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.

Electron pulse radiolysis coupled with transient absorption spectroscopy is the method of choice for measuring kinetics of radiation-induced species and 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 linac. The University of Wisconsin Nuclear Reactor Facility is a convenient source of neutron radiation that can be exploited for radiolysis experiments from room temperature 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

Researchers conducted experimental measurements of $(e\text{-})_{aq}$, H atom, and H_2 yields as a function of temperature. They carried out complementary experiments using neutrons, gamma radiation, and 3 MeV beta radiation. The chemistry exploited the following reactions:

$$\text{Radiolysis} \rightarrow (e\text{-})_{aq}, H, OH, H_2, H_2O_2$$

$$(e\text{-})_{aq} + N_2O + H_2O \rightarrow N_2 + OH\text{-} + OH \qquad (A)$$

$$H + CD_3CD_2OH \rightarrow HD + (CD_3CDOH)\cdot \qquad (B)$$

$$OH + CD_3CD_2OH \rightarrow HDO + (CD_3CDOH)\cdot \qquad (C)$$

Researchers used mass spectroscopy to measure the dissolved product gases N_2, H_2, and HD. The yield of N_2 was unexpectedly large in experiments above 200°C. The researchers determined that this was due to the presence of the hydroxyethyl radical (CD_3CDOH), which can also reduce N_2O to produce N_2, acetaldehyde, and OH radical. At room temperature, this reaction is too slow to be important; but, at 300°C it contributes a large fraction of the N_2 signal. Thus, it is necessary to run a second experiment with phenol (C_6H_6O) as a scavenger for OH and H atoms, using reaction (A) for the hydrated electrons.

Figure 1 shows results for 3 MeV beta radiolysis as a function of temperature at 250 bar. At the end of 2005, corresponding measurements in the neutron loop had not yet been completed.

Researchers also conducted experiments to measure key second-order recombination reactions in order to construct a comprehensive model of the chemistry. One reaction of key importance in reducing the effects of water radiolysis is the reaction of hydrogen atoms with oxygen. To measure this reaction at high temperature, researchers used electron pulse radiolysis with the following reaction scheme:

Pulse Radiolysis $\rightarrow$ OH, $(e\text{-})_{aq}$, H
$(e\text{-})_{aq} + N_2O + H_2O \rightarrow N_2 + OH\text{-} + OH$ (A)
$H_2 + OH \rightarrow H_2O + H$ (D)
$H + O_2 \rightarrow HO_2$ (E)

The experiment involved three pumps to mix water containing dissolved H_2, O_2, and N_2O gases. The production of HO_2 is followed by its transient absorption in the ultraviolet at 250 nm. A plot of rate constants vs. temperature for reaction (E) is shown in Figure 2.

Researchers did not expect the reaction rate at high temperature to decrease. In the gas phase, this reaction is extremely well studied. Scientists know from very high level *ab-initio* calculations that there is no reaction barrier for approach of an H atom at a 45 degree incident angle with respect to the O-O axis. Both reactants are hydrophobic, and there is no reason to think the solvent strongly perturbs the intermolecular potential. Researchers expected the reaction to be limited by diffusion, as shown by the dashed line in Figure 2. However, for H atoms approaching at 90 degrees or 0 degrees with respect to the O-O axis, there is a large barrier. The diffusion limit will only be reached if there are sufficient re-collisions (due to caging effect of the water solvent) to always find the 45 degree angle of approach. In high-temperature water, this is clearly not true.

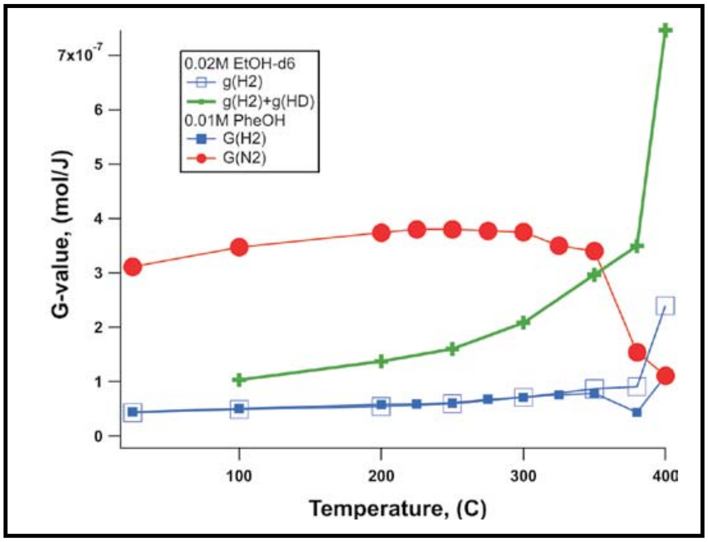


Figure 1. Radiolysis yields (G) as a function of temperature for H_2, N_2 (hydrated electron), and HD (H atoms) in radiolysis by 3 MeV beta, at 300 bar pressure.

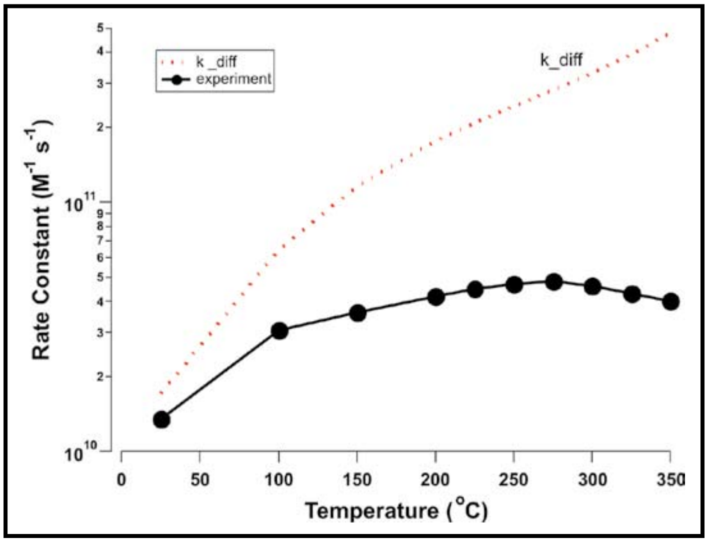


Figure 2. Rate constant for the reaction of H atoms with O_2 in water at 250 bar pressure. The dashed line is an estimate of the diffusion limit for this reaction.

The insight afforded by this observation can be used to understand and predict behavior for many other reactions of the important H and OH radicals in reactor cooling loops. The requirement for a particular approach angle will be typical, so most free radical recombinations will NOT be diffusion-limited in high-temperature water, and a turnover in reaction rates may be a general phenomenon. The one recombination which MUST be diffusion-limited is the simple reaction of two hydrogen atoms giving H_2 because there can be no angular component to the reaction path.

Planned Activities

Researchers compiled a new set of yields and rate constants for the radiation chemistry up to 350°C, a daunting objective given the large number of reactions involved and the inherent difficulty in correctly measuring second-order recombination reactions. In the course of this project, researchers measured numerous important reaction rates; others, already reported in the literature, were found to be incorrect. Work to measure second-order recombination in the supercritical water regime has barely begun. However, researchers feel confident that radiolysis yield measurements for high-temperature water reported here and elsewhere in the literature are fairly reliable.

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 focused on characterizing critical iron and nickel-based stainless steel alloys by testing them under well-controlled conditions to minimize in-service complexities. Researchers used 1) quantitative assessment of crack growth rates to isolate the effects of key variables and 2) high-resolution analytical transmission electron microscopy (ATEM) 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 were systematically interrogated for the first time. Researchers used novel mechanistic "fingerprinting" of crack-tip structures tied to thermodynamic and kinetic modeling of crack-tip processes to isolate causes of environmental cracking. They compared their experimental results against metallurgical analyses conducted on failed components removed from service from operating light water reactors (LWR).

This research strategy capitalized on unique national laboratory, industry, and university capabilities to generate basic materials and corrosion science results that will immediately impact the next generation of nuclear power systems. Project activities were integrated with existing NERI projects as well as U.S. and international projects that also address LWR degradation issues. This leveraged approach facilitated 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 was an essential first step toward elucidating environmental cracking mechanisms. Selecting these conditions was critical to producing degradation where crack initiation and advanced processes are best understood. By controlling material, environmental, and electrochemical conditions in high-temperature water to promote degradation by fundamentally different processes, researchers were able 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 conducted critical experimentation in several areas, including quantitative stress corrosion cracking (SCC) crack-growth-rate measurements on specially tailored stainless steels and nickel-based alloys in simulated LWR environments. Initial work systematically mapped matrix strength effects on intergranular SCC (IGSCC) in 300-series stainless steels and linked results to the behavior of irradiated core internals. Researchers 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. They also isolated the influence of grain boundary carbides and chromium depletion on IGSCC, documenting the detrimental effect of depletion and revealing that carbides can decrease crack-growth rates by approximately five times below that for precipitate-free boundaries. These tests provided 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. This task generated a library of high-resolution, crack-tip “fingerprints” for SCC in LWR structural materials. These fingerprints were then used to compare with in-service cracking and assess environment-assisted cracking mechanisms. 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 LWR service.

Researchers documented crack and crack-tip corrosion structures at near-atomic dimensions for environment-induced cracks in a number of different materials, including laboratory crack-growth samples of 304 and 316 stainless steel, nickel-based Inconel alloy-600, and laboratory U-bend samples of alloy 600 and alloy 690. In addition, they documented cracks in 304 and 316 stainless steel boiling-water-reactor (BWR) piping and core internals, 316 stainless steel pressurized-water-reactor (PWR) core internals, alloy 600 and alloy 182 PWR upper-head penetrations, and alloy 600 PWR steam generator tubing. For the first time, researchers were able to directly compare crack-tip characteristics established under well-controlled laboratory test conditions with those produced in actual LWR service.

Detailed comparisons were made between lead-assisted SCC of alloy 600 in high-temperature water for controlled laboratory tests and PWR secondary-side steam generator tube failures. Contrary to currently accepted theories, caustic crack solutions were not required in order to promote rapid IGSCC in mill-annealed alloy 600. Crack and crack-tip structures produced in laboratory tests with lead in simulated PWR water were nearly identical to those observed in many steam-generator tubes from service. However, laboratory tests in caustic solutions similar to those predicted for secondary-side crevices revealed corrosion products and Pb distributions quite different than observed for service. Transgranular (TG) SCC of thermally treated alloy 600 and 690 in Pb-doped environments was also investigated and found to occur in tandem with the development of a localized deformation structure at crack tips. Dislocation analyses at tips revealed the role of intense local plasticity in the crack-propagation process.

Researchers also investigated complex solidification microstructures and cracks in alloy 182 weldments that were removed from two failed PWR upper-head penetrations. They observed significant macro-segregation of manganese and niobium, but this did not appear to play a role in IGSCC that was identified in both cases. The cause of cracking in these weldments was clearly identified as IGSCC and not influenced by hot cracking during welding or repair welding. Important differences were found between service cracking from the Ringhals weldments and the Davis-Besse weld. They observed limited grain boundary precipitation on cracking interfaces for the Ringhals examples, while extensive carbide precipitation was present in the Davis Besse weldments. In addition, Davis-Besse crack corrosion products revealed that sulfur in the water may have contributed to the observed SCC. The observation of sulfur in the alloy 182 weld cracks was confirmed by even higher levels found in IGSCC crack oxides in the adjacent alloy 600 nozzle.

These service results showing sulfur in IGSCC cracks produced in PWR primary water prompted additional examinations, including primary-side cracks in two alloy 600 steam generator tubes and cracks produced during a crack-growth test on alloy 600 in simulated PWR primary water. Surprisingly high levels of sulfur impurity were again identified in cracks and at crack tips in one of the steam-generator tubes. However, the basic characteristics of penetrative grain boundary oxidation leading IGSCC in both the service and laboratory examples were unchanged by the presence of sulfur. As was the case for lead in secondary PWR environments, sulfur may accelerate the intergranular oxidation and cracking process but is not a root cause for degradation. In general, results show that a stress-assisted, grain boundary oxidation process drives intergranular attack and SCC of Ni-base alloy 600 in both PWR primary and secondary water.

The most comprehensive evaluations of IGSCC mechanisms from crack tips focused on stainless steel samples after crack-growth tests in BWR oxidizing water. Researchers documented characteristics for cold-worked 304SS, cold-worked 316SS, and sensitized 304SS and compared them to a BWR service example from an irradiated 304SS top guide. Unique crack-tip structures and compositions found in the service top-guide were not reproduced in the well-controlled laboratory test samples. Researchers observed dramatic selective oxidation of iron and chromium at the tips of oxide-filled cracks in the service example. Porous metal was present at the crack tips along with an extensive region of Ni-rich, Fe/Cr-depleted metal ahead of the tip. The laboratory crack-growth samples exhibited open cracks. Epoxy-filled cracks exhibited extremely tight crack tips with thin, Cr-rich wall oxides. Crack and crack-tip regions showed extremely high local deformation including shear bands and twinning. In general, these characteristics were consistent with a slip-oxidation SCC mechanism. Interestingly, somewhat blunt crack-tips were found in the sensitized 304SS sample where the Cr-depleted metal along grain boundaries was rapidly oxidized during crack advance.

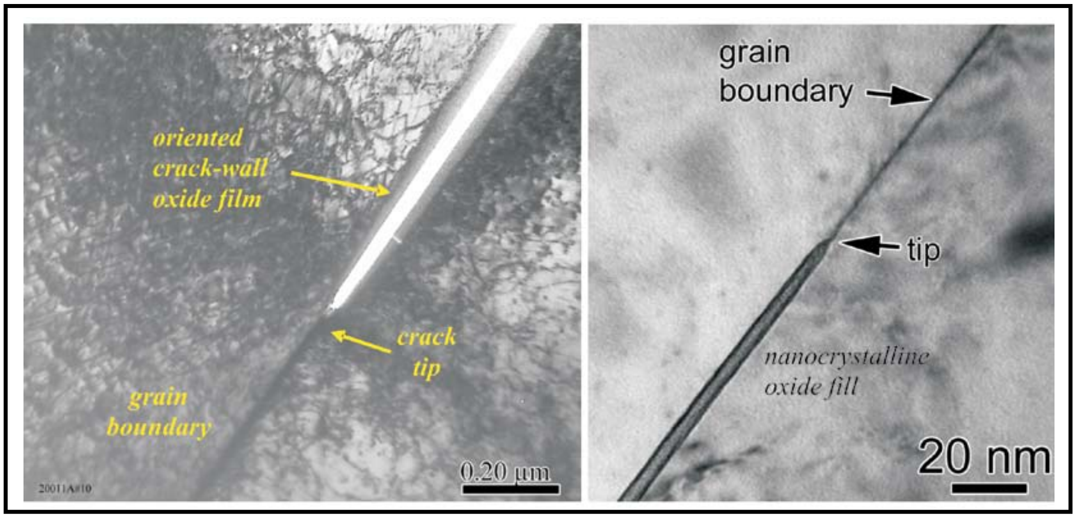


Figure 1. Representative crack tip regions for an Fe-base, austentic stainless steel produced in BWR oxidizing water (left) and for a Ni-base alloy 600 produced in PWR water (right).

Figure 1 illustrates the fundamental differences in crack-tip characteristics, showing a typical crack tip in Fe-base 304SS after propagation in oxidizing BWR water and a typical tip region in Ni-base alloy 600 after propagation in hydrogenated PWR water. Intergranular cracks in stainless steels form a protective, non-porous oxide film on crack walls. Crack widths decrease from over 100 nm to less than 10 nm, ending at open tips in heavily deformed metal. Alloy 600 shows distinctly different characteristics with narrow (~10 nm wide) penetrative oxide extending from many µm before ending in the grain boundary plane. The formation of a thin, non-porous, Cr-rich protective oxide at crack walls and at the crack tips in the 300-series stainless steels is consistent with the classical SCC slip-oxidation mechanism. In this mechanism, cracks propagate by localized dissolution or oxidation resulting from repeated breakdown of the protective Cr-rich film at the crack tip. These events are in agreement with the high-deformation structure (including shear bands) seen in the tip regions. Penetrative oxidation without significant deformation along grain boundaries in alloy 600 crack-tip regions suggests a quite different propagation mechanism. The extremely narrow dimensions of the cracked or attacked structure and the porous nature of the oxide fill suggest a stress-assisted, active-path, intergranular oxidation process driving degradation.

Modeling Crack-Tip Corrosion Reactions. The final key area of research was modeling the crack-tip corrosion reactions. This task focused on predicting thermodynamic stability of corrosion products, aqueous transport kinetics to crack tips, and alloy composition changes caused by tip corrosion. The Geochemists Workbench program calculates and plots stability diagrams based on electrochemical conditions, specified species, and reaction rate equations. Specifically, potential-pH stability was calculated for conditions of Pb aqueous chemistry and alloying elements that relate to crack tip corrosion reactions for stainless Fe and Ni alloys. Model calculations of Pb aqueous concentrations as low as 1 appm (atomic parts per million) promote $PbCrO_4$ formation but only at high potential and pH and not at conditions examined in the characterization task. The lack of observed Pb oxides is consistent with the lack of Pb oxides predicted by Geochemists Workbench®. Kinetic models for the transport of Pb to advancing cracks tips predicted aqueous composition and potential conditions required to maintain an adequate supply of Pb to the tip. Vacancy injection at an actively corroding tip was established as the only credible mechanism for the observed change in alloy composition and concentration profiles at tips. The proportional changes in composition were identical to that expected based on relative diffusivities of Fe, Cr, and Ni. This suggested that the apparent selective dissolution could be influenced by selective diffusion rates of the species.

Planned Activities

This project ended in September 2005. Selected research activities with nuclear power industry collaborators are continuing under independent funding primarily focused on root cause failure analysis in LWR components.

NUCLEAR ENERGY RESEARCH INITIATIVE

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

PI: Todd Allen, University of Wisconsin- Madison

Collaborators: Idaho 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 focused on improving the radiation tolerance of both austenitic and ferritic-martensitic (F-M) alloys projected for use in advanced light water and Generation IV reactor systems. The materials limitations of Generation IV components include higher temperature creep strength, dimensional stability, and corrosion/stress corrosion compatibility. This research focused on three approaches to increasing the radiation tolerance of these alloys: 1) grain boundary engineering, 2) microstructural/nanostructural control by matrix alteration, and 3) the addition of oversized solutes to the matrix. Each approach was 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. Matrix precipitate additions provide a high density of interfaces to promote recombination and trap helium, while also strengthening the matrix against high-temperature creep. Oversized solute additions will promote recombination by reducing RIS and altering the dislocation microstructure.

Research Progress

Grain Boundary Engineering. Grain boundary engineering involved 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) to reduce cracking susceptibility and improve 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. 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. As shown in Figure 1, improved creep strength results from the preservation of all the microstructural features decisive for creep strength in T91 and from increasing the fraction of low-angle boundaries. In addition to T91, the researchers also developed CSL-enhancement techniques for alloy 800H, a higher temperature alloy seen as a candidate for high-temperature, gas-cooled reactors. These treatments have proven to be effective methods of stabilizing oxides when 800H is exposed to supercritical water.

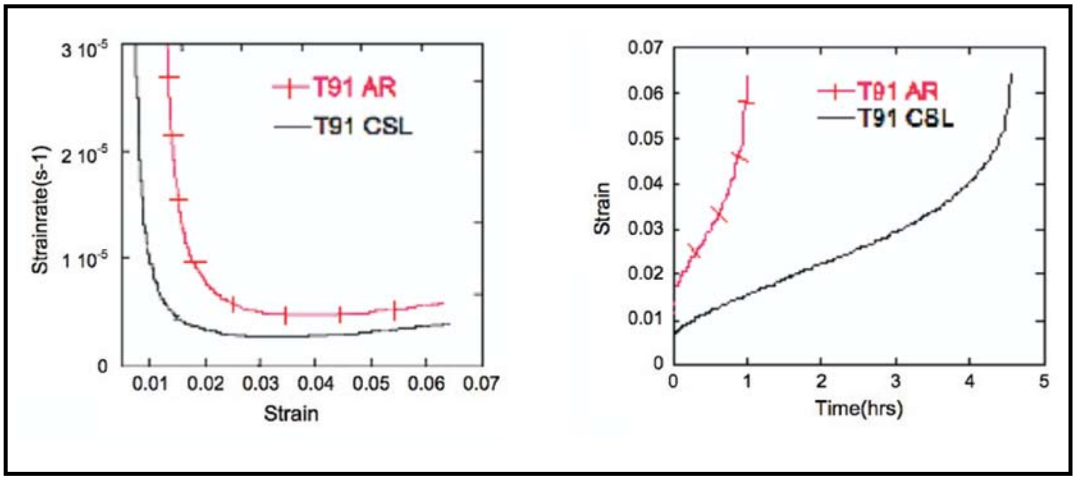


Figure 1. Plot of a) strain vs. time and b) strain rate vs. strain for creep test of alloy T91 at 220 MPa (600°C, argon). AR=as-received, CSL=enhanced grain boundaries.

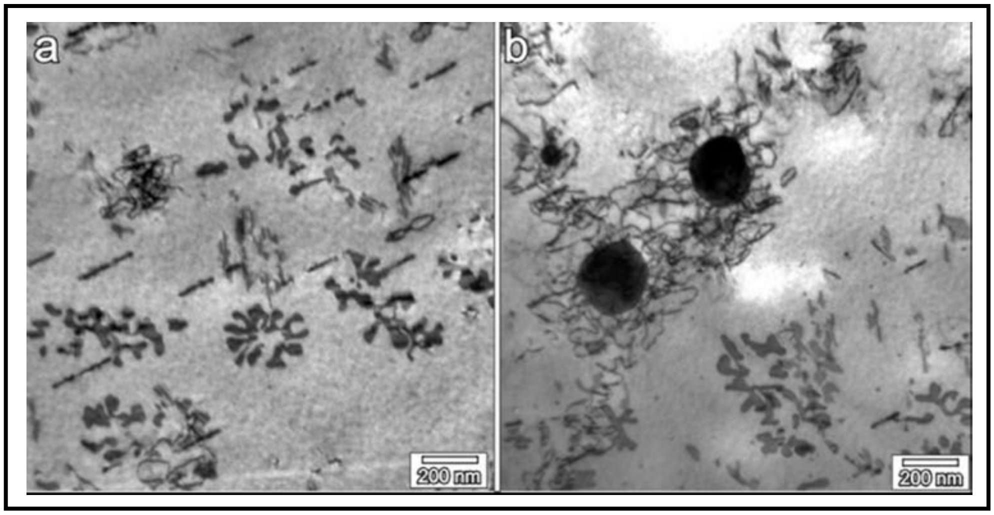


Figure 2. Crenulated Frank loops were very common in the FeCrNi + Zr alloy irradiated to 3.7 dpa at 431°C. The Zr-Ni particles shown in (b) often had dense tangles of dislocations and Frank loops surrounding each particle. The crenulated appearance may be due to precipitation or impurities that have segregated to the loops during irradiation.

Radiation Stability Following Matrix Alteration. The second major task was to examine the radiation stability of advanced alloys. Proton and heavy ion irradiation were performed to evaluate the radiation response of alloys 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 conventional fast reactor alloy. HCM12A was irradiated with 3.2 MeV protons and with 5 MeV nickel ions at 500°C. The hardness of HCM12A increases with dose, saturating after approximately 5 dpa with about a 70 percent increase in hardness. Using hardness-yield strength correlations, this corresponds roughly to a 520-550 MPa yield strength increase. Nothing in these irradiation studies indicated adverse properties in this alloy as compared to HT9. Irradiations showed that in ferritic-martensitic steels, chromium is enriched at boundaries during irradiation, differing from the radiation response in austenitic steels. Radiation of the 9 Cr ODS alloys showed that, over a temperature range of 500-700°C, the larger oxides are redistributed into a smaller oxide distribution, potentially leading to an increase in strength.

Oversized Solute Additions. The addition of oversized solutes was 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 the U.S. Department of Energy'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 of a 316 alloy without hafnium produced the least amount of swelling. Figure 2 shows unique microstructures that arise in zirconium-containing alloys under radiation. Although adding zirconium to a high-purity stainless-steel alloy reduced swelling at a 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.

Modeling of RIS and the effect of oversized solutes in Fe-Cr-Ni reveal that a high binding energy, on the order of 0.8–1.0 eV, is necessary to appreciably enhance recombination and reduce segregation. Oversized solutes are generally most effective in reducing RIS at temperatures of 400–550°C with dose rates of 10^{-5} to 10^{-6} dpa/s. The variation in effectiveness as a function of dose rate indicates that oversized solute additions may be more effective for ion irradiations but less effective for neutron irradiations.

Planned Activities

This project ended in September 2005. The grain boundary engineering studies will continue as part of work under the Generation IV initiative, specifically supporting the gas-cooled fast reactor and supercritical-water-cooled reactor project.

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

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 key objective of this research project was to develop systems to efficiently and selectively recover special-purpose isotopes that are added to primary coolant water in light water reactors (LWRs) for neutron flux control and water chemistry. Increasing nuclear fuel enrichment in existing and advanced pressurized water reactors (PWRs) 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 flux chemical shim. 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 was 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.

Research Progress

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. Then, 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. Following is a more detailed description of this work.

Boric acid readily forms covalent bonds with alcohol functionalities through the displacement of the boron hydroxyl group. Based on the equilibrium constants reported, reagents having two sets of phenolic hydroxyl groups generally form much more stable borate complexes than those of alcohols. It is anticipated that boric acid complexation by these ligands will occur at the neutral conditions present in reactor coolant water. Solution thermodynamic studies with other ions have demonstrated that substituting various amide substituents can affect the overall stability of the resultant complex by changing the charge of the net complex.

A number of ligands, including catecholamides, terephthalamides, and hydroxypyridinonates, are expected to more effective and feature higher binding constants than simple sugars with vicinal diols when supported to water soluble polyethylenimine (PEI). Using a number of synthetic chemical steps, researchers synthesized, characterized, and evaluated a series of novel boron chelating compounds and their resulting borate esters, Figure 1.

These ligands are capable of binding a range of ions and can be easily altered to affect complexation and solubility properties. Potentiometric and spectrophotometric titrations and ^{11}B-NMR spectroscopy (Figure 2) have been used as to study the structure, solution thermodynamics, and kinetics of borate complexes. The structural characteristics, kinetics, and solution thermodynamics of these systems (Figure 3) have been investigated at various conditions. The solution behavior of these species has been investigated and the formation constants have been in modeling efforts.

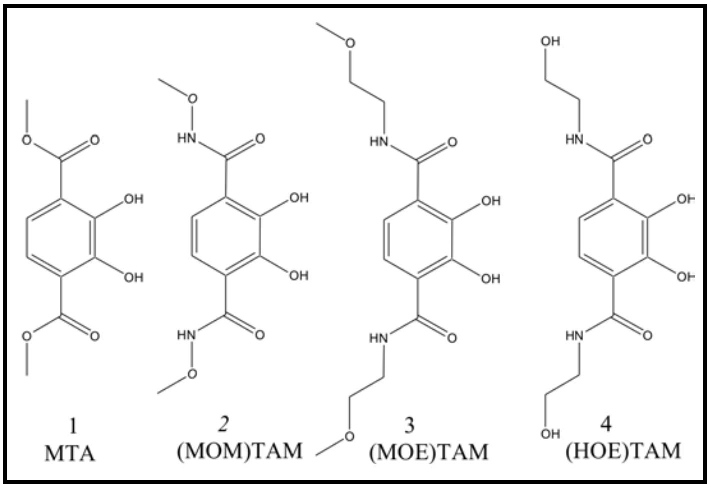


Figure 1. Series of terephthalamide borate ester complexes.

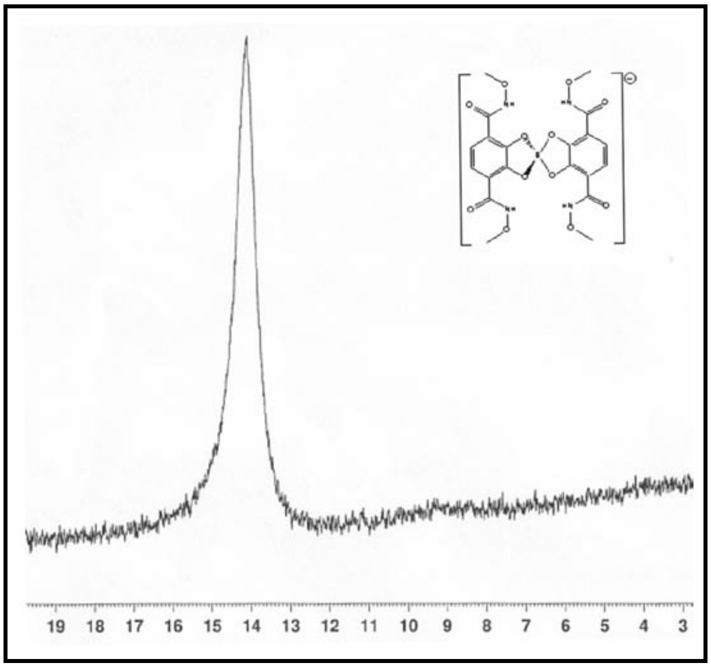


Figure 2. ^{11}B NMR Spectra obtained for dimethylammonium bis[MTA-*O,O'*] borate.

Researchers have shown that 0.1 M NaCl salt added to PEI/100 ppm boric acid (BA) solution had the effect of quenching the ion-pairing between the borate anion and the weak anion exchange sites of the PEI polymer. Adding 0.1 M NaCl to ¼-1,2-diol-PEI/100 ppm BA solution quenched borate ion-pairing and allowed for an estimate of ion-pairing to borate ester ratio. However, it was not known if salt addition could have some unanticipated effect on borate ester formation, which could bias the previous borate ester/borate ion-pairing ratio estimate. Thus, the team examined by ^{11}B-NMR the influence of adding a neutral D_2O solution of salt (NaCl) to the 2/3 and the 3/3-1,2-diol-PEI polymers in the presence of BA. Solutions containing 1 M in NaCl and 600 ppm boron were analyzed by ^{11}B-NMR and compared with their counterpart ones without NaCl. Surprisingly, the addition of NaCl resulted in very large increases in pD (similar to pH but with deuterium from the D_2O) and, as a consequence, in large chemical shifts of the BA/borate peak.

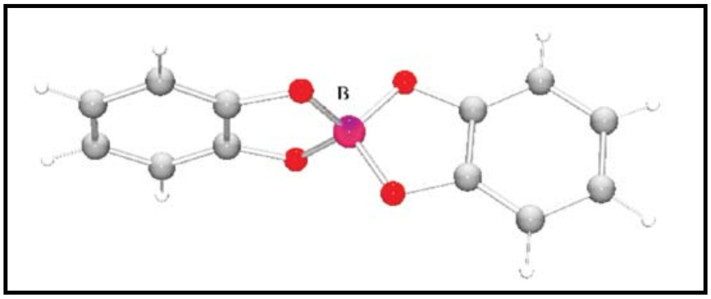


Figure 3. Boron complexation by ligands.

Easy and straightforward reversal of boron binding is a very important factor for recycling ^{10}B-enriched BA in the nuclear power industry. Temperature reversal of boron binding is more amenable to nuclear power applications than the more often-reported acid or base stripping. The facility of 1,2-diol-PEI polymers to perform as temperature-reversible boron chelators was investigated for the 2/3 functionalized PEI polymer. According to the ^{11}B-NMR studies, this polymer appears to give the optimum level of boron binding.

Researchers prepared this sample with 1 percent mass/volume polymer concentration and for boron concentrations ranging from 1,000 to 100 ppm. ^{11}B-NMR spectra data obtained from 25 to 65°C are summarized in Figure 4. These results indicate that an increase in temperature to 65°C suppresses most borate ester formation for all boron concentrations. In addition, binding of borates with anion exchange resins through ion pairing has been reported to decrease as temperature rises. Consequently, the water-soluble polymers proposed in this study for BA recovery are expected to perform efficiently with regard to reversible boron binding as well.

There are several approaches to addressing the problem of antimony (Sb) contamination. One is the selective back stripping of boron from the water-soluble polymers, leaving the Sb on the polymers. Once the soluble polymer is loaded with Sb to some predetermined level the polymer concentrate could be discarded as radioactive waste and be replaced with fresh polymer for binding more boric acid. Since the researchers were focusing on thermal stripping as the mode of boric acid recovery, they needed to determine if there is any selectivity in thermal stripping between boric acid and Sb. The best way to determine if the dilute solution species of Sb in the power plants can be removed is to do live tests at the power plants.

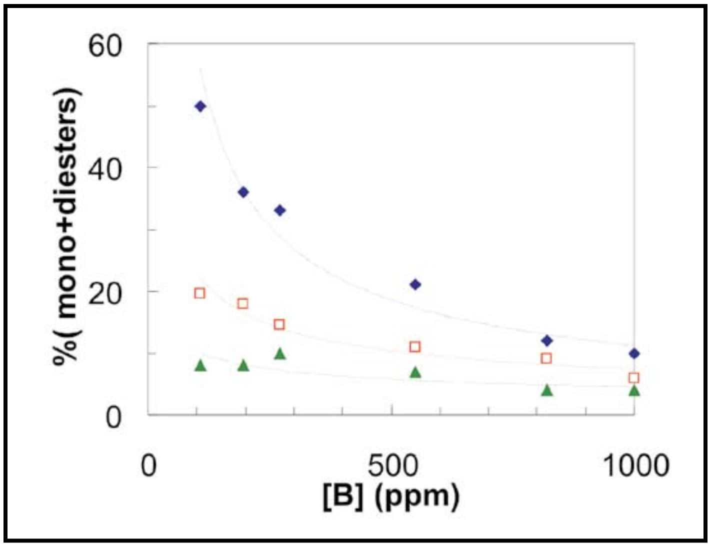


Figure 4. Relative abundance of mono- and diester for 2/3-PEI versus boron concentration at various temperatures: (♦) 25°C, (□) 45°C, and (▲) 65°C.

As reported in the power plant studies, it appears that some of these isotopes in coolant water exist as particulates that can be concentrated by simple ultrafiltration (e.g., iron, cobalt, and lanthanum were concentrated, but not antimony and manganese). Thus, it appears that it is possible to concentrate BA along with all contaminating isotopes that cause a problem in reactant cooling water and then selectively recover the boric acid. Some of them could be removed by ion-pairing through the weak cationic site on the polymeric backbone, but they might be removed with thermal stripping. Some could form nitrogen complexes as might be expected for the cobalt species, silver species, and perhaps even the chromium species. The Sb is expected to form complex esters similar to the boric acid esters.

Planned Activities

This project has been completed.

7. FY 2005 and 2006 NERI Project Summaries and Abstracts

In 2004, the focus of NERI research changed to exclusively fund R&D supporting the Generation IV, AFCI, and NHI programs. This research, reflected in the FY 2005 and 2006 NERI awards, is lead by U.S. universities and colleges in collaboration with the DOE National Laboratories and industry research organizations. This section contains FY 2005 project summaries, which include each project's participants, its objectives, a brief discussion of research progress made in 2005, and planned activities over the remainder of the project. The FY 2006 project abstracts similarly present each project's participants and objectives, along with a brief description of the proposed work scope. Following is a brief description of the three program areas that NERI currently supports.

Generation IV Nuclear Energy Systems Initiative

The Generation IV Nuclear Energy Systems Initiative is focused on developing new reactor systems to be deployed during the next 20 years. This program supports 19 NERI research projects: 13 awarded in FY 2005 and six new projects awarded in FY 2006. These projects are related to the seven research areas described below.

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 economically produce both hydrogen and electricity. The NGNP is in the first phase of project development and may lead to the actual design, construction, and future operation of an Advanced Nuclear Reactor-Hydrogen Cogeneration Demonstration unit. New research projects focus on validating reactor physics and core design analyses tools, developing and validating reactor thermal-hydraulic and mechanical design analyses tools, performing materials research and power-conversion unit assessments, and conducting 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 to: 1) demonstrate a full-scale unit in the 2015 timeframe; 2) demonstrate the Brayton Cycle for the full-scale unit; 3) test the exceptional safety capabilities of advanced very-high-temperature reactors; 4) obtain a Combined Operating License (COL) for the NGNP using NRC's proposed performance-based, risk-informed licensing process (10 CFR Part 53 now in development); and 5) support the development of other technologies involving hydrogen applications.

Supercritical Water-Cooled Reactor. Projects associated with this technology concentrate on showing the technical feasibility of a light water reactor operating above the critical temperature and pressure of water, thus producing low-cost electricity. The focus will be on five main areas: 1) design of safety systems and containment to handle abnormal events; 2) evaluation of dynamic power/flow instabilities; 3) corrosion and stress-corrosion cracking testing of materials for the core and vessel internals; 4) the investigation of basic thermal phenomena for the reactor; and 5) 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 that can be deployed 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. The research scope encompasses several milestone goals, such as concept development and completion of a paper discussing the implementation of these concepts. Specifically, research will be performed to generate a design for concept development and for establishing economic requirements and proliferation-resistance principles. Researchers will develop a design and data requirements document, revise the safety and licensing approach, establish a fuel qualification strategy, resolve materials and corrosion issues, submit a concept paper on implementation of fuel cycle centers, and ascertain 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, and 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. In FY03, NERI project researchers identified possible passive/semi-passive safety systems, initially tested CO_2 compatible materials, and identified candidate high-temperature fuels. Continued work in this area will include 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. For example, research needs to be performed to evaluate the economic feasibility of these new plants and to ensure the proliferation resistance (PR) and physical protection (PP) of the public. Major activities for FY05 through FY08 include 1) improving the NGNP analytical capabilities and validate these capabilities, 2) defining fast reactor modeling requirements for the GFR and LFR designs, and 3) improving the Generation IV evaluation methodologies and selecting preferred options, such as developing preferred methods for evaluating PR and PP measures and 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 for the design and construction of a small-scale demonstration experiment.

Advanced Fuel Cycle Initiative

The Advanced Fuel Cycle Initiative 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 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. This program supports 32 NERI research projects: 19 projects awarded in FY 2005 and 13 new projects awarded in FY 2006. These projects are related to the four research areas described below.

Separations. Separations research includes 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. FY 2005 and 2006 projects under this research area will focus on selecting a process for LWR spent fuel reprocessing including isotope separation and recovery; fabricating waste/storage modules (forms) for storage in a repository; demonstrating the one-group transuranic (TRU) 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/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, and fast spectrum systems). NERI research projects for the next three years will focus on continuing TRISO fuel development including shakedown testing, performance testing, and fission product transport and ATR (Advanced Test Reactor at INL) irradiation and PIE of mixed-oxide and inert matrix fuel (IMF), as well as nitride, metal, and dispersion fuels.

Transmutation Science and Engineering. 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 the Advanced Fuel Cycle Initiative, 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 ADS (Accelerator Driven Systems) technology.

Systems Analysis. This research area includes broad systems studies, transmutation system studies and integrated model development, fuel cycle proliferation resistance studies, economics analyses, and safety assessment. The objectives of research projects under this area are to analyze 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 spent fuel repository.

Nuclear Hydrogen Initiative

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 2019. This program supports eight research projects: three awarded in FY 2005 and five new projects awarded in FY 2006. These projects are related to the three research areas described below.

Thermochemical Cycles. The focus of projects under this research area is thermochemical cycles for nuclear application, such as sulfur-based cycles, calcium-bromine cycles, and alternative cycles; flowsheet methodology for comparing thermochemical cycles; and high-temperature interface requirements for heat exchangers and materials. Research planned for the next three years will consist of laboratory-scale demonstrations of candidate processes for sulfur-iodine, hybrid sulfur, and calcium-bromine; 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 construction of commercial units. Materials analysis for cells, heat exchangers, and separations will also be performed under this research area. Planned NERI projects will complete HTE cell and stack/module testing, complete the design and assembly of components for the integrated laboratory test unit by 2007, complete the design and fabrication of a pilot-scale experiment by 2013, and conduct tests for high-temperature heat exchangers and separations.

Reactor-Hydrogen Production Process Interface. Research in this area includes high-temperature heat exchanger (HX) design (including HX type, operating conditions, efficiency, and material qualification), implications of the 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 assessment of the process, infrastructure, and facilities requirements for the pilot plant and the balance-of-plant (BOP) design.

Directory of FY 2005 Project Summaries and FY 2006 Abstracts

Generation IV Nuclear Energy Systems Initiative

Advanced Fuel Cycle Initiative

Nuclear Hydrogen Initiative

Generation IV Nuclear Energy Systems Initiative Projects

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: January 2005

Project End Date: December 2007

Research Objectives

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

Researchers will study various promising candidate materials as well as materials whose surfaces have been modified by applying the novel process of Ionized Plasma Deposition (IPD). 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.

Objectives of this research project are to:

- 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, 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

Research Progress

Researchers used the Advanced Photon source to study ex-situ samples of 316 stainless steel, molybdenum, and spinel which had been exposed to lead at 1,000°C for 5 or 50 hours under an argon atmosphere. Because of the ex-situ nature of the experiments, they found that the lead was in an oxidized state on the samples when measured.

For the 5-hour exposures, both the molybdenum and stainless steel showed significant attack by lead, although the spinel sample seemed relatively unaffected. For the 50-hour exposures, the stainless and molybdenum showed significant flaking of the surface and the spinel also appeared to have some surface contamination.

The researchers conducted extended X-ray absorption fine structure (EXAFS) measurements on all the 5-hour samples and some of the 50-hour stainless steel samples, measuring both the material which flaked off the surface and the residual materials remaining on the sample surfaces. The other 50-hour samples were measured in November, during the 2005-3 run.

During the same period, they have been working on bringing the vacuum deposition device into working order and making initial Pb film depositions on a Mo substrate. Researchers prepared an initial sample and conducted measurements in order to determine the conditions under which they can probe the film and substrate separately by changing the incident angle of the beam.

Figure 1 compares experimental data on 316 stainless steel exposed to Pb at 1,000°C (black), the flaked off corrosion layer (red), iron oxide (green), and Fe metal foil (blue). Clearly, there is a significant amount of iron in the flaked off material. Researchers will be able to observe the first stages of corrosion in the planned *in-situ* experiments. The edge structure of the sample and the flakes shows an intermediate character between Fe metal and Fe oxide. The similarity between these two spectra is indicative of the fact that the team is likely probing only the reacted surface of the stainless steel at the 20 degree incident angle.

Figure 2 shows incident intensity-normalized data taken on the molybdenum sample coated with a thin, evaporated layer of Pb at 45 and 15 degree incident angles. The lower intensity at the smaller angle is indicative of the shallower probe depth. At the lower energy of the Fe edge, this effect is more pronounced.

Planned Activities

This year, researchers plan to begin *in-situ* studies at the Materials Research Collaborative Access Team (MRCAT) beamline facility at the DOE Advanced Photon Source. They have also procured a reactor chamber designed for insertion into a furnace which can be heated to 1,000°C. This chamber has a side port to allow fluorescent x-rays to reach the detector. They will prepare Pb-coated samples with defined angles of beam incidence and modify the chamber accordingly. These samples will be measured as a function of time.

In addition, researchers have identified a company to provide surface modification for this project. They will discuss coating steel samples with a spinel, as this is the material which appeared to be most resistant to attack by Pb vapor.

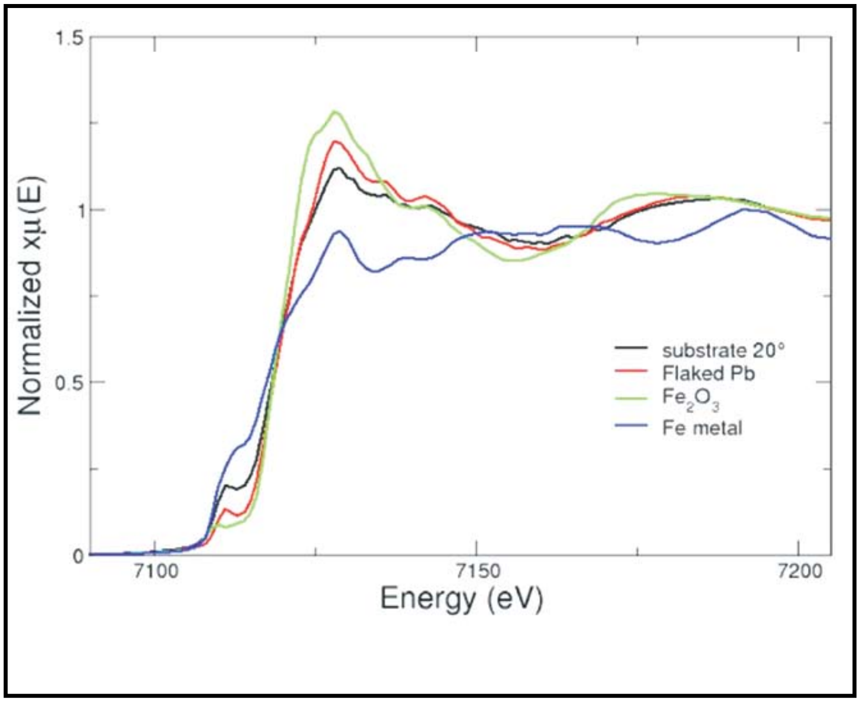


Figure 1. 316 stainless steel exposed to Pb at 1,000°C.

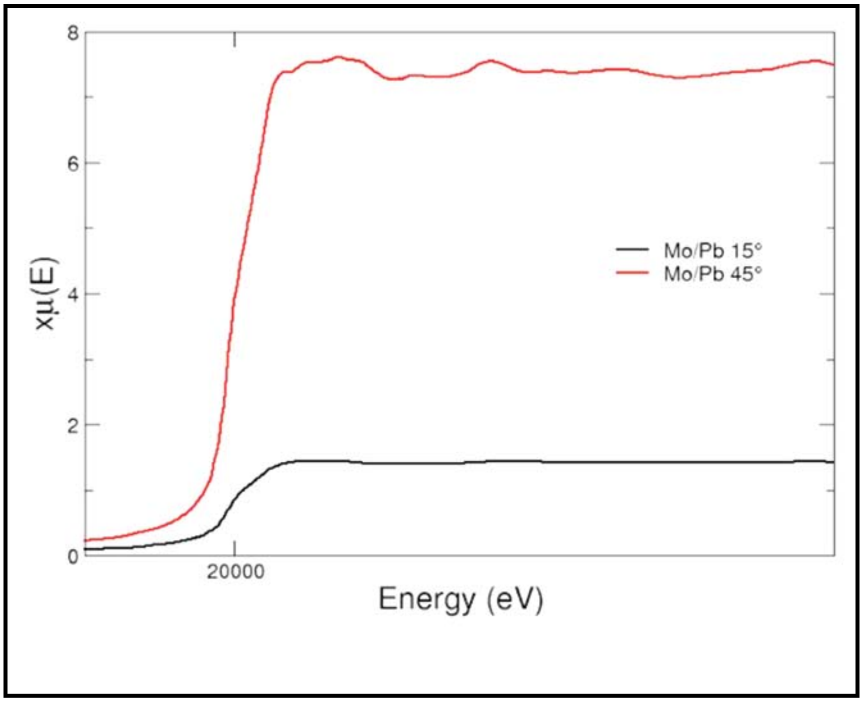


Figure 2. Intensity-normalized data on Pb-coated Mo at 15° and 45° incident angles.

NUCLEAR ENERGY RESEARCH INITIATIVE

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

PI: Michael J. Driscoll, Massachusetts Institute of Technology (MIT)

Collaborators: None

Project Number: 05-044

Project Start Date: April 2005

Project End Date: April 2008

Research Objectives

This project will develop a complete, integrated plant design for a gas-cooled fast reactor (GFR) based on the compact and highly efficient, direct supercritical carbon dioxide (S-CO_2) Brayton cycle (which is also under development at MIT for Generation IV service). This plant will be capable of performing a full spectrum of 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. Researchers will evaluate prospective plant designs using existing Nuclear Regulatory Commission (NRC) deterministic prescriptions and proposed risk-informed licensing requirements. They will also use probabilistic techniques to optimize the tradeoffs between economics and safety assurance.

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 challenges to the core and component materials. Because the S-CO_2 cycle can achieve high thermodynamic efficiency (approximately 44 percent to 51 percent) at modest temperatures (from 550 to 650°C), researchers can utilize already proven technologies in their design.

Major tasks of this project will focus on designing the reactor core and the decay heat removal systems. In designing the core, researchers will optimize features of the vented fuel concept using tube-in-duct assemblies, develop a pin-type core design as a benchmark, and confirm the capability of burning TRU and minor actinides. Safety system design, guided by probabilistic methods, will involve development of a decay heat removal system for accidents, shutdown, and refueling; improvement of emergency power, such as microturbines or fuel cells; and integration

Core		Comments
Fuel	UO_2 + BeO	LWR TRU fissile
Clad	ODS, MA 956	SiC a long range possibility
Configuration	tube-in-duct fuel assemblies, "hexnut" pellets, vented, orificed	pin-type core as fallback
Thermal-Hydraulics	axial peaking factor ≤1.3 radial peaking factor ≤1.3 power density ≥85 W/cc	vary BeO fraction to flatten power lower than GA GCFR of 1970's @ 235 W/cc
Burnup	≥120 MWd/kg (avg)	In single batch core
Safety Systems		
Auxiliary Loops	combined shutdown & emergency, 4 x 50% capable, forced convection; natural convection supplemented water boiler heat sink	based on MIT/CEA/ANL INERI project design. For P≥0.5MPa natural convection alone may suffice
Emergency Power	fuel cells to supplement diesels	projected to be more reliable than diesels alone in long run
Plant		
Power Cycle	supercritical CO_2 Brayton direct 4 x 300 MWe loops=1200MWe core exit/turbine inlet 550°C≤T≤700° pressure: 20 MPa	AGRs in UK use CO_2 coolant at 4 MPa and have T~650°C
Reactor Vessel	PCRV, PCIV or Steel	20MPa is challenge for PCRV and perhaps PCIV

Table 1. Initial GFR Plant Features.

of both active and passive means of shutdown assurance to preclude an anticipated transient without scram (ATWS). The final design will be optimized for integration with a high-temperature steam electrolysis process for hydrogen production, and an economic assessment will be conducted to estimate costs. 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.

Research Progress

Work has been initiated in most of the task areas. The principal achievement has been the development of an initial reference design that has features consistent with the project's goals, as shown in Table 1. The most significant design feature is the radially powered flattened core. As shown in Figure 1, the maximum peaking factor over core lifetime is only 1.2 for a batch-irradiated core driven to 120 MWd/kg average burnup. The key innovation that allows this performance is the use of radial variation of beryllium oxide (BeO) diluent in the fuel. The concurrent spectral softening also yields a coolant void reactivity of less than one dollar over core lifetime. Furthermore, the BeO increases fuel conductivity by about 50 percent. In conjunction with the use of "hexnut" shaped annular fuel pellets, instead of researchers' original plans to use VIPAC fuel, this considerably enhances the thermal-hydraulic performance of the vented tube-in-duct fuel assemblies.

Planned Activities

The most significant next step will be to carry out RELAP5-3D transient thermal hydraulic analyses of candidate core designs coupled to a complete plant model, including both the power conversion and shutdown cooling systems. Converging upon a suitable fuel assembly configuration is an essential prerequisite to demonstrating other important fuel cycle goals such as minor actinide burning. These RELAP calculations, in conjunction with PRA analyses, are also important to finalizing the shutdown cooling system design, including balancing between active and passive features as a function of pressure and post-shutdown time, since to date the design has been based on quasi-steady-state analyses.

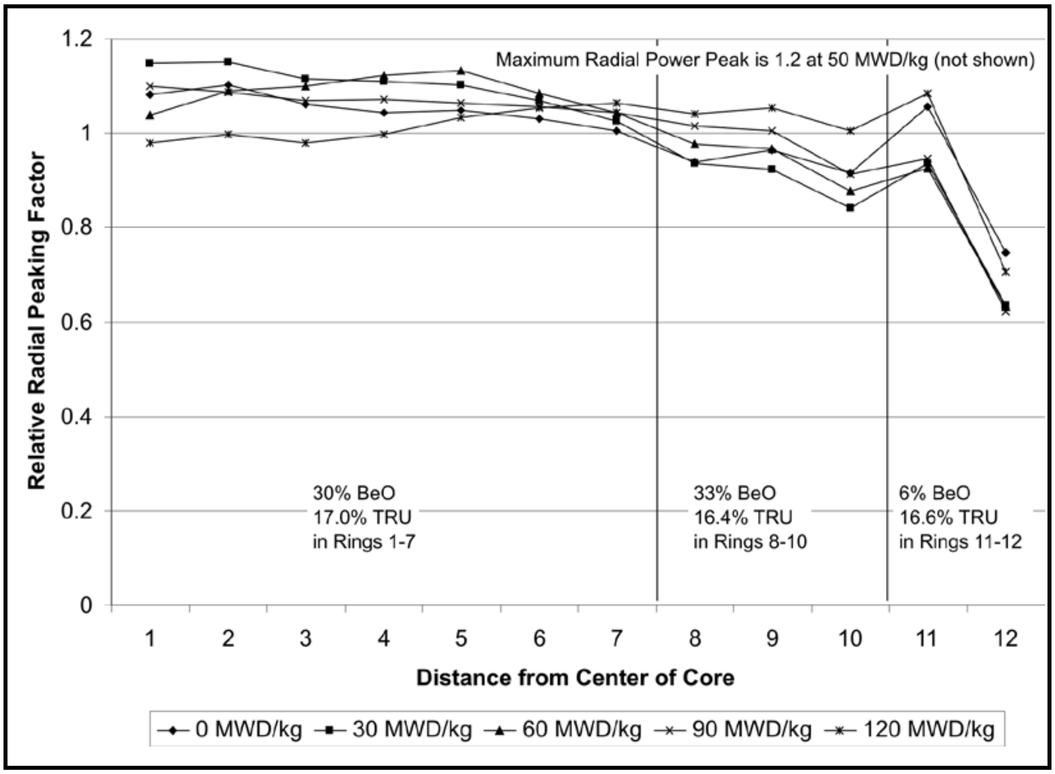


Figure 1. Reference power-flattened GFR core performance.

NUCLEAR ENERGY RESEARCH INITIATIVE

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

PI: Robert G. Odette, University of California, Santa Barbara

Collaborators: Oak Ridge National Laboratory, University of California, Berkeley

Project Number: 05-074

Project Start Date: March 2005

Project End Date: March 2008

Research Objectives

The primary objective of this research project is to develop an advanced high-temperature alloy class, called "nanostructured ferritic alloys" (NFAs), for use in advanced nuclear reactor systems. These alloys manifest remarkable creep strength, have excellent ductility, show good fracture toughness potential, and may also mitigate radiation damage. The researchers will address the combined effects of high temperatures (to ≥ 900°C) and high radiation dose on commercial alloys, especially MA957. They will also carry out model alloy experiments to better understand NFA thermo-kinetics and structure-property relations, in order to optimize processing paths and the balance of properties. They continue to collect data on and will conduct thermal aging experiments on 9Cr tempered martensitic steels (TMS) to assess embrittlement and high-temperature stability-aging limits.

The five major tasks in the program include:

- Develop a comprehensive mechanical property and microstructural database on MA957, other NFAs, and TMS, emphasizing the effects of irradiation on deformation and fracture; semi-empirical constitutive models, including high-temperature creep; and the master fracture toughness-temperature curve method for application to NFAs
- Conduct experiments on nano-microstructural evolutions and mechanical properties, for both commercial and model NFAs, and optimize processing paths, properties, and service lifetimes; thermal aging on NFAs and TMS; and post irradiation examination and data analysis of ongoing and future irradiation experiments
- Investigate advanced joining methods (e.g., solid-state welding and diffusion bonding) for MA957 and other NFAs that maintain beneficial NFA micro-nanostructures
- Develop models of the character, precipitation thermo-kinetics, and high-temperature thermal and irradiation stability of nm-scale precipitates in NFAs; the transport fate and consequences of helium; and compare the predictions to the results of thermal aging and irradiation experiments
- Conduct other activities, including collaboration in new irradiation studies and design of an advanced reactor surveillance-component monitoring program

Research Progress

Database and Physically Based Constitutive and Fracture Toughness Models. Researchers have continued to develop a large 9Cr TMS database to support developing models of irradiation hardening and embrittlement. This includes assessment of the effects of high helium levels in weakening grain boundaries and thus enhancing intergranular fracture that may lead to very severe embrittlement. They have added a significant number of tensile, Charpy, and fracture toughness data points to their database (approximately 1,600 data sets in total). They have collected, tabulated, and begun the analysis of tensile, creep, fracture, and microstructure data on MA957 and other NFAs. There are currently 246 data points in the NFA database. They have also developed a modified orientation-dependent master curve for extruded MA957 and identified the mechanisms leading to highly anisotropic, low toughness that is associated with both trigger particle impurity inclusions and crystallographic texturing.

Developing Processing and Alloy Stability Modeling Database for TMS and NFA. Researchers have procured approximately 800 g of mechanically alloyed Fe14Cr-YWT powders through a cost-sharing collaboration with ORNL and contracted for hot isostatic pressing (HIP) of the powders at 850, 1,000, and 1,150°C and 400 MPa. A second set of alloys, using the same type of powders, will be consolidated by hot extrusion at ORNL. Researchers will comprehensively characterize the micro-nanostructures in these alloys using various analytical techniques, including small angle neutron scattering (SANS), 3D atom probe tomography (APT), and scanning and transmission electron microscopy (SEM and TEM). They will combine this information with mechanical property data obtained from hardness, tensile, fracture toughness, and creep tests to establish micro-nanostructure property relations. Long-term aging at 900, 950, and 1,000°C of hardness, SANS, and utility coupons of MA957 have accumulated 1,375 hours of test time at temperature. Aging tests on two larger pieces of MA957 at 900°C and 1,000°C, to be used for various mechanical property tests, have accumulated 670 hours of aging time. Vickers microhardness and optical metallography carried out on tracking coupons aged at 900°C and 1,000°C for 735 hours showed a very slight decrease in hardness and no change in the coarse scale microstructure. The new model alloys and TMS, at lower aging temperatures, will be added to the aging matrix. Finally, researchers developed detailed plans for TEM studies to characterize the microstructures of in situ helium-implanted MA957 irradiated at 300 to 500°C and 5 to 10 dpa with 20 to 40 appm He/dpa. This collaborative effort will provide the first assessment of the transport and fate of helium in NFAs and the concept of helium management by trapping at the nano-features. These results will be compared to the corresponding observations on TMS as well as the predictions of multi-scale models.

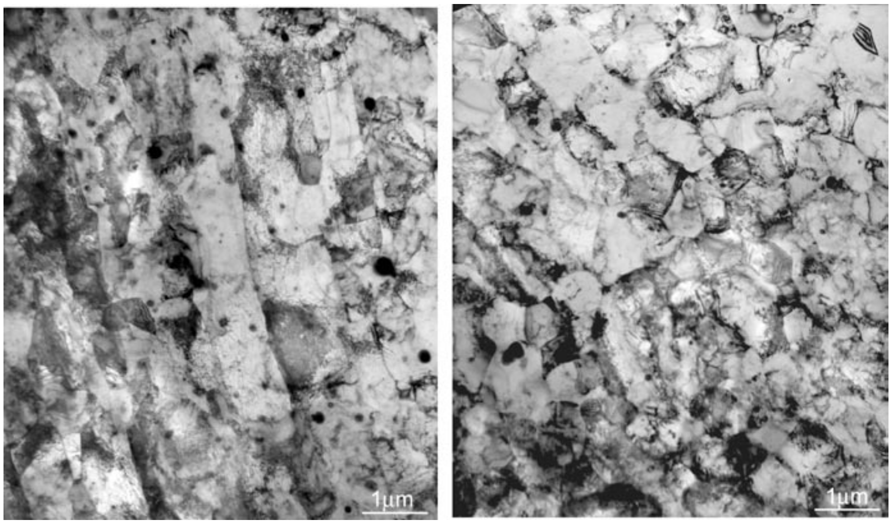


Figure 1. Microstructure of the FSW-welded MA957 (a) base and (b) weld metal.

Analysis parameters	Base metal	Weld metal - FSW	Weld metal - ESD
Average diameter (nm)	2.4	2.5	4.7

Table 1. Average diameter of nano-scale particles.

Development and Characterization of Advanced Joining Methods. Researchers used microhardness testing to measure the strength of MA957 joints created by friction stir welding (FSW) and Electro Discharge Welding (EDW) techniques, and characterized the stability of their nanofeatures using SANS and TEM. Examination of FSW welds showed a uniform fine-scale and equiaxed ferrite structure with significant density of dislocations in the joint region (Figure 1). Although the nano-scale particles were only slightly coarsened during the FSW welding, the electro-spark deposition (ESD) welds of MA957 contained porosity and the nano-scale particles coarsened significantly (Table 1). The microhardness of MA957 decreased slightly after the FSW welding, but was significantly degraded after the ESD welding, as shown in Figure 2. Thus, FSW appears to be a viable approach to joining NFAs.

Multi-scale Modeling of Alloy Performance. The team has developed a Lattice Monte Carlo (LMC) simulation code. They have also derived and verified the thermodynamic parameters describing various combinations of elements in the Fe-Y-O-Ti system by comparisons to published phase diagrams. Bond energies were obtained from the enthalpies of mixing within a regular-solution thermodynamics model supplemented by *ab-initio* density functional calculations. Simulations are performed on a body-centered cubic lattice, with oxygen diffusion on an octahedral interstitial sub-lattice, starting from a random super-saturated solution. The preliminary results support the initial formation of metastable coherent transition phase precipitates with a low oxygen-to-metal ratio, as observed by APT and SANS. The precipitates are remarkably stable, but at sufficiently high temperatures they eventually grow and transform to pyroclore $Y_2Ti_2O_7$ oxide phases. Researchers are also continuing collaborative modeling of helium transport, fate, and consequences in NFAs including the effects of He/dpa ratios on micro-nanostructural evolutions.

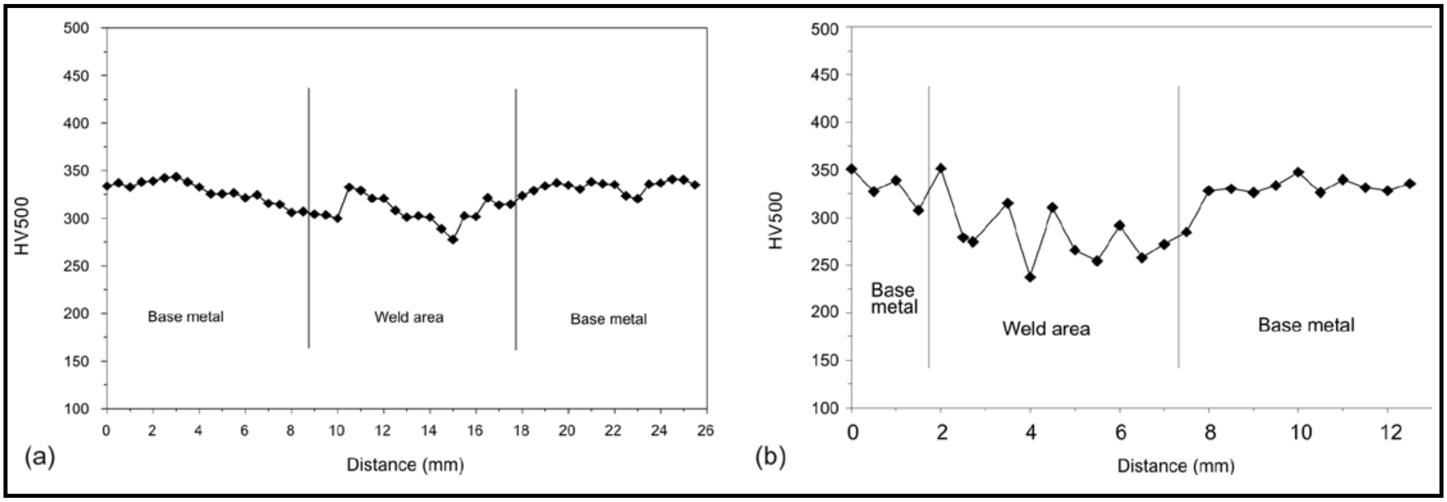


Figure 2. Microhardness profile across the weld of (a) FSW and (b) ESD-welded MA957.

Other Activities. The researchers have established a new collaboration with the Paul Scherrer Institute in Switzerland that will provide two capsules (pins) in the STIP 5 spallation proton irradiation (SPI) experiment. The capsules will include a wide variety of multipurpose specimens to characterize the effects of SPI on micro-nanostructural evolutions, as well as flow and fracture properties, including creep processes. An innovative approach to minimizing the very large temperature uncertainties in SPI experiments that are due both to high intrinsic heating rates and proton beam transients has been developed. The design seeks to more precisely control the temperature in critical specimen regions, like the process zone near a pre-crack tip. Better temperature control will be accomplished by reducing the number of interfaces to only one at the specimen-pin inner capsule wall. This interface will be engineered to provide a controlled and more precise temperature drop. Where they cannot be avoided, temperature gradients will be exploited to provide a basis for microhardness mapping of changes in flow properties as a function of temperature and dose; in some cases, gradients will be purposely enhanced to extend the range of accessible temperatures. The pins will operate at nominal centerline temperatures of approximately 300 and 600°C. The 300°C capsule will focus on the effects of high helium and hardening on fast fracture. The 600°C capsule will focus on the stability of NFA micro-nanostructures under irradiation and the ability of the nano-features to trap and manage high levels of helium. The capsule designs are supported by extensive finite element heat transfer analysis. For example, researchers have shown that the centerline temperature for the lower temperature pin can be controlled to 305±15ºC over most of the axial length (Figure 3). These experiments will produce an unprecedented database on the effects of SPI irradiations on the microstructures and mechanical properties of TMS and NFAs, including exploration of the effects of high helium on fast fracture methods to control and manage the damaging effects of high levels of helium.

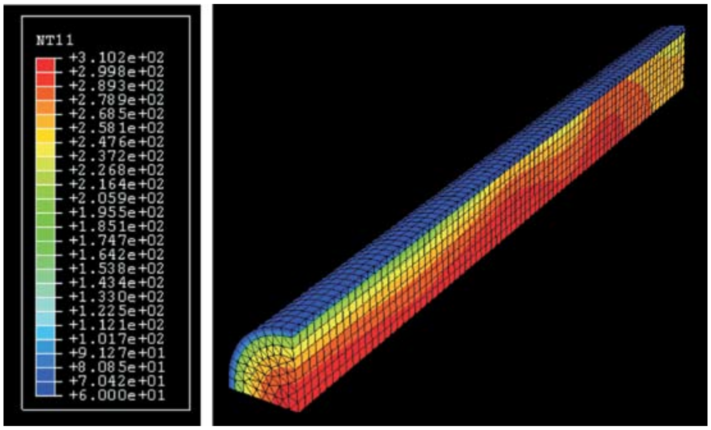


Figure 3. Temperature mapping over a 300ºC pin designed for the nominal power operation of SINQ.

Planned Activities

The accomplishments during the first year of this three-year project met or exceeded all the program goals. Following is a brief description of the major activities during the next project period.

Researchers will continue to build deformation, fracture, and microstructural databases on TMS and NFAs and develop physically based static constitutive, dynamic dispersion strengthening creep laws, and high strength NFA fracture toughness models. They will also analyze mechanical property and micro-nanostructural data on MA957 and the new model alloys in this task.

They will conduct microstructural and mechanical property characterization on new model alloys, including the extruded product, as well as MA957 base metal and solid-state joint material. Thermal aging studies will be continued and extended to the new model NFAs as well as TMS at lower temperatures. New small specimen methods will be developed and applied to measuring the high-temperature creep properties of NFAs. Extensive post-irradiation examinations will be carried out on helium-implanted NFAs from the ORNL High Flux Isotope Reactor (HFIR) irradiations.

Researchers will produce and characterize the microstructure and properties of diffusion-bonded NFA and NFA-dissimilar alloy joints. They will also acquire and characterize the microstructures of severely deformed MA957 with properties and microstructures typical of FSW-type joints.

They will continue to develop the LMC program emphasizing better treatment of misfit strain and excess vacancy effects, thermal coarsening, and the transition from coherent to partially oxide structures with pyroclore crystal structures with good lattice matching to the ferrite matrix. The simulated nano-feature structures will be compared to experimental observations obtained from TEM, SANS, APT, and positron annihilation spectroscopy. Work will also continue on the multi-scale helium transport, fate and consequences model, with considerable emphasis on comparison with new experimental data.

Finally, the design and fabrication of two capsules for irradiation in the STIP 5 experiment will be completed.

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: April 2005

Project End Date: April 2008

Research Objectives

In this project, researchers at the University of California, Berkeley, are studying advanced high-temperature heat transport and power conversion technology in support of the Nuclear Hydrogen Initiative and Generation IV. The work focuses on fundamental and applied questions for developing an advanced technology for high-temperature nuclear heat transport and power conversion using different combinations of gases (such as helium) and liquids (such as clean liquid salts).

This project contributes to four research areas. Following is a brief list of this project's contribution in each of those areas:

1) *Next generation nuclear plant (NGNP) intermediate heat exchanger (IHX) design.* During the first year, researchers developed design options for the 50 MW_t NGNP intermediate heat exchanger system to explore the major tradeoffs among a variety of intermediate heat transport fluids used for hydrogen production, such as high-pressure helium, intermediate-pressure helium, and liquid salt.
2) *Scaled transient heat transport integral experiments.* In this area, researchers will perform scaled, integral tests of transient high-temperature heat transport and heat exchange with gases and molten salt liquids. These experiments will generate data for model development and code verification.
3) *Multi-reheat helium Brayton power conversion.* In this research area, researchers will study the use of NGNP high-temperature Brayton cycle technology in multiple-reheat configuration for liquid-cooled, high-temperature Generation IV systems. Systems studied will include the modular lead-cooled fast reactor (LFR), advanced molten salt-cooled VHTR, and the Molten Salt Reactor (MSR). This technology will allow efficiency increases of 5 to 10 percent, while nearly doubling the power density.
4) *Advanced High Temperature Reactor (AHTR) design analysis.* This work will extend initial design studies of the MS-cooled AHTR as a potential high-temperature heat source for electricity and power production.

Research Progress

Researchers have completed the first requirements document for the NGNP intermediate heat exchanger, and have investigated liquid salt chemistry control methods. Because Tasks 3 and 4 are important to a liquid salt VHTR project being conducted by one of the collaborators, these tasks were undertaken first, delaying some planned Task 1 activities.

Following is a summary of the work that has been accomplished for each task:

Task 1. Researchers completed the initial functional requirements document for the NGNP intermediate loop, which describes the major design requirements for the application and use of liquid salt. One of most important viability and safety issues in using liquid salt as an intermediate heat transfer fluid for hydrogen production is chemical control. Ingress of sulfuric acid from hydrogen product loops into liquid salt results in strong corrosion of chromium from high-temperature metals and hydrofluoric acid generation. Because of the importance of this problem, it is receiving high priority and has been added to the scope of Task 1. After detailed literature reviews and thermodynamic study of salt chemistry control, the researchers propose to use cryolite (Na_3AlF_6) in solution with the liquid eutectic fluoride mixture LiF–NaF–KF (flinak) to keep oxygen potential low, and a liquid sodium bath to maintain favorable reducing conditions (fluorine and oxygen potentials). Based on experiments that have observed high solubility of beryllium metal in flibe (a molten salt of LiF and BeF_2), similar solubility may be observed for sodium in flinak. If high solubility does exist, dissolved sodium could provide local control of fluorine potential in the loop. The researchers have constructed an experimental facility to measure NaK solubility in liquid flinak salt. However, the first run of the experiment was unsuccessful due to several drawbacks in the design.

Task 2. The research team has written a paper summarizing the theoretical basis and main ideas for the scaled transient heat transport experiment. By using a category of light mineral oils to simulate liquid salts and adjusting the length, velocity, average temperature, and temperature difference scales of the experiment, it is possible to simultaneously match the Reynolds (Re), Froude (Fr), Prandtl (Pr), and Grashof (Gr) numbers in the scaled experiments. For example, the light mineral oil, Penreco Drakesol 260-AT, can be used to simulate the liquid salt flibe (Li_2BeF_4). At 110°C, the oil Pr matches 600°C flibe, and at 165°C, the oil Pr matches 900°C flibe. Re, Fr, and Gr numbers can then be matched with the length scale reduced to 40 percent, velocity scale to 63 percent, and temperature difference scale to 40 percent. The Weber number is then matched within a factor of two. Mechanical pumping power and heat input are reduced to about 1 percent of the prototype power inputs.

Helium fluid mechanics and heat transfer can be simulated by nitrogen following the same procedure. Helium at 750°C (1,023K) is similar to 130°C (403K) N_2 with the same length, velocity, and temperature scaling, and a pressure scale reduced to 11 percent of prototypical. Steady state and transient heat transfer to the steel and graphite structure can be reproduced with moderate distortion using Pyrex and high-thermal conductivity epoxies, respectively. Development of a preliminary experiment design and description for the Scaled High Temperature Heat Transfer (S-HT2) loop is underway and will be issued for review. With recent emphasis on development in LS-VHTR design work, the team is considering options to construct an S-HT2 loop to simulate high-temperature liquid salt fluid mechanics and heat transfer with light mineral oil in mixed-convection up-flow in heated channels, a flow regime of importance to the LS-VHTR.

Task 3. A study has been completed showing that the multi-reheat closed Brayton cycle provides an interesting option for indirect-cycle power conversion for 600 MW_t gas-cooled, high temperature reactors such as GT-MHR. The Brayton cycle exhibits several advantages: it has no net loss of efficiency compared to direct cycles; it has a fully non-nuclear power conversion system; and it operates at a higher average heat rejection temperature than an indirect combined cycle which allows economic dry cooling and desalination.

Researchers have also finished a preliminary analysis of coupling multiple reheat power conversion systems with advanced multi-effect distillation (MED) desalination systems. This can be used for producing potable water from seawater and other poor-quality water sources to further improve the economics. Closed gas Brayton cycles have an advantage over steam Rankine power cycles because gas cycles reject heat at a substantially greater average temperature. A preliminary study shows that with a water price at $0.60/$m^3$, 10 percent or more net revenue can be generated by producing desalinated water without affecting the electricity generation efficiency.

Task 4. The researchers believe it will be difficult to devise a workable insulation system for the LS-VHTR vessel. Therefore, they will examine uninsulated vessel designs in which the vessel remains at the salt pool temperature through an entire accident transient. Using high-temperature alloys with appropriate cladding, all in-vessel components are metallic and the IHX can also be located inside the reactor vessel. With the pool and vessel operating at a normal temperature of about 650°C and the core outlet temperature 750°C, the peak accident temperature will be about 850°C. The multiple-reheat helium Brayton cycles has about 48 percent thermodynamic efficiency at these temperatures and a 650°C vessel temperature is well outside the range where creep is an issue. Assuming up-flow in the reactor core, only the IHX will experience the core outlet temperature during normal operation.

Another key feature of this design is the use of a flibe primary loop, separate from the buffer salt in the reactor tank. This minimizes the amount of flibe needed, since it's expensive. The main advantage of flibe, besides neutronics, is that it has negligible gamma activity within a couple of minutes following reactor shutdown (like water does), so the pool provides effective shielding, eliminating a heavy shield cover over the pool as is needed for other coolants such as sodium. This could greatly simplify core access for in-service inspection and refueling. This new conceptual design for an all-metallic Advanced High Temperature Reactor with Metallic Vessel Internals (AHTR-MI) is being pursued. Figure 1 shows the schematic. The design will be demonstrated through a scaled pilot plant of approximately 50 MW_t.

The research team has published five papers on various aspects of this project, including two meeting presentations and three U.C. Berkeley reports:

- B. Laurenty, G. Fukuda, D.D. Damba, P.F. Peterson "Inhibiting corrosion by molten fluoride salts: investigation on Flinak," AIChE 2005 Annual Meeting, Cincinnati, Ohio, Oct. 31 – Nov. 4, 2005. (Selected for AIChE Nuclear Engineering Division Student Award for Best Paper).
- P. Bardet and P.F. Peterson, "Design of Scaled Integral Experiments for High Temperature Liquid Salt and Helium Fluid Mechanics and Heat Transfer," Eleventh International Topical Meeting on Nuclear Reactor Thermal Hydraulics, NURETH 11, Avignon, France, October 2-6, 2005.
- P.F. Peterson, H. Zhao, and G. Fukuda, "Functional Requirements Overview for a 50-MW(t) Liquid-Salt Intermediate Loop for NGNP," U.C. Berkeley Report UCBTH-05-007, Rev. B, December 29, 2005.
- P.F. Peterson and H. Zhao, "Design and Development Strategy for a First-Generation Commercial Advanced High Temperature Reactor with Metallic Vessel Internals (AHTR-MI)," U.C. Berkeley Report UCBTH-05-005, Rev. D, December 10, 2005.
- Per F. Peterson and Haihua Zhao, "An Optimized System for Multi-Effect Distillation Using Waste Heat From Closed Gas Brayton Cycles," U.C. Berkeley Report UCBTH-05-003, Rev. A, June 19, 2005.

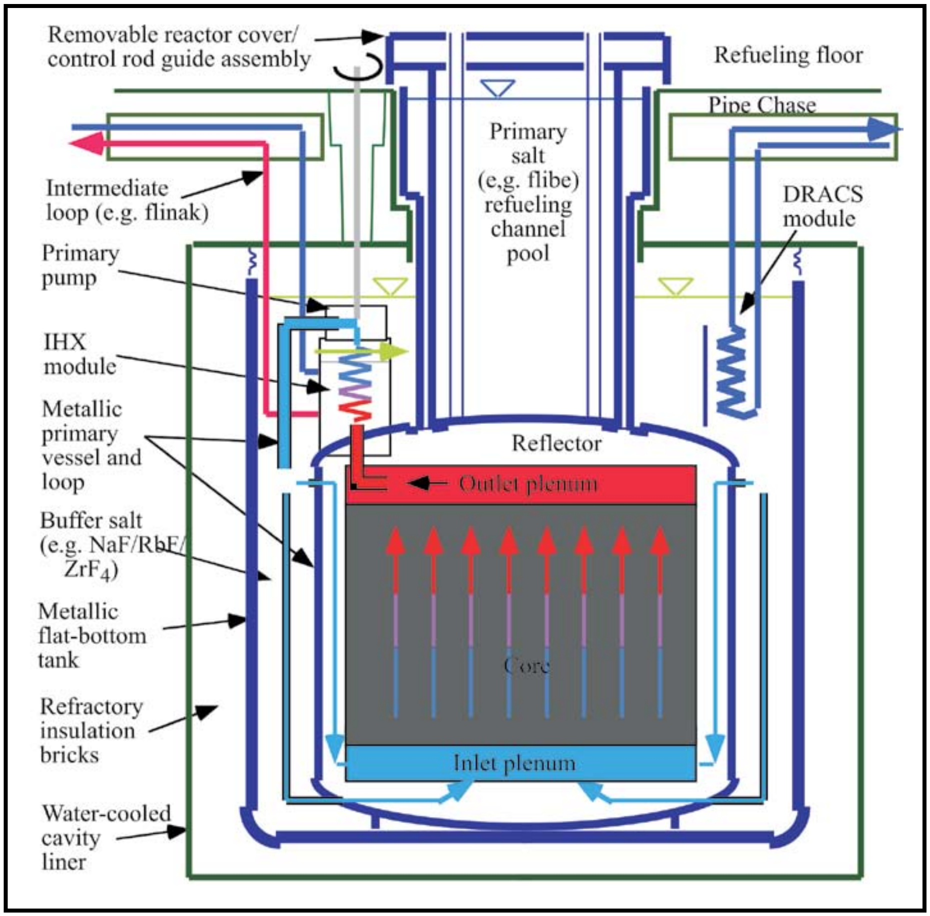


Figure 1. Schematic of all-metallic AHTR-MI.

Planned Activities

Progress on Tasks 2, 3, and 4 is ahead of schedule, while Task 1 is behind. Planned activities for Task 1 include completion of the Phenomena Identification and Ranking Table (PIRT) for the NGNP intermediate heat exchanger loop system and issuing the design and analysis reports. Additional activities for Task 2 include developing the experimental design and description for the Scaled High Temperature Heat Transfer (S-HT2) loop and issuing the design for review; demonstrating fabrication of PYREX components for the test loop and procuring other loop components; establishing cold, low-pressure fluid-mechanics characterization of the S-HT2 test loop, including measurement of gas flow distribution and liquid manifold flows; performing heated, full pressure steady state S-HT2 loop heat transfer tests; and performing transient heat transfer and structure thermal response testing using the S-HT2 test loop. For Task 3, the team plans to develop a pre-conceptual design and a design and analysis report for a lead fast reactor multiple-reheat helium Brayton cycle power conversion system and develop design and analysis reports for intermediate temperature and very high temperature AHTR multiple-reheat helium Brayton cycle power conversion systems. For Task 4, the team will develop low, intermediate, and high temperature AHTR heat transport loop designs, followed by scaled integral test designs, and will develop intermediate and very high temperature AHTR design and development plan reports.

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

Research Objectives

This project will develop quantitative safety goals for Generation IV nuclear energy systems. These safety goals will be risk based and technology independent. Researchers will develop the foundations for a new approach to risk analysis and a new operational definition of risk. This risk analysis approach 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. Results of this study will prove valuable to those government agencies charged with assuring the health and safety of the public as they manage, review, and approve these advanced reactor designs.

Research Progress

For the purposes of this project, risk is defined as the expected value of an undesirable consequence; that is, the product of frequency times consequence. Because these are highly uncertain events, frequency is interpreted as the annual probability of occurrence.

This project deals with three broad classes of risk:

1) **Risk to public health and safety.** This risk is a consequence of the movement and release of particulate, gaseous, or respirable quantities of radioactive materials to the environment due to accidents, impacting both the environment (ecological risk) and the health and safety of facility and co-located workers.
2) **Risk to public welfare.** This risk entails a threat to national and international security, and is a consequence of unauthorized movement of bulk materials (primarily fissile fuel: uranium and plutonium) and unauthorized release of fuel from the system to the environment.
3) **Risk to the environment.** This risk to the environment (ecological risk) includes accidental releases as well as intentional releases from any (geological) waste repository to the environment over very long time periods.

The measures used to quantify each risk will vary. Moreover, routine operations in a nuclear energy system can also lead to risk, such as radiation exposure to facility workers. Considerations aimed at reducing occupational risks, such as radiation protection, can also impact the design and will be treated at a later date.

Researchers formulated a technical approach that models a generic fuel cycle as a network, with nodes representing enrichment facilities, nuclear reactors, fuel storage facilities, reprocessing facilities, etc. These nodes are related to each other by material, energy, and information flows. They envision three types of risk: 1) the loss of bulk material from the network (e.g., fissile fuel such as plutonium) resulting in proliferation or public welfare risk, 2) the loss of particulate and gaseous material (e.g., fission products and respirable plutonium particles) from the network due to accidents resulting in a public health risk, and 3) the planned removal of material from the network (e.g., mill tailings, high-level radioactive waste) resulting in public health and ecological risk.

The researchers assumed that the U.S. Nuclear Regulatory Commission’s policy on safety goals in terms of quantitative health objectives will be applied to Generation IV reactors. However, the subsidiary goals utilized in risk-informed regulation, Core Damage Frequency (CDF) and Large Early Release Frequency (LERF), are specific to today’s light water reactors (LWR), and thus may not be applicable to the various Generation IV candidate systems.

Hence, there is a need for risk-based criteria that are "technology-neutral" wherever possible, especially during the down-selection process.

In those countries where there has been a significant effort on the part of a regulatory agency or a utility to define quantitative design objectives (QDOs) for nuclear power plants, there had been a consensus that the annual probability of a large atmospheric release of radioactivity, which researchers denoted as P(lr), should not exceed 10^{-6} per reactor year. "Large release" has rarely been precisely defined, but is usually taken to be 1 percent or more of the gaseous and volatile isotopes in a reactor core. However, the NRC, in its use of Risk-Informed Regulation, uses 10^{-5} as a limit for LERF as a surrogate for meeting the public health objective of one-tenth of one percent (0.001) of acute fatalities from all other causes.

A subsidiary design objective denoted by P(a) defines the maximum acceptable annual probability of accidents causing severe damage of the reactor core. Values for P(a) range from 10^{-6} per reactor year as adopted by the U.K. utilities, which implies little reliance on containment, to a value of 10^{-4} in the U. S. as a surrogate for meeting the public health objective of one tenth of one percent (0.001) of the background cancer risk (latent fatalities). Today, there is a view in the U. S. that an appropriate value for P(a) for advanced reactors should be 10^{-5}, meeting the NRC's Policy Statement on Advanced Reactors.

Because of the mathematical and conceptual difficulties in using a public health measure as a QDO, and to illustrate the risk-based approach described above, the research team began with values for P(a) and P(lr) that might be appropriate for Generation IV nuclear energy systems. In order to develop a set of QDOs, they will use a set of plausible probabilities based on experience with other nuclear systems. Two sets of QDOs are necessary: one set for protecting public health and one for protecting public welfare.

For the public health risk-based criteria, the researchers assumed that two types of events need to be considered:

- **Type 1:** Severe accidents leading to releases of respirable, gaseous or particulate matter, to the general environment surrounding the system, and sufficient to cause off-site actions such as the interdiction of foodstuffs, or the temporary evacuation of the public.
- **Type 2:** Severe accidents, even if there is not a significant release of particulate or respirable radioactive material to the general environment. Such an accident would undermine public confidence, both nationally and internationally, in the ability to operate nuclear power plants over long time periods. This would also constitute a very large economic risk.

Assuming that a Generation IV nuclear energy system should be of comparable public health risk to any other single nuclear facility, either nationally or internationally, such as a single nuclear power reactor, then the annual probability of a Type 1 accident resulting in a large release, P(lr), should be limited to 10^{-6} per year. Hence, the public in the vicinity of the system would be given the same level of assurance regarding a large release of respirable or particulate matter that they would have for other nuclear facilities worldwide.

With regard to the design target for Type 2 accidents that do entail significant radiological release, researchers assumed that these systems, satisfying the NRC Policy Statement on Advanced Reactors, will seek a goal for P(a) of 10^{-5} per year. Such a level of assurance would be more stringent than the level afforded current nuclear power reactors, from a public confidence viewpoint. However, given the public's concern for nuclear energy, such a level of assurance seems plausible.

For the criteria based on risk to public welfare, three types of events are considered: 1) loss of material accountability; 2) loss of material control; and 3) loss of material. In order to establish QDOs based on public welfare risk, the researchers assumed that loss of material accountability and control (MAC) leads to a loss of public confidence. Over a 100-year period, with 100 Generation IV systems in operation, a 0.1 percent chance (one chance in 10,000) can be assumed necessary to maintain public confidence in preventing Type 1 and Type 2 events. That is to say, the public might tolerate one event where there is a loss of material accountability and control, but not two events. This assumption translates into a P(MAC) on the order of 10^{-4} per year.

Because of the potentially large consequences associated with a loss of material (lm), researchers suggest a value of P(lm) on the order of 10^{-7} per year as a point of departure for discussion. The limit on P(lm) is an order of magnitude more stringent than the limit assumed for P(lr), which is meant to represent a greater significance attached to the release of bulk fissile material such as Pu than for respirable or particulate material such as Pu.

Accordingly, the four QDOs can be summarized as follows:

- Limit on the annual probability of a large accident: $P(a)=10^{-5}$/year

- Limit on the annual probability of a large release: P(lr)=10^{-6}/year
- Limit on the annual probability of a loss of material accountability and control: P(MAC)=10^{-4}/year
- Limit on the annual probability of a loss of material: P(lm)=10^{-7}/year

Researchers on this project will use these QDOs to develop functional design criteria, varying the four values for P(a), P(lr), P(MAC), and P(lm) to illustrate the sensitivity of the design to changes in the design targets. They will also take into account the major sources of uncertainty in the initiating events and in the event sequences themselves.

Planned Activities

Following is a summary of the tasks that will be performed to complete this research:

- Continue to 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 will account 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 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 Z. Podowski, Rensselaer Polytechnic Institute

Collaborators: The Royal Institute of Technology (KTH), Sweden

Project Number: 05-086

Project Start Date: March 2005

Project End Date: March 2008

Research Objectives

The primary objective of this project is to develop an accurate and carefully calibrated experimental database for supercritical water reactors (SCWRs). Specifically, researchers will develop correlations and models of fluid flow and heat transfer in these reactors. New and/or improved models are under development for future investigations of SCWR thermal-hydraulics under steady-state (stationary) and transient (dynamic) conditions. These models will be consistent with current computational capabilities of reactor systems, and will include a mechanistic approach to detailed flow and heat transfer phenomena. They will also incorporate the new, general, non-dimensional correlations that the researchers are formulating, which may still be phenomenological in form, but are based on sound physical concepts extending over a well-defined range of conditions. Researchers will validate the models against both currently available and newly acquired experimental data.

The focus of the work is on demonstrating that the proposed models will be applicable to study SCWR thermal-hydraulics under steady-state or transient conditions, using both system codes and computational fluid dynamics (CFD) codes. For numerical implementation application, researchers will use the advanced CFD code NPHASE. They will also conduct model testing and will validate the results against existing reactor systems analysis codes, such as RELAP.

Numerical simulations of supercritical water system dynamics will be based on the NPHASE code. The purpose of these simulations is to investigate flow and heat transfer conditions during transients associated with power and flow variations, as well as with system depressurization from supercritical to subcritical pressure and transition form forced to free convection.

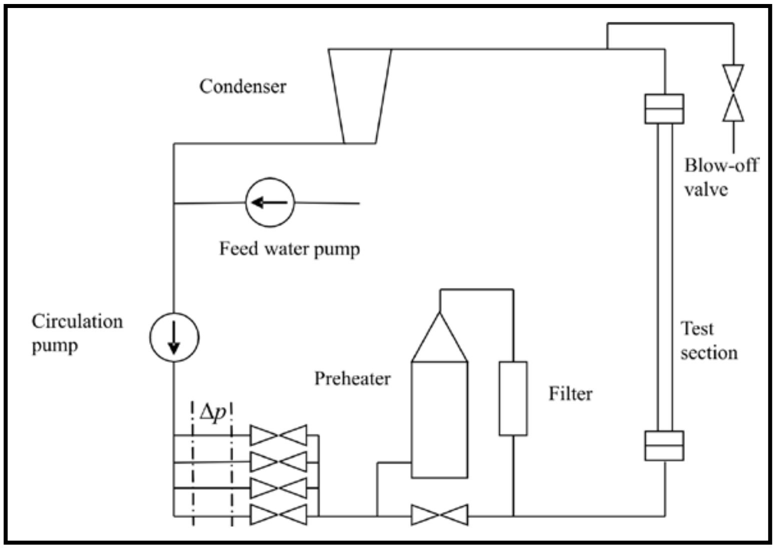


Figure 1. Schematic of supercritical water test loop facility.

Research Progress

Details of the organization and schedule of the experimental program for the Supercritical Water Test Loop (SCWTL) at KTH are being closely coordinated between RPI and KTH. The original loop was disassembled, moved to a new location, and reassembled. A schematic of the loop is shown in Figure 1. The results of initial experiments should be available in the spring of 2006.

Researchers are developing correlations and scalable models of coolant flow and heat transfer for application to thermal-hydraulic and safety analyses of SCWRs. They have conducted a literature review to collect and document the existing experimental data and models for SCWR thermal-hydraulic analysis. In addition, they have begun work on the mechanistic multi-dimensional modeling of flow and heat transfer in geometries similar to those that will be used in the SCWTL experiments. One of important issues for future detailed computer simulations is the development of an appropriate database for the physical properties of supercritical fluids and their derivatives. Data is necessary for such fluids as water and CO_2.

Researchers are developing multidimensional CFD simulation capabilities using the NPHASE code. They have carried out research to formulate a mechanistic, multi-dimensional modeling framework to use NPHASE in the scoping calculations and pre-test flow and heat transfer simulations in geometries similar to those that will be used in the SCWTL experiments. Sample results of preliminary calculations of a three-dimensional velocity field are shown in Figure 2.

Planned Activities

Researchers plan to continue this project by conducting the following tasks:

- Perform experiments using the SCWTL facility. Use the results of measurements to develop a carefully calibrated experimental database for the modeling of flow and heat transfer in water at supercritical pressures.
- Develop new mechanistic multi-dimensional models of turbulence and forced convection heat transfer for both water and CO_2 at supercritical pressures.
- Develop scalable correlations and simple models for application in system codes.
- Encode the new models in the NPHASE code and perform model testing and validation for both water and CO_2.
- Use the NPHASE code to perform numerical simulations of supercritical water systems corresponding to the anticipated designs of SCWR.

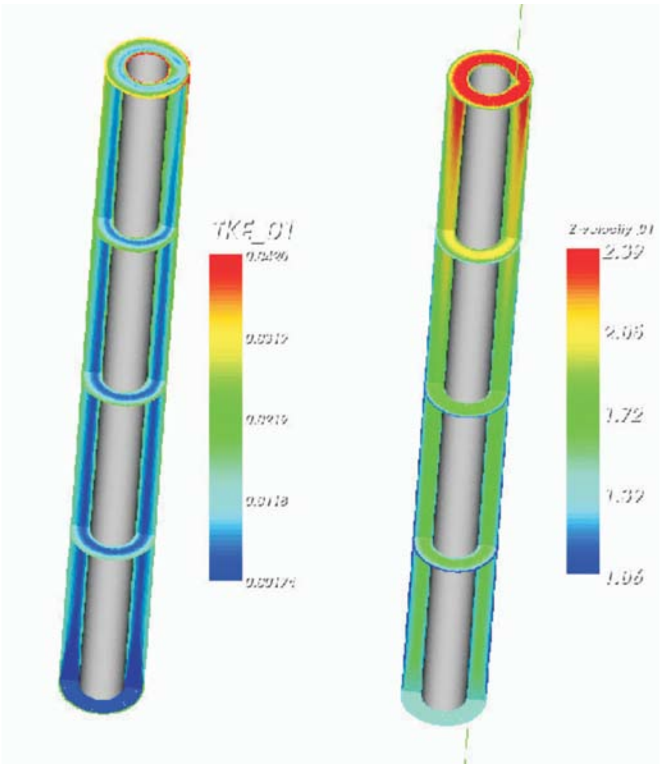


Figure 2. Three-dimensional velocity field.

- Actively collaborate with other research teams in the following areas:
  - Review experimental data for supercritical CO_2, for possible application in NPHASE simulations at RPI
  - Jointly develop improved models of turbulence for supercritical fluids, sharing the results of SCWTL experiments with foreign partners
  - Identify benchmark problems for simulations performed by the collaborating organizations

Novel Processing of Unique Ceramic-Based Nuclear Materials and Fuels

PI: Raman P. Singh, State University of New York at Stony Brook

Collaborators: Brookhaven National Laboratory

Project Number: 05-110

Project Start Date: April 2005

Project End Date: April 2008

Research Objectives

This primary objective of this project is to develop high-temperature refractory ceramic materials for Gas-Cooled Fast Reactors, including fuel, in-core components, and control elements. Refractory-based ceramics such as carbides, borides, and nitrides display a number of unique properties, including extremely high melting points, high 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 a conventional oven under an inert atmosphere. The materials produced will include metal ceramic carbides, mixed metal carbides, and unique metal silico-carbides. 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, manufacturing processes for fabricating a variety of unique high-temperature ceramic materials and fuels will be developed.

Research Progress

The research activities are broken into three primary tasks—materials processing, process modeling, and nuclear transport—discussed as follows.

Materials Processing. A key task of this effort is to fabricate silicon-carbide-based nuclear materials for both fuel and non-fuel components. This task has focused on fabricating composites comprised of a silicon-carbide matrix with hafnium carbide and uranium carbide/oxide inclusions. Researchers first acquired various raw materials and validated protocols for handling pyrophoric metal powders and radiological materials. They initiated processing of materials needed to fabricate hafnium carbide/silicon carbide composites, and they prepared Φ 25 x 30 mm preform plugs by compaction. The researchers then densified the plugs using the polymer infiltration and pyrolysis (PIP) procedure to form composites with a range of porosities.

The materials processing task also focused on generating kinetic data needed as input for the process modeling task. Samples of the polymer precursor are being heated from room temperature to 1,650°C to determine the evolution of several physical, chemical, and microstructural characteristics. These parameters include ceramics yield, determined from mass loss data and pycnometry; chemical changes up to pyrolysis temperatures, determined by Fourier transform infrared spectroscopy (FTIR); and microstructural changes beyond pyrolysis temperatures, from x-ray diffraction (XRD) and thermographic and differential thermal analyses (TGA/DTA). Experiments have been carried out for a small polymer precursor sample at different temperatures up to 900°C to investigate the pyrolysis process, and up to 1,650°C to investigate the amorphous to nanocrystalline conversion. The decomposition reaction in the experiments

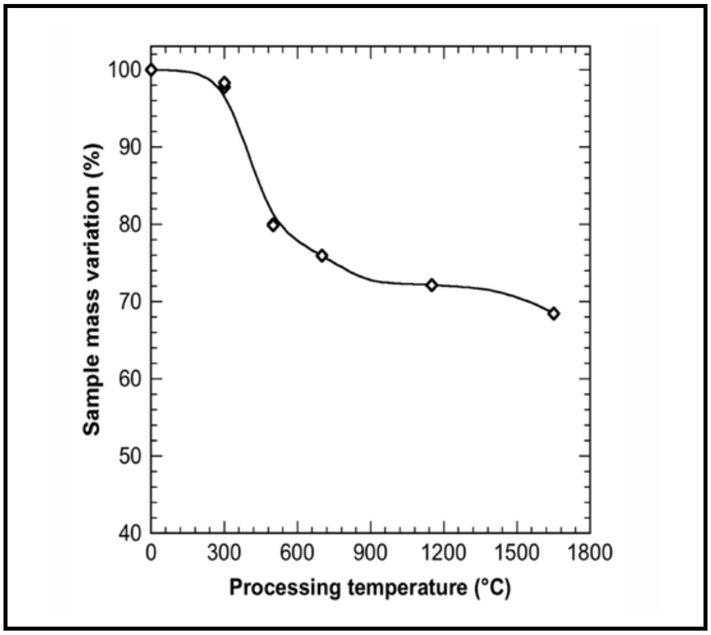


Figure 1. Data showing mass loss variation as a function of processing temperature, which indicates ceramic yield. Data is obtained from several experiments showing consistency.

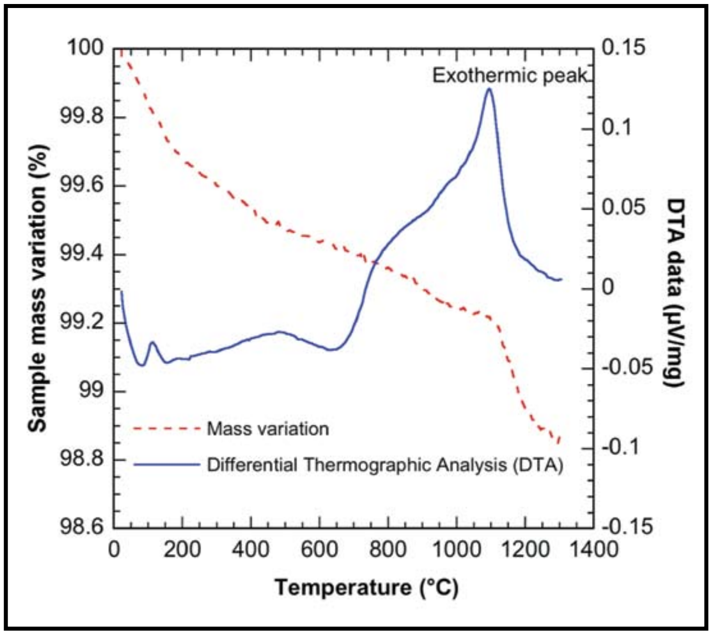


Figure 2. Mass loss variation (from TGA) and DTA data showing microstructural change beyond 900°C. The exothermic peak indicates crystallization.

results in the formation of silicon carbide and the release of gaseous decomposition products.

Figure 1 shows mass loss data as a function of temperature, to illustrate the ceramic yield. Figure 2 shows data from TGA and DTA, which indicates an exothermic reaction at approximately 1,100°C, signifying the onset of amorphous to nanocrystalline conversion in these materials. Detailed kinetic data is essential for accurate process modeling, which will then be used to guide material fabrication.

Process Modeling. Researchers have developed a porous media-based global process model for processing SiC-based metal-ceramic materials. This technique combines mass conservation, momentum conservation, energy conservation, reaction kinetics, gas transport, property variations, and crystallization process of a porous composite material. This model can simulate temperature or porosity changes, reaction rate evolution, gas pressure, and velocity. The heating rate is also included in the model so the heating condition and path for the experiments can be optimized based on the simulation result. The model can be used to guide the design of the system and process of experiments.

Besides the numerical pyrolysis rate, the analytical pyrolysis rate has been studied based on Arrhenius degradation kinetics in terms of a characteristic temperature and temperature range that is easily obtained from experiments. A driving force related to gases pressure and mass diffusion resistances is incorporated into the analytical pyrolysis rate equation, such that a more realistic volumetric pyrolysis rate equation is built to replace the simplified surface degradation kinetics. The pyrolysis kinetics parameters are generated experimentally, as outlined in the section on Material Processing, and substituted into the analytical pyrolysis equation.

Nuclear Transport: Material Composition and Fuel Configuration. The initial step in reactor core design involved the fuel element geometric shape. In the case of the gas-cooled reactor concept being considered in this study, two possible fuel element configurations were selected: a pin-type fuel-element-based core that is based on a similar design developed for an I-NERI Gas-Cooled Fast Reactor (GCFR) study, and a prismatic hexagonal block fuel element. The pin design consists of 271 fuel pins contained in a hexagonal can, with each fuel pin having a total length of 340.0 cm and an outer diameter of 0.845 cm. In the prismatic core design, fuel elements are assumed to be a prismatic hexagonal block containing fuel and appropriate cooling passages. The fuel can take the form of fuel “sticks” implanted in the solid material between the cooling passages or be infiltrated homogeneously in the solid component. Both these elements have silicon carbide as a base material. Initially, uranium carbide has been added to result in acceptable fuel.

The beginning-of-life fissile loading had to be chosen to be consistent with both criticality and safeguards requirements. The safeguards requirement will be satisfied by ensuring that the fuel enrichment remains less than 20 percent, which is the traditionally accepted value above which safeguards concerns are triggered. The criticality conditions are based on fissile loading used with the GCFR design, which had a core size similar to that of the current design. The GCFR ^{235}U fissile loading was 1,320 kg at 18 percent enrichment, giving a total uranium loading of 7,340 kg. The fuel volume of the currently designed core is 1,471 liters, which implies a uranium density of 4.989 g/cc in the fuel pellets.

At this stage, two assumptions are made regarding the composition of the uranium/silicon carbide pellet mixture: 1) a uranium metal (U) and silicon carbide mixture and 2) a mixture of uranium carbide (UC_2) and silicon carbide. In the first case, the uranium volume fraction was estimated by taking the ratio of the above uranium density to that of uranium metal (19.1 g/cc). The remainder of the pellet was assumed to be silicon carbide, and thus the nuclide number densities could be determined. In the second case, the original uranium number density was used to determine the reduced uranium carbide density. This value implied uranium carbide and silicon carbide volume fractions. Again, the nuclide number densities could be determined.

By comparing the atom and mass fractions for the two fuel compositions, it is apparent that carbon and silicon dominate the atom fractions, while uranium dominates the mass fractions. It was evident that uranium should contribute a total mass fraction of 67-70 percent of the fuel mixture, regardless of the method of creating the fuel pellet. In order to carry out criticality determinations, the atomic density (at/b-cm) is required. Atomic densities for the two fuel types postulated are given in Table 1. It is seen that the uranium number densities are the same for both cases, and only the carbon and silicon contributions change with fuel type.

Nuclide	**Case *a***	**Case *b***
Carbon	0.03569687	0.05003988
Silicon	0.03569687	0.02478658
U-235	0.002272787	0.002272787
U-238	0.01035385	0.01035385

Table 1. Atomic densities for the two possible fuel types.

Planned Activities

The material fabrication tasks will continue to focus on fabricating carbide and silico-carbide composite plugs for uranium, hafnium, zirconium, and niobium as various fuel and in-core materials. Additionally, reaction kinetics data will be generated for the neat polymer precursor containing uranium, hafnium, and other materials that are of interest in this investigation.

To complement the current global process model, a particle-based local model will be developed that is capable of predicting polymer-particle interaction, pore formation, and density variations. This local model will be incorporated into the global model. Finally, both the local and global process models will be modified to incorporate reaction kinetics due to the presence of uranium and other species of interest, along with the polymer precursor.

Fuel composition studies will include additional metal carbides, mixed into the fuel material, in order to change the material properties. An estimate of the behavior of fuel performance will be made following criticality and burn-up studies. Also, burn-up resulting from reactor operation will be used to determine the performance of the fuel once it contains fission products and higher actinides.

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

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

Collaborators: Los Alamos National Laboratory

Project Number: 05-114

Project Start Date: April 2005

Project End Date: March 2007

Research Objectives

The primary focus of this project is to address 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. The 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.

A central objective of this project is to further develop impedance spectroscopy for characterizing and monitoring corrosion in liquid metal systems. This advanced real-time corrosion monitoring method will then be used to study the kinetics of scale formation on selected alloys and to apply this understanding to develop new corrosion-resistant alloy compositions or surface treatment techniques.

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

- Develop the IS technique for lead and lead-bismuths systems
- Use enhanced IS techniques to measure the kinetics of oxide scale formation for a variety of materials and oxygen pressures at temperatures ranging from 400–700°C
- Develop improved surface treatments and alloy compositions for enhanced corrosion resistance

Research Progress

Significant progress on this project began in the summer of 2005, resulting in several areas of focus:

1) The need to find and/or develop a theoretical model of the defect concentrations of oxide layers and their corresponding impedance responses
2) The need to investigate the kinetic limitations of the gas dynamic equilibration method for oxygen control and to develop a work-around solution
3) The need to investigate the effect of the mechanical integrity of the oxide layer on the impedance response

Accordingly, the researchers launched a literature search to find an appropriate, physics-based model for the impedance responses of oxide layers. The point defect model developed by D. D. Macdonald, et al. (c. 1981), appears to be the best available model for this project, as it has been heavily referenced and built upon by other researchers for other corrosion systems and because it can interface well with Wagner's theory of gaseous, high-temperature oxidation. Currently, the assumptions made in the model are being re-evaluated and verified for liquid-metal systems.

The discovery of kinetic limitations of the gas dynamic equilibration method for oxygen control was unexpected, as it appears these limitations have not been addressed by other researchers. Essentially, gas phase equilibrium of the $H_2/O_2/H_2O$ system cannot be established in a reasonable timeframe below about 800°C. Due to the unreliability of determining oxygen content below 1 ppm via other methods, the need for assurance of the oxygen content of incoming gas is essential. Researchers devised a plan to work around these limitations by heating the incoming gas above 800°C to establish equilibrium, then quenching it to operating temperature and allowing it to scavenge oxygen from the lead-bismuth eutectic (LBE) via surface-catalyzed reactions. The necessary equipment to try this method has been acquired, but the experiments to verify this solution have yet to be conducted.

Mechanical integrity of protective oxide layers on the steel surfaces appears to be of utmost importance for the reliable use of IS techniques. The issue is that if the oxide layers have a significant defect in them—such as holes or cracks—or if there is a gap at the edge of the oxide layer where it meets up with another surface, then the measured impedance effectively drops to zero. Since LBE is electrically conductive, the electrical current flows through the LBE in the crack or gap, rather than through the highly resistive oxide layer, and thus the crack or gap shorts out the oxide impedance measurement.

Researchers are working on the development of a repeatable and quantitative experimental program to characterize this important effect of mechanical integrity. The plan is to poke holes in the oxide layers of steel samples using a micro-hardness measuring machine and then to compare the impedance responses of the samples before and after the holes were made. The ability to make measurable and repeatable holes using the micro-hardness machine should enable a systematic treatment of the phenomena.

As a starting point to verify the experimental techniques, researchers have duplicated some published results which were obtained with a similar experimental technique. An example of a typical impedance response for stainless steel 316L, pre-oxidized for 48 hours at 800°C and immersed for over 200 hours in LBE at 200°C, is presented in Figure 1. The parabolic effect of immersion time on the impedance response is shown in Figure 2.

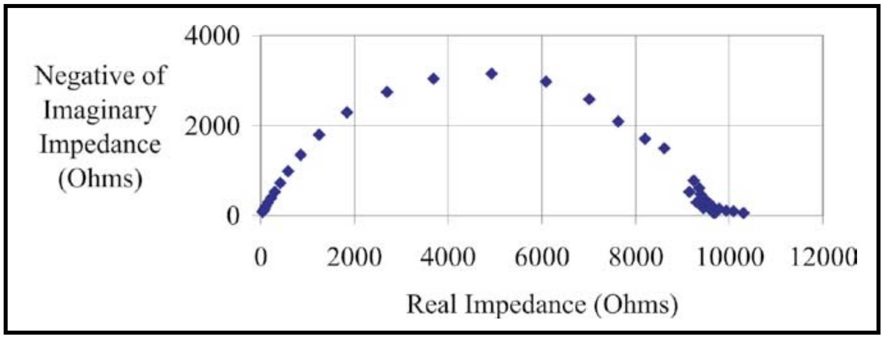


Figure 1. Complex plane ("Nyquist") plot of impedance response of SS316L, pre-oxidized for 48 hours at 800°C and immersed for over 200 hours in LBE at 200°C.

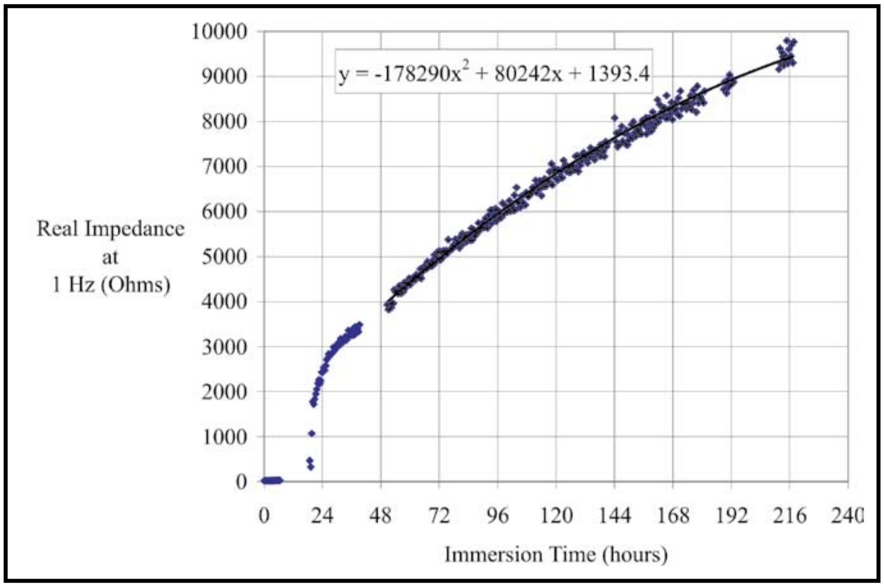


Figure 2. Effect of immersion time on the impedance response of the sample represented in Figure 1.

Planned Activities

Planned activities for the coming year include the continuation of the three focused research efforts described above: 1) verification and extension of the point defect model to liquid LBE corrosion, 2) experimental verification that heating and quenching the incoming gas can work around the kinetic limitations of oxygen control by gas dynamic equilibrium, and 3) experimentation to quantitatively determine the effect of mechanical integrity defects on the impedance response of oxide layers.

Successful completion of these activities will provide a strong base of confidence in the subsequent efforts to develop new IS probes; to characterize and analyze the IS responses of various materials; and to develop new alloys, surface treatments, and coatings to mitigate liquid-metal dissolution corrosion.

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: Los Alamos National Laboratory; Argonne National Laboratory; University of Michigan; Idaho State University; Washington State; the Belgian Nuclear Research Centre (SCK/CEN); Hokkaido University, Japan

Project Number: 05-116

Project Start Date: May 2005

Project End Date: April 2007

Research Objectives

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, and to extend this knowledge to increasingly complex iron alloy materials that are of interest for advanced reactor and accelerator-based applications. This information will be applied to the development of 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. Existing irradiation performance data was developed for the fast breeder program, but this database represents fast fission neutron spectra.

This project will address irradiation damage and effects in advanced reactor and accelerator-driven systems concepts. It 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. It 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 hydrogen/helium production in increasingly complex materials, ranging from the simplest single crystal iron, to polycrystalline iron and iron alloys, through advanced ferritic/martensitic steels.

The workscope consists of the following tasks:

- 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.

Research Progress

The objective of the first year of the research was to conduct proton and helium irradiations in single crystal, body-centered cubic (BCC) iron. These specimens were analyzed with positron annihilation spectroscopy and transmission electron microscopy to understand the effects of hydrogen and helium on the damage microstructure. Simulations were also performed to emulate the experimental conditions utilized and to follow the defect evolution processes.

Researchers completed irradiations of single crystal BCC iron for both the protons and helium. They conducted the 1 MeV proton irradiations at either 573K or 723K with doses ranging from 0 to 3 dpa. The helium (2.3 MeV) implantations ranged from 0 to 450 appm. The combination of both irradiations using helium and protons lead to four values of helium-to-damage ratios: 0, 10, 150, and ∞ (appm/dpa). The values of 10 and 150 appm/dpa were chosen since they are consistent with observed values in fusion and spallation systems, respectively.

Researchers pre-implanted polycrystalline and single crystal BCC Fe specimens with 3 and 30 appm of He (2.3 MeV), which were then thinned to expose the He implanted region and then subjected to *in-situ* transmission electron microscopy (TEM). The *in-situ* TEM utilized 250 keV protons to conduct uniform radiation damage throughout the specimens. They also conducted positron annihilation spectroscopy (Doppler broadening) with the single crystal BCC specimens that were irradiated with varying proton doses and helium concentrations. This beam positron annihilation technique allowed for depth profiling of the damage region of the specimens (Figure 1).

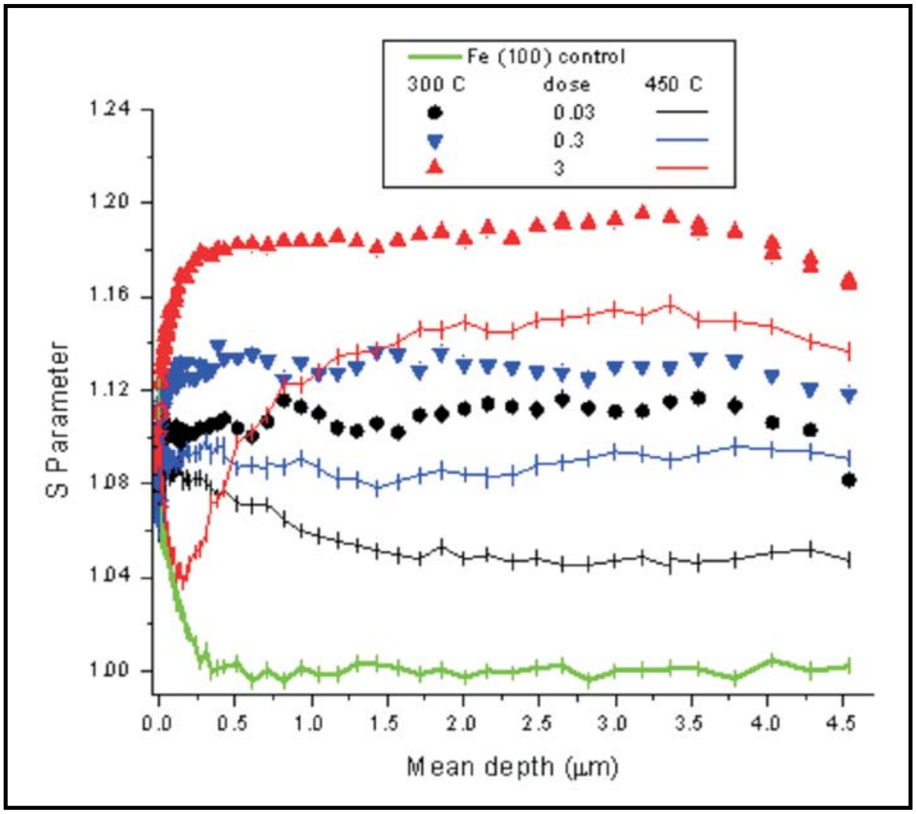


Figure 1. S parameter versus mean depth for the Doppler broadening results in single crystal BCC Fe for proton doses ranging from 0.03 to 3 dpa at 300°C and 450°C. Note that the 300°C specimens exhibit more open-volume defects than their 450°C counterparts at the same dose levels.

Molecular dynamics (MD) test simulations were conducted with WARP (a parallel MD code). These runs consisted of low-energy (2–10 keV) irradiations of the pure BCC iron and iron-helium systems at 573K and 723K with helium bubbles or helium interstitials of varying concentrations.

Planned Activities

During the next two years, this project will focus on the microstructural evolution caused by radiation damage in pure iron and iron alloys. The research in pure iron will continue on both the experimental and computational sides. This work will include additional molecular dynamics and kinetic Monte Carlo modeling of helium and hydrogen in BCC Fe. The experimental techniques will include transmission electron microscopy and positron annihilation spectroscopy for identifying and quantifying defects. The work with Fe alloys will examine how the alloying elements (i.e., chromium and carbon) affect the microstructural evolution when the system is exposed to proton irradiation and the presence of helium. This work will also be carried out with both similar experimental and computational techniques. These results will be compared to that of pure iron to understand the effects of the alloying elements.

NUCLEAR ENERGY RESEARCH INITIATIVE

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

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

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

Project Number: 05-143

Project Start Date: January 2005

Project End Date: December 2007

Research Objectives

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 from 900 to 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 up to 1,000°C. These are extremely challenging conditions for operating the metallic components in the intermediate heat exchanger and primary-to-secondary piping. Inconel 617, an advanced nickel-based alloy, has been identified as a leading candidate for such applications. However, material properties in a high-temperature, impure helium environment are not sufficiently understood to qualify this alloy for service. Therefore, this study will also investigate alloy and microstructure modifications to enhance material properties.

Research Progress

To date, researchers have completed the controlled-purity helium flow test system. The main purpose of this system is to provide a target gas mixture using pre-mixed gas bottles and a series of mass flow controllers. Figure 1 shows a schematic diagram of the components of the controlled impurity helium flow test system. The system consists of two basic subsystems: the controlled-impurity helium exposure system and the creep system. The controlled-impurity level helium gas mixture then passes through the test section, which consists of seven concentric

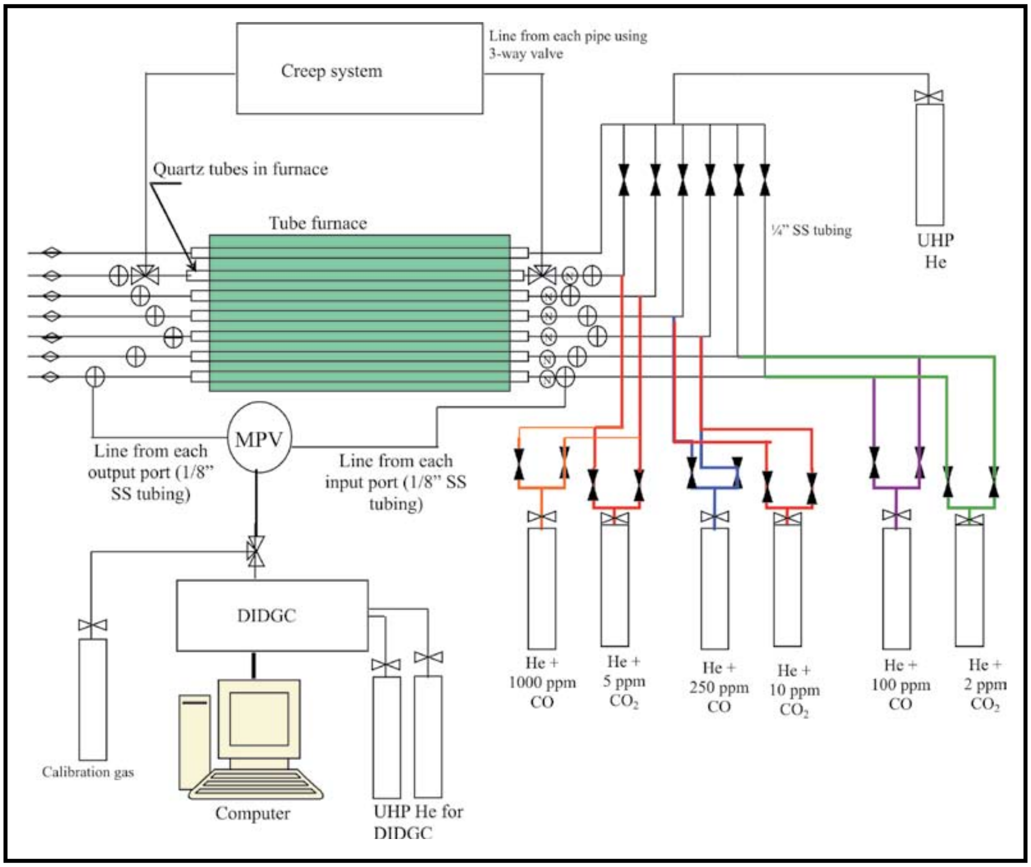


Figure 1. Schematic diagram of controlled-impurity helium flow system.

quartz tubes housed in a furnace. The discharge ionization detector (DIDGC) is used to analyze gas mixtures both before and after the furnace. With this system, exposure tests up to 1,000 hours will be conducted by exposing samples to He gas with known impurity gas concentration.

Five candidate alloys with compositions shown in Table 1 have been successfully cast using the Crystalox levitation cold crucible system. Microstructures of these alloys have been investigated by standard optical and scanning electron microscopy of metallographically prepared samples. (Figure 2). Homogenization studies of the five alloys were conducted and resulted in successful homogenization at a temperature of 1,316°C (2,400°F).

	Ni	Cr	Al	C	W	Re
J1-1	84		6.5		9.5	
J1-2	72.95	12	3	0.05	12	
J1-3	82.95	6	3	0.05	8	
J1-4	76.95	6	3	0.05	8	6
J1-5	79.95	6	3	0.05	8	3

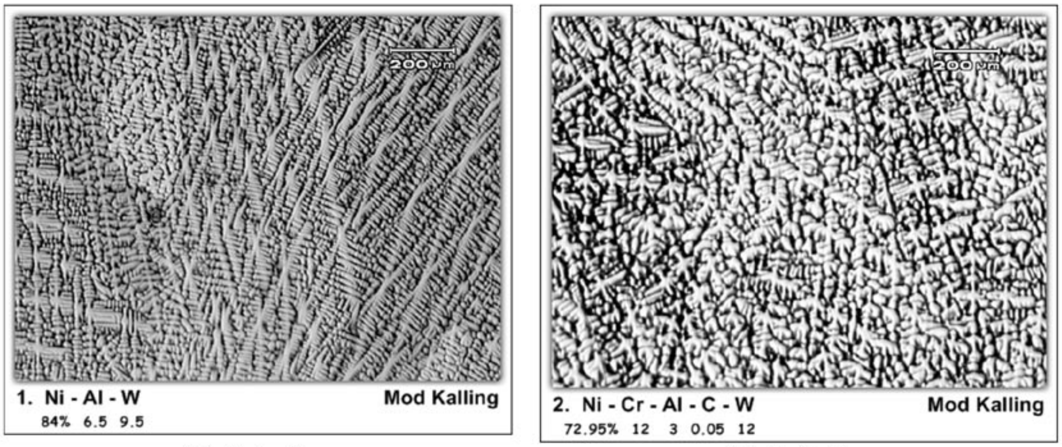
Table 1. Candidate alloys.

Planned Activities

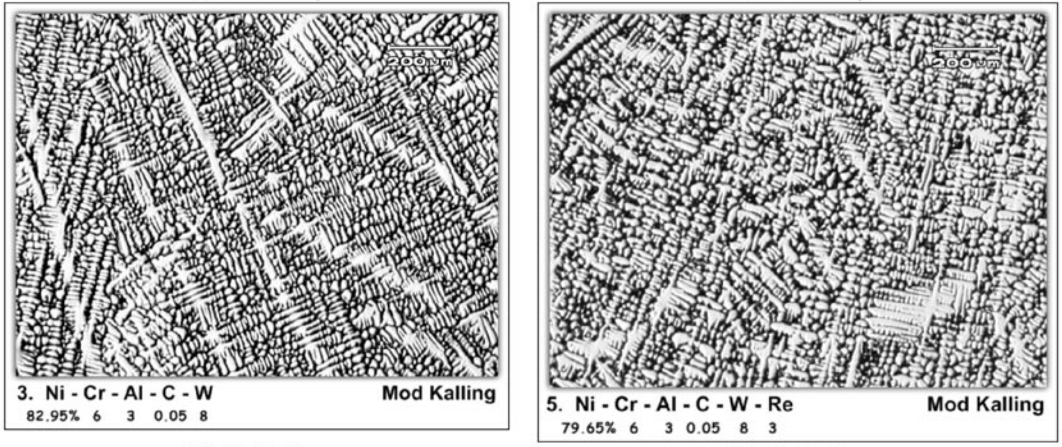
The flowing helium system is undergoing checkout and acceptance testing. Upon completion, researchers will conduct scoping experiments to determine the oxidation rates in a mixture of CO/CO_2 at temperatures between 850°C and 1,000°C.

Further confirmation of homogenization will be obtained from compositional analysis by microprobe studies that are underway. A set of 1,000°C compression creep tests will be performed on homogenized alloys to identify which alloys will be further developed.

Researchers are completing a custom tensile creep system designed for creep experiments in controlled He at temperatures up to 1,000°C. Following exposure test, researchers will perform experiments on creep behavior.



(a) J1-1 alloy (b) J1-2 alloy



(c) J1-3 alloy (d) J1-5 alloy

Figure 2. Microstructures of compositionally modified alloys by optical microscopy.

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: January 2005

Project End Date: December 2007

Research Objectives

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 increase efficiency while operating at high temperatures suitable for cogeneration of hydrogen. However, these same beneficial properties of supercritical CO_2 also present technical challenges to the regenerative heat exchanger design. To overcome these challenges, a compact heat exchanger is required.

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. Basic heat exchanger performance data are needed in order to develop an improved design for application to the supercritical CO_2 power conversion system.

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 or PCHE)
- Develop fluid flow and heat transfer simulation models and tools to support the evaluation of heat exchanger designs

Researchers will evaluate PCHE (or similar heat exchanger) performance for CO_2/CO_2 and CO_2/water heat exchange for the Brayton cycle application using a scaled experimental loop. Tests of two different designs of the

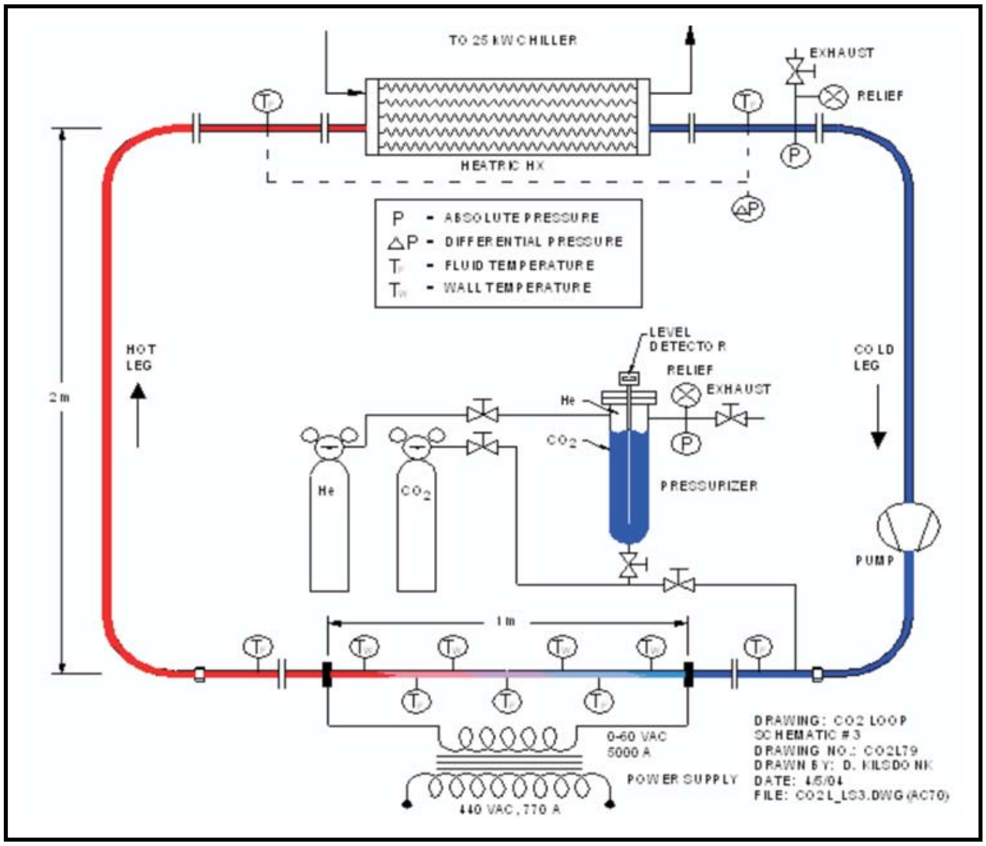


Figure 1. Schematic of operational water/supercritical CO_2 loop to test the PCHE.

compact heat-exchangers will be performed using the S-CO_2 loops to be put into operation at ANL-East. The two loops are: 1) low-pressure S-CO_2 /water and 2) low-pressure S-CO_2 /high-pressure S-CO_2. The objectives of the tests are as follows:

- Check the performance of the heat exchanger unit under design conditions, notably flow rate, pressure, and temperature
- Estimate the performance beyond design-basis such that the performance data can serve as a basis for model development and design optimization
- Compare the performance of PCHE (and/or compact heat exchange of similar design) to that of a reference heat exchanger design (e.g. shell-and-tube, helical coil, etc.)

A schematic of the low-pressure S-CO_2 /water loop is shown in Figure 1 and the solid model of the apparatus is shown in Figure 2.

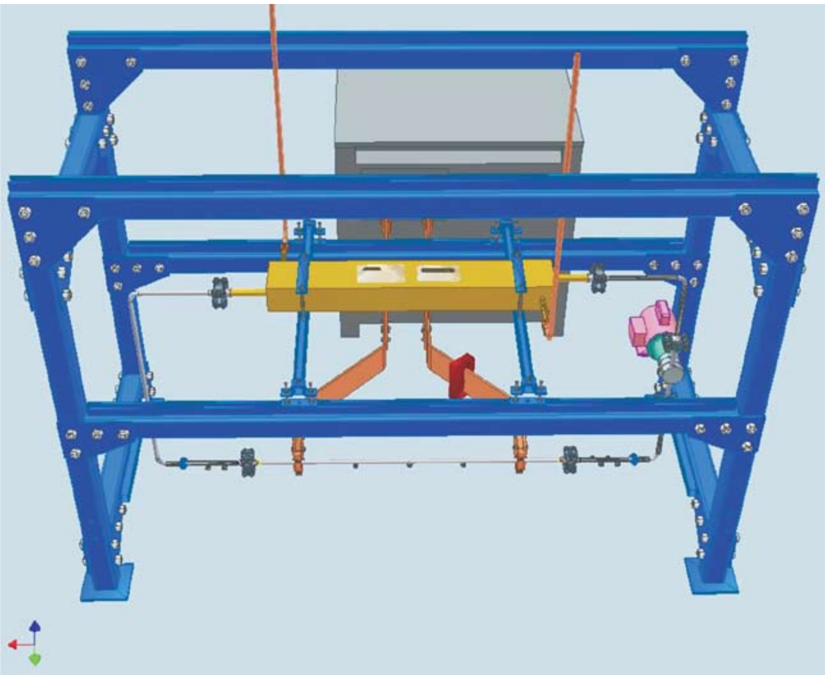


Figure 2. Front view of the water/supercritical CO_2 loop with the PCHE shown in brown.

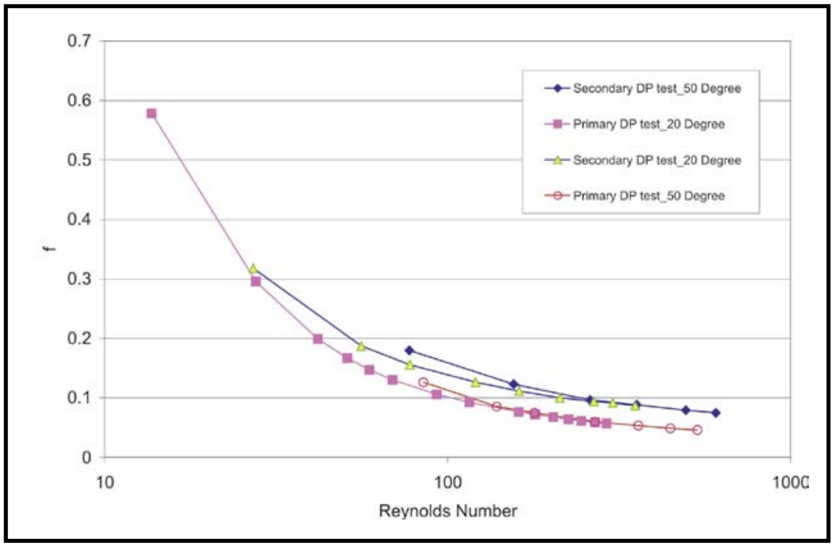


Figure 3. Isothermal friction factor measurements with hot and cold water.

Research Progress

Researchers have identified experimental needs for compact heat exchangers in the S-CO_2 Brayton cycle, including a review of S-CO_2 heat transfer data relevant to the recuperator/cooler application. They have also determined a range of conditions necessary to simulate the key phenomena that take place in recuperators and coolers. Researchers have initiated contacts with a vendor for procurement of a printed circuit heat exchanger (PCHE), identified as a promising candidate for the recuperator application, and have designed an experimental facility for performance testing of compact heat exchangers for S-CO_2. For CO_2/CO_2 heat exchange (i.e., recuperators, the facility will consist of two closed S-CO_2 loops: a low-pressure (about 7 MPa) and a high-pressure loop (about 20 MPa), connected via the PCHE. For CO_2/water heat exchange (i.e., coolers), only a low-pressure loop will be employed, provided by the existing SNAC test loop.

Researchers have developed a plan for testing the PCHE in the existing SNAC test loop. After replacing the existing tube-in-tube heat exchanger with the PCHE, they Initiated CO_2/water heat exchange testing, simulating the cooler for the CO_2 Brayton cycle.

Researchers also reconfigured the test loop for water/water heat exchange testing and friction factor measurements. The purpose of the latter is the desire to establish that the printed circuit pattern on the primary and secondary sides is similar (it is proprietary and not provided with the HX design data). The existence of similar patterns allows the assumption that heat transfer coefficients at identical Reynolds numbers will also be similar for water/water heat transfer tests. Due to physical limitations of the experimental setup for water/water tests, it is necessary to have the same heat transfer coefficient on each side of the HX to calculate a numerical value of the heat transfer coefficient. The friction factors have been measured with both hot and cold water and the results, plotted in Figure 3, show that the friction factors of each side are similar.

The researchers are also using these measurements to calibrate the secondary side DP sensor against the coriolis flow meter so they can use the DP sensor to measure mass flow during the water/water and water/CO_2 tests. This is necessary because the turbine flow meter used to measure secondary side water flow has proven too susceptible to electrical noise from both the power supply and pump speed control.

Planned Activities

The research team will conduct the following future activities:

- Begin developing an approach to evaluate the performance of compact heat exchanger designs via means such as computational fluid dynamics and heat transfer simulation capabilities. Consideration will be given to heat transfer enhancement schemes, modeling and rudimentary balance-of-plant estimates, considering design of the high/low pressure CO_2 loop.
- Conduct PCHE testing for CO_2/CO_2 heat exchange, simulating the low-temperature recuperator for the CO_2 Brayton cycle at CO_2 temperature ranges from 85°C to 150°C and recuperator conditions of 74 bar CO_2 on the primary side and 200 bar on the secondary. Assess test data for development of an improved heat exchanger model for accurate sizing of the recuperators and coolers.
- Evaluate results of PCHE (and/or equivalent) performance testing and identify needs for improvement for the supercritical CO_2 Brayton cycle application.
- Develop an approach and tools to evaluate the performance of compact heat exchanger designs in the intended application using CFD and heat transfer simulation capabilities; consider heat transfer enhancement schemes and modeling thereof. Software tools development will begin when the project commences. Specific model development using these tools is projected to substantially take place over the last year.

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: January 2005

Project End Date: December 2007

Research Objectives

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

The objective of this project is to investigate degradation of materials in the supercritical water environment. First, researchers will study representative alloys from the important classes of candidate materials to analyze their corrosion and stress-corrosion cracking resistance in supercritical water. This analysis will include ferritic-martensitic steels, austenitic stainless steels, and Ni-base alloys. Corrosion and stress-corrosion cracking (SCC) tests will be conducted at various temperatures and exposure times, as well as in various water chemistries. Second, emerging plasma surface modification and grain boundary engineering technologies will be applied to modify the near surface chemistry, microstructure, and stress-state of the alloys prior to corrosion testing. Third, researchers will examine the effect of irradiation on corrosion and stress-corrosion cracking of alloys in the as-received and modified/engineered conditions by irradiating samples using high-energy protons and then exposing them to supercritical water.

Research Progress

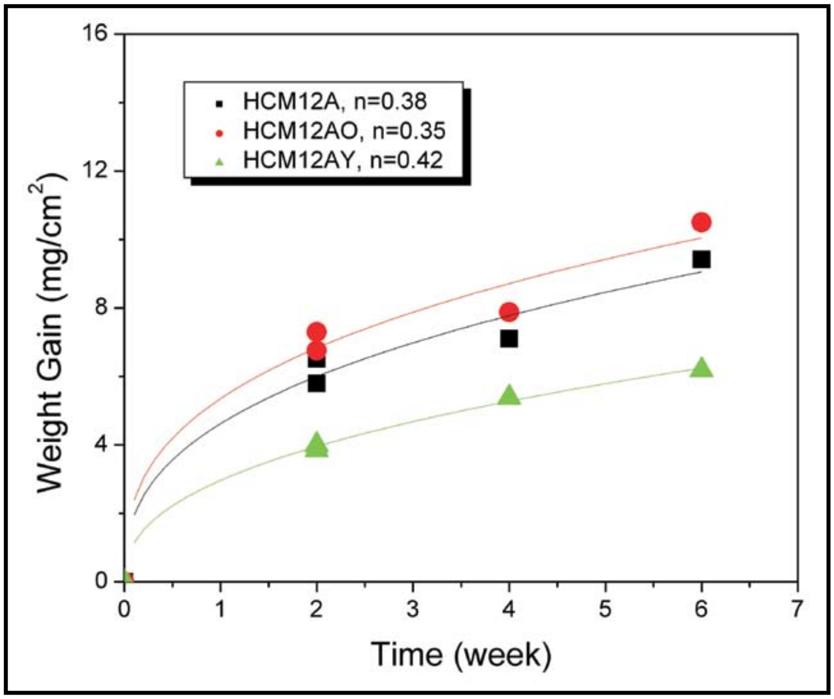


Figure 1. Weight gain of HCM12A as a function of exposure time in supercritical water (O=surface implanted with oxygen, Y=surface implanted with yttrium, and n is a power-law fitting parameter).

The first task performed this year was to separate the Wisconsin SCW loop into two separate loops, a thermal hydraulic studies loop and a corrosion/ SCC test loop. This separation allows for a greater flexibility in testing across multiple programs.

To understand corrosion performance at high temperature, a 600°C exposure over a six-week duration was completed. Eight different alloys were exposed, either in a polished condition or after having a surface implantation with oxygen or yttrium to change the surface chemistry. Additional samples were exposed after a grain boundary engineering treatment to examine the effect of grain boundary structure on oxide growth and stability.

Figure 1 shows the weight gain of a ^{12}Cr ferritic-martensitic steel as a function of time and surface treatment. The surface treatment with yttrium improves the corrosion resistance.

To understand radiation resistance of candidate alloys, researchers conducted an irradiation at 400 ± 10°C with 3 MeV protons to a dose of 7 dpa over a period of 8 days on alloys 316L, 690, and 800H. Beta counting indicated that all alloys were irradiated to the expected dose and that the sample-to-sample variation was small. Microhardness tests using a 25 g load were conducted to determine the degree of radiation hardening. Figure 2 shows that hardening was greatest in alloy 800H, followed by 316L and then 690. Typically, increased hardening due to irradiation corresponds with loss of ductility. Since the ductility is a desired property, smaller measured hardening is an indication of better radiation resistance.

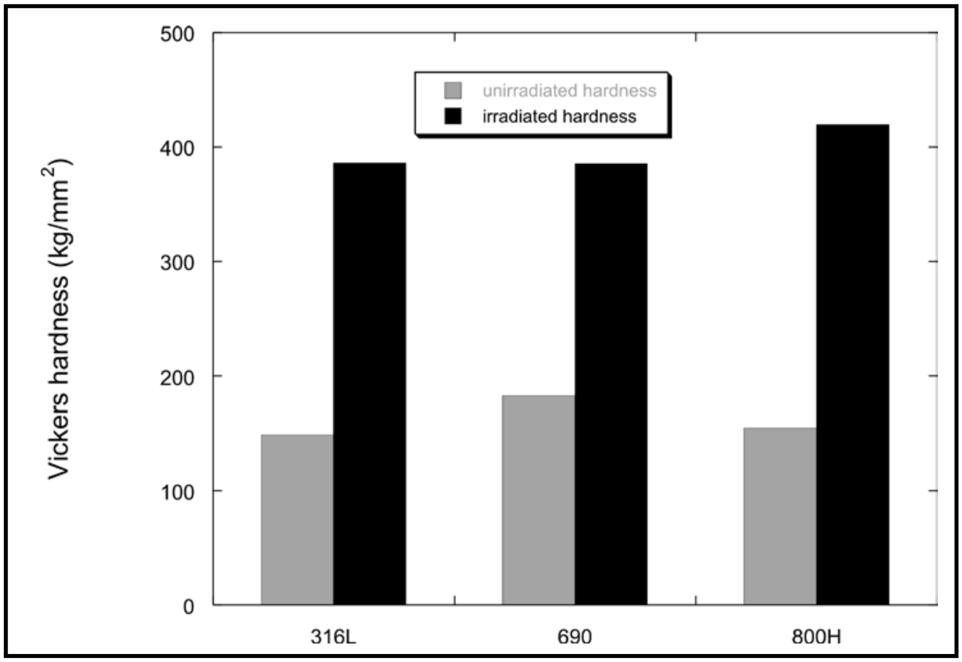


Figure 2. Hardness increase in candidate alloys due to irradiation at 400°C.

Planned Activities

To understand corrosion susceptibility, researchers will perform additional corrosion tests in the range of 350-600°C on samples with and without surface treatments and grain boundary engineering. Weld materials will be added to the experimental program. Samples will be examined using advanced microscopy techniques to gain mechanistic understanding of the oxidation process.

Transmission electron microscopy (TEM) bars will be prepared and initial TEM work will begin to characterize the irradiated microstructure. Stress corrosion cracking (SCC) samples will be tested in 400°C SCW to determine the susceptibility of the alloys in that fluid. In addition, TEM and SCC samples will be made from the D9 alloy and an irradiation to 3 dpa will be conducted at 400°C.

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: April 2005

Project End Date: December 2007

Research Objectives

Currently, two primary approaches exist for computational fluid dynamics (CFD) modeling of reactor systems. One approach is to use thermal/hydraulic analysis codes, such as RELAP, which model the entire plant using coarse nodes, but cannot predict small-scale flow details. Another approach is to use traditional CFD codes, such as FLUENT, which are adept at detailed flow and temperature predictions, but only 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, or Reynolds-averaged Navier-Stokes (RANS) equations. Because no single model can handle all geometries, research is needed to validate, modify, and improve CFD predictive capabilities.

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.

Research Progress

Geometry: Cylinder Array/Lower Plenum Model. Researchers compared the computational results from several different flow models against experimentally determined measurements. Specifically, they compared the k-epsilon $(k-\varepsilon)$, k-omega $(k-\omega)$, V2F (v^2-f), and differential Reynolds stress (RSM) turbulence models with particle image velocimetry (PIV) data, for flow across an array of cylinders representative of an idealized lower plenum model. In particular, results were compared over laminar to fully turbulent flow regimes (Reynolds numbers ranging from 237 to 55,920). Figure 1 shows minor pressure losses computed across the cylinder array. The results indicate that each of the turbulence models is able to predict the general trend of decreasing minor loss factor as a function of increasing Reynolds number. Over the range of Reynolds numbers, numerical results for all models are within approximately 10 percent of the experimental values. In addition, the models all closely approximate the slope of the experimental data. Overall, it is difficult to discern which model performs best.

Figure 2 shows recirculation lengths downstream of the wall-mounted cylinder (non-dimensionalized by the pitch) vs. Reynolds number. The experimental data shows

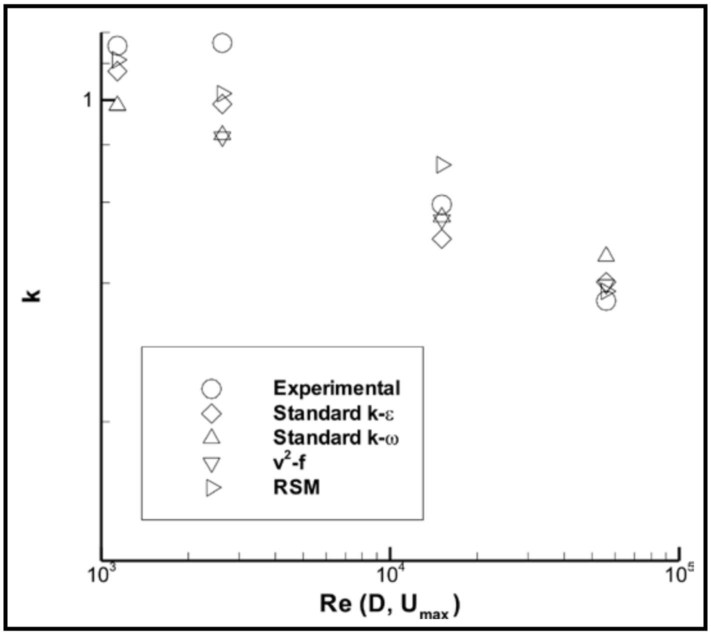


Figure 1. Minor pressure losses as a function of Reynolds number.

a slight increase, followed by a significant decrease in recirculation length as the Reynolds number is increased. The calculated results indicate considerable variation in the predictions of the various turbulence models. However, common among all models is a consistent over-prediction of the recirculation length at all but the lowest (laminar or near laminar) Reynolds numbers. The predictions using the $k-\omega$ model are furthest from the experimental data, while the $k-\varepsilon$ model predictions are the closest. Perhaps of greater concern is the fact that the RSM does not predict the correct sign of the slope, but predicts an increasing recirculation length with increasing Reynolds number over the entire range.

Geometry: Flow Along Rod Bundle with Spacer. Velocity distributions were measured in a prototype rod bundle consisting of two parallel rods downstream of a spacer grid using a two-dimensional particle image velocimetry (PIV) system. The measurements were taken in the Idaho National Laboratory matched-index-of-refraction (MIR) facility, which allowed the measurement of complete two-dimensional planes surrounding the model fuel rods. In all, a total of 33 planes were measured at two streamwise stations between two grid spacers which provided a full three-dimensional data set. The data provide benchmark velocity and turbulence measurements representative of flow phenomena and forced convection in complex reactor geometries.

Subsequently, the steady, three-dimensional Reynolds-Averaged-Navier-Stokes (RANS) equations were solved over this geometry using the general purpose CFD solver, FLUENT. Boundary conditions representing periodicity and symmetry conditions were applied where appropriate to reduce the model cell count, resulting in a fine grid discretized model consisting of approximately 900,000 cells. Results were computed using five different turbulence models: standard k-epsilon, realizable k-epsilon, shear stress transport (SST), k-omega, and V2f. All turbulence models were integrated to the wall, with a two-layer approach being used for the k-epsilon modls. Comparisons between experimental data and numerical results were made at streamwise locations 32, 80, 200, and 320 mm downstream of a grid spacer (which are streamwise periodic with a length of 40 mm, and a spacing of 440 mm). Results in terms of mean axial velocity profiles indicated that the k-epsilon and realizable k-epsilon models did a better job of predicting the experimental data than did the other three models. In particular, the k-omega, SST, and V2f models each showed a tendency to over-predict velocities in the narrow gap between the parallel cylinders and to over-predict the length of the wake behind the spacer. Results in terms of turbulence quantities were less conclusive.

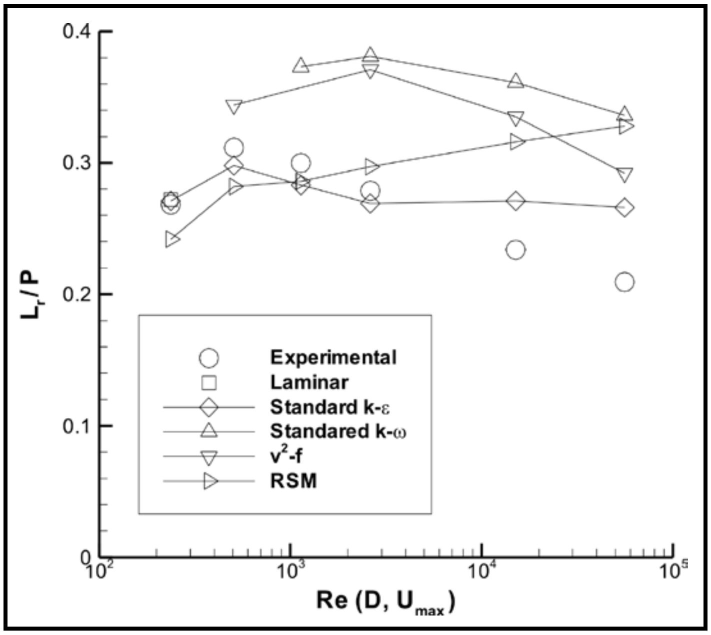


Figure 2. Dimensionless recirculation length vs. Reynolds number for the wall-mounted cylinder.

Planned Activities

The design for a rounded-exit jet experiment is nearly complete, and INL staff will shortly submit an order for the manufacturing of the model. Experiments on these rounded jet exits are planned in the INL MIR facility starting in July 2006 and lasting one month. Researchers will employ the new time-resolved, stereo PIV system, providing full 3-D data as a function of time.

The construction of the heated jet array facility has begun. The researchers anticipate that this construction will take about a month. Heated jet measurements using PIV will begin in November 2006, pending availability of the USU Experimental Fluid Dynamics Laboratory PIV system.

Fully three-dimensional RANS calculations for the cylinder array configuration will continue. Researchers will conduct Large Eddy Simulation (LES) calculations for the Shehata/McEligot heated tube data, which should be completed by the summer of 2006. They will also begin hybrid LES/DES (Detached Eddy Simulation) calculations for this geometry. Parallel jet calculations for both RANS and LES models will begin in the fall of 2006, and should be completed by the spring of 2007. During the final year of the project (2007), researchers will fully evaluate results of the test cases and propose any necessary modifications to the turbulence/subgrid scale models.

NUCLEAR ENERGY RESEARCH INITIATIVE

Ab-Initio-Based Modeling of Radiation Effects in Multi-Component Alloys

PI: Dane Morgan, Todd Allen, University of Wisconsin, Madison

Collaborators: None

Project Number: 06-006

Project Start Date: March 2006

Project End Date: March 2009

Project Description

The objective of this project is to develop an original, highly accurate thermokinetic model for austenitic stainless steels based on fundamental quantum mechanical calculations. New, more accurate microstructural models are necessary for the proposed Generation IV reactor designs in order to overcome limitations inherent in the existing models. This project will take advantage of the latest developments in *ab-initio* methods and alloy theory by developing a full thermokinetic model from quantum-mechanically-derived, atomic-scale energy parameters. The model will incorporate the true temperature- and composition-dependence of the diffusion constants and provide the missing information on interstitial motion.

Atomic-scale energy parameters for austenitic Fe-Cr-Ni will be calculated with *ab-initio* methods to establish accurate diffusion constants (unlike the approximate, extrapolated values currently in use). These parameters will be calculated using kinetic and standard Monte Carlo simulations combined with relationships derived from linear response theory. The resulting model will be refined by comparing simulated radiation-induced segregation (RIS) to a comprehensive database and validated against simulated and measured void growth in austenitic alloys. In order to demonstrate the general applicability of this approach and provide further validation, the model will be extended to ferritic Fe-Cr-Ni. While the complete *ab-initio*- to microstructural-modeling approach has been developed for binary alloys, this will be the first application to a ternary system and the first time that interstitials are included.

Workscope

This project will perform the following tasks in order to establish critical data and techniques for *ab-initio*-based modeling of microstructural development in complex engineering alloys:

- Perform initial *ab-initio* calculations of atomic-scale properties in pure elements and limited alloys
- Develop Fe-Cr-Ni RIS simulation and validation/refinement
- Perform more complete *ab-initio* calculations of atomic-scale properties in Fe-Cr-Ni alloy
- Develop Monte Carlo codes, Fe-Cr-Ni RIS simulation based on *ab-initio* values and Monte Carlo, and perform further validation and refinement
- Calculate diffusion constants, void growth modeling, and Fe-Cr-Ni RIS simulation
- Perform final validation/refinement by comparison to experiments
- Extend calculations and simulations to ferritic Fe-Cr model

NUCLEAR ENERGY RESEARCH INITIATIVE

Managing Model-Data-Introduced Uncertainties in Simulator Predictions for Generation IV Systems via Optimum Experimental Design

PI: Paul J. Turinsky, North Carolina State University

Collaborators: Idaho National Laboratory, Argonne National Laboratory

Project Number: 06-046

Project Start Date: March 2006

Project End Date: March 2009

Project Description

This research project addresses the understanding and management of uncertainties in modeling and simulation software due to the uncertainties that are inherent in the physical data utilized in the computer codes. This research will quantify the uncertainties and develop an optimum design for experiments to reduce these uncertainties. the approach will incorporate aspects of sensitivity analysis, uncertainty analysis, inverse theory, and mathematical optimization. To validate the capabilities being developed, researchers will apply the optimized design to a Generation IV nuclear energy system concept.

Specifically, this project will determine model uncertainties of key design attributes originating from the uncertainty associated with the nuclear data of a proposed Generation IV nuclear energy system. Researchers will design and optimize an experiment to reduce these model data uncertainties, utilizing the Idaho National Laboratory Zero Power Physics Reactor (ZPPR) facility as a test basis. They will conduct a simulated ZPPR experiment for the optimum design to obtain pseudo-observable core parameters, which will be utilized to calculate values of the adapted nuclear data. By understanding and quantifying uncertainties of complex engineering systems, appropriate design margins can be applied and rational cost-benefit decisions made.

Workscope

The following key activities comprise the project workscope:

- Determine which Generation IV core design to evaluate
- Define key design limiting responses of Generation IV core and design parameters/ instrumentation options for ZPPR core
- Establish neutronic models for Generation IV and ZPPR cores
- Develop cost models
- Evaluate key responses of the Generation IV core and sensitivity coefficients for ZPPR
- Develop simulation of ZPPR core and determine nuclear data matrix
- Determine matrix of key design limiting responses of Generation IV core
- Complete optimization of ZPPR core composition and instrumentation using virtual and design basis core models

NUCLEAR ENERGY RESEARCH INITIATIVE

Uncertainty Quantification in the Reliability and Risk Assessment of Generation IV Reactors

PI: Karen Vierow, Purdue University

Collaborators: The Ohio State University

Project Number: 06-057

Project Start Date: March 2006

Project End Date: March 2009

Project Description

The goal of this project is to develop practical approaches and tools for dynamic reliability and risk assessment techniques, which can be used to augment the uncertainty quantification process in probabilistic risk assessment (PRA) methods for Generation IV reactors. The objectives of the project are to develop practical approaches and computationally efficient software packages designed to test event tree completeness for Generation IV reactors, integrate a reactor safety code with PRA, and assess and propagate plant state uncertainties in the PRA analysis.

This project involves generating a practical dynamic event tree tool and assessing and quantifying uncertainty propagation. In Phase 1, current software for Dynamic Event Tree generation will be modified and linked to a best-estimate computer code (MELCOR for demonstration purposes). Key uncertainties in Generation IV modeling will be identified via the Phenomena Identification and Ranking Table technique. The integrated software package will be tested on selected, high-risk initiating events. In Phase 2, the computational efficiency will be improved by coupling the Dynamic Event Tree generation software with sampling software developed by Sandia National Laboratories. A purely numerical version of Devooght's and Dessar's uncertainty propagation methodology will be developed from a semi-analytical form and incorporated into the coupled best-estimate code/dynamic event tree generator package. Finally, the new software will be tested for selected initiating events.

Workscope

The following key activities/tasks will be performed:

Dynamic event tree generation

- Develop the SCHEDULER, PROBABILITY MODULE, and DATABASE MANAGEMENT SYSTEM and link to the MELCOR code
- Determine key uncertainties in reactor modeling and high-risk initiating events
- Test the software on selected initiating events

Uncertainty propagation and quantification

- Link the coupled MELCOR/DET generation scheme to the SNL Monte Carlo/Latin Hypercube Sampling tool
- Test for selected initiating events for the Pebble Bed Modular Reactor (PBMR)
- Modify the Devooght and Dessars methodology from semi-analytical to a purely numerical algorithm and incorporate into the PROBABILITY MODULE of the coupled MELCOR/DET generation scheme
- Test the new software for initiating events

NUCLEAR ENERGY RESEARCH INITIATIVE

An Advanced Neutronic Analysis Toolkit with In-line Monte Carlo Capability for VHTR Analysis

PI: William R. Martin and John C. Lee,
University of Michigan

Collaborators: Studsvik of America, Idaho National Laboratory, Los Alamos National Laboratory, General Atomics, Oak Ridge National Laboratory, and TransWare Enterprises, Inc.

Project Number: 06-068

Project Start Date: March 2006

Project End Date: March 2009

Project Description

The goal of this project is to develop, implement, and test a lattice physics code for very high temperature reactor (VHTR) neutronic design and analysis. This code is based on a production-quality lattice physics code used in LWR analysis and is augmented by Monte Carlo capability to treat resonance absorption in TRISO particle fuel. The approach takes advantage of the highly developed capabilities available for light water reactor neutronic analysis, in which lattice physics codes generate effective cross sections at the assembly level. These cross sections can be used in nodal codes to allow efficient calculation of global flux/power distributions and k_{eff} as a function of fuel depletion and temperature.

This project will incorporate the capability of the Monte Carlo code, MCNP5, directly into the lattice code, CPM-3, to establish "proof-of-principle." Code linking will be accomplished through an interface that will enable the MCNP5 capability to be extensible to other cross-section generation codes as well. This capability will be demonstrated by linking MCNP5 to CASMO-4. The resultant package will inherit the substantial downstream capabilities of CASMO-SIMULATE, including cross-section generation for global nodal analysis and depletion, systematic preparation of cross-section sets for accident analysis, and efficient fuel cycle analyses and assessment of alternative fuel management schemes. The final result will be a validated neutronics methodology for VHTR design and analysis, including cross-section generation, global reactor analysis, depletion, and fuel management.

Workscope

Tasks associated with this project are listed below:

- Design the interface (API) and modify CPM-3 code
- Develop VHTR test suite and verify CPM-3/MCNP5 methodology
- Specify VHTR design parameters
- Port API to CASMO-4
- Assess capability to treat low-lying plutonium resonances for Deep Burn application
- Develop validation test suite and validate coupled methodology
- Generate cross sections for nominal VHTR design
- Assess potential for analyzing pebble bed fuel

NUCLEAR ENERGY RESEARCH INITIATIVE

Improving Corrosion Behavior in SCWR, LFR, and VHTR Reactor Materials by Formation of a Stable Oxide

PI: Arthur T. Motta, The Pennsylvania State University

Collaborators: Westinghouse Electric Company

Project Number: 06-100

Project Start Date: March 2006

Project End Date: March 2009

Project Description

This project is designed to establish a technical basis for corrosion protection of candidate materials for three different types of reactors: the supercritical water reactor (SCWR), the lead-fast reactor (LFR), and the very high temperature reactor (VHTR). The materials to be studied include ferritic-martensitic steels, austenitic alloys, and Ni-based alloys. In order to understand the mechanisms associated with corrosion behavior in these materials, a systematic study will be conducted on the nature of protective films formed during corrosion tests in simulated reactor environments. The overall objective is to understand why certain alloys exhibit better corrosion behavior than others by examining the oxide microstructure. Alloys that resist corrosion develop a protective oxide layer that limits the access of corrosive species to the underlying metal, leading to stable oxide growth. The differences between a protective and a non-protective oxide are determined by the alloy chemistry and microstructure. Very small changes in microstructure can significantly affect corrosion rate.

The techniques to be used are microbeam synchrotron radiation diffraction and fluorescence and cross-sectional transmission electron microcopy on samples prepared using a focused ion beam. Examination by x-ray diffraction and fluorescence resolves the crystal structure, texture, and composition of oxide layers at the sub-micron level. By complementing the examination with transmission electron microscopy, the researchers will precisely determine the structure of these layers and their impact on corrosion behavior of the alloys.

Workscope

The following tasks comprise the primary project workscope:

- Procure and characterize initial set of alloy oxides formed in SCW, VHTR and LFR
- Perform oxide characterization by TEM and synchrotron microbeam
- Perform data reduction and analysis to identify oxide phases and determine elemental distribution; compare oxide characteristics of as-fabricated and surface-modified ferritic-martensitic alloy oxides formed in different environments
- Formulate hypotheses for corrosion mechanisms; decide on modified set of alloy oxides for new examinations to test and confirm the previous results
- Identify mechanisms of surface modification enhancement of corrosion resistance by formation of stable oxide

NUCLEAR ENERGY RESEARCH INITIATIVE

Multi-Scale Modeling of the Deformation of Advanced Ferritic Steels for Generation IV Nuclear Energy Systems

PI: Nasr M.Ghoniem, University of California-Los Angeles

Collaborators: California State University Northridge

Project Number: 06-109

Project Start Date: March 2006

Project End Date: March 2009

Project Description

The primary objective of this project is to use the multi-scale modeling of materials (MMM) approach to develop an improved understanding of the effects of neutron irradiation on the mechanical properties of high-temperature materials (i.e., 650-700°C, compared to the current 550°C limit). Presently, there are no physically based models for high-temperature, in-reactor deformation of advanced ferritic/martensitic steels. The objective of this project is to develop such a model.

Empirical potentials are not well developed for alloys and cannot easily adapt to local changes in chemistry caused by impurities. In this project, researchers will combine *ab-initio* calculations with traditional molecular dynamics (MD) simulations of dislocation-defect interactions. Particular emphasis will be placed on determining interactions with oxide and carbide precipitates, which control the ductility and high-temperature strength of steels, and on simulating properties of radiation-damaged steels as a function of neutron dose. The analysis will also determine interaction with nano-voids, precipitates, and self-interstitial clusters during irradiation. This information will be used to enhance a comprehensive model of radiation damage and in-reactor deformation. Predictions will be made for in-reactor deformation of simplified geometry, with full microstructure information linked with the deformation field.

Workscope

The following key activities comprise the project workscope:

- Develop and test the hybrid atomistic-continuum model for body-centered cubic (BCC) metals
- Apply the approach to study dislocation in a pure alloy (Fe-Cr) host
- Perform simulations of interaction mechanisms and determine stress relationships
- Extend model to twin boundaries and study dislocation properties in Fe-Cr alloys
- Evaluate effect of H, He, and C on properties and continue simulations to determine suitable rate theory parameters
- Apply to other materials and extend models
- Compare Rate Theory model with experimental data and model Generation IV vessel sections

Advanced Fuel Cycle Initiative Projects

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: March 2008

Project Description

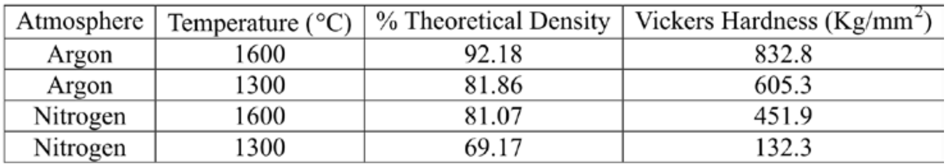
Fuel development under the Advanced Fuel Cycle Initiative (AFCI) will rely heavily on modeling physical mechanisms to predict performance, rather than conducting direct experimentation on nuclear materials or using empirical relationships derived from experimental data. This project will investigate a complementary strategy for fuel development based on performing experimental work on relevant surrogate materials. Researchers will study the structure-property relationship of nitrides and oxides in solid solutions using surrogate elements in order 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: 1) they will characterize the microstructure by measuring global texture evolution and local crystallographic variations, 2) determine mechanical properties (including fracture toughness, compression strength, and hardness) as functions of load and temperature, and 3) develop structure-property relations to describe mechanical behavior of the materials based on experimental data. Two new aspects of this research are 1) the use of crystallographic information in evaluating fuel performance and 2) the incorporation of statistical variations of microstructural variables into simplified models of the mechanical behavior of fuels. The primary objectives of this project are listed below. In conjunction with these activities, the research team will also carry out work with actual fuel materials.

- Develop processing procedures for pellet fabrication and characterize their properties
- Establish appropriate plutonium surrogates, fabricate samples, and optimize processing
- Translate techniques from surrogate processing to actual fuel; fabricate and characterize
- Establish models for high-temperature behavior, characterize properties, perform testing and analysis

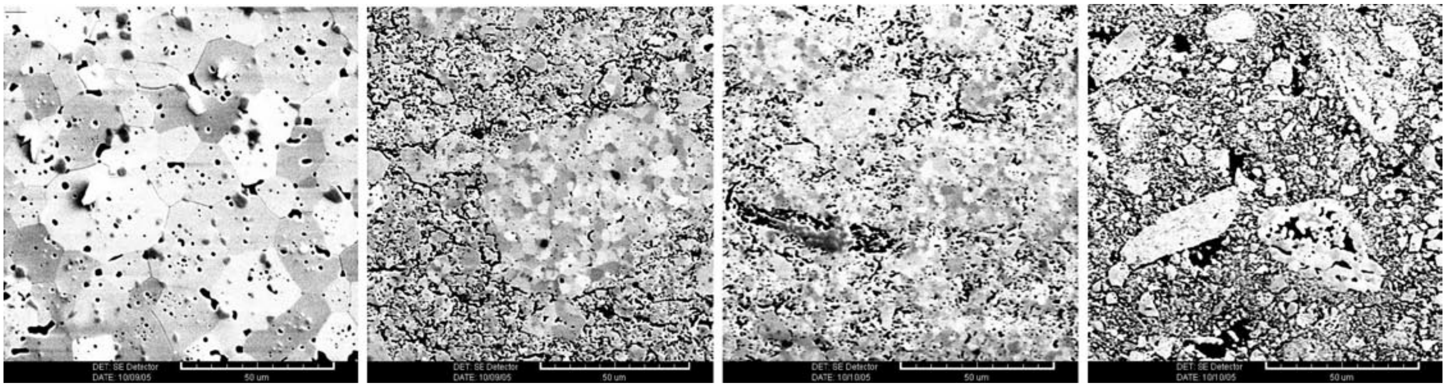
Research Progress

During the first year of this project, researchers have studied and elucidated various aspects of the processing, microstructure, and mechanical properties of sintered zirconium nitride (ZrN). They studied ZrN as a surrogate material for actinide nitrides, as well as a main constituent for inert matrix and transmutation fuel. One particular aspect that is central to this project is developing the capability to optimize processing conditions in order to control the resulting structure-property relations. Researchers conducted a parametric study of ZrN pellets sintered at various temperatures and gas atmospheres in order to identify optimal processing conditions. Some of the sintering conditions and resulting pellet densities and hardness are shown in Table 1.



Atmosphere	Temperature (°C)	% Theoretical Density	Vickers Hardness (Kg/mm^2)
Argon	1600	92.18	832.8
Argon	1300	81.86	605.3
Nitrogen	1600	81.07	451.9
Nitrogen	1300	69.17	132.3

Table 1. Sintering conditions, densities, and hardness values for sintered ZrN pellets.



(a) Argon 1,600°C (b) Nitrogen 1,600°C (c) Argon 1,300°C (d) Nitrogen 1,300°C

Figure 1. Typical microstructure of sintered samples. Scale bar is 50 µm in all images.

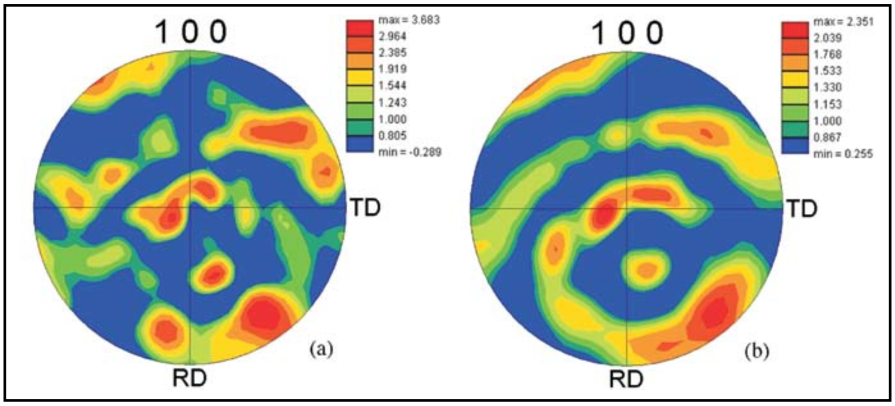


Figure 2. Crystallographic texture pole figures: (a) 1,600°C Ar; (b) 1,600°C N.

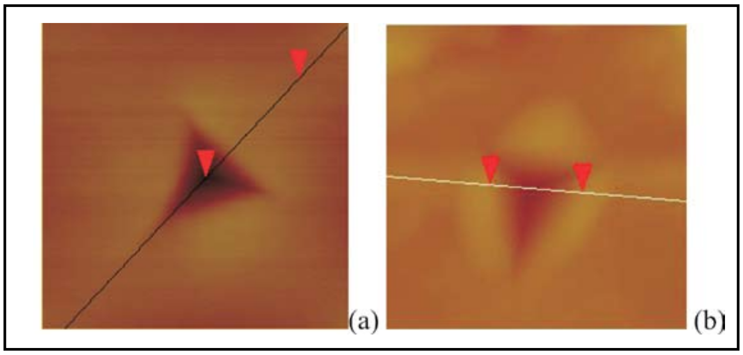


Figure 3. Surface topography around nanoindents in ZrN.

Note that samples sintered in argon result in substantially higher densities and hardness than similar samples sintered in nitrogen at the same temperature as well as other significant differences, as shown in Figure 1.

The samples sintered at 1,600°C, Figure 1a and Figure 1b, showed clear recrystallization and grain growth, but only the samples sintered in argon showed a fully developed microstructure. All other samples, particularly those sintered in nitrogen, showed a bimodal microstructure in which dense clusters are surrounded by a highly porous matrix. Note that sample sintered in argon at 1,300°C also shows extensive recrystallization, whereas the sample sintered in nitrogen is still in an earlier stage of particle consolidation and coalescence. The crystallographic texture of the samples was also measured for samples sintered at 1,600°C, as shown in Figure 2.

Figure 2 shows that the texture of the two samples appears similar, although the texture of the argon sample is sharper and has several localized peaks. These peaks are consistent with the presence of large grains (Figure 1a). The texture in the nitrogen-sintered samples is much smoother, which is a result of the presence of fine particles and small grains (Figure 1b). Taken together, these trends in sintered density and hardness, microstructure, and texture provide clear evidence that there is a dramatic difference in sintering kinetics between argon and nitrogen environments. Possible explanations for this may be related to higher density of point defects in argon-sintered samples due to nitrogen deficiencies, which can lead to faster kinetics. In addition, grain growth in samples sintered in nitrogen may be hindered by pore pressurization. Researchers continue to explore these and other mechanisms.

Another aspect of the mechanical behavior of ZrN that was investigated was surface-related plasticity. Through nanoindentation and Atomic Force Microscopy (AFM) experiments and Transmission Electron Microscopy (TEM) characterization, researchers found that dislocations can be present in significant quantities at the surface of ZrN. Furthermore, careful polishing can reduce the dislocation density at the surface to the point that a displacement excursion can be seen in a load-penetration curve during nanoindentation. Rough polishing increases the dislocation density and makes the displacement excursion disappear. Surface topography around the nanoindents (Figure 3) indicates sink-in for carefully polished samples (high-hardening rate) and pile-up for samples that underwent rough polishing (indicative of low-hardening rates as a result of pre-existing plastic deformation).

Planned Activities

Activities planned for the second year of this project include further investigations of the effects of sintering atmosphere and sample geometry in the sintering kinetics of ZrN by using gas mixtures such as Ar-H and N-H and different sintering die geometries. The kinetics of grain growth will be measured from these experiments as well as the evolution of the overall crystallographic texture and the local texture using Orientation Imaging Microscopy (OIM). The microstructure characterization to be performed for this purpose will be correlated with mechanical property measurements in additional samples made under different sintering conditions. The measurements will include hardness, fracture toughness, and compression strength. Creep experiments will be initiated using a 1,800°C furnace and mechanical testing setup that was recently acquired for this purpose. Deformation and failure mechanisms on samples used for mechanical testing will be evaluated using scanning and transmission electron microscopy, AFM, and OIM. The results will be used to propose basic structure-property relations in ZrN with emphasis on mechanical behavior.

Another important aspect of the planned activities for the second year is the selection of a second nitride to form a solid solution with ZrN to simulate the sintering kinetics of (Pu,Zr)N pellets for inert matrix fuels. A full battery of characterization techniques will also be used to understand structure-property relations in this case and some of these characterization techniques will be extended to Post-Irradiation Examinations (PIE) of irradiated fuels.

Finally, serial sectioning techniques will be implemented to perform material characterization via three-dimensional reconstruction of the microstructures. This will be used to gather statistical parameters in 3-D, including grain and pore sizes, as well as grain and pore shapes. The 3-D distributions will be compared with those obtained via 2-D sections and correction factors will be developed if the need arises. The reconstruction of the microstructure will also be used to produce geometric information to create preliminary finite element meshes to simulate the mechanical performance of nitride-based fuel.

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: January 2005

Project End Date: December 2007

Research Objectives

The major objective of this research project is to develop techniques for using self-propagating high-temperature synthesis (SHS) to produce ceramic nuclear fuels. The SHS process is used to synthesize advanced ceramic materials by capitalizing on the heat energy released from exothermic chemical reactions of the constituent raw materials. Researchers will first determine the fundamental SHS processing parameters using manganese as a surrogate for americium (Am) in order to produce dense Zr-Mn-N ceramic compounds. They will then transfer these fundamental principles to the production of dense Zr-Am-N ceramic materials. Due to the high vapor pressures of Am and americium nitride (AmN), there is concern about producing nitride ceramic nuclear fuels containing americium. Sintering results in retention problems of americium, which adversely affects the ability to synthesize a consistent product with desirable homogeneity, density, and porosity. Similar difficulties experienced during laboratory-scale process development for producing metal alloys containing americium have led researchers to abandon compact powder sintering methods and investigate other synthesis methods, such as SHS.

Another objective of this research program is to extend the SHS process for synthesizing Pu-Am-Zr-N and U-Pu-Am-N ceramic fuels. Researchers will obtain the fundamental SHS processing data to create these compounds using cerium (Ce) as the surrogate for Pu, manganese (Mn) as the surrogate for americium, and depleted uranium as the surrogate for uranium. The sequence for SHS synthesis of these ceramic fuels will be as follows:

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. 64 wt% $[(Ce_{0.5}Mn_{0.5})N]$ - 36 wt% ZrN

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. $(DU_{0.5}Ce0_{.25}Mn0_{.25})N$

Once sufficient fundamental data has been determined for these surrogate systems, the data will be transferred to INL for synthesis of Zr-Am-N, Pu-Am-Zr-N, and U-Pu-Am-N ceramic fuels.

Research Progress

The researchers have acquired stocks of zirconium (Zr), Mn, and manganese nitride (Mn-N) powders. Preliminary experiments using Zr and the Mn-N compound have been successful in yielding an SHS reaction. X-ray diffraction (XRD) analysis results showed the presence of Zr-N and Mn-Zr products. These data are the initial results in optimizing the Zr-Mn-N SHS process. Continuing experiments to optimize the reaction of Zr and Mn-N powders are being conducted, considering variations in stoichiometry, green density, and amount of diluent.

Researchers have performed differential thermal analysis (DTA) on the reaction between Zr + Mn-N reactants and Zr + Mn reactants and determined that they have an insufficient heating rate to generate reaction data. Thermogravametric analysis (TGA) will be conducted on the Mn-N compound to determine physical characteristics.

Investigation into the production of dense zirconium, manganese, and nitrogen (N) compounds has been initiated. Parts to modify existing laboratory equipment for the production of dense Zr-Mn-N compounds arrived and will be implemented by the end of the next quarter. A densification process using a Gleeble 1500 is being investigated as a preliminary step to using the induction hot press discussed in the proposal.

Preliminary research into the production of manganese nitrides (Mn-N) has been initiated. A self-igniting gel process using manganese nitrate is being explored. The resultant product of this method would be a powder that is usable in the current experimental set up. Further experimentation is required to determine the viability of this reaction.

Planned Activities

- SHS of porous Mn-N
- Differential thermal analysis of Mn + N_2 SHS reactions
- Optimization of SHS of Mn-N
- SHS consolidation of dense Zr-Mn-N compounds
- Combined SHS consolidation of dense Zr-Mn-N compounds
- Thermogravametric analysis of SHS formation of Zr-Mn-N compounds
- Additional surrogate studies for Pu, Np, Cm
- Optimization of SHS consolidation of dense Zr-Mn-N compounds
- Optimization of Am-N and Zr-Am-N SHS systems at INL
- Differential thermal analysis of Am-N and Zr-Am-N SHS systems at INL
- Optimization of SHS consolidation of Zr-Am-N system at INL

Minor Actinide Doppler Coefficient Measurement Assessment

PI: Nolan Hertel, Georgia Institute of Technology

Collaborators: Education, Research and Developmetn Association of Georgia Universities, Los Alamos National Laboratory

Project Number: 05-024

Project Start Date: April 2005

Project End Date: May 2007

Research Objective

The primary objective of this project is to assess the viability of measuring the Doppler coefficient of reactivity of minor actinides. Using a series of calculations, the researchers will estimate the change in reactivity resulting from a change in the operating temperature of small quantities (approximately 1-10 grams) of pure minor actinides. They will use the resulting data to design experiments for measuring Doppler coefficients.

Researchers are performing extensive scoping calculations as a function of five parameters (isotope, sample quantity, operating temperature, critical assembly, and data library) using the radiation transport code MCNP5. The objective is to determine the quantity of minor actinide material needed to cause a 10^{-5} change in reactivity due to a 200K change in temperature of the sample (e.g., from 800 to 1,000K). Researchers will use the results from these calculations to develop an experiment that will measure the Doppler coefficient of each isotope. Experiments will be developed for the FLATTOP and COMET critical assemblies at the DOE Critical Experimental Facilities. Several isotopes of plutonium, neptunium, americium, and curium are proposed for this study, including Pu-239, Np-237, Pu-238, Am-242m, Am-243, Pu-241, Am-241, and Cm-244.

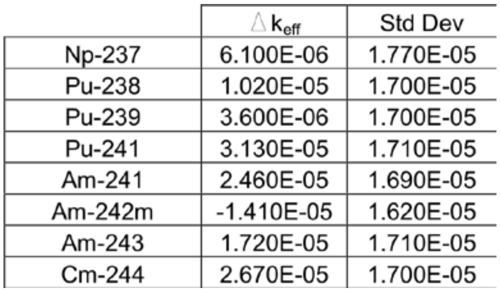
As part of the Advanced Fuel Cycle Initiative (AFCI), research using advanced fast reactors in combination with accelerator-driven systems for the transmutation of waste has shown the ability to reduce the amount of plutonium and transuranic materials for disposal. The Doppler coefficients of each isotope are needed so researchers can conduct safety assessments of transmutation systems for fuel with a significant content of minor actinides.

Research Progress

In FY 2005, researchers obtained information on the critical assemblies, types of experiments possible, and the MCNP models for the FLATTOP and the ZEUS critical experiment using the COMET assembly.

They modified the KCODE output from MCNP5 to print out k_{eff} and the associated statistical uncertainty data so that differences in the k_{eff} values on the order of 10^{-5} could be determined. These additional significant digits are needed to look for reactivity changes on the order hoped for. Upon completion of the code modification, they set up a dedicated computer cluster. The team also created cross-section files for MCNP at 800°C, 900°C, 1,000°C, and 1,100°C from the ENDF, JEFF, and JENDL tapes for each of the isotopes of concern, using the NJOY computer code

During this timeframe, researchers also completed initial calculations using 10 g samples of each isotope in the FLATTOP MCNP model. They created 480 input files for the FLATTOP calculations using the masses determined above, at temperatures of 800°C, 900°C, 1,000°C, and 1,100°C and for the three cross-section libraries. Nine of these 480 cases have been run. Each calculation requires 1,500 KCODE cycles with 6×10^6 neutrons per cycle, taking approximately three and a half days per calculation. Results of these calculations were used to determine five different masses for each isotope which will be investigated. For Np-237 and Pu-239, the masses ranged from 6 g to 22 g; for the other six isotopes, the masses ranged from 2 g to 18 g. Table 1 shows the Δk_{eff} for a temperature change from 800 to 1,000°C for each isotope from these calculations.



	Δk_{eff}	Std Dev
Np-237	6.100E-06	1.770E-05
Pu-238	1.020E-05	1.700E-05
Pu-239	3.600E-06	1.700E-05
Pu-241	3.130E-05	1.710E-05
Am-241	2.460E-05	1.690E-05
Am-242m	-1.410E-05	1.620E-05
Am-243	1.720E-05	1.710E-05
Cm-244	2.670E-05	1.700E-05

Table 1. Calculated change in k_{eff} resulting from temperature change from 800 to 1,000°C for 10 g samples of various actinides.

Planned Activities

Using the current computer cluster, the total time to run these 480 calculations would take approximately four and a half years. After discussing options to shorten the calculation time, the decision was made to use one of the 1,000+ processor machines at LANL to speed up the calculational program. While the access to the LANL computer is being finalized, additional computing clusters with the nuclear engineering program at Georgia Tech will be used when time is available. Following the 480 calculations for the FLATTOP assembly, scoping calculations for the COMET assembly will be made to determine the five different masses for each isotope. Once these masses have been determined, the 480 calculations for the second assembly will be made.

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: April 2005

Project End Date: April 2007

Research Objectives

The objective of this research is to validate and improve computational models for coating uranium fuel pellets with carbon and silicon carbide using the chemical vapor deposition (CVD) process in a spouting bed. Researchers will conduct a state-of-the-art computational study of the CVD process in order to further develop computational fluid dynamics (CFD) models of the reaction kinetics. This project will take a complementary approach using CFD 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. The specific tasks to achieve the objectives are to:

- 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 Multiphase Flow with Interphase eXchanges (MFIX) computer code
- Implement a polydispersity model in MFIX to account for effects of particle size distribution

Research Progress

Researchers have begun developing reaction kinetics model. The process by which the nuclear fuel pellets are coated is through the direct deposition of silicone carbide (SiC) from the gas phase of the pyrolytic decomposition of methyltrichlorosilane (MTS). They have completed the investigation of the potential energy surface (PES) for the unimolecular decomposition pathways of MTS using the GAMESS package.

Figure 1 shows the eight decomposition reactions that MTS may undergo in the gas phase, which can be classified into two categories:

1) The three-bond cleavage reactions displayed on the bottom right of the PES graph where MTS is broken into two radical fragments by breaking C-Si, C-H, or Si-Cl bonds

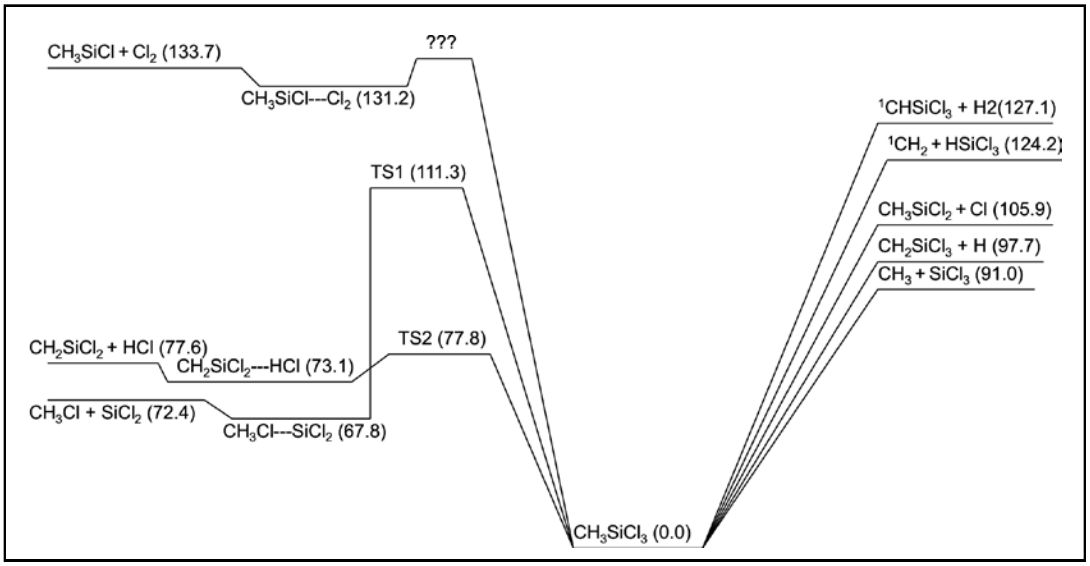


Figure 1. Potential energy surface (PES) of the decomposition pathways of MTS. The structures connected by dashed lines are molecular complexes. The numbers in parentheses are MP2/Aug-cc-pVDZ free energies at 0K, in units of kcal/mol.

2) The elimination/insertion and isomerization reactions where more significant structure rearrangement is involved

The two carbene (CH_2) elimination reactions on the top-right and the three reactions involving $SiCl_2$, CH_3SiCl, or HCl elimination on the left-hand side of the PES graph (Figure 1) belong to the second category. Usually the reactions of the first category have a much larger Arrhenius pre-exponential A factor than the reactions of the second category.

Numerous consecutive reactions may take place in the gas phase during and after the decomposition of MTS at various high temperatures from 1,000 to 2,000K, which is the typical temperature range for the chemical vapor deposition of SiC. Researchers have proposed a homogeneous chemistry mechanism consisting of 85 gas phase reactions (subject to change after further investigation). Twenty-six of these reactions have been found to have no well-defined transition state structures. The energetic and structural features of the transition states have been determined for 28 reactions at the MP2 level. However, the pathways for the remaining 31 reactions have not yet been studied. Application of transition state theory to a couple of reactions, such as the $CH_3 + H_2 \rightarrow CH_4 + H$ reaction, ensure that the MP2 calculations provide reasonable energy barriers and partition functions for the rate constant calculations.

As a starting point for the CFD modeling of complex chemistry, researchers compiled a list of homogeneous gas and heterogeneous reactions (along with reaction rates) involved in the thermal decomposition of MTS. The information on chemical kinetics will be incorporated into MFIX to model the combined effects of hydrodynamics, heat and mass transfer, and detailed chemistry involved in the CVD process. Prior to incorporating the equations (about 24 homogeneous reactions and 12 heterogeneous reactions), it is important to understand the effects of these kinetics on simplified models of mixing and to estimate the lower bounds on the computational costs involved in a full simulation. For this purpose, researchers have chosen the pair-wise mixing stirred reactor (PMSR) model for the simplified mixing model.

In the PMSR model, the particles are first arranged in pairs. As in the case for MFIX with chemical reactions, a fractional time-stepping algorithm is used to solve the species transport equations where the chemical source term is isolated from the mixing term. The mixing fractional time step consists of a pair evolving according to a first-order linear model, while the reaction fractional time step consists of solving a set of ordinary differential equations. Because of the wide range of chemical time scales involved in the rate expressions, the solution of the stiff system of ordinary differential equations (ODEs) during the reaction fractional time step is generally computationally intensive with direct integration methods. To speed up the solution of the stiff system of ODEs, the in situ adaptive tabulation (ISAT) algorithm, which is based on multi-linear interpolation, will be used. These PMSR models for decomposition of MTS are currently being implemented in a FORTRAN code, used in conjunction with CHEMKIN software.

Planned Activities

The remaining activities for the first phase include extending the SIMOMM embedded cluster method to model the carbon surface and its effects on the detailed mechanisms. A simple reaction kinetics scheme for the CVD process will be implemented into MFIX using ISAT and will be validated based on published literature. In addition, a surface growth model will be implemented into MFIX using the direct quadrature method of moments (DQMOM).

The second phase will continue the reaction kinetics modeling by investigating the mechanisms for H_2 interactions with MTS and begin modeling using the activation energies obtained from the electronic structure calculations. The work with MFIX will progress by implementing the reaction kinetics scheme as determined from the computational chemistry study. The model will be validated to determine any shortcomings and need for further improvements.

The final phase will initiate calculations of surface effects on the predicted rate constants. The MFIX code will be used to address scale-up issues for the process-size coater and study the hydrodynamics and particle mixing patterns.

NUCLEAR ENERGY RESEARCH INITIATIVE

On-Line Fuel Failure Monitor for Fuel Testing and Monitoring of Gas-Cooled Very High Temperature Reactors

PIs: Ayman I. Hawari, Mohamed Bourham, North Carolina State University

Collaborators: None

Project Number: 05-054

Project Start Date: January 2005

Project End Date: December 2007

Project Description

The primary objective of this project is to devise an accurate approach for detecting failures of TRISO (Tristructural Isotropic) 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 to develop techniques for detecting 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. Researchers will study various detection methods to differentiate the minute fission product signal from the background. As part of the VHTR fuel development program, they will conduct fuel failure experiments at the Advanced Test Reactor (ATR) at the Idaho National Laboratory (INL). Methods and instrumentation developed by this project are expected to be applicable to on-line fuel failure monitoring of VHTRs.

Research Progress

During the first year, this project focused on exploring the use of passive gamma-ray spectrometry techniques to detect and monitor the failure of TRISO-type fuel and to investigate its use in benchmarking the mechanistic failure models that are currently used to describe fuel failure.

As a first step, researchers focused on understanding the nature of the signal that can be generated upon the failure of a TRISO microsphere. From a gamma-ray spectrometry perspective, the consequence of TRISO failure will be the enhanced release of fission products into the gas flow of either a fuel testing experiment or a VHTR. Consequently, they estimated the concentrations of the relevant fission products gases (mainly krypton and xenon) as functions of irradiation time (i.e., burnup), that can be released upon failure. The calculations utilized the MCNP-MONTEBURNS-ORIGEN code system. Currently, researchers are assessing the accuracy of the produced concentrations (i.e., activities) to understand the impact of the lack of some of the nuclear data in the krypton and xenon production chains on the results.

Researchers will use gamma-ray spectrometry to understand the relationship between release models and the detected gamma-ray spectrum. Several methods are being examined that are based on different combinations of detection instrumentation and spectral analysis methods. Specific to this work, room temperature gamma-ray detectors such as high pressure xenon (HPXe) are being considered for performing the spectrometry. Figure 1 shows the HPXe detector and the ionization system that are currently under test. To enhance the success of these detectors, researchers are investigating the use of a system to extract the gas from the main flow stream and to ionize it and magnetically steer it to the detector. In this fashion, the relevant Kr and Xe signals can be detected in a minimum noise environment.

Researchers also tested the spectrometric performance of the HPXe detector. These detectors have been reported to suffer from resolution degradation due to microfonic effects. However, no such degradation was evident during

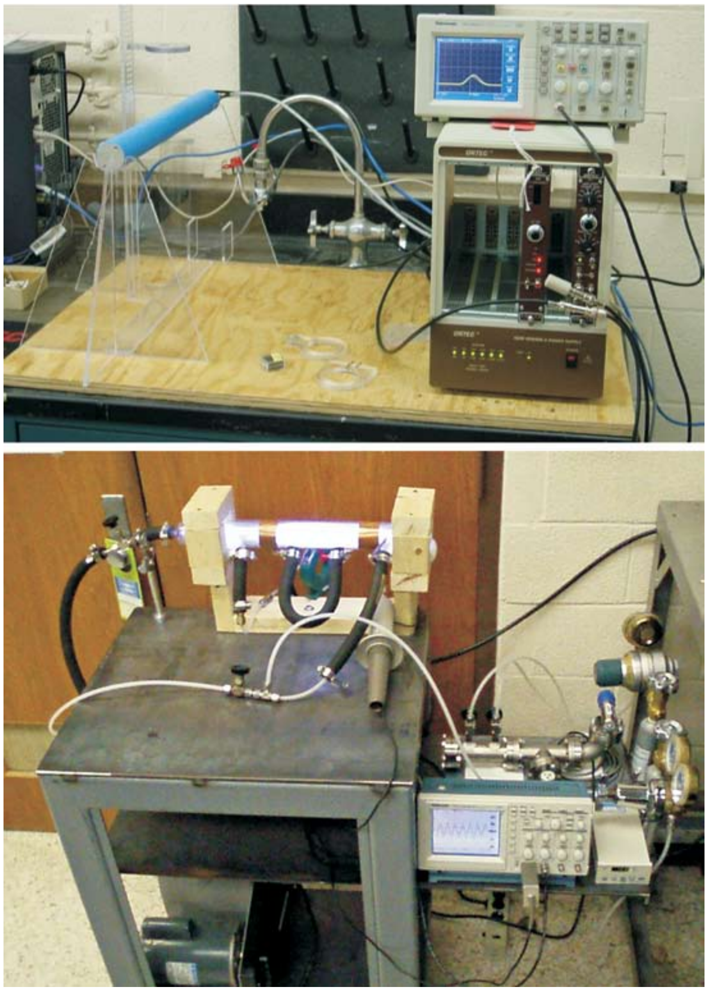


Figure 1. The experimental system, including the HPXe detector (top) and the gas ionization system (bottom).

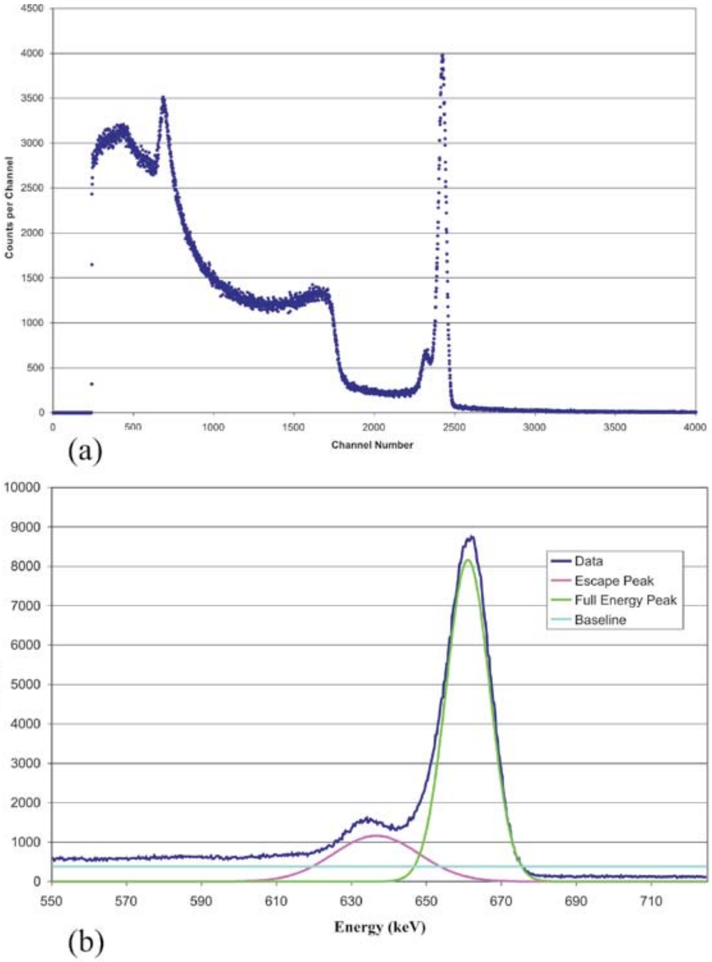


Figure 2. The measured HPXe gamma-ray spectrum of Cs-137 (a) and the details of the 662 keV full energy peak (b).

the testing period for the past few months. The measured resolution at the 662 keV energy of Cs-137 was found to be approximately two percent, which is the ideal value reported in the literature. An additional complication in the spectrometric performance of these detectors is the generation of a 28 keV xenon X-ray, which has a high probability of escaping from typical HPXe detectors. This escape peak results in partial deposition of the gamma-ray energy in the detector, which manifests itself in the appearance of a partial energy deposition peak at an energy that is 28 keV below the full energy peak. Figure 2 illustrates a typical gamma-ray spectrum of a Cs-137 source, details of the full energy peak, and the escape peak effect.

Since samples of radioactive Kr and Xe gases are not readily available, researchers are using MCNP simulations to predict the performance of the various detectors in this task. Gamma-ray source terms have been created for Kr and Xe based on the concentration estimated from the ORIGEN (burnup) calculations and added to the MCNP input file. In addition, information on the variation of the energy-dependent resolution of a given detector was also included. Figure 3 shows the predicted spectra for an HPXe detector for both Kr and Xe.

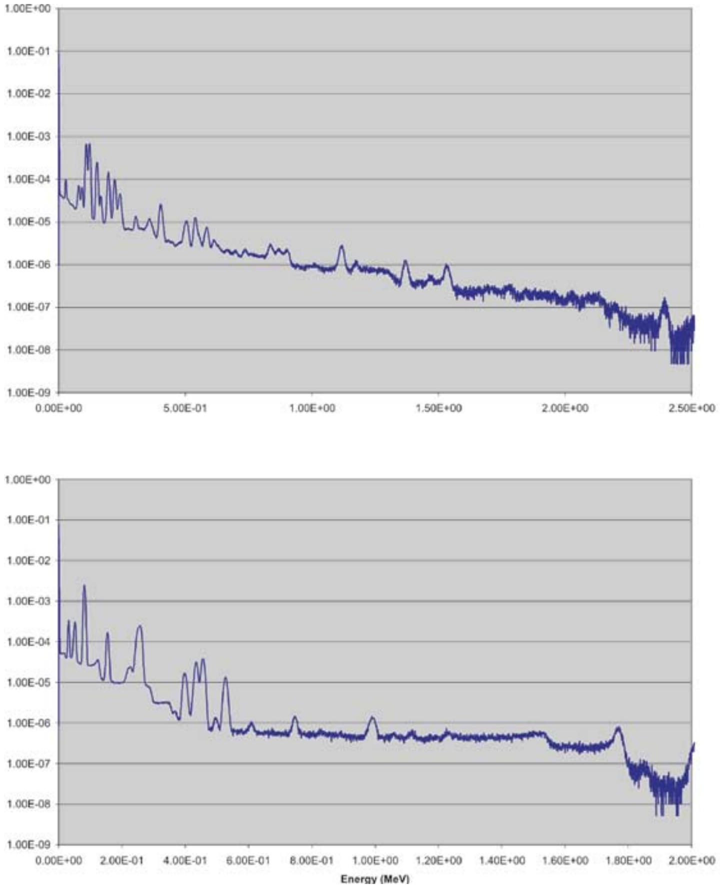


Figure 3. The MCNP simulated spectra for an HPXe detector - Kr (top) and Xe (bottom).

Planned Activities

Plans are currently underway to study additional detectors for possible utilization in this work. This includes high-resolution scintillation detectors that are based on lanthanum halide scintillators. In addition, various spectral analysis techniques will be devised for application with the low-resolution detectors in this application. Performance of the detectors with digital signal analysis systems will also be documented. The design of the gas extraction and steering system is expected to be finalized during the upcoming period. The performance of the system will be tested using non-radioactive Kr and Xe gases. In this case, the ionization and steering of these gases will be detected using optical spectroscopy methods. Furthermore, coordination will continue with INL staff to explore transferring the extraction and detection system to INL for testing at the ATR test loop.

Plutonium Chemistry in the UREX+ Separation Processes

PI: Alena Paulenova, Oregon State University

Collaborators: University of Nevada at Las Vegas and Argonne National Laboratory (ANL)

Project Number: 05-062

Project Start Date: April 2005

Project End Date: March 2008

Research Objectives

The objective of this project is to examine the chemical speciation of plutonium in UREX+ (uranium/tributyl phosphate) extraction processes for advanced fuel technology. 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. 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.

Research Progress

During this reporting period, researchers are completing the distribution and UV-Vis spectrophotometric studies and are focusing on the complexation and redox behavior of Pu(IV) with acetohydroxamic acid (AHA) in aqueous nitrate systems. First, they will perform extractions and UV-Vis spectrometry with U(VI) and several tetravalent cations, including thorium, cerium, and zirconium as chemical analog models for tetravalent actinides.

Researchers prepared a series of solutions with constant concentrations of metal and AHA at different concentrations of nitric acid and total nitrate (by adding lithium nitrate). They are generating UV-Vis spectra of these aqueous solutions for comparison with the distribution ratios of metals between the aqueous phase and TBP-organic phase, as measured by optical emission spectrometry (ICP-OES). The extracted nitrate is analyzed chromatographically and the extracted nitric acid and water potentiometrically.

The hydrolysis of AHA, present in acidic extraction systems, causes the formation of acetic acid and hydroxylamine, according to the following reaction:

$CH_3\text{-CONHOH} + H_2O + H^+ \rightarrow CH_3\text{-COOH} + NH_3OH^+$ [1]

Acetic acid formed in aqueous phase can be determined by anion chromatography (Figure 1); however, researchers used the much faster UV-Vis analytical method for determining unreacted acetohydroxamic acid via its complex with Fe(III).

The effect of acetohydroxamic acid on extraction of U, Th and Zr in 30% TBP is displayed in Figure 2. A small increase of distribution of $[UO_2]^{2+}$ and Th^{4+} in the presence of AHA is caused by extraction of AHA and its complexes to TBP. Extraction of AHA in the absence of metal was confirmed by UV-Vis. Back extraction of uranium, previously extracted to TBP phase from nitric acetic solutions, with aqueous AHA solution exhibited a strong decrease of distribution ratios of U (Figure 3).

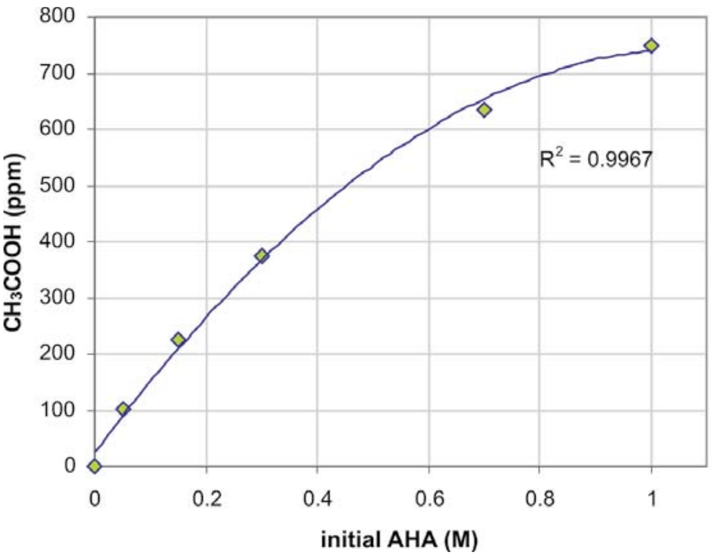


Figure 1. Content of acetic acid in aqueous phase after 3-hour agitation of solutions of AHA in 1.23M HNO_3 (back-extraction of uranium from 30 percent TBP in n-dodecane).

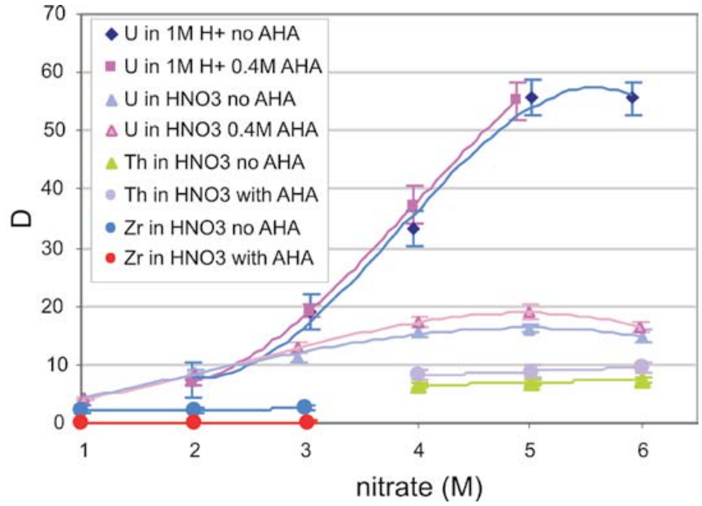


Figure 2. The effect of nitrate and AHA on distribution ratio of uranium, thorium and zirconium in 30%TBP/n-dodecane.

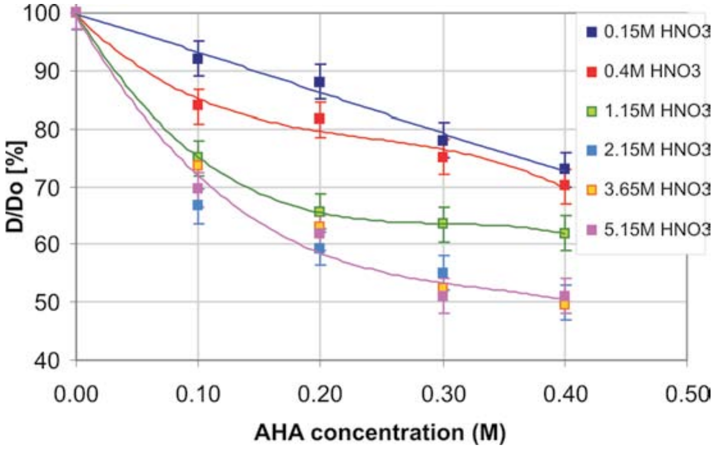


Figure 3. Back-extraction of uranium, previously extracted to 30 percent TBP in n-dodecane from various nitric acid solutions, as a function of AHA concentration in aqueous phase. Concentration of uranium = 0.0225M, D= U(org)/U(aq); D_0 = D at zero concentration of AHA.

Figure 4. Distribution of uranium between aqueous phase (orange-red complex of uranyl-hydraxamate, bottom phase) and organic phase (uranium nitrate in 30 percent TBP in n-dodecane, upper phase).

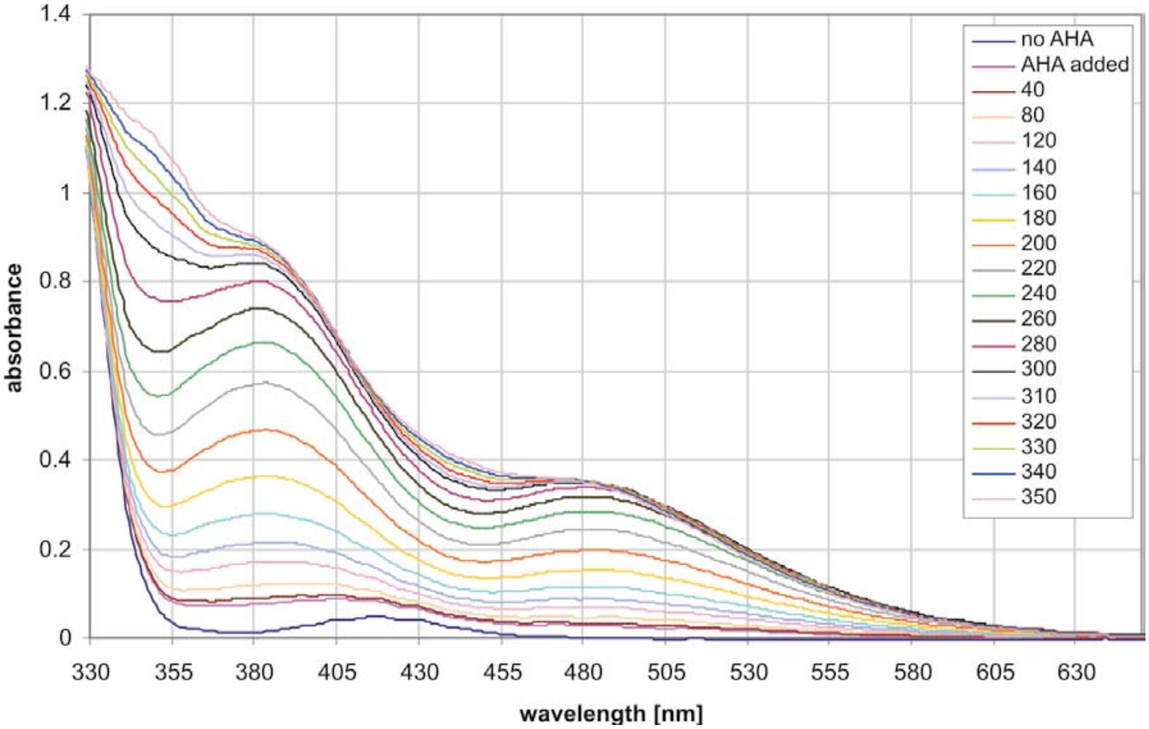


Figure 5. Spectrophotometrically monitored titration of acetohydroxamic acid (initial concentration 1.98×10^{-2} M) in the presence of 5×10^{-3} M U(VI) with 0.1 M KOH (a progressively increasing volume (μl) from the bottom up in the plot).

The researchers also initialized the UV-Vis study of the kinetics of acidic hydrolysis of acetohydroxamic acid. To evaluate the role of nitrous acid, they are performing kinetics measurements in both nitric and perchloric acid media, and the rate of hydrolysis is calculated as a function of H^+ concentration. The reduction of Fe (III) with hydroxylamine formed in the analytical reaction is much slower than complexation Fe (III) with AHA; therefore, with practical experience, reliable data on unreacted acetohydroxamic acid can be obtained.

Planned Activities

In the next period, researchers will:

- Perform the extraction and titration procedures with tetravalent plutonium, as well as plutonium in other oxidation states. For comparison, they will also include other actinides (Np, U, Am).

- Measure distribution of actinides between aqueous and organic phase under various conditions, such as different concentrations of nitric acid, total nitrates, or actinide ions (Figure 4).
- Determine plutonium and uranium speciation in all separation stages using techniques such as Raman and X-ray absorbance spectroscopy and small-angle neutron scattering.
- Utilize potentiometric and spectrophotometric (UV-Vis-NIR) titration and electrospray-mass spectroscopy to analyze these complex systems (Figure 5).

The research team has prepared three technical papers reporting their first results, for presentation at the Spring 2006 ACS Meeting in Atlanta (Nuclear Chemistry and Technology Division Symposium).

1. B. Matteson, J. Bruso, P. Tkac, A. Paulenova, "Speciation of Hexavalent U and Tetravalent Pu in UREX+ Extraction Systems," abstract submitted to Waste Management Conference, Tucson, AZ, 2006.
2. J. Bruso, P. Tkac, B. Matteson, A. Paulenova, "Reduction and Complexation Kinetics of M(IV) in the Presence of Acetohydroxamic Acid," abstract submitted to the ACS Annual Meeting, Atlanta, GA, March 2006.
3. B. Matteson, J. Bruso, P. Tkac, A. Paulenova, " Extraction Systems Effects of Nitrate on Extraction of Tetravalent Metals in UREX+," abstract submitted to the ACS Annual Meeting, Atlanta, GA, March 2006.

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

Collaborator: Argonne National Laboratory

Project Number: 05-066

Project Start Date: January 2005

Project End Date: December 2007

Research Objectives

The U.S. Department of Energy's Advanced Fuel Cycle Initiative (AFCI) is developing next generation processing methods for the recycling of transuranic (TRU) isotopes from spent nuclear fuel. These methods are expected to be phased into existence as advanced nuclear energy systems are implemented. Spent fuel recycling technologies will be established with the goal of closing the nuclear fuel cycle by the mid-21st century. This must include establishing reprocessing facilities and transmutation strategies within the United States. One potential scenario for nuclear energy implementation would involve the initial deployment of an advanced fast reactor for TRU transmutation within the next 50 years. While this transmutation strategy is being established, there is a need for interim storage of separated TRU isotopes.

The primary objective of this NERI project is to develop options for the proliferation-resistant storage of TRU oxides which will be isolated from spent fuel using aqueous uranium extraction (UREX+) processes. The UREX+ family of processes use sequential solvent extraction steps to isolate the large volume of spent uranium, selected high activity, or long half life fission products (e.g., cesium, strontium, and technetium), and the TRU elements (Figure 1). The TRU isotopes are a significant fission energy source, with ultra-long half lives and cross sections that make them fit for burnup in a fast neutron spectrum. The required TRU storage "lifetime" is estimated to be 50 to 60 years before a fast burner is available, so the design requirement of this project is set at 500 years of storage.

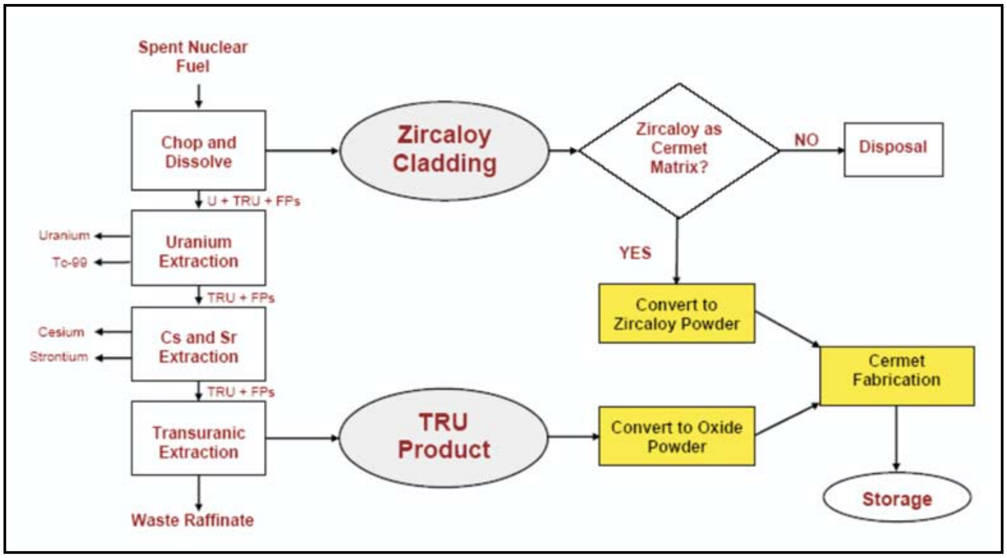


Figure 1. Flowsheet and logic diagram illustrating the relationship between the UREX+ process and the cermet TRU storage form.

The candidate storage forms include simple powder storage with engineered barriers and a manufactured metal matrix composite, or cermet, that will facilitate easy TRU burning after intermediate storage. The goals of the project are to:

- Develop the processing steps required to take the TRU products from aqueous nitrate solution to the final storage form
- Evaluate the impact of phenomena that govern durability of the storage form, material processing, and TRU utilization in fast reactor fuel

The researchers will develop processing logic and methods for converting actinide and lanthanide nitrate solutions into oxide powders, recycling irradiated zircaloy cladding to be used as the cermet matrix, and fabricating a zircaloy matrix cermet storage form. They will also use computational methods to evaluate the TRU burning performance of the cermet pin in a simulated fast reactor.

Research Progress

Figure 1 shows a high-level flowsheet for the UREX+ process combined with the TRU storage process logic of this project. In an early version, a portion of the rare earth fission product inventory was to be incorporated into the TRU nitrate stream as the result of their similar chemical behavior during separation. However, if the cermet storage pin is to be used as targets in a fast spectrum burner, the rare earth concentration will need to be kept very low to minimize neutron absorption and chemical degradation of the zircaloy matrix. Even so, the similarity of chemical behavior between these fission products and TRU makes lanthanide elements an imperfect, but natural, surrogate for actinide simulations.

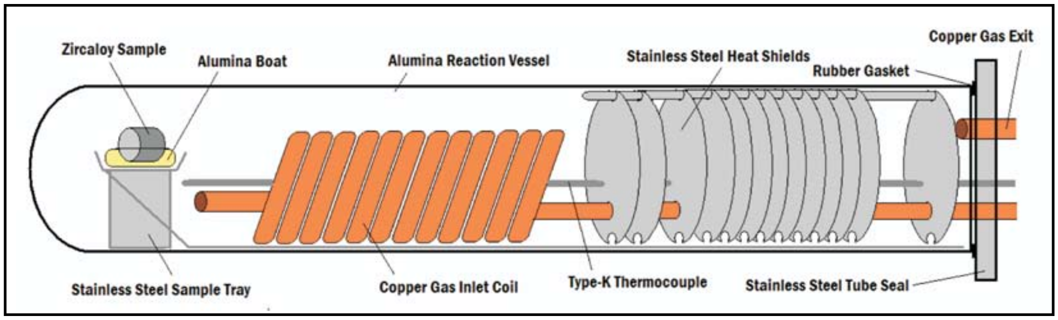
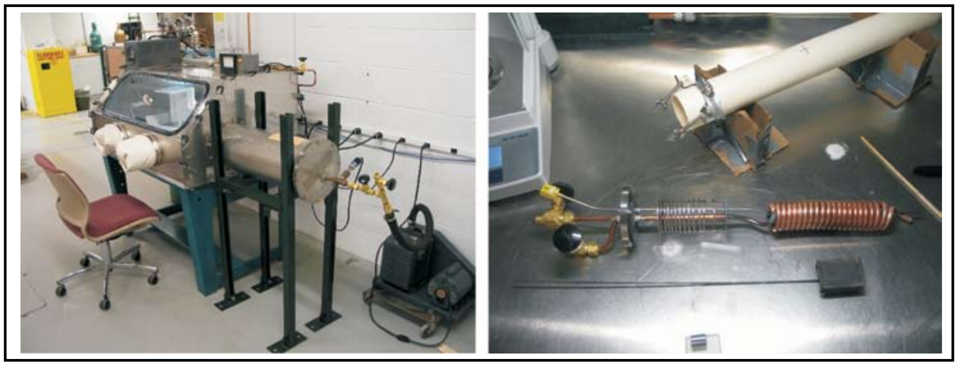


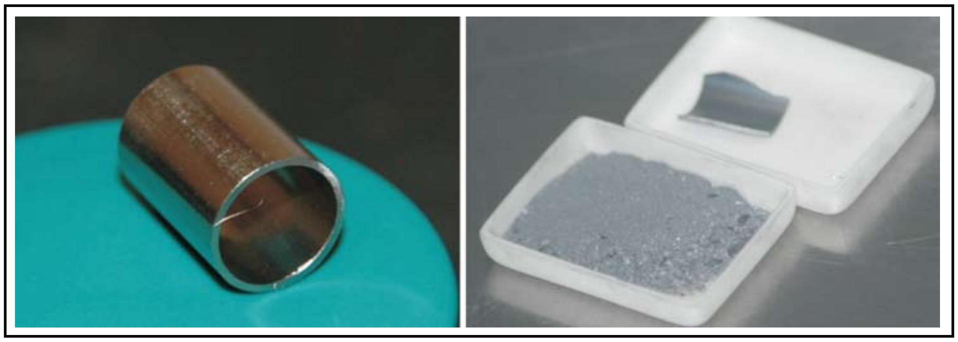
Figure 2. Schematic diagram of the hydriding experiment vessel.

The TRU cermet fabrication will involve three processes: 1) conversion of TRU to oxide powder or microspheres, 2) recovery of zircaloy cladding to form the cermet matrix, and 3) cermet fabrication. Researchers are evaluating TRU oxidation methods using direct denitration and sol-gel formation of oxide microspheres, based on the experience developed at Oak Ridge National Laboratory. They are designing experiments using uranium and rare earth isotopes to elucidate the basic phenomena behind co-precipitation and mixed oxide processing.



(a) Inert Atmosphere Glovebox (b) Hydride Equipment

Figure 3. Photograph of the (a) inert atmosphere glovebox and (b) hydride equipment designed and built for the zircaloy hydride process development.



(a) Zircaloy IV Tube (b) Zircaloy IV Hydride Powder

Figure 4. Photograph of (a) Zircaloy IV tube sample and (b) Zircaloy IV powder from a hydrided and crushed sample.

The zircaloy recovery process selected for development involves a hydride/dehydride operation with mechanical milling. The design is to divert a portion of the zircaloy waste that will become available when spent nuclear fuel is recycled. This material is attractive for the proposed cermet because zircaloy is already nuclear grade (i.e., the natural hafnium content has been removed). However, its use poses a challenge since the zircaloy will be oxidized by corrosion and fuel-cladding chemical interaction and it is modestly radioactive.

The hydride process is being developed to overcome and accommodate these challenges. The ultimate goal is to demonstrate a single equipment design to transform oxidized zircaloy cladding into metallic powder through hydride formation, mechanical milling, and dehydriding. The oxidized zirconium will either be sieved out of the powder or incorporated into the cermet matrix. Figure 2 shows a schematic diagram of the hydriding experiment vessel designed for this project. The vessel consists of a closed end ceramic tube, sample holder, gas supply coil, and thermal heat shields.

Figure 3 shows photos of the inert atmosphere glovebox set up for handling hydride powders and the disassembled process equipment inside of the glovebox. Figure 4 shows a representative sample from the starting Zircaloy IV tube stock along with an early photo of a hydrided zircaloy tube after grinding with a mortar and pestle. The initial process development results indicate that temperatures between 500°C and 600°C are required to embrittle the zircaloy under flowing Ar-5%H_2. Below 500°C, reduced solid state diffusion rates hindered the reaction kinetics. Above 600°C, β-zirconium is stabilized and has a high solubility for hydrogen; samples treated above 600°C maintained their shape and were too tough for grinding.

The final cermet fabrication process combines the products from the first two processes to fabricate a cermet pin using hot extrusion or an enhanced cold drawing technique which evolved from a previous NERI project for zirconium cermet pins. There is a need to have open porosity in the storage form matrix in order to accommodate the buildup of helium gas generated by alpha-decay. Initial calculations indicate that the helium would produce a large internal pressure over the 50 to 500 years of storage. Post storage extrusion may be used to densify the cermet matrix before inserting the TRU target into the fast reactor.

Planned Activities

In the second year of the project, researchers will continue to develop the process and conduct further computational simulations. Following is a summary of planned activities:

Process Development

- Continue the Zircaloy Recycle experiments to optimize the process variables and to discern the fundamental process mechanisms
- Design an all-in-one Zircaloy Recycle system to hydride, pulverize, and dehydride spent cladding into Zircaloy powder
- Continue cermet fabrication experiments by hot extrusion and cold drawing
- Begin co-precipitation and sol-gel experiments focused on understanding TRU denitration and oxidation using uranium and rare earth surrogates

Computational Simulation

- Complete criticality modeling of the TRU storage form options
- Complete models for heat generation and helium buildup (from α-decay) within the storage form
- Establish a system of simulations to evaluate the TRU burnup effectiveness in cermet and MOX fuels in a fast reactor

References

1. S.M. McDeavitt, et al., "Cermet Fuel Development for Advanced Nuclear Systems – (Th,U)O_2 in a Metal Matrix," Purdue University Technical Report PU/NE 05-04-FCML/1 (2005).
2. S.M. McDeavitt, T.J. Downar, and A.A. Solomon, "Cermet Fuels for Advanced Fuel Cycles and Transmutation," International Congress on Advances in Nuclear Power Plants (ICAPP '05), Seoul, South Korea, May 15-19, Paper No. 5602 (2005).

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: January 2005

Project End Date: December 2007

Research Objectives

This project investigates oxidation state adjustment (and control) methods to achieve selective partitioning of americium (Am) from lanthanides and curium (Cm) using conventional aqueous separations methods and materials. Increased application of MOX fuels and longer burnup times for conventional fuels will produce greater amounts of the transplutonium actinides americium and curium in spent nuclear fuel. Because the half-lives of some Am isotopes (^{241}Am - 433 yrs; ^{243}Am - 7,370 years) are significantly longer than those of the most important long-lived lanthanides or the most common isotopes of curium, berkelium and californium, the isolation of Am from other actinides and lanthanides represents an opportunity to minimize the volume of material requiring repository emplacement. Selective separation of Am isotopes also represents a rational approach to integrating transmutation into advanced nuclear fuel cycles as this process could significantly reduce the long-term radiotoxicity of the residues from spent fuel or from spent fuel reprocessing.

The chemistries of trivalent lanthanides and actinides under aqueous processing conditions are so similar that selective separation of Am is extremely challenging. Trivalent ion-recognition techniques are unlikely to provide an adequate thermodynamic basis for selective isolation of Am in an acceptable number of contacts. Kinetic features of the complexation reactions provide only slight hope for developing a successful selective separation of Am. Because the predominant Cm and lanthanide isotopes are short-lived, they are more amenable to near-surface disposal options.

Oxidized americium species are not expected to be inherently stable, but transient stability may be adequate for purposes of isolating Am from Cm and the lanthanides. Therefore, a key aspect of this investigation will be to identify the comparative redox stability of oxidized Am species under process-relevant conditions. Because it is important to minimize contact times with oxidizable macro constituents of the separation system, kinetic features (of redox, complexation, and phase transfer reactions) cannot be ignored.

Research Progress

The researchers have arranged for the delivery of the actinide isotopes necessary to conduct this research. Unexpected regulatory requirements delaying their receipt have been resolved. Adjustment of operational conditions was required and researchers have made improvements to the infrastructure, which are now nearly complete. Other available sources of radiotracer actinides that will prove useful in the longer term have been identified at other national labs, including a sample of ^{248}Cm.

Early lab work emphasized polyoxometallate synthesis and characterization and conventional solvent extraction from ammonium thiocyanate media. Researchers have pursued each of these avenues as necessary preambles to the stabilization of Am in unconventional oxidation states. They have conducted some initial studies of the effect of nonionic surfactants on actinide chemistry and are re-examining the latest literature on ionic liquids to determine the best path forward for this aspect of the program. On this latter issue, they ruled out a simple solvent-substitution approach, as prior work (elsewhere) has established that this approach is far too complex. Nevertheless, ionic liquids could play an important role to facilitate oxidation and stabilization of Am(III).

Planned Activities

During the next quarter, researchers will study the chemistry of actinides in the upper oxidation states in carbonate and bicarbonate solutions. This approach is seen as a means of increasing the lifetime of oxidized Am species, specifically Am(V,VI) to that regime suitable for conventional aqueous separations techniques. This line of investigation is unconventional, but could develop a variety of new options for selective separation of Am with variation in redox control parameters.

Several previous reports indicate that alkaline solutions could have a useful impact on creating and retaining oxidized Am species. It is also known that carbonate and alkaline solutions have the net effect of increasing the relative redox stability of oxidized actinides, Am(V, VI) relative to Am(III, IV). The researchers believe that they can significantly advance this activity by studying redox stable species like U(VI) and Np(V), and by using Np(V) and Pu(VI) as a spectrophotometric probes of the pentavalent and hexavalent oxidation states. Therefore, they will examine these chemistries as well.

Researchers have completed arrangements for the delivery of appropriate materials from the Pacific Northwest National Lab for the needed isotopes. Though their radiotracer operations and uranium chemistry operations are ongoing, they are still working with the Radiation Safety Office at WSU to get all of the nuclide handling procedures in place for intermediate scale (multiple milligram) actinide studies. Macro-actinide studies will be prominently featured in the experimental activities at the INL facilities.

The acquisition of an ozonizer will enable the production of oxidized Am species and the clean oxidation of other actinide ions, as needed. There are no specific plans to synthesize extractant molecules for this investigation, relying instead (at least initially) on commercially available extractant molecules. If structurally hindered reagents can provide a useful contribution to the net separation efficiency, researchers will pursue this option.

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: January 2005

Project End Date: December 2007

Research Objectives

Implementation of a closed-loop nuclear fuel cycle requires the use of mixed oxide (MOX) fuels containing plutonium. The use of MOX fuel, combined with longer irradiation times, results in increased production of trans-plutonium 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/or transmutation. Transmutation, whether by thermal reactor, fast spectrum reactor, or accelerator irradiation, is done most efficiently in the absence of lanthanide fission products, since they are neutron poisons. Because the lanthanide content is 100-fold higher in dissolved spent fuel than Am/Cm on a molar basis, it is very important to develop effective procedures for their mutual separation.

The most useful and versatile separations of these species are accomplished using reagents containing donor atoms that are "softer" than oxygen, specifically N, Cl^-, and S, which exhibit less ionic bonding character than oxygen and interact more strongly with actinides than lanthanides. Reagents that have been advanced in recent years (including the bistriazinylpyridine (BTP) extractants in France and thiophosphinic acid extractants in China and Europe) show promise for an efficient separation, but suffer various weaknesses, many relating to their instability in a radiation field or strongly acidic solutions.

This project investigates new chemical methods (or the improvement of existing methods) for selective separation of transplutonium actinide isotopes from fission product lanthanides using proven aqueous processing techniques. Soft-donor extractant systems based on ligands containing multiple soft donor nitrogen atoms, but distinct from the BTP class of extractants, represent one focus of this research program. A second thrust area is TALSPEAK-like chemistry, relying on the application of water-soluble soft-donor ligands like diethylenetriamine-N,N,N′,N″,N″-pentaacetic acid (DTPA) as the basis for the trivalent actinide-lanthanide selectivity. In each case, the researchers' initial focus is on developing the fundamental separation parameters in anticipation of later studies that will focus on coping with the inevitable deleterious effects of reagent degradation. A high priority in this project is mitigating as much as possible the effects of radiolytic and hydrolytic damage to reagents.

Research Progress

Researchers completed arrangements for the delivery of actinide isotopes, which was delayed unexpectedly by airborne release permitting requirements. Early lab work has emphasized the synthesis of a few new soft donor extraction reagents and preliminary work on the thiocyanate system. The polyaza extractant synthesis studies have progressed through an investigation of a combination amine-amide donor system to a multiple amine donor extraction system. Researchers have emphasized extraction from thiocyanate media and the application of multiple extractant (i.e., synergistic) solvents. Other aspects have received some investigation via electrochemistry.

The extractant design project features manipulation of the coordination geometry of the extractant molecules through the introduction of structurally hindered arrangements of donor atoms. It is anticipated that the energetic advantages in bond strength gained through the incorporation of multiple soft donor-atoms and structural alterations can have a "net" significant impact on separation efficiency (that is, energetic differences representing 1-2 hydrogen bonds can create separation factors of 10-100).

Solubility is an issue in some early studies of new extractant molecules, which has often proven to be either too low or too high in the "wrong" phase. Partitioning problems of extractant molecules are of less importance at this stage, as structural or solvent modifications can be developed at a later stage to optimize this feature. Researchers are most concerned with establishing the thermodynamic basis for the fundamental separation. Eventually, features of the kinetics of interactions will be investigated as they can have a significant impact on radiation protection strategies. The polyamine donor systems being investigated have potential either as extractant molecules or as DTPA-substitute molecules in TALSPEAK.

Planned Activities

Research will continue on extraction studies from thiocyanate or mixed-salt media using conventional extractant systems like CMPO or trioctylphosphine oxide (TOPO). The team has recently completed the synthesis of a new polyamine donor ligand system that appears promising in early tests. They have also completed a literature study of TALSPEAK separations systems. Future work will include increasing experimental focus on TALSPEAK-relevant chemical systems and writing a review paper for publication emphasizing opportunities for improvement in these systems.

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: April 2005

Project End Date: April 2008

Research Objectives

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 the understanding of 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.

Research Progress

Researchers on this project team have received bulk samples of 304 and 316L stainless steel, HT-9 alloys, 9Cr-1Mo steel, and EP823 and have processed these materials by equal channel angular extrusion (ECAE). Researchers performed annealing studies on ECAE-processed 316L SS and HT-9 metallic alloys to obtain thermal stability. It is anticipated that as-processed materials will have ultra-fine grain sizes on the order of submicron and annealing will induce grain growth at certain temperatures. A collaborator has written a proposal to request irradiation beam time at the synchrotron facility in Switzerland in order to perform irradiation tests on ECAE-processed HT-9 and 316L SS alloys with different grain sizes. These alloys will also be subjected to corrosion tests.

The following describes ECAE processing details of several alloys for this study:

ECAE processed alloys. Researchers processed 304, 316L, EP-823, HT-9, and 9Cr-1Mo steel samples at high temperature (600-700°C) and medium temperature (300°C). The starting materials are relatively strong and difficult to process at room temperature. The as-received samples are 0.25 inch in diameter and were cut into segments of around 1 inch length via electric discharge machining. The samples were placed in drilled holes and covered with stainless steel caps, then encapsulated in nickel cans machined in the ECAE shop. Their arrangement within the encapsulation can is shown in Figure 1. Processing conditions (temperature and routes) are listed in Table 1.

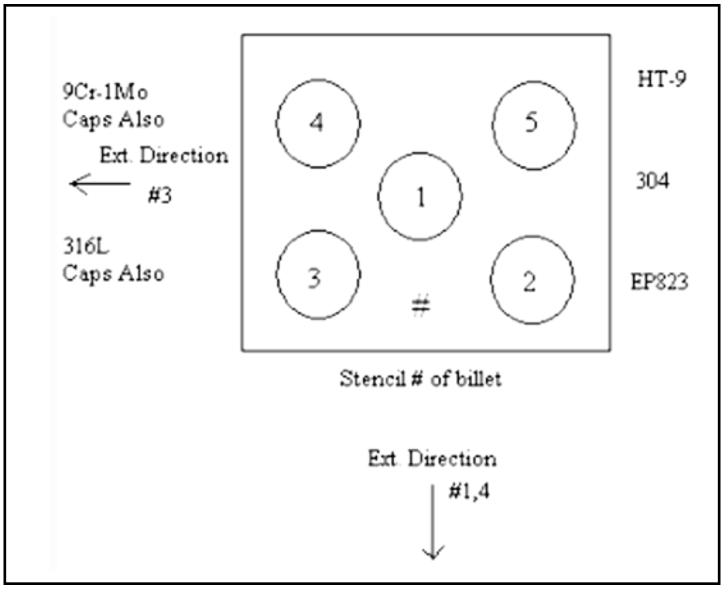


Figure 1. Top view of the Ni encapsulation can used to contain the materials for ECAE processing. Five pieces of alloys (0.25 inch diameter and around 1 inch in length) are embedded in the Ni can, separated by about 0.25 inch.

The samples were removed from the Ni cans and sectioned. Displacement during extrusion was a problem for several specimens, which became apparent in an optical micrograph of ECAE processed specimens, shown in Figure 2. When the samples were all extracted, they were halved using a diamond wafer saw.

The as-processed materials are expected to possess ultra-fine grain sizes, around a few hundred nanometers. Annealing should presumably enlarge the grain size. Detailed studies of microstructure and thermal stability are planned at Los Alamos for 316L stainless steel and HT-9, and at TAMU for the rest of the processed specimens.

Billet	Specimen	Pass/Route	Temperature (°C)
1	304 Stainless steel	1B	300
	304 Stainless steel	2B	300
2	EP823	not processed	
3	316L stainless steel	1A	700
4	9Cr-1Mo	1B	700
	9Cr-1Mo	2B	600

Table 1. Summary of billet processing.

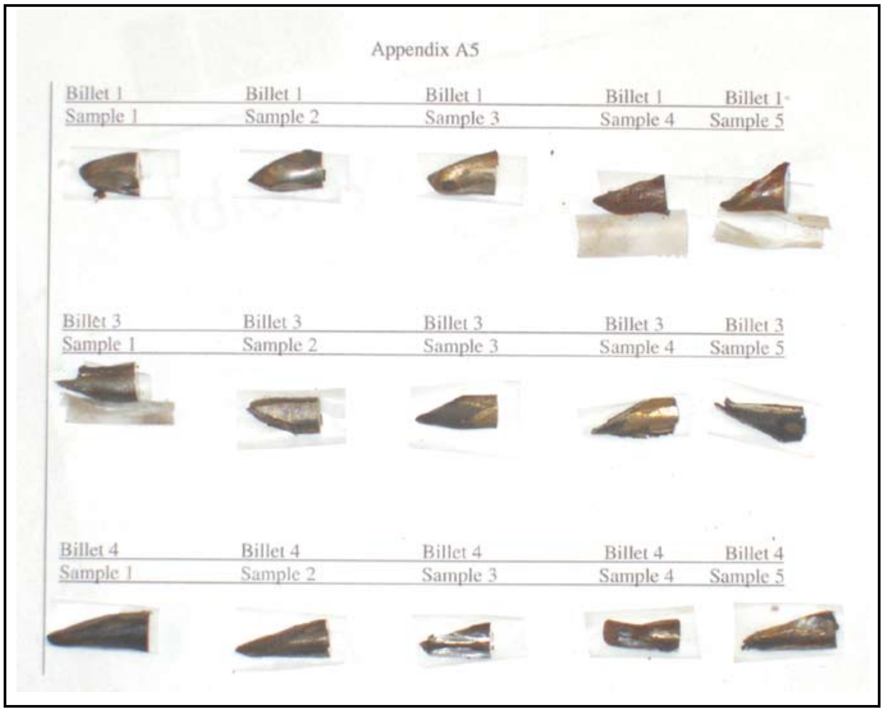


Figure 2. Optical micrograph of specimens processed by ECAE processing. The specimens include 304, 316L stainless steel, HT-9, EP823, and 9Cr-1Mo steels. Specimens with good deformability are typically indicated by a sharp and long taper edge.

Preparation of nanolayered thin films using magnetron sputtering technique. The research team has synthesized iron/tungsten (Fe/W) and aluminum/niobium (Al/Nb) nanolayer thin films using a magnetron sputter-deposition system. These metallic multilayers were sputter deposited at room temperature on silicon substrates. The constituent layers within the multilayers had equal thicknesses, which varied from 1 to 500 nm, and the number of bilayers deposited was such that the total multilayer film thickness was approximately 2 µm. Fe/W was chosen as both constituents have high melting points and could be beneficial for high temperature use in a nuclear reactor. Furthermore, both materials have relatively open crystal, body-centered cubic structure, which could enhance the capability of damage storage induced by radiation. Al/Nb was chosen because Al coatings may be corrosion resistant with the introduction of an oxide layer in lead-bismuth (Pb-Bi) cooled nuclear reactors. Preliminary nanomechanical testing shows that Fe/W multilayers have a maximum hardness exceeding 10 GPa, whereas Al/Nb multilayers possess a maximum hardness of around 5 GPa at a layer thickness of around 2 nm.

Researchers have also recently deposited 330 austenitic stainless steel (330 SS) thin films. Bulk 330 SS is known for improved corrosion resistance with the addition of nickel in the alloy and films have unusually high strength of about 7 GPa, an order of magnitude higher than that of bulk counterpart. The high strength in sputtered 330 SS thin films originates from high density growth twins. Such coatings could have potential application in nuclear reactors. Thermal stability of these coatings is being studied in order to ensure such materials can be used in nuclear reactors at an intermediate temperature range of approximately 500°C.

TEM micrographs of 330 SS films, given in Figure 3 (a), show that high density growth twins remain after annealing at 500°C for one hour. The average twin spacing is less than 10 nm, as revealed by high resolution TEM micrograph in Figure 3 (b). As a result of retention of such fine microstructures, the strength or hardness of films remain very high, around 7-8 GPa.

Such studies indicate that these high-strength austenitic stainless steel coatings can operate at intermediate temperature without the loss of high strength. The study has just been accepted for publication in *Applied Physics Letters* in 2005.

Equipment. A micro/nano hardness tester has been purchased for this project. The equipment has load and depth control capability, a load resolution of around 40 μN, and distance resolution of around 100 pm.

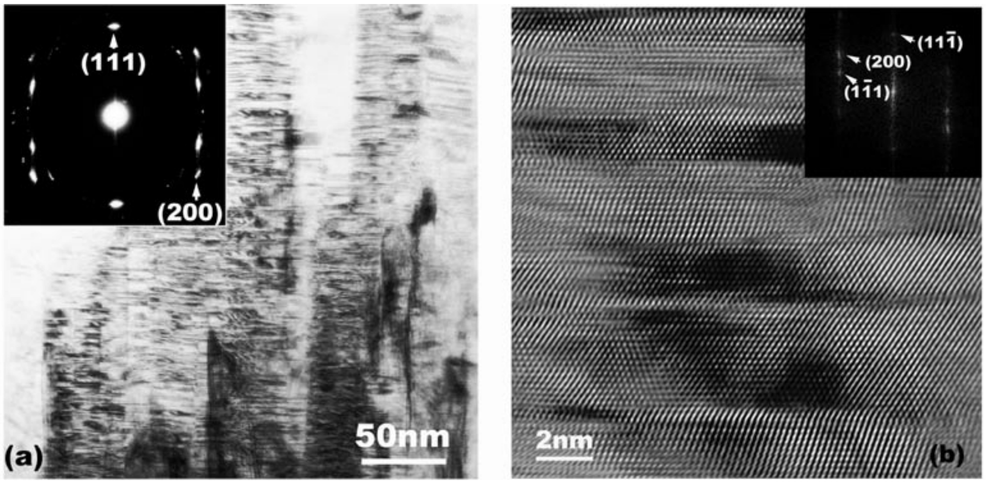


Figure 3. (a) Cross-sectional TEM micrograph of 330 SS films annealed at 500°C for 1h. (b) Cross-sectional HRTEM micrograph and the inserted FFT of the same film show the configuration of twins are almost identical to that of as-deposited films. The average columnar grain size increases from 30 nm for as-deposited films to 40 nm after annealing.

Planned Activities

Experiments are planned to test the mechanical properties of Fe/W and Al/Nb multilayer thin films systematically using nanoindentation. Researchers will examine plan-view and cross-sectional microstructures of these nanolayers with high-resolution transmission electron microscopy (HRTEM). The texture of these nanolayers will also be studied using x-ray diffraction equipment. Ion irradiation tests will be performed using an ion implantation system at LANL. Researchers also plan to examine the microstructure and mechanical properties (hardness and elastic modulus) of ECAE processed 304L SS, EP823, and 9Cr-1Mo alloys using the new micro/nano hardness tester. Such information will provide a basis for comparing the microstructure and properties of materials after irradiation or corrosion testing. Thermal stability of these bulk materials and nanolayers will be studied using differential scanning calorimetry (DSC). Nanolayer interface and grain boundaries in nanocrystalline or ultra-fine grain materials could provide improved radiation tolerance. It is important to determine if these structures will remain at a temperature that is suitable for use in nuclear reactors.

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: March 2008

Research Objectives

The objective of this project is to assess the possibility, advantages, and limitations of achieving ultra-long-life VHTR (Very High Temperature Reactor) configurations by utilizing minor actinides (MAs) as a fuel component. This analysis considers and compares the capabilities of pebble-bed and prismatic core designs with advanced actinide fuels to approach the reactor lifetime-long operation without intermediate refueling. The ultra-long life VHTR systems are developed and analyzed focusing on control, dynamics, safety, and proliferation-resistance during reactor lifetime-long autonomous operation.

To assure comprehensive, realistic assessment of the VHTR design and operation targeting passive safety confirmation, the adequacy of computational methods and models used to compute performance characteristics is supported by comparisons with experimental data covering an appropriate range of conditions. In addition to the experimental benchmark confirmation of the developed models and obtained results, the success of the project will be determined by its ability to demonstrate (or refute) the possibility, advantages, and limitations of achieving ultra-long life VHTR configurations by utilizing MAs as a fuel component.

This work consists of six tasks:

- Development of whole-core/system models with explicit multi-heterogeneity treatments
- Development of benchmark test problems to compare with experimental data
- Validation and verification of the VHTR models
- Analysis of uncertainty effects on VHTR performance characteristics
- Analysis of configuration variation capabilities to achieve ultra-long operation without refueling, maximize burn-up levels, and minimize reactivity swings
- Completion of 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 impact of increasing burn-up levels on the generated radioactivity.

This project investigates VHTRs with 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. Since both core designs, pebble beds and prismatic block stacks, 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 burnup.

The ultra-long core life approach reduces the technical need for additional repositories per decade of reactor operation and should improve marketability of the Generation IV VHTR by allowing worldwide deployment including in developing countries. Utilization of MAs facilitates development of new fuel cycles and supports fuel supply sustainability. Thus, this research comprehensively addresses both intermediate and long-term issues associated with high-level nuclear wastes and should demonstrate the potential of VHTR-based solutions.

Several technical challenges are associated with the analysis and development of ultra-long-life VHTR configurations with minor actinides as a fuel component:

- Whole core/system modeling with multi-heterogeneity treatments
- Model benchmarking, uncertainty effects of nuclear data, and design parameters including temperature dependence
- Error propagation during depletion calculations
- Large computational times affecting ability to consider many design configurations

A special effort is being made to verify that the computational modeling is consistent and realistic. The results will describe performance of the entire VHTR power unit and allow conclusions regarding the configuration's feasibility, performance, and possible directions for further analysis and development.

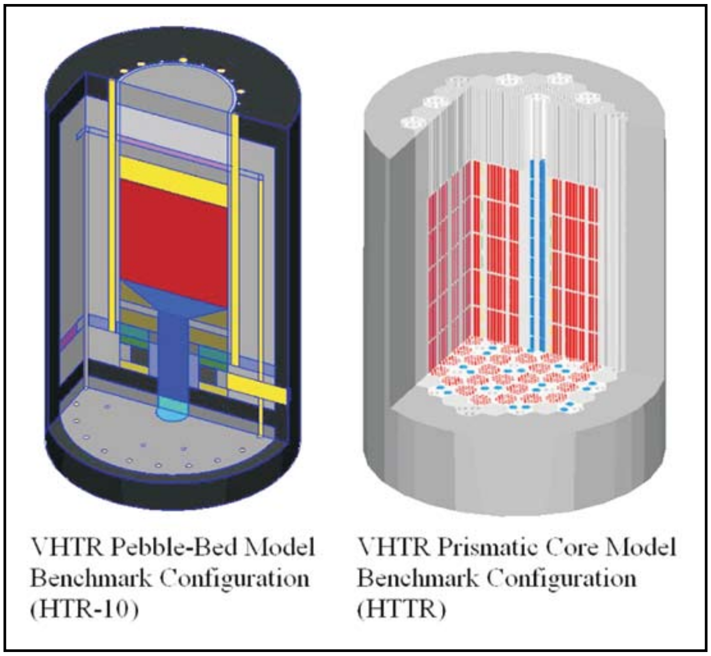


Figure 1. Benchmark configurations for the 3D whole-core VHTR model.

Research Progress

During the first year, researchers completed a literature review to support the analysis of VHTR configurations with advanced actinide fuels. Based on the survey results, they are developing an actinide fuel property database that will be used for realistic VHTR configurations with advanced materials.

To assure comprehensive, realistic assessment of the VHTR design and operation targeting passive safety confirmation, the adequacy of computational methods and models used to compute performance characteristics must be supported by comparisons with experimental data over an appropriate range of conditions. Current project tools include MCNP, SCALE, auxiliary tools to account for double heterogeneity, and the HeatWave suite. These tools form the system for coupled modeling. Validation data are available from power reactors as well as past critical experiments.

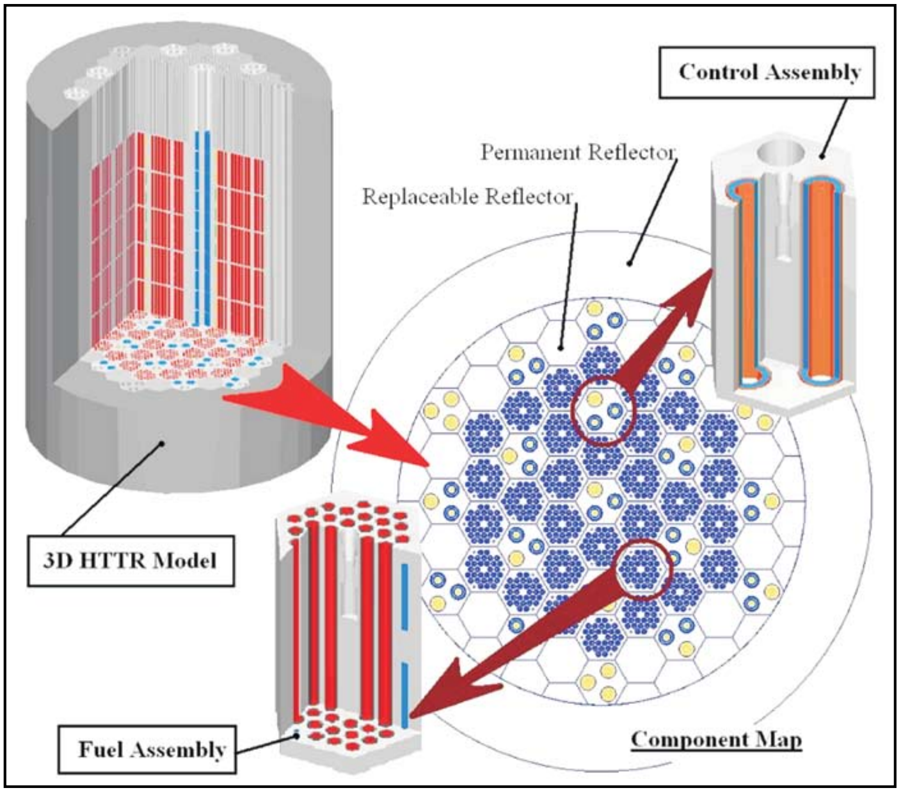


Figure 2. 3D whole-core model of the HTTR configuration (KENO3D visualization results).

The VHTR model benchmark sets, shown in Figure 1, were compiled based on the LEU-HTR PROTEUS (not shown in Figure 1, lattice-level model benchmarking including double-heterogeneity phenomena), HTTR, and HTR-10 experimental programs. Researchers have completed validation and verification of the VHTR models, including code-to-code and code-to-experiment benchmark studies.

The benchmark results obtained are in agreement with the available HTTR and HTR-10 data and confirm applicability of the chosen modeling approach. Figure 2 shows details of one of the developed and validated 3D whole-core models.

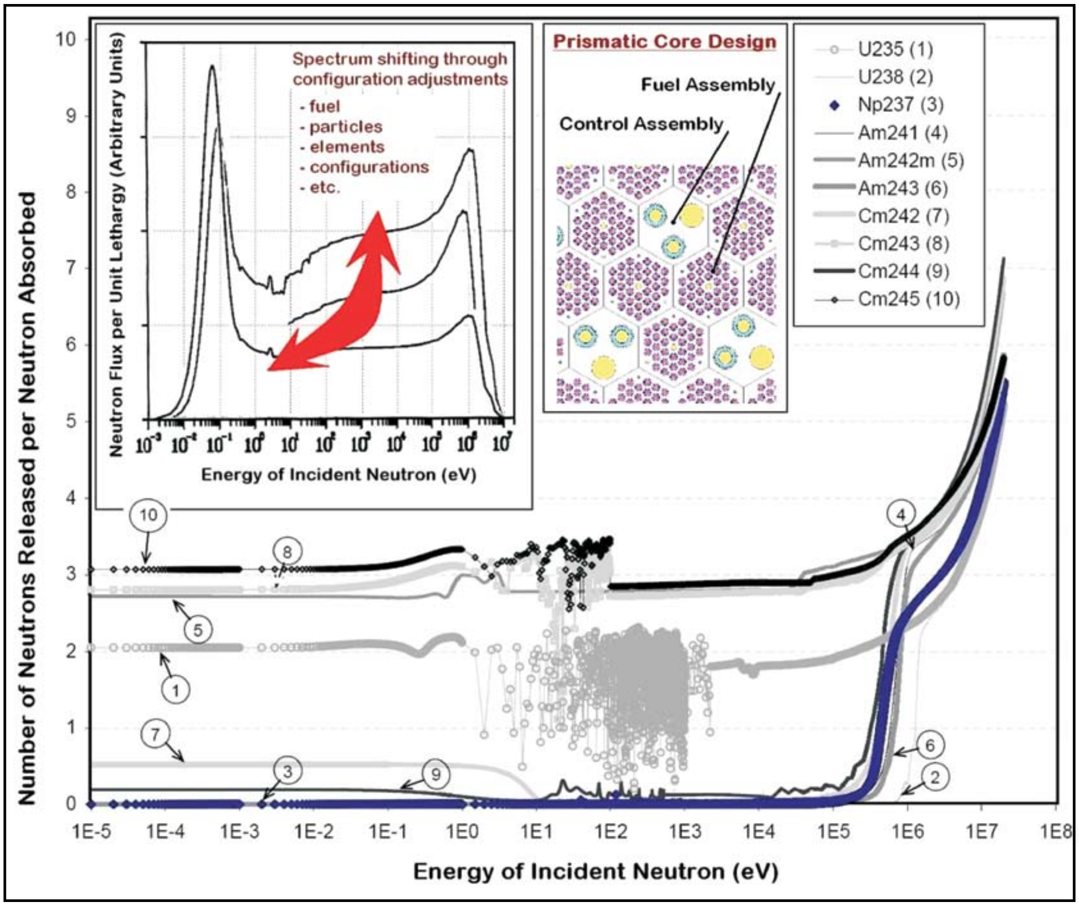


Figure 3. Spectrum shifting capabilities and neutronic performance of transuranic nuclides (SCALE modeling results).

The preliminary series of studies of the VHTRs with MAs are completed. Although further refinement is necessary, these preliminary results indicate promising performance characteristics.

As illustrated in Figure 3, depending on neutron spectra in a particular core configuration, neptunium, americium, and curium may serve as burnable poisons or fuel materials that contribute to small reactivity swings (self-stabilization) over prolonged irradiation periods yielding high levels of burnup. The main advantages of the resulting ultra-long-life VHTR configurations are their inherent capabilities for utilizing minor actinides from spent LWR fuel and for reducing spent fuel flows and handling per unit of produced energy, and their potential for autonomous operation with minimized maintenance.

Current results are in agreement with available data and confirm the chosen approach. Studies of VHTRs with MAs suggest promising performance. The developed VHTR configurations are being analyzed and compared, taking into account the existing systems and the Generation IV VHTR concepts with estimated characteristics. Using 3D whole-core models, researchers developed a variation analysis methodology.

Studies of VHTRs with MAs suggest promising performance. The first year results will be presented at the ANS annual meeting, ICAPP 2006, ICONE14, and PHYSOR 2006.

Planned Activities

During the second year of this project, researchers will complete the analysis of existing uncertainty effects on VHTR performance characteristics. This work will focus on effects due to nuclear data and design parameters. The uncertainty analysis will be followed by the reliability evaluation of the VHTR modeling. The developed and validated 3D whole-core/system models of VHTRs will be used in all studies.

Based on the first-year preliminary studies of the VHTR with MAs, the major thrust in the second year will be to begin the detailed analysis of configuration variation capabilities to achieve ultra-long operation without refueling, maximize burnup, and minimize reactivity swings. Using 3D whole-core/system models, researchers will develop a variation analysis methodology and then will apply it for studies of VHTR geometry and material variations.

In the third, final, year of the project, the following subtasks will be completed fulfilling objectives of the project:

- VHTR optimization to achieve ultra-long life
- Feasibility of ultra-long-life VHTRs with MAs
- Ultra-long-life VHTRs and transmutation
- Dynamics, safety, and control of VHTRs with MAs
- Fuel cycle of ultra-long-life VHTRs with MAs
- Proliferation-resistance of ultra-long-life VHTRs

These detailed studies account for key aspects of the developed VHTRs with minor actinides.

The research work advances as planned. Accomplishment of the project objectives is anticipated within the project timeframe.

NUCLEAR ENERGY RESEARCH INITIATIVE

Ambient Laboratory Coater for Advanced Gas Reactor Fuel Development

PI: Duane D. Bruns, University of Tennessee

Collaborators: Idaho National Laboratory, Oak Ridge National Laboratory, Iowa State University, University of Tennessee

Project Number: 05-118

Project Start Date: January 2005

Project End Date: December 2007

Research Objectives

Spouted fluidized bed coaters are used for applying coatings to tri-isotropic (TRISO) nuclear fuel particles for the advanced high temperature gas reactor (HTGR) and other Generation IV nuclear power reactors. The purpose of this project is to scale up coaters to production size by collecting detailed experimental measurements from the currently used 50 mm spouted bed coaters, intermediate size spouted beds (84 and 150 mm), and selected candidates for production scale coaters. In order to supply the required fuel particles for commercial reactors, 150 to 300 mm diameter coaters need to be designed. However, scale-up of the fluidization processes with chemical reactions poses unsolved problems. These processes are highly nonlinear, sensitive to boundary conditions and disturbances, and exhibit transients and unstable hydrodynamics. Further, the larger coaters will inevitably require distributors that are more sophisticated than the typical conical bottom.

Researchers will provide the experimental data to the ORNL chemical vapor deposition (CVD) fluidization reactor modeling team to validate computational fluid dynamics numerical models implemented in MFIX (Multiphase Flow with Interphase eXchanges). Validated models make parametric design simulations practical. Successive iterations between laboratory experimentation and simulations will target an optimal coater design. Researchers will use the experimental data to develop correlations for several spouted bed characteristics that may lead to scale-up relationships. This research is also expected to provide on-line monitoring and feedback control structures for coater operation.

Research objectives are to:

- Design and fabricate fluidized beds with changeable nozzles, distributors, and other components
- Develop LabVIEW software for operation, control, data collection/synchronization, data archiving, and feedback control
- Collect detailed experimental data, including local gas velocities and high-speed inlet pressure measurements (1,000 samples/second)
- Develop an experimental matrix of parameter, which may include: bed diameter, cone angle and throat diameter, selected distributors, bed load (amount of particles), type of gas, gas flow rate, as well as particle properties (surface characteristics, sphericity, size distribution, density, bulk density)
- Conduct image analysis using synchronized video from two cameras for spout shape (height and diameter), Poincaré sections for particle circulation, resident times distributions, and wall boundary conditions
- Develop correlations for spouted bed characteristics, such as minimum spouting velocity, bed pressure drop, spout shape, and scale-up
- Develop new measurement techniques and implement maturing technology, such as the ability to measure local particle flux, local particle momentum distribution, use fiber-optic probes for circulation studies, and local void fraction measurement
- Support instrumentation and control of hot coaters and address data measurement in the harsh coating environment of high temperature (2,000K) and chemical vapor deposition (coats everything)
- Apply chaos and nonlinear dynamics concepts for process monitoring and control (distributor restrictions, wall agglomerates, free agglomerates, hydraulic state, abnormal events)

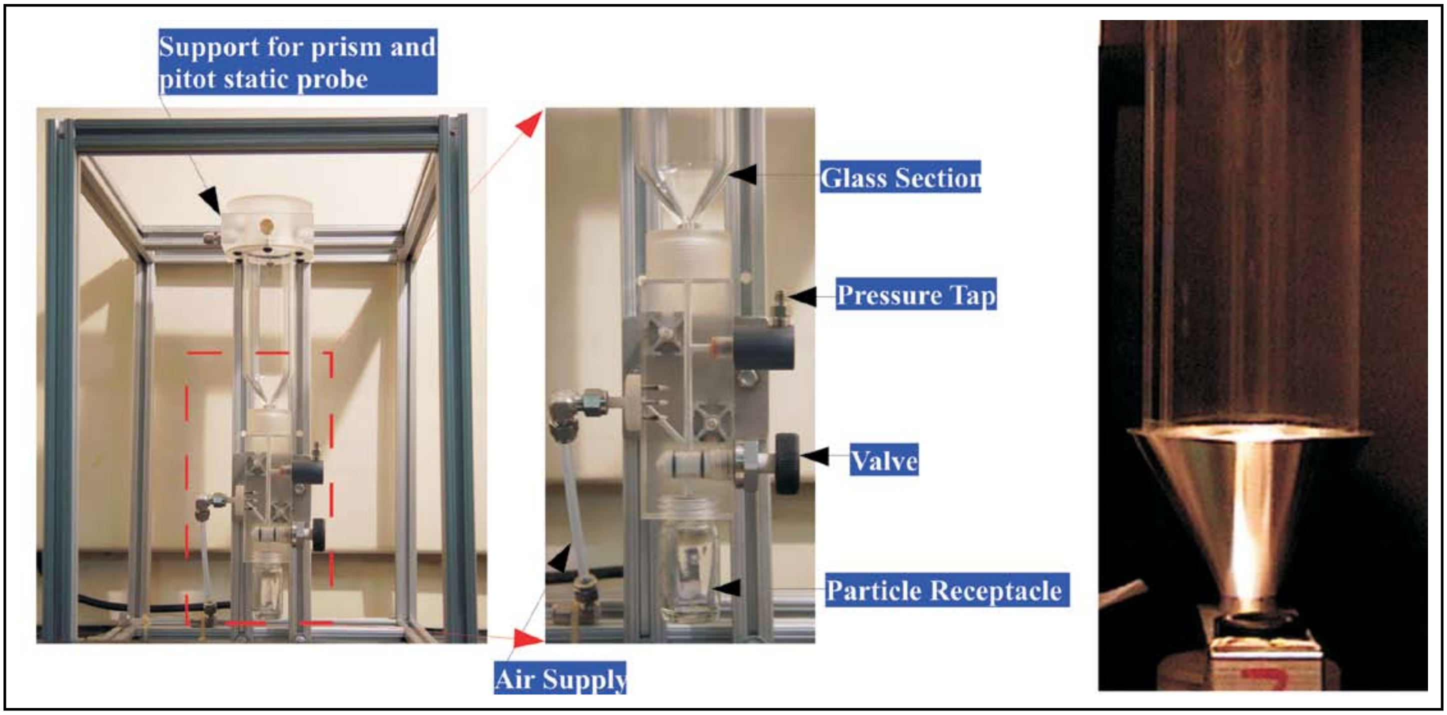


Figure 1. Optically clear beds provide access for image collection. Provisions are made for non-optical measurements as the fluctuating inlet pressure and Prandtl tubes for local gas velocity.

Research Progress

The spouted beds used for nuclear fuel particle coating are different than most spouted beds discussed in the literature as they are coating very dense particles and are very shallow (height of particles from the base of the cone/diameter of the bed < 1). However, the particles in the beds have a very narrow particle size distribution and are nearly spherical. Spouted beds of 50 mm diameter with flexibility to evaluate several configurations have been constructed and utilized for data collection. One configuration is shown in Figure 1.

Researchers are using a state-of-the-art National Instruments PXI system for all operation, data acquisition, and data archiving for the experiments, with much of the initial data analysis done on-line. They have developed correlations from the experimental data for minimum fluidization velocity, minimum spouting velocity, and pressure drop across the bed, comparing the results to literature where possible. It should be noted that most correlations in the literature are for substantially different process parameters.

The researchers have collected data from the 50 mm diameter bed for 7 sizes of surrogate ZrO_2 particles, 10 or more gas velocities, 4 cone angles and 4 bed loads. This extensive database has been used to validate the MFIX numerical models. Figure 2 show one frame from captured video of the spouted bed images at four different fluidization gas flow rates. Figure 3 shows detailed experimental local time-averaged radial velocities at 1 mm intervals for eight heights above the outlet of the throat. The simulation tracks the experimental values well at the centerline. More significantly, the model output compares well dynamically to lab data using the Fourier transform power spectrum as presented in Figure 4. However, the experimental results generally show one pinching amplitude peak, while the simulation shows two. Researchers are performing additional simulation experiments to understand this difference. Figure 5 shows visualized time-averaged simulation results for a vertical cross section through the center of the bed and a time-averaged image from the

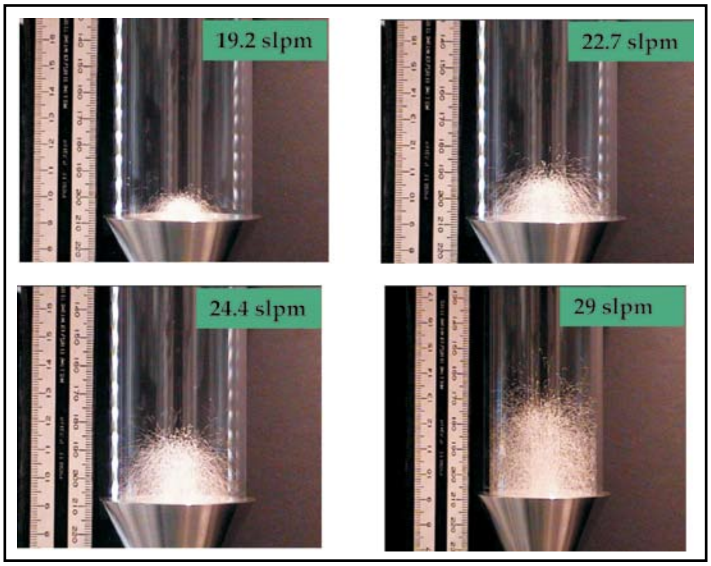


Figure 2. Fountain characteristics with varying gas flow rate for 55 grams of 500 μm ZrO_2 in a 60° cone.

experiment. Time-averaged images are used as the spout exhibits considerable variation as illustrated by the power spectrums shown in Figure 4.

Proof of principle has been demonstrated in the lab for a particle flux and momentum sensor. Detailed measurements are now being made with design version 4 of the sensor. Designs are being developed to test it in the hot surrogate coater at ORNL. Other approaches to develop or extend available sensors for fluidized bed measurements are being evaluated.

Image analysis software and procedures have been outlined to collect mixing and residence time distributions. The methodology for extracting spout height and diameter are illustrated in Figures 4 and 5. The images in Figure 4 are one frame from the digitized images. Figure 5 shows an image that is an average of many frames over a time period long enough that the spout has visited all of the states in its attractor. Analysis of this image will provide quantitative spout shape values.

Planned Activities

Activities planned for the next year include:

- Construct 84 and 150 mm beds with cone distributors and a limited number of distributors. The selected distributors will utilize the Coater Scale-up Plan, PLN-1975, and results from the MFIX modeling team. Data collection from these larger beds with cone distributors will be commissioned soon.
- Perform an added activity beyond the original proposal, which is the experimental evaluation of basic particle properties. These properties are coefficient of restitution and angle of repose for non-coated and coated surrogate particles on various surfaces. This will provide primary data for parameters in MFIX models.
- Measure spout height and diameter over a selected experimental parameter matrix and development of correlations.
- Demonstrate the particle flux and momentum sensor, ideally in a hot coater. Calibration equipment needs to be designed and constructed.
- Heat the spouted bed to approximately 400K, as developed for another project in the UT lab.
- Determine marked (e.g., colored) particle statistics for passage through a plane or a local volume; local particle velocities, flux, and void fraction; and wall boundary conditions from images of the 50 mm bed. The boundary conditions can be used in the numerical models.

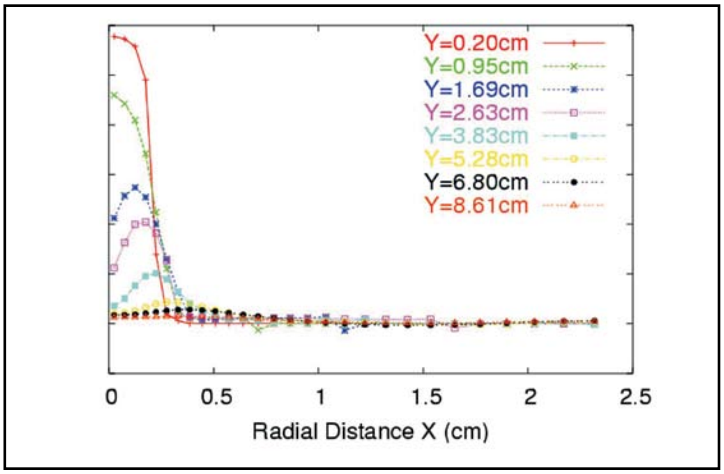


Figure 3. Radial variation of time-averaged axial gas velocity in the lab coater with 500 μm particles in a 60° cone.

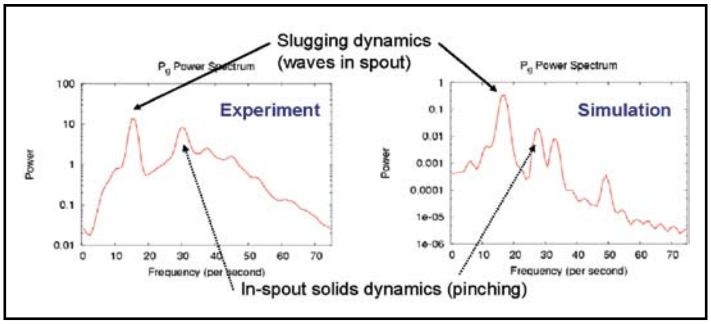


Figure 4. Power spectrum of the coater inlet gas pressure with 300 μm ZrO_2 particles: experiment versus simulation.

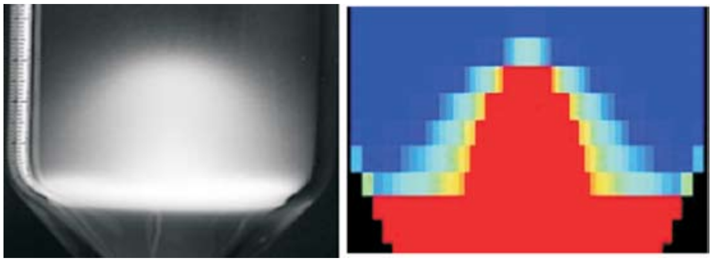


Figure 5. Visualized time-averaged results for a vertical cross section through the center of the bed: experiment versus simulation.

- Develop a low order ordinary differential equations model. Such a model would provide insight into understanding spouted bed behavior by identifying the important physical nonlinearities and process parameters. This type of model could be used for model-based control to improve the long-term operation of coaters.

Researchers plan to obtain particle trajectory information using the Computer Automated Radioactive Particle Tracking (CARPT) facility at Washington University in St. Louis. The research team will provide design information and sensor specifications. They will also collect high-frequency pressure measurements to compare with those already in the database and evaluate in conjunction with CARPT results, experiments at UTK and ORNL, and MFIX models.

A bilateral agreement has been made with Korea Maritime University to evaluate complex flows with quantitative flow visualizations such 4-dimensional particle tracking velocimetry (4D-PTV). They will provide free use of commercialized software; support in using the software; and guidance in specifying the needed high-speed cameras, optics, and lighting. The researchers are seeking funding opportunities.

NUCLEAR ENERGY RESEARCH INITIATIVE

Uncertainty Analyses of Advanced Fuel Cycles

PI: Laurence Miller, The University of Tennessee

Collaborators: Argonne National Laboratory

Project Number: 05-123

Project Start Date: March 2005

Project End Date: March 2008

Research Objectives

The objective of this research project is to evaluate a variety of issues that influence the sustainability of power generated by nuclear energy. The research will model various fuel cycle options and strategies for implementation through the use of a versatile computer program, Dynamic Analysis of Nuclear Energy System Strategies (DANESS), that was recently developed by personnel from Argonne National Laboratory (ANL). This code has capabilities to calculate numerical values of parameters that relate directly or indirectly to the endpoints cited below. In addition, uncertainties in the input data will be propagated through the computational model to determine distributions of values for the desired results; thus, the quality of results can be evaluated and understood. Also, the lack of knowledge that contributes to the uncertainty of results can be identified, permitting the researchers to understand where improvements in knowledge are needed and where additional resources should be invested.

Some of the options and strategies being addressed are described below. The work during the first year will focus on the first three objectives, whereas the remaining objectives are the focus of the second and third years.

1) Design and analyze advanced fuel cycles for light water reactors (LWRs)
2) Determine the effect of nuclear data and technological uncertainties on fuel cycle evaluations and evaluate the feedback of nuclear data requirements
3) Identify and assess the repository benefits of advanced fuel cycles
4) Determine the effect of uncertainties on repository benefit assessments
5) Conduct dynamic fuel cycle scenario studies to develop an understanding of the issues in the transition from thermal reactor to a mixed thermal/fast reactor fleet, or a fleet in which the majority of reactors are fast and/or accelerator-driven
6) Optimize the use of key resources, e.g., repository capacity and uranium ore, in the long term for advanced fuel cycles
7) Evaluate the optimal use of fast reactors and accelerator driven systems from both technological and market perspectives

Advanced fuel cycles offer the opportunity to significantly reduce the requirement for geologic repository space if fission products, fissile material, actinides, and fertile material are separated and processed effectively. In particular, the need for repository space is about two percent of that required for the once-through cycle if actinides and fissions are completely separated. Since implementation of advanced fuel cycles necessitates the construction of relatively expensive recycling facilities and of hard-spectrum reactors, it is important that uncertainties associated with deployment of these fuel cycles be understood and minimized.

Significant investments in infrastructure are needed to fully achieve the goal of near optimal management of repository space and overall resource requirements. Implementation of fuel cycles that both conserve repository space and utilize resources more effectively than the once-through cycle involves a number of decisions. These decisions must commit financial resources to achieve the goals associated with advanced fuel cycles. Some technological infrastructure that needs to be developed for improved fuel cycles include: 1) development of high burnup fuel, 2) utilization of mixed oxide (MOX) fuel, and 3) deployment of hard-spectrum reactors.

Research Progress

Researchers have investigated the time dependencies for implementing these technologies and evaluated their benefits. Figure 1 illustrates the effect of implementation time for integrating high burnup fuel into the current

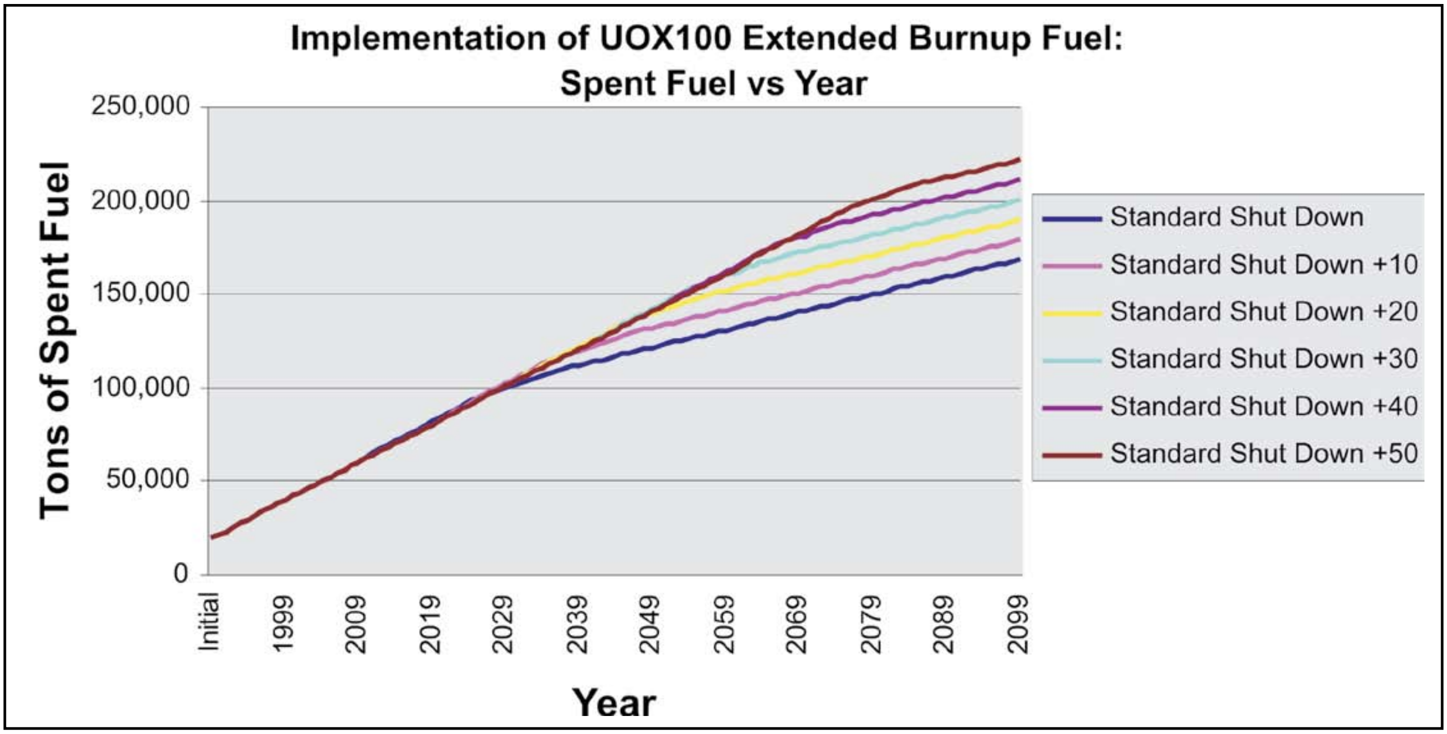


Figure 1. Spent fuel inventory with time implementation for UOX 100 at 0 percent growth.

U.S. reactor fleet, assuming zero percent growth generating capacity. The earliest possible implementation of high burnup fuel will provide the maximum benefit in terms of overall spent fuel inventory, with each decade removing an additional 10,000 tons from the waste stream by the year 2100.

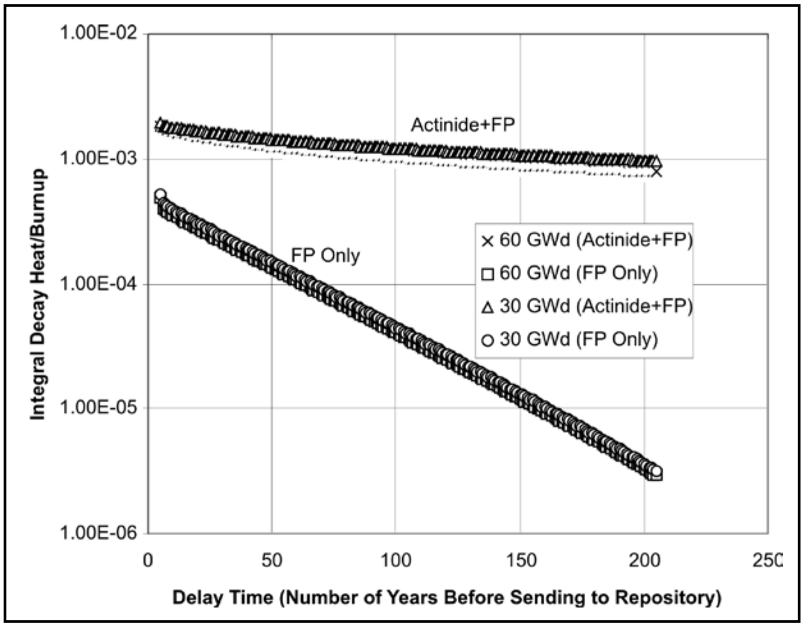


Figure 2. Ratio of integral decay heat deposited in the repository over 1,200 years for burnups of 30 GWd and 60 GWd with variable time between removal from the spent fuel pool and placement in a repository.

The researchers have completed studies evaluating uncertainties in the time of implementation for selected parameters in three fuel cycles, including high burnup fuel and MOX fuel with plutonium recycle. The assessment for hard-spectrum reactors is in progress. Some of the output variables included in these assessments are the following:
1) mass of plutonium in the repository at year 2100 per gigawatt-day (GWd) of energy produced, 2) integral heat produced over a 1,200 year period following closure of the repository, 3) mass of spent fuel, and 4) isotopic compositions of spent fuel. The benefit of using very high burnup fuel to 100 GWd per metric ton is only about 20 to 30 percent reduction of repository space requirement, if transition begins within the next 20 to 30 years. However, implementation of MOX fuel could achieve a factor of two or three in the reduction of repository requirements. Results for these studies are obtained from the DANESS computer code, which Argonne National Laboratory is developing for the purpose of nuclear fuel cycle evaluations including sensitivity and uncertainty analyses.

The impact of removing actinides from spent fuel is illustrated in Figure 2. Results in this plot correspond to the energy deposited in the repository during 1,200 years of residence as a function of cooling time before emplacement in the repository.

Results illustrated in Figure 2 show that disposal of actinides in fast reactors, rather than in repository, could provide a factor of 50 savings in repository requirements if cooling times of 100 are employed, and notably more if longer cooling times are considered.

Planned Activities

Researchers will focus on first-year objectives that are in progress. In particular, this will include: assessing data requirements, selecting fuel cycles to be considered in detail for the second year, and identifying repository benefits associated with advanced fuel cycles chosen for further study. The work during the second year will focus on determining the effects of uncertainties on repository benefit assessments and on the performance of dynamic fuel cycle scenario studies to develop an understanding of the issues in the transition from thermal reactor to a mixed thermal/fast reactor fleet, or a fleet in which the majority of reactors are fast and/or accelerator-driven.

In order to accomplish the second-year objectives, some of the required tasks are as follows:

- Review documentation on repository assessments and on dynamic fuel cycles scenarios
- Calculate the fractional contributions to uncertainties due to each parameter that has significant influence on repository requirements, such as repository space required per megawatt-hour of power generated
- Select cases for detailed study and develop corresponding strategies for chosen dynamic fuel cycles
- Conduct uncertainty assessments on dynamic fuel cycles selected for study.

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, Westinghouse Electric Corp.

Project Number: 05-125

Project Start Date: March 2005

Project End Date: March 2008

Research Objectives

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 include:

1) Developing a new methodology for the direct coupling of the pin-by-pin bundle loading control variables and the core-wide (bundle-by-bundle) optimization objectives
2) Extending this new methodology into a new application that includes control variables, objectives, and constraints designed to maximize incineration of targeted minor actinides

Although the first objective is expected to uncover considerable dormant thermal margin, the second (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 by investigating spent fuel treatment and recycling options for current generation LWRs and supporting the DOE's technical assessment of a second high-level waste repository. The general work scope entails:

- Performing within-bundle related developments. Defining candidate minor actinides; specifying control variables, objectives, and constraints at the bundle level; and generating families of lattices and bundles as a function of relevant parameters (exposure, void fraction, etc.).
- Performing core-wide activities. Defining bundle-to-core coupling, generating families of core loadings and corresponding operational strategies (control rod patterns, core flow), and validating diffusion theory analyses via transport theory benchmarks.
- Completing global integration of the project. Verifying proper functioning of models and generating optimized advanced BWR bundles and associated fuel cycle strategies.

Research Progress

Researchers are employing three different software packages for this project. First, DOE software obtained includes MCNP5, MONTEBURNS, ORIGEN, and SCALE, as distributed by RSICC at Oak Ridge National Laboratory (ORNL). Second, the University of Cincinnati (UC) and Westinghouse executed a software license and proprietary information agreement for the use of the IFIGEN/PHOENIX-4/POLCA-7 and the Core Master 2 (CM2) GUI interface. Last, a software license agreement was recently executed between UC and North Carolina State University (NCSU) to employ the FORMOSA-L lattice optimization code in this project. At this time, the DOE software and the PHOENIX-4 code have been delivered and installed in the UCNRE 22-node Beowulf cluster. Due to a few unexpected delays, the remainder of the Westinghouse software package (most notably POLCA-7 and Core Master 2) and the NCSU EPRC code, FORMOSA-L, are expected to be delivered in early 2006.

The first key "within-bundle" tasks carried out for this project involved the benchmarking and validation of MCNP5 models depleted with MONTEBURNS/ORIGEN, still considered an emerging technology. Benchmarking comparisons were carried out against the well-seasoned Westinghouse PHOENIX-4 industrial-grade code package, employing typical 10x10 modern BWR lattices. PHOENIX simulations were compared favorably against a few available standards at 0 percent, 40 percent, and 70 percent void; hot and cold operational conditions; and at a combination of controlled and uncontrolled conditions (with and without control blades).

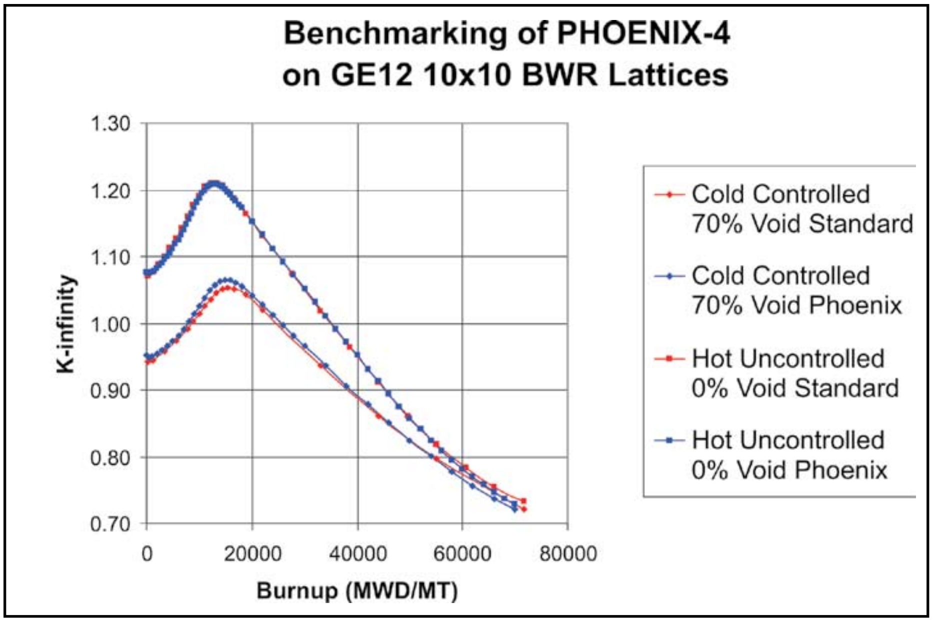


Figure 1. PHOENIX-4 benchmarking data with two representative cases - hot uncontrolled 0 percent void lattice and cold controlled 70 percent void.

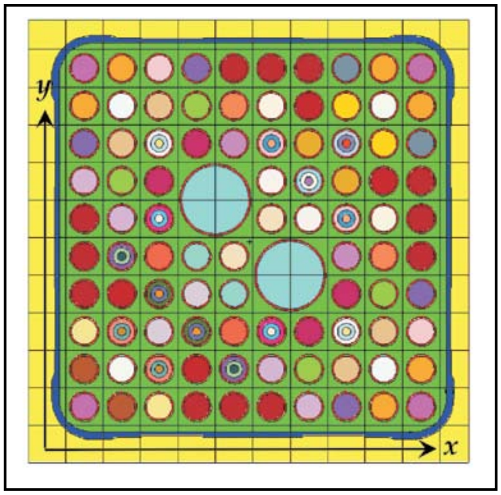


Figure 2. MCNP5 graphical rendition of the "dominant zone" axial region of a BWR GE12 10x10 fuel bundle. Each color denotes a different material region.

For ease of illustration, the plot shown in Figure 1 includes two representative cases: hot uncontrolled 0 percent void and cold controlled 70 percent void. This highlights the ability to properly model a modern 10x10 lattice under the range of typical operational conditions using this code. Figure 2 is an MCNP5 visual editor graphical rendition of the "dominant zone" axial region of a BWR GE12 10x10 fuel bundle that was modeled in this study. Each color denotes a different material region as modeled with the MONTEBURNS code for depletion purposes. Symmetry is maintained across the diagonal between the two large water holes in blue.

An important highlight of the MCNP/MONTEBURNS model includes the modeling of independent concentric material depletion region "rings" in pins with gadolinium loadings.

This technique is used to account for the high level of self shielding that occurs in highly absorbing materials, whereby these pins burn in a manner much similar to how an "onion peels," by layers. Accordingly, Figure 3 shows good agreements between the deterministic code results and the MCNP5/Monteburns approach for a 0 percent void, uncontrolled, hot case for the same lattice shown above.

Preliminary point models have been developed by employing the ORIGEN2.2 code to investigate the global impact of "spiking" typical BWR fuel with americium, neptunium, and curium minor actinides. Also, as shown in Figure 4, spatially detailed MCNP5/Monteburns models of the "vanishing zone" of a GE12 bundle were modeled with various arrangements of pins "spiked" with americium. Table 1 shows early results which tally the masses of these preloaded americium isotopes as a function of time extending up to approximately 36 months, in which it appears evident that a significant reduction in the inventory of the key americium isotopes is achievable. These and other related results have been submitted to the PHYSOR 2006 meeting (Vancouver) and to the ANS annual conference in Reno, NV.

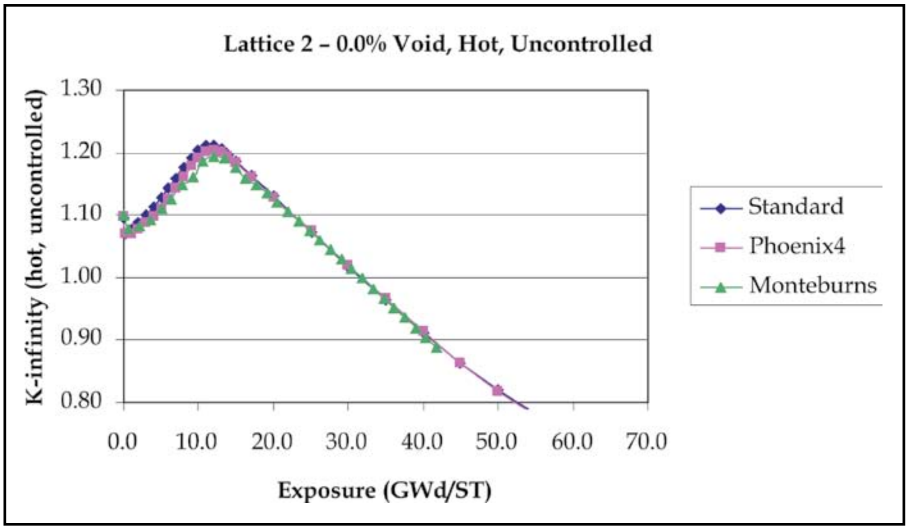


Figure 3. Comparison of PHOENIX-4 benchmarking data with MCNP/MONTEBURNS for the hot uncontrolled 0 percent void lattice.

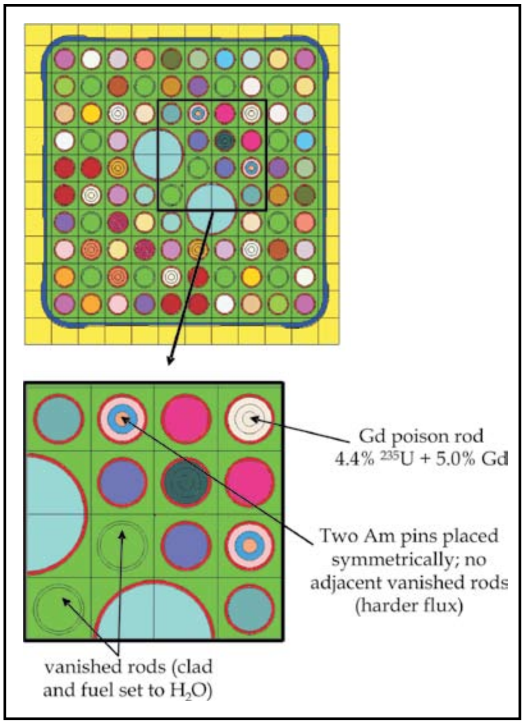


Figure 4. Spatial MCNP5/Monteburns models of the "vanishing zone" of a GE12 bundle, modeled with various arrangements of americium-spiked pins.

	Time steps (1 time step = 1.2 months)								
	1	4	7	10	13	17	22	27	30
241Am	6.401	5.423	4.386	3.391	2.486	1.462	0.6069	0.1984	0.0937
242Am	0.039	0.069	0.057	0.044	0.032	0.018	0.0074	0.0024	0.0011
243Am	1.061	0.974	0.913	0.851	0.797	0.714	0.6007	0.4979	0.4531

Table 1. Americium isotopic inventory (grams) in Am-spiked pins versus time.

Planned Activities

The plans for 2006 include developing models of BWR lattices with pins that have a blending of minor actinide contents similar to those scheduled for manufacturing and irradiation in the ATR facility in Idaho. Researchers also expect to initiate the coupling of the FORMOSA-L lattice optimization driver to the PHOENIX-4 lattice physics code to begin developing optimization objective functions and constraints that maximize minor actinide transmutation rates. This will lead to the "stacking" of lattices into actual three-dimensional bundles.

In late 2005, a representative of the Westinghouse BWR nuclear design staff provided training on the PHOENIX/POLCA and Core Master 2 codes, enabling the research team to carry out core-wide and multi-cycle studies of BWR bundles with minor actinide loadings during the second half of 2006.

Optimization of Oxide Compounds for Advanced Inert Matrix Materials

PI: Juan Nino, University of Florida

Collaborators: Imperial College of Science, Technology, and Medicine; Idaho National Laboratory

Project Number: 05-134

Project Start Date: April 2005

Project End Date: March 2008

Research Objectives

This research project is being conducted as part of DOE's Advanced Fuel Cycle Initiative program. Its objective is to model, design, synthesize, and characterize oxide ceramics that have optimal thermophysical properties for potential use as inert matrix fuel (IMF) in the current generation of light water reactors (LWR). To accomplish this, researchers will utilize a combined computational and experimental approach. Specifically, they will focus on the materials science aspects of developing oxide compounds and composites that exhibit minimal radiation swelling, high thermal conductivity, and excellent hot water corrosion resistance.

One important potential benefit of using optimized IMF materials rather than UO_2 fuel in current generation reactors is its higher thermal conductivity, which will considerably reduce the centerline temperature of the fuel. Lower stored heat in the fuel will minimize the impact of a loss-of-coolant accident. Further, use of an inert matrix will avoid formation of transuranic fission products by neutron capture, thereby allowing higher fuel burnup and reducing waste. Better corrosion resistance will make the IMF compatible with the coolant under cladding breach accidents.

Research Progress

The following summarizes the progress made in the first two quarters of this project:

- Researchers determined the processing routes and parameters for the synthesis of MgO-pyrochlore ceramic-ceramic (cercer) composites as part of the development of the experimental infrastructure. They also chose the solid state processing route and optimum milling times and identified calcination and sintering temperatures and heating profiles.
- Researchers identified suitable interatomic potentials as part of the development of the computational infrastructure for simulating the thermal conductivity of the candidate IMFs. In addition, they adapted previously utilized pyrochlore potentials and determined and tested new potential parameters for MgO.
- Researchers initiated solid state synthesis and characterization of the IMF candidates. The first set of MgO-$Nd_2Zr_2O_7$ sample pellets were successfully sintered to 99 percent of theoretical density. X-ray diffraction profiles of the individual phases, green and sintered composite pellets, are presented in Figure 1.

While there is no detrimental reaction between the MgO and the pyrochlore phases, analysis of the composite microstructure revealed non-homogeneous phase mixture with large regions of MgO phase. Such a microstructure can negatively impact the corrosion resistance of the IMF. Researchers modified the processing parameters, especially in the milling and mixing steps, to achieve a homogeneous microstructure.

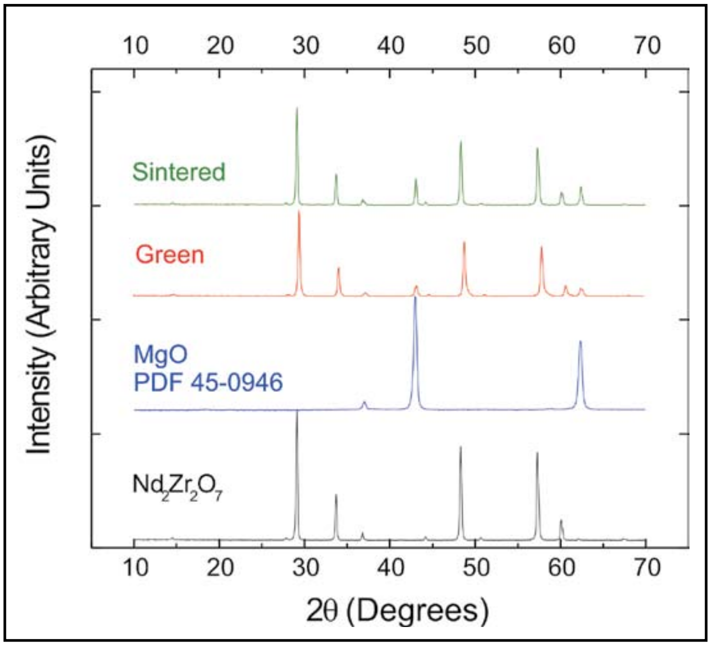


Figure 1. XRD of $Nd_2Zr_2O_7$ and MgO before and after sintering at 1,550°C for 8 hours.

- Sol-gel synthesis and characterization of $Nd_2Zr_2O_7$ was performed to investigate the effect of initial particle size on the final microstructure and corrosion resistance of the IMF composite. Phase pure pyrochlore formed after 6 hours at 1,200°C, which is 150°C lower and took half the time as solid state synthesis. In addition, the particle size was 30 nm, which can enable better microstructural control.
- Using initial simulation data, researchers calculated the thermal conductivity of $Nd_2Zr_2O_7$ to be 2.10 W/m-K at 1,473K. Figure 2 presents the thermal conductivity of the pyrochlore for different temperatures, which was determined through simulation. The closed symbols are an average over 1-1.75 million time steps; the open symbols are an average over 1.75-2.5 million time steps. The difference between the two is a measure of uncertainty of the simulated values. These simulated values are expected to be 30 percent larger than experimental values mainly because point defects and other microstructural defects are not considered in the calculations. However, the trends in the thermal conductivity behavior as a function of composition are well captured.
- Neutronic properties for candidate IMF composites were simulated using Monteburns and MCNP codes. The calculations also included mixed oxide (MOX) fuel and ZrO_2 for comparison. It was found that for the IMF composites with equal volumes of MgO and pyrochlore, the reactivity of the fuel is comparable to the MOX fuel only up to 20 MWD/kg equivalent burnup. This is due to the relatively large neutron absorption of neodymium and ytterbium. However, the MgO-ZrO_2 showed higher k_{eff} than MOX fuel, up to 50 MWD/kg. Based on these initial results, the IMF composition is being reformulated.

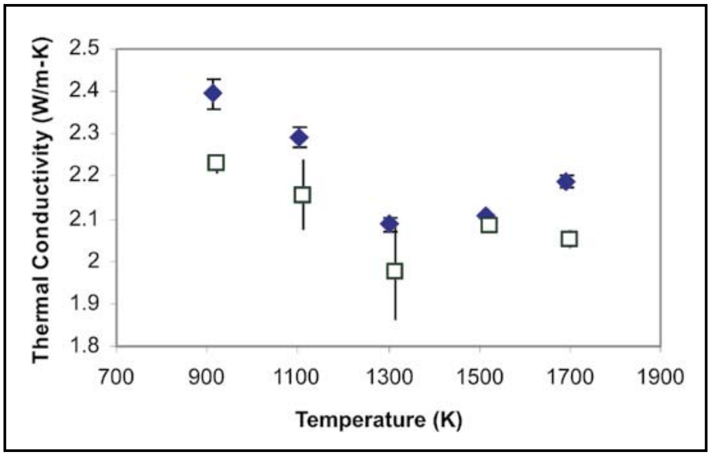


Figure 2. Thermal conductivity of $Nd_2Zr_2O_7$ determined from simulation.

Planned Activities

In line with the proposed tasks and timeline of the project, a complete set of MgO-pyrochlore will be synthesized for thermal conductivity measurements. The ongoing microstructural characterization will include a compositional study of the composite interface. In addition, corrosion and reprocessing studies of the composite IMF candidate materials will continue. The final compositions for the IMF candidates will be determined and inert powders will be synthesized. Finally, researchers will continue the thermophysical and neutronic property computational simulations on the composite IMF candidates which will further guide the experimental effort.

NUCLEAR ENERGY RESEARCH INITIATIVE

Synthesis and Optimization of the Sintering Kinetics of Actinide Nitrides

PI: Darryl P. Butt, Boise State University;
R. Baney, J. Tulenko, University of Florida

Collaborators: None

Project Number: 05-135

Project Start Date: August 2005

Project End Date: July 2008

Research Objectives

The primary objective of this project is to develop processing models and to perform an economic analysis on the synthesis of nitride-based nuclear fuels. Such fuels, particularly those based on uranium (UN) and plutonium (PuN), are candidates for the Advanced Fuel Cycle Initiative (AFCI). Nitride-based fuels have a combination of higher uranium loadings and higher thermal conductivity compared to other fuel forms, thus lower enrichment can be used. It is also 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 fully understood. Having a quantitative understanding of the mechanism and kinetics of densification and grain coarsening is critical to ensure both satisfactory product performance and favorable economics 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.

In this study, researchers will synthesize actinide and surrogate powders of varying morphologies and particle size. They will thoroughly characterize the powders and press them into fuel forms. Detailed sintering studies will be conducted in order to assess the specific rate equations and kinetics models as a function of time and temperature, grain size, and other processing variables. By applying these fundamental models and combining microstructural characterization, the researchers will determine rate limiting and process controlling mechanisms for 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.

Research Progress

The project PI moved his laboratory from the University of Florida (UF) to Boise State University (BSU) in August 2005. Consequently, much of the efforts this quarter were spent setting up facilities at BSU. Researchers have set up two controlled atmosphere furnaces for hydriding/nitriding with N_2 and up to 6 percent H_2, and they procured a third furnace for hydriding in pure H_2. All three systems will be equipped with mass flow control and vacuum capabilities as low as 10^{-6} torr. An induction furnace for use in high-temperature synthesis of small exploratory samples can achieve temperatures in excess of 2,000°C in ultra-high vacuum or controlled atmospheres.

The researchers have procured several powders for use in the project. As nitrides are not available in most cases, they will be synthesized in the laboratory from oxides, hydrides, or pure metals. Although the original program plan indicated that synthesis of powders by novel routes would not begin until the end of the first program year, researchers have begun this work early. Consequently, procurement of chemicals for this work is also underway. Plans are to synthesize UN and surrogates via amide, iodide, and silazane routes.

Initial work has been conducted to produce cerium nitride (CeN) surrogate powders. This work involved hydriding Ce metal, resulting in the conversion of approximately 10 grams of metal into pure CeH_x, as confirmed by X-ray diffraction. Completion of the gas-handling system is necessary to carry out the nitridation steps for conversion of the hydride to CeN. In addition, work has begun to assess the sintering kinetics of ZrN powders. The first pellets have been sintered and are currently being characterized.

Planned Activities

The major tasks associated with this project are to fabricate, procure, and characterize a sufficient quantity of powder to begin synthesis and fuel fabrication; fabricate and characterize fuel as a function of temperature, time, and grain size; fully assess the kinetics of sintering; and produce an economic analysis of nitride processing. These tasks are summarized below:

- Powder synthesis and characterization
- Fuel fabrication
- Microstructural characterization
- Assessment of atmospheric effects on densification and grain growth
- Model sintering kinetics and assess sintering mechanism
- Development of novel routes for nitride powder preparation
- Economic assessment

The Development of Models to Optimize Selection of Nuclear Fuel Materials through Atomic-Level Simulation

PI: Simon R. Phillpot, University of Florida

Collaborators: Commissariat à l'énergie Atomique (CEA), Los Alamos National Laboratory, Imperial College London

Project Number: 05-136

Project Start Date: April 2005

Project End Date: April 2008

Research Objectives

The objective of this project is to develop an advanced fuel performance calculational platform, based on the FRAPCON code, to optimize the selection of nuclear fuel materials. 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 on which they are based, nor can they be used for materials that lack experimental data. This project will develop detailed nuclear material properties input from databases derived from fundamental thermodynamic, atomic-level, and electronic-level calculations and simulations.

Development of this "first-principles-based" code will allow fuel development programs to reduce costly and time-consuming experimentation and transition to a selection process that is based on modeling, simulation, and analysis and that only will require confirmatory irradiation testing. 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.

This project consists of the following 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 those 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.

Research Progress

FRAPCON is hard-coded for a cylindrical fuel element with a gap, fuel clad, crud layer, and coolant. The code calculates the thermal distribution in the fuel at each of the user input axial nodes. To perform a calculation, the user inputs coolant inlet temperature, coolant channel equivalent heated diameter, time coolant mass flux, and axial linear heat generation rate, all of which are used to calculate the coolant bulk temperature through a single channel coolant enthalpy rise model. After this calculation, a temperature rise over the crud is calculated and the temperature at the outside of the cladding is determined. Using Fourier's Law, the temperature rise through the zirconium oxide and the clad is calculated. An annular gap conductance mode is used to find the temperature at the outside surface of the fuel. From this value, a temperature distribution is determined for the fuel pellet. This distribution assumes symmetry at the centerline and takes into account any fuel cracking effects.

FRAPCON consists of a series of subroutines to perform these calculations, most of which will not require modification to allow for first principles input. Work is currently underway to assess the individual importance of the five factors that determine the fuel thermal conductivity: 1) the conductivity of unirradiated urania, 2) the dissolved fission product factor, 3) the precipitated fission product factor, 4) the effect of porosity, and 5) the radiation effect. Initial efforts will focus on the first item, the conductivity of unirradiated urania as a function of temperature. Then the other four factors will be taken into account.

Thermal expansion can be simulated at the atomic level in a way that is directly analogous to an experimental approach. That is, the material is heated at zero pressure, and the temperature dependence of the lattice parameter is determined. Researchers used conventional atomic-level simulation methods with two different potentials, constructed by Grimes and Yamada, to simulate UO_2. Over the range of temperature examined (from 0 to 2,000K), uranium dioxide remains in the cubic fluorite structure. As expected, the lattice parameter is an approximately linear function of the temperature, which means that thermal expansion is only weakly temperature dependent. Linear fits to Fink's experimental compilation of thermal expansion data yielded a thermal expansion of $11.9 \times 10^{-6} K^{-1}$. By comparison, the researchers determined thermal expansion coefficients of, $7.02 \times 10^{-6} K^{-1}$, and $9.65 \times 10^{-6} K^{-1}$, respectively for Grimes' potential and Yamada's potential description of UO_2.

Using analogous simulation methods, the researchers determined the thermal conductivity, κ, of UO_2. The thermal conductivity calculated using the Yamada potential agrees quite well with the experimentally determined temperature dependence. However, the Grimes potential overestimates κ by almost a factor of three, compared to the experimental values at both 300K and 1,000K. Nevertheless, the trend is basically the same between the experimental data and the Grimes model; in both cases, the thermal conductivity drops by approximately 53 percent.

Researchers have concluded that the Yamada potential is superior to the Grimes potential in matching both the thermal expansion and thermal conductivity of UO_2. They note, however, that there is currently no parameterization of the Yamada potential for U^{3+} or U^{5+} ions, meaning that it can not yet be used for simulations of off-stoichiometric systems.

Cerium oxide is frequently used as a surrogate material for plutonium oxide. Researchers performed a detailed assessment of thermodynamic data associated with phase changes in the U-Pu-O and Ce-O systems. In reducing atmospheres at high temperatures, both compounds lose oxygen in a very similar way. The resulting Ce- and Pu-compounds show a distinct range of stoichiometry and are structurally and thermodynamically closely related. Numerous experimental data for the Ce-O phase diagram and related thermodynamic properties are published. Therefore, models and computer datasets with Gibbs free energy descriptions for the Ce-O system phases developed from the CALPHAD approach can be comprehensively tested and compared with available experimental data. This knowledge can then be transferred to the phase modeling and thermodynamic description of the Pu-O system where less experimental data are available.

Following this idea, the researchers assessed a thermodynamic data set for the central part of the Ce-O system covering the composition range between Ce_2O_3 and CeO_2 up to 2,000K. The thermodynamic modeling uses the so-called Compound Energy Formalism to develop Gibbs free energy functions for all phases in the system. The diagrams calculated from this dataset reproduce fairly well the experimental data from literature. The phase diagram and the following thermodynamic functions were calculated: 1) heat capacities, enthalpies of formation, and enthalpy increments of Ce_2O_3 and CeO_2; 2) partial enthalpies of oxygen in CeO_{2-x}; and 3) chemical potentials of oxygen in the CeO_{2-x} solid solution and in the two phase regions. Additionally, numerous tests and simulations were made for all thermodynamic functions not yet measured experimentally. For the first time, researchers developed a thermodynamic description of the Ce-O system that simultaneously describes the phase diagram data (including the solid solution phases) and all coupled thermodynamic functions.

The combination of binary and ternary thermodynamic datasets for the calculation of high-component systems requires a consistent modeling of all the stoichiometric and solid solution phases according to their crystal chemistry and the related thermodynamic and thermophysical data. A general international agreement on the reference states and modeling for the solution phases in the Compound Energy Formalism is necessary to avoid the development of non-compatible datasets. Such international collaboration is also important for the efficient progress of the work. Therefore, the researchers are actively collaborating with groups worldwide in the research area of the thermodynamics of nuclear fuels.

Planned Activities

Over the next research phase, researchers plan to complete the analysis and reconstruction of the FRAPCON code. The atomic-level simulations are being extended to PuO_2 and the Pu-U-N system. The tools required for the atomic-level simulation of radiation damage in the oxides and nitrides will be implemented into these codes during the next quarter, enabling simulations of radiation damage of UO_2 and its effect on the thermal transport properties. In the area of thermodynamic analysis, researchers will focus on a comprehensive assessment on the U-N, Pu-N, and U-Pu-N systems and continue to work on the U-Pu-O systems.

NUCLEAR ENERGY RESEARCH INITIATIVE

Development of TRU Transmuters for Optimization of the Global Fuel Cycle

PI: John C. Lee, University of Michigan

Collaborators: None

Project Number: 05-142

Project Start Date: January 2005

Project End Date: December 2007

Research Objectives

The objective of this project is to develop advanced fuel cycles for the transmutation of transuranic (TRU) elements in irradiated nuclear fuel from light water reactor (LWR) power plants. 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 disposing and storing irradiated nuclear fuel, such as 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 and fuel cycle economy.

Research Progress

Researchers have developed an LWR equilibrium search algorithm to systematically compare the performance of diverse fuel cycles for global fuel cycle analysis. The algorithm is similar to the REBUS-3 fast reactor methodology, but with macroscopic depletion capabilities typically employed for LWR analysis. Because neutrons in an LWR core reside primarily in the thermal and epi-thermal resonance region, varying degrees of spectral hardening over a fuel cycle entail a nonlinear evolution of the heavy metal (HM) inventory throughout the multi-recycling process leading to equilibrium. This LWR methodology projects the coupled nonlinear isotopic balance equations to a series of equations that are piece-wise linear in time. The integration of assembly-level lattice physics and global diffusion theory calculations allows iterations of microscopic reaction rates until the linearized isotopic balance equations yield a constant HM inventory. This equilibrium state allows for a direct comparison between LWR and fast-neutron transmuters.

To illustrate the usefulness of the LWR equilibrium methodology and to explore ways to efficiently transmute legacy plutonium in LWR spent nuclear fuel, the researchers have studied the use of thorium in an alternate pressurized water reactor (PWR) fuel cycle. The potential of thorium-based fuel to increase the net destruction of plutonium by eliminating the secondary production of new plutonium has led to the development of a thorium-based mixed-oxide (TMOX) PWR configuration. This configuration consists of (Th, Pu) O_2 pins loaded on the periphery of a standard 17x17 fuel assembly, with the remaining interior fuel pins loaded with (Th, ^{233}U)O_2. Researchers modeled an entire four-loop PWR core with TMOX assemblies and performed three-dimensional, global depletion calculations to arrive at a once-through equilibrium cycle, corresponding to the startup or initial recycle. Figure 1 shows that the TMOX configuration results in significant destruction of plutonium (95 percent depletion of ^{239}Pu and 70 percent depletion of total Pu) in once-through operation. This is

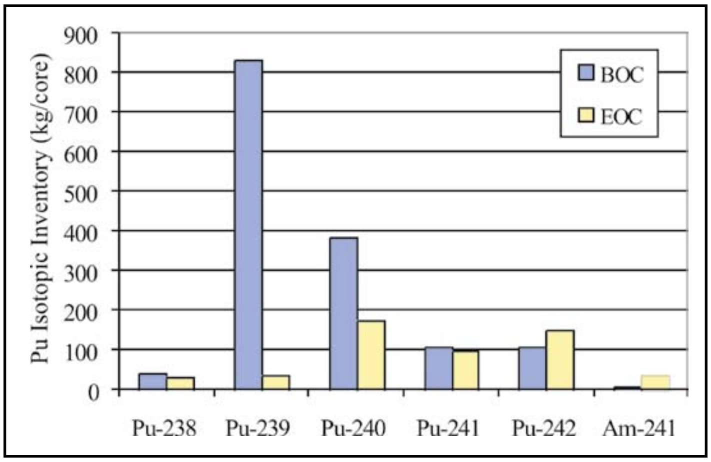


Figure 1. Isotopic plutonium inventory for a TMOX once-through equilibrium cycle.

comparable to the ^{239}Pu depletion rate of 98.1 percent reported for a deep-burn strategy in the driver fuel of the GT-MHR core. In addition to the significant reduction in Pu inventory, its quality is also significantly degraded, making the residual Pu virtually useless for weapons proliferation. In particular, the ^{239}Pu fraction is reduced from 58 weight percent at beginning-of-cycle (BOC) to 6 weight percent at end-of-cycle (EOC), while the ^{242}Pu fraction is increased from 7 percent at BOC to 26 percent at EOC.

The researchers have also studied a denatured thorium cycle, in which natural uranium is admixed with thorium in the TMOX configuration, so that the concentration of ^{233}U is limited to 12 weight percent of total uranium. This reduces the proliferation risk associated with ^{233}U produced from thorium. The denatured TMOX configuration results in 35 percent Pu destruction in a once-through equilibrium cycle. While this is a significant reduction of Pu, it is only half the 70 percent destruction fraction achieved by the regular TMOX cycle, as discussed above.

To assess the impact of introducing Th-based fuel into the overall fuel cycle strategy, researchers studied the once-through TMOX fuel cycle with the DANESS fuel cycle code. DANESS uses a dynamic systems modeling approach in various nuclear fuel cycle sectors and calculates the time-varying mass-flow chains, heat load, and economics of nuclear power plants and other associated fuel facilities. Initial studies with DANESS have focused on analyzing the fuel cycle mass-flow chains, which can be verified using a simplified fuel cycle model to benchmark the code. The benchmark case considered that the 103 existing U.S. nuclear power plants would operate until their current licenses expire and spent nuclear fuel will be sent to the Yucca Mountain repository starting in 2020. The simplified equations compared favorably with the DANESS calculation, as both models predicted a HM inventory of 96,000 tons for disposal in a spent fuel repository from the currently operating plants. Since the Yucca Mountain repository is currently limited to 70,000 tons HM, a second repository would be needed for this scenario. To lessen the repository requirements, researchers have explored an alternative scenario in which the current operating fleet shuts down as scheduled, and 48 new TMOX plants rated at 1.0 GWe are introduced. DANESS calculates that, with the fleet of TMOX reactors operating for 50 years to destroy the legacy plutonium, the HM burden at the Yucca Mountain repository would be reduced to 57,000 tons, well below the legal inventory limit.

Researchers on this project have also studied the use of a sodium-cooled fast reactor to improve repository performance. These studies have focused on a multi-tier strategy which first depletes Pu in LWRs, then destroys the remaining transuranic inventory in the fast reactor system. Typical fast transmuters fueled with legacy TRU tend to experience large reactivity swings over a fuel cycle. However, preliminary calculations indicate that by first exposing the legacy Pu to a once-through TMOX cycle in a PWR plant and then charging the resulting HM inventory into a fast reactor, the reactivity swing over a fuel cycle may be significantly reduced. This would reduce the safety concerns associated with fast-neutron transmuters.

Planned Activities

With the basic LWR equilibrium cycle methodology developed both for the assembly-level and whole-core fuel cycle calculations, the researchers will turn their efforts to the remaining project tasks. In particular, they will focus on developing a framework for optimizing the entire fuel cycle, not merely concentrating on in-core fuel optimization. The near-term objectives include:

- **Development of mathematical formulations.** Explicitly account for fuel cycle constraints, e.g., power peaking factor limit and reactivity swing over a cycle, in the overall fuel cycle optimization process. The fuel cycle optimization strategy should account for the goals of the Generation IV nuclear energy systems, including enhanced economics, proliferation risk reduction, and waste minimization. In particular, an optimized fuel cycle should represent processes that affect both the intrinsic and extrinsic proliferation risk measures and represent spent nuclear fuel transportation risk and repository performance.
- **Development of low-conversion-ratio fast-neutron transmuters.** In addition to continuing the development of denatured Th-based fuels in LWR and fast transmuters, researchers will optimize the performance of fast transmuters by incorporating heterogeneous core designs, including optimized TRU target configurations.
- **Development of a dynamic system model.** Researchers will enhance various modules in the DANESS fuel cycle system code and develop simplified models that could provide physical insights and guidance to evaluate diverse fuel cycles.

NUCLEAR ENERGY RESEARCH INITIATIVE

The Adoption of Advanced Fuel Cycle Technology Under a Single Repository Policy

PI: Paul P. H. Wilson, University of Wisconsin-Madison

Collaborators: Argonne National Laboratory

Project Number: 05-157

Project Start Date: April 2005

Project End Date: March 2008

Research Objectives

This project will develop models to study the impact of a national nuclear waste disposal policy that considers only a single spent fuel storage repository (i.e., 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), one of the near-term goals of the Advanced Fuel Cycle Initiative (AFCI).

Research Progress

Using the documents available on the licensing support network for the Yucca Mountain project, researchers conducted a literature review to investigate any existing specifications on waste packages and the analysis methods by which they are determined. This review identified performance measure for the repository as a whole and a number of specific criteria for the waste packages. However, most are based on the fact that these packages contain once-through light water reactor fuel.

Previous work by Argonne National Laboratory (ANL) has developed some simple specifications for the allowable heat load for packages emplaced in Yucca Mountain. In order to still meet the currently anticipated licensing basis, there are three simple limits: a limit on the instantaneous decay power at the time the forced convection ceases, a limit on the total released decay energy over a time period following the end of forced convection, and a limit on the instantaneous decay power at the time of emplacement in the repository.

Early indications are that repository loading will be limited by thermal considerations, not radiological. That is, any loading which meets the repository thermal performance limits will necessarily have radiological loadings that result in exposures below the stated limits. However, changes in radiological exposure limits and regulations during the fiscal year may have an impact on this conclusion.

The concept of *embodied energy* is best related directly to the quantity of fission products in any material stream since they are the only indicator of actual energy generation. This simplifies the modeling of embodied energy necessary to determine the cost per unit energy of different fuel cycle options. However, to best partition the costs of different fuel cycle processes, it may also be necessary to estimate the potential energy remaining in the material.

In general, the term embodied energy is used in the energy analysis field to account for the process input energy used to produce a material or item. For example, in a nuclear fuel cycle, the mining, conversion and enrichment processes would increase the embodied energy of a unit of nuclear reactor fuel due to the energy used in performing these processes. While this quantity is certainly important in a comprehensive fuel cycle analysis, this project will concentrate more on the theoretically available fission energy and the expended fission energy, known as *embodied potential energy* (EPE) and *embodied delivered energy* (EDE), respectively. Although many economic quantities are normalized by the delivered value, (often converted to electrical energy via a thermodynamic conversion efficiency), most fuel cycle study tools track mass and mass flows of materials within a fuel cycle scenario. Understanding both the potential and delivered energy of those materials in economic terms is necessary when performing economic comparisons of different scenarios. The simplest approach is to ascribe roughly 200 MeV of EPE to every remaining actinide atom and roughly 100 MeV of EDE to each fission product atom. This results in approximately 1 MWd_{th}/g of EPE for actinides and a similar quantity of EDE for fission products.

Associating the available energy exclusively with the actinides and the expended energy exclusively with the fission products preserves economic concepts that will be important for advanced fuel cycles. In particular, recycling of spent nuclear fuel results in a (primarily) fission product stream with a very high specific expended energy and an actinide stream with a very low specific expended energy but high specific available energy. This concept is directly related to the already demonstrated repository benefit of recycling spent nuclear fuel. The variable cost of constructing a deep geologic repository is dependent on the length of the tunnels and is independent of the EDE being disposed in those tunnels. By increasing the EDE of the material stored per unit length of tunnel, the economic utilization of the repository resource is improved.

Since different combinations of processes and reactor systems will have a wide array of total costs and also result in a wide array of EDE and EPE values for different material streams in the system, it is important to track the economic value of the streams through the use of the specific EDE and EPE quantities. Since EDE is most often related to the electrical energy and not the thermal energy, it is necessary to track this quantity as a separate characteristic of a material due to the variety of thermal efficiencies that may exist in a fuel cycle scenario. On the other hand, the thermal efficiency that will be applied to the utilization of the EPE is generally not known (or may even be varied). Therefore, this quantity need not be tracked as a separate characteristic of a material since it is directly related to the quantity of actinides in the material, a quantity which is generally tracked already.

Collaborations were established with Argonne National Laboratory (ANL) in order to align the goals of this project with the related research program underway in support of the AFCI project including Yucca Mountain repository modeling and fuel cycle scenario modeling with DANESS and DYMOND. Connections were also established with the SINEMA project being pursued under laboratory funding at Idaho National Laboratory. While the proposal for this project originally considered DANESS as the platform for implementing many of these developments, DYMOND, VISION, SINEMA or other options may be necessary to provide the necessary flexibility. Currently, none of the fuel cycle system study tools allow for economic optimization under resource (e.g., repository space) constraints, although such capability is fundamental to this project. Therefore, interactions between this project and SINEMA (or DYMOND/DANESS/VISION) will include support for economic optimization.

Planned Activities

The next period will focus on the conceptual development of appropriate economic optimization capability with a variety of time-dependent constraints, including interaction with the SINEMA development effort and revisions to DANESS/DYMOND (ANL) and VISION Idaho National Laboratory (INL). With many of the technical specifications largely determined by intervening analyses at ANL and elsewhere, the focus will be on the development of generic optimization capability within these fuel cycle study systems, both internal decision making algorithms and external optimization wrappers. Of particular interest to this project is economic optimization under the constraint of fixed repository space; however, other combinations of objective functions and constraints will be of generic interest to all those engaged in fuel cycle studies. In addition, the development of specific economic models to simulate different policy scenarios will also be important. Increased involvement of economists with specific expertise in dynamic systems modeling will assist in generating these models.

Radiation Stability of Candidate Materials for Advanced Fuel Cycles

PI: Todd Allen, James Blanchard, University of Wisconsin-Madison

Collaborators: None

Project Number: 06-007

Project Start Date: March 2006

Project End Date: March 2009

Project Description

Three types of advanced fuels play important roles in the Advanced Fuel Cycle Initiative (AFCI). These include inert matrix fuels (IMF) for reducing actinide inventories using light water reactors, coated particle fuels such as the TRISO fuel design being pursued for the very high temperature gas-cooled reactor, and ceramic-ceramic composites being pursued for the gas-cooled fast reactor. These candidate matrix materials are currently being irradiated in the Advanced Test Reactor and are slated for irradiation in the PHENIX fast test reactor. However, the limited neutron exposure and irradiation temperatures will be insufficient to fully characterize their microstructural stability.

This project will use proton irradiation to characterize the microstructural stability of ceramics being considered as matrix material for the advanced fuels. The researchers will use proton irradiation at temperatures between 600-900°C to understand the effect of radiation on lattice stability, phase change, void growth, and other microstructural features. They will also examine the effect of radiation on hardness and fracture toughness. Structural analysis will be performed to determine limiting performance and identify promising candidate materials. Numerous materials, including carbides, nitrides, and oxides, will be included in the test plan.

Workscope

The following tasks comprise the main elements of this project:

- Complete final selection of carbide, nitride, and IMF matrix materials to form basis of experimental program, coordinated with AFCI program neutron irradiations
- Characterize unirradiated microstructures using optical microscopy, SEM, XRD, and TEM
- Measure crack growth in unirradiated materials following Vicker's indentation
- Irradiate samples at temperatures between 600°C and 900°C to 3 dpa and analyze material properties to estimate change in fracture toughness

NUCLEAR ENERGY RESEARCH INITIATIVE

Solution-Based Synthesis of Nitride Fuels

PI: Ken Czerwinski, Thomas Hartmann,
University of Nevada, Las Vegas

Collaborators: Los Alamos National Laboratory,
Savannah River National Laboratory

Project Number: 06-012

Project Start Date: March 2006

Project End Date: March 2008

Project Description

The synthesis of nitride fuels for advanced reactor designs entails carbothermic reduction from the oxide to the nitride. This process step is based on solid-phase reactions involving a stepwise process from the metal oxide, to the carbide, and finally the nitride. This high-temperature, solid-phase synthesis process is plagued by impurities in the final nitride product and difficulties in production, creating major drawbacks in using nitride fuels for advanced reactor designs. If the nitride could be synthesized directly by a solution route, then the impurities and other synthesis problems could be eliminated, or at least minimized. The proposed solution route to nitride would also have the added benefit of providing several adjustable parameters that would allow control over the properties of the final solid product.

The objective of the proposed project is to develop a solution-phase process for synthesizing actinide nitrides for use in nuclear fuels. Recent work by the Los Alamos National Laboratory collaborators has investigated amido reactions in non-aqueous solvents. The non-aqueous synthetic route based on amido chemistry potentially provides property control over the nitride product similar to the sol-gel methods for actinide oxides. The resulting nitride product should be free of the impurities inherent in the carbothermic reduction technique.

Workscope

The project is divided into three primary tasks as outlined below:

- Perform non-aqueous coordination chemistry, involving development of amido coordination with uranium, neptunium, and transneptunium actinides
- Develop and characterize uranium nitride, neptunium nitride, and transneptunium nitrides from amido species
- Develop methods for the synthesis of actinide nitrides:
    - Identify crucial parameter of the solution-based synthesis of uranium nitride, neptunium nitride, and transneptunium nitride
    - Compare solution synthesis for uranium and neptunium nitride and initial identification of required methods for solid-solution synthesis
    - Perform experiments on the synthesis of uranium-neptunium solid solutions if large variations are identified
    - Compare solution synthesis methods for actinide nitride and identify required methods for solid-solution synthesis with different actinides

NUCLEAR ENERGY RESEARCH INITIATIVE

The Development and Production of Functionally Graded Composite for Lead-Bismuth Service

PI: Ronald G. Ballinger, Massachusetts Institute of Technology

Collaborators: Los Alamos National Laboratory

Project Number: 06-038

Project Start Date: April 2006

Project End Date: April 2009

Project Description

The use of lead and lead-bismuth eutectic as a coolant for advanced lead-cooled fast reactor systems and transmutation designs has been limited by the corrosive nature of the coolant on fuel cladding or structural materials. The existing upper limit of approximately 550°C for operation of these systems is insufficient. A new series of Fe-Cr-Si alloys have been developed that provide both a protective film over a wide range of oxygen potentials and, at the same time, provide minimal solubility in liquid metals, which may occur in crevices or other oxygen-depleted areas. An Fe-12Cr-2.55Si alloy has demonstrated a resistance to corrosion in Pb/Pb-Bi eutectic that will allow operation of materials at temperatures up to 700°C. Initial testing indicates resistance to stress corrosion cracking degradation in supercritical water systems. The formation of a protective dual oxide (Cr-based/Si-based) layer provides a high degree of protection.

The purpose of this project is to produce a system that meets both the corrosion and structural requirements of liquid Pb and supercritical water systems at temperatures up to 700°C and, at the same time, be suitable for applications in a neutron flux environment. The clad alloy can be fabricated in the form of welding wire that can then be used as an overlay for either an extrusion billet (for tubing/piping/cladding production) or as an overlay for more complex shapes. Functionally graded composites consisting of a corrosion-resistant layer on a structural alloy will be produced in two forms of tubing suitable for either piping or fuel cladding applications. Two structural alloy systems will be used for each product form: 1) quenched and tempered and 2) oxide dispersion strengthened (ODS), which will be fabricated using standard commercial practice. The materials produced will be tested for corrosion and structural properties.

Workscope

The following activities comprise the primary workscope of this project:

- Tubing production including overlay wire production, Q&T-based tubing production and weld overlay application, and ODS tubing production and weld overlay application
- Corrosion and microstructural characterization testing, analysis, and modeling including overlay wire, cladding product, and pipe/tubing product
- Mechanical properties testing

NUCLEAR ENERGY RESEARCH INITIATIVE

Flexible Conversion Ratio Fast Reactor Systems Evaluation

PI: Neil E. Todreas, Pavel Hejzlar, Massachusetts Institute of Technology

Collaborators: None

Project Number: 06-040

Project Start Date: March 2006

Project End Date: March 2008

Project Description

The objective of this project is to develop a feasible design for a flexible conversion ratio fast reactor system that will enable time-dependent management of fissile inventories and higher actinides. The goal is to produce a single reactor design capable of being configured to achieve 1) conversion ratios near zero in order to transmute legacy waste and 2) conversion ratios near unity such that the reactor can operate in a sustainable closed cycle. Researchers will develop two candidate liquid coolant core designs—lead and liquid salt—and perform comparisons to find the most promising approach. These designs will also be cross-compared against two other reactor designs—supercritical carbon dioxide and sodium-cooled reactors.

A consistent plant rating of 2,400 MWt will be used in both designs and the reactor cores will be cooled by forced circulation during normal operation to achieve high power density of at least 100 kilowatts/liter. To be more economically competitive, researchers will select power conversion systems that eliminate the intermediate heat transfer loop. Safety and core integrity will be achieved through a self-controllable reactor design that ensures safe shutdown for all key transients. Proliferation resistance will be enhanced by eliminating blankets and using transuranics as fuel without separating plutonium.

The outcome of this project will be lead alloy and liquid salt fast reactor core designs with flexible conversion ratios. These designs, coupled with the results of another NERI project on gas-cooled reactors being performed at MIT and work at the Argonne National Laboratory, will allow decision makers to consistently compare various large, economical, high-power density fast reactor concepts. Ultimately, this will help in the selection of the most attractive system for closed fuel cycles in countries with fuel service centers.

Workscope

The following tasks will be performed:

- Develop lead alloy-cooled reactor designs with unity and zero conversion cores
- Develop liquid salt-cooled reactor designs with unity and zero conversion cores
- Cross compare the various designs by assembling data for sodium-cooled and gas-cooled fast reactors and Comparing sodium-, gas-, lead alloy-, and liquid salt-cooled reactors

NUCLEAR ENERGY RESEARCH INITIATIVE

Development and Utilization of Mathematical Optimization in Advanced Fuel Cycle Systems Analysis

PI: Paul J. Turinsky, North Carolina State University

Collaborators: Argonne National Laboratory

Project Number: 06-047

Project Start Date: March 2006

Project End Date: March 2009

Project Description

This project will develop a model with the capability to mathematically optimize deployment strategies for reactor and fuel cycle facilities. The model will consider economic, energy, environmental, and nonproliferation resistance metrics of the fuel cycle based on a stochastic optimization approach.

The DYMOND/VISION code will be employed to model various fuel cycle scenarios of interest. Optimization will be completed over a user-specified planning horizon to allow proper treatment of the future impact of current decisions. The system will be capable of optimizing such factors as fuel enrichment, fabrication, power generation, interim fuel storage, and reprocessing/separation deployment strategies, subject to constraints such as isotopic mass availability and energy generation demand. These capabilities will assure objectivity with minimal human intervention, thereby providing higher confidence when utilizing the results in policy decision making.

The goal of this work is to develop a multi-objective mathematical optimization approach that will provide quantitative data to decision makers on such items as the cost of margin (for example, the relationship of repository thermal margin to energy costs). This data can be used to establish policies concerning incentives and requirements to encourage the evolution of the preferred commercial nuclear reactor, fuel, and fuel cycle facilities.

Workscope

This project will encompass the following primary tasks:

- Specify objective functions, constraints, and decision variables to be considered
- Assess coupling approaches to add optimization capability to DYMOND/VISION
- Modify DYMOND/VISION as required to support the optimization engine
- Develop single-objective optimization capability using Simulated Annealing approach
- Exercise the optimization capabilities that have been developed
- Develop single- and multi-objective optimization capability using the Genetic Algorithm approach
- Introduce parallel processing capability for the Genetic Algorithm approach and integrate the optimization engine within DYMOND/VISION, if possible

NUCLEAR ENERGY RESEARCH INITIATIVE

Engineered Materials for Cesium and Strontium Storage

PI: Sean M. McDeavitt, Purdue University

Collaborators: None

Project Number: 06-058

Project Start Date: March 2006

Project End Date: March 2009

Project Description

The mission of the Advanced Fuel Cycle Initiative (AFCI) is to develop next generation recycling methods for spent nuclear fuel that will enable sustainable nuclear energy production. Closing the nuclear fuel cycle requires reprocessing spent fuel to recover the long-lived components that still have useful energy content, while immobilizing the waste fission products in stable forms.

While the primary AFCI focus is on actinide recovery and recycle, the disposition of fission product waste also requires the development of innovative solutions. Idaho National Laboratory (INL) is currently leading the effort to engineer processing technology to immobilize cesium (Cs) and strontium (Sr) isotopes using a fluidized bed steam reformer.

The objective of this NERI project is to investigate the fundamental science behind the steam reforming process while generating candidate Cs/Sr storage materials and characterizing their behavior. This will complement the development of large-scale steam reformer at INL. The chemical precursors to the reforming system will be designed and tested to produce ceramic storage from candidates for evaluation. The ceramic products will be characterized to quantify their leach resistance, phase structure, and thermophysical properties. The end result will provide a design basis for large-scale process definition.

As the majority of the radiation and decay heat in fission product waste comes from Cs-137 and Sr-90, isolating these products for short-term decay storage (~150 years) eases the design requirements on long-term repository disposal. It is notable that a small fraction of the fission products yttrium, barium, and rubidium tend to be extracted by the Cs/Sr separation schemes, so the immobilization host must also accommodate minor levels of these isotopes.

Workscope

This project consists of the following two major tasks:

- *Synthesize candidate ceramics for Cs and Sr storage.* This includes experiments to define process variables, feed compositions, and the product form.
- *Characterize the Cs/Sr-bearing ceramic products.* This task includes the characterization of critical behavior and attributes of the candidate compounds, such as Cs/Sr leach resistance, ceramic phase structures, and thermophysical properties.

This task will use a lab-scale steam reformer to generate a variety of Cs/Sr bearing materials (Figure 1).

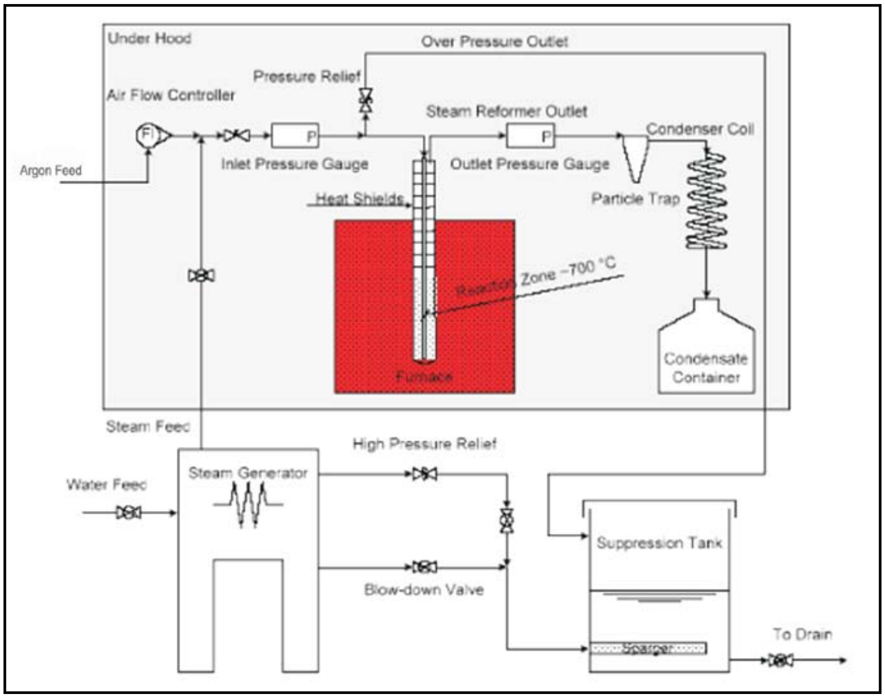


Figure 1. Schematic diagram of the Purdue University steam reformer for the fabrication of candidate Cs/Sr storage forms.

The initial list of candidate immobilization forms includes stable aluminosilicates as well as aluminate, silicate, titanate, zirconate, and hafnate ceramics (e.g., Chabazite, Cs_2ZrO_3, Cs_2SiO_3, $SrAl_2O_4$, $SrTiO_3$, $SrZrO_3$, $SrHfO_3$ as well as more complex ceramics). Producing these ceramics will require precursors such as chlorides or organometallics which can introduce Al, Si, Ti, Zr, or Hf into the fluidized bed steam reforming reaction chamber. As the Cs/Sr precursors and some of the candidate reactants are either liquids or solids with a volatile response to high temperature, multiple feed lines may be required to prevent reactions before entering the reaction vessel.

Feasibility of Recycling Plutonium and Minor Actinides in Light Water Reactors Using Hydride Fuel

PI: Ehud Greenspan, University of California, Berkeley

Collaborators: Massachusetts Institute of Technology, Argonne National Laboratory, University of Tokyo

Project Number: 06-065

Project Start Date: March 2006

Project End Date: March 2008

Project Description

This project is designed to assess the feasibility of improving the plutonium (Pu) and minor actinide recycling capabilities of pressurized water reactors (PWRs) by using hydride instead of oxide fuels. Use of hydride instead of oxide fuel is expected to offer the following improvements in PWR performance:

- Smaller inventory of Pu per core to provide the same cycle length with lower fuel cost
- Higher discharge burnup thereby reducing the amount of heavy metal inventory that must be handled or disposed of per unit energy generated
- Incineration of a larger fraction of the loaded Pu in one pass through the core thereby reducing the Pu inventory faster, decreasing the amount of actinides in the waste streams, and improving the economics of Pu incineration
- Smaller fissile Pu fraction in the discharged fuel, higher spontaneous neutron yield, and higher decay heat per gram of Pu and per gram of transuranic (TRU) thereby improving proliferation resistance of discharged fuel
- Somewhat smaller neutron source strength and decay heat per fuel assembly due to reduced TRU amount resulting in simplified handling of discharged fuel assemblies
- Unique prompt negative reactivity insertion mechanism due to spectrum hardening as a result of fuel temperature increase thereby enhancing PWR safety

In addition, there appears to be a good synergy between hydride fuel and the pyro-chemical separation process thereby improving the economics and proliferation resistance of recycling in PWRs.

Workscope

There will be four major parts to this project:

- Identify promising hydride fuel assembly designs for recycling Pu and minor actinides in PWRs including zirconium-hydride based; thorium-hydride based; and composite U, Th-hydride, Zr-hydride fuel.
- Conduct a comprehensive system analysis to identify the best recycling options by comparing fuel cycle characteristics of the most promising hydride fuel assembly designs to the fuel cycle characteristics of oxide or inert matrix fuel assemblies.
- Perform a comprehensive safety analysis of PWR cores loaded with hydride fuel to evaluate whether the design is licensable.
- Assess the compatibility of hydride fuel with water and cladding materials at typical PWR operating conditions.

NUCLEAR ENERGY RESEARCH INITIATIVE

Accelerator-Based Study of Irradiation Creep of Pyrolytic Carbon Used in TRISO Fuel Particles for the Very High Temperature Reactors (VHTR)

PI: Lumin Wang and Gary S. Was, University of Michigan

Collaborators: Oak Ridge National Laboratory; Idaho National Laboratory

Project Number: 06-113

Project Start Date: March 2006

Project End Date: March 2009

Project Description

Pyrolytic carbon (PyC) is one of the structural materials in the TRISO fuel particles which will be used in the next generation of gas-cooled very-high-temperature reactors. When the TRISO particles are under irradiation, creep of the pyrocarbon layers can cause radial cracking leading to catastrophic particle failure. Therefore, a fundamental understanding of the creep behavior of PyC during irradiation is required to predict the overall fuel performance.

The primary objective of this project is to characterize the creep behavior of PyC through a systematic program of accelerator-based proton irradiation and in-situ measurements under stress at various temperatures between 400°C and 1,200°C. Test data will be analyzed to determine creep coefficients, which will then be correlated to existing coefficients measured under neutron irradiation. In addition, initial experiments on the transport of select fission products (e.g., Ag and Sr) in PyC under irradiation and stress will be conducted by implanting ions into the sample surface. The PyC microstructure will be studied with advanced analytical transmission electron microscopy (TEM).

Workscope

The project will perform the following activities:

- Modify the ion accelerator target chamber and test overall system design and operation (temperature control and creep measurement during proton irradiation)
- Characterize PyC samples
- Conduct low-temperature irradiation (400°C and 600°C) on the reference microstructure (unloaded) and perform data analyses
- Conduct low-temperature irradiation on the microstructure under stress and perform data analyses
- Conduct high-temperature irradiation (800°C, 1,000°C, and 1,200°C) and perform data analyses
- Perform irradiation of samples with pre-implanted fission product and cross-sectional TEM analyses
- Conduct final data analyses and empirical modeling

NUCLEAR ENERGY RESEARCH INITIATIVE

Development of Acetic Acid Removal Technology for the UREX+ Process

PI: Robert M. Counce, James S. Watson, University of Tennessee

Collaborators: Oak Ridge National Laboratory

Project Number: 06-116

Project Start Date: March 2006

Project End Date: March 2009

Project Description

The focus of this proposal is to select and scientifically verify the most appropriate technology to separate acetic acid from the strip solution of the UREX+ process. Specifically, this technology will separate the acetic acid and recover nitric acid so that it can be recycled and reused in the process. Leading technologies will be screened on the basis of rudimentary estimates, including economics, safety, development time, health, and environmental metrics. The candidate technologies will include distillation, extraction, crystallization, and chemical reaction. Flowsheets will be developed for the two best technologies selected in the initial screening studies. These technologies will be subjected to more rigorous comparison based on the above metrics. A bench-scale evaluation of the most appropriate technology will demonstrate process viability. Reports for each phase will document the development activities and provide a plan for applying this technology.

Workscope

Following is a summary of the principal tasks involved in this project:

- Selection and screening of leading technologies
- In-depth analysis of leading technology including development of flowsheets
- Experimental verification of selected technology

NUCLEAR ENERGY RESEARCH INITIATIVE

Separation of Nuclear Fuel Surrogates from Silicon Carbide Inert Matrix

PI: Ronald Baney and James Tulenko,
University of Florida

Collaborators: None

Project Number: 06-126

Project Start Date: March 2006

Project End Date: March 2008

Project Description

Silicon carbide has become one of the prime candidates for inert matrix nuclear fuel materials for burning plutonium or for the transmutation of long-lived actinides. Studies carried out by the University of Florida have shown that this material holds excellent neutronic and thermal performance capabilities. A silicon carbide matrix fuel (IMF) may run with a centerline temperature that is only a few hundred degrees above the coolant temperature. Research supported worldwide has focused on silicon carbide as a suitable matrix for reprocessing spent fuel. However, techniques for processing and separating transuranic species and unspent fuel from a silicon carbide matrix have not been expertly identified.

This research seeks to fill this gap in knowledge by investigating novel techniques for separating surrogate materials from silicon carbide. Ceria will be used as a surrogate material for plutonium oxide. This work will establish a protocol for the dissolution and separation of silicon carbide from ceria using technologies compatible with traditional fuel handling. Researchers will conduct a thorough literature search of all processes capable of SiC separation, such as molten metals, low melting silicides, electronic industry etching processes of silicon carbide, and super critical water. The two most promising processes will be selected through screening criteria that take into account economics as well as adaptability to existing reprocessing technologies. Materials containing two different levels of CeO_2 will be processed by the selected process will be processed using dissolution and separation techniques that will co-dissolve processed samples with metal silicides. These dissolution techniques will be studied for ways of separating the matrix from urania and ceria. Ceria and other surrogate materials will be separated from the SiC matrix in a fashion comparable to methods reported in the literature. The resultant samples will be characterized for ease of separation, degree of safety (with regards to chemicals used and reaction products produced), and cost.

Workscope

The following primary tasks will be performed for this project:

- Examine literature for published reclamation and recovery processing of silicon carbide/metal and select the most viable methods according to defined screening criteria (e.g., processing complexity, economics, ease of implementation with current and/or potential processing equipment, and safety)
- Construct or purchase processing equipment
- Prepare two ceria/silicon carbide ratio compositions by hot pressing
- Evaluate the selected dissolution processing methods for ease and rate of dissolution of the SiC/CeO_2
- Develop processes for phase separation of CeO_2 from the dissolution process stream
- Recommend processes for further optimization and scaling

NUCLEAR ENERGY RESEARCH INITIATIVE

Enhancements to High Temperature In-Pile Thermocouple Performance

PI: John Crepeau, University of Idaho

Collaborators: Idaho National Laboratory

Project Number: 06-134

Project Start Date: March 2006

Project End Date: March 2008

Project Description

There are insufficient data to characterize the performance of new reactor materials in high temperature, radiation conditions. To evaluate candidate material performance, robust instrumentation is needed that can survive these conditions. Traditional methods for measuring temperature in-pile degrade at temperatures above 1,080°C or de-calibrate due to transmutation. During the last year, the Idaho National Laboratory launched an effort to develop temperature measurement methods suitable for long duration, high temperature in-pile testing. Initial results indicate that specialized alloy thermocouples fabricated from doped molybdenum/niobium-1 percent zirconium have the potential to provide the desired accuracy for long-duration tests. Although the performance of these thermocouples appears promising, there are several options that can potentially enhance their lifetime and reliability.

The major research objective of this project is to quantify the impact of candidate enhancements related to alloy materials, geometry, and fabrication techniques on thermocouple performance. Based on these evaluations, improved thermocouple designs will be fabricated and tested at high temperatures for long durations to quantify thermocouple accuracy, reliability, and lifetime. Ultimately, an optimized thermocouple design will be recommended for high temperature in-pile testing.

Workscope

This project comprises the following major tasks:

- Evaluate alternate alloys
    - Ductility and embrittlement assessments
    - Stability and thermal cycling demonstration tests
- Evaluate alternate geometry
    - Fabrication of alternate geometries
    - Stability and thermal cycling demonstration tests
- Evaluate alternate fabrication techniques
    - Fabrication of alternate geometries
    - Stability and thermal cycling demonstration tests
    - Optimized thermocouple design

NUCLEAR ENERGY RESEARCH INITIATIVE

Design and Development of Selective Extractants for An/Ln Separations

PI: Robert T. Paine, University of New Mexico

Collaborators: Washington State University, Idaho National Laboratory

Project Number: 06-137

Project Start Date: March 2006

Project End Date: March 2009

Project Description

This project is designed to remove transuranic elements from spent nuclear fuel for storage or for reuse in transmutation processes. The proposed method will develop an efficient aqueous separation scheme for 1) recovering Am and Cm from the acidic liquid remaining after UREX+2 processing, which is used to remove U, Pu, Np, and Cs/Sr, and 2) separating trivalent actinides from the fission product lanthanide ions. The project focuses on the continued development and optimization of the "NOPOPO ligands," which were previously demonstrated to be effective under laboratory-scale conditions, and which show promise as a large-scale process add-on to UREX+2.

The primary objectives of this project are to design, synthesize, and fully characterize the performance of 2,6-*bis*(phosphinomethyl)pyridine N,P,P´-trioxides (NOPOPO) as potential reagents for separating Am, Cm, and fission product lanthanides from other transuranics and fission products and for acting as a separations "platform" for the mutual separation of Am/Cm from the lanthanides. Preliminary analyses indicate that one member of the family of ligands, $(EtHx)_4$NOPOPO, offers improved separation of Am^{3+} and Ln^{3+} ions from acidic aqueous solutions compared to the CMPO ligand used in the TRUEX process. The research is expected to produce a best-case extractant compound, which will undergo a complete round of synthesis optimization and performance characterization with a realistic raffinate simulant representative of the UREX+ process.

Workscope

The specific tasks will include:

- Optimize the NOPOPO synthesis
- Conduct further extraction testing of $(EtHx)_4$NOPOPO
- Continue the study of phase compatibility and hydrolysis
- Characterize the radiolytic stability
- Design and synthesize new NOPOPO derivatives with improved solubility, phase compatibility, and stability characteristics
- Conduct extractive testing of the new derivatives to determine their relative efficacy
- Conduct focused testing on the best-case extractant compound to optimize synthesis and characterize performance with realistic waste solutions
- Perform a cost analysis for the new process

NUCLEAR ENERGY RESEARCH INITIATIVE

Microwave Processing of Simulated Advanced Nuclear Fuel Pellets

PI: David E. Clark and Diane Folz, Virginia Polytechnic Institute and State University

Collaborators: University of Tennessee, Los Alamos National Laboratory

Project Number: 06-141

Project Start Date: March 2006

Project End Date: March 2009

Project Description

The primary goal of this work is to sinter simulated nitride fuel pellets using microwave energy as an alternative method for manufacturing advanced nuclear fuel pellets. Researchers will characterize the sintered pellets, consisting of non-radioactive materials with properties similar to actual pellets, with respect to density and grain morphology. The research team will employ two different heating methods: a combination of microwave and conventional heating (microwave hybrid heating) and direct microwave sintering.

Work performed to date suggests that the overall sintering schedule for advanced nuclear fuel pellets could be modified to achieve the desired density (80 ± 10 percent theoretical density) without losing highly volatile critical components such as americium and dysprosium oxides and nitrides.

The following assessment metrics have been established:

- Demonstrate the ability to sinter simulated oxide and nitride fuel pellets to 85–95 percent theoretical density using microwave energy
- Demonstrate that sintered pellets lose less than 10 weight-percent dysprosium during the sintering process

Workscope

The primary objective of this work will be achieved through the following tasks:

- Fabricate green pellets of ZrO_2 for microwave heating (direct and hybrid) experiments
- Design and construct heating caskets/susceptors for microwave hybrid heating
- Bring on-line high-power microwave ovens for high-temperature sintering experiments
- Evaluate sintered pellets for density (Archimedes & pycnometry) and microstructure (XRD, SEM)
- Based on results of this work, recommend a path forward

Nuclear Hydrogen Initiative Projects

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

Project Number: 05-006

Project Start Date: April 2005

Project End Date: April 2008

Research Objectives

Of the numerous 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. However, because the S-I cycle involves complex, highly non-ideal phase behavior and reactions, many of the performance projections associated with this technology are based on uncertain and incomplete data. Thus, basic research on thermodynamic measurements and physical property models of this cycle are needed to successfully develop this technology for a future hydrogen economy.

This project will focus on three areas: thermodynamic measurements, physical property modeling, and process modeling. Researchers will use a unique integrated approach that will allow for highly efficient exploration and feedback among these focus areas. That is, they will use initial properties and process modeling development 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 most critical for minimizing remaining process uncertainties.

The results of this project will be transferred to the nuclear hydrogen community by process simulation of the SI cycle with reactive distillation of hydrogen iodide (HI).

Research Progress

Preliminary properties and process modeling work by the project team indicate that the HI decomposition section of the S-I cycle (see Figure 1), where mixtures of HI, I_2, and water are present at elevated temperatures and pressures, should be a priority for experimental measurements. To carry out this work, researchers have selected the continuous-flow apparatus (CFA) technique (see Figure 2), which has been previously used to measure complex phase behavior for a variety of systems at elevated temperatures and pressures. However, significant modifications to the existing CFA will be required, as mixtures of HI, I_2, and water present unique challenges, particularly in the area of corrosion. Therefore, a major task in this project to date has been to identify those aspects of the existing CFA that will require redesign and/or materials substitution.

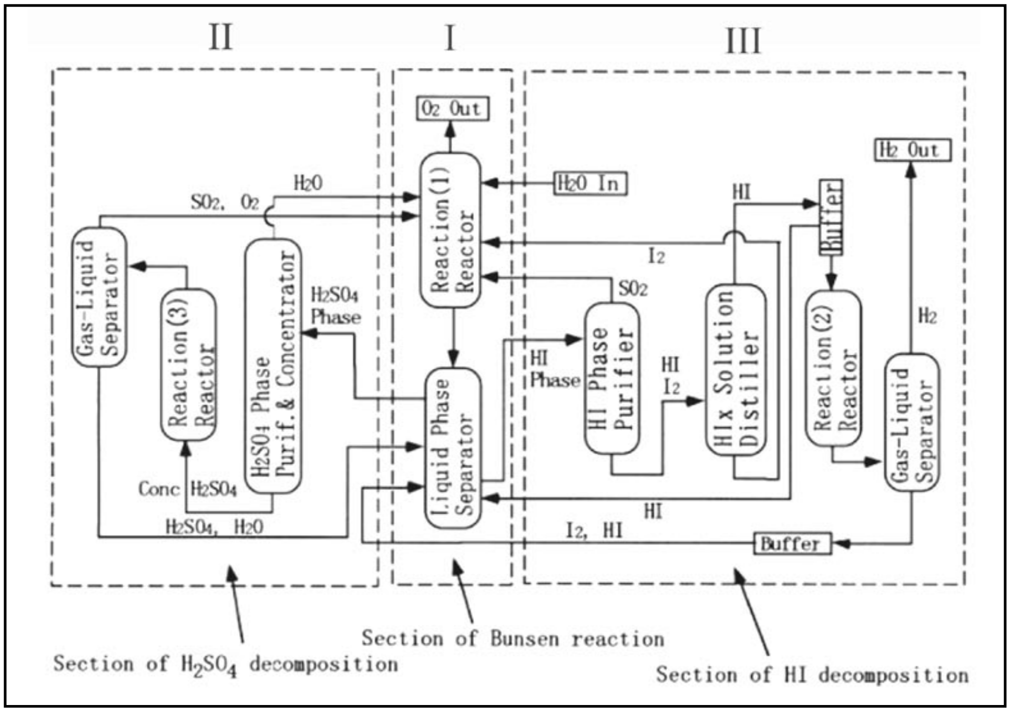


Figure 1. Simplified schematic (from Sakurai et al., 1999) of the S-I process.

Based on a corrosion study of $HI–I_2–H_2O$ mixtures, tantalum (Ta) and Ta alloys have been identified as appropriate materials of construction for all wetted surfaces at elevated temperature and pressure. However, tantalum does have its problems: 1) no vendors exist for the Ta high-pressure fittings (e.g., ferrules, glands, unions, and tees) required for assembling the CFA (fortunately, high-pressure Ta tubing is available); 2) Ta and its alloys are difficult to machine, requiring much more care and significantly longer times compared to steel; and 3) the cost of tantalum is high, about an order of magnitude higher than stainless steel. Nevertheless, the choice of tantalum was necessary for this work because of its excellent corrosion-resistance properties for the systems of interest.

Researchers are currently fabricating the high-pressure fittings for the CFA. They have conducted initial tests on the fabrication of three different types of tantalum fittings to evaluate their quality. Each tantalum fitting looks very similar to its steel counterpart, with little or no observed problems with threading, holes, or smoothed edges. Pressure testing of completed parts to 5,000 psi was also successful. The major concern with these fittings is the time and cost required to fabricate them. About 50 fittings are needed at a cost of $12,000 (with 75 percent attributed to labor) to manufacture the desired number of fittings. Machining of a Ta–2.5W block to make the wetted inner-chamber piece of the view cell (see Figure 2) has an estimated cost of $8,000. Fabrication of these parts is expected to get underway in early November 2005.

Initial properties and process modeling work by the project team indicate that measurement of liquid-liquid equilibria (LLE) for the I_2–water system at elevated temperature and pressure should be the first experimental priority. Therefore, researchers conducted tests to determine whether the interface of iodine and water at elevated temperatures could be observed in the view cell (see Figure 2), and, if not, what method of interface detection was suitable. Because molten iodine is highly colored, they were concerned that the interface between the iodine and water liquid layers would be difficult or impossible to see, as the mutual solubility of the components increased with temperature. Pyrex tubes containing iodine and water were sealed under vacuum and then heated in an oil bath to temperatures above 113°C, which is the normal melting point of iodine. At temperatures of approximately 150°C, the liquid-liquid interface between the iodine-rich and water-rich phases could not be observed using normal lighting. Fortunately, the interface between the I_2 and water layers could be easily observed with an infared detector. Researchers will therefore use this setup for their measurements.

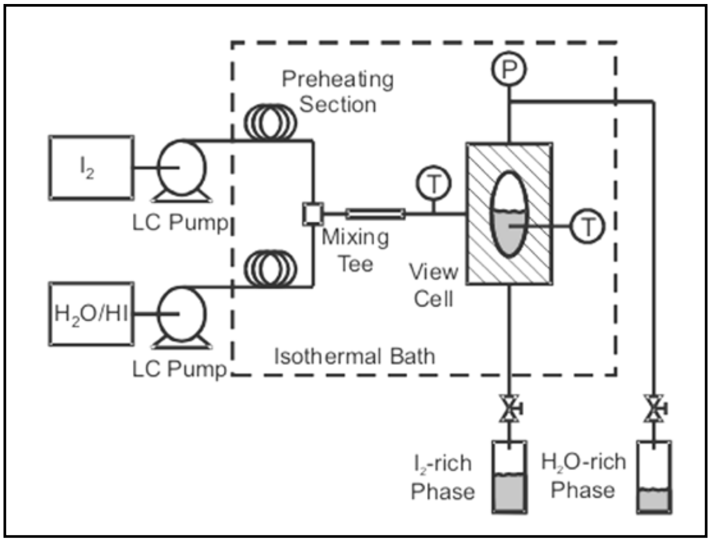


Figure 2. Schematic of continuous-flow apparatus.

The team received training on the use of Aspen Plus for process modeling, with an emphasis on the modeling of electrolyte systems, as these are a key part of the S-I cycle. The training described in detail the basis and results for Sections I-III of the S-I cycle (see Figure 1). In addition, the team conducted test evaluations with the Aspen Plus simulations of various sections of the S-I process to gain experience with the complexities of the simulation program and modeling. For example, a study of the effects of changing pressure on the reactive distillation column in Section III showed precisely where higher pressures resulted in better separation and significantly reduced energy requirements for the reboiler and condenser. Because the results were found to be sensitive to the modeling, they have provided initial direction for this experimental program.

As a way of building experience with the S-I cycle, researchers initiated thermodynamic analyses of the overall water-splitting process and of Sections II and III. The goal is to articulate a benchmark set of best-possible outcomes for reversible and essentially model-independent systems that can be used to compare proposed S-I processes with real equipment and irreversibilities. While many estimates of the efficiency of the S-I cycle have been calculated, they are often based on different idealizations, use energy requirements that do not reflect the real constraints of the S-I cycle, or include estimates from property models with significant uncertainties. This work is an attempt to standardize such comparisons. A review of this work was presented at the 2005 AIChE Annual Meeting in November 2005.

Planned Activities

A standard requirement for the measurement of quality experimental data is validating the experimental apparatus after its construction. Thus, in the winter of 2006, the researchers plan to use their continuous-flow apparatus to measure liquid-liquid equilibria for the benzene–water binary at temperatures similar to those to be used for I_2-water, that is, from 150 to 300°C. They have chosen this system because reliable data exist in the literature for comparison and mutual solubilities in each phase are similar to what is expected for I_2-water, so operating procedures should be similar. In the spring of 2006, the goal is to complete the fabrication of all Ta components, assemble the CFA with its new Ta components, and then carry out LLE measurements for the I_2-water system at temperatures up to the upper critical solution temperature of this system, estimated to occur at 300-350°C. By late summer 2006, researchers plan to begin work on the ternary HI-I_2-water system. The development of spectroscopic methods for determining the in situ chemical species (e.g., iodine complex concentrations) at elevated operating temperatures and pressures are also of interest, for both the binary and ternary systems mentioned above.

In spring 2006, researchers will complete work on the thermodynamic analysis of the overall water-splitting process and of Sections II and III (see Figure 1) of the S-I cycle. By summer 2006, they will complete the development of Aspen Plus property and process models for all streams in the current S-I process, including uncertainties due to experimental error and lack of data. These models will be used for sensitivity studies and to guide the selection of experimental conditions for the ternary HI-I_2-water system. Once this new experimental data is obtained, improvements can be made to the initial properties and process models for the S-I cycle. Finally, these updated process modeling results will be used to identify additional experiments most critical for minimizing remaining process uncertainties.

NUCLEAR ENERGY RESEARCH INITIATIVE

Silicon Carbide Ceramics for Compact Heat Exchangers

PI: Dennis C. Nagle, Johns Hopkins University

Collaborators: None

Project Number: 05-032

Project Start Date: April 2005

Project End Date: April 2008

Project Description

Generation IV nuclear reactors will require high-temperature hydrogen-to-helium heat exchangers capable of prolonged operation at temperatures over 900°C. Current block graphite heat exchanger materials cannot operate unprotected in this environment because of hydrogen gasification reactions that consume the graphite. The primary focus of this research project is to develop unique, impervious, net-shaped, silicon carbide materials for the next generation of nuclear power plants. Silicon carbide and other refractory carbides have very high thermal conductivities, and they are thermo-chemically stable at elevated temperatures. Therefore, they are excellent candidate materials for use in high-temperature gas heat exchangers.

Because traditional carbide processing techniques cannot produce silicon carbide with the complex geometries needed for nuclear energy heat exchanger designs, the major objective of this program is to establish an innovative and feasible way to fabricate fully dense and configurable silicon carbide (SiC) monoliths from carbon substrates that can be easily machined into desired shapes. Technologies developed in this project will have a direct impact on both new heat exchanger design and novel fabrication techniques for interconnections between metal and ceramic components in future nuclear reactors. Major tasks of this project are to:

- Establish a liquid silicon infiltration method for fabricating configurable SiC monoliths from carbon substrates
- Develop a glass-sealing technology crucial to eliminating residual porosity in the silicon carbide in order to achieve hermetic sealing in high-temperature hydrogen and helium atmospheres
- Study the micro-structures of prepared SiC, characterize the thermal and mechanical properties, and measure gas permeability
- Develop models to define the optimum channel configuration for SiC, including pressure drops and heat transfer rates related to the configuration and geometry
- Manufacture a heat exchanger prototype
- Test the developed heat exchanger to determine heat transfer capability and pressure drops
- Compare experimental data to model predictions

Research Progress

During the first year, researchers focused on materials synthesis, with the goal of establishing a proven method to design and produce a carbon matrix that can be converted into a fully dense silicon carbide monolith without deformation during the liquid silicon infiltration process. Research activities for this task were as follows:

- Prepare and characterize monolithic carbon pre-forms derived from microcrystalline cellulose and other polymeric precursors with desired density and micro-structural characteristics
- Create and study monolithic SiC of various microstructures and densities formed via Liquid Silicon Infiltration (LSI) techniques with desired volumetric and dimensional characteristics
- Fine-tune LSI techniques to allow complete and efficient conversion to fully dense SiC

In order to create high-quality, impervious, net-shaped SiC heat exchanger materials with liquid silicon infiltration, researchers need to develop carbon pre-forms with low density and high porosity. Initially, they employed microcrystalline cellulose as the carbon precursor. However, carbon derived from microcrystalline cellulose has a density of between 0.6 to 0.7 g/cm^3, which is much lower than the ideal carbon density of 0.96 g/cm^3 for the formation of fully dense silicon carbide.

Microcrystalline Cellulose/Phenolic Resin Mass Ratio	Carbon Substrate Apparent Density (g/cc)	Silicon Carbide Sample Apparent Density (g/cc)^A
100:0	0.60	
70:30	0.760	2.69
65:35	0.739	2.76
60:40	0.846	2.91
55:45	0.777	2.70
0:100	1.42	

A: Theoretical density of silicon carbide is 3.2g/cc.

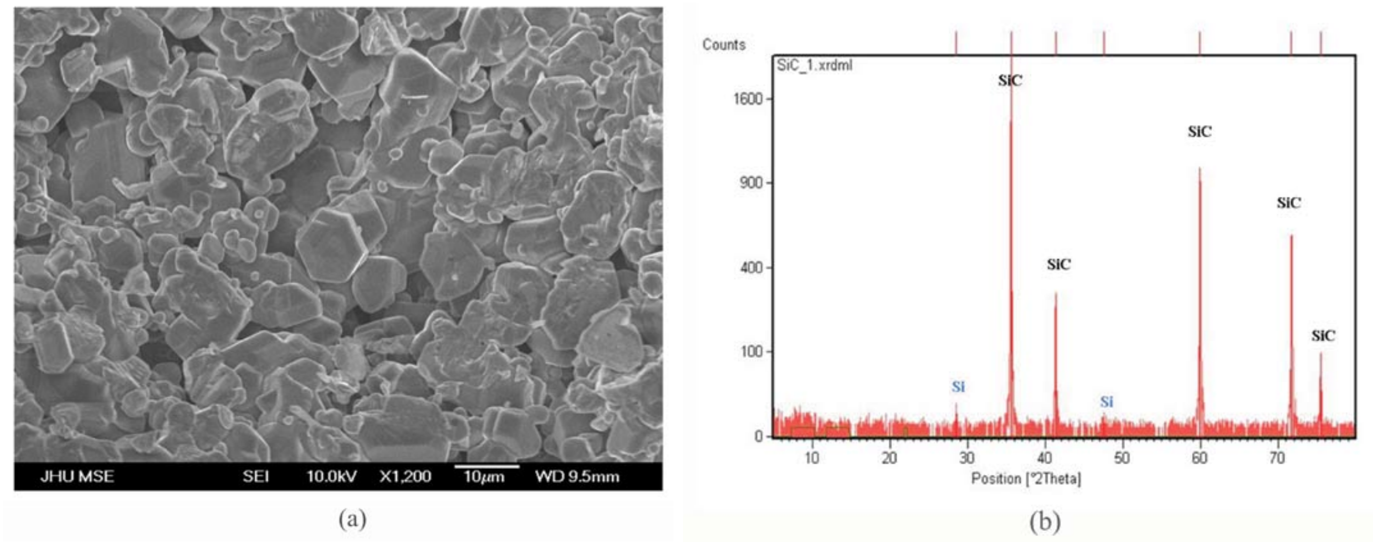
Table 1. Silicon carbide samples prepared from carbon substrates with a liquid silicon infiltration method from carbon substrates derived from cellulose/phenolic resin composites.

To adjust the apparent density and porous structure of carbon matrix to achieve the best infiltration results, the team employed mixtures of microcrystalline cellulose and phenolic resin as precursors to form carbons with the desired structure and density. Carbon derived from phenolic resin has a density of about 1.5 g/cm$_2$; therefore, carbon prepared from a mixture of microcrystalline cellulose and phenolic resin with the appropriate mass ratio would give the desired density.

For the carbonization process, researchers adopted a hybrid procedure of low-temperature atmospheric pyrolysis followed by high-temperature carbonization in argon to obtain carbons with micron-scale porosity in order to achieve effective liquid silicon infiltration and carbide formation.

In the silicon infiltration process, carbon disks were placed in graphite crucibles together with a thin layer of fine silicon powder (325 mesh) on each side of the disk (upper and lower). These crucibles were then placed in a high-temperature graphite furnace at temperatures of about 1,700ºC for one hour under vacuum to enhance the liquid silicon metal infiltration process and silicon carbide conversion. After studying the samples produced by this process, researchers discovered that silicon carbide disks with apparent density of up to 90 percent can be achieved with just one infiltration cycle on carbon pre-forms produced with this technique (Table 1). Figures 1 (a) and (b) show the SEM image and XRD pattern of the 60:40 SiC sample.

Together with the liquid phase infiltration method developed on this project, the researchers are also studying a low-temperature gas-solid reaction approach for converting carbon into refractory carbides. The basis of this approach is that carbide-forming elements, such as silicon, hafnium, zirconium, and niobium, can be transported as reactive gaseous species onto carbon surfaces and react to form carbides at temperatures much lower that those required for solid/solid carbide formation reactions. When a high vapor pressure, high valence metal compound such as $SiCl_4$ is placed together with silicon metal at an elevated temperature, equilibrium is established between the metal and the gas and an intermediate low valence gaseous species is produced.



(a) (b)

Figure 1. (a) SEM image of silicon carbide prepared from carbon derived from cellulose/phenolic resin composite with a ratio of 60:40, (b) X-ray powder diffraction pattern showing silicon carbide as the major phase.

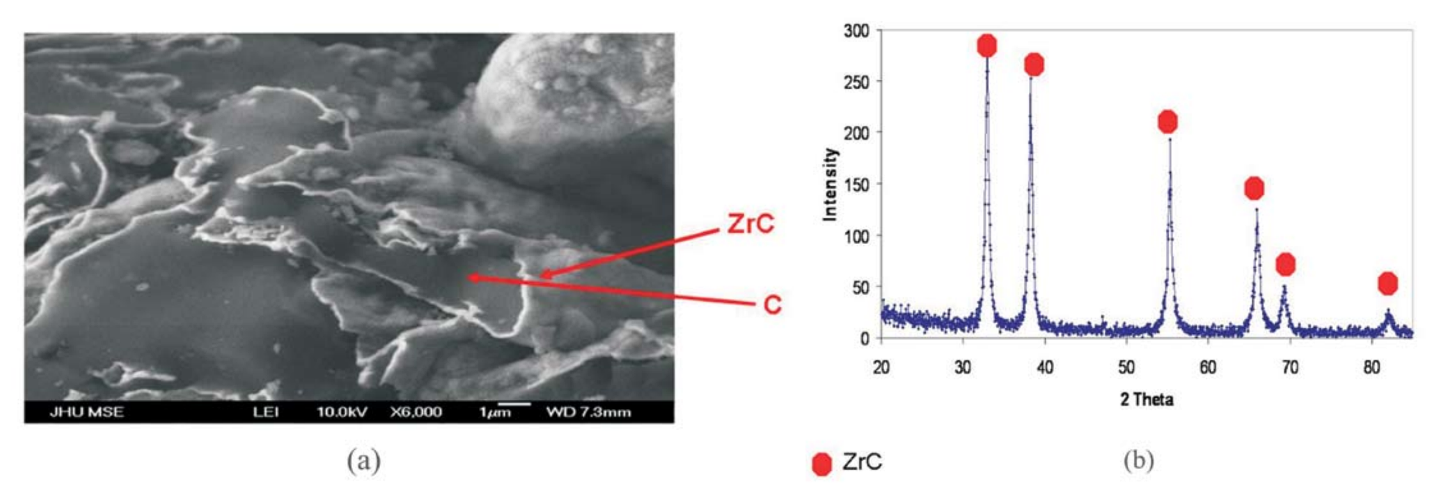


Figure 2. (a) Zirconium carbide coating on carbon prepared with a gas-solid phase method, (b) X-Ray pattern identifying zirconium carbide as major phase.

$$SiCl_4(gas) + Si\ (solid) \rightarrow 2SiCl_2{}^*(gas) \qquad (1)$$

The transient species $SiCl_2{}^*$ is a meta-stable compound and can only exist in low concentration. When this intermediate compound reaches the carbon surface, it will dissociate into Si and $SiCl_4$ and simultaneously react with carbon to form carbide:

$$2SiCl_2{}^*(gas) + C \rightarrow SiC + SiCl_4$$

Based on this mechanism, the researchers have studied gas/solid phase reactions of metal/carbon systems, including Hf-C, Zr-C, Si-C, and Nb-C, in both sealed high-temperature reactors and in high-temperature molten salt baths at temperatures between 800 to 1,000°C. They have successfully prepared HfC, ZrC, NbC, and SiC coatings and/or nanoparticles. Figures 2 (a) and (b) show the SEM image and XRD pattern of the ZrC sample

The importance of this gas-solid phase carbide formation technique to this research project is twofold. First, gas phase carbon-to-carbide conversion can be an effective method in eliminating both micron-sized and nano-scale porosities in SiC monoliths produced with the liquid phase method. Secondly, the vapor phase can enter smaller pores more effectively than the liquid metal. This will be achieved by first depositing carbon inside these pores with gas phase hydrocarbon decomposition/infiltration followed by gas phase carbide conversion. Secondly, dense carbide coatings/films can be formed on carbon surfaces, which provides a possible mechanism of active sealing/binding at high temperatures.

Additional progress achieved in this project include preliminary theoretical modeling of micro-scale heat exchanger design and building of a hermeticity testing facility based on a gas chromatography unit. Current research activities are focused on the detailed analysis of carbonization process of cellulose and phenolic resin precursors in order to gain a better understanding and control over the formation of carbon substrates with optimum microstructure.

Planned Activities

Based on progress achieved during last year, researchers are planning the following activities for further study:

- Continue optimization of liquid silicon infiltration techniques to increase density and homogeneity of prepared silicon carbide. This task involves more detailed studies on both the carbonization process and the infiltration process.
- Study the effectiveness of gas-solid phase carbide conversion method to further reduce the porosity of silicon carbide prepared from liquid phase infiltration methods.
- Develop glass sealing techniques to improve the hermeticity of silicon carbide.
- Evaluate mechanical properties and thermal conductivity of prepared silicon carbide at the operating temperatures of nuclear energy heat exchangers (900 to 1,000°C).
- Attempt to fabricate silicon carbide with larger dimensions (both in width and in thickness) and with complex shapes/configurations (curves, micro-channels).
- Model the heat exchange process and study silicon carbide heat exchanger design.

NUCLEAR ENERGY RESEARCH INITIATIVE

Molten Salt Heat Transport Loop: Materials Corrosion and Heat Transfer Phenomena

PI: Kumar Sridharan, University of Wisconsin-Madison

Collaborators: None

Project Number: 05-154

Project Start Date: April 2005

Project End Date: March 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 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. In summary, the research objectives of this project are: 1) to design, fabricate, and operate molten fluoride salt capsules in a flow loop under prototypic NGNP conditions using appropriate geometric scaling; 2) to 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; and 3) to document the observed corrosion effects and heat transport performance in integral and separate-effects databases.

Research Progress

Researchers began this project by developing an in-depth understanding of various issues pertaining to corrosion testing in a molten salt environment, in particular the safety and salt purity issues. They created a thermodynamic database of all relevant fluorides of cations in both the materials and the salts for the prediction of corrosion of alloying constituents in molten fluoride salt environments. The researchers began developing the experimental hardware. They finalized a design for a static corrosion cell to evaluate corrosion performance of candidate alloys for use in a molten salt environment. They conducted initial 1,000°C/500 hour dry tests (without salt or sample) to establish the integrity of the outer stainless steel capsule. These tests were followed by the examination of crack growth and the oxidation at the welds under the scanning electron microscope (SEM).

Steps were taken to procure additional materials and equipment needed for this project. The research team placed an order for the procurement of an atmosphere-controlled, high-temperature furnace (up to 1,200°C) to carry out the high-temperature corrosion tests. They also ordered high-purity FliNaK salt (46.5LiF-11.5NaF-42KF mol percent) from Electrochemical Systems, Inc., and a smaller trial batch of FliNaK from Idaho National Laboratory. Finally, the team procured six candidate, mostly code-certified alloys for initial corrosion testing. Sectioning of the samples to the appropriate size has been completed and surface preparation and base material has been initiated.

Researchers initiated studies on heat transfer modeling in molten salts that will aid in the construction of the molten salt corrosion loop for heat and mass transfer studies. These heat transfer calculations will provide a technical basis for the construction of the molten salt heat transfer loop. For the design of the molten salt loop, a thermal-convective code originally developed to study natural convection of supercritical water has been modified and applied to the molten FliNaK medium. The fluid and thermal properties of FLiNaK were input into the code to allow for this modeling. Since the heat capacity value was valid for only a small temperature range above the melting point of FliNaK, researchers compared their values with established ones. They also compared values for thermal conductivity and found these numbers to be in close agreement with those reported by Williams et al., at Oak Ridge National Laboratory. A graphical comparison of the different reported values of thermal conductivities are shown in Figure 1.

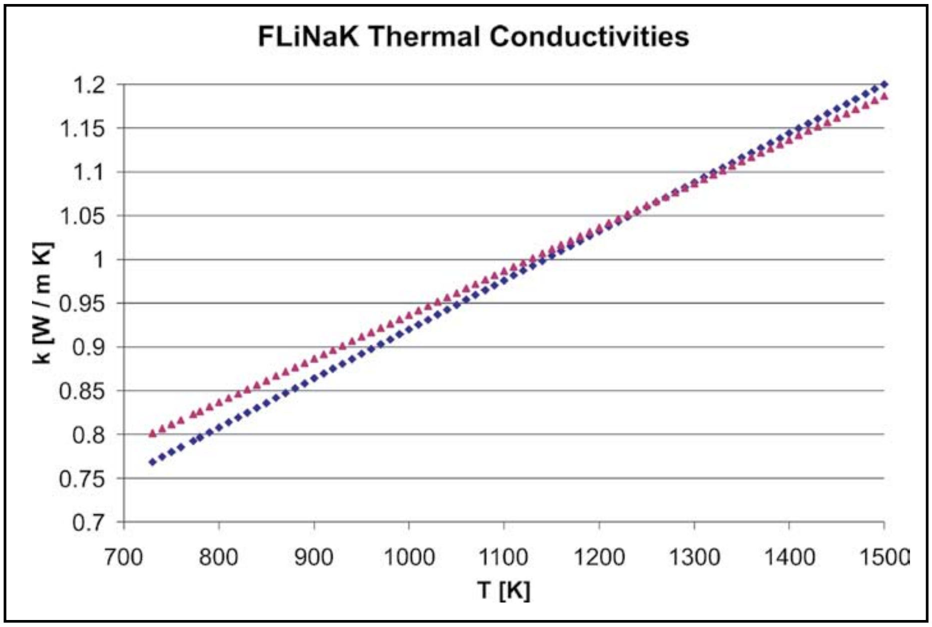


Figure 1. The thermal conductivity used in modeling FLiNaK follows closely to the correlation by Russian researchers, as reported by Williams.

Planned Activities

The first phase of the experimental plan involves testing in static corrosion capsules at 1,000°C for 500 hours. The initial evaluation will be performed on six candidate alloys with their selection based on high-temperature strength, air oxidation resistance, code certification status, and previous studies. The alloys include Hastelloy-X, Hastelloy-N, Haynes-230, Ni-200, Incoloy-800H, and Nb-1Zr. The testing will be performed in FLiNaK salt because it is representative of a class of fluoride salts being considered as heat transfer media. FLiNaK has a melting point of 454°C and a boiling point of 1,570°C. To avoid cross-contamination and to ensure reproducibility, each capsule will contain a single alloy, in triplicate. The second phase of experimentation will involve testing the best and worst performing alloys from the previous experiment separately in a FLiNaK environment but with the addition of a small quantity of a reducing agent, such as zirconium.

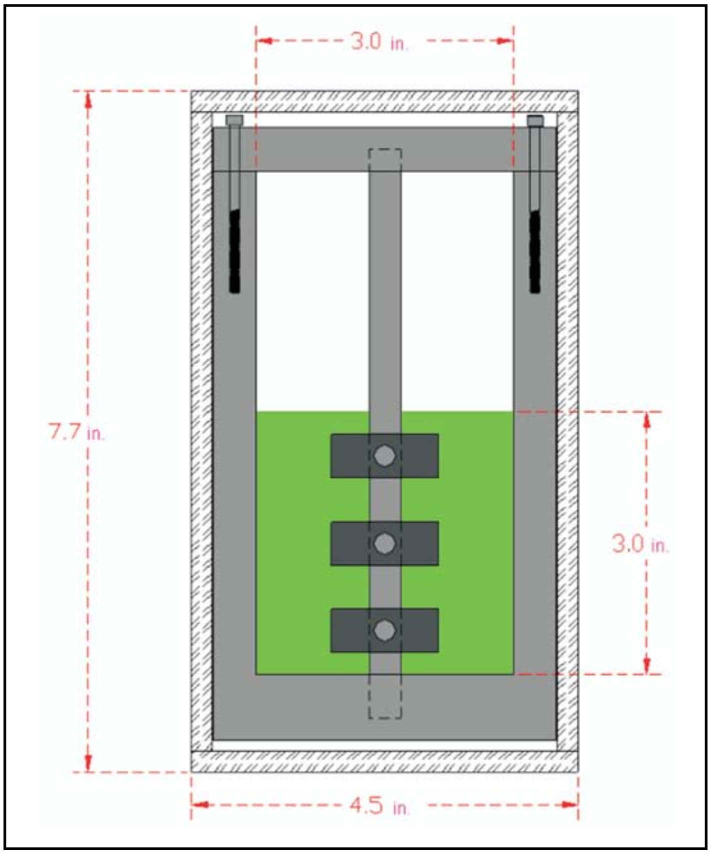


Figure 2. Cross-sectional view of the experimental capsule for corrosion testing of alloys in molten salt (Dimensions are in inches).

Figure 2 shows a schematic illustration of the cross-section of the experimental capsule for corrosion evaluation. Graphite screws attach the samples to a central graphite rod that is then inserted into a graphite crucible. The graphite crucible will then be encapsulated in a stainless steel containment vessel and molten FLiNaK salt will be transferred into the crucible using pressure differentials and airtight transfer systems. The capsules will then be welded shut, removed from the argon atmosphere, and inserted into an atmosphere-controlled furnace. Three capsules will be tested simultaneously at 1,000°C for 500 hours. Upon completion of this high-temperature exposure, the salt temperature will be lowered to 500°C and capsules will be inverted to allow the still molten FLiNaK to drain away from the samples before freezing.

Post-testing evaluation of the samples will employ a variety of analysis techniques. Weight-loss measurements will provide a rapid determination of the extent of attack due to elemental dissolution. Scanning electron microscopy will be used to examine micro-structural features of the corrosion attack. Auger Electron spectroscopy and Electron Spectroscopy for Chemical Analysis will be used for near-surface chemical analysis of the grain boundaries. X-ray diffraction will be used to determine the phases present in the alloys and to provide an indication of the corrosion products. Additionally, salt samples from the corrosion tests will be analyzed for elements leached from the alloy samples using inductively coupled plasma mass spectroscopy.

For the construction of the molten salt corrosion loop, graphite is being considered as the material of choice for the main loop components. Graphite is compatible with the molten salts and is able to withstand temperatures well above 2,000°C. In addition, the loop would consist of two removable test sections, which would be made of each of the candidate alloys for testing corrosion and transport of corrosion products from the hot leg to the cold leg of the loop. For joining graphite components, we are examining new technologies that may aid in more effective joining of graphite components. For example, a graphite epoxy, Resbond 931-1, forms a 99.9 percent pure graphite bond and is currently being evaluated as a possible method for joining graphite components. Small capsule tests are being designed to evaluate the strength and compatibility of this epoxy in a FLiNaK environment.

NUCLEAR ENERGY RESEARCH INITIATIVE

Ni-Si Alloys for the S-I Reactor-Hydrogen Production Process Interface

PI: Joseph W. Newkirk, University of Missouri-Rolla

Collaborators: Idaho National Laboratory

Project Number: 06-024

Project Start Date: March 2006

Project End Date: March 2009

Project Description

The goal of this project is to develop materials suitable for use in the sulfuric acid decomposition loop of the sulfur-iodine thermochemical cycle for nuclear hydrogen production. Materials must possess both acceptable corrosion resistance and sufficient ductility for component fabrication and avoidance of catastrophic failure. Nickel-silicon intermetallics show promise for such critical applications as the sulfuric acid vaporizer, vapor superheater, and the decomposer. Past work indicates that adding minor alloying elements to Ni_3Si provides significant ductility at room temperature (7-10 percent elongation at failure). This is a unique property for high silicon materials, which are usually brittle. Ni_3Si can also be easily joined by traditional methods such as welding. In addition, preliminary studies indicate that it has excellent corrosion resistance.

In this work, Ni_3Si will undergo further development to maximize ductility and corrosion resistance while reducing cost. The effects of adding elements such as niobium, boron, and iron will be analyzed. Microalloying may also be used to improve resistance to expected corrosive impurities in the sulfuric acid processing stream, such as iodine. Finally, the extent to which iron can be substituted for nickel in Ni_3Si without adversely affecting ductility or corrosion resistance will be studied.

The mechanical properties of these new materials will be documented over a range of temperatures and strain rates. The results will be used to improve material properties and microstructure. As a final test, corrosion-resistant materials will be subjected to flowing H_2SO_4 at temperatures and pressures comparable to the actual sulfuric acid processing loop (120-400°C at <10,000 psi) in order to measure corrosion rates. Various fabrication techniques will also be exercised by forming prototype plates, pipes, and forgings.

Workscope

The research will be divided into four areas:

- Materials development for ductility and corrosion resistance
- Thermal and mechanical property characterization
- Corrosion resistance in sulfuric acid
- Fabrication issues

NUCLEAR ENERGY RESEARCH INITIATIVE

Microstructure Sensitive Design for Materials in Solid Oxide Electrolyzer Cell

PI: Hamid Garmestani and Guillermo Goldsztein, Georgia Tech Research Corporation

Collaborators: Pacific Northwest National Laboratory

Project Number: 06-027

Project Start Date: March 2006

Project End Date: March 2009

Project Description

An efficient and cost-competitive method for producing hydrogen is to split water by solid oxide electrolyzer cells (SOEC) utilizing heat and electricity from a high-temperature nuclear reactor. Two major hurdles must still be overcome: 1) improving performance and life cycle of SOECs and 2) developing a method for large-scale fabrication of uniform and efficient electrodes. To address these challenges, this project will focus on analyzing transport properties of porous media and solid oxide fuel cells, and developing a set of multi-scale mathematical tools for describing classes of microstructures that allow efficient and fast computation of material properties. The research team will fabricate gradient porous electrode materials using both aerosol assisted chemical vapor deposition (AACVD) and spray pyrolysis methods. In addition, they will conduct experiments guided by microstructure sensitive design (MSD) techniques to optimize material microstructure design for best performance.

Workscope

The following activities comprise the primary project workscope:

- Develop mathematical methods for characterizing material microstructure
- Perform experimental work for fabricating gradient porous electrodes on a large scale
- Model the cathode microstructures and fabricate samples using CVD, AACVD, and spray pyrolysis methods

NUCLEAR ENERGY RESEARCH INITIATIVE

Dynamic Simulation and Optimization of Nuclear Hydrogen Production Systems

PI: Paul Barton, Mujid Kazimi, Massachusetts Institute of Technology

Collaborators: None

Project Number: 06-041

Project Start Date: April 2006

Project End Date: April 2009

Project Description

An important alternative to producing hydrogen from fossil fuels is producing it via nuclear power. This project is part of a research effort to design a hydrogen plant and its interface with the reactor. The natural interdependence between design and operational decisions for integrated nuclear hydrogen production systems necessitates a modeling and simulation environment that can capture the physical design descriptions and map these to steady-state and dynamic predictions of the hypothesized system behavior. Simulating these systems can provide invaluable information for the next step, whether this step is an experiment or design decision. The simulated environment must be adaptable, flexible, and expandable.

This project will develop a dynamic modeling, simulation, and optimization environment for nuclear hydrogen production systems. A hybrid discrete/continuous model design will capture both the continuous dynamics of the nuclear plant, hydrogen plant, and their interface, along with discrete events of the overall system. This will allow researchers to study plant operations and accident scenarios. Researchers can also use it to conduct parameter estimation studies to identify possible improvements in materials, mechanical design, and safety issues. The seamless connection between modeling and simulation can help establish optimal control schemes. These schemes can then be tested in the model.

Workscope

The following activities comprise the scope of work for this project:

- Conduct superstructure analysis to screen and compare hydrogen production alternatives
- Identify key characteristics for subsystems as components of the overall system
- Survey existing reactor and hydrogen production models for suitability
- Integrate new and/or existing models into the overall system
- Validate the models with available data
- Establish scenarios for simulation, including start-up, shutdown, off-normal, and accident
- Perform dynamic simulation studies of alternatives for hydrogen production
- Identify relevant process parameters and conduct dynamic optimization studies
- Compare performance, efficiency, and other factors for hydrogen process alternatives
- Study optimal control policies and streamline model improvement

NUCLEAR ENERGY RESEARCH INITIATIVE

High-Performance Electrolyzers for Hybrid Thermochemical Cycles

PI: John W. Weidner, University of South Carolina

Collaborators: Sandia National Laboratory, Savannah River National Laboratory, Argonne National Laboratory

Project Number: 06-054

Project Start Date: March 2006

Project End Date: March 2009

Project Description

This project provides the scientific basis for developing high-performance electrolyzers for use in two candidate thermochemical cycles for producing hydrogen from nuclear power: the hybrid sulfur process and the modified calcium-bromine cycle. This project will build on the successful application of the proton exchange membrane (PEM) electrolyzer for producing hydrogen through the conversion of water and sulfur dioxide ($H_2O + SO_2 \rightarrow H_2SO_4 + H_2$) and the dissociation of hydrogen bromide ($HBr \rightarrow Br_2 + H_2$). There are still a number of challenges in making these thermochemical cycles commercially viable, including 1) reducing the high cost of platinum and ruthenium catalysts, 2) minimizing SO_2 crossover (a serious lifetime-limiting phenomenon), and 3) finding suitable operating conditions for optimal electrolyzer performance and cycle efficiency.

This project will explore methods of improving the utilization of platinum and ruthenium in the cathodes and anodes, along with replacing them with other materials (metal alloys, silicides, mixed metal oxides, or a combination). New membranes are being developed with low gas crossover and high conductivity, mechanical stability, and temperature resistance that enable operation at higher temperatures and pressures. The researchers will evaluate the most promising catalysts and membranes in the PEM electrolyzer over a range of conditions (i.e., 30-120°C, 1-10 atmospheres, 50-80 percent conversions) and will apply mathematical models to optimize cell and process performance.

Workscope

This project will perform the following four tasks:

- Synthesize and characterize low-cost nanostructured electrocatalysts and supports for use in the anode and cathode
- Synthesize and characterize new membranes that operate at higher temperatures and minimize SO_2 crossover
- Conduct performance studies to optimize electrolyzer operation
- Perform mathematical modeling and process simulation to optimize operation of the electrolyzer and thermochemical cycles

Development of Efficient Flowsheet and Transient Modeling for Nuclear Heat Coupled Sulfur Iodine Cycle for Hydrogen Production

PI: Shripad T. Revankar, Purdue University

Collaborators: None

Project Number: 06-060

Project Start Date: March 2006

Project End Date: March 2009

Project Description

The main goal of this project is to develop a flowsheet for the closed-loop sulfur iodine (SI) cycle for nuclear hydrogen production. This flowsheet will use current advances in acid decomposition and product gas separation to achieve high thermal efficiency. It will result in the development of transient analysis methods for the SI cycle. Although the closed-loop SI cycle has recently been demonstrated on a bench scale, several challenges remain, such as maintaining stable operation, enhancing efficiency, obtaining thermodynamic data for the reactions, coupling to a high-temperature nuclear reactor, and determining transient behavior of the coupled system. This project will develop models to study transient performance of the closed-loop SI cycle. In addition, this research will explore several alternatives to SI cycles that have been proposed.

A model for the General Atomics (GA) SI cycle flowsheet will be developed using ASPEN PLUS and benchmarked with flowsheet simulation results. Flowsheet modifications will be considered using new membrane techniques for efficient separation of the process gases in the SI cycle. A comparative study of a modified flow sheet with the GA flowsheet will then be performed to identify further technical challenges and research needs. A transient analysis model for the nuclear-hydrogen coupled plant will also be developed for use as a tool to derive process control strategies and logic for coupled plant transient scenarios.

Workscope

Specific objectives of the project are to:

- Perform benchmark flowsheet analysis of the baseline SI cycle
- Investigate membrane techniques for HI and H_2SO_4 decomposition and separation processes
- Perform comparative flowsheet analyses of the modified cycles
- Develop component-wise SI cycle models for application to the transient analysis
- Perform preliminary analysis of transient behavior of the closed-loop SI cycle

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