



International Nuclear Energy Research Initiative

2006 Annual Report



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Foreword

The International Nuclear Energy Research Initiative (I-NERI) was established by the U.S. Department of Energy (DOE) Office of Nuclear Energy (NE) in Fiscal Year 2001 to conduct research and development (R&D) with international partners in advanced nuclear energy systems development. I-NERI was created in response to recommendations made by the President's Committee of Advisors on Science and Technology in a report entitled *Powerful Partnerships: The Federal Role in International Cooperation on Energy Innovation*. This annual report describes the I-NERI program's mission, organization, goals, accomplishments, and future plans.

I-NERI supports scientific and engineering R&D linked to the principal research programs sponsored by the DOE-NE: the Generation IV Nuclear Energy Systems Initiative, the Advanced Fuel Cycle R&D program, and the Nuclear Hydrogen Initiative. International collaboration is an important part of NE's R&D effort. I-NERI is designed to foster international partnerships to address key issues affecting the future global use of nuclear energy. Through international collaboration, DOE can effectively leverage its limited economic resources, more quickly expand the knowledge base of nuclear science and engineering, and establish valuable intellectual relationships with researchers from other countries.

Current I-NERI collaborators include Brazil, Canada, the European Union, France, Japan, and the Republic of Korea. In FY 2006, DOE initiated four more projects with the Republic of Korea, two with France, and one each with Japan, Brazil, and Italy. This *I-NERI 2006 Annual Report* provides a description of those new projects, along with a comprehensive summary of the progress of each collaborative research project initiated since FY 2003, based on information submitted by the projects' principal investigators. Forging these partnerships enhances the participation of the United States within the global nuclear community, building an international consensus on such critically important issues as designing proliferation-resistance into advanced nuclear systems and expanding the benefits of nuclear power.

Canno R. Spinger

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

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1.0 Introduction

The International Nuclear Energy Research Initiative (I-NERI) supports the *National Energy Policy* by conducting research to advance the state of nuclear science and technology in the United States. I-NERI sponsors innovative scientific and engineering research and development (R&D) in cooperation with participating countries. The research performed under the I-NERI umbrella addresses key issues affecting the future of nuclear energy and its global deployment. A link to the program can be found at the NE website http://www.nuclear.gov.

This *I-NERI 2006 Annual Report* serves to inform interested parties about the program's organization, progress of collaborative research projects undertaken since FY 2003, and future plans for the program. Following is an overview of each section:

- Section 2 discusses background information on the series of events that led to the creation of the I-NERI program. The countries and international organizations participating in current I-NERI collaborative agreements are also presented.
- Section 3 presents an overview of program goals and objectives, a work scope summary for the three constituent program areas, a description of the I-NERI organization, and an overview of funding since the program's inception.
- Section 4 provides a summary of programmatic accomplishments, highlighting key activities for each year of the program, areas of research under each bilateral agreement, and a profile of participating organizations. This section also provides a summary of the projects completed in FY 2006.
- Sections 5 through 11 present details of the R&D work scope for current I-NERI collaborative projects with Brazil, Canada, the European Union (EU), France, Japan, the Republic of Korea, and the Organization for Economic Cooperation and Development (OECD), respectively. For each participating country or organization, the report presents an index of projects and a summary of technical accomplishments achieved in FY 2006.

2.0 Background

In January 1997, the President of the United States requested his Committee of Advisors on Science and Technology (PCAST) to provide a strategy to ensure that the U.S. has a program to address the nation's energy and environmental needs for the next century. In a report responding to this request, the PCAST Energy R&D Panel determined that ensuring a viable nuclear energy option was essential to help meet U.S. future energy needs.

Specifically, the panel recommended that the U.S. Department of Energy (DOE) should implement a properly focused R&D effort in order to address the principal obstacles to continuing a viable nuclear energy option. In response to these recommendations, DOE established the Nuclear Energy Research Initiative (NERI) in 1999. Information and annual reports on the NERI program are available at the NERI website:

http://nuclear.energy.gov/neri/neNERIresearch.html

Recognizing the need for an international component of the NERI program, PCAST issued a report in June 1999 entitled *Powerful Partnerships: The Federal Role in International Cooperation on Energy Innovation*, which promotes "bilateral and multilateral research focused on advanced technologies for improving the cost, safety, waste management, and proliferation resistance of nuclear fission energy systems." The report states, "The costs of exploring new technological approaches that might deal effectively with the multiple challenges posed by conventional nuclear power are too great for the United States or any other single country to bear, so that a pooling of international resources is needed..."

In response to PCAST recommendations, I-NERI was established in FY 2001. This international component of NERI is designed to enhance DOE's ability to leverage its limited funding for nuclear technology research with additional funding from other countries. It responds to the Department's directive to improve cost performance, increase proliferation resistance, enhance safety, and improve the waste management of future nuclear energy systems. To date, seven bilateral I-NERI collaborative agreements have been fully implemented between DOE and the following international partners:

- Commissariat à l'Energie Atomique (CEA) of France
- Republic of Korea Ministry of Science and Technology (MOST)
- The Nuclear Energy Agency (NEA) of the Organization for Economic Cooperation and Development (OECD)
- European Atomic Energy Community (EURATOM)
- Department of Natural Resources Canada (NRCAN) and Atomic Energy of Canada Limited. (AECL)
- Brazilian Ministry of Science and Technology (MST)
- Agency of Natural Resources and Energy of Japan (ANRE) and the Ministry of Education, Culture, Sports, Science, and Technology of Japan (MEXT)

Since the program's inception, a total of 66 projects have been initiated:

- 18 with France
- 25 with the Republic of Korea
- 11 with the European Union
- 7 with Canada
- 2 with Brazil
- 2 with Japan
- 1 with OECD

DOE is engaged in discussions with the Republic of South Africa (agreement expected in FY 2007), with the intent of establishing additional collaborations in the future.

3.0 I-NERI Program Description

3.1 Mission

I-NERI's mission is to sponsor innovative scientific and engineering R&D in cooperation with international partner countries. This mission includes the DOE directive to address key issues affecting the future use of nuclear energy and its global deployment by improving cost performance, increasing proliferation resistance, enhancing safety, and improving the waste management of future nuclear energy systems.

3.2 Goals and Objectives

Through its mission, the I-NERI program is designed to foster closer collaboration among international and U.S. researchers, improve communications, and expand the sharing of nuclear research information. In order to accomplish its assigned mission, the I-NERI program has established the following overall objectives:

- To develop advanced concepts and scientific breakthroughs in nuclear energy and reactor technology in order to address and overcome the principal technical and scientific obstacles to expanding the global use of nuclear energy
- To promote collaboration with international agencies and research organizations in order to improve the development of nuclear energy
- To promote and maintain a nuclear science and engineering infrastructure in order to resolve future technical challenges

Through the I-NERI program, the DOE Office of Nuclear Energy (DOE-NE) has coordinated wide-ranging discussions among governments, industry, and the worldwide research community regarding the development of Generation IV nuclear energy systems, advanced fuel cycles, and nuclearhydrogen technologies.

3.3 International Agreements

In order to initiate an international collaboration, a government-to-government agreement must first be in place. These agreements are the vehicles to conduct Generation IV Nuclear Energy Systems Initiative (Generation IV), Advanced Fuel Cycle (AFC), and Nuclear Hydrogen Initiative (NHI) R&D with member countries of the Generation IV International Forum (GIF). Of the 13 chartered GIF members, 7 have signed the Framework Agreement to pursue international collaboration: Canada, the European Union, France, Japan, the Republic of Korea, Switzerland, and the United States. In July 2006, the GIF voted unanimously to extend offers of membership to China and Russia. These two countries officially signed the GIF Charter in November 2006 at the Policy Group meeting in Paris and have one year to sign the Framework to become full members.

The GIF partners are in the process of establishing multilateral agreements to conduct R&D among several member countries. In the meantime, I-NERI's bilateral agreements enable U.S. researchers to establish international R&D collaborations to begin developing nextgeneration nuclear energy systems. The United States has established bilateral I-NERI agreements with six of these member countries: Brazil, Canada, European Union, France, Japan, and the Republic of Korea.

3.4 Program Organization and Control

The DOE-NE manages I-NERI, with advice from the Nuclear Energy Research Advisory Committee (NERAC). NE's Office of International Partnerships, under the Office of Corporate and Global Partnership Development, negotiates and establishes the bilateral agreements; the Office of Nuclear Power Deployment, with the support of the technical Program Directors and Country Managers, administers the selection and funding of awards; and the Idaho Operations Office (ID) negotiates and monitors the cooperative agreements with U.S. entities. Once projects are initiated, the Offices of Gas Reactor Deployment and Advanced Fuel Cycle Research and Development manage the R&D work under these agreements. The National Laboratories generally assign the principal investigator to conduct the scientific research for each project.

The U.S. appoints technical coordinators for each of the R&D areas of the agreement. These are the National Technical Directors (NTDs) or Technical Directors (TDs), who are assisted by System Integration Managers (SIMs). There are seven NTDs, representing each technology area of the Generation IV program (systems analysis, fuels, materials, and energy conversion) and Advanced Fuel Cycle R&D (chemical separations, transmutation, and system design and evaluation). The NHI program has

three TDs for its major research areas (thermochemical cycles, high temperature electrolysis, and systems interface).

The SIMs manage technologies for each of the six Generation IV reactor concepts:

- Very High-Temperature Reactor (VHTR)
- Supercritical Water Reactor (SCWR)
- Gas Fast Reactor (GFR)
- Lead Fast Reactor (LFR)
- Sodium Fast Reactor (SFR)
- Molten Salt Reactor (MSR)

Integration of the three R&D program areas at the national level is accomplished by appointing a Country Coordinator for each bilateral I-NERI agreement. This individual represents the United States in bilateral meetings, negotiates areas of collaboration, selects new projects, and evaluates existing projects. Each collaborating country establishes a similar function. The I-NERI Program Manager manages the overall implementation of the agreements and administers all international collaborations under these agreements.

The NTDs, SIMs, and TDs assist the Country Coordinators in identifying cooperative research areas and defining specific work scopes. They review periodic progress reports and provide advice on future direction. They also participate on a panel of technical experts to formally evaluate the projects during annual project reviews.

Figure 1 illustrates the DOE organizational structure for control and administration of the I-NERI program.

3.5 Funding

I-NERI is an important vehicle for enabling international R&D in Generation IV, AFCR&D, and NHI technology on a leveraged, cost-shared basis. In addition, I-NERI allows bilateral collaborations between the U.S. and GIF countries until multilateral agreements are established.

Each country in an I-NERI collaboration provides funding for their respective project participants. The U.S. contribution is based upon current-year budgets.



Figure 1. I-NERI Organizational Structure.

Actual cost-share amounts are determined jointly for each selected project. The program's goal is to achieve approximately 50-50 matching contributions from each partner country.

To date, I-NERI projects have received a total research and development investment of \$188.5 million: \$101.6 million contributed by the U.S. and \$86.9 million by international collaborators. International investment is:

- \$15.0 million from Canada
- \$30.2 million from France
- \$24.4 million from the Republic of Korea
- \$2.7 million from Japan
- \$12.3 million from the European Union
- \$2.3 million from Brazil

Funding provided by the U.S. may only be spent by U.S. participants. I-NERI projects typically last three years and are funded annually by the Generation IV, AFCR&D, and NHI programs.

3.6 Work Scope

In FY 2004, DOE restructured the I-NERI program to support the objectives of NE's principal R&D programs: Generation IV, AFCR&D, and NHI. The work scope of all current I-NERI projects is directly linked to the scientific and engineering needs of these programs.

Candidate project work scopes are jointly developed by the U.S. and the collaborating country. The NTDs review the technical quality and budget of the proposed joint projects in order to make recommendations to the Country Coordinators. The Country Coordinators select R&D projects based on conformance with the bilateral agreement and current Generation IV, AFCR&D, and NHI programmatic needs.

Following is an overview of the individual work scopes for NE's three R&D programs:

Generation IV Nuclear Energy Systems Initiative. The Generation IV program is developing next-generation nuclear energy systems that offer advantages in the areas of economics, safety, reliability, and sustainability, with a goal of commercial deployment by the year 2030. Using a technology roadmap created by member countries of the Generation IV International Forum, six reactor concepts were deemed most promising. As previously listed, these designs include: the gas-cooled fast reactor, lead-cooled fast reactor, molten salt reactor, sodium-cooled fast reactor, supercritical water-cooled reactor, and the very hightemperature reactor. Current U.S. research priorities are focused primarily on the VHTR and SFR.

Generation IV has eight technology goals:

- To provide sustainable energy generation that meets clean air objectives and promotes long-term availability of systems and effective fuel utilization for worldwide energy production
- To minimize and manage nuclear waste, notably reducing the long-term stewardship burden in the future and thereby improving protection for public health and the environment
- To increase assurances against diversion or theft of weapons-usable materials
- 4) To ensure high safety and reliability
- 5) To design systems with very low likelihood and degree of reactor core damage
- 6) To create reactor designs that eliminate the need for offsite emergency response
- 7) To ensure that systems have a clear life-cycle cost advantage over other energy sources
- 8) To create systems that have a level of financial risk that is comparable to other energy projects

Advanced Fuel Cycle R&D. This initiative responds to the 2001 *National Energy Policy* recommendation that the United States "... develop reprocessing and fuel treatment technologies that are cleaner, more efficient, less waste-intensive, and more proliferation-resistant." These technologies are key components of the fuel cycles in the Global Nuclear Energy Partnership program and may also be of value to the Generation IV nuclear energy systems initiative.

Research under this initiative focuses on recycling, fuel treatment, and conditioning technologies that have the potential to dramatically reduce the quantity, toxicity, and thermal content of spent nuclear fuel, thus decreasing the requirements for geological disposal. The AFCR&D mission is to develop proliferation-resistant spent nuclear fuel treatment and transmutation technologies in order to enable a transition from the current once-through nuclear fuel cycle to a future sustainable closed nuclear fuel cycle.

Nuclear Hydrogen Initiative. The NHI program supports the President's Hydrogen Fuel Initiative. The goal of this initiative is to develop the technologies and infrastructure to economically produce, store, and distribute hydrogen for use in fuel cell vehicles and electricity generation. Hydrogen can be produced using a variety of technologies, each of which has its advantages and limitations. The primary advantage of nuclear energy production technologies is the potential ability to produce hydrogen in large quantities at a low cost without the emission of any greenhouse gases.

The goal of the NHI program is to demonstrate the commercial-scale, economically feasible production of hydrogen using nuclear energy by the year 2020. This initiative conducts R&D on enabling technologies, demonstrates nuclear-based hydrogen production technologies, studies potential hydrogen production schemes, and develops deployment alternatives to meet future needs for increased hydrogen consumption.

4.0 I-NERI Program Accomplishments

The I-NERI program began in the second quarter of FY 2001, initially focusing on developing international collaborations, program planning, and project procurements. Since the inception of the program, 66 projects have been awarded and 31 of these completed.

4.1 Programmatic Accomplishments

Following is a brief description of the program's primary accomplishments from FY 2001 through FY 2005, an overview of accomplishments during FY 2006, and planned accomplishments for the upcoming fiscal year.

FY 2001-2005 Accomplishments. In FY 2001, the first year of the I-NERI program, DOE signed collaborative agreements with the Republic of Korea and France. By the end of FY 2001, the U.S./France collaboration was initiated and four projects were awarded. A competitive procurement was conducted for the U.S./Republic of Korea collaboration.

In FY 2002, the U.S. and the Republic of Korea (ROK) awarded the first six projects. The OECD signed on with a new collaboration agreement, under which one new project was awarded. One new project was initiated under the U.S./France collaboration.

In FY 2003, five new awards were initiated under the U.S./Republic of Korea collaboration. DOE also signed I-NERI cooperative agreements with the European Commission, Canada, and Brazil.

In FY 2004, I-NERI researchers completed two U.S./ France projects that were awarded in FY 2001. Seven new projects were added to the U.S./Canada collaboration, eleven with France, six with Korea, and eight research projects with the European Union. Japan became an I-NERI participant during this fiscal year, signing a new cooperative agreement.

During FY 2005, four new projects were initiated with the Republic of Korea, two with the European Union, one with Brazil, and one with Japan. The latter represent the first research projects undertaken through the U.S. bilateral agreements with Brazil and Japan. Researchers completed work on eight cooperative projects begun in FY 2001 through 2003. U.S. contributions in FY 2005 totaled \$17.0 million and international contributions totaled \$18.7.

FY 2006 Accomplishments. During FY 2006, nine new research projects were initiated under the agreements with Japan, EURATOM, Brazil, France and the Republic of Korea. Twelve projects begun in FY 2004 were completed last year, along with one each started in FY 2003, FY 2005, and FY 2006.

Following is a summary of noteworthy accomplishments achieved during FY 2006:

- Completed 15 research projects:
 - ♦ one with Brazil
 - three with Canada
 - two with the European Union
 - ♦ five with France
 - ♦ four with the Republic of Korea
- Initiated nine new collaborative research projects
 - the first with Italy under the existing EURATOM agreement
 - ♦ a second with Brazil
 - ♦ a second with Japan
 - ♦ four with the Republic of Korea
 - ♦ two with France
- Completed FY 2006 annual project performance reviews for the European Union, France, and Republic of Korea bilateral collaborations.

Planned FY 2007 Activities. During FY 2007, DOE plans the following international activities:

- Initiate new cooperative projects under existing agreements
- Sign a new cooperative research agreement with South Africa
- Continue pursuing cooperative agreements with the United Kingdom, Argentina, and other prospective partner countries

4.2 Current I-NERI Collaborations

Brief descriptions of the current I-NERI collaborations are provided in the sections that follow. Descriptions of the work scopes, listings of funded projects, and brief project status reports are provided in Sections 5 through 11 for I-NERI R&D projects undertaken with Brazil, Canada, the European Union, France, Japan, the Republic of Korea, and OECD, respectively.

Table 1 presents a breakdown of the number of project awards for each country by fiscal year. Figure 2 shows the distribution of projects by each of the three major program areas since the program's inception. (It should be noted that prior to 2004 all of the projects were related strictly to the Generation IV initiative.)

Collaborator	FY 01	FY 02	FY 03	FY 04	FY 05	FY 06	Total
France	4	1		11		2	18
Republic of Korea		6	5	6	4	4	25
OECD-NEA		1					1
EURATOM				8	2	1	11
Canada				7			7
Brazil					1	1	2
Japan					1	1	2
Total	4	8	5	32	8	9	66

Table 1. Number of projects awarded.



Figure 2. Project distribution by program area.

Brazil. Cooperative research projects with the Brazilian Ministry of Science and Technology (MST) takes place primarily in the areas of advanced nuclear fuels, fuel cycles, and materials, based on a bilateral agreement signed June 20, 2003. Cooperative research with Brazil entails instrumentation, operations and control, and human interaction of the Integral Primary System Reactor (IPSR), along with an investigation into shared resources for multiple modular reactor designs.

Canada. The U.S./Canada collaboration includes R&D in the areas of nuclear hydrogen production, advanced fuel cycles, and supercritical water-cooled reactor technology. The collaborating agency in Canada is Atomic Energy of Canada Limited (AECL).

European Union. The collaborating agency for Europe is the European Atomic Energy Community (EURATOM), an international organization composed of the members of the European Union (EU). The U.S./EU collaboration focuses on R&D proposals in the areas of fuels and materials, advanced reactor design, and transmutation.

France. The collaborating agency in France is the Commissariat à l'Energie Atomique (CEA). The U.S./ France collaboration focuses on developing Generation IV advanced nuclear system technologies that will enable the U.S. and France to move forward with cutting-edge R&D that will benefit a range of anticipated future reactor and fuel cycle designs. This collaboration is conducting research in advanced gas-cooled reactors, fuels and materials, nuclear hydrogen production, and radiation effects.

Japan. An agreement was signed with the Agency of Natural Resources and Energy (ANRE) of Japan on May 26, 2004. The areas of collaboration under this agreement are supercritical water reactors, innovative light water technologies, oxide fuel processing for light water reactors, fuel technologies using solvent extraction, and radioactive waste processing. On February 8, 2005, DOE and Japan's Ministry of Education, Culture and Sports, Science and Technology (MEXT) signed the Implementing Arrangement concerning cooperation in these innovative nuclear energy R&D technologies.

Republic of Korea. The participating agency in the Republic of Korea is the Ministry of Science and Technology (MOST). The U.S./Republic of Korea collaboration encompasses advanced technologies for improving the cost, safety, and proliferation resistance of nuclear energy systems. Research includes instrumentation and controls, advanced light water reactors, fuel cycles and materials, gas-cooled and supercritical water-cooled reactors, and nuclear hydrogen production. The U.S./Republic of Korea I-NERI projects have been selected competitively through an independent peer-evaluation process.

OECD. The U.S. teamed with the 28-member Nuclear Energy Agency of the Organization for Economic Cooperation and Development to conduct reactor materials experiments and associated analyses involving concrete-fuel interactions and core cooling. The U.S. funding team consisted of the U.S. Nuclear Regulatory Commission and DOE.

4.3 Program Participants

Following is an organizational profile of the seven bilateral agreements and a complete list of the I-NERI program participants. In addition, it presents the level of U.S. university student participation in active I-NERI projects during 2006.

U.S. Collaborators

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

Industry Organizations

General Atomics Westinghouse Electric Gas Technology Institute

Universities

Iowa State University Massachusetts Institute of Technology Ohio State University Pennsylvania State University Purdue University University of California-Santa Barbara University of California-Santa Barbara University of Florida University of Florida University of Florida University of Maryland University of Maryland University of Michigan University of Notre Dame University of Wisconsin

International Collaborators

Industry Organizations Atomic Energy of Canada Limited Eletronuclear Framatome-ANP, Lyon Gamma Engineering **Award Profiles.** Table 2 provides a detailed breakdown of the participating organizations. Figure 3 illustrates the number of current I-NERI collaborators by type of organization.

Hitachi Works Hitachi, LTD Korea Hydro and Nuclear Power Company Toshiba Corporation Società Informazioni ed Esperienze Termoidrauliche

Universities

Cheju University Chosun University Chungnam National University École Polytechnique de Montréal Hanyang University Korean Maritime University Pusan National University Pusan National University Seoul National University Tohoku University University of Manchester University of Manitoba University of Sherbrooke University of Tokyo University of Bordeaux

Governmental Organizations

Brazilian Ministry of Science and Technology Chalk River Laboratories Commissariat à L'Energie Atomique (CEA) Electricité de France Ente per le Nuove Tecnologie, l'Energia e l'Ambiente Instituto de Pesquisas Energéticas e Nucleares (IPEN)) Japan Atomic Energy Research Institute (JAERI) Joint Research Center Institute for Transuranium Elements (ITU) Japan Atomic Energy Agency (JAEA) Korea Advanced Institute of Science and Technology (KAIST) Korea Atomic Energy Research Institute (KAERI) Korean Electric Power Research Institute (KEPRI) Organization for Economic Cooperation and Development/Nuclear Energy Agency (OECD/NEA)



Figure 3. I-NERI participant profile.

U.S. Student Participation. As noted in Figure 3, 12 U.S. universities and colleges participated in I-NERI research projects in 2006. Approximately 86 students from these institutions worked on active I-NERI research projects during that year. Their distribution by university degree level is noted in Figure 4.



Figure 4. 2006 I-NERI U.S. student participation profile.

4.4 Completed I-NERI Projects

This year marked the completion of fifteen I-NERI projects—twelve begun in FY 2004, one remaining project from FY 2003 and one each from FY 2005 and FY 2006.

Based on the documented accomplishments, it is apparent that I-NERI's goals and objectives continue to be satisfied. Collaborative efforts between the public and private sectors in both the U.S. and partnering international countries have resulted in significant scientific and technological enhancements in the global nuclear power arena. The international collaborations have forged lasting ties that will continue promoting the strong infrastructure necessary to overcome future challenges to the expanded use of this vital source of clean and reliable power. In conjunction with parallel efforts undertaken by the NERI program, Generation IV, AFCR&D, and NHI, this program has helped to revive the Nation's leadership role in international nuclear R&D. The resulting technological and scientific advances will ensure the U.S. remains competitive in both the global and domestic nuclear energy marketplaces.

The following table identifies the 15 projects completed in 2006. More detailed information on each can be found in the corresponding project summaries in Sections 5 through 11 of this report.

Project Number	Title	Lead Organization
2006-001-B	Design Strategies and Evaluation for Sharing Systems at Multi-Unit Plants	Oak Ridge National Laboratory
2004-001-C	High-Temperature Electrolyzer Optimization	Argonne National Laboratory
2004-005-C	Supercritical Water-Cooled Reactor Stability Analysis	Argonne National Laboratory
2004-006-C	Thermal-Hydraulic Benchmark Studies for SCWR Safety	Idaho National Laboratory
2004-002-E	Development of Fuels for the Gas-Cooled Fast Reactor	Idaho National Laboratory
2004-007-E	Molten Salt technology for Reactor Applications	Oak Ridge National Laboratory
2004-004-F	SiC/SiC for Control Rod Structures for Next Generation Nuclear Plants	Idaho National Laboratory
2004-005-F	Assessment of Existing Physics Experiments Relevant to VHTR Designs	Argonne National Laboratory
2004-006-F	GFR Physics Experiments in the CEA-Cadarache MASURCA Facility	Argonne National Laboratory
2004-008-F	Development of Generation IV Advanced Gas-Cooled Reactors with Hardened/Fast Neutron Spectrum	Argonne National Laboratory
2004-009-F	Development of Fuels for the Gas-Cooled Fast Reactor	Idaho National Laboratory
2003-020-К	Advanced Corrosion-Resistant Zirconium Alloys for High Burnup and Generation IV Applications	The Pennsylvania State University
2004-003-K	Development of Advanced Suite of Deterministic Codes for VHTR Physics Analysis	Argonne National Laboratory
2004-006-K	Alternative Methods for Treatment of TRISO Fuels	Argonne National Laboratory
2005-003-К	Improvement of the Decay Heat Removal System for the VHTR	Argonne National Laboratory

Table 3. I-NERI projects completed in 2006.

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5.0 U.S./Brazil Collaboration

The U.S. Department of Energy (DOE) and the Brazilian Ministry of Science and Technology (MST) established a bilateral agreement on June 20, 2003. Secretary of Energy Spencer Abraham signed the agreement for DOE and Brazilian Minister of Science and Technology, Roberto Amaral, signed for MST. The first collaborative project under this agreement was awarded in FY 2005.

5.1 Work Scope Areas

R&D topics for the U.S./Brazil collaboration include:

- Advanced reactor development for future-generation
 nuclear energy systems
- Advanced reactor fuel and reactor fuel cycle integration
- Life cycle management and upgrading of current operating reactors
- Advanced fuel and material irradiation and use of experimental facilities
- Environmental and safety issues related to new reactor and fuel cycle technologies
- Fundamental nuclear science and engineering

5.2 Project Summaries

One new project was awarded during FY 2006. A listing of the I-NERI U.S./Brazil projects that are currently underway follows, along with a summary of the accomplishments achieved in FY 2006.

Directory of Project Summaries

2005-001-В	Development of Advanced Instrumentation and Control for an Integral Primary System Reactor 1	.3
2006-001-В	Design Strategies and Evaluation for Sharing Systems at Multi-Unit Plants	.7

Development of Advanced Instrumentation and Control for an Integrated Primary System Reactor

PI (U.S.): David Holcomb, Oak Ridge National Laboratory (ORNL)

Project Number: 2005-001-B

PI (Brazil): Antonio Barroso, Instituto de Pesquisas Energéticas e Nucleares (IPEN) Project Start Date: March 2005

Project End Date: September 2007

Collaborator: Westinghouse Electric Company

Research Objectives

Integral primary system reactors (IPSRs) have distinctive instrumentation and control (I&C) configurations and requirements compared to traditional external loop light water reactors (LWRs). The overall objective of this project is to develop specialized I&C technologies that are not directly transferable from external loop LWRs. When this project began, no systematic assessment of the I&C requirements for IPSRs was available to compare with known LWR technologies. Therefore, performing a detailed review of the instrumentation requirements for an IPSR became a leading task.

A previously identified instrumentation challenge for IPSRs is obtaining an accurate, in-vessel water level measurement. Conventional measurements are almost impossible because of the irregular path imposed by the shape of the vessel's internal structural components, such as the pressurizer bottom plate, riser, and control rod drive mechanisms. Two candidate systems are currently under development: 1) an ultrasonic, torsional waveguidebased level measurement technique and 2) a cooled-fluidbased, lance-type probe with advanced signal processing algorithms.

An additional objective of this work is to assess areas of plant operation and control where IPSR features and operating modes require innovative approaches. For example, the medium size and modularity of the IPSR provide economic incentives for deployment of multiple reactor modules in a single nuclear park. Co-generation, which produces desalinated water, district heating, industrial steam, or hydrogen in addition to electrical power, is an attractive option for modular reactors sited in areas that already have sufficient electrical generation capacity to supply baseload power requirements. In order to fully use the energy available from all reactors in a nuclear park, the balance-of-plant must be reconfigurable to allow variable co-generation with changes in the electrical load. In order to optimize multi-modular and/or reconfigurable operation, a hierarchical supervisory control system needs to be developed to overlay the individual unit control system. Thus, the hierarchical control development task will maximize the utilization efficiency of the powerpark resources while minimizing staffing requirements.

The final technical area of this project is to develop guidelines for the operator to interact with plant controls and protection systems. IPSRs are typically characterized by long transient evolution times due to the large thermal inertia of the primary system. Advanced designs, such as the Westinghouse International Reactor Innovative & Secure (IRIS), are unique in that they are capable of responding to almost any operational condition or accident without requiring operator action. Due to the advantageous thermal characteristics of IPSRs compared to external loop LWRs, the operator interaction with the control/protection systems needs to be redesigned. Emergency procedure guidelines and control room architecture must also take into account the possibility of controlling multiple modules from a single control room. Human/machine interfaces that reflect the distinctive characteristics of IPSRs will be evaluated as this project progresses.

Research Progress

A primary project focus during FY 2006 has been developing prototype instrumentation to provide accurate and reliable measurement of in-vessel coolant levels. Two candidate systems are currently under development: 1) ORNL is pursuing an ultrasonic, torsional waveguide-based level measurement technique and 2) IPEN is developing a cooled lance-type liquid-level probe featuring advanced signal processing to yield a continuous liquid-level measurement.

Researchers have developed an advanced experimental prototype for the ultrasonic, torsional



Figure 1. Conceptual schematic showing configuration of ultrasonic torsional-wave based, in-vessel level measurement system.

wave-based level measurement system and demonstrated basic system functionality in a laboratory environment. As part of the prototype development process, they created custom signal processing software to interpret the measured signals and implemented prototype measurement electronics. To support the system design, they also developed a dynamic, 3-D finite element model for the transduction process and components. To facilitate IPSR in-vessel environmental survival, the design incorporates high-temperature and high-pressure tolerant materials throughout the sensor body. Figure 1 shows the conceptual layout of the level measurement system.

The reactor transient analysis task, completed this year, focused on reviewing the existing computer models of the IRIS reactor that are suitable for transient analyses. The goal of the evaluation was to determine what changes are needed to make the models applicable for control system design and simulation of plant dynamic response. Westinghouse served as the project lead for all of the reactor modeling and analysis tasks. Researchers reviewed and revised two previously developed reactor models to reflect the needs of this project: 1) a highfidelity, detailed RELAP model and 2) a low-order simulatortype MODELICA model. The RELAP model is intended for reference and benchmarking analyses. However, because its running time may amount to hours and even days, it is not suitable for repetitive simulations of transients needed to optimize control systems and instrumentation usage. MODELICA, on the other hand, executes much faster while still providing adequate results for modeling control systems. In particular, researchers optimized the MODELICA simulation for speed to enable its effective use.

During the past year, researchers have made considerable progress in developing supervisory control systems for IPSRs and have examined issues related to the control of modules in co-generation mode (e.g., for water desalination). Work has begun on the final project task for operator control and protection system interaction.

Planned Activities

During the final project year, the team will develop an engineering prototype of an ultrasonic torsional level measurement device and demonstrate its operation under typical in-vessel operational conditions. The system will include all of the significant components required to function on-line, inside a reactor vessel. The engineering prototype development phase of the task will apply the lessons learned from the testing, simulation, and redesigning phase. The system control software will also be refined to focus on operational performance as opposed to measurement system diagnostics. Further, the engineering prototype transducer system will be packaged to withstand the rigorous in-vessel mechanical, electromagnetic, and thermal environments. Researchers plan to integrate testing of the engineering prototype system with the IRIS reactor component testing taking place at an Italian facility.

Researchers will complete the supervisory control system development and operation control and protection interaction tasks in the third project year. Under this task, functional requirements and the high-level characteristics of the supervisory hierarchical control system will be finalized. In the operator control and protection interaction task, identified differences in emergency response guidelines between large LWRs and the IRIS reactor will serve as a basis for IPSR emergency response guideline definition. I-NERI — 2006 Annual Report

Design Strategies and Evaluation for Sharing Systems at Multi-Unit Plants

PI (U.S.): Richard Wood, Oak Ridge National Laboratory

PI (Brazil): Antonio C. Barroso, Instituto de Pesquisas Energéticas e Nucleares (IPEN)

Collaborators: Westinghouse Electric Company, Eletronuclear

Project Number: 2006-001-B

Project Start Date: March 2006

Project End Date: September 2006

Research Objectives

The objective of this project is to identify and resolve technical issues associated with implementing and managing multiple modular nuclear plants co-located on a single site. Small reactors (150-350 MWe), such as the International Reactor Innovative and Secure (IRIS), are attractive for international near-term deployment because of their flexibility for staged deployment, lower initial financial investment, and suitability for co-generation. The research team selected IRIS for this project because participating organizations are members of the IRIS Consortium. The results of this work are expected to be applicable to most small modular reactor concepts, such as the Pebble Bed Modular Reactor.

Modularity of small reactors has been identified as a significant factor in achieving potential capital cost savings. However, to realize these economic benefits, researchers must evaluate the system engineering implications of modularity on the design, construction, commissioning, and operation of a multi-unit plant. The success of locating multiple modular nuclear plants on a single site lies in: 1) the ability to share plant process systems, auxiliary systems, and instrumentation and control and 2) the feasibility of sharing control room resources such as space, human-system interfaces, information systems, and personnel.

Project plans involve assessing the feasibility of sharing resources and evaluating the impact of common systems on risk, performance, reliability, and cost. The objective is to demonstrate strategies and methods to optimize the flexible characteristics of modular small reactors. This work provides the foundation for the development of a comprehensive evaluation methodology.

Research Progress

During the project's first year, researchers determined which reference plant systems were suitable for sharing among multiple units. They also identified the key considerations for assessing sharing strategies and developed fundamental tools to evaluate the impact of shared systems. The IRIS reference plant arrangement provides twin units as shown in Figure 1. According to Westinghouse, the twin arrangement maximizes sharing of components between the two modules and allows operation of a completed unit to commence while construction of the subsequent unit is underway.

It was determined that four factors influence the sharing of systems—economics, safety, operability, and licensing. To maximize the economics of sharing, all systems and structures must be considered. With respect to licensing considerations, regulations allow the sharing of systems. Of approximately 100 systems in nuclear power plants, about three-fourths do not impact safety and are not evaluated in the plant's probabilistic risk assessment. In addition to these factors, researchers must consider numerous metrics—some of which may be conflicting.

Researchers have compiled a list of plant systems that can be considered for sharing. They also established key considerations needed to develop a decision support methodology that defines a cost-effective approach to multi-modular implementation and preserves the safety characteristics of advanced plants. The findings of this investigation, while focused on the IRIS concept, can be applied to other multi-modular reactor concepts as well.



Figure 1. IRIS plant layout arrangement with two sets of twin units (1,340 MWe total per station).

Development of tools to support further evaluation of the impact of shared systems was also accomplished in the first year. A mathematical model representing the heat transfer and fluid flow dynamics of the IRIS helical coil steam generator was implemented to provide a means to accurately simulate performance and stability in a multimodular plant configuration. The enhanced simulation capability supports investigation of dynamic effects arising from shared systems and enables supervisory control development. This tool permits representation of limitations in performance (e.g., boiling instability at low flow) that have to be considered when evaluating sharing options. Simulation tests have shown that the model is successful in demonstrating these features.

The next step for this research would be to process the lists of candidate systems for sharing and qualitatively evaluate the benefits and drawbacks. Utilization of economic, risk, and simulation tools can facilitate comparative studies. The safety impact of sharing must also be assessed in detail.

Planned Activities

This project was terminated in September 2006 as a result of funding shortfalls and management re-prioritization of the Brazilian collaborating organizations. There are no further activities planned as part of this project.

6.0 U.S./Canada Collaboration

The Director of DOE-NE, William D. Magwood IV, signed a bilateral agreement on June 17, 2003, with the Assistant Deputy Minister of the Department of Natural Resources Canada, Ric Cameron, and the Senior Vice-President Technology of Atomic Energy of Canada Limited, David F. Torgerson. The first U.S./Canada collaborative research projects were awarded in FY 2004.

6.1 Work Scope Areas

R&D topical areas for the U.S./Canada collaboration include:

- Hydrogen production by nuclear systems
- Sustainable and advanced fuel cycles
- Supercritical water-cooled reactor concepts

6.2 Project Summaries

In FY 2004, the initial year of the collaboration, seven research projects were initiated. These projects continued into FY 2006, with three being completed during the year. A listing of the I-NERI U.S./Canada projects that are currently underway follows, along with summaries of the accomplishments achieved in FY 2006.

Directory of Project Summaries

2004-001-C	High-Temperature Electrolyzer Optimization	21
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PI (U.S.): Richard D. Doctor, Argonne National Laboratory	Project Number: 2004-001-C		
	Project Start Date: May 2004		
PI (Canada): Atomic Energy of Canada Limited	- j		
(AECL)	Project End Date: December 2006		
Collaborators: Chalk River Laboratories, Idaho			
National Laboratory (INL)			

High-Temperature Electrolyzer Optimization

Research Objectives

The objective of this project was to thermally optimize a combined nuclear hydrogen production facility consisting of a high-temperature electrolysis (HTE) plant powered by a very high-temperature gas reactor. Researchers conducted two primary analyses to achieve economic optimization: 1) a computational fluid dynamics (CFD) analysis of individual flow channels in the HTE cells and 2) a balance-of-plant process design analysis.

Two basic cell configurations were proposed: 1) tubular and 2) planar, which is also termed "bipolar" or "flat plate." In this research, the CFD and electrochemical modeling were limited to flow pattern analysis within a single, planar HTE cell. Heat transfer and electrochemical efficiencies will degrade, depending on the extent the flow field of each cell stack deviates from uniformity.

To achieve the highest net efficiency and the lowest cost, researchers must completely understand the thermal and flow processes inside and outside of the cell. The results of HTE cell computational fluid dynamics models were combined with balance-of-plant process design studies from the Aspen[™] code to facilitate a workflow for overall plant design optimization. This enabled researchers to fully evaluate the required energy transfer, temperatures and the overall system efficiency of different system designs.

Using the results from an economic model building on Aspen[™], researchers compared HTE cells operating at a variety of hydrogen-steam feed inlet ratios against conventional electrolysis. Specifically, they evaluated two NorskHydro 50,000 nm³/h commercial electrolyzers and an HTE system, each delivering 9 tonnes/hour of hydrogen (H_2) .

Research Progress

Computational Fluid Dynamic Model of HTEcells. The researchers developed a model combining the governing electrochemical mechanisms with the heat transfer and fluid dynamics processes taking place during operation of the HTE cells. The finite element model combines an electrochemical (EC) module developed inhouse with the commercially available computational fluid dynamics (CFD) code, STAR-CD. This model simulates a three-dimensional HTE cell and calculates the local EC kinetics of the cells coupled to the mass- and heat-balances of the gaseous flow and the solid medium. The main coupling between the EC and CFD modules is through the temperature profile: 1) the CFD module provides a temperature field for the EC module to generate the current density distribution using temperature-dependent electrochemical parameters, while 2) the EC module provides species and heat generation rates, based on the calculated current density, for the CFD module to generate a consistent temperature profile.

The representative model of an HTE cell (Figure 1) consists of 12 layers, each divided into 40x40 elements, giving 19,200 elements per cell in the complete finite element model. The sensitivity of results with further division into a finer mesh was negligible.



Figure 1. Effect of the hydrogen electrode limiting current density on the (a) polarization and voltage efficiency, (b) average Nernst potential, and (c) average temperature in the HTE cells.

Researchers conducted a sensitivity analysis to investigate the influence of the following parameters on efficiency, temperature, and current density profiles:

- Electrochemical properties (i.e., oxygen electrode exchange current density and hydrogen electrode limiting current density)
- Hydrogen and nitrogen mass fractions at the cathode inlet
- Flow configuration (cross-flow and parallel)

For each case, they studied the polarization behavior of the simulated cell with an applied potential of 0.8–1.6V. The next steps would be to investigate HTE materials and operating conditions, vary the model inputs accordingly, and compare these data against experimental results for polarization, temperature, and flow.



Balance-of-plant process design and economics. To achieve the highest net efficiency and the lowest cost, researchers must thoroughly understand the thermal and flow processes outside of the HTE cells. Using the ASPEN[™] software package, they conducted the balance-of-plant process design or "flowsheet" analysis for high-temperature electrolysis. They settled on a production target of 9 tonnes per hour of dry hydrogen at 413.7 bar (6,000 psig), assuming a 50 percent conversion rate of steam to hydrogen. Directly quenching the hot hydrogen product with feed water to generate steam may prove a practical route to maximize heat recovery from the process. Air stripping the hot oxygen product gained further heat recovery. The researchers performed the following case studies:

1) Moderate pressure operation (3 bar), for inlet feeds of:

- 50% H₂O: 50% H₂
- 67% H₂O: 33% H₂
- 75% H₂O: 25% H₂
- 90% H₂O: 10% H₂
- 2) High pressure operation (15 bar), for inlet feeds of:
 - 75% H₂O: 25% H₂

In each case, the researchers considered a schematic of the solid oxide electrolysis cells as they would appear in the balance-of-plant configuration, the major stream compositions, temperatures, pressures, and thermodynamic properties. They also evaluated the overall system configuration and connectivity options, including various options for recycle streams and heat exchangers.



Figure 2. Direct and recycle steam feeds show an optimum at 33% H_2 :67% steam for 50% steam conversion per pass mapping into tabular results for H_2 economics for various electric/heat costs.

The optimum energy balance for the conservative HTE case occurred at 67 percent steam and 33 percent H_2 (see Figure 2), which was also the most economical ratio. Although the cost analysis for delivering nuclear heat requires further refinement, high-temperature electrolysis is attractive at current industrial electricity rates of \$52.70/ MWh (as reported by the U.S. Energy Information Agency for 2005.) Hydrogen production by HTE would cost \$3.71/ kg H_2 , while the NorskHydro system would cost \$3.88/kg.

To investigate the economic feasibility of these processes, researchers need to integrate the analysis of fluid transport and heat transfer with financial considerations. Table 1 presents a side-by-side comparison of the optimal HTE case with the NorskHydro system. Although a high-temperature electrolysis hydrogen production plant will involve significant up-front investment, this comparative economic analysis provides a basis for system selection.

Planned Activities

This I-NERI project has been completed.

System	NorskHydro		HTE - 67% steam	
0	Capita	Cost	Capital Cost	
Cost Component	Percentage	\$K	Percentage	\$K
Direct Costs				
Electrolyzer stack	26.80%	\$37,788	22.85%	\$43,785
Electrolyzer shell & manifold piping	2.85%	\$4,019	2.43%	\$4,656
High-temperature electrolysis equipment	0.00%	\$0	20.47%	\$39,228
Electric Power Transformer	7.00%	\$9,870	2.98%	\$5,718
Electric Power Invertor/Conditioning	21.00%	\$29,610	8.95%	\$17,155
Compressor	13.35%	\$18,824	10.88%	\$18,824
Recycle compressor	0.00%	\$0	2.47%	\$3,489
Support Facilities				
Pipe, valves, fittings, $H_2 \& O_2$ tanks	7.00%	\$9,870	7.00%	\$13,412
Process instruments and controls	7.00%	\$9,870	7.00%	\$13,412
Electrical equipment and materials - support	5.00%	\$7,050	5.00%	\$9,580
Structural support, insulation, paint	10.00%	\$14,100	10.00%	\$19,160
Direct Costs Sub-total	100.00%	\$141,000	100.04%	\$191,600
Indirect Costs *				
Erection and installation labor	22.00%	\$31,020	22.00%	\$42,152
General Facilities	7.00%	\$9,870	7.00%	\$13,412
Engineering Fees	10.00%	\$14,100	10.00%	\$19,160
Profit	15.00%	\$21,150	15.00%	\$28,740
Indirect Costs Sub-total		\$217,140		\$295,064
Total Costs				
Direct + Indirect Costs Sub-total		\$358,140		\$486,664
Interest & Inflation	0.00%	\$0	0.00%	\$0
Total Plant Investment-TPI		\$358,140		\$486,664
Royalties	0.60%	\$846	0.60%	\$1,150
Start-up Costs	1.00%	\$1,140	1.00%	\$1,916
Commissioning & spare parts	0.35%	\$1,242	0.35%	\$1,687
TOTAL PROJECT COSTS		\$361,638		\$491,417

Table 1. Comparison of capital costs for NorskHydro and HTE.

* Indirect costs are expressed as a percentage of Direct Costs Subtotal.

Development of Inert Matrix Fuels for Plutonium and Minor Actinide Management in Water Reactors

PI (U.S.): J. Carmack, Idaho National Laboratory (INL)	Project Number: 2004-002-C
	Project Start Date: October 2004
PI (Canada): P. Boczar, Atomic Energy of Canada Limited (AECL)	Project End Date: September 2008
Collaborators: University of Florida, Los Alamos National Laboratory, Brookhaven National	

Research Objectives

Laboratory

The objective of this project is to develop inert matrix fuels (IMFs) for the stabilization or burn-down of plutonium in existing commercial power reactors. IMFs offer the advantage of more efficient destruction of plutonium (Pu) and minor actinides (MA) relative to mixed oxide (MOX) fuel. Efficient plutonium reduction results in 1) greater flexibility in managing plutonium inventories, 2) improved strategies for disposing of MAs, and 3) potential fuel cycle cost savings. Because fabrication of plutonium- and MA-bearing fuel is expensive relative to uranium oxide (UO_{3}) , a cost benefit can be realized by reducing the number of Pu-bearing elements required for a given burn rate. In addition, the choice of matrix material may be manipulated either to facilitate fuel recycling or to improve proliferation resistance by making plutonium recovery extremely difficult. Furthermore, IMF having high thermal conductivity may provide operational and safety benefits. The resulting lower fuel temperatures can improve operating and safety margins and increase rated reactor power.

The Canada deuterium uranium (CANDU) reactor is the model design considered in this study, as it offers flexibility in plutonium management and MA burning. It can use a full core of IMF, containing either Pu or a plutonium-actinide mix with Pu destruction efficiencies greater than 90 percent and actinide destruction exceeding 60 percent. The advanced CANDU reactor (ACR) offers additional possibilities in the design of an IMF bundle, allowing additional fissile material in the center region of the bundle to improve actinide burning. The ACR would provide flexibility for managing both plutonium and MA from the existing light water reactor (LWR) fleet. Many of the fundamental principles concerning the use of IMF, such as fuel/coolant compatibility, fuel fabrication, and fuel irradiation behavior, also apply to other types of LWRs and supercritical water-cooled reactor (SCWR) technology. An IMF with high thermal conductivity would be particularly beneficial to any SCWR concept.

Research Progress

This project was inactive in 2006. Progress to date includes:

Fuel Selection. The researchers established goals for IMF development, assessed IMF candidates against this baseline, and selected several fuel materials for further study. They analyzed neutronic behavior (Pu burn rate and reactivity swings), fuel thermal and irradiation performance, and in-core corrosion resistance based on existing knowledge.

Fabrication Development. Researchers developed and qualified fabrication methods for inert-matrix fuels (MgO-ZrO₂ and Zr-metal matrix) and performed test fabrication and characterization. Figure 1 is a photograph of a plutonium-bearing MgO-ZrO₂ matrix pellet and Figure 2 shows a zirconium-metal matrix fuel sample. They initiated development of a candidate silicon carbide (SiC) IMF composition and fabricated and characterized preliminary fuel samples.



Figure 1. Photograph of dual-phase MgO-ZrO₂-PuO₂ pellet.

Figure 2. UO_2 particles extruded in a Zr-metal matrix.

Characterization. Researchers completed the experimental characterization of candidate IMF and matrix materials. They evaluated microstructure, thermal and mechanical properties, corrosion testing, and ionbeam irradiation to determine suitability for in-reactor irradiation testing and to provide a basis for fuel behavior modeling. Further refinement of the fabrication processes is necessary.

Fuel modeling. The team has constructed a finite element model using the ABAQUS code to model the phase structure of both heterogeneous and homogenous inert matrices. This code allows researchers to estimate key fuel properties, such as thermal conductivity and density, which are needed to support reactor irradiation experiment design and qualification. They conducted simulations with a plutonium-only SiC IMF and one also containing actinides. Plutonium, with or without actinides, was added to the outer two rings, while the central seven pins contained gadolinium burnable poison to suppress initial reactivity, reduce the refueling power ripple, and ensure negative void reactivity.

For the plutonium-only case, assuming 250 g of Pu per bundle and a refueling rate of 15 bundles per day, each CANDU unit can annihilate approximately one metric ton of plutonium per year. At an 80 percent capacity factor, 94 percent of fissile Pu is destroyed. Bundle and channel power remain within current licensing limits.

Fuel in the plutonium-plus-actinides simulation consisted of 356 g of Pu and 44 g of actinides (neptunium, americium, and curium). Sixty percent of the actinides and 90 percent of the plutonium are destroyed in this simulated process, amounting to approximately 70 kg of actinides and 860 kg of Pu per reactor per year.

Reactor physics, safety, and licensing analysis. Researchers performed four-bundle neutronic, thermal-hydraulic, and transient analyses of proposed inert matrix materials and compared the results with similar analyses of reference uranium-oxide fuel. The results will be used to screen the general feasibility of utilizing specific inert

matrix fuel compositions in existing and future light water reactors. Compositions identified as feasible will require further detailed analysis coupled with rigorous experimental testing and qualification.

Irradiation Testing and Post-irradiation Examination. A screening irradiation test designated LWR-2 is currently in the design and planning stages at the Advanced Test Reactor (ATR) at Idaho National Laboratory. This test will include a variety of candidate IMF compositions. Researchers will report post-irradiation examination (PIE) results from a preliminary irradiation test (LWR-1a) conducted on a sample of advanced MOX that incorporates approximately 4,000 ppm neptunium oxide. Figure 3 is a neutron radiograph of the LWR-1a rodlet following exposure in the ATR.

Figure 3. Neutron radiograph of $94\% dUO_2-6\%~RGPuO_2-<1\%~NpO_2~LWR-1a~MOX$ rodlet irradiated to 9 GWd/tU burnup.

SiC is expected to have adequate resistance to structural damage from neutrons, but fission fragments can cause damage. In order to simulate this damage, the researchers bombarded SiC-IMF and several other candidate materials with high-energy (72 MeV) iodine ions. Following exposure, profilometry examinations to detect any relief in the polished surface showed that only Zirconia, SiC, and UO_2 were unaffected under all test conditions. This result supports, but does not prove, that SiC-IMF will be stable in the reactor.

Planned Activities

Future efforts are expected to focus on fabricating fuel test specimens to be included in the LWR-2 irradiation test. This test will include a variety of candidate IMF compositions, as shown in Table 1, fabricated for both out-of-pile characterization and in-reactor irradiation tests. Figure 4 shows a schematic diagram of an LWR-2 test assembly rodlet that will contain one of the individual fuel specimens. The test assembly was originally planned to be installed in the spring of 2007.



Figure 4. 3-D representation of LWR-2 fuel irradiation test pin.

Designation	Description	Composition	Fabricator
LWR-2-A	RG-MOX at high burnup (LWR-1a) (U,Pu)O ₂		INL
LWR-2-B	Advanced MOX (Np, Am additions)	(U,Pu,Np,Am)O ₂	INL
LWR-2-C	IMF replacement to reference MOX	(Pu)O ₂ - MgO-ZrO ₂	INL
LWR-2-D	IMF (Np addition)	(Pu,Np)O ₂ , MgO-ZrO ₂	INL
LWR-2-E	IMF replacement to Advanced MOX	(Pu,Np,Am)O ₂ , MgO-ZrO ₂	INL
LWR-2-F	Sphere Pac target (low fertile matrix)	$UO_2 - AmO_2$	ORNL
LWR-2-G	Sphere Pac target (inert matrix)	PuO ₂ or ZrO ₂ - AmO ₂	ORNL
LWR-2-H	PuO ₂ Cercer - Cermet	PuO ₂ , YSZ or Mo (if feasible)	EURATOM
LWR-2-I	Pu - FeCr Ferritic (ITU fabrication)	Pu, Fe/Cr	EURATOM
LWR-2-J	SiC Inert Matrix	UO ₂ -SiC IMF	AECL
LWR-2-K	Advanced IMF (Univ. of Florida)	PuO ₂ - MgO-ZrO ₂ -Nd or Yb	UF/INL
LWR-2-L	Zr metal inert matrix	(PuO ₂ , Zr)	INL
LWR-2-M	WG-MOX at high burnup (LWR-1a)	(U, Pu)O ₂	INL

Table 1. Summary of IMF compositions for LWR-2.

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Evaluation of Materials for Supercritical Water-Cooled Reactors

PI (U.S.): D. Wilson, Oak Ridge National Laboratory

PI (Canada): H. Khartabil, Atomic Energy Canada Limited (AECL)

Collaborators: University of Wisconsin, University of Michigan, University of Notre Dame, University of Sherbrooke

Research Objectives

The goal of this project is to establish candidate materials for supercritical water reactor (SCWR) designs and to evaluate their mechanical properties, dimensional stability, and corrosion resistance. This project will address critical issues related to radiation stability, corrosion, and stress corrosion cracking performance in candidate materials for the SCWR.

Research Progress

Researchers performed corrosion testing of candidate ferritic/martensitic (F/M) steels, austenitic steels, and nickel-based alloys in both subcritical and supercritical

water at temperatures of 360 to 600°C for periods of time up to 1,026 hours. Surface modifications and grain boundary engineering (GBE) treatments were applied on selected material samples.

Figure 1 presents weight gains for most of the materials tested. Weight gain due to oxidation in supercritical water (SCW) was typically smaller but less predictable on austenitic stainless steels than in F/M steels. Nickel-based alloys showed a fairly good corrosion resistance in an SCW environment and had the lowest weight gain of all the tested materials. A 9Cr oxide-dispersion Project Number: 2004-003-C

Project Start Date: October 2004

Project End Date: September 2007

strengthened (ODS) steel showed the lowest weight gain among the F/M materials.

Figure 2 shows significant oxidation at the grain boundaries at the internal oxidation zone of the ODS steel. Furthermore, yttrium (Y) segregated to the grain boundary area in the internal oxidation layer.

Figure 3 shows a specimen surface modified by adding a layer of yttrium. An yttrium-rich Y–Fe–O oxide layer formed after exposure to SCW at 500°C, separating the magnetite into two layers with different microstructures. This may have influenced the diffusion of cations and/ or anions, resulting in significantly improved corrosion resistance.



Figure 1. Comparison of weight gain data for the candidate materials after exposure to subcritical/supercritical water with different oxygen concentrations for \sim 333 h.



Figure 2. TEM image showing grain morphology and boundaries in the internal oxidation layer formed in 9Cr ODS steel after 1,026 hours exposure to 500°C SCW.

Austenitic alloy 800H showed low weight gain in SCW; however, it suffered from extensive spallation of its outer magnetite scale. The researchers performed grain boundary engineering (GBE) to mitigate this. As shown in Figure 4, the scale on the GBE-treated alloy, 800H, became compact and continuous as compared to that on the control sample, which correlates with the higher weight gain. Increasing the fraction of low- Σ coincidence site lattice boundaries led to improved oxide stability.

The researchers conducted stress corrosion cracking (SCC) experiments on samples of alloys 316L and 690 irradiated with 3 MeV protons to a dose of 7 dpa (at

Figure 3. SEM image showing the magnetite layer microstructure in 9Cr ODS steel coated with a thin yttrium film after exposure to 500° C SCW for 667 hours.

400 and 500°C, and 25 MPa pressure). The deaerated condition resulted in a dissolved oxygen concentration below 10 ppb and outlet water conductivity below 0.1 μ S/ cm. A measure of the extent of cracking is shown in Figure 5. At 400°C, the cracking susceptibility of 316L irradiated to 7 dpa is 1.1 times higher than an unirradiated sample and 2.8 times higher for alloy 690. This increases to 6 times for alloy 316L and 48 times for alloy 690 at 500°C.

The irradiated microstructure is dominated by small (7 nm) faulted Frank loops at 400°C, and 25 nm Frank loops with voids at 500°C. Irradiation hardening was greater at the lower temperature, which agrees with the dispersed



Figure 4. Effect of GBE treatment on the corrosion resistance of austenitic alloy 800H after exposure to SCW at 500°C.


Figure 5. Influence of irradiation on the cracking susceptibility of alloy 316L and 690 in 400°C and 500°C SCW.

barrier-hardening model. Hardening was also greater on 316L than 690 at both temperatures. Irradiation resulted in grain boundary depletion of Cr, Fe, and Ni enrichment, with segregation increasing with dose. The higher temperature resulted in slightly greater radiation induced segregation (RIS). However, neither RIS nor hardening satisfactorily accounts for the changes in SCC susceptibility with irradiation temperature. To improve the cracking behavior of alloys 316L and alloy 690, the researchers used GBE to enhance the coincident site lattice boundary (CSLB) fractions—hot rolled between 982 and 1,149°C and then annealed at 1,038 to 1,093.3°C. Due to the presence of chromium carbides in the as-received (AR) condition, it was necessary to perform a short anneal at 1,100°C to dissolve the carbides. The resulting structure had a very high CSLB fraction of 76 percent, which researchers reduced by compressing the sample and recrystallizing at 1,100°C for 2 hours. Another compression and 7.5 minute anneal at 1,000°C reduced the CSLB fraction to 35 percent, leaving grain size unchanged (Figure 6). Researchers will test the effect of the CSLB fraction on the cracking behavior of alloys using this sample.

Work continued on characterizing the corrosion rates of candidate alloys. Initial test results showed a rapid increase in corrosion rates above 550°C, which agrees with data on other alloys in SCW. In the initial tests, researchers measured the temperature dependence of the corrosion rate of type 403 stainless steel at up to 650°C. They are developing a detailed database of corrosion rates to compare with existing data and to aid in selecting promising candidate alloys. Researchers also measured the corrosion rate of Inconel 690 and A106B carbon steel and characterized the corrosion films using secondary ion mass spectrometry (SIMS) and Raman spectroscopy. Although analysis is still underway, the results are consistent with the formation of an iron-chromium oxide and/or oxyhydroxide.



Figure 6. Comparison of two alloy 690 conditions showing large differences in CSLB fraction: left – 76%, right – 32%.

Researchers re-started work on the deposition of anti-corrosion coatings with the atmospheric pressure plasma jet, concentrating on handling the metalloorganic precursor materials (zirconium-tert butoxide) and spectroscopic characterization of the plume. They observed spectroscopic evidence for the break-up of precursor molecules in the plasma jet plume and are optimizing the process conditions for deposition of uniform zirconium dioxide films.

For the sol-gel dip coating process, researchers continued assessing the corrosion resistance that ZrO_2 films impart on various steels in SCW, considering parameters such as coupon preparation, colloid preparation, dipcoating procedure and sintering procedure. Modifications to the colloid distillation process allowed distillation under vacuum. They examined the surfaces of the coated coupons using auger scanning electron microscopy, secondary ion mass spectrometry (SIMS), and atomic force microscopy. X-ray diffraction examinations showed that the sintering process converts amorphous ZrO_2 colloid to the tetragonal ZrO_2 phase. SIMS examination of surface films formed on coated steel show a chromium-rich oxide between the metal surface and the ZrO_2 coating, damaging the coating integrity.

Additionally, researchers performed Monte Carlo calculations of the yields Ge_{aq} and of $(Ge_{aq} + GOH + GH)$ as functions of water density in SCW. Calculated yield curves cannot reproduce the measured yields at low water densities (e.g., below about 0.3 g/cm³), as shown in Figure 8. Calculations of the ratio of "initial" H atom yields to



Figure 8. Comparison between Monte-Carlo predictions and experimental data.

hydrated electron as a function of water density gave relatively constant values near unity, regardless of density. This compares well with the literature at 360-380°C, but fails to reproduce the strong increase observed below 0.5 g/cm³. Discrepancies between experimental data and the Monte Carlo simulation results indicate that SCW radiolysis is dominated by the gas-phase component.

Planned Activities

Researchers plan to conduct the following activities over the next fiscal year:

- Continue corrosion studies, including evaluating promising candidate materials, water chemistry effects, radiation effects, and temperature effects
- Continue efforts to optimize corrosion performance through surface modification and grain boundary engineering
- Continue to investigate deposition of corrosion-resistant coatings using the plasma jet
- Continue optimization of the sol-gel ZrO₂ coatings
- Evaluate the efficacy of pressurized tubes for studying SCC
- Continue evaluating alloys 316L and 690 to determine the dose and temperature dependence of SCC; irradiate alloys D9 and 800H to provide complementary data
- Perform SCC testing on CSLB optimized specimens of alloys 690 and 316L in the unirradiated and irradiated states
- Continue work to understand SCW radiolysis
 - Develop a Monte-Carlo program to simulate the radiolysis of water vapor
 - Identify the effects of phase differences and assess physical or/and chemical basis
 - Develop molecular dynamics calculations for SCW over a wide range of densities
 - Simulate radiolysis of SCW to quantitatively determine radiolytic yields

ACR Hydrogen Production for Heavy Oil Recovery

PI (U.S.): J. S. Herring, J. E. O'Brien, Idaho National Laboratory (INL)

PI (Canada): R. Sadhankar, Atomic Energy of Canada Limited (AECL)

Project Number: 2004-004-C

Project Start Date: June 2004

Project End Date: May 2007

Collaborators: Chalk River Laboratories

Research Objectives

The objective of this project is to analyze the feasibility of using the advanced CANDU reactor (ACR) for nuclear hydrogen production and as a source of steam for enhanced oil recovery. For hydrogen production, researchers are investigating the use of a high-temperature electrolysis (HTE) process coupled to the ACR. HTE has been proposed as an efficient hydrogen production technology. This process uses electrical power and high-temperature process heat, typically supplied by an advanced high-temperature reactor, to produce hydrogen. An advantage of HTE over thermochemical hydrogen production is its much smaller high-temperature process heat requirement. Consequently, HTE can potentially be driven by a conventional nuclear reactor, using lower temperature nuclear heat supplemented by electrical resistance (ohmic) or combustion-based heating, to achieve the necessary HTE operating temperature (800–900°C).

Researchers will develop a preliminary conceptual design for the electrical/thermal integration of an ACR for hydrogen production and the production of steam for oil sands heating and mobilization. They will assess the costs of hydrogen and steam production for comparison with the economics of low-temperature electrolysis (LTE) and other means of hydrogen production. The economic analysis will include an evaluation of hydrogen and steam production using fossil fuels in the Athabasca oil sands project under construction in Alberta, considering the price of petroleum recovered.

Research Progress

The researchers accomplished the following specific activities in FY 2006:

- Incorporated a 1-D integral electrolyzer model into a HYSYS ACR plant model coupled to a low-temperature electrolysis plant
- Performed scoping simulations to assess the effects of operating conditions, pressure, gas flow rates, stack resistance, and gas compositions (e.g., steam vs. air sweep vs. no sweep)
- Evaluated various system configurations of pumps, compressors, and turboexpanders
- Compared the hydrogen-production performance of the ACR-LTE system to an ACR-HTE system based on detailed modeling

Following is an overview of the research progress made to date.

As shown in Figure 1, the team developed a model of a high-temperature electrolysis plant coupled to an ACR using the HYSYS analysis code. The HYSYS model includes a custom electrolyzer module which enabled the team to evaluate various system configurations and operating conditions in order to assess ACR-HTE concepts and perform system optimization. Based on these results, they developed an ACR-HTE conceptual design and compared performance to a baseline ACR-low-temperature electrolysis system.



Figure 1. HYSYS process flow diagram for an ACR-700 HTE plant, with air sweep.

In order to perform the technical evaluation, researchers considered the basic thermodynamics of HTE and developed detailed system-level modeling capabilities, which they used to establish a preliminary conceptual design of the overall process. A conventional LTE system coupled to an ACR served as the baseline process for comparison. The HYSYS model included a custom, onedimensional, electrolyzer module incorporated directly into the code that enabled the team to determine operating parameters (such as voltage, gas outlet temperatures, and electrolyzer efficiency) for any specified inlet conditions (such as gas flow rates, current density, cell active area, and external heat loss or gain).

Using HYSYS, researchers defined a detailed process flow diagram that included all of the components present in an actual plant (such as pumps, compressors, heat exchangers, turbines, and the electrolyzer). To validate the one-dimensional electrolyzer model, they compared results against a 3-D computational fluid dynamics (CFD) model developed using FLUENT. This indicated overall thermalto-hydrogen efficiencies around 33–34 percent, with only a weak dependence on current density. These values compare favorably with corresponding overall thermalto-hydrogen efficiencies achievable with low-temperature electrolysis. The team carried out a preliminary economic analysis to assess the market for using hydrogen for oil sands development. It is clear that hydrogen demand will be huge (upward of 6,000 tonne/d) and the industry is searching for alternatives to steam reforming of natural gas. A contract study is being put in place to solidify understanding of the hydrogen market in northern Alberta and how the oil sands industry plans to produce it.

Over the past fiscal year, the team performed additional HYSYS analyses to further the HTE system modeling experience. They developed a slightly revised process-flow diagram for the air-sweep case that includes a two-stage air compressor with intercooling on the sweep gas. This more realistically represents the compression process for the large compression ratio required. They also considered an additional modification to the process flow diagram— eliminating the intermediate heat exchanger (IHX)—which would allow direct steam supply from the steam generator to the HTE process loop. This strategy avoids the irreversibilities associated with heat transfer across the IHX. Also, because this modification requires that the HTE loop be operated at the same pressure as the power-cycle loop, it allows for higher pressure hydrogen delivery.

The biggest change in the revised analyses was the imposition of a constraint on the heat exchangers to operate with a realistic 95 percent effectiveness, rather than using 5°C minimum approach temperatures as was done previously. The only exception is the steam generator, which required a slightly higher effectiveness of 96.6 percent in order to maintain the required pressure values on both the power-cycle and high-temperature electrolysis sides of the overall process.

The revised analysis yielded power-cycle and hydrogen production efficiencies that were both 2–3 percent lower, compared to the original values, reflecting increased irreversibilities associated with the heat exchanger effectiveness constraint. Results of the revised analysis with direct-steam to the HTE process (no IHX) showed similar efficiencies, but with a higher hydrogen delivery pressure of 7 MPa. If the delivery pressure is taken into account, these results are promising.

Planned Activities

Over the upcoming period, researchers will complete their assessment of the market and opportunity for nonconventional hydrogen production. After completing the ongoing conceptual design study, they plan to conduct additional economic analyses.

Supercritical Water-Cooled Reactor Stability Analysis

PI (U.S.): Won Sik Yang, Argonne National Laboratory (ANL)

PI (Canada): Hussam Khartabil, Atomic Energy of Canada Limited (AECL)

Collaborators: Massachusetts Institute of Technology, University of Manitoba, École Polytechnique de Montréal Project Number: 2004-005-C

Project Start Date: October 2004

Project End Date: September 2006

Research Objectives

The objective of this project was to improve the understanding of potential instability problems of the supercritical water cooled reactor (SCWR). The aim was to develop improved stability analysis tools and to define appropriate ranges for important design parameters that will ensure system stability using a combination of experimental and analytical techniques. Researchers planned to develop two codes: 1) a frequency domain linear stability analysis code applicable to SCWR thermalhydraulics and thermal-nuclear coupled stabilities and 2) a thermal-hydraulic stability analysis tool for studying the adequacy of SCWR sliding pressure startup and shutdown procedures.

Researchers planned to assess the suitability of the CATHENA and SPORTS codes for performing flow stability analysis at supercritical conditions and to implement needed modifications. Additionally, they planned to investigate coupled neutronic and thermal-hydraulic instabilities that can arise in a pressure tube reactor running at supercritical conditions using the neutron transport code DRAGON, together with a suitable thermalhydraulics code to carry out the analyses.

For the experimental study, researchers investigated flow instability of supercritical fluids using an existing supercritical CO₂ loop at ANL. The Canadian investigators constructed an experimental loop to study parallel-channel flow instabilities of water at supercritical conditions, using CO₂ and water as modeling fluids. They also planned to perform single and multi-channel flow instability tests.

Research Progress

Researchers developed the frequency domain linear stability code, SCWRSA, for thermal-nuclear coupled stability analysis of SCWRs. In order to investigate the effects of water rods used in the current U.S. reference SCWR design, the research team employed single-channel thermal-hydraulics models for the coolant channel and water rod, along with point kinetics neutronics and onedimensional fuel heat conduction models. They developed a multi-channel thermal-hydraulics analysis capability by combining these single-channel models with a parallelchannel flow distribution model. The multi-channel capability can model parallel channels either with or without a water rod. It can also model either a vertical or horizontal core configuration.

The steady-state flow distribution among parallel thermal-hydraulics channels was calculated under a fixed total flow rate and an equal pressure drop boundary condition; the coolant and water-rod flow rates were simultaneously determined by taking into account the heat transfer between coolant and water rod. Time-dependent behavior of water in the inlet plenum was approximated by two bounding inlet boundary conditions: 1) instantaneous mixing and 2) constant mixed-mean enthalpy of the water in the inlet plenum. The instantaneous mixing boundary condition neglects the time delay of mixing in the inlet plenum, while the constant mixed-mean enthalpy boundary condition assumes that water in the inlet plenum is maintained at steady-state enthalpy during the time of interest. Preliminary verification tests of both the single-channel and multi-channel analysis capabilities of SCWRSA have been performed using models derived from the U.S. reference SCWR design. Although individual assemblies can be represented as separate channels using the multichannel capability, the team used two-channel models in these tests because of the lack of information on the core power distribution (except for the target values of power peaking factors).

The team analyzed both the Dittus-Boelter and the Jackson heat transfer coefficient correlations. They observed that heat transfer between coolant and water rod affects the steady-state flow distribution. As shown the Bode diagram in Figure 1, the decay ratios obtained with the two-channel models were smaller than those found with the single, average-channel models. This is because the two-channel model includes hot channel assemblies that introduce larger Doppler and coolant density feedbacks than the average-channel assemblies. The instantaneous mixing approximation produces slightly larger decay ratios than the constant mixed-mean enthalpy approximation. The decay ratio for thermal-nuclear coupled stability estimated with the two-channel models was less than 0.17, well below the limit of 0.25 traditionally imposed for BWR stability.



Figure 1. Bode diagram for thermal-nuclear coupled stability of twochannel vertical test problem with water rods.

The research team also developed a thermal-hydraulic stability analysis tool to study the adequacy of SCWR sliding pressure startup and shutdown procedures. Based on a single-channel thermal-hydraulics model, they developed stability boundary maps for density wave oscillations for both supercritical and subcritical pressure conditions, and applied them to the U.S. reference design. They used a model at supercritical pressures consisting of three regions: 1) a "heavy fluid" region, 2) a mixed "heavy fluid"/"light fluid" region similar to the homogeneousequilibrium two-phase mixture, and 3) a "light fluid" region. Two important non-dimensional groups, a pseudosubcooling number and an expansion number, were defined as controlling the stability. Plotting the stability maps on a plane consisting of these two numbers using a frequency domain analysis of the channel shows that the U.S. reference design operates in the stable region with a large margin. Sensitivity studies produced results consistent with the trends of the earlier (subcritical pressure) two-phase flow models.

During the sliding pressure start-up operation, a two-phase steam-water mixture at subcritical pressure occurred in the reactor core. Researchers applied a nonhomogeneous (i.e., drift-flux) non-equilibrium two-phase flow model to this condition. The characteristic equation was numerically integrated, and the stability boundary maps were plotted on the traditional subcooling number versus phase change number plane. These maps were used to modify the sliding pressure start-up strategies of the SCWR to avoid thermal-hydraulic flow instabilities.

The researchers completed their assessment of the CATHENA and SPORTS codes and implemented the modifications needed to perform flow stability analysis at supercritical conditions. The thermo-physical properties at supercritical conditions were added to CATHENA using the properties package from the National Institute of Standards and Technology (NIST). Because SPORTS already has supercritical properties, the assessment compared a simple linear stability analysis for consistency. The results showed very good agreement between the linear model and the SPORTS code. It appeared that SPORTS is more suitable for performing stability analysis.

Using the preliminary CANDU-SCWR fuel design, researchers created input files for the DRAGON code. Water properties at supercritical conditions were implemented into the code to model the large density variations as temperature increase beyond the critical point. Researchers completed preliminary studies of the relationship between the thermal properties of materials in the cell (water density and fuel temperature) and the effective multiplication constant. In addition, they explored a strategy to couple DRAGON with a thermal-hydraulic code. In a related NERI project, researchers performed natural circulation tests with horizontal heating and cooling sections at the existing supercritical CO_2 loop at ANL. They did not observe flow instabilities with the loop, although previous numerical studies indicated the existence of such instability when the fluid is heated through the pseudocritical point. The system remained stable for a variety of system perturbations. The effects of the pressurizer, which were not included in previous numerical studies, were investigated. Researchers concluded that the pressurizer was not responsible for the discrepancy between experiment and calculations. Continued modeling work to identify the source of calculated instabilities is necessary. Construction of the parallel channel instability loop in Canada was completed in 2005. The initial plan was to build the loop to withstand operation at supercritical conditions and then build two test sections: one for CO_2 and one for water. However, the plan was modified so that a single test section could accommodate both fluids, due to the complexity of the design option with interchangeable test sections. Since then, AECL designed and constructed a data acquisition and control system. This system was sent to the University of Manitoba for installation.

Planned Activities

This project was completed in September 2006.

Thermal-Hydraulic Benchmark Studies for SCWR Safety

PI (U.S.): J. R. Wolf, Idaho National Laboratory (INL)

PI (Canada): H. Khartabil, Atomic Energy of Canada Limited (AECL)

Collaborators: École Polytechnique de Montréal, University of Manitoba, Rensselaer Polytechnic Institute

Research Objectives

The original objective of this project was twofold: 1) to address the critical issues associated with measuring heat transfer to supercritical water at prototypical conditions in a supercritical water reactor (SCWR) and 2) to develop tools to predict SCWR thermal transients. In addition to using supercritical water, researchers planned to use surrogate fluids at supercritical conditions. These alternative fluids can provide valuable insight into physical phenomena and can significantly reduce the cost and time to complete an experimental program.

At the 2006 U.S-Canadian I-NERI review meeting, it was recommended that this particular collaboration be suspended because of changing priorities in both the U.S. and Canada that cut funding for SCWR research.

Research Progress

The U.S. was to design and build a four-rod test section that would be used to determine heat transfer characteristics under prototypical supercritical water conditions. However, due to the reduced project scope, completing the test section will be the last thermalhydraulic activity. All additional thermal-hydraulic activities including the actual fabrication of the test section and the associated testing have been cancelled. Table 1 provides the requirements to which the test section was designed and Figure 1 provides a detailed schematic of the test section itself.

Project Number: 2004-006-C

Project Start Date: October 2004

Project End Date: September 2006

The Canadian side identified necessary modifications to the existing CO_2 loop, but major component purchases (e.g., pump) were delayed due to insufficient funds. Work on the CO_2 bundle simulator (which must be identical to the water bundle simulator for fluid scaling studies) was stopped because of uncertainties regarding construction of the water bundle simulator. Alternatives are being investigated, including getting access to existing water data from Russia.

Planned Activities

There are no further activities planned for this project. At the 2006 U.S.-Canadian I-NERI review meeting, it was recommended that this particular collaboration be suspended because of changing priorities and lack of U.S. SCWR funding.



Figure 1. Schematic of the four-rod test section.

Bundle geometry	2x2, square tube insert		
Maximum Pressure	25 MPa (3625 psia)		
Test section water inlet temperature (enthalpy window) both for coolant and moderator (individually chosen); two independent moderator flows	280 to 488°C		
Maximum test section water outlet temperature	550°C		
Maximum test section power flux	1,500 kW/m²		
Heater rod diameter	10 mm or equivalent		
Heater rod heated length	1 m		
Inlet length (unheated)	0.5 m		
Outlet length (unheated)	0.5 m		
Number of spacer grids	≥3		
Range of Coolant Mass flux	200 – 1,000 kg/m²s		
Number of heater rods	4 (one replaceable dummy rod (square))		
Rod spacing to diameter ratio (pitch)	1.15		
Direct or indirect heated rods	Indirect		

Table 1. Requirements of the four-rod test section.

Thermochemical Hydroger	Production	Process	Analysis
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PI (U.S.): M. Lewis, Argonne National Laboratory (ANL)

PIs (Canada): S. Suppiah and J. Li, Atomic Energy of Canada Limited (AECL)

Collaborators: University of Nevada Las Vegas Research Foundation, University of Ontario Institute of Technology (UOIT)

Research Objectives

The objective of this project is to determine the cost of hydrogen production using an optimized, low-temperature, copper-chloride (Cu-Cl) cycle. Proof-of-principle experiments showed that all of the reactions in the Cu-Cl had reasonable yields and kinetics. Also, ASPEN simulations, which relied on estimated thermodynamic

data, showed a promising efficiency of 41-44 percent, based on the lower heating value (LHV). Current experimental and modeling activities are designed to more accurately evaluate the Cu-Cl cycle and determine chemical viability, engineering feasibility, and, ultimately, a cost estimate.

Research Progress

During this research period, researchers focused on further understanding the hydrolysis reaction,

 $2CuCl_2 + H_2O \rightarrow Cu_2OCl_2 + 2HCl (g).$

This reaction is challenging because of a competing reaction and requirement for excess water to obtain high yields. Researchers began experimental work to determine optimum experimental variables. They designed, built, and tested a new hydrolysis reactor design, which is shown in Figure 1. Figure 2 shows the X-ray diffraction (XRD) patterns of the solid products obtained from one hydrolysis experiment conducted at 375°C, with lines for Cu_2OCl_2 , CuCl, and hydrated and anhydrous CuCl₂. The Cu_2OCl_2 is the desired product of the hydrolysis reaction. The CuCl may be a product from either a competing reaction (CuCl₂ \rightarrow CuCl + $1/_2$ Cl₂) or from decomposition of the Cu₂OCl₂. The CuCl₂ is unreacted material that researchers believe is produced in the hydrated form

Project Number: 2004-007-C

Project Start Date: June 2004

Project End Date: May 2007



Figure 1. Static vertical bed reactor for studying the reaction of $CuCl_2$ with steam.



Figure 2. XRD pattern for a product formed during the hydrolysis reaction at 375°C.



Figure 3. Heat capacity measurements for Cu_2OCl_2 versus temperature using adiabatic calorimetry (squares and circles), differential scanning calorimetry (dashes), the sum of heat capacity values for CuO and CuCl₂, and primary literature values. The inset figure suggests a solid-to-solid transition in the heat capacity versus temperature curve around T = 70 K.

during handling, as CuCl₂ is very hygroscopic. The high molar ratio of steam to copper and the presence of this unreacted material indicates that the current reactor design does not promote good contact between the steam and CuCl, powders. In order to obtain quantitative data for the various copper species, researchers applied Rietveld analysis to compare the integrated peak intensities of the most intense peak in test samples to calibration standards. However, the results were not completely satisfactory so the team is considering other analytical techniaues.

Researchers conducted experiments at different temperatures, flow rates, and steamcopper molar ratios. Preliminary XRD data suggest that 375°C is an optimum temperature and that a steam-copper molar ratio of 17 gives reasonable conversion to the desired products. However, that ratio is too high to be practical and further work, such as a new reactor design, is planned to reduce this parameter.

In addition to the experimental work, researchers continued modeling the cycle as a whole. Sensitivity studies of the hydrolysis reaction showed that a very small difference (11 kJ per mol) in the free energy of formation of Cu₂OCl₂ changed the predicted amount of oxychloride formed by a factor of 7. Consequently, the team synthesized a sample of very high purity Cu,OCl, and measured its enthalpy of formation at 298.15 K and its heat capacity as a function of temperature. Figure 3 shows the data for the latter. Researchers calculated the free energy of formation from these measurements and will update the model with these new results.

Researchers also started work on the electrochemical reaction, investigating two options to produce hydrogen and cupric chloride:

Option 1: 2CuCl + 2HCl(a) \rightarrow H₂ (g) + 2CuCl₂ or

Option 2: $4CuCl \rightarrow 2Cu + 2CuCl_2$ $2Cu + 2HCl \rightarrow H_2 (g) + 2CuCl$

The researchers successfully conducted two proofof-principle electrochemical experiments: 1) hydrogen production at AECL, and 2) deposition of copper powders and subsequent release from the cathode at ANL. Figure 4 shows the electrochemical cell used for copper deposition and release. Copper is deposited on the cathode; after the current is reversed, the copper falls to the bottom of the cathode compartment.

Planned Activities

Researchers will further define the experimental parameters that have the greatest impact on the hydrolysis reaction, optimize the parameters via experimental work and modeling, and study the performance of both types of electrochemical cells. The optimization at ANL will involve impedance measurements to determine whether the voltage requirements are due to mass transfer or kinetics for a range of concentrations of the various solutions and for different electrode materials.

Researchers hope to complete preliminary cost estimates for producing hydrogen with the Cu-Cl cycle using both options for the electrochemical reaction. As the simulations become more robust, interactions with AECL and UOIT will increase to promote further studies of coupling between the chemical facility and the nuclear plant.



Figure 4. (a) A titanium screen on which copper powders are deposited; (b) the titanium screen after reversal of the current; and (c) copper powders on the bottom of the cell.

7.0 U.S./European Union Collaboration

The U.S. Department of Energy (DOE) and the European Atomic Energy Community (EURATOM), an international organization composed of the members of the European Union, signed a bilateral agreement on March 6, 2003. Secretary of Energy Spencer Abraham signed the agreement for DOE and Commissioner for Research, Phillipe Busquin, signed on behalf of EURATOM. In 2004, the U.S. and EURATOM selected the first ten projects for collaboration.

7.1 Work Scope Areas

R&D topical areas for the U.S./EU collaboration include:

- Reactor fuels and materials research
- Advanced reactor design and engineering development
- Research and development related to the transmutation of high-level nuclear waste
- Transmutations-related system analyses

7.2 Project Summaries

Ten projects were approved in FY 2004 and one in FY 2006. A listing of the I-NERI U.S./EU projects that are currently underway follows, along with summaries of the accomplishments achieved in FY 2006.

Directory of Project Summaries

2004-001-E	Development of Inert Matrix Fuels for Plutonium and Minor Actinide Management in LWRs	49
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Development of Inert Matrix Fuels for Plutonium and Minor Actinide Management in LWRs

PI (U.S.): J. Carmack, Idaho National Laboratory (INL)	Project Number: 2004-001-E	
	Project Start Date: October 2004	
PI (Europe): J. Somers, Joint Research Center Institute for Transuranium Elements (JRC-ITU)	Project End Date: September 2008	
Collaborators: Los Alamos National Laboratory, Oak Ridge National Laboratory (ORNL)		

Research Objectives

The goal of this project is to develop feasibility data on using inert matrix fuels (IMF) and minor actinide targets in the existing fleet of light water reactors (LWRs). The only viable option for active management of plutonium and minor actinides over the next 10–20 years is in commercial reactors. There is worldwide interest in using the existing LWR fleet for fuel cycle scenarios to burn down plutonium (Pu) and stabilize minor actinide (MA) inventories.

Because of this interest in existing water-cooled reactors for plutonium and MA management, all research and development concepts being analyzed in this project will be suitable for loading into present-day and nearterm (Generation II and III) power reactor designs. This project will generate comparative data on the fabrication, properties, and irradiation behavior of several IMF fuel candidates to provide valuable information on their feasibility for managing plutonium and MA in LWRs.

Inert matrix fuels offer several advantages over conventional uranium matrix mixed oxide (MOX) fuel for plutonium management. IMF allows for more efficient destruction of Pu since the significant reduction of ²³⁸U precludes the breeding of additional plutonium—greater efficiency in reducing Pu provides improved flexibility in managing inventories. With IMF, there is a potential to lower fuel cycle costs due to the reduced number of rods required to effect a given plutonium burn rate. In addition, IMF can be used to dispose of minor actinides, particularly americium and neptunium. The choice of matrix material may be manipulated to facilitate either fuel recycling or direct disposal and plutonium recovery can be made relatively straightforward or extremely difficult in order to control proliferation. High thermal conductivity inert matrix fuels may also have operational and safety benefits. Cermet fuel, for example, operates at very low temperatures, which can improve operating and safety margins or increase rated reactor power.

Research Progress

This project was inactive in 2006. Progress to date includes:

Fuel Selection. Researchers established goals for IMF development, assessed IMF candidates against this baseline, and selected several materials for further study. They analyzed neutronic behavior (Pu burn rate and reactivity swing), fuel thermal and irradiation performance, and in-core corrosion resistance based on existing knowledge.

Fabrication Development. Researchers developed and qualified fabrication methods for inert-matrix fuels (MgO-ZrO₂ and Zr-metal matrix) and performed test fabrication and characterization. Figure 1 is a photograph of a plutonium-bearing MgO-ZrO₂ matrix pellet and Figure 2 shows a Zr-metal matrix fuel sample.

The team developed two dust-free fabrication routes permitting the routine achievement of high-density zirconium-yttrium oxide $(Zr,Y)O_2$ pellets. In the first, cerium nitrate (a surrogate for the actinide) was infiltrated into $(Zr,Y)O_2$ sol gel beads. Following calcination, these beads were milled, then pressed and sintered. Because of the milling step, this process is not applicable to americium (Am). In the second (Am-compatible) process, the $(Zr,Y)O_2$



Figure 1. Photograph of dual-phase MgO-ZrO₂-PuO₂ pellet.

Figure 2. UO_2 particles extruded in a Zr-metal matrix.

beads were softened by adding carbon in the sol gel feed solution, which was later removed by pyrolysis in the calcinations step. The resulting "softer" beads permitted high actinide content infiltration and easier pressing. Both methods yielded pellets of similar quality and greater than 90 percent of the theoretical density.

Research on CERMET fuels has been ongoing with molybdenum (Mo) as the metallic matrix. Although mainly associated with the FUTURIX irradiation tests in the Phénix reactor, cermet samples with (Pu0.73Th0.23)O₂/Mo have also been prepared. In this case, the ceramic (Pu,Th)O₂ phase was prepared from PuO₂ beads produced with sol-gel, infiltrated with thorium nitrate and subsequently calcined to convert the nitrate to the oxide. The ceramic phase was diluted to 40 volume-percent by adding Mo and blending prior to pressing. Densities of 90 percent of the theoretical value were obtained. A photo of the surface of the pellet is shown in Figure 3.



Figure 3. Cermet (Pu0.73Th0.23)O₂/Mo (40:60 vol%) pellet end face.

Characterization. Researchers completed their experimental characterization of the candidate IMF and matrix materials. They analyzed microstructure, thermal and mechanical properties, corrosion testing, and ionbeam irradiation in order to determine suitability for in-reactor irradiation testing and to provide a basis for modeling fuel behavior. Further refinement of fabrication processes is necessary.

Fuel Modeling. The team constructed a finite element model using

the ABAQUS code to model the phase structure of both heterogeneous and homogenous inert matrices. This code allows researchers to estimate key fuel properties (such as thermal conductivity and density), which is needed to support reactor irradiation experiment design and qualification.

Reactor Physics, Safety, and Licensing Analysis. Researchers performed four-bundle neutronic, thermalhydraulic, and transient analyses of proposed inert matrix materials and compared the results to similar analyses for reference UOX fuel. They will use the results for screening purposes. The goal is to identify the feasibility of using specific IMF compositions in existing and future LWRs. Compositions identified as feasible will require further detailed analysis and rigorous experimental testing and qualification.

Irradiation Testing and PIE. The LWR-2 screening irradiation test at the Advanced Test Reactor (ATR) at Idaho National Laboratory is currently in the design and planning stages. This test will include a variety of candidate IMF compositions, as shown in Table 1. Researchers will report post-irradiation examination (PIE) results from a preliminary irradiation test (LWR-1a) on a sample of advanced MOX incorporating approximately 4,000 ppm neptunium oxide. Figure 4 is a neutron radiograph of the LWR-1a rodlet following exposure in the ATR.

Figure 4. Neutron radiograph of 94% dUO₂ – 6% RGPuO₂ - <1% NpO₂

sample PROF-3 irradiated to ~9 GWd/tU burnup.

Following an intensive discussion period, researchers finally abandoned the MILE irradiation program in Beznau. In parallel, they submitted a STREP proposal to the European Commission. If accepted, this will provide JRC-ITU an opportunity to irradiate fuels in two European reactors (the BR2 in Belgium's Mol nuclear research center and the Borsele LWR in the Netherlands).

Planned Activities

Future efforts will focus on fabricating fuel test specimens to be included in the LWR-2 irradiation test planned for the ATR. This test will include a variety of candidate IMF compositions, as shown in Table 1. The team will fabricate fuel specimens for both out-of-pile characterization and in-reactor irradiation tests. Figure 5 shows a schematic diagram of an LWR-2 test assembly rodlet that will contain one of the individual specimens. Researchers plan to install the test assembly in the spring of 2007.



Figure 5. 3-D representation of a LWR-2 fuel irradiation test pin.

Designation	Description	Composition	Fabricator
LWR-2-A	RG-MOX at high burnup (LWR-1a)	(U,Pu)O ₂	INL
LWR-2-B	Advanced MOX (Np, Am additions)	(U,Pu,Np,Am)O ₂	INL
LWR-2-C	IMF replacement to reference MOX	(Pu)O ₂ - MgO-ZrO ₂	INL
LWR-2-D	IMF (Np addition)	(Pu,Np)O ₂ , MgO-ZrO ₂	INL
LWR-2-E IMF replacement to Advanced MOX		(Pu,Np,Am)O ₂ , MgO-ZrO ₂	INL
LWR-2-F	Sphere Pac target (low fertile matrix)	$UO_2 - AmO_2$	ORNL
LWR-2-G	Sphere Pac target (inert matrix)	PuO ₂ or ZrO ₂ - AmO ₂	ORNL
LWR-2-H	PuO ₂ Cercer - Cermet	PuO ₂ , YSZ or Mo (if feasible)	EURATOM
LWR-2-I	Pu - FeCr Ferritic (ITU fabrication)	Pu, Fe/Cr	EURATOM
LWR-2-J	SiC Inert Matrix	UO ₂ -SiC IMF	AECL
LWR-2-K	Advanced IMF (Univ. of Florida)	PuO ₂ - MgO-ZrO ₂ -Nd or Yb	UF/INL
LWR-2-L	Zr metal inert matrix	(PuO ₂ , Zr)	INL
LWR-2-M	WG-MOX at high burnup (LWR-1a)	(U, Pu)O ₂	INL

Table 1. Summary of IMF compositions for LWR-2.

Development of Fuels for the Gas-Cooled Fast Reactor

PI (U.S.): M. Meyer, Idaho National Laboratory (INL)

PI (Europe): J. Somers, Joint Research Center Institute for Transuranium Elements (JRC-ITU) Project Number: 2004-002-E

Project Start Date: October 2004

Project End Date: September 2006

Collaborators: Oak Ridge National Laboratory

Research Objectives

The objective of this project is to develop silicon carbide matrix, uranium-carbide dispersion fuel in two forms suitable for gas-cooled fast reactor (GFR) service: 1) a hexagonal block with coolant holes throughout and 2) a pin-type dispersion fuel with integral silicon-carbide cladding. Because fuel operating parameters and physical requirements for the GFR are outside of the current experimental nuclear fuel database, many basic viability issues need to be addressed experimentally to demonstrate the feasibility of the proposed fuels.

The GFR-F1 test in the Advanced Test Reactor (ATR) at INL and the FUTURIX-MI test in the French Phénix reactor address basic issues regarding the irradiation behavior of the "exotic" refractory materials required for GFR fuel service in a neutron-only environment. Ultimately, proofof-concept can only be demonstrated through irradiation testing of fissile-bearing specimens. The GFR-F2 fuel irradiation test in the ATR is a fuel behavior test that will provide the first true indication of fuel feasibility.

Research Progress

Current GFR fuels are based on a dispersion of (U,Pu)C coated particles in a silicon-carbide (SiC) matrix. The U.S. has two reference fuel forms: 1) large hexagonal blocks with coolant holes drilled throughout and 2) a refractory clad pin-type fuel. Both the block-type and the rod-type fuel will be fabricated through reaction bonding; however, the starting material preforms are fabricated in a slightly different fashion.

The method of reaction bonding used for the blocktype fuel fabrication starts with a polymer-derived carbon preform. The polymer is produced with a specific amount of a pore-forming agent that creates the appropriate porous microstructure. The polymer is then cured and pyrolyzed to produce a porous carbon preform that can be cast and/or machined into final shape. The preform is then infiltrated with molten silicon which reacts with the carbon to form SiC. The microstructure must be tailored to produce a fully infiltrated sample with minimal residual silicon (less than 5 volume-percent). The microstructure is dependent upon the amount of pore former used in the original polymer mixture.

The disadvantage of this process for fuel fabrication is the sample shrinks on the order of 50 percent during pyrolyzation. If fuel spheres are incorporated at this point (with no filler), the preform will crack due to shrinkage of the matrix around the stable fuel particles. To control this shrinkage, a filler powder must be added. Throughout FY 2005, sample reproducibility plagued this process. However, a graphite element vacuum/inert atmosphere furnace installed in FY 2006 greatly improved the reproducibility of results. Researchers concluded that much of the previous variability can be attributed to poor atmospheric control.

Previous results show that an 80:20 ratio of filler powder to polymer is required to reduce the shrinkage below 5 percent. The filler materials dramatically affect the green, or pre-infiltrated, microstructure. Filler powder composition also affects the infiltrated microstructure. Researchers conducted a series of parametric studies using 70, 80, and 90 percent β -SiC filler material, with the remainder carbon in the form of carbon black and graphite. After pyrolysis, the samples were weighed to determine mass loss and dimensionally analyzed to characterize volumetric shrinkage. The carbon formed during pyrolysis

Sample	Pressing Pressure (MPa)	Volume (cm³)	SiC (vol %)	Carbon (vol %)	Porosity (vol %)	Porosity/ Carbon	Infiltration Results
90-1	17.2	0.45	45.0	15.7	39.3	2.50	Center portion not infiltrated
90-2	34.4	0.40	49.5	17.2	33.2	1.92	Surface only
90-3	51.7	0.41	51.2	17.8	31.0	1.74	Surface only
80-1	17.2	0.38	38.3	22.6	39.1	1.74	Delamination, friable
80-2	34.4	0.28	42.4	25.6	32.0	1.25	Large cracks
80-3	51.7	0.33	43.9	26.1	30.0	1.15	Large cracks
70-1	17.2	0.33	31.7	28.4	39.9	1.40	Delamination, friable
70-2	34.4	0.26	35.5	31.7	32.8	1.04	Delamination, friable
70-3	51.7	0.32	37.1	32.6	30.4	0.93	Cracks, slightly friable
90-4	8.6	0.42	36.3	12.0	51.7	4.31	Friable
90-5	17.2	0.35	44.6	15.1	40.4	2.68	Dense sample

Table 1. Results from pyrolyzation and infiltration of pellets with varying filler SiC loadings and pressing pressures.

was calculated based on the initial polymer loading and mass loss. Based on known densities of each constituent, researchers calculated the volume fraction of each component in the microstructure, as listed in Table 1.

The samples containing 90 percent filler remained intact but did not fully infiltrate. In many cases, only the surface reacted to form a dense SiC outer layer. Samples with 80 percent SiC filler fully infiltrated, but many large cracks were formed that filled with excess silicon. The 70 percent SiC filler samples also fully infiltrated; however, the resulting samples were friable and had no structural integrity. They often delaminated upon infiltration. Figure 1 shows typical pellets from each of the three groups after infiltration.

The researchers conducted further experimentation to refine the fabrication process, noting significant differences in final microstructure with changes in the pressure applied during pelletization. Table 1 lists the specific parameters used and resulting pellet volume fractions. Infiltration results varied from a fully infiltrated dense pellet (90-5) to a friable pellet with no structural integrity. Figure 2 shows the microstructure of the fully dense, infiltrated pellet, with less than 5 percent porosity and very little residual silicon.

During reaction bonding, carbon reacts with silicon to form SiC, resulting in a volume expansion of 2.36 times relative to the initial carbon substrate. Because of this, the porosity-to-carbon ratio must exceed 2.36 to ensure adequate space for volume expansion during the formation of SiC. Samples that remained intact while exhibiting appreciable infiltration had porosity-to-carbon ratios of 2.5 and 2.6. Lower ratios cracked during infiltration. Excessive porosity leads to a friable sample with little or no structural integrity. If adequate silicon is present, it will fill the pores, increasing the amount of free silicon to unacceptably high levels.



Figure 1. Typical pellets after infiltration: (A) 90% SiC filler, (B) 80% SiC filler, and (C) 70% SiC filler.



Figure 2. Resulting SiC microstructure of a pellet with 90% SiC filler material and a porosity-to-carbon ratio of 2.68 (90-5).

Pore size and morphology were also found to be important. As transport of silicon relies on a network of interconnected porosity, infiltration does not take place if the pores are too small or disconnected. In the case of small pore size, silicon will start infiltrating at the preform surface, but the channels quickly close due to the formation of SiC. This leads to a SiC-coated preform with no reaction-formed SiC in the interior. The high particle volume loading required for the GFR will also undoubtedly reduce the transport paths for silicon. A process must, therefore, be developed to ensure evenly spaced particles completely surrounded by the SiC precursor materials, with the proper pore size to allow transport of molten silicon throughout the sample. Further process development is required to produce fully infiltrated dense samples with a high fuel loading using this technique.

Research targeted at developing pin-type fuel with integral SiC cladding was not conducted in FY 2006 due to funding limitations. The only carbon source used in this fabrication process is carbon powder added to SiC powder. The processing route chosen was based on wet mixing of SiC and graphite powder. Researchers found that a-SiC (70 percent -100+200 mesh and 30 percent -325 mesh) and graphite (-325 mesh) in a 75:25 ratio, mixed with 25 weight-percent glycol and pressed at 9,000-18,000 psi and 1,550°C, yields well-infiltrated samples of matrix material, as shown in Figure 3. Adding spheres to the matrix, however, resulted in severe cracking of the infiltrated samples, indicating that further optimization is required in order to incorporate fuel particles into the matrix.



Figure 3. Microstructure of reaction bonded SiC using the proper ratio of particles sizes and compositions.

In conjunction with the fuel fabrication activities, researchers conducted several material irradiation experiments, including heavy ion irradiation studies of ceramics (ZrN, TiC, TiN, ZrC, and SiC). All the irradiations were conducted at 800°C and dose of 10 and 70 dpa, 1 MeV krypton ions from the IVEM-TANDEM facility at Argonne National Laboratory. Irradiation performance of ZrC and ZrN under these conditions was poor, with the materials showing severe lattice expansion. The performance of TiC and TiN was much improved, however, with lattice expansion reduced by a factor of four. The SiC samples exhibited virtually no lattice expansion. The GFR-F1 low-dose material experiment irradiated commercially procured SiC, TiC, ZrC, AIN, TiN, and ZrN in the Advanced Test Reactor at INL. The experiment was not instrumented and researchers maintained a calculated specimen temperature of approximately 1,000°C by varying heat transfer from the experiment capsules with differing fill gas compositions. The first experimental capsules are currently awaiting post irradiation examination.

Reaction bonding appears to be a viable path for fabricating a dense SiC matrix-dispersion fuel suitable for the GFR, although continued process development is needed to optimize fuel particle distribution, to produce uniform infiltration of a specimens with high particle loading, and to reduce free silicon content.

Planned Activities

No further work on this project is planned. Funding is not expected in FY 2007 due to changing U.S. nuclear R&D priorities.

Lead (Pb) Fast Reactor Engineering and Analysis

PI (U.S.): James J. Sienicki, Argonne National Laboratory (ANL)

PI (Europe): Hartmut U. Wider, Joint Research Center of the European Commission, Institute for Energy (JRC-IE) Project Number: 2004-003-E

Project Start Date: November 2004

Project End Date: November 2007

Collaborators: None

Research Objectives

The objectives of this project are to further develop the analysis base and experimental database for the leadcooled fast reactor (LFR). Europe is conducting extensive research and development on the European lead-cooled system (ELSY) to demonstrate the technical feasibility of an LFR. This reactor is economical and especially effective at burning minor actinides (MAs), and is consistent with the goals of the Global Nuclear Energy Partnership (GNEP) and the Generation IV Nuclear Energy Systems Initiative. ANL is carrying out R&D on the Small Secure Transportable Autonomous Reactor (SSTAR), a proliferation-resistant LFR for deployment in developing nations. This design incorporates features for fissile self-sufficiency (providing efficient utilization of uranium resources), autonomous load following (making it suitable for small or immature grid applications), and a high degree of passive safety, all of which are also consistent with GNEP and Generation IV goals, especially for reactors for international deployment.

Research Progress

Experiments performed under a contract with JRC/IE evaluated whether fission products with low melting and boiling temperatures, such as iodine, cesium, and cesium-iodide, would be absorbed by lead-bismuth eutectic (LBE) following a postulated fuel failure. The experiments showed that LBE absorbs these radionuclides at 400 and 600°C. Cesium forms inter-metallic compounds in LBE while iodine reacts with the lead to form PbI₂.

Using computational fluid dynamics (CFD) techniques, researchers calculated the steam released immediately after a postulated steam generator tube rupture (SGTR) in reactor systems having the configuration envisioned for the European Facility for Industrial Transmutation (EFIT) and ELSY. In the exploratory calculations, the released steam was calculated to fill the steam generator volume, rising to the free surface and blocking the heavy liquid metal coolant flow in that steam generator. This would divert the coolant flow through the second intact steam generator at a higher velocity. The codes were also compared with Japanese data on nitrogen and water injection into LBE.

Researchers developed a concept for a new experimental facility to investigate SGTR phenomena in a tube bundle geometry and generate data for code validation in three phases: 1) nitrogen gas injection into water, 2) superheated steam injection into Pb (approximately 500 Kg), and 3) supercritical steam injection into Pb. JRC/IE shared their experimental results with ANL, including high-temperature uni-axial time-to-failure creep rupture experiments at 70 MPa stress. Researchers found that at temperatures close to 900°C, the time-to-failure is less than 30 minutes.

Preliminary work has started on experiments investigating the viability of disposing LBE and lead in seawater following plant decommissioning. This is relevant to potential SSTAR cassette core shipping accidents at sea. Lead, bismuth, and LBE will be exposed to tap water, seawater, and artificially clean seawater at 50°C for four months. For the same nine material pairs, separate tests will include electrolysis-induced corrosion.

Researchers evaluated the new approach of locating the heat exchangers in the riser above the core. This would reduce the likelihood that steam bubbles could be transported to the core following a postulated SGTR event and would naturally result in a lower reactor vessel wall temperature during normal operation. On the other hand, the design reduces the effectiveness of coolant natural circulation, resulting in higher core temperatures in an unprotected loss-of-flow accident.

Using CFD code calculations, researchers investigated the performance of an air natural circulation Reactor Vessel Auxiliary Cooling System (RVACS) for decay heat removal. They investigated two different 600 MWe LFR concepts and one 900 MWe LFR design. For a protected total loss-ofpower accident, the RVACS design was unable to prevent the core outlet temperature from exceeding the structural steel creep limit. Therefore, they concluded that this design may require a more efficient emergency decay heat removal approach.

Using calculations on the performance of oxidefueled burner cores in which minor actinides are homogeneously distributed throughout the entire core, rather than in blanket regions, researchers determined that the homogeneous cores have a large positive coolant temperature reactivity coefficient due to the large mass of lead coolant involved. The coolant temperature reactivity coefficient can be reduced by inserting thermalizing pins containing BeO, CaH₂, or UZrH_{1.6}. By calculating an unprotected loss-of-heat sink accident for a 600 MWe LFR, in which the pumps function normally but the steam generators stop removing heat after 20 seconds, researchers found that the power passively decreases to about 15 percent nominal within 1,000 seconds, after which the system coolant and structural temperatures rise to about 830°C. However, the structures retained their integrity.

ANL researchers have further developed and improved the pre-conceptual design and configuration for the SSTAR reactor system (Figure 1) by reducing the vessel height; incorporating a thermal baffle to protect the reactor vessel near the Pb-free surface; and configuring fuel pin grid spacers, control rods, control rod guide tubes, drivelines, and drives. An automatic control strategy developed for the SSTAR supercritical carbon dioxide (S-CO₂) Brayton cycle energy converter, in principle, should enable autonomous load following by the reactor over the full range of power, from 0 to 100 percent nominal load



Figure 1. Illustration of 20 MWe (45 MWt) Small Secure Transportable Autonomous Reactor (SSTAR).

demand. This scheme is especially suited for deployment on small or immature grids. The strategy consists of turbine bypass control over the complete range of load demand combined with inventory control above 35 percent nominal load. The SSTAR S-CO₂ Brayton cycle energy converter has been improved by incorporating centrifugal compressors that provide wider operating range, especially near the critical point, and can better handle potential twophase flow in certain accident sequences relative to the former axial flow compressors.

Calculations were initiated for an international neutronics benchmarking problem on an LBE-cooled, mixed nitride-fueled core using the same neutronics codes applied to SSTAR. This initiative is part of an International Atomic Energy Agency (IAEA) research program on small reactors without on-site refueling. Researchers formulated a safety design approach for SSTAR consistent with NRC and IAEA requirements. They also developed a new transient fuel pin design computer code that integrates time-dependent fission gas release from nitride fuel, local cladding thermal creep, and the remaining unoxidized cladding. The method incorporates parabolic kinetics to determine penetration of the oxide layer into the metal and linear kinetics for dissolution of the oxide trailing surface by the LBE or Pb coolant.

Planned Activities

The following activities will be conducted during the next fiscal year.

- Additional experiments will be carried out investigating iodine, cesium, and cesium-iodide behavior in LBE under severe accident conditions at 800-900°C. Of particular interest is the behavior of iodine under these conditions, since the boiling point of PbI₂ is 831°C.
- SGTR experiments will be performed over the next two years.
- CFD analyses will analyze LFR behavior following postulated SGTR.

The focus of LFR development in the U.S. is now shifting towards a near-term deployable LFR test demonstrator implementing innovative engineering features that should allow lead technology to demonstrate its economic potential and industrial attractiveness. In addition, an LFR for international deployment that operates at lower system temperatures than SSTAR would enable the use of existing cladding and structural materials that have been shown to be corrosion resistant when used with active oxygen control at temperatures below 550°C.

Proliferation Resistance and Physical Protection Assessment Methodology

PI (U.S.): Ike Therios, Argonne National Laboratory (ANL)

PI (Europe): Giacomo Cojazzi, Joint Research Center of the European Commission (JRC) Project Number: 2004-004-E

Project Start Date: January 2004

Project End Date: September 2008

Collaborators: None

Research Objectives

The purpose of this project is to develop a systematic method for evaluating and comparing proliferation resistance and physical protection (PR&PP) of Generation IV nuclear energy systems, including their fuel cycle facilities and operations, as recommended by the *Generation IV Roadmap*. The methodology will be applicable from the early development stages throughout the full lifecycle. One main objective is to establish an iterative process in which the PR&PP performance of the system is included in the evolution of the design.

The Generation IV International Forum (GIF) created a PR&PP expert group in December 2002 and tasked them with leading the development project. This expert group currently includes participants from U.S. national laboratories (ANL, BNL, INL, LANL, LLNL, ORNL, PNNL, and SNL), academia, international experts from six GIF member countries (Canada, European Union, France, Japan, Republic of Korea, and the United Kingdom), the International Atomic Energy Agency, and observers from the U.S. State Department and the U.S. Nuclear Regulatory Commission.

Research Progress

To focus development of the methodology, the group adopted the following definitions of proliferation resistance and physical protection:

Proliferation resistance is that characteristic of a nuclear energy system that impedes the diversion or undeclared production of nuclear material, or misuse of technology, by the host state in order to acquire nuclear weapons or other nuclear explosive devices. *Physical protection (robustness)* is that characteristic of a nuclear energy system that impedes the theft of materials suitable for nuclear explosives or radiation dispersal devices, and the sabotage of facilities and transportation, by sub-national entities and other non-host state adversaries.

Figure 1 illustrates the methodology. For a given system, researchers first define a set of **challenges** then assess the **response** of the system to these challenges, expressed in terms of **outcomes**.

CHALLENGES	SYSTEM RESPONSE	\rightarrow	OUTCOMES
Threats	PR & PP		Assessment

Figure 1. Framework for PR&PP evaluation.

The methodology is organized in a progressive approach that allows evaluations to become more detailed and more representative as the system design matures. The progressive approach is intended to maximize early, useful feedback to designers throughout each phase, from basic process selection, to the detailed layout of equipment and structures, to facility demonstration testing.

The system response is expressed in terms of proliferation resistance and physical protection *measures*. The term *measures* is used by the PR&PP group to identify the few measurable, high-level parameters that can be used to express proliferation resistance (PR) or physical protection (PP). Usage of this term should not be confused with another common meaning that indicates a set of external actions or procedures applied to material and facility control and protection. Researchers will first evaluate measures for individual *segments* and then aggregate them for *complete pathways*. Aggregating results permits pathway comparisons and system assessment.

For proliferation resistance, the measures are:

- Proliferation technical difficulty
- Proliferation cost
- Proliferation time
- Fissile material type
- Detection probability
- Detection resource efficiency

For *physical protection*, the measures are:

- Probability of adversary success
- Consequences
- Physical protection resources

The researchers are developing specific quantitative indices and metrics to evaluate the proliferation resistance measures. In addition to the above PR pathway measures, they have defined the more holistic concept of *safeguardability* as "the ease of putting an effective and efficient safeguards system into operation." This concept can be used at early design stages as a type of checklist to identify the features to be considered for safeguarding all or part of a nuclear energy system. This can result in improved integration of PR with safeguard approaches and can provide hints to the designers at early design stages.

During the past year, researchers further developed the methodology by applying it to the nuclear fuel pyrochemical reprocessing facility, part of the hypothetical Example Sodium Fast Reactor (ESFR) system. The demonstration study examined proliferation pathways from a select set of material balance areas within the pyroprocessing fuel cycle facility. They used three different implementations of the pathway analysis methodology to assess proliferation resistance measures: 1) qualitative, 2) event tree/fault tree, and 3) Markov. The scenario assumes that the host state diverts transuranic materials (TRU), found in different chemical and physical forms at different stages of the fuel cycle, in order to acquire a significant quantity of Pu in a year's time. Different diversion strategies and diversion rates have been investigated. The assessment considers both intrinsic and extrinsic barriers to proliferation. Processing of diverted material is assumed to take place in a clandestine facility. Metrics and scales are identified for the six proliferation resistance measures and qualitative and quantitative results are presented in both tabular and graphical form. The researchers compared results of the three assessment approaches and drew preliminary observations and issued lessons learned from the demonstration study.

The PR&PP methodology provides a framework to answer a wide variety of security-related questions for nuclear energy systems and to optimize these systems to enhance their ability to withstand the threats of proliferation, theft, and sabotage. The PR&PP methodology provides the tools to assess nuclear energy systems with respect to the security-related goals for Generation IV technologies to be "a very unattractive and the least desirable route for diversion or theft of weapons-usable materials, and provide increased physical protection against acts of terrorism."

PR&PP analysis is intended to be performed simultaneously with initial hazards identification and safety analysis, at the earliest stages of system design—when initial flow diagrams and physical arrangement drawings are being developed. The structure of the PR&PP methodology bears strong similarity to safety analysis techniques. The PR&PP methodology systematically identifies the security challenges a system may face, evaluates the system response to these challenges, and compares outcomes. The outcomes are expressed in terms of measures, which reflect the primary information that a proliferant state or an adversary would consider in selecting strategies and pathways to achieve their objectives.

By understanding the features of a facility or system that could provide more attractive pathways, the designer can introduce barriers that systematically make these pathways less attractive. When this is not possible, for example in reducing the attractiveness of the proliferation pathways provided by uranium enrichment, the analyst can highlight where special institutional measures may be required to provide appropriate levels of security, such as assured fuel supply and return regimes. The PR&PP methodology provides a high degree of flexibility to the analyst. Researchers anticipate that approaches to performing PR&PP evaluations will evolve over time, as the literature and examples of assessments expand. They also expect that the number of tools for identifying targets, assessing system response and uncertainty, comparing pathway outcomes, and presenting results will increase, as will the range of questions that can be answered and insights gained from PR&PP studies.

Planned Activities

This I-NERI project will continue with the development of the PR&PP evaluation methodology.

Researchers will conduct analyses on diverse nuclear systems with diverse analytic objectives to further understand appropriate application of analytic methods to PR&PP analysis and to establish a baseline standard/norm for such analysis.

Characterization of Nuclear Waste Forms and Their Corrosion Products

PI (U.S.): J. Cunnane, Argonne National Laboratory

PI (Europe): V. Rondinella, Joint Research Center Institute for Transuranium Elements (JRC-ITU)

Collaborators: Pacific Northwest National Laboratory

Project Number: 2004-005-E

Project Start Date: TBD

Project End Date: TBD

Project Abstract

This project has not yet received any funding. As a result, no work has been accomplished so the annual report remains in abstract form.

The objective of this project is to understand the formation of altered phases and their effect on the behavior of waste forms (spent fuel and/or conditioning matrix) during storage and through contact with groundwater. Researchers will investigate the development of waste alteration caused by large accumulation of alpha decay damage (structural, property changes) and the formation of secondary phases on the waste form surface and its effect on the waste corrosion behavior (e.g., corrosion rate and formation of layers blocking further corrosion on the waste form surface).

The goals of this research are to:

- Monitor the effects of radiation damage accumulation by measuring relevant quantities/properties (e.g., lattice parameter, macroscopic swelling, hardness, and thermophysical properties) and characterizing the microstructure using techniques such as transmission electron microscopy (TEM)
- Investigate recovery mechanisms and study the accumulation/release behavior of helium (He) in different materials, comparing irradiated fuels and alpha-doped fuels/matrices

- Evaluate possible relationships between observed property changes and corrosion behavior through experiments of aqueous corrosion of "aged" materials (e.g., preferential etching sites and isotopic fractions of released actinides)
- Examine corrosion mechanisms and characterize solid corrosion products formed during the aqueous alteration of spent nuclear fuel using X-ray photoelectron spectroscopy, TEM/energyloss spectroscopy (TEMEELS), scanning electron microscopy/energy dispersive spectrometry (SEM-EDS), and X-ray diffraction (XRD)
- Determine the fate of various radionuclides following their release from altered spent fuel, especially in terms of secondary phase formation and co-precipitation phenomena
- 6) Characterize re-precipitated phases on leached surfaces in terms of composition and potential effects on corrosion

The research may also include an exchange of suitable samples (e.g., "alpha-doped" materials) for full characterization by both U.S. and E.U. collaborators.
Nitride Fuel Fabrication Research

PI (U.S.): S. Voit, Los Alamos National Laboratory (LANL) Project Number: 2004-006-E

PI (Europe): S. Fernandez, Joint Research Center Institute for Transuranium Elements (JRC-ITU)

Project Start Date: October 2004 Project End Date: September 2007

Collaborators: None

Research Objectives

The objective of this research is to compare fabrication and property data of nitride fuels, focusing on fastspectrum fuels that can be used in liquid-metal reactors or accelerator-driven systems (ADS). Researchers are investigating two fuel forms: 1) non-fertile fuel designated for ADS and 2) low-fertile fuel for liquid-metal fast reactors. They have chose a non-fertile composition of 36wt%ZrN(Pu_{0.50}Am_{0.50})N for initial development and a lowfertile composition of $(U_{0.50}Pu_{0.25}Am_{0.15}Np_{0.10})N$.

Research Progress

This section summarizes fabrication and characterization results for the Futurix-FTA nitride fuel. Table 1 shows process parameters used in the *low-fertile* fabrication and Table 2 summarizes chemistry results. The reduction in solutionization, synthesis, and sintering temperatures for

Element	Specification Value	Actual Result
Plutonium (Pu)	23.7 wt. %	24.43 wt. %
Uranium (U)	47.1 wt. %	47.23 wt. %
Americium (Am)	14.3 wt. %	12.6 wt. %
Neptunium (Np)	9.4 wt. %	7.7 wt. %
Carbon (C)	≤3,000 ppm	1,275 ppm
Oxygen (O)	≤3,000 ppm	1,130 ppm
Nitrogen (N)	5.5 wt. %	5.63 wt. %

Table 2. Summary of low-fertile chemistry results.

the low-fertile composition reduced the americium boil-off. The data show only a 1.7 weight-percent deviation from the specification value, and an average of 85.4 percent theoretical density.

Low-Fertile Processing Parameters					
Solutionization		Zinc Stearate	0.2 wt. %		
Time	12 hours	PEG	0.2 wt. %		
Temperature	1,400°C	SPEX Mill Time	45 minutes		
Atmosphere	Atmosphere Ar		215 MPa		
Carbothermic Reduction/Nitridization		Sintering			
Time	20 hours	Time	12 hours		
Temperature	1,550°C	Temperature	1,600°C		
Atmosphere	$N_2 - N_2 / 6\% H_2$	Atmosphere	Argon		
C:O Ratio	1.25				

Table 1. Futurix-FTA low-fertile processing parameters.

Researchers took optical micrographs from low-fertile sample pellets. The microstructure of an axial pellet section (Figure 1) lacks the endcapping that was prevalent in previous fabrications. X-ray diffraction (XRD) results show a single-phase solid solution, which agrees with the AFC-1G low-fertile material (Figure 2). The residual weighted profile parameter, a measure of the "goodness of fit" between the observed and calculated patterns, is 14.1 percent for the low-fertile sample model. These values are in the expected range for refinements of powder X-ray diffraction data recorded from multi-phase samples.

Scanning electron microscopy (SEM) microprobe data (Figure 3) show some heterogeneity on the microscale (10–20 percent variation in actinide concentration over 5–10 μ m), with no apparent macroscopic heterogeneity. The results agree with the single-phase solid solution shown by XRD.

Process parameters for the *non-fertile* fabrication run are given in Table 3, while Table 4 summarizes the chemistry results. There are two important results: 1) multiple grind and re-sinter of the non-fertile material resulted in a significant americium loss (data show 23.3 weight-percent deviation from the specification value) and 2) the residual carbon value is significantly higher due to the reprocessed material with added carbon-based organics. The average theoretical density was 82.4.



Figure 3. Low-fertile SEM microprobe element maps with line scan. Upper: plutonium (Pu), americium (Am), and neptunium (Np); Lower: uranium (U), nitrogen (N), and an elemental line scan.



Figure 2. Low-fertile XRD pattern.

Non-Fertile Processing Parameters				
Solutionization		Zinc Stearate	0.2 wt. %	
Time	12 hours	PEG	0.2 wt. %	
Temperature	1,400°C	SPEX Mill Time	45 minutes	
Atmosphere	Ar	Pressing Pressure	58 MPa	
Carbothermic Reduction/Nitridization		Sintering		
Time	20 hours	Time	12 hours	
Temperature	1,550°C	Temperature	1,700°C	
Atmosphere	$N_2 - N_2/6\%H_2$	Atmosphere	Argon	
C:O Ratio	1.05			

Table 3. Futurix-FTA non-fertile processing parameters.

As for the low-fertile sample, the typical microstructure of non-fertile pellets (Figure 4) also lacks the endcapping that was prevalent with previous fabrications. Of particular interest is a "rind" observed around the circumference of the non-fertile pellet, shown on an optical macrograph of a radial pellet section in Figure 5. The phenomenon is most likely due to a local density effect resulting from an americium depleted zone.

The XRD results in Figure 6 show a two-phase mixture, which agrees with the AFC-1G non-fertile material: 1) an actinide nitride phase with a small amount of zirconium (Zr) in solution and 2) a zirconium-nitride phase with a small amount of actinides in solution. The residual weighted profile parameter of 17 percent for the non-fertile sample model is in the expected range for refinements of powder X-ray diffraction data.

SEM microprobe data of the non-fertile samples (Figures 7 and 8) show some heterogeneity on the microscale (10–20 percent variation in actinide concentration over 5–10 μ m), again with no macroscopic heterogeneity apparent. The results agree with the single-phase solid solution and two-phase mixture shown by XRD. The americium-depleted zone is apparent, extending approximately 200 μ m into the pellet.

Element	Specification Value	Actual Result
Pu	29.9 wt. %	37.83 wt. %
Am	30.2 wt. %	6.9 wt. %
Zr	31.5 wt. %	38.8 wt. %
С	≤3,000 ppm	11,800 ppm
0	≤3,000 ppm	2,620 ppm
Ν	8.3 wt. %	8.41 wt. %

Table 4. Summary of non-fertile chemistry results.



Figure 4. Non-fertile optical micrograph.



Figure 5. Non-fertile optical macrograph.



Figure 6. Non-fertile XRD pattern.

Planned Activities

The actinide nitride fuel development and fabrication work has yielded valuable nitride fuel processing information for the Advanced Fuel Cycle R&D program. There are several areas in which improvements must be made before actinide nitrides can be seriously considered as a transmutation fuel, as described below.

Minor actinide (MA) volatilization. Preliminary solutionization results do not show appreciable MA retention with respect to non-solutionized pellets.



Figure 7. Non-fertile SEM microprobe element maps.

However, it may be worthwhile to investigate solutionization of the matrix constituents during the nitridization step. Since volatilization is temperature dependent, reducing the nitridization and sintering temperatures through the study of atmosphere effects and sintering aids should be considered. Additionally, alternative techniques such as hot pressing and inductive heating may show promise.

Residual carbon and oxygen. Additional optimization of the carbothermic reduction (CTR) process is needed in order to reduce the residual carbon and oxygen content further. Researchers must

improve their understanding of CTR atmosphere effects and the form in which the material is carbothermically reduced. Improvements can also be made by more closely controlling the handling and storage of nitride materials during processing.

Compositional dependent behavior variation.

Numerous complexities of actinide nitrides need to be addressed. Although similar complexities are also expected for other fuel compositions, the responses will be different. Since a truly closed fuel cycle will have variable feedstock, it is also important to determine the sensitivity of the fabrication process to a range of compositions. The goal is to understand how the behavior of actinide nitrides varies with composition, so the fabrication process can accommodate the specific nitride fuel mix determined by a separations facility.



Figure 8. Non-fertile SEM microprobe line scan.

Molten Salt Technology for Reactor Applications

PI (U.S.): David F. Williams, Oak Ridge National Laboratory (ORNL)

PI (Europe): R. Konings, European Union's Joint Research Center-Institute for Transuranium Elements (JRC-ITU) Project Number: 2004-007-E Project Start: October 2004 Project End: September 2006

Collaborators: None

Research Objectives

Molten fluoride salts have been studied for use as fuel and as coolant for fluid-fueled nuclear reactor systems. Clean molten salts are candidates to transfer heat from a very high-temperature reactor (VHTR) to a thermochemical hydrogen production plant. Researchers are also studying their use as a coolant in fast-spectrum reactors.

In order to develop molten salt technology, a firm scientific basis is essential. The objectives of this project are to strengthen the scientific basis for molten salt technology and to address the important technological issues surrounding salt purification and on-line removal of impurities. A key issue is the impact of higher operating temperatures and larger temperature gradients on the salt's behavior and corrosive effects on reactor structural materials.

In the U.S., Europe, and Japan, renewed interest in molten salt coolants, the Generation IV molten salt reactor (MSR) concept, and partitioning and transmutation studies have led to new research programs. For example, the JRC-ITU research focuses on the assessment of physicochemical properties and calculation and measurement of phase diagrams of fluoride salts. The Institute is also studying fuel salts for use in thorium/uranium (Th/U) fuel cycles, transuranium transmutation cycles, and as coolant salts.

Research Progress

During FY 2006, researchers extended the evaluation of salts from primary-coolant applications to secondarycoolant applications. They also performed a preliminary economic evaluation and experiments on some novel salt systems. Primary coolants for high-temperature thermalspectrum reactors are restricted to the family of fluoride salts, but secondary coolants and fast-spectrum coolants can be constituted from fluoroborate or chloride salts. Researchers conducted a comprehensive evaluation of these additional coolant salts on the basis of their physical properties, including melting point, vapor pressure, density, heat capacity, viscosity, and thermal conductivity. Table 1 provides a summary of the results. Salts with lower formula weight exhibited superior heat-transfer metrics, but the least expensive salt constituents (NaCl, MgCl₂, KCl, NaF, AlF₂) are associated with salts with higher formula weights.

The team prepared high-purity LiF-KF-ZrF₄ and LiF-KF-AlF₃ salts and conducted density and calorimetric measurements on a number of compositions. Calorimetric measurements indicate relatively low melting point LiF-KF-ZrF₄ mixtures (approximately 400°C) for concentrations near 40 mol% ZrF₄, which may be suitable for future applications. For tests with the LiF-KF-AlF₃ salts, the team focused on the solubility of trivalent rare-earth fluorides. They found that the solubility of PrF₃ was lower than expected and much less than that exhibited in a simple alkali-fluoride (LiF-KF). This contrasts with the high solubility of rare-earth fluorides in ZrF₄-rich salts reported by researchers working in the area of infrared-transparent glasses.

Planned Activities

This project has ended. The U.S. investigator has issued reports and open literature articles based upon the decision that this project is complete.

				Не	at-Transfer Pr	operties at 70	O°C
Coolant	Formula Weight (g/mol)	Melting Point (ºC)	900°C Vapor Press (mm Hg)	ρ Density (g/cm3)	Ср Heat Capacity (cal/g-°C)	μ Viscosity (cP)	<i>k</i> Conductivity (W/m-K)
LiF-NaF-KF	41.3	454	0.7	2.02	0.45	2.9	0.92
LiF-NaF-RbF	67.7	435	0.8	2.69	0.236	2.6	0.62
LiF-BeF ₂	33.0	460	1.2	1.94	0.577	5.6	1.0
NaF-BeF ₂	44.1	340	1.4	2.01	0.52	7	0.87
LiF-NaF-BeF ₂	38.9	315	1.7	2.00	0.489	5	0.97
LiF-ZrF ₄	95.2	509	77	3.09	0.292	> 5.1	0.48
NaF-ZrF ₄	92.7	500	5	3.14	0.280	5.1	0.49
KF-ZrF ₄	103.9	390	1.2	2.80	0.251	< 5.1	0.45
Rb-ZrF ₄	132.9	410	1.3	3.22	0.200	5.1	0.39
LiF-NaF-ZrF ₄	84.2	436	~ 5	2.92	0.350	6.9	0.53
KF-AIF ₃	69.7	560	~ 8	1.80	0.320	1.4	0.5
LiCI-KCI	55.5	355	5.8	1.52	0.287	1.15	0.42
LiCl-RbCl	75.4	313		1.88	0.213	1.30	0.36
NaCl-MgCl ₂	73.7	445	< 2.5	1.68	0.262	1.36	0.50
KCI-MgCl ₂	81.4	426	< 2.0	1.66	0.276	1.40	0.40
NaF-NaBF ₄	104.4	385	9500	1.75	0.36	0.90	0.40
KF-KBF ₄	109.0	460	100	1.70	0.312	0.90	0.38
RbF-RbF₄	151.3	442	< 100	2.21	0.218	0.90	0.28

Table 1. Properties of candidate secondary-coolant salts for high-temperature applications.

Use of an Ionization Chamber in Fission Cross-Section Measurements

PI (U.S.): Tony Hill, Los Alamos National Laboratory (LANL)

Project Number: 2004-009-E

PI (Europe): P. Rullhusen, Institute for Reference Materials and Measurement (IRMM) Project Start Date: October 2004

Project End Date: October 2007

Collaborators: None

Research Objectives

The aim of this collaboration is to pool resources and competencies in cross-section measurement to produce improved fission cross-section data files for several key isotopes. Researchers need accurate data files on major and minor actinides for the design, optimization, and safety assessment of accelerator driven systems (ADS), fast reactors, and waste transmuters as well as on actinides relevant to the thorium-uranium fuel cycle. This work will contribute to the new ENDF B/VII and JEFF 3.1 libraries.

Research Progress

To date, the team has published the results of $^{231}Pa(n,f)$ cross-section measurements up to 3.5 MeV incident neutron energy and finalized cross-section measurements in the energy range: $E_n = 15$ to 21 MeV. Neutron production was based on the T(d,n)⁴He reaction using a solid TiT target. The neutrons produced are no longer mono-energetic above 17 MeV. Special care was taken to correct the measured fission cross-sections using data from a similar investigation of activation cross-sections at IRMM. For the uncertainty calculation, researchers considered the most conservative background scenarios, resulting in asymmetric point-wise uncertainties whose upper limit may reach 45 percent. The cross-section values obtained are considerably lower than most of the other values found in the literature. However, they agree with recent cross-section calculations made in collaboration with scientists from Bucharest University, including the most recent fission cross-section measurements performed for ^{233,231}Pa at IRMM and recent surrogate measurements below 10 MeV (Figure 1). The new experimental data and calculations have been

presented and included in the new International Atomic Energy Agency evaluation of the Th-U fuel cycle.

Due to scheduling of the Van de Graaff accelerator facility at IRMM, researchers postponed measuring the ²³³U fission cross-section as a function of incident neutron energy and deferred expanding experimental data to cover the incident neutron energy range from 4 and 10 MeV.

Researchers included the ²³⁷Np fission cross-section data in a new ENDF/B-VII evaluation. The data were also sent to French evaluators at CEA-Cadarache who determined that they are consistent with the MASURCA integral experiment benchmarks between 100 and 800 keV. The CEA evaluators also extracted a number of resonance parameters near thermal energies to record precision, which validated the measurement capability at the low-energy end of the incident neutron spectrum. This



Figure 1. The new ²³¹Pa fission cross-section results are shown in blue, along with a number of previous measurements. The ENDF and JENDL evaluations are also shown.

experiment has been fully documented and submitted for peer review and publication.

The fission cross-section measurement program at the Los Alamos Neutron Science Center (LANSCE) continued this year, focusing on ²⁴²Pu and ²⁴⁰Pu. These isotopes were a high priority due to large discrepancies in the data libraries in the fast region. The ENDF/B-VII and JENDL3.3 evaluations, shown in Figures 2 and 3, demonstrate the discrepancy of these libraries in the fast region. The data and evaluations include the contribution of a 10⁻⁴ ²⁴¹Pu impurity in the sample, which has little impact in the fast region. To complete the measurements, data for ²⁴¹Pu and ²³⁹Pu (Figures 4 and 5) were collected for background subtraction. The ²⁴¹Pu and ²³⁹Pu data are of high quality and will be analyzed in the upcoming year.

Planned Activities

The commissioning of the double-gridded chamber will



Figure 2. $^{242}\text{Pu}(n,f)/^{235}\text{U}(n,f)$ cross-section ratio as a function of incident neutron energy.



Figure 3. $^{240}\text{Pu}(n,f)/^{235}\text{U}(n,f)$ cross-section ratio as a function of incident neutron energy.

continue so that design benchmarks can be developed. The goal is to use the chamber to carry out precision fission cross-section measurements on highly active targets by combining signals from both fission fragments for triggering against radioactive alpha decays. Successful comparisons with well-measured cross sections will be required before a larger chamber is designed and built. The advanced detector will allow data to be taken on many samples simultaneously.

Researchers will continue to analyze the LANSCE fission data and expect to release the results for ²³⁹Pu, ²⁴¹Pu and ²⁴²Pu to evaluators in FY 2007. There will be limited beam available at LANSCE beginning in June 2007. Researchers plan to carry out measurements based on priorities set in the Transmutation Physics Working Group and expect to publish the results for ²³¹Pa above 15 MeV. However, further measurements are postponed due to overload of the Van de Graff facility at IRMM.



Figure 4. $^{\rm 241}\text{Pu}(n,f)/^{\rm 235}\text{U}(n,f)$ cross-section ratio as a function of incident neutron energy.



Figure 5. $^{239}\text{Pu}(n,f)/^{235}\text{U}(n,f)$ cross-section ratio as a function of incident neutron energy.

Nuclear-Assisted Hydrogen Storage and Safety Issues

PI (U.S.): Richard Vilim, Argonne National Laboratory (ANL)

Project Number: 2004-010-E

Project Start Date: May 2005

PI (Europe): Heinz Wilkening, Joint Research Commission–Institute for Energy (JRC-IE)

Project End Date: September 2007

Collaborators: None

Research Objectives

The objectives of this project are to identify and assess the key technical and economic issues associated with generating nuclear hydrogen and to define the remaining analytical and experimental work needed to achieve maturity of the nuclear hydrogen process. In this project, researchers are working with hydrogen production cycles previously identified as having a reasonable basis for technical and economic viability. The three main tasks are to:

- 1) Perform analytical research on the safety aspects of hydrogen production, storage, and transport
- Compare the efficiency, complexity, reliability/ availability, and economy of different hydrogen production and distribution systems
- Conduct a comparative analysis of Generation IV reactor types for hydrogen generation focusing on the engineering linkage between the hydrogen production plant and the nuclear heat source

Research Progress

In the second year of this project, researchers focused on designing and developing a model of a hightemperature electrolysis system (HTSE) plant coupled to a very high temperature reactor (VHTR). During this time, they identified the components in the plant, developed models for the components, and integrated the model to obtain a working steady-state simulation for the combined plant. This model is being used to investigate the sensitivity of overall plant efficiency to equipment configuration. It is also being used to investigate how best to operate such a plant. Since hydrogen production driven by nuclear power is a relatively new concept, control and safety issues must be understood before a prototype plant can be built. Both reactor safety and operability of the hydrogen plant are affected by separation distance between the two facilities, the former by the possibility of hydrogen inventory detonation and the latter by potential stability problems from large transport times.

Figure 1 shows the plant configuration that was analyzed, which consists of a 600 MWt VHTR and an HTSE plant with an electrical load of 275 MWe and a thermal load of 50 MWt. The electrolyzer pressure is set to 5 MPa and the inlet temperature to 850°C. The heat to raise the temperature of the reactants comes from energy recuperated from the product stream and from heat supplied by intermediate heat exchangers. The electrolyzer products pass through an expansion step to exit the plant at 70°C and 1.25 MPa. The gas phase of the products by mole fraction is 0.98 hydrogen and 0.02 water vapor.

The manner in which the HTSE process is operated determines the quality and quantity of reactor heat needed and has safety and reliability implications. If the electrolytic cell is operated at a higher voltage where resistive heating is significant, then it may be preferable to use recuperated ohmic heat to raise the final temperature of the reactants rather than reactor heat. The bulk of the energy needed to raise the temperature of the reactants is added in the vaporization step, which occurs at the relatively low temperature of 265°C. For the gas reactor, this temperature approaches that of the waste heat rejected to the ultimate heat sink; therefore, from an efficiency standpoint, this heat is inexpensive. Also, because a conventional heat exchanger technology is applicable, the HTSE process could be engineered to require minimal highquality reactor heat. Furthermore, a heat exchanger in the primary system at the reactor outlet may be unnecessary.



Figure 1. Network diagram for integrated VHTR/HTSE system.

Because a relatively small amount of high-temperature heat is needed, this heat may be added by electrical heaters in the chemical plant—without a significant efficiency penalty. High-temperature heat exchangers operating in the chemical plant, rather than on the nuclear side, might significantly improve the safety and maintainability of the nuclear plant.

Planned Activities

With the completion of this preliminary coupled plant design, representative numbers for the hydrogen production rate and inventory in the electrolyzer are available for investigating safety. There is still some question as to the size of the downstream gas inventory associated with on-site handling prior to shipping for off-site storage. This question will be addressed in FY 2007 in collaboration with EURATOM.

Transient safety analyses of hydrogen combustion will be performed using a computational fluid dynamics code. These analyses will address how environmental conditions (e.g., wind) and man-made structures (e.g., blast berms) can affect the potential combustion energy from an accidental hydrogen release. These analyses will provide images such as the computer simulation in Figure 2, which shows the hydrogen molar fractions (from which the flame front can be inferred) for a hydrogen pipeline leak in the vicinity of buildings. This data will be used to answer the important question: How far must the nuclear plant be separated from the hydrogen plant?



Figure 2. Hydrogen molar fractions in pipeline leak.

Experimental Investigation of Small Break LOCAs in Coupled Vessel/Containment Integral Reactors

PI (U.S.): Milorad Dzodzo, Westi Corp.	ghouse Electric Project Number: 2006-001-E
DI (Italy): Fosco Rianchi Ente n	Project Start Date: May 2006
Tecnologie, l'Energia e l'Ambiente	(ENEA) Project End Date: March 2010
Collaborators: Oak Ridge Nation	l Laboratory,

Collaborators: Oak Ridge National Laboratory, Società Informazioni ed Esperienze Termoidrauliche (SIET)

Project Abstract

The purpose of this project is to experimentally verify the behavior of integral reactors during accident conditions. The Global Nuclear Energy Initiative (GNEP) includes international deployment of smaller-scale, grid-appropriate reactors with fully passive safety systems, such as the International Reactor Innovative and Secure (IRIS). IRIS offers advantages over traditional passive safety features with its inherent, design-based approach to coping with small break loss-of-coolant accidents (LOCA) that does not rely on dedicated safety systems for coolant injection. The integral configuration of IRIS (without the primary loop external to reactor vessel) also precludes the possibility of a large break LOCA.

During a small break LOCA, the reactor vessel depressurizes due to heat removal and condensation by the integral steam generators, while the pressure in the high design pressure small spherical containment increases from the steam release through the break. The two pressures equalize relatively quickly, nullifying the pressure differential that drives the coolant egress, thus terminating the LOCA without any coolant injection or need for operator intervention. Numerous computer simulations performed under a variety of conditions indicate that the core remains safely covered at all times. However, an accurate and comprehensive experimental investigation is necessary to validate analytic tools and confirm this result. This work will require extensive modifications and upgrades of SIET's existing AP600 test facility to represent the characteristics of an integral rather than a loop-type reactor and the vessel/containment coupling. Researchers will develop an analytical program to guide testing. The four-year duration supports the IRIS goal of submitting the Design Certification application in 2010.

The project entails the following tasks:

- Design a small break LOCA experimental facility for the coupled vessel/containment configuration that also allows investigation of other accident scenarios
- Review existing QA plans and update as necessary to satisfy IRIS integral testing needs
- Perform pre-test analyses to guide and evaluate the actual tests
- Procure components and assemble the equipment necessary to modify, construct, and commission the test facility
- Conduct the test matrix, including shakedown tests
- Evaluate results and prepare a comprehensive report to be incorporated into the safety documentation required by the IRIS application for NRC final design approval

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8.0 U.S./France Collaboration

U.S. Secretary of Energy Spencer Abraham and the CEA Chairman, Pascal Colombani, signed a bilateral agreement on July 9, 2001, to jointly fund innovative U.S./French research in advanced reactors and fuel cycle development. The U.S./France collaboration was the first I-NERI agreement to be fully implemented; 18 U.S./France collaborative research projects have been awarded since FY 2001.

8.1 Work Scope Areas

R&D topical areas for the U.S./France collaboration include:

- Advanced gas-cooled reactors
- Advanced fuel and materials development
- Radiation damage simulation
- Hydrogen production using nuclear energy

8.2 Project Summaries

Five research projects begun in FY 2001 and 2002, the initial years of the collaboration, have been completed previously, along with five more in FY 2006. Work is continuing on 6 collaborative projects initiated in FY 2004 and two new projects started in FY 2006.

A listing of the I-NERI U.S./France projects that are currently underway follows, along with summaries of the accomplishments achieved in FY 2006.

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Hydrogen Process to High-Temperature Heat Source Coupling Technology

PI (U.S.): Steven Sherman, Idaho National Laboratory (INL)

Project Number: 2004-001-F

PI (France): Dominique Barbier, Commissariat à l'Energie Atomique (CEA) – Cadarache

Project Start Date: August 2004

Project End Date: July 2007

Collaborators: None

Research Objectives

This project will develop the technology for coupling a high-temperature heat source to hydrogen (H_2) production processes for the Nuclear Hydrogen Initiative. The primary focus of this I-NERI research is to develop the Sulfur-Iodine (S-I) cycle process technology. Specific research objectives are to:

 Propose design schemes and component connections, compare their characteristics and select a component arrangement that will reduce heat losses for the following interfaces:

Reactor

- └→ Intermediate heat exchanger
 - → High-temperature step
 - └→ Medium-temperature step
 - └→ Low-temperature step
- Evaluate design solutions for heat transmission and exchangers including technical and industrial feasibility, flexibility of coupling schemes, and conversion energy losses.
- Develop potential design schemes.
- Develop a model and evaluate the complete heat balance for each design scheme, analyzing the use of energy (electricity and/or heat) in relation to hydrogen production. This model will help researchers understand the process behavior during normal operation, transient, and accidental conditions. The information from the models and simulations will provide the data needed to perform availability and safety analyses. It will also provide interface data for the economic evaluation and cost estimation work teams.

Research Progress

The most promising current process for very high temperatures, in terms of efficiency, is the S-I closed thermochemical cycle. That is the basis for the specific studies being undertaken on coupling a nuclear and hydrogen plant. Following is a summary of the research team's progress.

- Researchers evaluated pre-design options of a co-generation plant, mainly based on efficiency optimization. They considered several design schemes to meet the very flexible operational requirements for nuclear electricity generation and hydrogen production, taking into account criteria on industrial feasibility and economics.
- The team developed a design of the entire fluid transfer circuit between the primary heat exchanger and the preliminary hydrogen plant chemical flow sheet. This design is closely linked to a safety approach based on defense-in-depth principles, allowing analysis of all system configurations under normal, incident, and accident operating conditions.
- They developed a dedicated multi-code platform. This platform mainly concerns the use of codes developed within the CEA framework:
 - Pre-design codes for stationary nuclear plant operation (links to H₂ plant with mainly heat sinks and source boundary conditions)
 - ♦ Conceptual design of the coupling fluid circuits
 - ♦ Hydrogen plant design
 - Nuclear safety codes extended to chemical plant operation for the final safety report

- Researchers focused on an alternative hydrogen production process—high-temperature electrolysis. CEA is developing a specific electrolyzer model to improve thermodynamic efficiency and to develop a flow sheet compatible with industrial-scale H₂ production.
- Researchers evaluated the feasibility of some specific components, mainly the high-temperature heat exchanger with the SO₃ decomposer and the design of a test loop for mock-ups.

The U.S. research team advanced the science of coupling a future large-scale hydrogen production facility to a high-temperature heat source (such as a Generation IV nuclear reactor), focusing on:

- Materials
- Heat exchanger designs
- System designs
- Safety

Researchers tested materials to determine their suitability to withstand the very high temperatures and aggressive environments of the coupling process. They tested the tensile properties of Hastelloy C-22, C-276, Incoloy 800H, Waspaloy, Zr-705, and Inconel 617 at temperatures up to 600°C. Inconel 617 and Waspaloy were the best performers, followed by 800H. Future measurements will be extended to 1,000°C. Researchers also studied high-temperature deformation of these materials, concluding that ductile failure occurred at elevated temperatures. Optical microscopy revealed conventional solution-annealed microstructures. Ceramic and carbon/silicon-carbide (C/SiC) materials were also tested to identify candidate fabrication methods for forming flow channels in plates, and for laminating and infiltrating multiple plates. Thermal stress was analyzed at full heat exchanger scale. The high-temperature heat exchanger plates need to be redesigned based on the stress analysis results.

The leading choice of liquid salt being considered for use as a heat transfer fluid for the system interface is FLiNaK (46.5% LiF, 11.5% NaF, and 42% KF by weight at 454°C). Researchers performed an initial study of FLiNaK to examine corrosion control mechanisms and methods for controlling the formation of HF on H_2SO_4 in-leakage. They also commissioned a Molten Salt Working Group to bring together experts on liquid salt technology.



Figure 1. Liquid salts are being investigated as possible heat transfer media.

Researchers used several different techniques to investigate compact heat exchanger designs, including thermal/mechanical stress modeling using ANSYS 9.0; fluid flow using FLUENT and MATLAB; parametric studies of strip fin thickness, gap length, channel dimensions, fin length, pitch, sizing, grid independence; and development of parallel processing capability. Researchers are analyzing intermediate heat exchanger designs using ceramic and composite material for helium/molten salt interactions. They are also analyzing the optimization of an offset strip fin design for a compact, ceramic-composite heat exchanger. Figure 2 shows a prototype heat exchanger design.



Figure 2. Prototype configuration of a silicon-carbon heat exchanger.

With the thermal-hydraulic modeling of a system interface, researchers examined the relative efficiency differences among basic configurations. They compared 2 MPa helium, 7 MPa helium, and liquid salt (FLiNaK, NaBF₄-NaF) and evaluated metallic material constraints. This work concluded that an interface pressure of 7 MPa would exceed metal creep constraints in the interface/ process heat exchanger using a simple heat exchanger design. However, the team found that liquid salt boosts efficiency 6 percent over 7 MPa helium, with a 30 percent smaller system volume. Researchers also studied aspects of the systems interface in more detail, including parallel versus concentric pipe arrangement, pipe diameters and temperature losses versus distance, and stress and creep rupture limits of printed circuit heat exchangers.

An initial probabilistic risk assessment of the separation distance required between the nuclear plant and the hydrogen facility resulted in a minimum spacing of 110 meters. Incorporating protective barriers and other technologies into the design may reduce this to as low as 60 meters.

Planned Activities

Planned work will support materials corrosion studies, studies of liquid salts and liquid salt redox control to reduce materials corrosion problems, and the design of laboratoryscale heat transfer loops for testing heat exchangers and other components. Future work includes:

- Performing materials corrosion studies under static conditions to support system interface development and design of the sulfuric acid systems in the S-I hydrogen production process.
- Designing a sulfuric acid flow loop that would be capable of exposing test samples to liquid and vapor sulfuric acid with contaminants, under pressure for hundreds of hours. Preparing the necessary safety analyses, parts lists, etc., to a sufficient level of detail to authorize construction and use.
- Studying liquid salt redox control methods for reducing materials corrosion. Investigators will synthesize or purchase FLiNaK and NaBF₄–NaF; characterize impurities in the salt; employ the salt purification techniques of hydrofluorination, filtration, and electroplating; assess their effectiveness and utility at a larger scale; and construct test equipment for assessing materials corrosion, metals dissolution, and redox control for future experiments.
- Exploring different cladding techniques and clad material/base metal options with the goal of defining suitable combinations that can be used to fabricate HIx decomposition section components, such as boilers and heat exchangers. Fabricating samples and prototypes using the chosen fabrication routes and testing for corrosion resistance and mechanical stability.
- Continuing to develop and improve steady-state models of the system interface and related components to support future safety analyses and estimates of process efficiencies.

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OSMOSE - An Experimental Program for Improving Neutronic Predictions of Advanced Nuclear Fuels

PI (U.S.): R. Klann, Argonne National Laboratory	Project Number: 2004-002-F
PI (France): J. Hudelot, Commisariat à l'Energie Atomique (CEA) – Cadarache	Project Start Date: October 2004
Collaborator: CEA-Valrho	Project End Date: September 2007

Research Objectives

The goal of the OSMOSE program (Oscillation in Minerve of Isotopes in "Eupraxic" Spectra) is to create a database of the measured reactivity effect of minor actinides in known neutron spectra. This database will be used as an international benchmark. Researchers will measure very accurate integral reaction rates in representative spectra for the actinides that are important to future nuclear system designs, particularly the Generation IV reactor program and the Advanced Fuel Cycle Initiative. The work will provide the experimental data that is needed for improving basic nuclear data files.

The OSMOSE program is generic; that is, it will measure

Element	Isotope
Thorium	²³² Th
Uranium	233U 234U 235U 236U 238U
Neptunium	²³⁷ Np
Plutonium	²³⁸ Pu ²³⁹ Pu ²⁴⁰ Pu ²⁴¹ Pu ²⁴² Pu
Americium	²⁴¹ Am ²⁴³ Am
Curium	²⁴⁴ Cm ²⁴⁵ Cm

Table 1. Important heavy nuclides.

reaction rates over a broad range of isotopes and spectra corresponding to specific experimental lattices in the CEA-Cadarache **MINERVE** reactor (thermal, epithermal, moderated/fast, and fast spectra). OSMOSE will provide precise experimental, integral, absorption crosssection data for a majority of the heavy nuclides important to reactor and nuclear fuel cycle physics, as shown in Table 1.

Researchers can use these accurate measurements of minor actinides in various spectra—from over-moderated thermal spectra to fast spectra—in coordination with international nuclear data to characterize deficiencies in the basic nuclear data libraries, identify their origins, and propose paths towards correcting them.

Research Progress

During FY 2006, researchers made technical progress on the OSMOSE project in reactor modeling, sample fabrication, experimental measurements, and data analysis. Following is an overview of accomplishments in each area.

Reactor Modeling. Reactor modeling efforts had two major focuses: 1) to generate reactivity-worth estimates of the samples to support the data analysis of the experiment measurements and 2) to generate spectra for the OSMOSE configurations for comparison to Generation IV systems.

Researchers completed a series of calculations of the reactivity-worth of the OSMOSE samples in the MINERVE reactor with the R1-UO2, R2-UO2, and R1-MOX core configurations using a deterministic model based on the REBUS code system. To be consistent with the two-dimensional modeling being performed by CEA and to improve the sensitivity of the calculations, they generated new deterministic models using the DRAGON code.

Within the MINERVE reactor, different spectra were achieved by changing the experimental lattice. The R1-UO2 configuration corresponds to a light water reactor (LWR) loaded with UO_2 ; the R1-MOX corresponds to an LWR loaded with a mixed oxide matrix; the R2-UO2 corresponds to an over-moderated LWR spectrum; and MORGANE/S corresponds to an epithermal spectrum representative of under-moderated reactors. The spectra in the central sample channel of the R1-UO2 and R2-UO2 configurations

Comparison of the Spectral Composition						
		MINEF	VHTR			
Energy Kange	R1-UO2 R2-UO2 R1-MOX MORGANE/S			Uranium	DBMHR	
< 1 eV	0.2045	0.3645	0.1232	0.0444	0.2851	0.0240
1-10 eV	0.0598	0.0620	0.0638	0.0467	0.0845	0.0856
10 eV – 1 keV	0.1408	0.1281	0.1561	0.1607	0.2021	0.2687
1 keV - 110 keV	0.1823	0.1440	0.2022	0.2515	0.2260	0.3257
> 110 keV	0.4125	0.3014	0.4548	0.4967	0.2023	0.2960
Total	1.0	1.0	1.0	1.0	1.0	1.0

Table 2. Comparison of the spectra for several Generation IV systems.

were calculated with the MCNP code, using a 99 energy group structure. Researchers used the deterministic code, DRAGON, with the same two-dimensional mini-lattice (11x11 pins) model and 172-group neutron library to validate the MCNP results. They observed that the spectra calculated by DRAGON and MCNP agreed well for the configurations. The spectra were then compared to several representative spectra for Generation IV systems.

Sample Fabrication. Preparation of the OSMOSE samples is a multi-stage process which includes purifying the isotope feedstock materials, creating an oxide material, mixing and fabricating sintered oxide fuel pellets, assembling the fuel pellets into the sample, and welding the clad for double-encapsulation.

The last material prepared was the ²³³U sample, procured without ²³²U or other decay products. The material was purified and processed in a glove box. All of the remaining isotopes are now ready for pellet fabrication.

Researchers fabricated the second set of pellets composed of six samples:

- $UO_2 + {}^{238}PuO_2$
- $UO_2 + {}^{240}PuO_2$
- $UO_2 + {}^{241}PuO_2$
- $UO_2 + {}^{241}AmO_2(1)$
- $UO_2 + {}^{241}AmO_2(2)$
- ThO₂

They obtained good agreement with the specifications, although the mean density of several samples appeared a little low. Cladding and the welding took place in glove boxes due to the radioactivity of the actinide material. After CEA-Cadarache accepted the six sample pins, the samples were delivered to MINERVE. Researchers have performed the isotopic and chemical characterization of the doped actinides and have completed the analysis of the pellets from the first batch of samples. The results do not show any important values far from the target values, which means that the manufacturing was performed without pollution or cross-contamination of the samples. Arrangements were made to ship pellet samples to ANL to perform a cross-comparison of the chemical analysis results and to reduce the uncertainties on the sample compositions.

Experimental Measurements. Over the course of the fiscal year, several series of measurements have been performed. Oscillation measurements were conducted in the R1-UO2 configuration using the first set of OSMOSE samples (²³⁹Pu, ²⁴²Pu, ²³²U/Th, ²³⁴U, URE, ²³⁷Np, and natural U).

CEA shipped the second set of samples, consisting of ²⁴⁰Pu, ²³⁸Pu, ²⁴¹Am (2 samples), ²⁴¹Pu and pure ²³²Th, from Marcoule to Cadarache and performed modified conversion ratio measurements in the R1-UO2 core configuration. They also conducted oscillation measurements in the R1-UO2 core configuration using the second set of OSMOSE samples, completing measurements in the R1-UO2 configuration for all of the OSMOSE samples that are currently available. Then, the core was reloaded with the R1-MOX configuration. Safety measurements and calibration of the pilot rod in the R1-MOX core configuration were completed. Following that, CEA began oscillation measurements of the OSMOSE and calibration samples in the R1-MOX core configuration.

Data Analysis. Researchers obtained excellent agreement (within 1 σ) between APOLLO2 and DRAGON calculation results for ²³²Th, ²³⁹Pu, and ²⁴²Pu, with JEF2.2 and JEFF3.1 data libraries. The results with ENDF/B-VI are consistent for the ²³²Th and ²³⁹Pu samples. However, there are discrepancies for ²³⁴U and ²³⁷Np between the two models. These may be a result of potential material balance or self-shielding differences in the models. Further investigation is ongoing to resolve the differences.

Researchers achieved good agreement (within 2 σ) between APOLLO2 and DRAGON calculation results and experimental results for all cases except ²³⁷Np and ²³⁴U. For ²³⁷Np, they obtained consistent calculation-to-experimental (C/E) results for the two samples using the APOLLO model. However, experimental results underestimated the calculation by about 8 percent with JEF2.2 and 13 percent with JEFF3.1. As the ²³⁷Np signal is mainly due to both thermal and epithermal captures, it is not possible to identify whether the thermal part of the capture cross section is underestimated, the integral resonance, or both. The C/Es obtained in future core configurations should allow more precise identification of the origin of the deviation between calculation and experimental results.

The DRAGON model results appear to show a systematic bias such that all of the calculated values are higher than the experimentally measured values, although the model has not yet been validated. Otherwise, results are reasonably consistent. Following are several potential sources of bias and discrepancy between these results and those from the APOLLO2 model:

- Self-shielding effects may not have been appropriately considered in collapsing from the continuous energy cross-sections to the multi-group cross-sections, especially in the case of the minor actinides
- The DRAGON model did not account for the slight variation in sample height as discussed in the APOLLO2 model
- There may be a small bias in the calibration curve based on the model of the calibration samples
- There may still be some minor differences in the compositions of the samples, which are still being investigated

Planned Activities

Several activities related to reactor modeling are planned for FY 2007. Researchers will perform pre-analysis estimates of the reactivity effect of the OSMOSE samples in the MORGANE/R, MORGANE/S, and R2-UO2 core configurations. Additionally, the results of the OSMOSE samples in the R1-UO2 and R1-MOX configurations will be studied and re-analyzed as the results of the chemical analysis become available. The main goal of reactor modeling efforts will be to finalize the models for the R1-UO2 and R1-MOX configurations and to prepare a benchmark report for these configurations and experimental results.

The team will study the OSMOSE samples in the planned core configurations to determine the relevance of these configurations to the Generation IV program. Specifically, the configurations will be compared to block-type very high-temperature (VHTR) and gas-cooled reactor (GCR) systems. Although initial spectra have been previously obtained and compared, a more detailed assessment must still be performed.

The last five samples—²⁴⁴Cm, ²⁴⁴⁺²⁴⁵Cm, ²⁴³Am (x 2) and ²³³U—will be fabricated and shipped. Researchers will complete experiments for the first and second sets of OSMOSE samples and calibration samples in the R1-MOX configuration, along with measurements for the third set of samples (including the two Curium samples).

OSMOSE samples will be oscillated in the R2-UO2 configuration from October 2007 to March 2008, followed by five months in the MORGANE-R configuration. Researchers will request a safety report and authorization for both of these core configurations, reflecting the adaptation of the new calculation scheme to the MORGANE-R loading (MOX 11 percent with fuel pins in a hexagonal pitch). A separate safety authorization will be needed for measuring curium samples in all core configurations.

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Thermal-Hydraulic Analyses and Experiments for GCR Safety

PI (U.S.): Richard R. Schultz, Idaho National Laboratory

Project Number: 2004-003-F

Project Start Date: January 2005

Project End Date: January 2008

PI (France): Denis Tenchine, Commissariat à l'Energie Atomique (CEA)

Collaborators: Argonne National Laboratory (ANL); Utah State University

Research Objectives

The objective of this collaborative research project is to provide experimental and computational benchmark data for improving and validating gas-cooled reactor (GCR) thermal-fluid-dynamics codes. These codes are proposed for evaluating decay heat removal concepts and designs of the very high-temperature reactor (VHTR) and gas-cooled fast-spectrum reactor (GFR). The complex geometries and wide temperature ranges found in these reactor designs can lead to significant variations in gas thermodynamic and transport properties during loss-of-flow and loss-of-coolant accident scenarios and during reduced power operations.

Research Progress

This research effort is divided into two areas: 1) benchmark analyses and validations and 2) benchmark experiments for complex GCR geometries. Below is an overview of the research progress that has been made in each area.

BENCHMARK ANALYSES AND VALIDATIONS

Feasibility Study for VHTR Water-Cooled RCCS. Researchers performed a scaling analysis to demonstrate that the ANL Natural Convection Shutdown Heat Removal Test Facility (NSTF) is capable of modeling a water-cooled reactor core cooling system (RCCS) at a prototypic-scale laterally and half-scale vertically. Researchers must validate this scaling analysis by evaluating steady-state and transient performance of a reference RCCS design using computational fluid dynamics (CFD) techniques and system codes. This is challenging due to strong 3-D effects, turbulent flows, validity of heat transfer correlations, nucleate boiling in the tubes, water flashing during transients, and two-phase-flow phenomena.

Figure 1 illustrates the experimental system, based on the pebble bed modular reactor (PBMR), which consists of a series of oval standpipes surrounding the reactor vessel to provide natural convection and radiative cooling of the reactor cavity during both normal and off-normal conditions. During normal operation, water is pumped to the standpipes from a heat sink outside the containment. During off-normal conditions, forced convection flow and the ultimate heat sink are presumed to be lost, so water is drawn passively by natural convection to remove decay heat and maintain acceptable fuel temperatures.

Multi-scale modeling of GCR core. Researchers coupled a whole-core modeling approach (porous media) and a subassembly description (CFD approach) using the Trio_U code. They chose a domain decomposition method consisting of two simulations of the same domain subzone, each able to influence the other through boundary conditions or volumetric source terms.

The Trio_U application uses a porous media approach for the whole core and 3-D CFD methods at the subassembly level. Boundary conditions established on temperature transmit data from the porous media to the CFD approach, and a volumetric thermal source term passes data from the CFD approach to the porous media.



Figure 1. Sketch of NSTF system modifications for water-based RCCS testing.

The GCR core is made up of hexagonal subassemblies with plate-type fuel elements, as shown in Figure 2. Researchers simulated a channel blockage on the central subassembly in the CFD description to study the consequences on the porous media scale, including the effect of the volumetric thermal source term.



Figure 2. Porous media domain with coarse mesh, showing details in the central subassembly.

Turbulent Boundary Layer Equations (TBLE). Previous work in a plane channel configuration has shown that the TBLE model provides improved results compared with standard wall models. The TBLE model solves the Reynolds-averaged Navier-Stokes (RANS) thin boundary layer equations in order to supply the large-eddy simulation (LES) computation with the wall shear stress. Researchers incorporated this model into the Trio U code for both structured and unstructured grids and applied it to Generation IV reactor systems.

In 2006, researchers introduced a variable-density version (TBLE- ρ) to simulate the non-isothermal, unsteady, turbulent wall-bounded flow in GFRs. This method is based on solving four equations: streamwise and spanwise momentum, temperature, and the state equation. Results of testing in a plane channel configuration prove that TBLE- ρ recovers the TBLE model in quasi-isothermal flows. For highly non-isothermal cases, the revised

model yields improved velocity and temperature results and gives acceptable skin friction and wall heat flux (<5 percent error). In future work, researchers will continue validating and deploying these features into the Trio_U code.

CFD modeling of HTR-10. The HTR-10 reactor is a pebble bed High-Temperature GFR designed and operated by the Institute of Nuclear Energy Technology (INET) in China. Researchers are benchmarking steady-state temperature distributions against experimental results and an analytical model created with the ARCTURUS CFD code (part of the CAST3M fluid and structural mechanics code).

Figure 3 depicts the original and revised benchmarks in the radial and axial directions, which show no significant impact on the key-temperatures results. The maximum fuel temperature of 875°C occurred in the lower core, where average power density was 2.33 W/cm³. Table 1 shows the maximum temperatures inside the pebbles.



Figure 3. HTR-10–CAST3M/ARCTURUS benchmark results: (a) radial and (b) axial temperatures.

Component	Maximum Temperature
Fuel Element Center	889°C
Surface	865°C
Side Reflector	520°C
Bottom Reflector	808°C

Table 1. Maximum temperature of the main pebble components.

This benchmark exercise demonstrated the capabilities of the CAST3M/ARCTURUS code to simulate steady state temperature fields, including complex convective, conductive, and radiative heat transfer. Some discrepancies between the calculated and the measured values still exist, especially in by-pass regions where convective heat transfer is important. Additional experimental results may be necessary to evaluate the model under transient conditions.

BENCHMARK EXPERIMENTS

Experimental Measurement of Flow in VHTR.

Researchers began developing the computational tools needed to predict the thermal-hydraulics conditions and safety margins of the VHTR design. They designed and built matched-index-of-refraction (MIR) fluid dynamics experiments to develop benchmark databases for CFD solutions of the momentum equations, scalar mixing, and turbulence models for typical VHTR plenum geometries. The MIR experiments use optical techniques to measure flow characteristics without disturbing the flow field. The refractive index of the working fluid, light mineral oil, is maintained by controlling temperature in the test section to within 0.1°C. Figure 4 illustrates the flow in the lower plenum of the prismatic VHTR, which can be considered as multiple buoyant jets into a confined density-stratified crossflow with obstructions. The arrows in the figure provide intuitive examples of expected paths. The flow rate (or Reynolds number) increases from right to left as more incoming jets participate.



Figure 4. Examples of possible flow paths in the lower plenum of a typical prismatic VHTR concept.

Researchers fabricated the scale model shown in Figure 5, which consists of quartz posts and half-posts to simulate flow features found in the VHTR design and match the refractiveindex of the working fluid. Figure 6 shows the results of initial mean velocity vector measurements of two operating jets. Flow is from top to bottom in a vertical plane, which is illuminated by a thin laser light. Higher velocities (yellow/orange/red) may be observed in the central regions of the jet inlet ducts (upper half) and below the duct inlets in the lower plenum area (bottom half). Lower velocities (blue/purple) occur along the walls of the jet inlet ducts (upper half) and between the ducts (bottom center).



Figure 5. Plan view of scale model for the lower plenum experiment.

Planned Activities

Following are the activities researchers will conduct over the next fiscal year:

- Continue to validate the TBLE-p model and deploy its features in the Trio_U code
- Conduct additional experiments to evaluate the CAST3M/ARCTURUS model under transient conditions
- Continue developing computational tools and MIR testing needed to predict the thermal-hydraulics conditions and safety margins of the VHTR design



Figure 6. Mean velocity vectors of two inlet jets.

SiC/SiC for Control Rod Structures for Next Generation Nuclear Plants

PI (U.S.): William Windes, Idaho National Laboratory (INL)

PI (France): Jacques Lamon, University of Bordeaux

Collaborators: Oak Ridge National Laboratory (ORNL), Pacific Northwest National Laboratory, Commissariat à l'Energie Atomique (CEA)

Research Objectives

This research project was designed to investigate issues surrounding the development of tubular geometry SiC/SiC composite material for control rod and guide tube applications in a very high-temperature reactor (VHTR). Fiber-reinforced ceramic composites are considered the only viable choice to be used for these primary safety components because they must be capable of withstanding the extremely high temperature and dose levels inside the reactor core. Researchers planned to study the mechanical, thermal, and radiation-damage response of composites fabricated in the U.S. and France.

The Department of Energy (DOE) recently decided to eliminate SiC composites from its Next Generation Nuclear Plant (NGNP) research program. Therefore, all SiC research tasks have been reduced or eliminated for the next fiscal year and the I-NERI collaboration with France will end, including planned irradiation programs at Oak Ridge National Laboratory (ORNL) and INL using French micro-composite material, specimen testing/comparisons, and collaborative development of improved SiC composites.

Research Progress

Project Number: 2004-004-F

Project Start Date: October 2005

Project End Date: September 2006

Researchers began designing and fabricating new microcomposite samples for irradiation stability studies inside the ORNL High Flux Isotope Reactor (HFIR). These new microcomposite samples utilize the superior French interphase materials with the hope of producing a better composite for irradiation applications.

Mechanical Testing Development. Researchers fabricated a number of tubular test samples in order to perform a series of mechanical tests on tubular composite materials, which are shown in Figure 1. These samples were fabricated in order to support the development of ASTM-approved testing standards for these composite materials. ASTM "robustness" and "round-robin" tests will be used to develop specific testing standards for room- and high-temperature applications. These ASTM-approved tests are required to validate the composites for use within a nuclear environment. Initial mechanical testing will involve only low-temperature tensile tests where the strength of the tubular composite is compared to flat plate composite specimens. Later tests will be conducted at high (900°C) and very high (1,100°C) temperatures to determine the effects of temperature on composite strength.



Figure 1. Tubular and flat test samples fabricated for ASTM test standard development.

The University of Bordeaux has agreed to be one of the testing laboratories participating in these initial testing development activities. All participating laboratories (including the University of Bordeaux) were provided with at least four "straight" tubular composite specimens for tensile testing, representing the minimum number of samples for statistically significant results. All laboratories have agreed to test these initial composite samples in uniaxial tension at room temperature and share their results.

Irradiation Stability Testing of Mini-Composites. The French SiC composite structures have a unique interphase structure that can provide superior irradiation performance to these ceramic matrix composite (CMC) materials. They have pioneered the use of 2-D woven SiC/ SiC composites and also nanoscale-multilayered pyrolytic carbon/silicon carbide interphases. These improved interphase composites could provide superior irradiation stability to the 2-D braided composite structures currently being proposed for the U.S. and French programs. The French have agreed to begin designing and fabricating new micro-composite samples using this innovative fabrication approach. These micro-composite samples would be irradiated inside HFIR to levels of 5–10 dpa, which are comparable to expected NGNP values.

Planned Activities

All planned activities have been canceled due to the termination of U.S. funding.

The University of Bordeaux has agreed to assist with future high-temperature mechanical testing planned after the room-temperature tests have been completed. These tests were to be conducted at high (900°C) and very high (1,100°C) temperatures to determine the effects of temperature on tubular composite strength.

If the irradiation studies demonstrated a clearly superior composite structure from the French fabrication method, tubular samples were to be fabricated for insertion into the composite irradiation creep capsule for testing in the Advanced Test Reactor (ATR) at INL. Researchers intended to compare these new composite structures "head-to-head" with the current U.S. fiber and preform fiber architectures for irradiation creep testing.

Assessment of Existing Physics Experiments Relevant to VHTR Designs

PI (U.S.): Temitope A. Taiwo, Argonne National Laboratory (ANL)

PI (France): Robert Jacqmin, Commissariat à l'Energie Atomique (CEA)

Project Number: 2004-005-F

Project Start Date: October 2003

Project End Date: September 2006

Collaborator: Idaho National Laboratory

Research Objectives

The objective of this project was to assess experimental data and benchmark tests applicable to the qualification of Generation IV system design and analysis physics tools. The research team identified existing data and evaluated its suitability for use in the qualification and quality assurance computer codes and databases designed for reactor physics analysis of the very high-temperature reactor (VHTR). This work supports subsequent efforts to document the benchmark specifications and measured results in a standard format for use in VHTR software quality assurance efforts.

In this work, the team:

- Evaluated the adequacy of existing critical experiments and nuclear data
- Defined target accuracies for pertinent core parameters
- Conducted sensitivity studies to assess the relevance of experiments to VHTR
- Identified additional integral experiments and/or nuclear data evaluation and measurements that are required

Research Progress

Over the course of this project, the researchers accomplished the following tasks:

 Completed an assessment of experimental tests and measurements performed since the early 1960s that could be used to validate and verify reactor physics codes and data for VHTR/Next Generation Nuclear Plant (NGNP) analysis

- Evaluated the potential impact of nuclear data uncertainties on a number of performance parameters (core and fuel cycle) for the prismatic block-type VHTR
- Evaluated design target accuracies, which could be relevant in successive design phases, and evaluated nuclear data improvement requirements (see Table 1)
- Performed a preliminary assessment of the relevance of the existing experimental tests to a prismatic blocktype VHTR by comparing the values of core parameters for the systems to that of a representative reactor
- Evaluated and analyzed results of experimental tests to create standard benchmark problems that can be used to validate computer codes and nuclear data
- Worked with international groups (OECD/NEA and the Generation IV International Forum) to improve the analytical tools (code and data) for analyzing advanced reactor systems, with a focus on the VHTR

The team found that the temperature and burnup operating range of the VHTR requires an extensive experimental component to thoroughly validate the reactor physics calculations, including Monte Carlo calculations. This will entail a significant follow-on effort. The team has identified the steps that must be taken to justify, define, and perform the new experiments.

The researchers also completed two technical documents on VHTR benchmarking during this past fiscal year.

Parameter	Target accuracy (1σ)
Criticality	300 pcm (operation) 500 pcm (safety)
Local power (in fuel compact)	6%*
Burn-up (cycle length)	1% (~ 500 MWd/t)
Doppler coefficient	20%
Moderator temperature coefficient	1 pcm/°C
Beta-effective (β _{eff})	10%
Prompt neutron lifetime	10%
Control rod worth Integral Differential 	10% 15% (locally)
Nuclide inventories at end of life Main fissile isotopes Fertile isotopes Minor actinide/fission products 	4% 5% 20%
Poisons	< 3% (capture)
Shutdown margin	10%
Fuel decay heat	30%**
* 2% in pin-wise fission rate of fresh fuel + 4% in	main fissile isotope concentration

 2% in pin-wise fission rate of fresh fuel + 4% in main fissile isotope concentration of irradiated fuel

** 20% on radio-nuclide concentrations + 10% on decay half-lives and energies

Table 1. Uncertainty requirements proposed for $\rm UO_2\mathchar`-$ and $\rm PuO_2\mathchar`-$ fueled HTRs (source: AREVA-NP and CEA).

Planned Activities

This project is complete.

For the proposed additional experiments, the researchers recommended leveraging existing international activities (e.g., HTTR, HTR-10, and ASTRA). Obtaining all the pertinent data from international organizations is a potential problem due to its proprietary nature. To facilitate sharing of this information, researchers will pursue bilateral or multi-lateral agreements. Additionally, since new integral measurements cannot be ruled out, they will also pursue costeffective approaches for creating high-quality measurements in collaboration with other local and international organizations. In this case, they recommended that an experts group consisting of analysts and experimentalists be convened to provide justification for the experiments and estimates of the associated costs.

GFR Physics Experiments in the CEA-Cadarache MASURCA Facility

PI (U.S.): Temitope A. Taiwo, Argonne National Laboratory (ANL)

Project Number: 2004-006-F

PI (France): Robert Jacqmin, Commissariat à l'Energie Atomigue (CEA)

Project Start Date: October 2003

Project End Date: September 2006

Collaborators: None

Research Objectives

Gas-cooled fast reactor (GFR) designs are being developed to meet Generation IV goals of sustainability, economics, safety, reliability, proliferation resistance, and physical protection. CEA-Cadarache investigated core physics issues relevant to Generation IV GFR designs through an experimental program known as ENIGMA—the **E**xperimental **N**eutron **I**nvestigation on **G**as-reactors at **MA**SURCA. The objectives of ENIGMA are to 1) define configurations of the CEA-Cadarache MASURCA reactor which have neutronic characteristics similar to the candidate GFR designs and 2) extend the validation domain of the neutronics tools to the design and licensing calculations of future GFRs.

This I-NERI project supported the ENIGMA program. The objectives of this research were to:

- 1) Improve analytical models for GFRs based on evaluation of experimental results
- Establish broader international participation in the ENIGMA program (including justification, definition, and design of experiments)

Research Progress

Following is a list of the tasks that researchers accomplished during previous years.

 Defined core configurations and measurements for the first phase of the ENIGMA project. The results showed that the significant material differences and neutronic characteristics between the GFR cores and typical sodium-cooled fast reactor cores justify the planned experimental campaign. They also showed the similarity between the planned MASURCA experiments and the GFR concepts and the potential contribution of these experiments in reducing uncertainties in core physics integral parameters. These results were reported in a paper entitled *Investigation* of the Similarity of Reactor Physics Experimental Configurations Planned in the CEA MASURCA Facility to Gas-Cooled Fast Reactor Concepts.

- Analyzed past experiments in the ANL ZPR-9 gascooled reactor configurations. The results support the need for additional experiments for the advanced GFRs.
- Outlined the planned ENIGMA experiments to facilitate the collaborative definition of the GFR physics measurements, which were done as part of this project.
- Completed a study of possible configurations for the pre-experimental phase of the ENIGMA project. The findings are documented in a joint report entitled *Impact of Spectral Transition Zone in Reference ENIGMA Configuration*. The results indicate that significant variations in the neutron flux can be obtained by the use of transition zones, with small perturbations in the core reactivity state. Using a graphite transition zone produces a significantly softer spectrum, while using the void transition zone produces a significantly harder spectrum compared to the reference case.

Over the past fiscal year, the researchers defined and evaluated central control rod experiments for the ENIGMA program. They performed various calculations, such as boron-carbide (B_4C) rod pattern studies, to establish optimal experimental configurations. Such experiments are useful for characterizing GFRs in the presence of control rods and for assessing the adequacy of existing fast reactor analysis tools for calculating control rod worths and power distribution distortions near absorbers. Researchers

determined the number of additional assemblies required to restore core criticality (or critical core radius) under various B_4C rodlet configurations and analyzed control rod worths for each configuration. Additionally, they presented flux distributions and reaction rate distributions for a few nuclides.

The various proposed experiments were summarized in a joint ANL-CEA report entitled *Control Rod Studies for ENIGMA Configurations*. The results showed the need for more assemblies at the core periphery as the number of absorber rodlets introduced into the core increases. The importance of spatial heterogeneity for the control rod experiments was also evident, indicating the flexibility available in achieving desired rod worths for the ENIGMA program.

CEA researchers loaded the ENIGMA reference core into the MASURCA facility and achieved criticality during 2006. This first critical core (Figure 1) was similar to that originally calculated by the CEA and ANL teams, although minor modifications were necessary. Safety related measurements conducted at the facility to date include: pilot rod reactivity worth, safety rod reactivity worth, differential/integral rod worths, reactivity worth of peripheral fuel cells, temperature coefficient in the range $20-35^{\circ}$ C, and delayed neutron fraction using the Feynman- α method and power calibration. Fission rate distribution and spectral indices measurements are planned.

Planned Activities

This I-NERI project is complete; however, much more work is still needed to define the ENIGMA experimental program in full detail, with inputs from measurement experts at both CEA and ANL. It is particularly important to establish what new materials and equipment will be necessary for subsequent phases of the ENIGMA program. Renovation of the MASURCA facility will continue, with current estimates that it will be available for experiments by early 2011.



Figure 1. ENIGMA core center with 32 absorber rodlets (pattern #2 with 8 absorber rods).

Evaluation of Materials for Gas-Cooled Fast Reactors

PI (U.S.): Todd R. Allen, University of Wisconsin

PI (France): Jean-Louis Seran, Commissariat à l'Energie Atomique (CEA)

Collaborators: University of Michigan, Pacific Northwest National Laboratory (PNNL)

Project Number: 2004-007-F

Project Start Date: August 2004

Project End Date: August 2007

Research Objectives

Both France and the United States have a shared interest in developing advanced reactor systems that employ inert gas as a coolant. Currently, there is an insufficient amount of physical property data to qualify candidate materials for gas-cooled fast reactor (GFR) designs. The goals of this project are to establish candidate metallic and ceramic materials for GFR designs and to evaluate their mechanical properties, dimensional stability, and corrosion resistance.

The first goal of this project is to improve hightemperature creep strength and resistance to environmental attack by optimizing grain boundary structural orientations, known as grain boundary engineering (GBE). Thermal-mechanical treatment is performed on GFR candidate alloys to maximize the fraction of low-energy boundaries. Following treatment, the changes to microstructure are characterized. Researchers will focus on Alloy 800H, which is an austenitic alloy designed for high-temperature boiler components; Alloy 617, which is a nickel-based austenitic alloy; and on T-91, which is a low-carbon (9Cr-MoVNb) ferritic-martensitic alloy designed for lower temperature boiler components.

The second goal of this project is to characterize the radiation resistance of candidate GFR metallic materials. Metallic materials have not typically been used for high-dose core components in GFR applications. Therefore, researchers will determine the radiation response of these alloys by examining the changes in the microstructure of samples that are irradiated with high-energy ions and, when available, neutrons from a test reactor. The focus is on Alloy 800H.

Research Progress

Grain boundary engineering (GBE) of Inconel Alloy 800H has been successfully performed by means of thermomechanical processing (TMP). The fraction of low- Σ coincidence site lattice boundaries (CSLBs) had been significantly increased and was stable at temperatures up to 600°C for 1,000 hours.

Similar methodology has been extended to optimize Incoloy Alloy 617. Researchers studied the effects of thickness reduction levels, annealing times, and TMP cycles on grain boundary character distribution (GBCD) of the material. The results indicate that five cycles of TMP with approximately 5 percent thickness reduction, followed by annealing at 1,100°C for 90 minutes, produced the highest fraction of low- Σ CSLBs and the lowest fraction of random boundaries.

The effect of cyclic TMP on the fraction of the low- Σ CSLBs components for the as-received and GBE-treated samples is shown in Figure 1. It is clear that the fractions of twins (Σ 3) and twin variants (Σ 9 and Σ 27) were greatly enhanced by TMP. This phenomenon is consistent with the universal GBCD feature of face-centered cubic polycrystalline materials. The sample treated with the 5-cycle TMP possesses the maximum fraction of Σ 3 boundaries (0.64), which is about 4.4 times that of the as-received sample and approaches the theoretical limit for the Σ 3 fraction (2/3).



Figure 1. Fraction of low- Σ CSLBs of the as-received (AR) and GBE-treated samples with 1 to 6 cycles of thermomechanical processing.



Figure 2. Thermal stability of the GBCD of as-received and GBE-treated samples with 2- and 5-cycle TMP (2^{nd} and 5^{th}) after exposure to air at 850°C and 1,000°C for 4 and 6 weeks.

The team evaluated the thermal stability of GBE-treated samples by annealing at 850° C and $1,000^{\circ}$ C for 4 and 6 weeks. Two GBE-treated samples with 2- and 5-cycle TMP were selected for the test due to their special GBCD characteristics, i.e., the highest fraction of $\Sigma1$ and low- Σ CSLBs. As-received samples were also tested at the same time for comparison.

Figure 2 shows the GBCD results of the annealed samples. Annealing slightly increases the fraction of random boundaries, but the annealing-induced GBCD variation is very small for samples with 5-cycle TMP compared to the other samples. This limitedtime annealing test indicates that the use of GBE-treated sample with a low fraction of $\Sigma 1$ and high fraction of low- Σ CSLBs is stable at temperatures up to 1,000°C. The preliminary thermal stability results provide some insights on the applicable conditions of GBE-treated materials, although thermal stability tests with much longer test times are needed.

In continuation of the work conducted earlier on the microstructural analysis of as-received, crept, and optimized alloys T91, additional TEM analysis was performed. The results are very similar to those obtained by SEM analysis done earlier. Table 1 gives the results of the TEM analysis. Results confirm that both carbide and subgrain growth are linked to the creep strain, and by reducing the creep rate the CSLE condition slows carbide and subgrain growth.

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the average subgrain size in the CSLE condition is smaller compared to the AR condition. There is also evidence that hardening due to longrange internal stresses is dependent on subgrain size, with smaller grains being stronger. Thus, both a higher volume fraction of subgrains and the presence of smaller

subgrains improve the creep strength in the

Sample	Carbide size M ₂₃ C ₆ (nm)	Subgrain size (µm)	Test Duration (hrs)	
AR	365±15	0.52±0.1	NA	
CSLE	360±17	0.50±0.1	NA	
*AR-550-165	560±13	1.20±0.1	2,177	
*CSLE-550-165	570±15	1.18±0.1	2,829	
AR-550-160	615±20	1.50±0.1	2,829	
CSLE-550-160	538±18	1.25±0.1	2,829	
* Test conducted to a strain of 2%.				

Table 1. Quantitative micorstructural parameters for T91 in different conditions.

Figures 3 and 4 show the TEM images of tempered lath and carbide sizes respectively for the samples CSLE-550-160 and AR-550-160. There is a 30 percent higher fraction of $\Sigma1$ boundaries with misorientation angle <3° in

dislocations in the vicinity of these boundaries, by X-ray line profiles and of HOLZ lines in convergent beam electron microscopy. Further

TEM analysis also confirmed that

CSLE condition by inducing long-range internal stresses which opposes the applied stress and thus reduces the effective stress for creep.





Figure 3. TEM images showing tempered lath sizes after creep test in a) CSLE-550-160 and b) AR-550-160.



Figure 4. TEM images showing carbide sizes after creep test in a) CSLE-550-160 and b) AR-550-160.

Planned Activities

Impact testing. The fracture toughness of the GBE-treated samples is expected to be improved due to the highly increased fraction of low-energy low- Σ CSLBs. Impact tests following ASTM E23-05 (standard test method for notched bar impact testing of metallic materials) will be performed to evaluate the effect of GBE on fracture toughness.

Cyclic oxidation testing. Samples will be heated to and held at specific temperatures for a period of time and then cooled to room temperature. Multiple heating-hold-cooling cycles will be performed to obtain weight change as a function of cycles, which represents thermal shock resistance of the oxide scale. Samples will be exposed to air and environment with controlled oxygen content.

Irradiation damage studies are complete under this I-NERI project, but samples of 800H being irradiated in the Advanced Test Reactor will be available for analysis at a future date.

Stress-dip tests of T91. Since both subgrain size and volume fraction affects creep behavior, it becomes important to incorporate both the subgrain size and volume fraction as variables in the creep rate equation. The general Norton's creep rate equation will be modified to incorporate these terms. Further stress-dip experiments have been planned to evaluate the internal stresses associated with the subgrain structure. Constant structure creep tests will also be conducted to obtain the dependence of creep rate on the size of subgrains.
Development of Generation IV Advanced Gas-Cooled Reactors with Hardened/Fast Neutron Spectrum

PI (U.S.): T.Y.C. Wei, Argonne National Laboratory

PI (France): J. Rouault, Commissariat à l'Energie Atomique (CEA) – Cadarache

Collaborators: Brookhaven National Laboratory, General Atomics, Massachusetts Institute of Technology, Idaho National Laboratory Project Number: 2004-008-F

Project Start Date: March 2005

Project End Date: February 2006

Research Objectives

The objective of this project was to design a gas fast reactor (GFR) that features a high level of safety, full recycling of actinides, high proliferation resistance, and economic attractiveness. This project was a continuation of a previous I-NERI project (2001-002-F) which evaluated possible GFR models, focusing in particular on a 2,400 MWt design. Researchers planned to develop a pre-conceptual GFR design based on the following parameters:

- **Fuel choice**. Dispersed fuel in plate sub-assemblies as the reference; SiC-clad pellets in pin sub-assemblies as a back-up. Design studies used a carbide actinide compound, but nitride remains a possible candidate
- **Unit size**. 2,400 MWt
- Power density. 100 MW/m³
- **Decay heat removal**. Natural convection passive approach combined with active low power circulators, in a mix to be refined. Alternative options (conduction paths, heavy gas injection, etc.) may also be considered
- **Balance-of-plant**. A direct Brayton cycle remains the reference design, but researchers are considering the indirect super-critical CO₂ cycle and other combined cycle alternatives using gas mixtures with an equivalent cycle efficiency

Work on the design and safety of this plant option proceeded in coordination with the multilateral Generation IV International Forum (GIF) collaboration plan, pending successful completion of the Project Arrangement. The goal was to develop core and system designs, safety rules and approach, safety systems, and transient analysis.



Figure 1. 2,400 MWt indirect cycle primary system.

Research Progress

The first year of the research work concluded in February 2006. With the reorientation of U.S efforts towards the sodium fast reactor (SFR) design, this project was not considered for continuation so no further joint work will be pursued.

Figures 1 and 2 summarize the concluding status of this work, showing the primary system layout for the 2,400 MWt indirect cycle and direct Brayton cycle options, respectively. In both cases, the vessel is metal with a hot plenum shroud, annular cross ducts, bottom entry control rods, and top refueling. Figure 2 shows a concrete guard containment with a steel liner; the metal guard containment alternative is not presented.

Planned Activities

This project ended in February 2006.



Figure 2. 2,400 MWt direct cycle plant layout.

Development of Fuels for the Gas-Cooled Fast Reactor

PI (U.S.): Mitch Meyer, Idaho National Laboratory (INL)

Project Number: 2004-009-F

Project Start Date: October 2004

Project End Date: September 2006

PI (France): N. Chauvin, Commissariat à l'Energie Atomique (CEA)

Collaborators: Oak Ridge National Laboratory, Purdue University

Research Objectives

The objective of this project is to develop silicon carbide matrix, uranium carbide dispersion fuel in two forms that are suitable for gas-cooled fast reactor (GFR) service: 1) a hexagonal block with coolant holes throughout and 2) a pin-type dispersion fuel with integral silicon carbide cladding. Because fuel operating parameters and physical requirements for the GFR are outside the current experimental nuclear fuel database, many basic viability issues will need to be experimentally addressed to demonstrate the feasibility of the proposed fuels.

Ion irradiation testing will be conducted to simulate material behavior at high irradiation doses. The GFR-F1 test in the Advanced Test Reactor (ATR) at INL and the FUTURIX-MI test in the French Phénix reactor address basic issues regarding the irradiation behavior of the "exotic" refractory materials required for GFR fuel service in a neutron-only environment. Ultimately, proof-of-concept for GFR fuel can only be demonstrated through irradiation testing of fissile-bearing specimens.

Research Progress

Current GFR fuels are based on a dispersion of uranium– and plutonium–carbide [(U,Pu)C] coated particles in a silicon carbide (SiC) matrix. The U.S. has two reference fuel forms: 1) large hexagonal blocks with coolant holes drilled throughout and 2) a refractory clad pin-type fuel. Both the block and the rod fuel types will be fabricated through reaction bonding. The starting material preforms for the two types of fuel are each fabricated in a slightly different manner.

The method of reaction bonding used for the blocktype fuel fabrication starts with a polymer-derived carbon preform. The polymer is produced with a specific amount of pore-forming agent that creates the appropriate porous microstructure. The polymer is then cured and pyrolyzed to produce a porous carbon preform that can be cast and/ or machined into shape. The preform is then infiltrated with molten silicon which reacts with the carbon to form SiC. The microstructure must be tailored to produce a fully infiltrated sample with minimal residual silicon (targeted at less than 5 volume-percent). The preform microstructure is dependent upon the amount of pore former used in the original polymer mixture. The disadvantage of this process for fuel fabrication is that the sample shrinks on the order of 50 percent during pyrolysis. If fuel spheres are incorporated into a polymer-derived preform (with no filler), the preform will crack due to shrinkage of the matrix around the stable fuel particles. To control this shrinkage, a filler must be added.

Previous results show an 80:20 ratio of filler powder to polymer is required to reduce the shrinkage below 5 percent. The filler materials dramatically affect the green, or pre-infiltrated, microstructure. Filler powder composition also affects the infiltrated microstructure. A series of parametric studies were conducted using 70, 80, and 90 percent β -SiC filler material, with the remainder carbon in the form of carbon black and graphite. After pyrolysis, the samples were weighed to determine mass loss and dimensionally analyzed to characterize volumetric shrinkage. The carbon formed during pyrolysis was calculated based on the initial polymer loading and mass loss. Based on known densities of each constituent,

Sample	Pressing Pressure (MPa)	Volume (cm³)	SiC (vol %)	Carbon (vol %)	Porosity (vol %)	Porosity/ Carbon	Infiltration Results	
90-1	17.2	0.45	45.0	15.7	39.3	2.50	Center portion not infiltrated	
90-2	34.4	0.40	49.5	17.2	33.2	1.92	Surface only	
90-3	51.7	0.41	51.2	17.8	31.0	1.74	Surface only	
80-1	17.2	0.38	38.3	22.6	39.1	1.74	Delamination, friable	
80-2	34.4	0.28	42.4	25.6	32.0	1.25	Large cracks	
80-3	51.7	0.33	43.9	26.1	30.0	1.15	Large cracks	
70-1	17.2	0.33	31.7	28.4	39.9	1.40	Delamination, friable	
70-2	34.4	0.26	35.5	31.7	32.8	1.04	Delamination, friable	
70-3	51.7	0.32	37.1	32.6	30.4	0.93	Cracks, slightly friable	
90-4	8.6	0.42	36.3	12.0	51.7	4.31	Friable	
90-5	17.2	0.35	44.6	15.1	40.4	2.68	Dense sample	

Table 1. Results from pyrolyzation and infiltration of pellets with varying filler SiC loadings and pressing pressures.

researchers calculated the volume fraction of each component in the microstructure, as listed in Table 1.

The samples containing 90 percent filler remained intact but did not fully infiltrate. In many cases, only the surface reacted to form a dense SiC outer layer. Samples with 80 percent SiC filler fully infiltrated, but many large cracks were formed that filled



Figure 1. Typical pellets after infiltration: (A) 90% SiC filler, (B) 80% SiC filler, and (C) 70% SiC filler.

with excess silicon. The 70 percent SiC filler samples also fully infiltrated; however, the resulting samples were friable and had no structural integrity. They often delaminated upon infiltration. Figure 1 shows typical pellets from each of the three groups after infiltration.

The researchers conducted further experimentation to refine the fabrication process, noting significant differences in final microstructure with changes in the pressure applied during pelletization. Table 1 lists the specific parameters used and resulting pellet volume fractions. Infiltration results varied from a fully infiltrated dense pellet (90-5) to a friable pellet with no structural integrity. Figure 2 shows the microstructure of the fully dense, infiltrated pellet. As can be seen, the pellet is dense with less than 5 percent porosity and very little residual silicon.

During reaction bonding, carbon reacts with silicon to form SiC, resulting in a volume expansion of 2.36 times relative to the initial carbon substrate. Because of this, the porosity-to-carbon ratio must exceed 2.36 to ensure adequate space for volume expansion during the formation of SiC. Samples that remained intact while exhibiting appreciable infiltration had porosity-to-carbon ratios of 2.5



Figure 2. Resulting SiC microstructure of a pellet with 90% SiC filler material and a porosity-to-carbon ratio of 2.68 (90-5).

and 2.6. Lower ratios cracked during infiltration. Excessive porosity leads to a friable sample with little or no structural integrity. If adequate silicon is present, it will fill the pores, increasing the amount of free silicon to unacceptably high levels.

Pore size and morphology were also found to be important. As transport of silicon relies on a network of interconnected porosity, infiltration does not take place if the pores are too small or disconnected. In the case of small pore size, silicon will start infiltrating at the preform surface, but the channels quickly close due to the formation of SiC, leading to a SiC-coated preform with no reactionformed SiC in the interior. The high particle volume loading required for the GFR will also undoubtedly reduce the transport paths for silicon. A process must, therefore, be developed to ensure evenly spaced particles completely surrounded by the SiC precursor materials, with the proper pore size to allow transport of molten silicon throughout the sample. Further process development is required to produce fully infiltrated dense samples with a high fuel loading using this technique.

Research targeted at developing pin-type fuel with integral SiC cladding was not conducted in FY 2006 due to funding limitations. The only carbon source used in this fabrication process is carbon powder added to SiC powder. The processing route chosen was based on wet mixing of SiC and graphite powders. Researchers found that a-SiC (70 percent -100+200 mesh and 30 percent -325 mesh) and graphite (-325 mesh) in a 75:25 ratio, mixed with 25 weight-percent glycol and pressed at 9,000-18,000 psi and 1,550°C, yields well-infiltrated samples of matrix material, as shown in Figure 3. Adding spheres to the matrix,



Figure 3. Microstructure of reaction bonded SiC using the proper ratio of particles sizes and compositions.

however, resulted in severe cracking of the infiltrated samples, indicating that further optimization is required in order to incorporate fuel particles into the matrix.

In conjunction with the fuel fabrication activities, researchers conducted several material irradiation experiments, including heavy ion irradiation studies of ceramics (ZrN, TiC, TiN, ZrC, and SiC). All the irradiations were conducted at a temperature of 800°C and doses of 10 and 70 dpa with 1 MeV krypton ions using the IVEM-TANDEM facility at Argonne National Laboratory. Irradiation performance of ZrC and ZrN under these conditions was poor, with the materials showing severe lattice expansion. The performance of TiC and TiN was much improved, however, with lattice expansion reduced by a factor of four. The SiC samples exhibited virtually no lattice expansion.

The GFR-F1 low-dose material experiment irradiated commercially procured SiC, TiC, ZrC, AlN, TiN, and ZrN in the Advanced Test Reactor at INL. The experiment was not instrumented and researchers maintained a calculated specimen temperature of approximately 1,000°C by varying heat transfer from the experiment capsules with differing fill gas compositions. The first experimental capsules are currently awaiting post irradiation examination.

Planned Activities

The FUTURIX-MI irradiation experiment will be inserted into the Phénix reactor in France in 2007. Initial planning began for the GFR-F2 fuel irradiation test in the ATR. However, U.S. funding re-allocations due to the Global Nuclear Energy program and its focus on the sodiumcooled fast reactor will not allow this work to continue. Due to changing funding priorities, no further work is planned in the area of GFR fuel development. I-NERI — 2006 Annual Report

PRA-Aided Design of Advanced Reactors with an Application to GFR Safety-Related Systems

PI (U.S.): G. Apostolakis, Massachusetts Institute of Technology (MIT)	Project Number: 2004-010-F
PI (France): N. Devictor, Commissariat à	Project Start: August 2004
l'Energie Atomique (CEA)	Project End: August 2007

Collaborators: None

Research Objectives

The objective of this project is to develop a conceptual design of a decay heat removal system for the gas-cooled fast reactor (GFR). This system will function during both normal modes of operation (including shutdown and refueling) and accident conditions (such as following a loss-of-coolant accident [LOCA]). Researchers will evaluate the system under a range of scenarios, including station blackout and anticipated transients without scram (ATWS).

If GFRs are to meet the high expectations for safety assurance established for new reactor designs, then they must employ a reliable decay heat removal system. Probabilistic risk assessment (PRA), which has matured over the last 30 years, is expected to play a key role in all aspects of system design and safety. PRA will allow the designers to build on the vast array of applications already developed for light water reactors and other reactor types.

Two major issues must be addressed to take full advantage of PRA capabilities for advanced reactors:

- PRA models for passive systems will have to be developed.
- The use of PRA in design implies that probabilistic goals exist to determine the acceptability of a design. However, the current licensing framework is largely deterministic, which is not expected to change substantially in the near future.

The U.S. Nuclear Regulatory Commission (NRC) and International Atomic Energy Agency (IAEA) are working to establish probabilistic goals. This raises the issue of whether the design must satisfy both sets of criteria, particularly if a design option meets the probabilistic goals but fails the deterministic criteria.

Research Progress

The reliability assessment of passive systems focuses on the thermal-hydraulic aspects, i.e., a subset within IAEA passive systems classification that is characterized by moving working fluids, either with or without moving mechanical parts such as check or relief valves. There are several reasons for this emphasis. The most important involve the uncertainties surrounding model predictions and system performance and the variety of possible failure mechanisms that may affect these systems.

Since system failure is no longer characterized by the failure of an active component, a new failure description is required. The concept of "passive function failure," introduced by Burgazzi in 2003, describes failure in terms of a load exceeding the capacity of the system at a certain location. The load is represented as a time-dependent vector that contains a set of performance parameters describing the system safety status. Failure occurs whenever one or more of these parameters exceeds the corresponding value on the capacity vector. Two models are needed: one to predict the system performance and another to describe its capacity.

This project will develop capacity descriptions and the corresponding reliability calculation schemes when no formal models exist. The capacity vector and its time behavior will be specified by aggregating expert opinions. Since the capacity of the system is a function of its state, the capacity changes as the state changes. Introducing a value for time greatly affects the reliability calculation, which is a serious issue for transient calculations. In addition, guidelines are required for choosing different failure representations when facing different situations; that is, different capacities need different treatments, given the nature of their intent. For example, whether the analysis is done on a regulatory framework affects the form of the reliability assessment and the corresponding description of the capacity. Three descriptions are proposed: 1) deterministic, 2) epistemic, and 3) aleatory capacities.

Researchers have developed a RELAP5-3D[®] model of a fully passive, redundant, two-loop decay heat removal system attached to a 600 MW helium-cooled fast reactor. They are using this as the system performance model to demonstrate the applicability of the reliability calculations. The researchers are examining propagation of uncertainties through the model using Latin Hypercube Sampling of over seven independent parameters. The work captures system dynamic behavior during the transient following a LOCA to verify that the performance criteria are satisfied. The researchers have gained insight into features of the system dynamics under different operational conditions that were useful in development of the model. They can also offer a perspective on the future steps to improve the description of passive system failure mechanisms.

Researchers have analyzed the passive cooling of a helium-cooled fast reactor in quasi-steady state post-LOCA conditions. They used an importance sampling Monte Carlo technique to propagate the epistemic uncertainties and to calculate the probabilities of function failures for comparison with an active design. Previous work showed that active systems are more reliable than passive for two- and three-loop designs, while the passive design was more reliable in the four-loop configuration. By employing larger uncertainties in some physical parameters, derived from more recent correlation formulas, and by modifying the previous results, the researchers showed that the failure probability of the active four-loop configuration is lower than for the passive design. These results need to be coupled with those from the transient study to give a complete view of the reliability of the passive decay heat removal system design.

Planned Activities

Researchers will continue to finalize the analysis of time-dependent behavior of a passive decay heat removal system during LOCA transients in a helium-cooled GFR. Time-dependent situations are complex and their inclusion in PRA models is challenging. By analyzing transient failure modes, researchers will obtain a more precise picture of the decay heat removal system based on passive phenomena. This is a necessary step before including passive thermal-hydraulic systems in the PRA model.

The researchers have evaluated the similarities and differences of their PRA methodologies. They have found that methodologies are similar, but lack an explicit treatment of transients; this will constitute a major task for the next period.

Finally, the team will study specific decay-heat removal systems, including the behavior of passive and active systems during LOCA transients, to develop probabilistic criteria for determining the acceptability of a design.

Thermochemical Hydrogen Production Process Analysis

PI (U.S.): M. Lewis, Argonne National Laboratory	Project Number: 2004-011-F
PI (France): P. Carles, P. Anziew, and J.M. Borgard, Commissariat à l'Energie Atomique	Project Start: October 2004

Project End: September 2007

Collaborators: None

(CEA)

Research Objectives

There are two objectives of this I-NERI project: 1) to develop a consistent methodology for evaluating the potential of a given thermochemical cycle to produce hydrogen with the use of nuclear heat, and 2) to use this methodology to identify the most promising hydrogen cycles among the 200+ proposed in the literature. The metrics described within the methodology include chemical viability, energy efficiency, and engineering feasibility. In this way, all of the cycles will be compared on a consistent basis.

Research Progress

The researchers have identified alternative thermochemical cycles and are developing a consistent evaluation methodology based on the CEA concept of a cycle's efficiency changing as the level of knowledge increases. analysis requires the same heat and work inputs, but also includes equilibrium data to identify unrealistic process conditions, problematic separations, unexpected byproducts, etc. Researchers may adjust reaction conditions from values in the literature in order to maximize yields, minimize the formation of competing product, and reduce recycle. Available kinetic data can adjust reaction conditions to provide reasonably fast reaction rates. Using the same methodology allows cycles to be compared on a consistent basis and focuses process development efforts. The scoping flowsheet methodology defined components of the first two levels.

The researchers evaluated several promising alternative cycles, including the copper-sulfate (Cu-SO₄), zinc-sulfate (Zn-SO₄), magnesium-chloride (Mg-Cl) and copper-chloride (Cu-Cl) cycles. Table 1 shows the calculated efficiencies. The data for the Cu-SO₄ cycle show a wide spread in calculated values, indicating a lack of optimization. More

The methodology consists of two levels: Level 1 rapidly screens previously identified thermochemical cycles with reasonable realism, and Level 2 identifies problems and unrealistic conditions. The screening only considers heat and work inputs and assumes stoichiometric reactions, resulting in the maximum theoretical efficiency. The Level 2

Cycle	Level	Efficiency % (LHV)	Maximum Temperature, °C	Other Conditions
	1	46.0	850	1 mol water
Cu-SO ₄	2	38.1	1,100	1 mol water
	2	30.7	Maximum Temperature, °C 850 1 1,100 1 1,200 1 550 1 550 1 1,200 1 550 1 1,200 1 1,200 1 1,200 1 1,200 1 1,200 1 1,200 1 1,200 1 1,200 1 1,200 1 1,200 1 1,200 1 1,400 2 600 1 600 2	10 mols water
	1	45.0	550	1 mol water
Cu-Cl	2	43.9 From Aspen-Plus®	550	Excess water and HCI
7= 00	1	40.5	850	1 mol water
211-504	2	40.8	1,400	2.7 mols water
	1	35.2	600	1 mol water
ivig-Cl	2	30.0–33.1	850 1 mol w 1,100 1 mol w 1,200 10 mols 550 1 mol w 550 1 mol w 550 1 mol w 550 1 mol w 550 2 mol w 1,400 2.7 mol w 600 1 mol w 600 2 mol w	2 mols water

Table 1. Efficiency calculations for the alternative cycles evaluated with the scoping flowsheet methodology.

accurate efficiency calculations for the Cu-Cl cycle will require new thermodynamic measurements, as well as further optimization. The preliminary study of the $Zn-SO_4$ cycle indicates that required reaction temperatures are too high for a nuclear heat source, while the efficiency of the Mg-Cl cycle is relatively low. Unless further compelling information is obtained, the researchers no longer consider these two cycles viable. They are currently calculating the efficiency of a fifth cycle, cerium-chloride (Ce-Cl).

Researchers have nearly completed proof-of-principle work for the Cu-Cl, Cu-SO₄, and the Ce-Cl cycles. The results of laboratory analyses indicate that these cycles are chemically viable. Since they also have acceptable efficiencies, further examination is warranted.

Researchers have identified the key parameters that influence the cost of hydrogen production by such hightemperature processes as thermochemical cycles and electrolysis. For thermochemical cycles, raw material investment, maintenance, heat exchanger costs, and energy recovery are the important parameters. For hightemperature electrolysis, the important parameters are the heat exchange and investment cost for the electrolyzer, as well as the assumed electrolyzer lifetime and its ability to handle shutdowns and recycle events. Researchers are quantifying general screening criteria for potential thermochemical cycles as part of this effort.

Planned Activities

In their original plans, the research team specified that they would evaluate flowsheets for the baseline cycles, including one or more sulfur cycles and possibly high-temperature electrolysis. They have deferred this work until after developing Level 3 analysis methodology, which consists of a detailed engineering feasibility study of the most promising alternative cycles. This analysis will include values for common equipment parameters, such as the efficiencies of pumps, compressors, and furnaces. In addition, researchers will fix boundary conditions, such as hydrogen outlet pressure and purity, so that all cycles may be evaluated on a consistent basis. Some challenges associated with engineering feasibility include developing energy-efficient methods for separations and the removal of excess water, maintaining high-temperature operations under aggressive, oxidizing conditions, identifying corrosion-resistant materials for use at high temperatures and moderate pressures, and developing new technologies for difficult separations. The Level 3 evaluation will consider technical options to meet these challenges. Various U.S. universities will partner in this effort.

Sulfur-Iodine Integrated Laboratory-Scale Experiment

PI (U.S.): Paul Pickard, Sandia National Laboratories

Project Number: 2006-001-F

Project Start Date: April 2006

PI (France): Philippe Carles, Commissariat à l'Energie Atomique (CEA)

Project End Date: April 2009

Collaborators: General Atomics

Abstract

The Nuclear Hydrogen Initiative (NHI) is investigating high-temperature thermochemical cycles for the production of hydrogen with nuclear energy. Thermochemical cycles use a series of chemical reactions to generate hydrogen and oxygen from water at lower temperatures than direct thermal dissociation. The sulfur-iodide (S-I) cycle is one of the leading candidates due to its technical maturity and its potential high efficiency.

The objective of this project is to evaluate the key technology issues associated with using the S-I cycle to produce hydrogen. Researchers will build on an earlier I-NERI project (2002-001-F) that developed the process chemistry and materials technology for each of the three major S-I reaction sections and investigated their operation as stand-alone units.

For this research project, the team will integrate the component reaction sections at a single location to perform a series of closed-loop laboratory-scale experiments. Results of the integrated laboratory-scale experiments will establish the technical basis to assess the viability of the S-I cycle for nuclear hydrogen production. In addition, the team will develop control systems for closed-loop operations, select interface components between sections, and create the safety documentation necessary to conduct the integrated laboratory-scale experiments.

Developing and demonstrating the S-I cycle will include the following sequence of activities:

- 1) Develop, construct, and test the individual laboratoryscale sections
- 2) Ship sections to the experiment site and assemble as an integrated, closed-loop system
- 3) Conduct integrated, laboratory-scale S-I experiments and analyze the results

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High-Temperature Nickel-Based Alloys for VHTR Applications: Mechanical and Corrosion Testing

PI (U.S.): Richard Wright, Idaho National Laboratory

Project Number: 2006-002-F

Project Start Date: January 2006

Project End Date: December 2008

PI (France): P. Billot, Commissariat à l'Energie Atomique (CEA)

Collaborators: Oak Ridge National Laboratory, Electricité de France, Framatome

Abstract

The very high-temperature reactor (VHTR) is a helium-cooled reactor operating with outlet temperatures exceeding 950°C. There are two main advantages of this concept—high yields for energy generation and the ability to supply high-temperature process heat for hydrogen production. However, the high temperatures impose challenging design requirements on structural materials, particularly for the intermediate heat exchangers.

Nickel-based alloys are the most suitable materials for high temperatures, particularly Inconel Alloy 617 and Haynes 230. However, these alloys are not fully ASME code-qualified for nuclear applications. Basic data are needed to achieve a complete understanding of their behavior at high temperatures. Researchers must investigate the mechanical properties of these alloys and study surface/subsurface corrosion effects caused by helium impurities. As component integrity must be demonstrated over the entire operational lifetime (on the order of 100,000 hours), the evolution of properties over time must be taken into account, specifically the effects of thermal aging. In this work, researchers will conduct experimental thermal aging studies of Alloy 617 and Haynes 230 through short-duration (500 hours) and medium-duration (5,000 hours) exposure tests of material coupons and specimens. Through these experiments, they will evaluate the microstructural evolution under thermal aging conditions and determine the effect of thermal aging on Charpy and tensile test data. They will use the results of these studies to develop a model capable of extrapolating the effects of thermal aging on materials properties over a very long duration in order to predict component lifetimes. Finally, they will validate the model by conducting long-exposure testing on material specimens to 15,000 hours. I-NERI — 2006 Annual Report

9.0 U.S./Japan Collaboration

An exchange of notes was signed by U.S. Assistant Secretary John Wolf, for Secretary of State Colin Powell, and Mr. Keiichi Katakami, Minister of the Japanese Embassy, for Ambassador Kato, on April 22, 2004.

To implement this bilateral collaboration, the Japanese government decided to sign implementing arrangements using the following two organizations: the Agency of Natural Resources and Energy of Japan (ANRE) and the Ministry of Education, Culture, Sports, Science, and Technology of Japan (MEXT). ANRE is the office responsible for nuclear technology for the Ministry of Economy, Trade and Industry (METI).

On May 26, 2004, the implementing arrangement with ANRE was finalized and signed by Mr. William D. Magwood IV, then Director of DOE-NE, and Mr. Kusaka, Director-General of ANRE. An annex regarding the I-NERI collaboration was signed on June 10, 2004, by Mr. Shane Johnson, Deputy Director for Technology, DOE-NE, and Mr. Shigeru Maeda, Director for Nuclear Energy Policy, Nuclear Policy Division, ANRE/METI.

On February 8, 2005, the United States Department of Energy (DOE) and Japan's MEXT signed the Implementing Arrangement concerning cooperation in the field of research and development of innovative nuclear energy technologies (I-NERI).

9.1 Work Scope Areas

Areas of Collaboration with ANRE

Supercritical Water-Cooled Reactor

Areas of Collaboration with MEXT

- Innovative nuclear reactor technologies
- Innovative processing technologies; and
- Innovative fuel technologies using solvent extraction
 - 9.2 Project Summaries

The first project was awarded under this cooperative agreement in FY 2005, with the second awarded early 2006. A listing of the I-NERI U.S./Japan projects that are currently underway follows, along with summaries of the accomplishments achieved in FY 2006.

Directory of Project Summaries

2005-001-J	Development of Materials for Supercritical-Water-Cooled Reactor	119
2006-001-J	Preliminary Assessment of Irradiation Performance of Zirconium Carbide (ZrC) TRISO Fuel Particles	125

Development of Materials for Supercritical-Water-Cooled Reactor

PI (U.S.): Gary Was, University of Michigan

Project Number: 2005-001-J

PI (Japan): Hideki Matsui, Tohoku University

Collaborators: Idaho National Laboratory, University of Wisconsin-Madison, University of Tokyo, Toshiba Corporation, Hitachi, Ltd. Project Start Date: October 2004

Project End Date: September 2007

Research Objectives

The objective of this project is to evaluate the irradiation durability and corrosion performance of select materials that have been proposed for use in the supercritical water-cooled reactor (SCWR). Researchers will analyze the materials' resistance to stress corrosion cracking and conduct neutron irradiation tests under simulated SCWR conditions.

Material development is a critical issue for SCWR development. Previous studies in this area have been limited to screening commercial alloys and potential new materials via simulated irradiation tests and un-irradiated corrosion testing. To develop a detailed SCWR system design, researchers need a more thorough understanding of material behavior under specific SCWR conditions and a comprehensive materials database.

In this work, researchers will subject selected commercial alloys and newly developed alloys to microanalysis and corrosion tests in supercritical water. They will evaluate long-term reliability via phase stability tests and long-duration corrosion tests. Through these experimental studies, they will gain sufficient understanding of the material's behavior under SCWR conditions. This knowledge will enable them to construct a materials database which is needed for designing fuel cladding and reactor core components.

Research Progress

During the past fiscal year, researchers received irradiated samples, conducted initial constant extension rate tensile tests (CERT) in deaerated supercritical water (SCW) at 400°C, and completed the benchmarking experiment for crack growth rate testing in SCW. They selected 21 tensile samples of alloy JPCA for testing in supercritical water. These samples were irradiated in the Fast Flux Test Facility (FFTF) at temperatures of 390–520°C and doses of 26.9–43.9 dpa. Four samples irradiated at 407 or 427°C and tested in 400°C deaerated SCW exhibited intergranular stress corrosion cracking, low strain to failure, and low reduction in area. From existing data on the tensile behavior of this alloy tested in room temperature air, the SCW environment appears to have resulted in significant embrittlement. Although one sample failed in the grip area, its fracture surface was completely intergranular. Figure 1 shows representative fracture images of the cold-worked samples following the CERT test. Table 1 summarizes the results of these experiments.

All specimens exhibited intergranular cracking. K]K9 failed at a pinhole, but no other samples showed evidence of deformation or cracking in this area. Samples irradiated at 407°C to 41.1 dpa exhibited less elongation to rupture, less reduction of area, and more intergranular cracking than those irradiated at 427°C to 43.9 dpa. Cold working had a negative effect on irradiation-assisted stress corrosion cracking (IASCC), as these samples exhibited more intergranular cracking than solution-annealed samples.

Researchers conducted a crack growth rate test on a compact tension specimen of type 316 stainless steel in pure water under subcritical and supercritical conditions. In the supercritical regime, the crack growth rate decreased with increasing temperature and the effect of increased dissolved oxygen reduced the rate compared to the deaerated case. Researchers analyzed the fracture surface using a scanning electron microscope (SEM), which revealed intergranular stress corrosion cracking (SCC) in both subcritical and supercritical water. Comparison of



Figure 1. Fracture surface of sample K]K9 (CW-407°C/41.1 dpa) after CERT experiment in 400°C deaerated SCW, showing completely intergranular cracking, and detail showing crack branching.

Sample	Irradiation condition**	Yield stress (MPa)	Max stress (MPa)	Strain to failure total/plastic (MPa)	Reduction in area (%)	Intergranular Cracking (%)
K[L0	SA-407°C/41.1 dpa	760	766	5.25/1.5	6	20
K]K9*	CW-407°C/41.1 dpa	850	850	5.70/1.5	NA	100
K[L2	SA-427°C/43.9 dpa	762	785	6.75/2.2	7	7
K]L2	CW-427°C/43.9 dpa	800	844	7.25/3.0	11	13.7

Table 1. Summary of CERT experiments in deaerated SCW at 400°C.

* sample failed in grips ** SA=solution annealed, CW=cold worked



Figure 2. Crack growth rate vs. temperature across the subcriticalsupercritical line for 0.5T CT unsensitized type 316L stainless steel in pure water.

crack length measurements showed that the direct current potential drop (DCPD) method was within 30 percent of SEM measurements at the fracture surface. Crack blunting by rapid oxidation in the supercritical regime during a crack growth rate (CGR) test, as opposed to film rupture by the higher strain rate of a CERT test, may explain the apparent contradiction of the temperature dependence of cracking in CGR vs. CERT tests. Figure 2 shows the crack growth rate, oxidation rate, and water density as a function of temperature.

Researchers continued corrosion testing of candidate alloys, including ferritic/martensitic (F/M) steels, austenitic steels, and nickel-based alloys. They applied surface modifications and grain boundary engineering treatments on selected materials and investigated the corrosion response. Figure 3 compares the weight gain for materials tested over a range of temperatures. Weight gain due to oxidation in SCW is typically smaller but less predictable in austenitic stainless steels than in F/M steels. Nickelbased alloys show a fairly good corrosion resistance in an SCW environment and have the lowest weight gain of all the tested materials. A 9Cr oxide dispersion strengthened (ODS) sample showed the lowest weight gain among the F/M steels.



Figure 3. Comparison of weight gain data for the candidate materials after exposure to subcritical/supercritical water with different oxygen concentrations for \sim 333 hours.

Figure 4 illustrates the significant oxidation at the grain boundaries in the internal oxidation zone of the ODS steel. This is believed to change the mechanism of the outward growth of the oxide scale. Furthermore, yttrium segregated to the grain boundary area in the internal oxidation layer may retard the diffusion of cations and improve the corrosion properties in SCW. For a sample surface modified by adding a layer of yttrium (Y), the cross sectional image (Figure 5) shows an Y-rich Y-Fe-O oxide layer that separates the magnetite into two layers with different microstructures. The Y-Fe-O layer significantly improved corrosion resistance.

Austenitic alloy 800H showed low weight gain in supercritical water; however, it also suffered spallation of its outer magnetite scale. One approach to solve the spallation issue is grain boundary engineering (GBE) treatment. As shown in Figure 6, the scale on the GBEtreated alloy 800H became compact and continuous compared to the control sample. Optimizing the grain boundary character distribution of the base metal led to improved oxide stability.



Figure 4. TEM image showing grain morphology and boundaries in the internal oxidation layer formed in 9Cr ODS steel after 1,026 hrs exposure to 500°C SCW.



Figure 5. SEM image showing the microstructure of the magnetite layer formed in 9Cr ODS steel coated with a thin yttrium film after exposure to SCW at 500°C for 667 hours.



Figure 6. Effect of GBE treatment on the corrosion resistance of the austenitic alloy 800H after exposure to SCW at 500°C.

Japanese efforts focused on creep, long-term corrosion, and SCC testing of candidate materials. The research efforts in each of these areas are described below.

Creep test preparation. Detailed examination of design requirements has lead to an expanded creep test matrix, including pressurized tube creep (PTC) tests in Japan's JOYO reactor. Among the alloys in the matrix, SUS316 and SUS310 can be fabricated using standard methods, but alloy 690 requires an optimized process. Researchers fabricated helium-filled PTC specimens and loaded them into JOYO for irradiation. **Long-term corrosion tests.** Corrosion tests with de-aerated water on SUS316 and zirconium (Zr)-modified steel show linear weight gain to 2,000 hours; the maximum weight gain of approximately 300 mg/dm² was observed in SUS316L. Ni-based alloys and SUS310 and its derivatives show much lower weight gain and tend to saturate after 500 hours at levels about 5 mg/dm², which implies dissolution is occurring along with oxidation. Figure 7 shows a schematic of the general corrosion test apparatus for radioactive samples being installed in the hotlab facility of Tohoku University. Hot runs using irradiated specimens will be performed in FY 2007.

Figure 7. SCRW loop for corrosion test of irradiated specimens.

Alloy	Sub-critical pure water	SC pure water	
Sensitized stainless steel	GB SCC	No SCC sensitivity Micro-cracks	
Non-sensitized stainless steel	No SCC sensitivity	Micro-cracks	
Ferritic stainless steel	No SCC sensitivity	No SCC sensitivity	
Ni-based alloys	No SCC sensitivity	Micro-cracks	
INCONEL 625		GB fracture	
Ti alloys	No SCC sensitivity	TG fracture	

Table 2. SCC test results.

SCC behavior. Results of SCC tests in subcritical and supercritical water are summarized in Table 2. Although only alloy 625 and titanium alloys exhibit SCC sensitivity, micro-cracks are frequently observed close to the fracture point. Tests conducted in a vacuum yielded similar micro-crack, implying that SCW is not responsible for their formation. Further study will be conducted to examine the mechanism of micro-crack formation and to assess their significance.

Helium embrittlement. In thermal reactors, helium generation in nickel-bearing alloys may be an issue since they are susceptible to helium embrittlement. Researchers calculated 100 to 400 appm helium generation for the design lifetime of the cladding material, depending on alloy type, with displacement damage of 15 dpa. Specimens implanted with helium to a depth of 1–3 μ m at 100 appm were tensile tested in vacuum at 700°C and the fracture surface examined by SEM. Some alloys exhibited an intergranular fracture surface, while others showed ductile failure. Since intergranular fracture was often observed throughout the sample cross-section, it is likely that helium diffused out of the implanted region toward the back surface. Detailed analysis is underway.

Planned Activities

Researchers will continue CERT tests on JPCA alloy samples irradiated in the FFTF in order to determine the effect of dose, irradiation temperature and water chemistry on stress corrosion cracking in supercritical water. They will also conduct experiments to determine the crack growth rate behavior of annealed 316L and alloy 690 in supercritical water as a function of temperature. Corrosion studies will concentrate on surface coatings, the effect of radiation on corrosion, and long-term performance.

The most important tasks in FY 2007 will be post irradiation examination (PIE) of samples irradiated in the JOYO reactor. The PIE will include general corrosion tests, mechanical tests, TEM, and measurements of pressurized tube specimens. At the conclusion of this project, researchers will identify suitable materials for SCWR applications. I-NERI — 2006 Annual Report

Assessment of Irradiation Performance of Zirconium Carbide (ZrC) TRISO Fuel Particles

PI (U.S.): Yutai Katoh and Lance Snead, Oak Ridge National Laboratory (ORNL)

Project Number: 2006-001-J

Project Start Date: March 2006

PI (Japan): Kazuhiro Sawa, Japan Atomic Energy Agency (JAEA) Project End Date: February 2009

Agency (JAEA)

Collaborators: Idaho National Laboratory

Research Objectives

Zirconium carbide (ZrC) is a leading candidate to replace silicon carbide (SiC) as a coating material for TRISO fuel particles. However, researchers must first determine the fundamental radiation effects of ZrC in order to assess its viability as a fuel coating. The objectives of this project are to: 1) clarify the fundamental irradiation response of ZrC, both in the form of a coating on surrogate TRISO particles and in a zone-refined, high-purity, polycrystalline form; 2) evaluate statistical fracture strength properties of developmental ZrC coatings before and after irradiation; and 3) develop a failure model of ZrC TRISO particles for fuel performance evaluation and safety design.

This project consists of the following tasks:

- 1) Conduct post-irradiation examination (PIE) of zonerefined, high-purity ZrC samples to understand baseline irradiation effects
- 2) Characterize mechanical properties of unirradiated developmental ZrC coatings
- 3) Perform high-temperature neutron irradiation of ZrCcoated surrogate fuel particles and conduct PIE
- 4) Study thermomechanical response of ZrC-coated particles using the PARFUME model
- 5) Examine chemical interactions between ZrC and key fission products to identify potential degradation
- 6) Irradiate fueled UO₂-ZrC TRISO-coated particles

Research Progress

Over the past fiscal year, researchers completed neutron irradiation of zone-refined, high-purity ZrC samples in the High Flux Isotope Reactor (HFIR) at ORNL. They successfully disassembled the irradiation capsules and subjected the first set of specimens to strain measurement to generate irradiation creep data. They used the bend stress relaxation (BSR) method to evaluate ZrC irradiation creep. Figure 1 provides a view of the BSR irradiation creep assembly and an example of a crept ZrC specimen. Assuming a linear stress dependence of the irradiation creep rate, the irradiation creep compliance of ZrC above 1,030°C appears reasonably constant at approximately 0.8x10⁻⁶ (MPa-dpa)⁻¹, which is comparable with SiC. However, at 700°C, the irradiation creep strain of ZrC is approximately an order of magnitude greater than SiC. Further investigation is necessary to determine the cause of this very substantial irradiation creep in ZrC at relatively low temperatures. However, researchers are encouraged by the excellent irradiation creep resistance at higher temperatures.

Figure 1. Bend stress relaxation holder assembly for irradiation creep study of refractory ceramics (upper), and ZrC sample after creep deformation to 3.7 dpa at 640° C.

Use of a bromide process optimized the commercialscale coating of ZrC surrogate particles. This process involves depositing a ZrC layer through a pyrolytic reaction of zirconium bromide (ZrBr₄), methane (CH₄), and hydrogen in a spouted-bed coater, as shown in Figure 2. In the first year, researchers modified the coating system for improved safety and temperature control and conducted a parametric survey to establish a correlation between the deposition condition and coating quality. After several iterations of deposition and analysis, they achieved a homogeneous ZrC coating layer with good stoichiometry (illustrated in Figure 3) and subsequently produced six batches of surrogate particles for pre-irradiation characterization and irradiation testing.

Figure 2. Coating equipment for ZrC-TRISO fuel particles.

The team has also started developing novel inspection methods to determine coating properties such as density, thickness, C:Zr ratio, and ZrC defect fraction. Gas pycnometry appears to be a feasible technique for conducting density measurements of coated layers. Combined application of inductively coupled plasma (ICP) mass spectrometry and oxidation methods also appear promising for determining the C:Zr ratio.

In order to accurately characterize ZrC coatings, it is essential to develop a technique to remove the pyrolytic carbon (PyC) layers. However, the highly oxidative reactivity of ZrC may prevent burning the PyC, as is common for SiC-TRISO systems. Researchers attempted several removal methods during this reporting period. Both mechanical sand blasting and plasma etching techniques appear promising. Chemical techniques, such as alkali activation, reduction by hydrogen, and etching with fluorine compounds, exhibited poor results.

Planned Activities

Researchers plan to accomplish the following activities during the next fiscal year:

- Continue PIE of zone-refined ZrC samples, while designing a new vehicle for irradiation of surrogate ZrC TRISO fuel particles at HFIR
- Develop parametric coating tests for further quality improvement, larger batch sizes, and production of a continuous ZrC-OPyC coating
- Perform non-irradiated and irradiated characterization of the developmental coatings
- Conduct modeling and code development for fuel performance evaluation
- Study fission products/ZrC chemical interactions
- Possibly conduct an irradiation study of ZrC-TRISO fuel particles

Figure 3. Optical micrographs of the developmental IPyC and ZrC coatings on surrogate zirconia kernel.

10.0 U.S./Republic of Korea Collaboration

Director of DOE-NE, William D. Magwood IV, signed the first bilateral I-NERI Agreement on May 16, 2001, with Dr. Chung-Won Cho, Director General of Korea's Atomic Energy Bureau, signing for the Republic of Korea's Ministry of Science and Technology. The first U.S./Republic of Korea collaborative research projects were awarded in FY 2002, with a total of 21 projects awarded to date.

10.1 Work Scope Areas

R&D topical areas for the U.S./Republic of Korea collaboration include:

2002 projects:

- Instrumentation, controls, and diagnostics
- Advanced light water reactors (LWR)
- Advanced LWR fuels and materials technology
- LWR safety technology
- Advanced LWR Computational methods 2003 projects:
- Next generation reactor/fuel cycle technology
- Innovative nuclear plant design
- Advanced nuclear fuels and materials

2004 projects:

- Advanced gas-cooled fast reactor
- Hydrogen production by nuclear systems
- Advanced fuels and materials development
- Supercritical water-cooled reactor concepts

2005-6 projects:

- Next-generation reactor/fuel cycle technology (higher efficiency, lower cost, improved safety and proliferation resistance)
- Advanced nuclear fuels
- Hydrogen production by nuclear systems

10.2 Project Summaries

In FY 2002, the initial year of the collaboration, six projects were awarded, with five additional projects awarded the following year and six in FY 2004. The final remaining FY 2003 project was completed during the past fiscal year, along with three FY 2004 projects. Four new collaborative projects were awarded to Korean partners in FY 2005, and an additional four in FY 2006.

A listing of I-NERI U.S./ROK projects that are currently underway, completed last year, and newly awarded follows, along with summaries of FY 2006 accomplishments and abstracts of the new projects.

Directory of Project Summaries

2003-020-К	Advanced Corrosion-Resistant Zirconium Alloys for High Burnup and Generation IV Applications	129
2004-001-K	Screening of Gas-Cooled Reactor Thermal-Hydraulic and Safety Analysis Tools and	
	Experiment Database	131
2004-002-K	Investigation of Heat Transfer in Supercritical Fluids for Application to the	
	Generation IV Supercritical Water-Cooled Reactor (SCWR)	135
2004-003-K	Development of Advanced Suite of Deterministic Codes for VHTR Physics Analysis	137
2004-004-K	Development of Voloxidation Process for Treatment of LWR Spent Fuel	139
2004-005-K	Development and Test of Cladding Materials for Lead-Alloy Cooled Transmutation Reactors	141
2004-006-K	Alternative Methods for Treatment of TRISO Fuels	145
2005-001-K	Supercritical Carbon Dioxide Brayton Cycle Energy Conversion	147
2005-002-К	Development of HyPEP, A Hydrogen Production Plant Efficiency Calculation Program	151
2005-003-K	Improvement of the Decay Heat Removal System for VHTR	155
2005-004-K	Development of Head-end Pyrochemical Reduction Process for Advanced Oxide Fuels	157
2006-001-K	Core Design Studies for Sodium-Cooled TRU Burner Reactors	161
2006-002-К	Separation of Fission Products from Molten LiCI-KCI Salt used for Electro-refining of Metal Fuels	163
2006-003-K	Development of Crosscutting Materials for the Electrochemical Reduction of	
	Actinide Oxides Used in Advanced Fast Burner Reactors	165
2006-006-К	VHTR Environmental and Irradiation Effects on High-Temperature Materials	167

Advanced Corrosion-Resistant Zirconium Alloys for High Burnup and Generation IV Applications

behavior.

PI (U.S.): Arthur T. Motta, Pennsylvania State University

PI (Korea): Yong Hwan Jeong, Korea Atomic Energy Research Institute (KAERI)

Collaborators: Westinghouse Electric Co., University of Michigan, and Hanyang University

Research Objectives

The objectives of this project were to 1) demonstrate a technical basis for improving the corrosion resistance of zirconium-based (Zr) alloys in extreme operating environments of light water reactors (e.g., high burnup, boiling, high temperature, aggressive chemistry) and 2) investigate the feasibility of using advanced zirconiumbased alloys in a supercritical water (SCW) environment. Model alloys were designed to isolate specific features of the microstructure that affect the formation of the protective oxide layer to study their impact on the corrosion rate. Researchers examined the fine structure of oxide layers formed on the model alloys and compared corrosion kinetics.

A key aspect of this program was to rationalize the differences in corrosion kinetics among alloys by identifying differences in the structure of the protective oxide formed on each alloy. To find the structural differences in the oxides, researchers used a number of advanced techniques for characterizing the metal and oxide and comparing differences between the oxide structure and the original microstructure of the alloy. These techniques included submicron-beam synchrotron radiation diffraction and fluorescence, cross sectional transmission electron microscopy (TEM), transmitted light optical microscopy, and nano-indentation.

Research Progress

Researchers have completed long-term corrosion tests to obtain detailed corrosion kinetics data and to assess the susceptibility of model zirconium alloys to stable or accelerated oxide growth in water at 360°C or 500°C—the

latter temperature relevant to the proposed supercritical water reactor. The team performed studies to address the specific role of alloying elements in promoting stable and protective oxide growth. They corrosion-tested 30 model alloys for over 400 days in 360°C water, and for 150 days to over 300 days in 500°C supercritical water. They also conducted limited testing in lithiated water. The oxides were later examined using advanced techniques to identify which characteristics were associated with protective oxide

The results from both low- and high-temperature corrosion testing showed a wide range of corrosion behavior. These differences manifested themselves in the kinetic parameters obtained for each alloy and in their susceptibility to breakaway corrosion. Several of the alloys, especially those in the zirconium-iron-chromium (Zr-Fe-Cr) class, showed protective behavior throughout the long-term autoclave tests both at high and low temperatures. The conclusions reached from corrosion testing were:

- In comparison with other alloys being considered for the supercritical water reactor (SCWR), the Zr alloys showed higher corrosion rates than austenitic alloys, but lower rates than ferritic-martensitic and 12Cr steels. Figure 1 shows the comparative weight gains between the best Zr alloys obtained in this study and ferritic martensitic alloys studied elsewhere.
- 2) Significant concerns exist in terms of creep rates and other properties, but this research indicates that proper alloying additions induce protective oxide growth in model Zr alloys. From a corrosion point of view, the Zr alloys should be considered as possible candidate materials for application in the SCWR.

Project Number: 2003-020-K

Project Start Date: February 2003

Project End Date: July 2006

davs.

Figure 1. Weight gain versus exposure time for zirconium-based model alloys tested under different autoclave conditions (SCW or steam).

The researchers also characterized oxides to determine which structures are associated with protective and nonprotective behavior. The main conclusions were the following:

- There are characteristic differences in the oxide-tometal interface regions of protective oxides compared to those of non-protective oxides. In particular, the presence of two interfacial oxide phases—a highly oriented tetragonal phase and a sub-oxide phase—were associated with protective behavior. Figure 2 shows a series of diffraction patterns of a protective oxide layer formed in supercritical water taken using microbeam synchrotron radiation. A large peak near the interface (marked T in the figure) shows the presence of the tetragonal phase associated with protective behavior.
- 2) The overall crystallographic texture of oxides revealed clear differences between protective oxides and nonprotective oxides. The growth direction of the oxides was similar but the distribution of oxide poles about the growth direction was considerably more anisotropic in the non-protective oxides.

3) The mechanisms of oxide formation and transition/

Figure 2. Diffraction intensity versus two-theta angle versus position in

the oxide for Zr1.0Cr0.2Fe corroded in 500°C supercritical water for 150

breakaway appear similar between alloys and for highand low-temperature corrosion testing.

Planned Activities

The project has concluded and the results are being published in peer-reviewed journals and conferences. The corrosion results show that certain zirconium alloys can be considered as candidate materials for some hightemperature applications. The knowledge gained from this study concerning protective oxide formation will enable researchers to design better alloys whose microstructure and composition induce stable oxide growth. This knowledge can also extend to other uses of Zr alloys in nuclear reactors, such as in extended burnup where further improvements in corrosion resistance are beneficial.

Screening of Gas-Cooled Reactor Thermal-Hydraulic and Safety Analysis Tools and Experiment Database

PI (U.S.): T.Y.C. Wei, Argonne National Laboratory (ANL)

Project Number: 2004-001-K

Project Start Date: June 2004

PI (Korea): W.J. Lee, Korea Atomic Energy Research Institute (KAERI)

Project End Date: May 2007

Collaborators: Idaho National Laboratory (INL)

Research Objectives

This research project supports development of the very high-temperature reactor (VHTR), one of six reactor technologies under the Generation IV International Forum (GIF). The Department of Energy selected this system to demonstrate the production of hydrogen in conjunction with emission-free electricity. The Korean Ministry of Science and Technology (MOST), under their Nuclear Hydrogen Development and Demonstration (NHDD) project, selected the pebble bed reactor (PBR) and prismatic modular reactor (PMR) technologies for further evaluation.

This project addresses the thermal-hydraulic and safety analysis capabilities that will be used in reactor plant analysis and auditing. Specific objectives of this research are to 1) develop a formal qualification framework, 2) conduct initial filtering of the existing databases, and 3) perform preliminary screening of the tools used in thermalhydraulic and safety analyses.

Research Progress

Of all the Phenomenon Identification and Ranking Table (PIRT) events screened to date, core flow distribution has been identified as one of the most important safety criteria. Steady-state core flow and bypass at normal operation set the core initial peaking conditions for the accident and transient scenarios. Uncertainty in the initial flow conditions of the core would not only affect fuel margins at steady-state, but would also propagate into the uncertainties in the safety criteria for other events in the plant duty cycle. In the case of a PMR, past experience indicates that core bypass flow could be a significant proportion of total core flow due to 1) the inter-block gaps, 2) radial and axial manufacturing and refueling tolerances for the graphite blocks, 3) irradiation swelling and cracking, and 4) distribution of the thermal expansion.

Similarly, in the case of a PBR, flow bypassing the core central regions could be a significant proportion of total core flow due to 1) random pebble and coolant void distribution, 2) edge effects at the center and side reflectors, and 3) the lateral leakage through the "loose-fitting" graphite pieces which form these reflectors.

Modeling core flow distribution is necessary. To further explore the modeling needs that address bypass flow phenomena, researchers performed sensitivity calculations to evaluate the relative importance of underlying subphenomena to the safety criteria. Eventually, these sensitivity studies will be utilized in the scaling analysis to filter out non-dimensional groups of lesser importance and to weight the contribution of each phenomenon in the uncertainty assessment.

This work was completed and results obtained for both the generic PMR and the generic PBR options. Figures 1 and 2 show the core block coolant leakage flow pathways for the PMR option, and the GAS-NET code network representation developed to calculate the sensitivities of associated flows.

The GAS-NET results of the PMR sensitivity calculations show that the behavior of the design seal at the core support structure is most significant. The next most important factor is the stacking surface fit for block elements at the core periphery. The other factors are much less significant.

Figure 1. Potential leakage pathways in PMR block core.

Figure 2. GAS-NET network presentation of PMR potential leak paths.

Assessing peak temperature in the PBR core relies on three individually predicted quantities: 1) volumetric rate of power generation in the hottest pebble in the bed, 2) coolant flow rate over the surface of that pebble, and 3) heat transfer coefficient associated with that flow rate and pebble bed configuration. The current assessment only focused on identifying thermal hydraulic modeling capability and needs, relying upon existing core neutronics analysis methodology to predict the peak rate of power generation with acceptable accuracy.

This research provides an initial assessment of the sensitivity of predictions of the core thermal hydraulic characteristics to the uncertainty in pebble size and pebble coolant void distribution. As a basis, researchers used experimentally derived correlations for the pressure loss and heat transfer coefficients taken from the open literature. They concluded that the largest sensitivity factor is the pebble bed void distribution.

KAERI continues to address the initial scaling analysis by working on both PBR and PMR plant analyses to obtain prototypic conditions. They performed iterative analyses for refined steady state core power and temperature distributions, alternating between VSOP-94 for core physics and GAMMA for the core thermo-fluids to better define temperature feedback. The modeling focused on the reference PBR and PMR operating conditions: 490°C reactor pressure vessel (RPV) inlet temperature and 950°C outlet temperature at a pressure of 70 bars.

The accident analyses were carried out for the refined HPCC (high pressure conduction cooldown) and LPCC (low pressure conduction cooldown) models using the MARS-

GCR and GAMMA codes, respectively. The PBR GAMMA model is given in Figure 3. The analysis results showed that both the core and RPV temperatures remain within the safety limit.

After obtaining results for the 600 MWt PMR and a 400 MWt PBR, researchers carried out a scaled-down calculation for a 100 MWt PMR. These results show that safety is enhanced due to the increased radiation surface area of the scaled-down RPV.

The researchers are also performing an initial filter of existing experimental databases based on the PIRTs. They conducted a database survey of nuclear reactors and experimental facilities. The reactor category was subdivided into research and commercial reactors. Experimental facilities were subdivided into large-scale and small-scale gas loops for Separate Effects Tests (SET) and component tests. PMR reactors include Dragon, Peach Bottom Unit 1, Fort St. Vrain, and the HTTR. PBR reactors include AVR, THTR, and HTR-10. The list of multipurpose test loops includes HENDEL and KVK, and the SET or Component test loops include NACOK, PBMM, and CTFL.

Experimental and reactor information has been filtered based on the design issues, major phenomena, and components investigated. To review design issues, researchers assembled the high-level information through the PIRTs and then classified them according to phenomena. Test-matrix tables were structured by three parameters: 1) accidents, 2) phenomena, and 3) components. A set of seven accidents have been defined that correspond to those evaluated for the PIRT work. An additional 11 phenomena and 14 components, also based

Figure 3. PBR GAMMA model.

on PIRT results, were utilized in the structure of the testmatrix tables, which include a corresponding list of facility capabilities. "Quick-look" reference tables briefly describe the designs and operating conditions of the facilities and are hyperlinked to the test-matrix tables and the reference documents for user convenience.

Researchers initiated a preliminary scoping analysis for the IAEA CRP-5 steady state temperature distribution benchmark problem HTR-10, used to assess the GAMMA multi-dimensional thermo-fluid model. In general, GAMMA results compare favorably with HTR-10 measurements, except for a large deviation near the bottom reflector and the discharge zones which may be due to measurement error. However, there is a need for further improvement of GAMMA, especially in modeling the cone-shaped lower core and reflector zones. Researchers used the MARS-GCR code to conduct preliminary scoping of the IAEA PBMM steady state power conversion unit performance benchmark. Nitrogen has been implemented as the main working fluid in MARS-GCR for the assessment, and compressor and turbine models have been incorporated into the code. The calculated results show acceptable agreement with the measurements.

Planned Activities

Due to budget uncertainties, the U.S. side will suspend work on all tasks except for PIRT refinements. However, KAERI will continue work and proceed according to plan. In particular, planned KAERI sub-tasks are

- IAEA PBMM transient benchmark using MARS-GCR
- OECD neutronics and thermo-fluid coupled analysis using MARS/MASTER
- ASME CFD benchmark using LILAC

I-NERI — 2006 Annual Report

Investigation of Heat Transfer in Supercritical Fluids for Application to the Generation IV Supercritical Water-Cooled Reactor (SCWR)

PI (U.S.): J.R. Wolf, Idaho National Laboratory (INL)

PI (Korea): Y.Y. Bae, Korea Atomic Energy Research Institute (KAERI) Project Number: 2004-002-K

Project Start Date: October 2004

Project End Date: September 2007

Collaborators: Rensselaer Polytechnic Institute (RPI)

Research Objectives

The objectives of this project are twofold: 1) to address issues associated with measuring heat transfer to supercritical water under prototypical supercritical water reactor (SCWR) conditions and 2) to develop tools to predict SCWR thermal transients. Because of the lack of phase change in the SCWR core, these reactors cannot use design criteria based on critical heat flux. The commonly accepted practice is to specify cladding temperature limits that must be met during different transient events. However, there is little information about heat transfer to supercritical water in a reactor environment.

In addition to supercritical water, researchers will also use surrogate fluids such as supercritical carbon dioxide (CO_2) as experimental media, which will provide valuable insight into the physical phenomena taking place. The lower critical temperatures and pressures of surrogate fluids can significantly reduce the cost and time to complete the experimental program.

Research Progress

One of the initial objectives was to design and construct a bundle test section for installation in Framatome's Benson Facility supercritical water test loop. Researchers have developed requirements for the bundle test section and completed the design. All design documents have been distributed to the appropriate collaborators. However, due to budget reductions, the test section will not be fabricated.

The final test section design for the Benson Loop is shown in Figure 1.

Figure 1. Benson loop supercritical water test section.

KAERI has completed construction of the CO_2 test facility. Commissioning of the facility is underway, including reduction of electrical noise and tuning. The researchers have completed testing and data analysis for the 4.4 mm tube, along with testing for the 9 mm tube. Results of numerical simulation of flow through these two tubes have been shared with the research team in order to establish boundary conditions for testing. The single rod test has recently begun and limited test results are reported here. Numerical simulation of flow through an annular passage of 8 mm ID and 10 mm OD is being performed. In addition, researchers will perform pre-test analysis to predict experimental behavior of an eccentric single rod test assembly. Sample test results are shown in Figure 2. Researchers analyzed specific benchmark problems with the RPI NPHASE computational fluid dynamics (CFD) code and will compare results to both experimental data and FLUENT simulations. Analysis performed with these two different codes will yield additional insight into the physical processes taking place in the tests. Researchers also expect to identify areas where additional code development is needed.

Planned Activities

Although the test section has been designed and requirements completed, as previously discussed, it will not be fabricated due to budget restrictions. No additional work is planned in this area. Researchers will continue to perform additional supercritical CO_2 tests and will benchmark the NPHASE CFD code using data from these tests.

Figure 2. Test results for 9 mm ID tube.

Development of Advanced Suite of Deterministic Codes for VHTR Physics Analysis

PI (U.S.): Temitope A. Taiwo, Argonne National Laboratory (ANL)

Project Number: 2004-003-K

PI (Korea): K. S. Kim, Korea Atomic Energy Research Institute (KAERI)

Project End Date: September 2006

Project Start Date: October 2003

Collaborators: Seoul National University (SNU)

Research Objectives

The objective of this project was to develop an advanced suite of deterministic computer codes for designing and licensing prismatic very high-temperature reactors (VHTR). Researchers investigated the capabilities of currently available physics analysis tools, focusing on VHTR modeling requirements. Based on this assessment, they identified required enhancements to the lattice physics, whole-core, and fuel cycle modeling capabilities. To implement these enhancements, the team developed an advanced suite of codes based on the conventional twostep lattice and the whole-core calculational approach, as applied to block-type VHTR designs.

To perform the VHTR analysis, the researchers also adapted the 3-D whole-core transport code DeCART, which was developed for light water reactor (LWR) applications under a previous I-NERI project. This code represents local heterogeneity explicitly without homogenization, eliminating the approximations and laborious generation of multigroup constants needed for the two-step approach. Other code features include using a multigroup crosssection library directly without group condensation and incorporating pin-wise thermal-hydraulic feedback. The team performed necessary verification and validation tests of the new suite of codes using appropriately defined benchmark problems and available experimental data.

Research Progress

The researchers previously accomplished the following tasks:

Assessed existing computational tools. These assessments focused on the static and depletion analysis capabilities of the tools to identify their

usefulness for the neutronics design and analysis of VHTRs. The researchers identified neutronics characteristics to be simulated in the core models along with Monte Carlo and deterministic capabilities for their representation. Based on these assessments, two separate code systems were selected for the conventional two-step core analysis procedure: 1) WIMS8 and DRAGON for generating multigroup cross sections and DIF3D/REBUS-3 for whole-core physics and fuel cycle analyses and 2) HELIOS lattice code and the MASTER whole-core analysis code. In order to use HELIOS, which is not capable of modeling coated fuel particles explicitly as are WIMS8 and DRAGON, researchers developed an indirect method, called the "reactivity-equivalent physical transformation" (RPT) procedure. RPT captures the self-shielding effect of randomly distributed fuel particles by representing a fuel region as an equivalent cell of two homogeneous zones.

- **Completed initial code modifications**. Researchers modified these code suites for applicability to VHTR analysis and performed preliminary verification tests. The results indicated that these code suites were working properly and provided reasonably accurate solutions compared to high-fidelity Monte Carlo solutions.
- **Benchmarking**. The team evaluated existing data in order to create benchmark problems for use in validating the codes.

During FY 2006, researchers made additional modifications to the code suites to make them suitable for VHTR analysis. First, they identified limitations in code capabilities and incorporated modifications to provide the

needed features. The DRAGON lattice code has been modified to produce data for the assembly pin power distribution and the surface fluxes and currents in order to generate cross-section equivalence parameters. A better representation of the surface parameters is needed in the future, since only temporary models have been developed.

Researchers developed a cross-section manager toolkit (X-MANAGER) to help automate routine jobs and simplify data management. This toolkit runs DRAGON depletion jobs for different temperatures and prepares the cross sections required for the REBUS-3/DIF3D wholecore simulation package. Researchers have developed procedures for generating cross sections for different core zones. They have also developed utility codes to generate cross section data using the HELIOS lattice code and to process the data for use in the MASTER whole-core analysis code. The latter code has been extended to functionalize and interpolate microscopic and macroscopic *multigroup* cross sections. Additionally, the team implemented a thermal feedback calculation module for prismatic VHTR cores.

For the REBUS-3/DIF3D package, code features that require development or upgrade for VHTR analysis were identified and the required modifications implemented. These modifications include new cross section interpolation routines, routines for handling large data files, modules for thermal feedback calculations, and code upgrades to utilize cross section equivalence parameters. The researchers have completed preliminary verifications of these code modifications.

A primary effort this year has been incorporating a hexagonal transport calculation kernel in the multigroup, spatially-heterogeneous DeCART code to analyze the heterogeneous VHTR core problem using the method of characteristics (MOC). The code was developed for light-water reactor cores under a previous I-NERI project, For this project, researchers developed a modular ray tracing scheme to reduce the memory size allocated for tracking the hexagonal block array. The scheme uses path linkage to connect the modular rays between neighboring blocks. Additionally, a multigroup unstructured coarsemesh finite difference (CMFD) acceleration scheme was implemented to more efficiently obtain the transport solution. Researchers analyzed test problems of varying difficulties using this code and compared eigenvalue and power distribution results to Monte Carlo solutions calculated with MCNP. Following are the problems they analyzed:

- BLOCK-1, a standard VHTR prismatic block graphite assembly, with regular hexagonal cells for fuel compacts and coolant holes
- BLOCK-2 incorporates a large hole for control rod position
- BLOCK-3 has a central fuel handling hole, but otherwise similar to BLOCK-1
- The 2-D core is composed of these assembly blocks (i.e., BLOCK-1, -2, and -3)

Table 1 shows the preliminary results of eigenvalue comparisons for the VHTR assembly blocks and 2-D whole-core problems, indicating that the code is working properly. Researchers have also obtained good results for a numerical benchmark based on the Compact Nuclear Power Source experiments.

The collaborators published four topical reports on cross section/code development, REBUS-3/DIF3D enhancements, HELIOS/MASTER procedures, and benchmark calculations.

Planned Activities

This I-NERI project is complete. However, researchers will continue their efforts to develop, verify, and validate the code suites and the DeCART code for VHTR analysis.

thus has an existing Cartesian transport kernel.

Problem	Code	Parameter	300 K	600 K	900 K
	MCNP	k _∞	1.53112	1.48272	1.44922
BLUCK-1	DeCART 190g	Δ ρ, pcm	-98	-8	-43
BLOCK-3	MCNP	k _∞	1.53382	1.48853	1.45417
	DeCART 190g	$\Delta \rho$, pcm	-8	-27	-13
BLOCK-2	MCNP	k _∞	1.54717	1.50096	1.46770
	DeCART 190g	$\Delta \rho$, pcm	-51	20	20
2-D CORE	MCNP	k _{eff}	1.43657	1.41004	1.38729
	DeCART 190g	Δρ, pcm	56	-	-

Table 1. Comparison of DeCART and MCNP Eigenvalues.
Development of Voloxidation Process for Treatment of LWR Spent Fuel

PI (U.S.): B.R. Westphal, Idaho National Laboratory (INL)

PI (Korea): J.J. Park, Korea Atomic Energy Research Institute (KAERI)

Collaborator: Oak Ridge National Laboratory (ORNL)

Research Objectives

This research project is developing a head-end fuel treatment process for light water reactor (LWR) spent fuel, known as voloxidation, as part of the Advanced Fuel Cycle R&D initiative. Voloxidation involves oxidizing uranium oxide fuel at high temperature, using either air or oxygen, in order to separate fuel from the cladding, reduce particle size, and remove volatile fission products. When used as a head-end step for aqueous or pyrochemical treatment processes of spent nuclear fuel, voloxidation provides three important advantages:

- Separation of fuel from the cladding, which would simplify process flow sheets by excluding the cladding constituents from the fuel constituents. Segregation of cladding may also generate less high-level waste.
- Decrease in fuel particle size as a result of the oxidation cycle, which may increase the efficiency of downstream dissolution processes.
- Removal of problematic constituents from the fuel prior to downstream treatment operations. Gaseous fission products such as cesium-137, krypton-85, xenon-133, technetium-99, carbon-14, and tritium (³H) may be removed prior to fuel dissolution, thus simplifying the process flow sheets and yielding more flexible waste treatment operations.

This project focuses on process development in three general areas:

 Measurement and assessment of the release behavior of volatile and semi-volatile fission products from the voloxidation process.

- Assessment of techniques to trap and recover gaseous fission products.
- Development of process cycles to optimize fuel cladding separation and fuel particle size.

Project Number: 2004-004-K

Project Start Date: June 2004

Project End Date: April 2007

Research Progress

Researchers continued conducting hot experiments to assess the release behavior of volatile and semi-volatile fission products during voloxidation using irradiated spent oxide fuels. They performed tests with carbon-14, krypton, and cesium fission gases (14C, 85Kr, and Cs) to determine their release fractions during an oxidation/reduction cycle. For ¹⁴C and ⁸⁵Kr, the release characteristics are similar and linked to the degree of grain boundary diffusion. For cesium, researchers achieved a release fraction over 90 percent for voloxidation cycle temperatures to 1,250°C (Figure 1). Additional tests to study the effects of temperature, pressure, oxidative gases, and cladding on fission product removal showed that higher temperatures, vacuum conditions, and declad fuel improve removal efficiencies. A significant improvement was noted for cesium, with a removal of 99 percent for vacuum conditions above 1,000°C.

Researchers performed additional tests with surrogate materials at bench-scale to evaluate the performance of trapping media for the volatile fission products. Specifically, they conducted experiments with a calcium-based filter under varying operating conditions to maximize the trapping of ruthenium, ¹⁴C, and technetium. A temperature of 600°C was optimal for trapping efficiencies over 99 percent.



Figure 1. Cumulative release fraction of cesium during voloxidation at 1,000°C and 1,500°C.

Because the original equipment used for irradiated voloxidation testing required major modifications for trapping volatile off-gases, researchers designed a new offgas treatment system (OTS) for the hot cell environment. The new OTS (Figure 2) includes four separate heating zones—one for oxidation of the spent fuel and three for collection of specific volatile fission products. The three targeted fission products are cesium, ruthenium, and iodine, which will be collected on fly-ash, calcium-based, and zeolite filters, respectively. Other secondary fission products to be trapped include rubidium, cadmium, technetium, and ¹⁴C. During the past year, researchers completed the OTS design, fabrication, and qualification, and installed it in the hot cell facility for irradiated operation.

Planned Activities

Using filter media developed at KAERI, researchers will initiate testing with spent oxide fuel to evaluate the trapping efficiency of the filters and optimize the operating parameters with respect to

removal efficiency. Following an initial series of tests with the new OTS, they will extensively analyze filter media from the three trapping zones to determine the collection efficiency of the desired volatile fission products. Analyses may include gravimetric, gamma spectroscopy, chemical, X-ray diffraction, and electron microscopy to determine the extent of fission product retention. Based on these data, further tests will optimize the voloxidation process for both the removal and collection of the volatile fission gases of interest.



Figure 2. (a) Schematic of the OTS and (b) photograph installed in hot cell facility.

Development and Test of Cladding Materials for Lead-Alloy Cooled Transmutation Reactors

PI (U.S.): N. Li, Los Alamos National Laboratory
(LANL)Project Number: 2004-005-KPI (Korea): T. Song, Korea Atomic Energy
Research Institute (KAERI)Project Start Date: June 2004Project End Date: May 2007

Research Objectives

The objective of this research project is to develop accurate, reproducible methods for measuring the fuel cladding corrosion rate and corrosion product redistribution in lead-alloy flow loops under conditions similar to those of transmutation reactors. Researchers are exploring both subcritical and critical systems in support of the Advanced Fuel Cycle R&D (AFCR&D) and Generation IV lead-alloy-cooled fast reactor (LFR) programs. They will test new cladding materials, including silicon and chromium-containing alloys, to identify suitable candidates and to define directions for innovative and evolutionary cladding material developments. They expect to develop internationally acceptable procedures for measuring corrosion rates by conducting tests with a set of controlled materials.

Collaborators: Seoul National University (SNU)

This project consists of the following tasks: 1) loop corrosion measurements and benchmarking, 2) reference electrode development and benchmarking, and 3) corrosion product transport and low-temperature embrittlement research. The collaborating organizations will define the materials and test conditions. The research team will incorporate improved oxygen sensors, accurate flow meters, and coolant chemistry monitoring techniques into the process. They will systematically construct data sets to characterize the effects of chemical and metallurgical variables on corrosion resistance in transmutation reactor environments. Objectives will be achieved when all three laboratories reproduce consistent test results on candidate cladding materials over a range of coolant conditions.

Research Progress

Since U.S. research funding was greatly reduced, LANL did not perform all originally planned activities and will defer them to the third year. The following results have been achieved:

Loop Corrosion Measurement and Benchmarking. The objective this task is to obtain lead-bismuth (Pb-Bi) and Pb corrosion data of steel samples using the loops developed by each collaborator. Researchers are testing conventional reactor materials and have selected new cladding materials, including silicon (Si) and chromium (Cr)-containing alloys, to identify suitable candidates for further development.

During the past year, researchers completed construction of two Pb-Bi loops (at KAERI and SNU) and a Pb loop at KAERI. They performed additional Pb-Bi corrosion tests using the LANL DELTA loop at 535°C. In addition, they pre-tested some samples using the Pb-Bi loop at SNU and KAERI's Pb loop (Figure 1).

Reference Electrode Development and Benchmark. In this task, the team is developing solid-state yttriastabilized zirconia (YSZ) membrane sensors to measure the oxygen content in lead-alloys. The corrosion data cannot be analyzed without this information.



Figure 1. (a) Pb and Pb-Bi loops at KAERI; (b) 450°C testing at KAERI; (c) HELIOS at SNU.

Researchers have established fabrication procedures and completed the fabrication and calibration of nine sensors using Bi/Bi_2O_3 reference material and tantalum (Ta) wire. Calibrations were performed at two sets of conditions: 1) 350°C using hydrogen and oxygen directly and 2) 450°C using hydrogen and water in oxygen control (Figure 2).

Corrosion Product Transport and Low-Temperature

Embrittlement. Researchers are developing a systematic framework to construct a set of system corrosion kinetics models. This framework will incorporate interface oxidation and scale removal with hydrodynamic transport and redistribution of corrosion products. The theoretical calculation method can be applied to each organization's test loops to predict dissolution and precipitation and to study lowtemperature embrittlement. The team has completed development of a framework for measuring corrosion products. This model can be used to compare measurements with predicted values. Researchers have also designed and fabricated the embrittlement test facility and established a test plan.





Figure 2. (a) Oxygen sensor calibration apparatus at SNU; (b) a calibration data graph.



Figure 3. (a) Unifying the reported corrosion test data based on LANL's model; (b) SNU design of corrosion product measurement device; (c) SNU device to measure low-temperature liquid metal embrittlement.

Planned Activities

During the remainder of this project, the research team plans to accomplish the following activities:

- Conduct extended test of Pb-Bi eutectic (LBE) above 500°C in DELTA
- Conduct LBE test at 570°C for 500 hours
- Conduct Pb test at 550-600°C for 500 hours
- Conduct LBE test at 450°C for 1,000 hours
- Perform online corrosion measurement
- Test calibrated oxygen sensors in loops
- Measure corrosion products in loops and compare to model
- Perform liquid metal embrittlement tests and analyze results



Alternative Methods for Treatment of TRISO Fuels

PI (U.S.): Christine T. Snyder, Argonne National Laboratory (ANL)

Project Number: 2004-006-K

Project Start Date: May 2004

PI (Korea): Eung-Ho Kim, Korea Atomic Energy Institute (KAERI)

Project End Date: December 2006

Collaborators: None

Research Objectives

Current methods in treating spent tri-isotropic (TRISO) coated fuel particles call for dry mechanical crushing and/or grinding to breach the outer pyrolytic carbon and silicon-carbide layers of the fuel particle. The objectives of this project were to 1) explore alternative methods of breaching the protective layers and 2) investigate new separation methods for the resulting fines and fuel.

The Advanced Fuel Cycle R&D (AFCR&D) initiative and the KAERI Advanced Spent Fuel Conditioning Process (ACP) are developing strategies for separating and transmuting the transuranic elements (plutonium, neptunium, americium, and curium) and long-lived technetium and iodine fission products (⁹⁹Tc and ¹²⁹I) contained in spent nuclear fuel. Most of the high-level radiotoxic material in nuclear waste destined for direct disposal consists of these long-lived fission products and the transuranics that can be transmuted, recycled, and/or disposed of in smaller volume. TRISO-coated fuel particles from hightemperature gas reactors (HTGR) will be part of this inventory.

As shown in Figure 1, TRISO fuel consists of a microspherical kernel of uranium oxide 450-600 μ m in diameter encapsulated in the following layers: 1) a low-

density, porous carbon buffer layer 55-60 μ m thick adjacent to the fuel kernel; 2) an inner isotropic pyrolytic carbon (PyC) layer 35-40 μ m thick; 3) a silicon carbide layer approximately 35 μ m thick; and 4) an outer, dense PyC coating approximately 45 μ m thick.

Coated fuel particles are mixed with a graphite matrix and sintered to form a fuel compact. These fuel compacts are contained by fuel rods that are approximately 35 mm in diameter and 600 mm in length, which are inserted into vertical holes in a graphite block. When placed into the HTGR core, helium gas coolant flows through gaps between the holes and the rods.

Research Progress

Researchers investigated several promising technologies for breaching the TRISO coating for this study. Following were the methods considered, ranked in order of effectiveness:

- 1) Radio Frequency Induction
- 2) Microwave
- 3) Ultrasound
- 4) Laser



Figure 1. TRISO fuel particle structure.

Each of these techniques uses various methods of interaction with the coating material to cause rapid heating that leads to breaching of the fuel kernel. The interaction induces thermal stress cracking and debonding in the coating layers by thermal expansion of residual internal gas. This results in a rupture of the TRISO particle coating. These methods may be combined with a subsequent quenching to enhance stress on the material properties.

Promising separation technologies that were considered for this study can also be ranked in order of effectiveness:

- 1) Fluidization separator
- 2) Cyclone separator
- 3) Electrostatic precipitation

Separation was carried out according to the differences in density or size of the particles. The fluidization separator used in this project was equipped with a vibrator for effective separation of breached particles from kernels. The cyclone was simple, durable, and easy to maintain as it had no moving parts. Cyclone separators use centrifugal forces generated by a rapidly spinning gas to separate solids. Larger particles migrate to the wall of the cyclone and proceed downward while the smaller particles tend to exit upwards with the overflow.

The electrostatic precipitator imparts an electrical charge to the particles passing through a high-intensity electrical field. The electrostatic precipitator can be used for collecting very fine powders from sub-micron up to 60 microns.

Planned Activities

The research objectives and funding are concluded. In this project, the research team evaluated each breaching and separations method independently. In future work, they may investigate conceptual models that combine two or more techniques into a single process for breaching and separation.

Supercritical Carbon Dioxide Brayton Cycle Energy Conversion

PI (U.S.): J. Sienicki, Argonne National Laboratory (ANL) Project Number: 2005-001-K

Project Start Date: October 2005

Project End Date: September 2008

PI (Korea): S.O. Kim, Korea Atomic Energy Research Institute (KAERI)

Collaborators: None

Research Objectives

The objectives of this project are to develop supercritical carbon dioxide $(S-CO_2)$ Brayton cycle energy conversion systems and evaluate their performance when coupled to advanced nuclear reactors. Researchers are focusing on three Generation IV designs: 1) the sodium-cooled fast reactor (SFR), 2) the very high-temperature reactor (VHTR), and 3) the lead-cooled fast reactor (LFR). The S-CO₂ Brayton cycle is expected to increase plant efficiency and reduce balance-of-plant costs relative to a Rankine steam cycle operating at the same reactor core outlet temperature. The project is organized into the following tasks:

- Develop preliminary S-CO₂ Brayton cycle layouts for the KALIMER-600 SFR and the gas turbine modular helium reactor (GT-MHR), a reference plant for the VHTR
- Develop overpressure protection for sodium-to-CO₂ heat exchangers and optimize KALIMER-600 turbine design
- Develop TURB1D analysis code for turbine design, 3-D computational fluid dynamics code for turbine performance, and model centrifugal compressors
- Develop designs for the Small Secure Transportable Autonomous Reactor-Liquid Metal (SSTAR-LM) LFR
- Identify transients and S-CO₂ Brayton cycle control schemes for load following

- Develop system level codes to model S-CO₂ Brayton cycle and a natural circulation LFR
- Develop S-CO₂ Brayton cycle energy converters and evaluate performance for the SFR, VHTR, and LFR

Research Progress

All objectives for the first year of the project have been met. Following is an overview of progress made throughout the year.

Researchers developed a preliminary $S-CO_2$ Brayton cycle energy converter layout (Figure 1) for the 600 MWe (1,523 MWt) KALIMER-600 SFR, one of three reference SFR plants under the Generation IV International Forum (GIF). They calculated a net plant efficiency of 41.3 percent with an intermediate sodium circuit incorporated in the design,



Figure 1. Schematic illustration of S-CO₂ Brayton cycle coupled to KALIMER-600.



Figure 2. Schematic illustration of S-CO, Brayton cycle coupled to a GT-MHR.

compared to 39.4 percent for a superheated Rankine steam cycle. They also developed an overpressure protection system concept for the shell-and-tube sodium-to- CO_2 heat exchangers using Design Institute for Emergency Relief Systems (DIERS) decision tree procedures, consisting of a rupture disc set to fail at 1.0 MPa pressure and release sodium into a dump tank.

Researchers also developed a preliminary S-CO₂ Brayton cycle concept coupled to a 600 MWt GT-MHR and calculated a net plant efficiency of 53.7 percent, compared to 48.0 percent for a conventional design (Figure 2).

The team has developed a suite of supporting analytical tools:

- The one-dimensional computer code TURB1D to optimize a KALIMER-600 turbine design
- A three-dimensional Computational Fluid Dynamics (CFD) analysis for turbine performance, applied to two stages
- Models for centrifugal compressors, used to develop preconceptual compressor designs and performance maps for the 20 MWe (45 MWt) SSTAR LFR, demonstrating that centrifugal compressors provide a wider operating range between stall and choking near the critical point than axial flow compressors

- Steady-state one-dimensional and multidimensional analysis capabilities for compact heat exchangers (using CFX 5.7.1 CFD), incorporating Printed Circuit Heat ExchangerTM concepts for sodium-to-CO₂ heat exchangers, recuperators, and coolers
- Identified system transients and investigated possible S-CO₂ Brayton cycle control schemes for KALIMER-600 load following, adapting the MARS computer code (based on RELAP5)
- Exploratory experiments to investigate the basic nature of chemical reactions between sodium and CO₂ between 200 and 600°C

In addition to this work, ANL researchers developed a systems-level plant dynamics computer code to model the $S-CO_2$ Brayton cycle coupled to a natural circulation 181 MWe (400 MWt) STAR-LM LFR plant.

Previous Brayton cycle analysis codes used simplified ideal gas approximations for turbomachinery design and performance, which are not suitable for CO₂ property variations, especially sharp changes in the vicinity of the critical point. This new code solves timedependent, one-dimensional energy, mass, and momentum conservation equations for the compressible S-CO₂ fluid, plus the turbomachinery shaft dynamics equation on a multinodal representation of the complete S-CO₂ Brayton cycle energy converter. The code modeling includes the major secondary components (such as turbine, main compressor, and recuperators), control systems, and a simple systems model for a LFR.

Researchers developed an optimal control strategy for the SSTAR LFR that enables autonomous load following over the complete range of electrical grid load demand (essentially 0 to 100 percent), using steady state analysis computer codes. The strategy involves turbine bypass control over the entire range of load demand with inventory control between 35 and 100 percent load demand. Incorporating centrifugal compressors increased the operating range for inventory control beyond that previously calculated for axial compressors. Researchers also carried out initial Plant Dynamics Code calculations for a set of STAR-LM LFR operational transients, including a decrease in load demand followed by a return to full load. The results, shown in Figure 3, demonstrate the controllability of the plant and autonomous load following using turbine bypass combined with inventory control. Code development efforts have benefited from a commercial custom-designed turbomachinery vendor's suggestions to use centrifugal versus axial compressors and suggestions to improve the turbine and compressor models by including diffusers and more realistic calculations of turbomachinery efficiency.

Planned Activities

Researchers will continue to develop the $S-CO_2$ Brayton cycle energy converters and their components and evaluate their performance for the SFR, VHTR, and LFR.



Figure 3. Plant dynamics code simulation of STAR-LM response to reduction and increase of electrical grid load demand—(a) turbine and compressor work, (b) compressor stall check, (c) valves control action, (d) Brayton cycle pressures.

Development of HyPEP, A Hydrogen Production Plant Efficiency Calculation Program

PI (U.S.): C. Oh, Idaho National Laboratory

Project Number: 2005-002-K

PI (Korea): Y. J. Lee and W. J. Lee, Korea Atomic Energy Research Institute (KAERI)

Project End Date: September 2008

Project Start Date: October 2005

Collaborators: Argonne National Laboratory

Research Objectives

This research project will evaluate and optimize cycle efficiencies for producing hydrogen and electricity in a very high-temperature reactor (VHTR). Systems for producing electricity and hydrogen are complex and the calculations associated with optimizing these systems are intensive, involving a large number of operating parameter variations and many different system configurations. This research project will produce the HyPEP computer model, which is specifically designed to be an easy-to-use and fast-running tool for evaluating nuclear hydrogen and electricity production facilities. The HyPEP computer model accommodates flexible system layouts and its cost modeling capability makes it well-suited for system optimization.

Specific activities of this research are designed to develop the HyPEP model into a working tool, including 1) identifying major systems and components for modeling, 2) establishing system operating parameters and calculation scope, 3) establishing the overall calculation scheme, 4) developing component models, 5) developing cost and optimization models, and 6) verifying and validating the computer code. Once the HyPEP code is fully developed and validated, it will be used to execute calculations on candidate system configurations.

Research Progress

The research team began the first task of this project, which includes defining the scope of system modeling in HyPEP by identifying and hierarchically categorizing the major systems, components, and the operating parameters of the Nuclear Hydrogen Development and Demonstration (NHDD) facility and the Next Generation Nuclear Plant (NGNP). During the first three months, they evaluated and selected a programming language, set up the overall model/calculation requirements, and established the hierarchical system/component modeling system. The component hierarchy has been defined and the overview is shown in Figure 1. The results will form the basis of the plant modeling interface of HyPEP and help define the thermal-hydraulic processes and phenomena to consider.

Researchers have begun formulating the numerical solution scheme and established the following overall outline. The node-link-block is the basic fluid/heat flow network scheme:

- Node component handles chemical reactions for the high-temperature electrolysis and thermo-chemical systems. It represents thermal-hydraulic volume with scalar properties (e.g., volume, mass, molar fraction, energy, pressure, temperature).
- *Link component* models flow between nodes and has such properties as mass flow rate, pressure drop, and the scalar properties of the donor-node.
- *Block component* represents the solid structures that conduct or generate heat and provides models for the solid-to-fluid boundaries where convection occurs.

The basic equations consider the steady-state mass and energy transport of reactive multi-species fluid mixtures. General thermo-dynamic table search routines have been written and are being tested. The routines are developed with options for user-added property tables.

Researchers have evaluated four programming languages – Visual C++, Visual Basic, Fortran, and Delphi – and selected Delphi for the HyPEP development. Delphi is an object-oriented version of the PASCAL language and has versions for the .NET as well as the win32 environment. As HyPEP will have a strong



Figure 1. Hierarchical system/component of HyPEP.

hierarchical structure, the object-oriented program language was deemed to best suit the programming style. The language comparison is summarized in Table 1.

The development Graphical User Interface (GUI) for the alpha version of HyPEP has been completed. A screen capture of the GUI for the HyPEP alpha version is shown in Figure 2. The HyPEP alpha version is being programmed using the win32 version of the Delphi language.

Various routines have been developed for the GUI including component generation and deletion, dragdrop of components, on-screen manipulation of the components, automatic line generation for the link component, connection maintenance, and thermal hydraulic system build up. The definitions for the object classes TNode and TLink (for Node and Link components) which form the base components for the HyPEP alpha have been setup and are also being tested in the prototype windows form.

The TNode and TLink Objects have been derived from the graphic objects of the main canvas using the inheritance of the Object Oriented Program paradigm. In this way, the correspondence between the object in the graphic canvas and the thermalhydraulic component can be ensured. Preliminary component templates have been created with a total of 32 components categorized into 9 component categories.

Features/Comments	DELPHI	C++	Visual Basic	Fortran
Base Language	Pascal	С	Basic	Fortran
Win32 Version	Delphi2005	C++	MS Visual Basic	Compaq, Lahey etc.
NET Version Common Language Reuntime	Delphi2005	MS C# Borland C# etc.	MS Visual Basic	Intel Fortran
OOPS Capability	Very Good	Very Good	Good	No
Structured Programming	Good	Good	Good	Poor
Body of ready-made scientific software	Small	Small	Small	Large
Code Reusability	Good	Good	Fair	Poor
Web Programming Capability	Good (ASPX, ActiveX)	Good	Good (ActiveX)	Poor
Integrated Development Environment	Good	Good	Good	Good
Rapid Application Development (RAD)	Good	Fair	Fair	Poor

Table 1. Comparison of major programming languages.



Figure 2. Screen Capture of the HyPEP alpha version.



Figure 3. Required electrical work done on the electrolyzer.

As part of system integration, INL and ANL developed a high-temperature electrolysis model, calculated the power requirement for high temperature electrolysis (HTE), and implemented the results into the codes. INL also calculated the sizing of all the heat exchangers in the coupled VHTR and HTE.

Figure 3 shows results of electrical work done on the electrolyzer. As shown in Figure 3, the increase in current density reduces the number of cells and increases the electrical work on the electrolyzer.

Planned Activities

At the end of year 2006, it is expected that a working preliminary version (alpha version) of the HyPEP program will be produced. Major planned R&D activities in 2007 are as follows:

Basic Equation Setup and Numerical Solution Development. Researchers will set up steady state mass and energy conservation equations for Flow Network. A numerical solution scheme will be developed for flexible network topology. Ancillary routines to obtain T/H properties, heat transfer correlations, pressure drop correlations, etc., will be developed and coded. The numerical scheme will be coded to produce a preliminary working code, and verification and refinement are planned for the next Fiscal Year.

GUI Development. The main activity will be developing the object class definitions, which will also be used in numerical coding development. The GUI support routines for component generation and deletion, dragdrop, connection maintenance, flexible layout, etc., will be developed and coded.

Component Model Development.

Component specific models will be developed. The models to develop include 1) VHTR and IHX model, 2) Power Conversion Unit (PCU) T/H Model (Brayton Cycle and Rankine Cycle), 3) HTE T/H Model, and 4) Thermo-Chemical Model (I-S Cycle). The Node, Link and Block component will be further developed. The palette for the on-screen input/output template for the component will be defined and developed.

NHDD System Model & Preliminary Calculation. With the interim version of the HyPEP, a simple NHDD System layout will be modeled and preliminary calculations will be carried out to confirm integrity of the numerical scheme and the ancillary routines.

Component Sizing Model Development. Models will be developed to evaluate the component sizing. Because the model will basically rely on a database, the main task will be to collect and categorize a reliable database.

Cost Model Development. Researchers will develop a simple model (based on the database) to calculate the overnight construction cost and an algorithm to calculate simple interest.

HyPEP Verification & Validation. HyPEP will be assessed against established codes such as HYSYS or ASPEN for the program verification and validation.

Development of System Optimization Method. A calculation method/procedure will be developed for the optimization of a Nuclear Hydrogen Production Facility. The HyPEP program will be used as the main tool in the method.

System Integration. All the sub-models in the VHTR with PCU and hydrogen plant will be integrated for steady-state analyses. This task includes transient analyses for the control system development.

Improvement of the Decay Heat Removal System for VHTR

PI (U.S.): T.Y.C. Wei, Argonne National Laboratory (ANL)

Project Number: 2005-003-K

PI (Korea): Y. S. Sim, Korea Atomic Energy Research Institute (KAERI) Project Start Date: October 2005

Project End Date: October 2006

Collaborator: Idaho National Laboratory (INL)

Research Objectives

The objective of this project was to improve performance of the reactor cavity cooling system (RCCS) of the Very High-Temperature Reactor (VHTR). The RCCS is a major component in safely and passively removing decay heat. Water-cooled and air-cooled RCCS concepts have been proposed that rely either on natural convection or gravity drain to discharge decay heat to the environment. The preliminary Phenomena Identification Ranking Tables (PIRTs) developed under a separate I-NERI project identified various phenomena that are important for these decay heat removal accident scenarios, and particularly to the RCCS.

Improving the RCCS is an important step in licensing the VHTR. This project has a number of tasks aimed at improving the performance of the RCCS in passively removing core decay heat during VHTR accident conditions. Following were the three goals of this project:

- To improve RCCS performance through a combination of well-focused experiments that provide data for model validation and design performance testing
- 2) To improve and validate the analysis methodology
- To improve the designs of innovative heat transfer concepts

The system can also accommodate a beyond-design basis accident involving depressurization of the primary coolant system accompanied by a loss of station electric power. Under this scenario, heat transfer occurs by passive conduction-radiation from the reactor core to the vessel boundary, and then primarily by radiation from the vessel wall to the RCCS. Upon DOE approval, the Natural Convection Shutdown Heat Removal Test Facility (NSTF) at ANL will be refurbished for both the water- and air-cooled options in order to conduct experiments to validate and improve the RCCS analysis tools. The objective is to acquire the model/ code validation data for natural convection and radiation heat transfer in the VHTR reactor cavity and the reactor cavity cooling system from experiments performed at the NSTF.

A coupled Fluent-RELAP analysis tool developed by INL will be used to analyze various RCCS design concepts and KAERI will develop a radiation-enhanced heat transfer mechanism. Validation studies will be carried out during the course of the project as the experiments are conducted.

Research Progress

Researchers worked on feasibility plans to refurbish the NTSF for VHTR reactor cavity cooling system experiments, since the facility was originally constructed for air cooling tests in sodium reactors. The results of scaling studies identified the important non-dimensional parameters for each separate-effects study. Based on these results, researchers determined the range of experimental conditions as well as the appropriate scale that most effectively simulates actual system behavior. They also developed a first-level modification plan for the NSTF facility. Figure 1 shows the as-built facility.



Figure 1. Natural convection shutdown heat removal test facility (NSTF).

To improve the RCCS by utilizing radiation structures, researchers developed the required analysis models and systems and investigated heat transfer characteristics. Based on the investigation results, they have successfully devised an RCCS design concept which has substantial heat removal capacity. This design is expected to be among the final candidate concepts for improving the RCCS.

Planned Activities

The U.S. suspended participation in these tasks on October 1, 2006. At this point, U.S. funding for the continuation of the project is uncertain.

Development of Head-end Pyrochemical Reduction Process for Advanced Oxide Fuels

PI (U.S.): S. D. Herrmann, Idaho National Laboratory (INL)

Project Number: 2005-004-K

PI (Korea): S. M. Jeong, Korea Atomic Energy Research Institute (KAERI)

Project End Date: September 2008

Project Start Date: October 2005

Collaborators: None

Research Objectives

Pyroprocessing can be a very effective method for recovering heavy metals and producing stable waste forms from the highly radioactive fission products in spent nuclear fuel. This is of great interest for the purpose of closing the fuel cycle for Generation IV fast reactors. However, pyroprocessing was originally developed for treating metal fuels, while the primary feed material for this process may be oxide spent fuel. A proposed solution to this problem is to develop a process for converting spent oxide fuel into a metallic form.

The United States and the ROK have been active in developing pyrochemical conversion methods for producing feed material that is compatible with pyroprocessing. Both countries favor this electrolytic reduction method, referred to as "oxide reduction." While significant advances have been made in oxide reduction over the last decade, including electrolytic reduction, a number of important technical issues need to be resolved in order to properly assess implementation of this technology.

This research project will advance the design of an economical, high-throughput oxide reduction process by focusing on two technical issues: 1) the effect of fission products and 2) process scalability.

Research Progress

During this fiscal year, researchers completed a series of bench-scale pyrochemical processing experiments with irradiated oxide fuel. These experiments involved the electrolytic reduction of six batches of spent oxide fuel, followed by electrorefining the reduced product to recover refined uranium. Researchers performed the experiments in an electrochemical apparatus that was positioned inside a hot cell at INL's Hot Fuel Examination Facility. The objectives were to 1) determine the extent of reduction of metal oxides in the spent fuel following the reduction process, 2) determine the decontamination factors for the uranium separation from fission products in the electrorefining process, 3) examine the distribution of fuel constituents between the salt and fuel phases in each process, and 4) assess the possible effect of accumulated fission products on either process.

The team performed six electrolytic reduction runs in succession with a single loading of molten salt. In each run, they loaded crushed oxide fuel (nominally 50 g) into a permeable steel basket immersed in 500 milliliters of molten lithium chloride salt (LiCl – 1 wt% $Li_{0}O$) at 650°C. An electric current applied between the steel basket cathode and a platinum wire anode submerged in the salt effected the reduction of the fuel to metal in the basket, while simultaneously forming oxygen gas on the platinum wire surface. A schematic of the electrolytic reduction cell is shown in Figure 1. They extracted samples of the salt before and after each reduction run for chemical and radiochemical analyses. The analytical results quantified those fission products (notably, cesium, barium, strontium, and iodine) that accumulated in the salt. The fuel baskets were removed from the salt after each run and placed in sealed storage containers, except for the third run. Researchers sectioned that basket, as shown in Figure 2. They extracted and analyzed a fuel sample from the sectioned basket, which revealed a 99.7 percent reduction in uranium oxide. They also quantified the reduction of other metal oxides in the reduced fuel.



Figure 1. Hot-cell electrochemical apparatus for bench-scale electrolytic reduction and electrorefining experiments. (electrolytic reduction setup shown with cylindrical fuel basket, spiral-wound platinum anode, centerline thermocouple, and reference electrode submerged in molten salt electrolyte).



Figure 3. Refined uranium metal deposits from reduced spent oxide fuel.

In the electrorefining runs, the team submerged each basket of reduced fuel into a molten salt of LiCI-KCl (eutectic)–UCl, at 500°C. An electric current applied between the reduced fuel basket anode and a submerged steel rod cathode effected the dissolution of uranium metal in the fuel basket and its simultaneous deposition onto the steel cathode as refined uranium. The researchers generated multiple cathode rods of refined uranium from the complete dissolution of reduced oxide fuel in each basket. Figure 3 illustrates refined uranium deposits on a cathode rod. Researchers



Figure 2. Post-test section of reduced fuel basket.

extracted samples of the salt before and after each electrorefining run and also collected a refined uranium fuel sample from one of the cathode rods in each run. The salt and fuel samples were subjected to chemical and radiochemical analyses. The analytical results revealed the distribution of fission products within the electrorefining process, as well as the decontamination factors for the refined uranium. After six successive runs with irradiated oxide fuel, researchers observed no deleterious effects of fission products from either the electrolytic reduction or the electrorefining process.

Using KAERI's Advanced Spent Fuel Conditioning Process (ACP) facility, researchers performed two electrolytic reduction runs at engineering scale with unirradiated uranium oxide, as shown in Figure 4. Each run involved the reduction of 10 kg of SIMFUEL—unirradiated U₂O₂ loaded with non-radioactive oxides of Ba, Ce, La, Mo, Sr, Cs, U, Zr, Ru, and Nd in proportion to those contained in typical spent fuel after 43,000 $\rm MWd/MT_{\rm HM}$ burnup and 10 years decay. They loaded the SIMFUEL into a cylindrical, porous, magnesia container within a reduction vessel, configured with a center electrode contacting the fuel and six platinum anodes around the central magnesia container. Then they filled the vessel with approximately 90 kg of LiCl-3 wt% Li₂O and heated the system to 650°C. A controlled electric current applied to the cell reduced the oxide fuel to metal, while forming and discharging oxygen gas. Once the reduction was complete, they off-loaded the salt from the reduction vessel and allowed the system to cool. The reduced fuel was removed and sampled at various locations within the bed, revealing uranium-oxide reduction in excess of 99 percent.



Figure 4. Engineering-scale reduction vessel with internal cut-away view (left) and installed in acp facility (right).

Researchers also performed bench-scale electrolytic reduction tests to define the reaction kinetics in support of a process model for high-throughput operations. Specifically, they initiated a matrix of electrolytic reduction tests for 10 g loadings of U_3O_8 , varying the applied charge from 20 to 120 percent of theoretical in 20 percent increments for constant applied currents of 0.1, 0.2, 0.4, and 0.8 amperes. The team is currently measuring the extent of uranium oxide reduction for each of the 24 runs.

Planned Activities

Over the next period, the research team plans to conduct a series of four electrolytic reduction runs with a single salt loading using irradiated oxide fuel in the hot-cell electrochemical apparatus at INL. The oxide fuel for the tests will consist of irradiated UO_2 fuel that has been subjected to a voloxidation process to form U_3O_8 . The primary variables in the four electrolytic

reduction runs are fuel basket containment material (i.e., porous stainless steel and porous magnesia) and the Li_2O concentration in the LiCl salt (i.e., 1 to 3 weight-percent). Through these electrolytic reduction runs, the researchers will be able to assess the possible effects of accumulated fission products on the reduction process. They will also perform experiments investigating the use of zeolites for the removal of fission products from LiCl.

Researchers will perform three electrolytic reduction runs at engineering-scale with SIMFUEL in KAERI's ACP Facility, involving salt ingot fabrication and the formation of metal product. In addition, the team will complete the matrix of electrolytic reduction tests at bench scale. Portions of the bench-scale test matrix will be repeated with stainless steel baskets in order to compare the efficacy of stainless steel and porous magnesia as containment materials.

Core Design Studies for Sodium-Cooled TRU Burner Reactors

PI (U.S.): Won Sik Yang, Argonne National Laboratory (ANL)

Project Number: 2006-001-K

Project Start Date: January 2007

PI (Korea): Yeong-Il Kim, Korea Atomic Energy Research Institute (KAERI)

Project End Date: September 2009

Collaborators: None

Project Abstract

This project will develop conceptual core designs for sodium-cooled fast reactors to transmute recycled transuranic (TRU) elements. Transuranics are the dominant contributors to spent fuel radiotoxicity, longterm heat, and dose. The objectives are to 1) develop the core designs for TRU burner reactors, 2) perform relevant verification and validation analyses, and 3) evaluate the performance enhancements provided by innovative safety design features.

The burner core design task will examine various design concepts to enhance the TRU transmutation rate under practical design constraints and to develop optimum core designs for different conversion ratios and power levels. Researchers will also investigate design options that minimize the burnup reactivity swing and improve reactivity feedback coefficients. The design method verification and validation will consist of high-fidelity simulations and analysis of fast critical experiments. In particular, detailed benchmark analyses will be performed for the ZPPR-15, ZPPR-21, and BFS-73-1 critical experiments. Researchers will analyze the sensitivities of cross sections and computational methods on neutronics performance parameters and evaluate the uncertainties of performance parameters.

For evaluation of safety performance enhancements, researchers will analyze unprotected accident sequences with the ANL SASSYS-1 and KAERI SSC-K safety analysis codes for various combinations of innovative design features, focusing on inherent plant performance. They will also demonstrate enhancements to safety margins provided by advanced modeling techniques.

Separation of Fission Products from Molten LiCl–KCl Salt Used for Electro-refining of Metal Fuels

PI (U.S.): Michael Simpson, Idaho National Laboratory (INL)

Project Number: 2006-002-K

PI (Korea): In-Tae Kim, Korea Atomic Energy Research Institute (KAERI)

Project End Date: September 2009

Project Start Date: January 2007

Collaborators: None

Project Abstract

The objective of this project is to develop a process for selectively removing fission products from the lithiumchloride–potassium-chloride (LiCl–KCl) eutectic salts used in pyroprocessing of spent nuclear fuel. Separating fission products from the salts is critical to minimizing the waste volume generated from pyroprocessing. The current technology for disposing of these fission products is nonselective, discarding the LiCl–KCl salts along with the fission products, thus creating large quantities of high-level radioactive waste. Since the eutectic salts do not need to be discarded, developing and implementing a selective process can significantly reduce the waste volume. Two basic approaches are being considered: 1) salt-zeolite ion exchange and 2) selective precipitation of fission products. Researchers have previously investigated both of these methods. However, a complete assessment and comparison of the options has not yet been accomplished. Additional data for each technology option are needed in order to assess the efficiency of removing fission product and associated processing costs. For this project, each laboratory will perform additional experiments and will work together to assess which technology is more attractive. Initially, they will focus on filling in data gaps for salt-zeolite ion exchange and for selective fission product precipitation. In the final year, both labs will perform an integrated demonstration using salt-containing fission products. A systems analysis will determine if one or multiple technologies should be implemented into the reference pyroprocessing flowsheet.

Development of Crosscutting Materials for the Electrochemical Reduction of Actinide Oxides Used in Advanced Fast Burner Reactors

PI (U.S.): Christine T. Snyder, Argonne National Laboratory	Project Number: 2006-003-K
PIs (Korea): Fund-Ho Kim and Jong-Hyeon Lee	Project Start Date: January 2007
Korea Atomic Energy Research Institute (KAERI)	Project End Date: September 2009
Collaborator: University of Illinois at Chicago	

Project Abstract

The objective of this project is to use surface engineering and coating technology to improve the chemical, thermal, and corrosion performance of the pyroprocessing vessel and related structural components. The conditions found in the oxide reduction step during spent nuclear fuel pyroprocessing represent one of the most aggressive thermal and chemical environments encountered by structural materials. When subjected to the convective flows generated by the pyro-metallurgical process, the high-temperature molten salts, coupled with oxygen gas, have been shown to corrode the most advanced nickel-based alloys and ceramic and metal coatings. In addition, the vessel might become exposed to uranium, plutonium, and other reactive elements during processing of the spent nuclear fuel. These elements may not only reduce oxides that are components of a coating material, thereby resulting in degradation, but may also produce low-temperature eutectic alloys.

The goal of this research is to develop a coating system that combines the thermal barrier qualities of graded ceramic layers with the corrosion protection of a metallic bond coat. One method to achieve this goal is to design coatings with progressive complexity. That is, developing combinations of materials to produce a "functional" gradient in the material properties. The gradients in functionally graded materials (FGM) are primarily guided by material property considerations.

In this project, the research team will focus on developing ternary and quaternary multicomponent oxides as well as a compatible bond-coating material to strengthen the adhesion of the ceramic coating to the metal substrate. This research will generate unique materials solutions to advance the design limits presently imposed upon structural alloy materials in the harsh environment found in spent nuclear fuel oxide reduction.

VHTR Environmental and Irradiation Effects on High-Temperature Materials

PI (U.S.): Dane F. Wilson, Oak Ridge National Laboratory	Project Number: 2006-006-K	
PI (Korea): Woo-Seog Ryu Korea Atomic Energy	Project Start Date: TBD	
Research Institute (KAERI)	Project End Date: TBD	
Collaborators: None		

Project Abstract

This project will focus on selecting and qualifying high-temperature materials for next-generation reactors. Researchers will investigate irradiation and environmental effects and develop a materials handbook containing properties of high-temperature metallic materials.

Specifically, they will study the effects of a helium environment on the mechanical properties of hightemperature metallic alloys proposed for use in the very high-temperature gas reactor (VHTR). Researchers will evaluate the effects of irradiation on high-temperature metallic alloys using Korea's High-Flux Advanced Neutron Application Reactor (HANARO) and the adjacent Irradiated Materials Evaluation Facility (IMEF). Finally, they will capture the results in a *Generation IV Materials Handbook* containing a database of high-temperature materials.

11.0 U.S./OECD Collaboration

The U.S. and an international consortium under the auspices of the OECD-NEA signed a bilateral I-NERI Agreement in March 2002. The U.S./OECD-NEA collaboration has resulted in only a single project to date, "Melt Coolability and Concrete Interaction," which was completed in FY 2005.

11.1 Work Scope

There are no ongoing projects with OECD. Previous R&D topical areas for the U.S./OECD-NEA collaboration included:

- Resolving ex-vessel debris coolability issues through a program that focuses on providing both confirmatory evidence and test data for the coolability mechanisms identified in the melt attack and coolability experiments integral effects tests
- Addressing remaining uncertainties related to long-term, two-dimensional, molten core-concrete interaction under both wet and dry cavity conditions

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