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UNITED STATES OF AMERICA

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Annual Report

International Nuclear Energy Research Initiative

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Foreword

The International Nuclear Energy Research Initiative (I-NERI) was established by the U.S. Department of Energy, Office of Nuclear Energy (DOE-NE) in Fiscal Year (FY) 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 Initiative, and the Nuclear Hydrogen Initiative. International collaboration is an important part of NE's R&D efforts. 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 2007, DOE initiated seven more projects with the Republic of Korea and two with Canada. This I-NERI *2007 Annual Report* provides a description of these new projects, along with a comprehensive summary of the progress of each collaborative research project initiated since FY 2004. Forging these partnerships enhances the participation of the United States within the global nuclear community, and helps build an international consensus on critically important issues such as designing proliferation-resistance into advanced nuclear systems and expanding the benefits of nuclear power.

R. Shane Johnson
Principal Deputy Assistant Secretary for Nuclear Energy
U.S. Department of Energy

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INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE

1.0 Introduction

The International Nuclear Energy Research Initiative (I-NERI) supports the *National Energy Policy* by pursuing international collaborations to conduct research that will advance the state of nuclear science and technology in the United States. I-NERI promotes bilateral and multilateral scientific and engineering research and development (R&D) with other nations. Innovative research performed under the I-NERI umbrella addresses key issues affecting the future of nuclear energy and its global deployment by improving cost performance, enhancing safety, and increasing proliferation resistance of future nuclear energy systems. A link to the program can be found at the U.S. Department of Energy Office of Nuclear Energy (DOE-NE) website: http://www.nuclear.gov.

This *I-NERI 2007 Annual Report* serves to inform interested parties about the program's organization, progress of collaborative research projects undertaken since FY 2004, and future plans for the program. Following is an overview of each section:

- **Section 2** discusses background information on the 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 2007.
- **Sections 5 through 10** present details of the R&D work scope for current I-NERI collaborative projects with Brazil, Canada, the European Union (EU), France, Japan, and the Republic of Korea, respectively. For each participant, the report presents an index of projects and a summary of technical accomplishments achieved in FY 2007. There are no current projects with the Organization for Economic Co-operation and Development (OECD).

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 for addressing the nation's energy and environmental needs during 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 implement a properly focused (R&D) effort to address the principal obstacles to continuing a viable nuclear energy option. In 1999, DOE responded to these recommendations with the establishment of the Nuclear Energy Research Initiative (NERI). Information about the program is 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 the same year that NERI was established, entitled *Powerful Partnerships: The Federal Role in International Cooperation on Energy Innovation*. This report promoted "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..."

I-NERI was established in FY 2001 in response to PCAST recommendations. 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, while also providing the United States greater credibility and influence in international activities associated with the application of nuclear technologies. I-NERI is fully integrated into the principal research programs of DOE's Office of Nuclear Energy (NE), including Generation IV Nuclear Energy Systems Initiative (Gen IV), Global Nuclear Energy Partnership (GNEP), the

Advanced Fuel Cycle Initiative (AFCI), and the Nuclear Hydrogen Initiative (NHI). This integration makes NE's entire advanced nuclear R&D available for international collaboration on a cost-shared basis.

To date, DOE has implemented bilateral I-NERI collaborative agreements with the following international partners:

1) Commissariat à l'énergie atomique (CEA) of France
2) Republic of Korea Ministry of Science and Technology (MOST)
3) The Nuclear Energy Agency (NEA) of the Organisation for Economic Co-operation and Development (OECD)
4) European Atomic Energy Community (EURATOM)
5) Department of Natural Resources Canada (NRCAN) and Atomic Energy of Canada Limited (AECL)
6) Brazilian Ministério da Ciência e Tecnologia (Ministry of Science and Technology – MST)
7) Agency of Natural Resources and Energy (ANRE) and the Ministry of Education, Culture, Sports, Science, and Technology (MEXT) of Japan.

Since the program's inception, a total of 80 projects have been initiated:

- 19 with France
- 32 with the Republic of Korea
- 15 with the European Union
- 9 with Canada
- 2 with Brazil
- 2 with Japan
- 1 with OECD

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 bilateral and multilateral collaboration with international agencies and research organizations to improve the development of nuclear energy
- To promote and maintain a nuclear science and engineering infrastructure to meet future technical challenges

Through the I-NERI program, 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 nuclear-hydrogen 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 R&D under Generation IV, AFCI, and NHI with member countries of the Generation IV International Forum (GIF). The 13 charter countries of the GIF include Argentina, Brazil, Canada, China, EURATOM, France, Japan, Republic of Korea, Russia, South Africa, Switzerland, and the United Kingdom, and the United States. China signed the Framework Agreement to pursue international collaboration this year, adding to the seven previous signatories: Canada, the European Union, France, Japan, the Republic of Korea, Switzerland, and the United States. The GIF member countries have established multilateral agreements to conduct R&D in four areas. Through the Department of Energy, the U.S. became a partner in the System Arrangements for sodium fast reactors and very high-temperature reactors in February 2006 and November 2006, respectively.

In the meantime, I-NERI's bilateral agreements enable U.S. researchers to establish international R&D collaborations to begin developing next-generation nuclear energy systems. Negotiations to establish new bilateral agreements are underway with the Republic of South Africa and the United Kingdom.

Extensive multilateral agreements are in place with the 21 countries participating in the Global Nuclear Energy Partnership (GNEP). AFCI is the domestic component of GNEP, through which the U.S. and collaborating nations are developing and demonstrating the technology to close the nuclear fuel cycle.

GNEP Membership

Australia
Bulgaria
Hungary
Canada
China
France
Ghana
Italy
Japan
Jordan
Kazakhstan
Lithuania
Poland
Republic of Korea
Romania
Russia
Senegal
Slovenia
Ukraine
United Kingdom
United States

3.4 Program Organization and Control

DOE-NE manages the I-NERI program. 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 (NE-3), with the support of the technical Program Directors and Country Managers, administers the selection and funding of awards. The Idaho Operations Office (ID) negotiates and monitors the cooperative agreements with U.S. entities. Once projects are initiated, the Office of Gas Reactor Deployment and the Office of 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 United States appoints a scientist from the national laboratories to serve as technical coordinator for each of the R&D areas of the agreement. A corresponding Program Manager (Generation IV and NHI) or Campaign Manager (AFCI), drawn from the ranks of DOE-NE headquarters staff, provides management support for each of these research areas under the Program Director. There are four National Technical Directors (NTDs), representing each technology area of the Generation IV program (systems analysis, fuels, materials, and energy conversion) and seven Campaign Directors (CDs) addressing development needs for the AFCI program in the areas of transmutation fuel, separations technology, fast reactors, safeguards, waste forms, systems analysis, and grid-appropriate reactors. The NHI program has three Technical Directors (TDs) for its major research areas (thermochemical cycles, high-temperature electrolysis, and systems interface).

System Integration Managers (SIMs) assist the NTDs by managing the technologies for each of the Generation IV reactor concepts that DOE participates in:

1) Very High-Temperature Reactor (VHTR)
2) Supercritical Water Reactor (SCWR)
3) Gas Fast Reactor (GFR)
4) Lead Fast Reactor (LFR)
5) Sodium Fast Reactor (SFR)

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, TDs, and CDs 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.

3.5 Funding

I-NERI is an important vehicle for enabling international R&D in Generation IV, AFCI, 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.

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 participants have committed a total research and development investment of $192 million: $104 million contributed by the U.S. and $88 million by international collaborators. International investment is:

- $15.2 million from Canada
- $30.2 million from France
- $25.1 million from the Republic of Korea
- $2.7 million from Japan
- $12.5 million from the European Union
- $2.3 million from Brazil

Funding provided by the United States may only be spent by U.S. participants. I-NERI projects typically last three years and are funded annually by the Generation IV, AFCI, 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, AFCI, 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, TDs, and CDs 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, AFCI, 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 GIF member countries, the following six reactor concepts were deemed most promising: the gas-cooled fast reactor, lead-cooled fast reactor, molten salt reactor, sodium-cooled fast reactor, supercritical water-cooled reactor, and the very high temperature reactor. Current U.S. research priorities are focused on the VHTR and SFR.

Generation IV has eight technology goals:

1) 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
2) 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
3) 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 off-site 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 Initiative. The AFCI 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. 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 comprise the domestic R&D component of the Global Nuclear Energy Partnership program and may also be of value to the Generation IV nuclear energy systems initiative. GNEP is an international cooperation that seeks to expand the peaceful use of nuclear energy through the development of advanced fuel cycle technologies that reduce the risk of proliferation.

Research under this initiative focuses on recycling, fuel treatment, and conditioning technologies that have the potential to dramatically reduce the quantity, radioactivity, and thermal content of spent nuclear fuel materials requiring geological disposal, thus greatly expanding a repository's effective capacity.

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. The goal of the NHI program is to demonstrate the commercial-scale, economic viability of hydrogen production 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.

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 or the consumption of fossil fuel resources.

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, 80 projects have been awarded, 50 of which have been completed.

4.1 Programmatic Accomplishments

Following is a brief description of the program's primary accomplishments from FY 2001 through FY 2006, an overview of accomplishments during FY 2007, and planned accomplishments for the upcoming fiscal year.

Historical Accomplishments (FY 2001-2006). 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 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.

In FY 2006, 14 new research projects were initiated under the agreements with Japan, EURATOM, Brazil, France, and the Republic of Korea. Twelve projects that had begun in FY 2004 were completed as well as one from each of the following years: FY 2003, FY 2005, and FY 2006.

FY 2007 Accomplishments. During FY 2007, nine new research projects were awarded: seven with the Republic of Korea and two with Canada. Nineteen projects that had begun in 2004 and 2005 were completed this year. The following summarizes noteworthy accomplishments during the past year.

- Completed 19 research projects with five I-NERI collaborators:
  - Canada (3)
  - Euratom (5)
  - France (5)
  - Japan (1)
  - Republic of Korea (5)
- Initiated 9 new collaborative research projects:
  - Canada (2)
  - Republic of Korea (7)
- Conducted annual project performance reviews and bilateral program planning meetings during FY 2007 with:
  - Republic of Korea
  - France
  - Canada

FY 2008 Planned Activities. DOE plans the following international activities:

- Initiate new cooperative projects under existing agreements
- Conduct annual project review/bilateral meetings with international partners (France, Korea, Euratom, Canada)
- Continue pursuing new cooperative agreements with South Africa and other prospective partner countries

Collaborator	FY 01	FY 02	FY 03	FY 04	FY 05	FY 06	FY 07	Total
France	4	1		11		3		**19**
Republic of Korea		6	5	6	4	4	7	**32**
OECD-NEA		1						**1**
EURATOM				8	2	5		**15**
Canada				7			2	**9**
Brazil					1	1		**2**
Japan					1	1		**2**
Total	**4**	**8**	**5**	**32**	**8**	**13**	**9**	**80**

Table 1. Number of I-NERI projects awarded.

4.2 Current I-NERI Collaborations

Following are brief descriptions of the current I-NERI collaborations. 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. (Prior to 2004, all of the projects were related strictly to the Generation IV initiative.) Figure 3 shows the international portion of I-NERI project funding from 2001-2007. Figure 4 shows the number of projects for each international collaborating country for the same period.

Brazil. Cooperative research projects with the Brazilian Ministério da Ciência e Tecnologia (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 with 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 and advanced fuel cycles. The collaborating agency in Canada is Atomic Energy of Canada Limited (AECL).

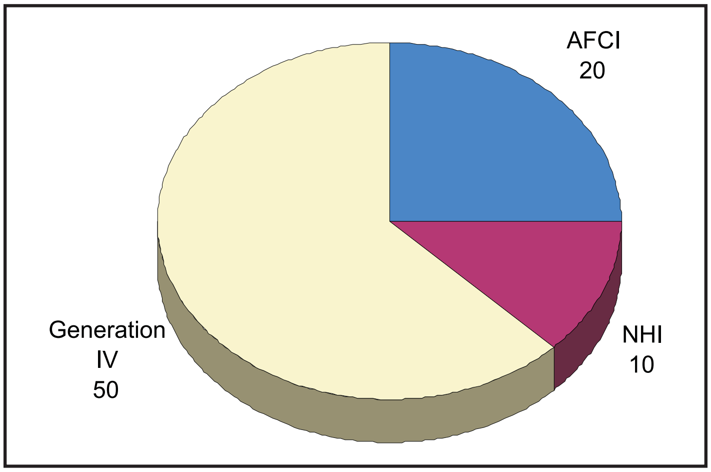


Figure 2. Project distribution by program area.

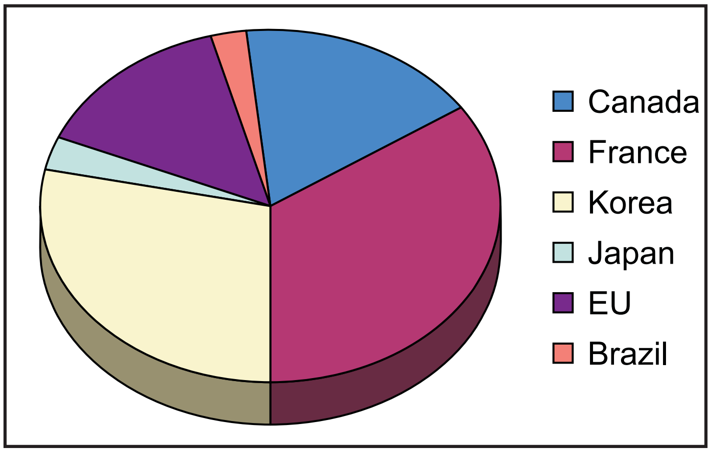


Figure 3. Total Budget Share by Collaborating Country (2001-2007).

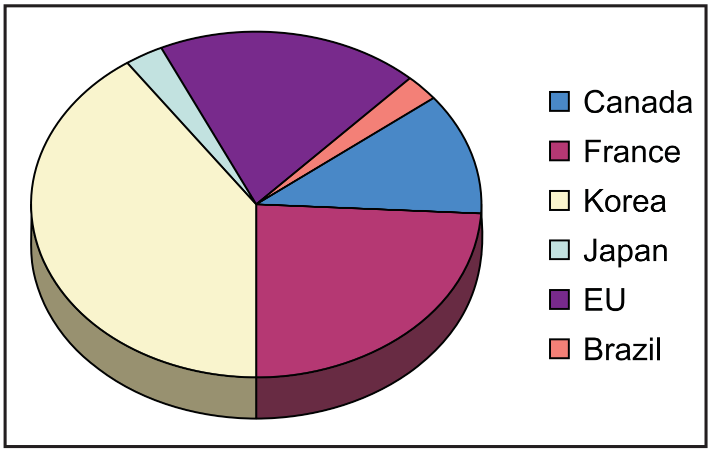


Figure 4. Number of Projects by Collaborating Country (2001-2007).

European Union. The collaborating agency for Europe is the European Atomic Energy Community (EURATOM), an international organization comprised 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'énergie 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 Japan's Agency of Natural Resources and Energy (ANRE) 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 the Japanese Ministry of Education, Culture, Sports, Science, and Technology (MEXT) signed an Implementing Arrangement concerning cooperation in innovative nuclear reactors, processing, and fuel 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 fuel cycles and materials, gas-cooled and supercritical water-cooled reactors, sodium-cooled fast reactors, nuclear hydrogen production, and next-generation reactor and fuel cycle technology. Research projects for collaboration have been selected competitively through an independent peer-evaluation process.

OECD. The U.S. teamed with the 28-member Nuclear Energy Agency, a specialized group within the Organisation for Economic Co-operation and Development (OECD), to conduct reactor materials experiments and associated analyses involving concrete-fuel interactions and core cooling.

4.3 Program Participants

Following is a complete list of the I-NERI program participants (Table 2) and their affiliation (Figure 5). In addition, this section presents the level of U.S. university student participation in active I-NERI projects during 2007.

U.S. Student Participation. One of the I-NERI program goals is to spur development of nuclear-related education and research opportunities at U.S. universities. As noted in Figure 5, a total of 16 universities and colleges participated in I-NERI research projects during FY 2007. Approximately 62 students from these institutions worked on active I-NERI research projects during the year, distributed by degree level as shown in Figure 6.

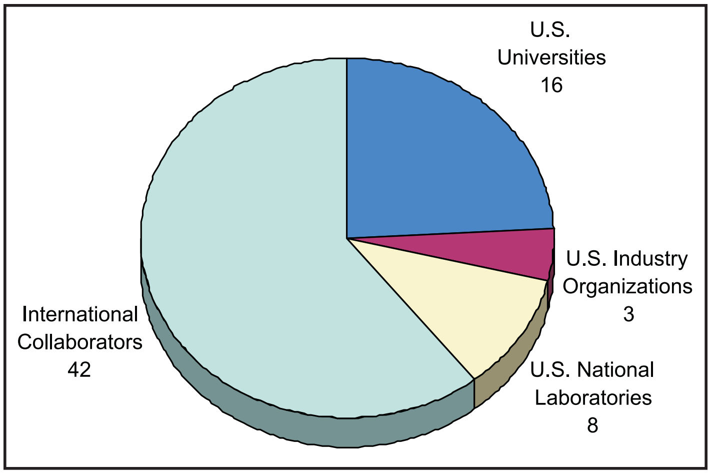


Figure 5. I-NERI participant profile.

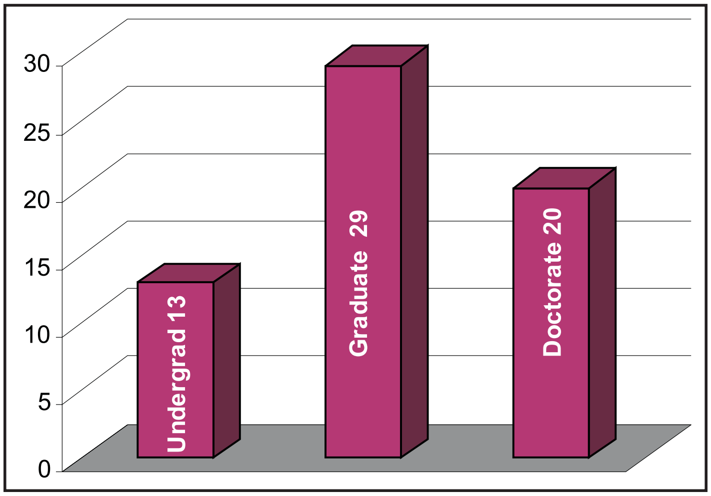


Figure 6. 2007 I-NERI U.S. student participation profile.

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

- Gas Technology Institute
- General Atomics
- Westinghouse Electric

Universities

- Iowa State University
- Massachusetts Institute of Technology
- Ohio State University
- Pennsylvania State University
- Purdue University
- Rensselaer Polytechnic Institute
- University of California-Santa Barbara
- University of Florida
- University of Idaho
- University of Illinois at Chicago
- University of Maryland
- University of Michigan
- University of Nevada-Las Vegas
- University of Notre Dame
- University of Wisconsin
- Utah State University

International Collaborators

Industry Organizations

- Atomic Energy of Canada Limited
- Eletronuclear
- Framatome-ANP
- Gamma Engineering
- Hitachi Works
- Hitachi, LTD
- Institute for Reference Materials and Measurement
- Korea Hydro and Nuclear Power Company
- Società Informazioni ed Esperienze Termoidrauliche
- Toshiba Corporation

Universities

- Cheju National University
- Chosun University
- Chungnam National University
- École Polytechnique de Montréal
- Hanyang University
- Korea Maritime University
- Pusan National University
- Seoul National University
- Tohoku University
- Université Bordeaux
- Université de Sherbrooke
- University of Manchester
- University of Manitoba
- University of Ontario Institute of Technology
- University of Tokyo

Governmental Organizations

- Chalk River Laboratories
- Commissariat à l'énergie 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 Centre-Institute for Transuranium Elements (ITU)
- Joint Research Centre-Institute for Energy
- Japan Atomic Energy Agency (JAEA)
- Korea Advanced Institute of Science and Technology (KAIST)
- Korea Atomic Energy Research Institute (KAERI)
- Korea Electric Power Research Institute (KEPRI)
- Laboratoire des Composites Thermostructuraux (LCTS)
- Ministério da Ciência e Tecnologia
- Organisation for Economic Co-operation and Development-Nuclear Energy Agency (OECD/NEA)

Table 2. I-NERI participating organizations.

4.4 Completed I-NERI Projects

This year marked the completion of 19 I-NERI research projects—17 of which were initiated in FY 2004 and two in FY 2005.

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 entities have resulted in significant scientific and technological enhancements in the global nuclear power arena. The international collaborations have forged lasting ties that will continue to promote 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, AFCI, 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.

Table 3 identifies the 19 projects that were completed in 2007. More detailed information on each can be found in the project summaries provided in subsequent sections of this report.

Project Number	Title	Lead Organization
2004-003-C	Evaluation of Materials for Supercritical Water-Cooled Reactors	Oak Ridge National Laboratory
2004-004-C	ACR Hydrogen Production for Heavy Oil Recovery	Idaho National Laboratory
2004-007-C	Thermochemical Hydrogen Production Analysis	Argonne National Laboratory
2004-003-E	Lead-Cooled Fast Reactor Engineering and Analysis	Argonne National Laboratory
2004-005-E	Characterization of Nuclear Waste Forms and Their Corrosion Products	Argonne National Laboratory
2004-006-E	Nitride Fuel Fabrication Research	Los Alamos National Laboratory
2004-009-E	Use of an Ionization Chamber in Fission Cross-Section Measurements	Los Alamos National Laboratory
2004-010-E	Nuclear-Assisted Hydrogen Storage and Safety Issues	Argonne National Laboratory
2004-001-F	Hydrogen Process to High-Temperature Heat Source Coupling Technology	Idaho National Laboratory
2004-002-F	OSMOSE—An Experimental Program for Improving Neutronic Predictions of Advanced Nuclear Fuels	Argonne National Laboratory
2004-003-F	Thermal-Hydraulic Analyses and Experiments for GCR Safety	Idaho National Laboratory
2004-007-F	Evaluation of Materials for Gas-Cooled Fast Reactors	University of Wisconsin
2004-011-F	Thermochemical Hydrogen Production Process Analysis	Argonne National Laboratory
2005-001-J	Development of Materials for Supercritical Water-Cooled Reactor	University of Michigan
2004-001-K	Screening of Gas-Cooled Reactor Thermal-Hydraulic and Safety Analysis Tools and Experiment Database	Argonne National Laboratory
2004-002-K	Investigation of Heat Transfer in Supercritical Fluids for Application to the Generation IV Supercritical-Water Cooled	Idaho National Laboratory
2004-004-K	Development of Voloxidation Process for Treatment of LWR Spent Fuel	Idaho National Laboratory
2004-005-K	Development and Test of Cladding Materials for Lead-Alloy Cooled Transmutation Reactors	Los Alamos National Laboratory
2005-002-K	Development of HyPEP, A Hydrogen Production Plant Efficiency Calculation Program	Idaho National Laboratory

Table 3. I-NERI projects completed in 2007.

5.0 U.S./Brazil Collaboration

The U.S. Department of Energy (DOE) and the Brazilian Ministério da Ciência e Tecnologia 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 project continued during FY 2007, with no new projects awarded. A listing of the I-NERI U.S./Brazil project that is currently underway follows, along with a summary of the accomplishments achieved in FY 2007.

Directory of Project Summaries

INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE

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

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

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

Collaborator: Westinghouse Electric Company

Project Number: 2005-001-B

Program Area: AFCI

Project Start Date: March 2005

Project End Date: March 2008

Research Objective

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. As no systematic assessment of I&C requirements for IPSRs was available when this project began, performing a detailed review of the instrumentation requirements 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 waveguide-based level measurement technique and 2) application of advanced signal processing algorithms to a cooled-fluid-based, lance-type probe.

An additional objective of this work is to assess areas in plant operation and control where unique IPSR features and operating modes require innovative approaches. The medium size and modularity of the IPSR provide economic incentives for deploying 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 vary co-generation with hourly changes in 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. The hierarchical control development task will maximize the utilization efficiency of the power-park resources while minimizing staffing requirements.

The final technical area of this project is to develop guidelines for operator interaction with the plant controls and protection systems. Advanced IPSR designs, particularly the Westinghouse International Reactor Innovative & Secure (IRIS), are unique in that they are capable of responding to almost any operational condition or accident without operator intervention. The large thermal inertia of their primary system gives these reactors long transient evolution times. Due to the advantageous thermal characteristics of IPSRs compared to external loop LWRs, the operator's 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.

Research Progress

A primary project focus during FY 2007 was developing prototype instrumentation to provide accurate and reliable measurement of in-vessel coolant levels. Two candidate systems are currently under development: 1) an ultrasonic, torsional waveguide-based level measurement technique and 2) a cooled lance-type liquid-level probe featuring advanced signal processing to yield continuous liquid-level measurement. Project staff developed an engineering test grade experimental prototype for the ultrasonic torsional wave based level measurement system and demonstrated system functionality in a laboratory environment.

Development efforts had two main thrusts: 1) redesign mechanical components for optimal performance and 2) refine electronics to minimize noise while maximizing the signal gain. All of the required mechanical components and the in-vessel electrical interconnections were fabricated from materials suitable for the IPSR vessel environment. Presently, the system is undergoing final assembly and checkout before high-temperature, high-pressure testing. The IPSR test facility has been designed and fabricated, and is in process of installation and testing. The test chamber consists of a code-rated pressure vessel with external heaters and a control system to allow comparison water level measurement. Figure 1 shows representative ultrasonic reflection data acquired with the prototype level measurement system in a laboratory environment. The return time of the torsional signal (peak at approximately 2.5 ms in Figure 1) varies linearly with the depth of probe immersion. The grey plane in the figure highlights the frequency used for comparison measurements.

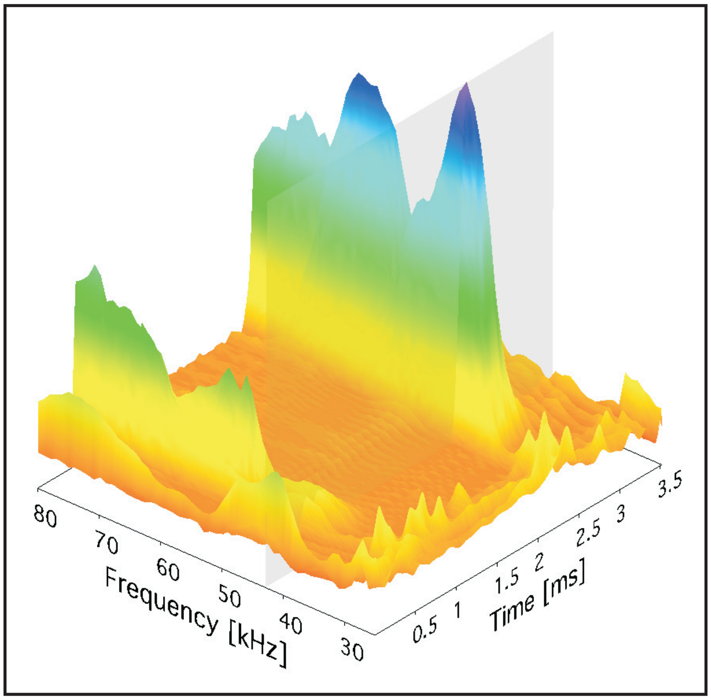


Figure 1. Ultrasonic reflection signal for prototype level measurement system.

The second task focuses on creating a hierarchical, supervisory control scheme based on the IRIS reactor. The main effort of this task has been to accommodate multiple modules and products interconnected through the feedwater and steam systems. Researchers developed the technical basis for a hierarchical control system which 1) subdivides the component systems to make the control problem tractable and 2) coordinates at a supervisory control level in order to maintain stability of the combined system while meeting system performance specifications.

A central focus in support of the overall control system design task has been developing the mathematical model representing the heat transfer and fluid flow dynamics of the IRIS helical coil steam generator. The model applies the moving boundary approach to track the points of transition between heat transfer regimes on the secondary side. The major regimes of heat transfer in the helical coil steam generator are the subcooled, nuclear boiling, film boiling, and superheat regimes. The remainder of the model for the primary and tube metal is built around the four control volumes separated by these three moving boundaries. The model is successful with a relatively small number of control volumes because the distributed parameters vary smoothly within each region, leading to very straightforward integral average formulations for heat transfer, friction pressure loss, and void fraction. The integral average formulations are all based on the steady-state distributions of temperature, steam quality, and pressure drop along the tube. Consequently, the calculations are very accurate near steady state and give an excellent approximation for transients in which the distributed parameters remain close to the steady state distribution. The model is appropriate for normal operation, control studies, and other mild upsets. The simulation is capable of operating over the full range of power.

Planned Activities

During the final project months, the team will package and demonstrate an engineering prototype ultrasonic torsional level measurement device and demonstrate the device 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.

6.0 U.S./Canada Collaboration

The Director of 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

Three of the research projects initiated in FY 2004, the initial year of the collaboration, were completed in FY 2007; one of this group remains active. Two new projects were awarded during the past 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 2007.

Directory of Project Summaries

INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE

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

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

PI (Canada): P. Boczar, Atomic Energy of Canada Limited (AECL)

Collaborators: University of Florida, Los Alamos National Laboratory (LANL), Brookhaven National Laboratory

Project Number: 2004-002-C

Program Area: AFCI

Project Start Date: October 2004

Project End Date: September 2008

Research Objectives

The objective of this project is to develop inert matrix fuels (IMFs) for the stabilization or burnup of plutonium (Pu) in existing commercial power reactors. IMFs offer the advantage of more efficient destruction of plutonium 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_2), 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, IMFs 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. Figure 1 shows UO_2 in a zirconium (Zr) matrix.

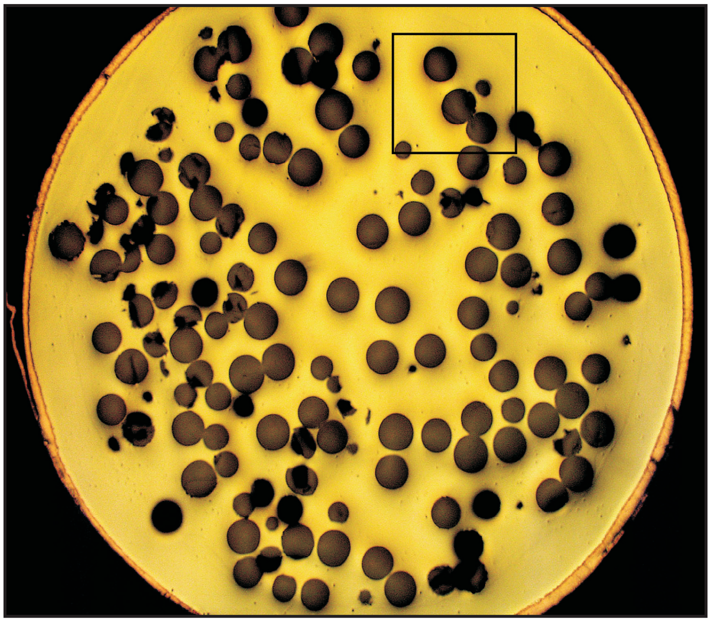


Figure 1. UO_2 particles extruded in a Zr-metal matrix.

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

Progress over the past year of this investigation can be summarized as follows:

Synthesis and Characterization. The specific heat capacity of the composite was measured using a differential scanning calorimeter. When the measured specific heat capacity was compared to a value calculated from the rule of mixtures, researchers found that the calculation was a poor approximation for the measured specific heat capacity. Three batches of composite material were synthesized using each of four different processing methods (ball milling, magnetic bar stirring, mortar and pestle, and Spex milling), then three pellets fabricated from each batch. Figure 2 shows a typical pellet. Thermal diffusivity results indicate that processing may affect the *intra*-batch, but not the *inter*-batch sample variation. Each processing method can be characterized by an average thermal conductivity that may not vary significantly across a population of samples. The thermal diffusivity was also measured on the pellets fabricated for the sintering kinetics study. There is evidence that as the microstructure evolves, there is an effect on the thermal conductivity of the samples. As the sintering temperature and time increased from 1,550°C to 1,650°C, and from 8 hours to 24 and 100 hours, respectively, there was a corresponding drop in thermal conductivity.

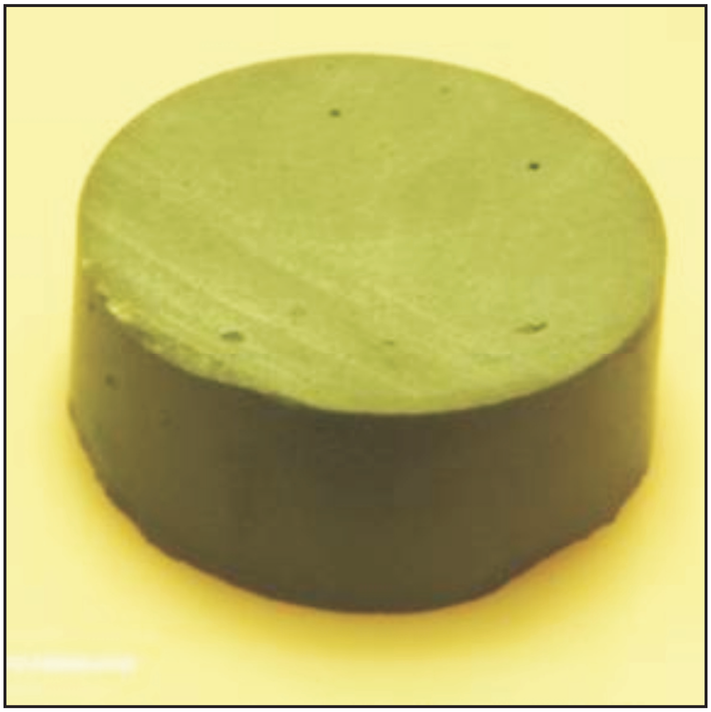


Figure 2. Photograph of dual-phase $MgO-ZrO_2-PuO_2$ pellet.

Spinel Candidate Material Selection. The Mg_2SnO_4 sample previously irradiated by 1 MeV Kr^{2+} ions at 50 K and fluence of 5×10^{19} ions/m^2 was reexamined at room temperature using transmission electron microscopy (TEM) and energy dispersive spectroscopy (EDS). After thermal recovery, the amorphous sample turned polycrystalline. Selected area diffraction (SAD) patterns indicate that the crystal structure for major inner grains is spinel. EDS results and bright field (BF) images indicate that phase separation occurred along the edge of the focused ion beam (FIB) sample and the thickness of this fringe is about 1 µm. The researchers performed a TRIM calculation and converted the amorphization doses into displacement per atom (dpa). In the simulation, 40 eV displacement threshold, Ed, energy was chosen for all atoms in the Mg_2SnO_4 target. Researchers then calculated the corresponding amorphization dose at 50 and 150 K to be 6.25 and 12.49 dpa, respectively. To simulate fission fragment effects such as swelling, follow-up collaboration with LANL is being explored in order to generate a heavy ion source at fission energy levels by using a high-energy proton source (800 MeV) to bombard a bismuth foil.

Corrosion Testing. The $MgO-Nd_2Zr_2O_7$ sample with 70 volume percent MgO made by gel-casting was tested at 300°C and saturation pressure for 10 days. Due to the irregular shape of the sample, only a qualitative experiment was conducted. The sample withstood hydration induced stresses and did not fracture, which shows improved hydration resistance compared with the ball mill produced $MgO-Nd_2Zr_2O_7$ composite, which fractured after one hour exposure to 300°C deionized water.

Thermophysical Property Simulation. The researchers developed a general computer code for the generation of MgO-NDZ composites of various morphologies. This code generates structures in a manner similar to that used for polycrystals, as described in previous reports. A systematic study of $MgAl_2O_4$ spinel and structures with 25, 50, 75, and 100 percent cation inversion has shown that thermal expansion is independent of the degree of inversion. Simulations to determine the dependence of thermal conductivity on composite microstructure and the degree of inversion are underway.

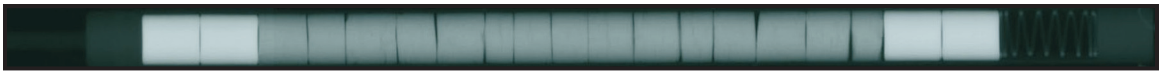


Figure 3. Neutron radiograph of 94%dUO_2–6% RGPuO_2–<1% NpO_2 LWR-1a MOX rodlet irradiated to 9 GWd/tU burnup.

Planned Activities

This project was predicated on the use of thermal reactor spectra for the transmutation of minor actinides. In February 2006, the Department of Energy decided to focus efforts on the transmutation of minor actinides in fast spectrum systems, thereby eliminating the need for separation of specific actinide streams. With this decision, the work packages and authorizations for research of thermal spectrum transmutation have been redirected to fast reactor transmutation and all work on thermal transmutation terminated. Currently, this agreement is being renegotiated to redirect focus of research to fast reactor transmutation fuels.

INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE

Evaluation of Materials for Supercritical Water-Cooled Reactors

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

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

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

Project Number: 2004-003-C

Program Area: Generation IV

Project Start Date: October 2004

Project End Date: September 2007

Research Objectives

The goal of this project was to establish candidate materials for supercritical water-cooled reactor (SCWR) designs and to evaluate their mechanical properties, dimensional stability, and corrosion resistance. This project addressed critical issues related to radiation stability, corrosion, and stress corrosion cracking performance in candidate materials for 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 up to 1,026 hours. They applied surface modifications and grain boundary engineering (GBE) treatments 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 for austenitic stainless steels than F/M steels. Nickel-based alloys showed fairly good corrosion resistance in an SCW environment and had the lowest weight gain of all the tested materials. A 9Cr oxide-dispersion strengthened (ODS) steel showed the lowest weight gain among the F/M materials.

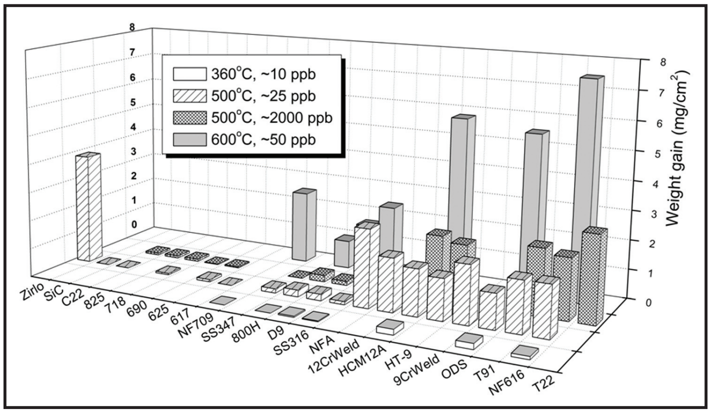


Figure 1. Comparison of weight gain data for the candidate materials after exposure to subcritical/supercritical water with different oxygen concentrations for ~333 h.

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 YFeO 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.

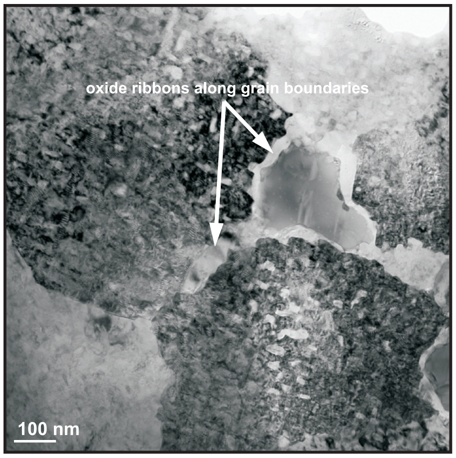


Figure 2.TEM image showing grain morphology and boundaries in the internal oxidation layer formed in 9Cr ODS steel after 1,026 hours exposure to 500°C SCW.

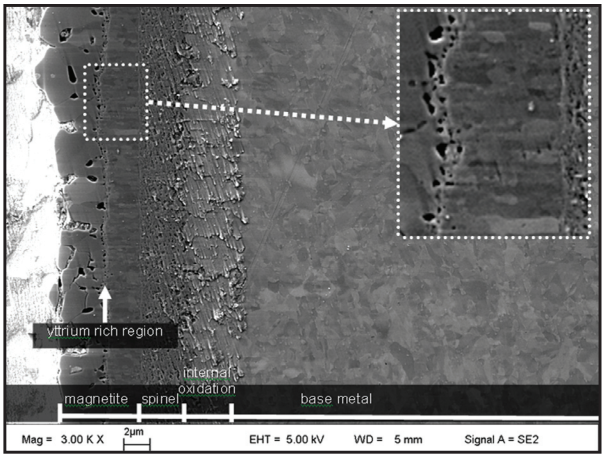


Figure 3. SEM image showing the magnetite layer microstructure in 9Cr ODS steel coated with a thin yttrium film after exposure to 500°C SCW for 667 hours.

Austenitic alloy 800H showed low weight gain in SCW; however, it suffered from extensive spallation of its outer magnetite scale. The researchers performed 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.

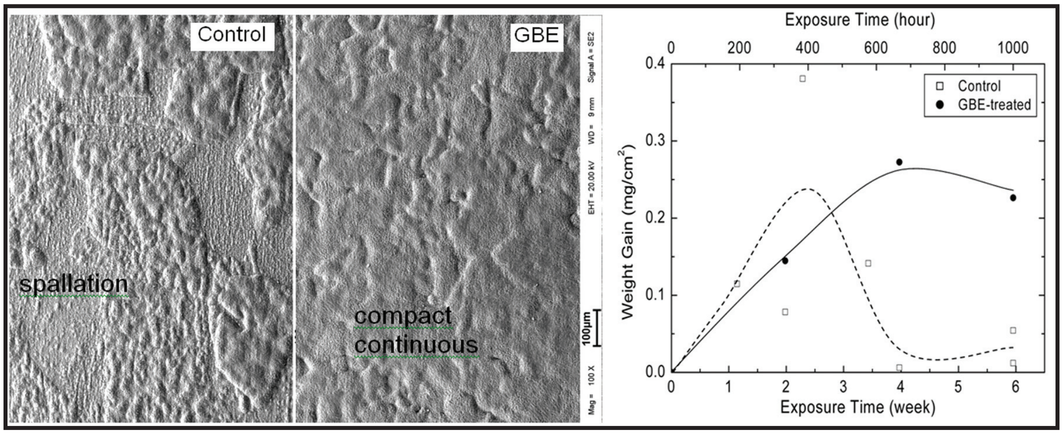


Figure 4. Effect of GBE treatment on the corrosion resistance of austenitic alloy 800H after exposure to SCW at 500°C.

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 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 μS/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 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.

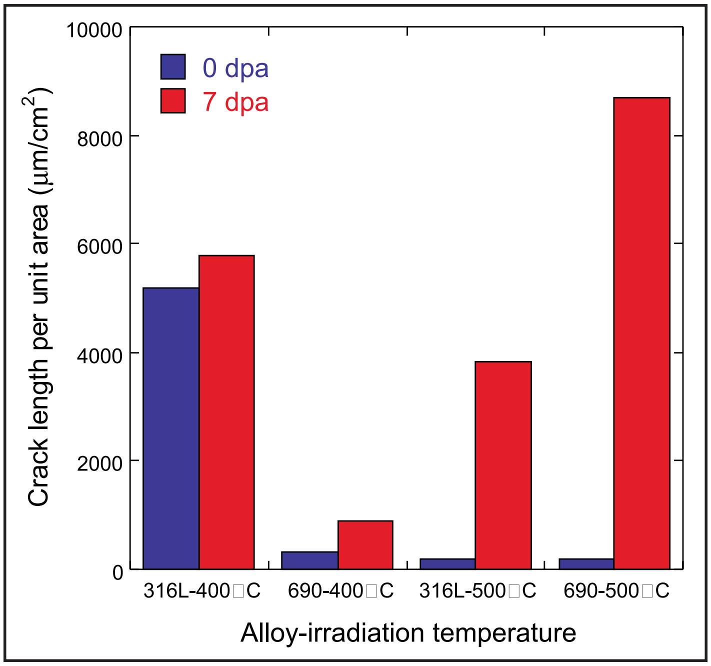


Figure 5. Influence of irradiation on the cracking susceptibility of alloy 316L and 690 in 400°C and 500°C SCW.

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 1,149 and 982°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.

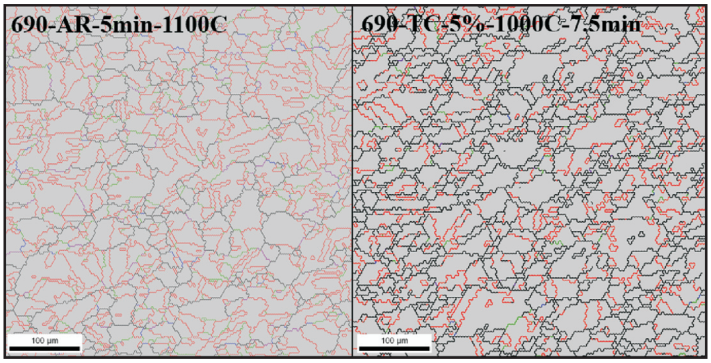


Figure 6. Comparison of two alloy 690 conditions showing large differences in CSLB fraction: left – 76%, right – 32%.

Work continued to characterize the corrosion rates of candidate alloys. Initial test results showed a rapid increase in corrosion rates above 550ºC, 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.

Researchers re-started work on the deposition of anti-corrosion coatings with the atmospheric pressure plasma jet, concentrating on handling the metallo-organic 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, dip-coating 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, 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/cm^3), as shown in Figure 7. Calculations of the ratio of "initial" H atom yields to 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/cm^3. Discrepancies between experimental data and the Monte Carlo simulation results indicate that SCW radiolysis is dominated by the gas-phase component.

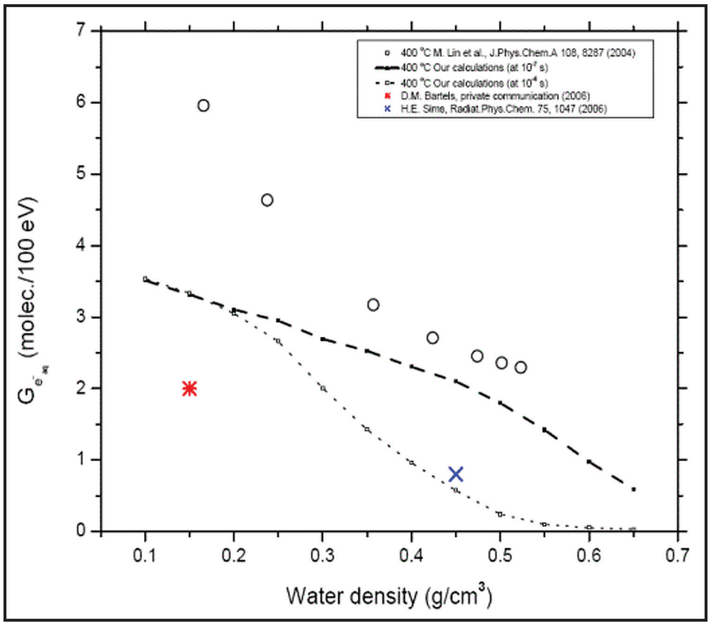


Figure 7. Comparison between Monte-Carlo predictions and experimental data.

Planned Activities

This project has been completed.

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)

Collaborators: Chalk River Laboratories

Project Number: 2004-004-C

Program Area: NHI

Project Start Date: June 2004

Project End Date: May 2007

Research Objectives

The objective of this project was 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. Researchers investigated the use of a high-temperature electrolysis (HTE) process coupled to the ACR, which 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 operating temperature of 800–900°C.

In this project, researchers developed 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 assessed the costs of hydrogen and steam production for comparing the economics of low-temperature electrolysis and other means of hydrogen production. The economic analysis included an evaluation of hydrogen and steam production using fossil fuels in the Athabasca oil sands project under construction in Alberta, Canada, considering the price of petroleum recovered.

Research Progress

The researchers have accomplished the following activities:

- Incorporated a 1-D integral electrolyzer model into a HYSYS ACR plant model coupled to a low-temperature electrolysis (LTE) 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 design 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.

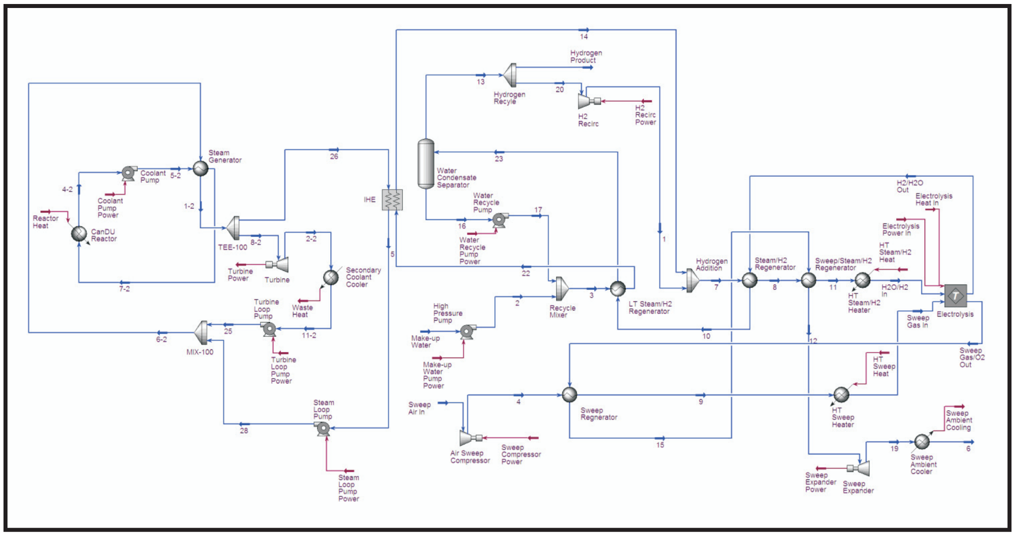


Figure 1. HYSYS process flow diagram for an ACR-700 HTE plant with air sweep.

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, one-dimensional, 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. Results obtained indicated overall thermal-to-hydrogen efficiencies about 33–34 percent, with only a weak dependence on current density. These values compare favorably with corresponding overall thermal-to-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.

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

This I-NERI project has concluded.

INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE

Thermochemical Hydrogen Production Process Analysis

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, University of Ontario Institute of Technology (UOIT)

Project Number: 2004-007-C

Program Area: NHI

Project Start Date: June 2004

Project End Date: May 2007

Research Objectives

The objective of this project is to estimate the cost of hydrogen production using the low-temperature, copper-chloride (Cu-Cl) cycle. The steps necessary to meet this objective are the following: 1) complete proof-of-principle experiments for the thermal reactions that demonstrate high yields and relatively fast kinetics; 2) complete proof-of-principle experiments for the proposed electrolysis reactions; 3) develop a conceptual process design and calculate the efficiency of the process using Aspen and performance targets; 4) use the Aspen flowsheet as the basis for the H2A cost analysis. Current experimental and modeling activities are designed to more accurately evaluate the Cu-Cl cycle and solve the technical challenges that are observed in the proof-of-principle work. The process design and the cost estimates are to be optimized and refined as work progresses.

Research Progress

Hydrolysis reaction. During this research period, work focused on further understanding the hydrolysis reaction,

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

One technical challenge is the requirement for excess steam to produce high yields of the Cu_2OCl_2 and hydrogen chloride (HCl) compounds. The amount of excess water required to accomplish this is quite high. Figure 1 shows the effect of temperature and the steam-to-copper molar ratio on the Cu_2OCl_2 yield. The molar steam-to-copper ratio must be about 17 for high yields below 450°C, according to the Aspen model. A second challenge is the presence of a parasitic reaction in which $CuCl_2$ decomposes to give CuCl and chlorine. The Aspen model predicts that the decomposition reaction should be very small, as shown in Figure 2. However, experimental data indicates that the decomposition reaction is significant and the amount of CuCl in the solid product can be as high as 25 weight-percent of the total product.

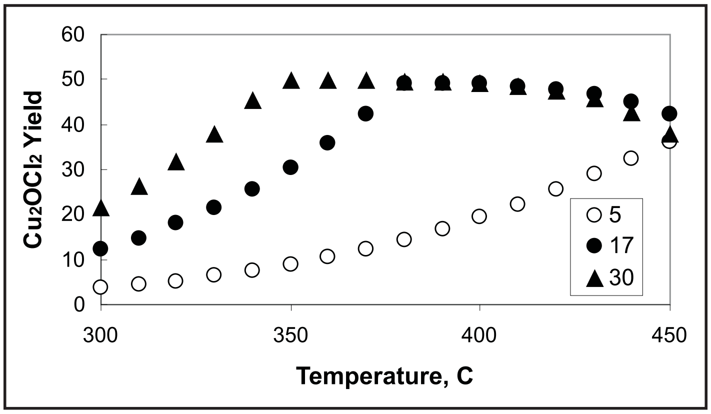


Figure 1. Effect of temperature and three steam-to-copper molar ratios (5, 17, and 30) on the yield of Cu_2OCl_2.

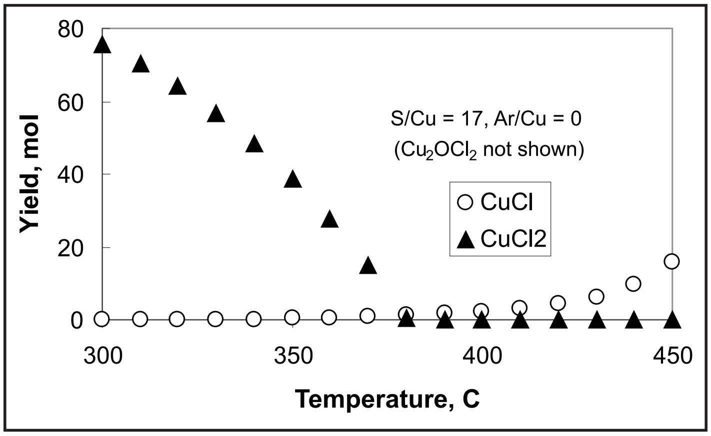


Figure 2. Effect of temperature on $CuCl_2$ decomposition.

Work has concentrated on the reactor design for the hydrolysis reaction. Researchers studied the $CuCl_2$ hydrolysis reaction,

$$2CuCl_2\ (s) + H_2O\ (g) \Leftrightarrow Cu_2OCl_2\ (s) + 2HCl\ (g),$$

using two types of fixed bed reactors: fluidized bed and rotary. The objective was to determine how the reactor design influenced both the hydrolysis reaction and the competing $CuCl_2$ decomposition reaction, $2CuCl_2$ (s) $\Leftrightarrow$ 2CuCl (s) + Cl_2 (g).

Some results of the fixed bed experiments are:

1) Tests with dehydrated $CuCl_2$ showed 17 to 27 weight-percent CuCl in the solid product. The Cu_2OCl_2 yield increased with increasing flow rates (50 to 200 ml/min) of the humidified carrier gas, but was still relatively low at 48 to 75 percent. These tests used reactor Design A shown in Figure 3.

2) The next set of six tests used hydrated, sieved 150-250 μm $CuCl_2$ particles and a 200 ml/min flow rate of argon (Ar), also with reactor Design A. The amount of CuCl in the solid products varied between 8 and 16 percent. Researchers hypothesized that the bed porosity increased due to the release of the water of hydration in situ. This, coupled with the high Ar flow rate, resulted in improved mass transfer. All but one of the samples contained 80-89 weight-percent Cu_2OCl_2. The variation is due to differences in test duration and steam/Cu molar ratio. The amount of CuCl varied from 8 to 16 weight-percent, nearly a two-fold reduction compared to the tests with dehydrated $CuCl_2$.

The conclusion from these two sets of tests was that the Ar flow rate is critically important for high yields of the desired product and that hydrated $CuCl_2$ probably provides increased porosity in the bed.

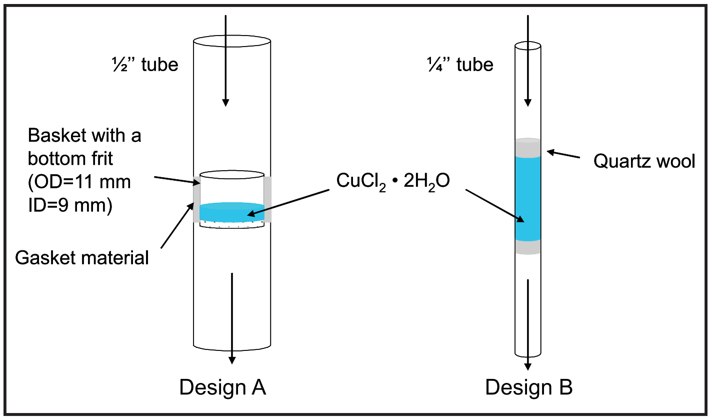


Figure 3. Schematic of two types of fixed bed reactors.

The reactor was then changed to Design B, as shown in Figure 3, which had a length-to-diameter (L/D) bed ratio ten times larger to improve mass transfer. Researchers measured how changes in various parameters such as particle size, test temperature, and test duration affected CuCl formation. Because of the high costs, the products were only analyzed for CuCl. However, $CuCl_2$ is brown and Cu_2OCl_2 is black, enabling a qualitative estimate of the conversion to Cu_2OCl_2.

In general, tests with smaller particles, lower test temperatures, larger steam-to-copper molar ratios, and shorter durations showed less CuCl formation in the solid product. All of these tests also showed a significant amount of unreacted brown material, or $CuCl_2$, suggesting poor mass and heat transfer. The black Cu_2OCl_2 material tended to be free flowing and located near the walls of the reactor and on the top of the sample, as shown in Figure 4, while the $CuCl_2$ was usually aggregated.

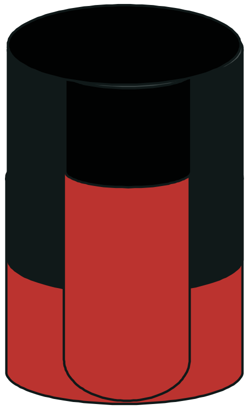


Figure 4. Distribution of products obtained in tests with shorter durations, lower temperatures, and lower steam-to-Cu ratios ($CuCl_2$ is brown and Cu_2OCl_2 is black).

Fluidized Bed Reactor. Researchers conducted a series of tests with a fluidized bed reactor in which the Ar carrier gas entered from the bottom rather than the top. In these tests, the total flow rate of both steam and Ar was fixed at 200 ml/min, while the individual rates of Ar and steam were varied. The fluidization of the ball milled material was minimal. The S/Cu ratio varied from 16 to 112. One unexplained result was that all of the products from these fluidized bed experiments showed undissolved material remaining from preparation for the CuCl analysis, which consisted of dissolving the samples in ferric ammonium sulfate reagent containing 20 percent HCl. At this time, researchers speculate that this undissolved material is CuO, which was previously detected by x-ray diffraction (XRD) on similar samples from fluidized bed reactor experiments. A second unexplained result is that all of the products contained brown aggregated material, indicating a significant amount of $CuCl_2$ remained unreacted, presumably because of poor mass transfer in the center of the bed.

Rotary Reactor. Researchers completed the safety review for the rotary reactor. Initial experiments performed showed significant difficulties in operating the equipment. Consequently, this approach was not deemed sufficiently promising to continue.

Summary of Reactor Tests. None of the reactors tested were considered satisfactory. CuCl was observed in all of the products. Analysis of these results indicated that mass and heat transfer were poor in these reactor designs. Therefore, researchers designed a new reactor that uses a nebulizer to inject a $CuCl_2$ solution, which should improve mass and heat transfer. Equipment is being assembled and testing will start in FY 2008.

Status of Analytical Test Methods. In the 2006 annual report, researchers noted concerns in using XRD for analyzing the solid products of the hydrolysis reaction. Since then, they have determined that XRD is not an optimum method for analyzing mixtures of Cu_2OCl_2, $CuCl_2$, and CuCl, which are typically observed as solids from the hydrolysis reaction. The oxygen in the air can oxidize the CuCl and moisture in the air can promote the conversion of Cu_2OCl_2 to atacamite, an insoluble copper hydroxy chloride at room temperature. A wet analytical method was subsequently developed. However, it is very expensive and researchers have analyzed recent samples only for CuCl in order to reduce costs.

The Electrolyzer (AECL). Researchers built and tested two electrolyzers: 1) a static system and 2) a flowing (or continuous) system. They examined six types of commercially available membranes in the static system using a variety of combinations of electrode materials for direct H_2 production. Membranes supplied by the Ameridia Division of Eurodia Industrie (ACM and AMV) and (Asahi Glass Engineering Co., Japan (ASV) have been identified as the better candidates for this application in light of their resistance to Cu^{2+} ion migration across the membrane. Testing in the static system has been carried out with cathodic electrolyte of 1.5 M CuCl in 6 M HCl solution and anodic electrolyte of 6 M HCl solution at room temperature. This effort on the static system is to help determine testing conditions and materials that will be used in the continuous system. Results presented in Table 1 are directly useful for selection of electrode materials for the continuous system. Applied potentials presented are the minimum required to produce H_2 for the specific combination of membrane and electrode. The minimum potential required for a certain membrane varied depending on electrode materials tested. H_2 could be produced at a potentials as low as about 0.55V with some combinations of membranes and electrode materials.

The researchers designed and constructed a continuous electrochemical cell experimental unit at AECL, as shown in Figure 5. The unit is designed for continuous operation on a daily basis with flowrates ranging from 1–10 ml/min for anodic and cathodic solutions. The electrolyzer cell, in Figure 6A, is a commercially available unit made with carbon graphite and is temperature controlled. The system is designed to be flexible in its operation so that the variables that affect cell performance can be evaluated systematically.

Membrane	Anode	Cathode	Minimum Required Potential (V)	Demonstration Duration (min)
1-ACM	Pt	316 Stainless Steel	0.66	18
1-ACM	C	316 Stainless Steel	0.71	24
1-ACM	C	Pt	0.55	15
1-ACM	Pt	C	0.55	15
1-ACM	C	C	0.61	16
AMV	Pt	Brass	0.82	22
AMV	Pt	316 Stainless Steel	0.77	21
AMV	Pt	C	0.77	20
AMV	Pt	Pt	0.55	18
AMV	C	316 Stainless Steel	0.71	20
AMV	C	Pt	0.77	20
ASV	Pt	Pt	0.55	12
ASV	Pt	Brass	0.82	18
ASV	Pt	316 Stainless Steel	0.72	20
ASV	C	Pt	0.54	12
ASV	C	Brass	0.82	20
ASV	C	316 Stainless Steel	0.72	20

Table 1. Minimum potential required for H_2 production in the static system.

Researchers also carried out experimental studies of direct H_2 production by electrolysis of HCl/CuCl at room temperature in the continuous system shown in Figure 5. Based on the results of the static system, the ACM membrane was tested with a carbon paper anode and platinum (Pt) deposited on carbon paper as the cathode, to demonstrate and investigate the direct H_2 production. In all tests carried out, 1.5 M CuCl in 6 M HCl solution and 6 M HCl solution were used as cathodic and anodic electrolytes, respectively, at the same flow rate of 5 ml/min. Hydrogen was produced at a potential as low as 0.50 V at an initial current density of 2.1 $mA \cdot cm^{-2}$ in some tests, although testing was conducted mainly at about 0.65 V in this study. Figure 6B shows the hydrogen bubbles generated in the electrolyzer cell. Thus, the researchers have experimentally demonstrated the concept of direct H_2 production by electrolysis of HCl/CuCl system in the continuous system.

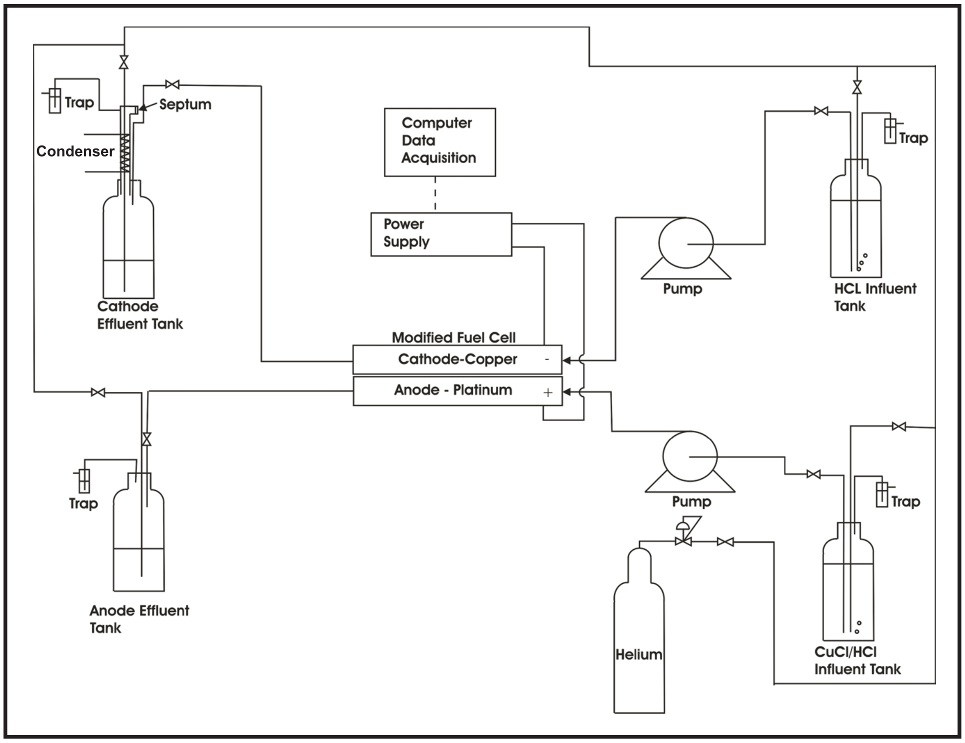


Figure 5. Schematic of the flowing electrolyzer developed at AECL.

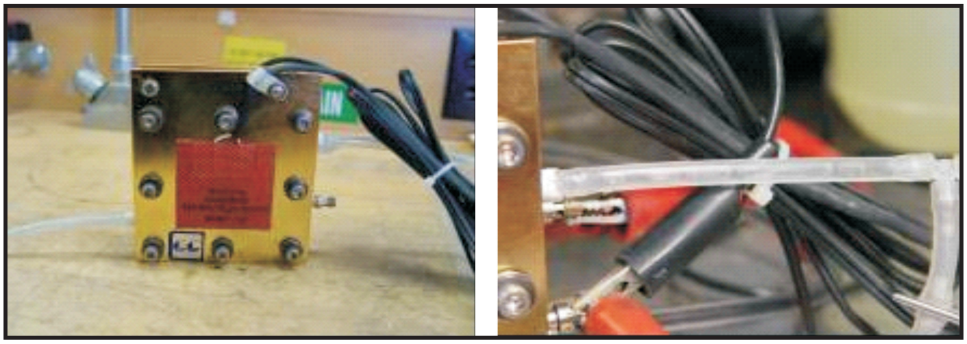


Figure 6. Pictures of (a) electrolyzer cell and (b) direct H_2 production demonstration by electrolysis of HCl/CuCl system.

Planned Activities

Although this project has concluded, research will continue for another three-year period under I-NERI project 2007-002-C with the same title. ANL researchers will continue developing the hydrolysis reactor with objectives of increasing the Cu_2OCl_2 yield, minimizing the decomposition of $CuCl_2$ into Cl_2, and reducing the steam-to-copper molar ratio. A related NERI-C project has also been funded to advance the knowledge base for three thermochemical hydrogen cycles: Cu-Cl, Ca-Br, and the active metal alloy cycles. Additional collaboration is planned with several other Canadian universities.

INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE

Hydrogen Production Using High Temperature Electrolysis

PI (U.S.): Stephen Herring, Idaho National Laboratory; Richard Doctor, Argonne National Laboratory (ANL)

PI (Canada): Sam Suppiah and Donald Ryland, Atomic Energy of Canada Limited (AECL)

Collaborators: University of Ontario Institute of Technology

Project Number: 2007-001-C

Program Area: NHI

Project Start Date: July 2007

Project End Date: March 2010

Project Abstract

This project will address research and scale-up issues associated with the use of energy efficient, high temperature solid-oxide electrolysis cells (SOEC) to produce hydrogen from steam. Researchers will use a detailed economic model developed at ANL to compare the economics of high- and low-temperature electrolysis cell designs. The project has three primary objectives: 1) economic analysis of electrolysis processes; 2) handling of high-temperature oxygen in high-temperature electrolysis (HTE) systems; and 3) high-temperature materials for the cell stack and heat exchangers.

HTE systems operate at high temperature, presenting significant technical issues and hazards in handling the hot gas streams, especially oxygen. An integrated laboratory scale (ILS) experiment will dilute the 850°C oxygen stream with air and cool it to room temperature before being exhausted. Researchers will develop methods for safely handling the 3 $Nm^3 \cdot h^{-1}$ of hot oxygen produced (2.1 kg O_2/hr), along with processes for recovering heat from the oxygen stream. Proper selection of materials for the key components—high-temperature heat exchangers, cell stack components, and interconnecting piping—is critical to achieving high performance of the overall HTE process. Researchers will conduct a series of corrosion tests to analyze material performance in both the high temperature electrolysis cell and the balance of the plant.

The following tasks comprise the main elements of this project:

- Provide input for safe handling of oxygen streams
- Conduct economic analysis of low temperature electrolysis systems
- Identify suitable materials for HTE systems and analyze their performance
- Develop process for heat recovery from hot oxygen streams
- Conduct economic analysis of low temperature and high temperature electrolysis systems

INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE

Thermochemical Hydrogen Production Process Analysis

PI (U.S.): M. Lewis, Argonne National Laboratory

PIs (Canada): S. Suppiah and L. Stolberg, Atomic Energy Canada Limited (AECL)

Collaborators: University of Ontario Institute of Technology, Pennsylvania State University, Tulane University, University of South Carolina

Project Number: 2007-002-C

Program Area: NHI

Project Start Date: May 2007

Project End Date: May 2010

Research Objectives

The objective of this project is to obtain the cost of hydrogen production using the low-temperature, copper-chlorine (Cu-Cl) cycle. The steps necessary to meet this objective are the following: 1) develop a pilot plant-scale unit for the hydrolysis reactor that provides for high mass and heat transfer, 2) develop a Cu-Cl electrolyzer that produces hydrogen at the cathode and meets the performance target, 3) update the Aspen flowsheet with new experimental data, and 4) calculate hydrogen production costs. Current experimental and modeling activities are designed to more accurately evaluate the Cu-Cl cycle and solve the technical challenges that have been identified.

Reaction		Temperature (˚C)
(1) Hydrolysis	$2CuCl_2(s) + H_2O(g) \rightarrow Cu_2OCl_2(s) + 2HCl(g)$	340-400
(2) Decomposition	$Cu_2OCl_2(s) \rightarrow \frac{1}{2}O_2(g) + 2CuCl(s)$	450-530
(3) Electrolysis	$CuCl(s) + HCl\ (aq) \rightarrow CuCl_2(s) + \frac{1}{2}H_2(g)$	100

Table 1. Reactions in the copper-chlorine cycle.

Research Progress

The copper-chlorine cycle thermochemical cycle can be used to produce hydrogen using nuclear or solar heat. Several types of nuclear reactors can be used as a heat source, such as the CANDU Mark 2 supercritical water reactor being developed in Canada, the lead-cooled reactor, or the high-temperature gas reactor. All these provide heat near or above 600°C, the maximum temperature required for the cycle. Solar heat can be obtained from commercially proven tower technology.

The copper-chlorine cycle consists of the three major reactions shown in Table 1, all of which have been demonstrated in proof-of-concept experiments. The two thermal reactions, hydrolysis of cupric chloride ($CuCl_2$) and decomposition of copper oxychloride (Cu_2OCl_2), have been proven. In bench-scale experiments, all of the oxygen was recovered at 530°C from Reaction (2). AECL recently demonstrated the electrolytic reaction in which $CuCl_2$ is produced at the anode and H_2 at the cathode.

During this quarter, researchers focused on the following three areas: 1) the design of the hydrolysis reactor, 2) development of the Aspen flowsheet, and the 3) H2A cost analysis for hydrogen production using solar heat.

Hydrolysis Reaction. Researchers designed a new reactor in which mass and heat transfer should be improved in comparison with fixed beds. This reactor uses a nebulizer to inject a $CuCl_2$ solution, as shown in Figure 1. Preliminary testing indicates that the concept has merit

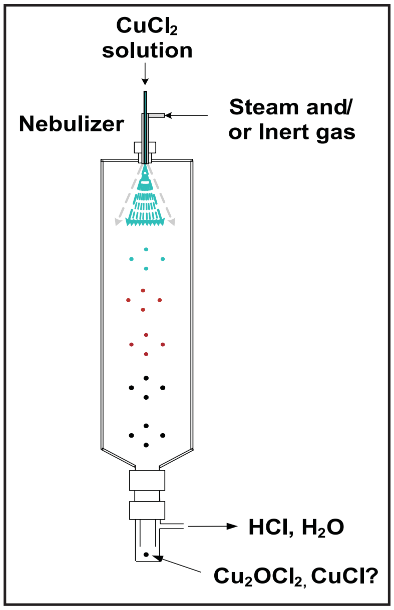


Figure 1. Nebulizer reactor design.

but some modifications are necessary to increase the Cu_2OCl_2 yield. Most of the $CuCl_2$ remained unreacted in early experiments, which researchers suspect was caused by residence time in the heated zone being too short. The team will install equipment to add superheated steam between 300-400°C and will use a 40-inch furnace rather than a 12-inch furnace.

Conceptual Process Design and Aspen Flowsheet Development. Researchers completed a conceptual process design for the cycle that consists of two sections: 1) the electrolyzer and crystallizer, and 2) the hydrolyzer and oxychloride decomposition reactors, shown in Figures 2 and 3, respectively. In the first section, the electrolyzer is run near 100°C at 24 bar. The catholyte is a hydrochloric acid (HCl) solution and the anolyte is a solution of HCl, dissolved CuCl, and $CuCl_2$. Hydrogen is produced at the cathode at pipeline pressure and Cu(I) is oxidized to Cu(II) at the anode. The anode effluent is transferred to the crystallizer, which operates at 15°C, and some of the $CuCl_2$ crystallizes. The supernatant is transferred to the anode feed tank where recycle CuCl is added. The $CuCl_2$ solid or slurry is transferred from the crystallizer to the hydrolysis reactor, where additional steam is added. The Aspen model indicates that a steam-to-copper molar ratio of 14 is required for operation at 400°C. The $CuCl_2$ is converted to Cu_2OCl_2 (a dry free-flowing powder) in the hydrolysis reactor, then gravity flows to the oxychloride decomposition reactor where it decomposes to liquid CuCl and oxygen.

Researchers developed an Aspen Plus flowsheet using this process design. They balanced energy and mass, calculated the heat exchanger duties and shaft work, and optimized heat recovery with pinch analysis. Because there are no performance data for the electrolyzer needed in the Cu-Cl cycle, researchers used the current hybrid sulfur cycle performance targets, i.e., a cell EMF of 0.5 V and current density of 500 mA/cm^2. The hydrogen production efficiency from this flowsheet was 44 percent based on lower heating value (LHV), with 2,400 kg of H_2 produced per day.

H2A Cost Analysis. The funds for completing the H2A cost analysis were obtained from the Department of Energy Office of Energy Efficiency (DOE-EE). Under this program, a solar heating source was to be included in the cost of hydrogen production. Researchers estimated the hydrogen production cost using the H2A hydrogen analysis methodology and estimated capital and operating costs for the thermal processes using Capcost software [R. Turton et al., in *Analysis, Synthesis and Design of Chemical Processes* (2nd ed.), Prentice Hall, 2003]. Costs for the electrolyzer

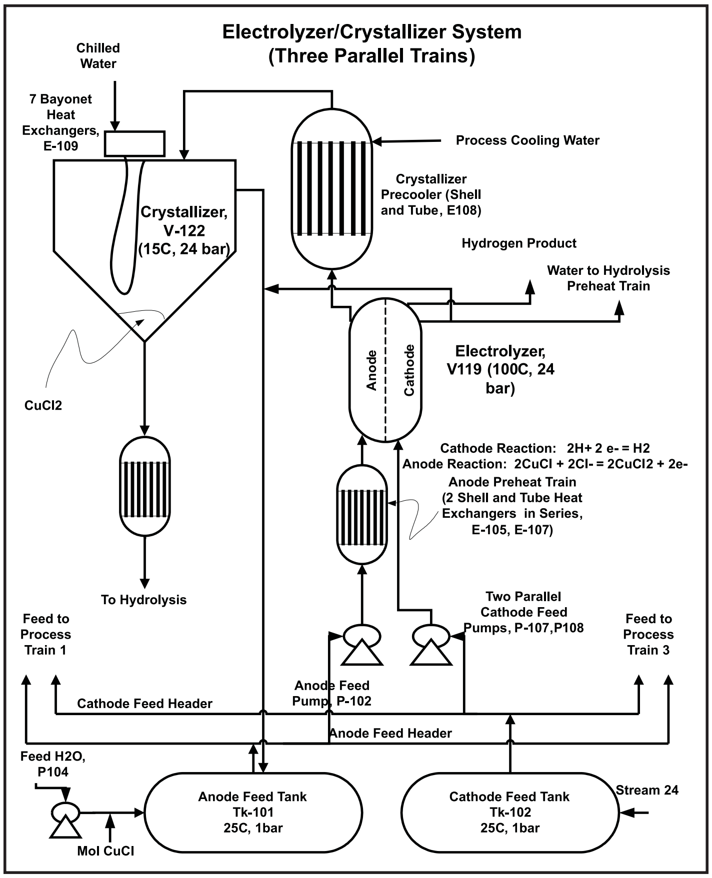


Figure 2. The electrolyzer and crystallizer units in the conceptual process design.

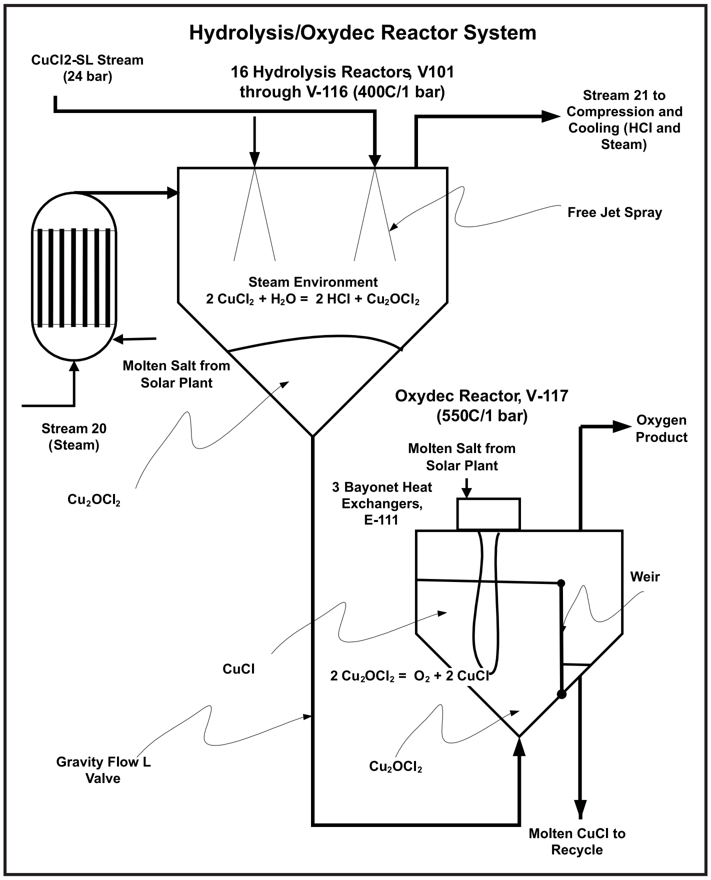


Figure 3. The hydrolysis and oxychloride decomposition reactors.

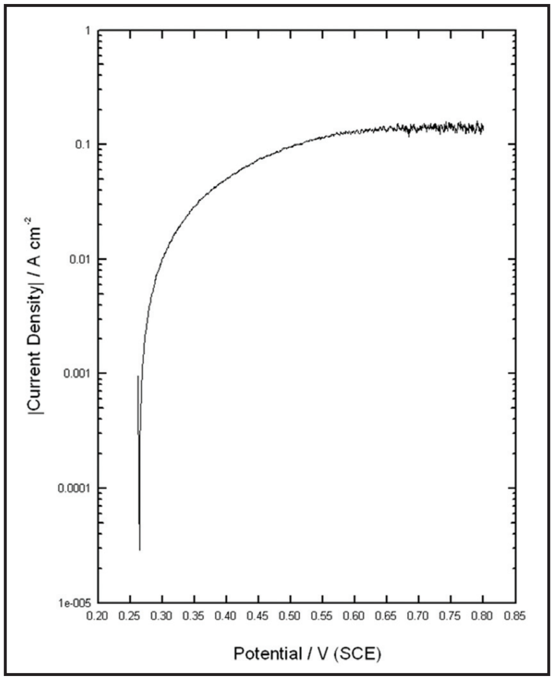


Figure 4. Half-cell polarization curve recorded for a platinum electrocatalyst in an unstirred 1.5 M CuCl in 6 M HCl argon purged solution at ambient temperature. The electrode area and scan rate are 1.79 cm^2 and 0.2 mV s^{-1}, respectively.

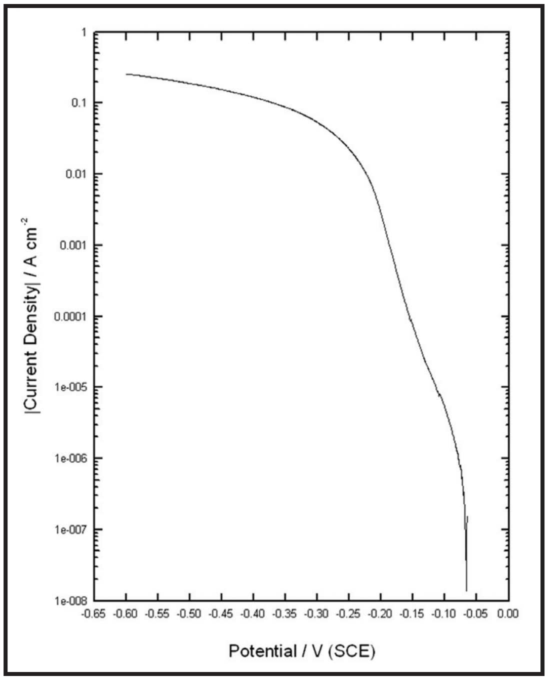


Figure 5. Half-cell polarization curve recorded for a Pt electrocatalyst in an unstirred 6 M HCl argon purged solution at ambient temperature. The electrode area and scan rate are 1.79 cm^2 and 0.2 mV s^{-1}, respectively.

were assumed to be the same as those for the hybrid sulfur electrolyzer, \$2,500/m^2. Other key assumptions were the following: 1) the Chemical Engineering Plant Cost Index (CEPCI) was 486; 2) porcelain-coated steel would not be corroded and the coating would add 6 percent to the cost of carbon steel; 3) the crystallizer would operate as defined in the model, based on literature data; and 4) the hydrolysis reactor would provide high heat and mass transfer, resulting in a residence time of 5 seconds. The cost of hydrogen produced using solar heat was \$4.29/kg H_2 when 125 MT (metric tons or 1000 kg) of hydrogen are produced per day for 2015 technology and \$2.81 for 2025 technology. If the costs for the solar heating plant are stripped out and the costs for nuclear electricity and heat were included parametrically as \$60/MWh(e) and \$20/MWh(t), the resulting cost for producing hydrogen is \$3.10/kg for 2015 technology. Both the Aspen flowsheet and the H2A cost analysis are currently under review and will be updated whenever new experimental data indicate changes should be made.

Electrolyzer (AECL). In one version of the Cu-Cl thermochemical cycle, hydrogen is produced electrochemically according to the following electrolysis reaction given in Table 1:

$$2CuCl(s) + 2HCl(aq) \rightarrow H_2(g) + 2CuCl_2(s) \qquad (1)$$

This electrochemical reaction can be separated into anodic and cathodic half-cell reactions. The half-cell electrochemical reaction that takes place at the anode is:

$$2CuCl(s) + Cl\text{-}(aq) \rightarrow 2CuCl_2(s) + 2e^- \qquad (2)$$

In this half-cell reaction, cuprous chloride (CuCl) is oxidized to cupric chloride ($CuCl_2$). The half-cell reaction that takes place at the cathode, in which protons are reduced to form hydrogen gas, is given by the following equation:

$$2H^+(aq) + 2e^- \rightarrow H_2(g) \qquad (3)$$

Researchers carried out electrochemical experiments to study the half-cell reactions given by Equations 2 and 3 using a standard corrosion cell with three electrodes. The polarization curve obtained for the anodic half reaction is shown in Figure 4, while the polarization curve for the cathodic half-cell reaction is shown in Figure 5. The data in these figures are presented as semi-logarithmic plots of the absolute value of the current density versus the applied potential. The potentials are reported versus the saturated calomel reference electrode (SCE). In both half-cell experiments, a platinum electrocatalyst was used as the working and counter electrodes.

The polarization curve presented in Figure 4 was obtained for the platinum electrocatalyst in a 1.5 M CuCl in 6 M HCl solution. The data show that for a 0.249 V overvoltage, a current density of 0.1 A cm^{-2} (100 mA cm^{-2}) can be obtained. When the electrode potential reaches values above 0.6 V, the polarization curve displays a well-defined plateau. The value of the current density in this region is 0.14 A cm^{-2}. The current density in this region is called a limiting current. It occurs when the electron transfer kinetics is fast, while the transfer of the electroactive species to the electrode-solution interface is slow or inefficient. When the solution is well stirred, a limiting current is no longer observed in this region and the current increases with increasing potential. The current density in this potential region is larger than the limiting current density when the solution is stirred. The polarization curve presented in Figure 5 was obtained for the platinum electrocatalyst in a 6 M HCl solution. The data shows that a current density of 0.1 A cm^{-2} can be obtained at an overvoltage of 0.302 V. Incidentally, at this current density, hydrogen bubbles are rapidly generated at the electrode surface. The data in Figure 5 show that the hydrogen evolution reaction is not under mass transfer control under the present set of experimental conditions. But rather, it is under activation control. This implies that the current density should show a dependence on the nature of the electrocatalyst.

Data from the polarization curves in Figures 4 and 5 suggest that the full-cell electrolysis cell should be able to generate a current density of 0.1 A cm^{-2} at a voltage of 0.551 V.

Planned Activities

Researchers will continue the development of the hydrolysis reactor with the objectives of increasing the Cu_2OCl_2 yield, minimizing the Cl_2 yield from $CuCl_2$ decomposition, and reducing the steam-to-copper molar ratio. They are also supporting work to identify HCl sorbents in a steam/HCl stream. If funds permit, they will start work to verify the solubility properties of $CuCl_2$ and CuCl in HCl aqueous solutions at different temperatures.

A related Nuclear Energy Research Initiative-Consortium (NERI-C) project that was recently funded is concerned with advancing the knowledge base for three cycles: the Cu-Cl, the Ca-Br, and the active metal-alloy cycles. All of these cycles involve an electrolytic reaction, and the proposed work will develop advanced electrochemical technologies that will lead to overall improvements in cycle performance. The work will focus specifically on the following: 1) development of membranes, electrocatalysts, electrode materials, and membrane electrode assemblies (MEA) for all of the cycles; 2) separation of reaction products; 3) identification and modeling of species involved in the electrochemical processes; and 4) flowsheet analyses to guide the experimental program towards higher efficiency and lower cost processes. Methods for concentrating solutions using electrodialysis and possibly other techniques will also be investigated. The participants in the NERI-C will also collaborate with their colleagues at the University of Ontario Institute of Technology.

The modeling work and the experimental work will be iterative. As understanding of the chemistry increases and the technology evolves, researchers will update and optimize the Aspen flowsheets for these cycles to maximize efficiency and reduce production costs.

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

Of the seven projects approved in FY 2004 that remained active during the past year, 5 have been completed. One new collaboration with Italy was established in 2006 under the EURATOM agreement. A listing of the I-NERI U.S./EU projects that are currently underway follows, along with summaries of the accomplishments achieved in FY 2007.

Directory of Project Summaries

INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE

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

PI (U.S.): J. Carmack, Idaho National Laboratory

PI (Europe): J. Somers, Joint Research Centre Institute for Transuranium Elements (JRC-ITU)

Collaborators: University of Florida, Los Alamos National Laboratory (LANL)

Project Number: 2004-001-E

Program Area: AFCI

Project Start Date: October 2004

Project End Date: September 2008

Research Objectives

Commercial power reactors are the only viable option for short- to mid-term (10–20 years) active management of plutonium and minor actinides (MAs). Thus, there is worldwide interest in the use of the existing commercial reactor fleet for fuel cycle scenarios involving burn down of plutonium and stabilization of minor actinide inventories. This project seeks to develop feasibility data related to the use of inert matrix fuels (IMFs) as fuel and MA targets in the existing fleet of light water reactors (LWRs). IMF offers potential advantages over conventional uranium matrix mixed oxide (MOX) fuel for plutonium management as it allows more efficient destruction of plutonium, since the exclusion (or significant reduction) of U^{238} from the fuel precludes breeding additional plutonium. Greater efficiency in plutonium reduction results in improved flexibility in managing plutonium inventories. Also, there is a potential for fuel cycle cost savings due to the reduced number of rods required to effect a given plutonium burn rate. In addition, IMF can be used in strategies for the disposition of minor actinides, particularly americium and neptunium.

The choice of matrix material may be manipulated to facilitate either fuel recycling or direct disposal; plutonium recovery can be made relatively straightforward or extremely difficult. Inert matrix fuels having high thermal conductivity may also have operational and safety benefits. Cermet fuel (Figure 1), for example, operates at very low fuel temperatures, a property that can be used to increase operating and safety margins or uprate reactor power. Minor actinide targets contain high MA contents and are designed for heterogeneous MA burning schemes. In order to use existing water-cooled reactors, all research and development concepts considered in this proposal will be suitable for loading into present-day and near-term (Generation II and III) LWR designs. Completion of this research project will result in comparative data on the fabrication, properties, and irradiation behavior of several IMF fuel candidates. This data will provide valuable information on the feasibility of IMF for plutonium and minor actinide management in LWRs.

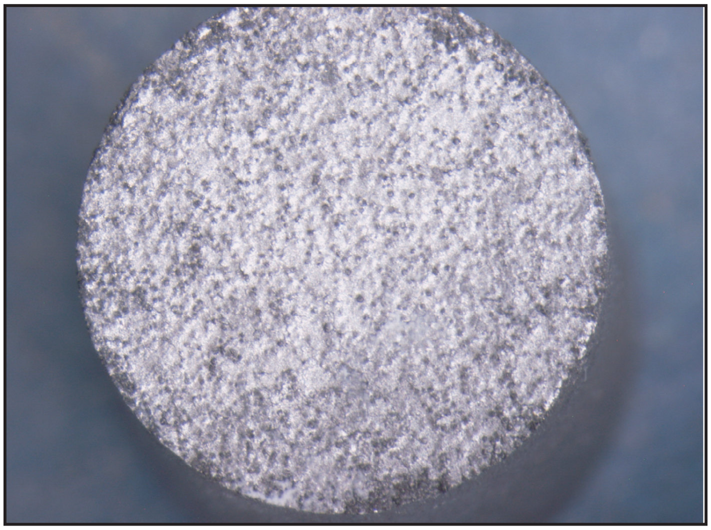


Figure 1. End face of Cermet pellet $(Pu0.73Th0.23)O_2/Mo$ (40:60 vol%).

Research Progress

Following is a description of the progress that researchers have made over the past year.

Composite Synthesis and Characterization. The specific heat capacity of the composite was measured using a differential scanning calorimeter. When the measured specific heat capacity was compared to a value calculated from the rule of mixtures, researchers found that the calculation was a poor approximation for the measured specific heat capacity. Three batches of composite material were synthesized using each of four different processing methods (ball milling, magnetic bar stirring, mortar and pestle, and Spex milling), then three pellets fabricated from each batch. Figure 2 shows a typical pellet. Thermal diffusivity results indicate that processing may affect the *intra*-batch, but not the *inter*-batch sample variation. Each processing method can be characterized by an average thermal conductivity that may not vary significantly across a population of samples. The thermal diffusivity was also measured on the pellets fabricated for the sintering kinetics study. There is evidence that as the microstructure evolves, there is an effect on the thermal conductivity of the samples. As the sintering temperature and time increased from 1,550°C to 1,650°C, and from 8 hours to 24 and 100 hours, respectively, there was a corresponding drop in thermal conductivity.

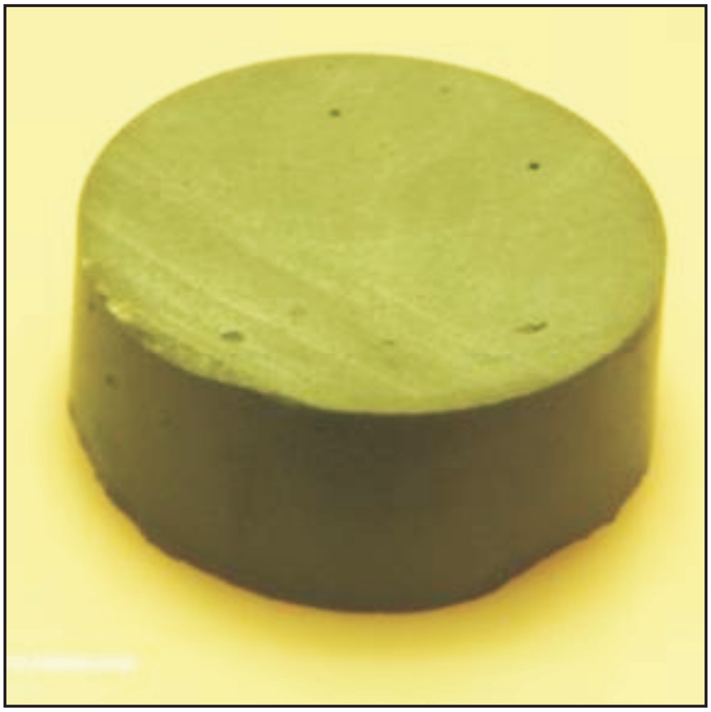


Figure 2. Photograph of dual-phase MgO-ZrO_2-PuO_2 pellet.

Spinel Candidate Material Selection. The Mg_2SnO_4 sample previously irradiated by 1 MeV Kr^{2+} ions at 50 K and fluence of 5×10^{19} ions/m^2 was reexamined at room temperature using transmission electron microscopy (TEM) and energy dispersive spectroscopy (EDS). After thermal recovery, the amorphous sample turned polycrystalline. Selected area diffraction (SAD) patterns indicate that the crystal structure for major inner grains is spinel. EDS results and bright field (BF) images indicate that phase separation occurred along the edge of the focused ion beam (FIB) sample and the thickness of this fringe is about 1 μm. The researchers performed a TRIM calculation and converted the amorphization doses into displacement per atom (dpa). In the simulation, 40 eV displacement threshold energy was chosen for all atoms in the Mg_2SnO_4 target. Researchers then calculated the corresponding amorphization dose at 50 and 150 K to be 6.25 and 12.49 dpa, respectively. To simulate fission fragment effects such as swelling, follow-up collaboration with LANL is being explored in order to generate a heavy ion source at fission energy levels by using a high-energy proton source (800 MeV) to bombard a bismuth foil.

Corrosion Testing. The MgO-$Nd_2Zr_2O_7$ sample with 70 volume percent MgO made by gel-casting was tested at 300°C and saturation pressure for 10 days. Due to the irregular shape of the sample, only a qualitative experiment was conducted. The sample withstood hydration induced stresses and did not fracture, which shows improved hydration resistance compared with the ball mill produced MgO-$Nd_2Zr_2O_7$ composite, which fractured after one hour exposure to 300°C deionized water.

Thermophysical Property Simulation. The researchers developed a general computer code for the generation of MgO-NDZ composites of various morphologies. This code generates structures in a manner similar to that used for polycrystals, as described in previous reports. A systematic study of $MgAl_2O_4$ spinel and structures with 25, 50, 75, and 100 percent cation inversion has shown that thermal expansion is independent of the degree of inversion. Simulations to determine the dependence of thermal conductivity on composite microstructure and the degree of inversion are underway.

Planned Activities

This project was predicated on the use of thermal reactor spectra for the transmutation of minor actinides. In February 2006, the Department of Energy decided to focus efforts on the transmutation of minor actinides in fast spectrum systems, thereby eliminating the need for separation of specific actinide streams. With this decision, the work packages and authorizations for research of thermal spectrum transmutation have been redirected to fast reactor transmutation and all work on thermal transmutation terminated. Currently, this agreement is being renegotiated to redirect focus of research to fast reactor transmutation fuels.

INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE

Lead-Cooled Fast Reactor Engineering and Analysis

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

PI (Europe): Hartmut U. Wider, Joint Research Centre, Institute for Energy (JRC-IE)

Collaborators: None

Project Number: 2004-003-E

Program Area: Generation IV

Project Start Date: November 2004

Project End Date: November 2007

Research Objectives

The objectives of this project are to further develop the analysis base and experimental database for the lead-cooled fast reactor (LFR). Europe is conducting extensive research and development on the European Lead-cooled SYstem (ELSY) to demonstrate the technical feasibility and economic competitiveness of an LFR for burning minor actinides (MAs). The U.S. is carrying out R&D on the Small Secure Transportable Autonomous Reactor (SSTAR), a modular proliferation-resistant LFR intended for international 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. Both reactor designs are consistent with the goals of the Global Nuclear Energy Partnership (GNEP) and the Generation IV Nuclear Energy Systems Initiative.

Research Progress

Researchers investigated three alternative concepts for burning minor actinides in the ELSY core: 1) a homogenous mixture of 5 percent MAs with the oxide fuel, 2) MAs strictly in external blankets/targets, and 3) MAs both in the fuel and external blankets. They found that the latter configuration, with an active core height of 1.2 or 1.3 m, would be most effective, consuming approximately 115 kg of MAs per year while maintaining or increasing the mass of plutonium (Pu). The use of plutonium-thorium-MA (Pu-Th-MA) fuel was also examined in a regime designed to burn 89 kg of MAs per year while generating more than 1,000 kg of uranium-233 (^{233}U). This can be used to fuel converter or breeder CANDU reactors in a Th–^{233}U fuel cycle, enabling a faster growth rate in future nuclear capacity than breeding Pu in sodium-cooled fast reactors.

Experiments were performed at the Forschungszentrum Karlsruhe under a contract with JRC/IE to evaluate 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 temperatures between 400 and 600°C. Cesium forms inter-metallic compounds in LBE, while iodine reacts with the lead to form PbI_2, that has a melting temperature of 850°C. However, formation of PbI_2 did not consume all of the iodine; some iodine escaped absorption and was released from the LBE free surface.

Researchers developed a design for a new experiment investigating LFR steam generator tube rupture phenomena in a tube bundle. The experiment will inject supercritical water at 25 MPa and 480°C into a 10 cm diameter pipe 80 cm in height with flowing Pb and simulated heat exchanger tubes. The bottom of the pipe closes upon rapid pressurization, simulating a design feature of the ELSY steam generators. Injection of 3 liters of supercritical steam simulates the failure of eight steam generator tubes. Other experiment results from JRC/IE were shared 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.

Using computational fluid dynamics (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. They found that for a protected total loss-of-power accident, the RVACS design was unable to prevent core outlet temperature from exceeding the structural steel creep limit. Therefore, they concluded that the designs may require a more efficient emergency decay heat removal approach.

Researchers analyzed the thermal hydraulic conditions in an ELSY, open square-lattice fuel assembly postulated to have a total inlet flow blockage. Calculations with the CFX10 CFD code show that crossflow between the fuel pins facilitates heat removal from the fuel pins such that the open pin assembly design overcomes the blockage with the maximum coolant temperature in the lower velocity region above the blockage reaching 919°C.

Postulated unprotected loss-of-flow (ULOF) and unprotected loss-of-heat sink (ULOHS) accidents for ELSY were calculated with the EAC-2 computer code. In the ULOF accident, the core power decreases to about 85 percent nominal due to the negative reactivity feedbacks from the oxide fuel Doppler and axial expansion while the core outlet temperature increases to about 750°C. For the ULOHS accident, the power is calculated to decrease to about 20 percent nominal during the first 1,000 seconds while the coolant temperature rises to about 930°C. Reactivity feedback from radial expansion of the core support plate was not incorporated in the calculation as a conservatism; its inclusion would cause power to decrease more rapidly to near decay heat levels and reduce the magnitude of the temperature increase. Researchers 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-CO_2) Brayton cycle energy converter enables autonomous load following by the reactor over the full range of power from 0 to100 percent, and controllability of the S-CO_2 Brayton cycle near the critical point. This scheme is especially suited for deployment on small or immature electrical grids. The strategy consists of: 1) turbine bypass control for fast changes in heat removal at all load levels, 2) additional inventory control between 90 and 35 percent nominal load (limited by the inventory control tank volume) to maintain cycle temperatures, 3) cooler bypass control to maintain the CO_2 temperature at the main compressor first stage inlet (i.e., the lowest temperature in the cycle) above the critical temperature (30.98°C) to avert the onset of two-phase flow, 4) cooling water flow rate control designed to prevent full opening or closing of the cooler bypass valve, and 5) compressor surge prevention systems to preclude surges that could cause compressor damage under postulated accident scenarios.

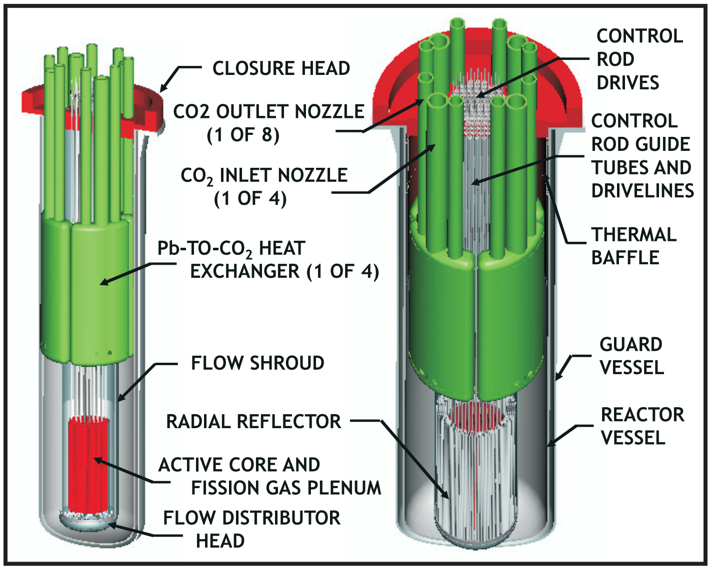


Figure 1. Illustration of 20 MWe (45 MWt) Small Secure Transportable Autonomous Reactor (SSTAR).

Calculations were carried out for an international neutronics benchmarking problem on an LBE-cooled, mixed nitride-fueled core using the same neutronics codes applied to SSTAR. This work is part of an International Atomic Energy Agency (IAEA) research program on small reactors without onsite 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.

The researchers carried out an initial scoping investigation to determine the viability of a near-term deployable, 100 MWt, Pb-cooled, metallic-fueled, forced flow, LFR demonstration test reactor that operates at low temperatures (peak cladding temperature below approximately 550°C). Such a design would enable the use of existing materials (e.g., T91 and Type 316 stainless steel) that have been shown resistant to LBE corrosion with active oxygen control at temperatures below approximately 550°C.

Planned Activities

This I-NERI project is complete.

INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE

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)

Collaborators: None

Project Number: 2004-004-E

Program Area: Generation IV

Project Start Date: January 2004

Project End Date: September 2008

Research Objectives

The purpose of this project is to develop a systematic method for evaluating and comparing the proliferation resistance and physical protection (PR&PP) of Generation IV nuclear energy systems, including 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 an expert group—the PR&PP Working Group—in December 2002 and tasked them with leading the development project. This expert group includes participants from U.S. national laboratories (Argonne, Brookhaven, Idaho, Los Alamos, Lawrence Livermore, Oak Ridge, Pacific Northwest, and Sandia National Laboratories), academia, international experts from five GIF members (Canada, EURATOM, France, Japan, and Republic of Korea), the International Atomic Energy Agency, and observers from the U.S. State Department and the U.S. Nuclear Regulatory Commission.

Research Progress

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 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.”

To focus development of the methodology, the Working Group has 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 → 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" identifies the few measurable, high-level parameters that can be used to express proliferation resistance (PR) or physical protection (PP). Researchers will first evaluate measures for individual segments and then aggregate them for complete pathways. Aggregating results permits pathway comparisons and system assessment.

Proliferation Resistance measures:

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

Physical Protection measures:

- 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 is expected to result in improved integration of PR with safeguard approaches and can provide positive indications to the designers early in the design process.

The Working Group developed the methodology with the aid of the previous Development Study and Demonstration Study, based on an example sodium fast reactor (ESFR). The ESFR is a hypothetical nuclear energy system consisting of four sodium-cooled fast reactors of medium size co-located with an on-site dry fuel storage facility and a pyrochemical spent fuel reprocessing facility. These studies showed that the methodology can be applied to practical cases. However, review of this initial work has indicated areas for further development, such as the use of expert elicitation as a routine component of a PR&PP analysis. Further progress on the PR&PP methodology required a more comprehensive evaluation of a complete reactor/fuel cycle system to gain practical experience within the application process, to discern the needs for further methodology development and presentation of results, and to confirm the usefulness and usability of the evaluation methodology. In particular, the Working Group was requested to demonstrate that designers can obtain practical guidance through application of the methodology. Another request was to demonstrate the capability to apply the PR&PP evaluation framework at different levels of detail, corresponding to different efforts and resources. These formed the basis of a two-year case study begun in FY 2007.

The major objectives of the case study are to:

1) Exercise GIF PR&PP methodology for a complete Generation IV reactor/fuel cycle system
2) Demonstrate that the methodology can generate meaningful results, based on the comparison of different design options
3) Provide examples of PR&PP evaluations for the Implementation Guide and future users of the methodology in order to facilitate the transition to other studies (as planned by Japan and France) and facilitate ongoing collaborative efforts (e.g., INPRO) and associated programs (e.g., GNEP)

The comparison of design options is a key goal of the methodology. Towards this aim, particular care has been exercised in the selection of the reactor/fuel cycle system for the case study. Because the ESFR system with an associated fuel cycle facility has already been well characterized, it has been selected as the reference case. Whereas the case study focuses on facility-level PR&PP questions, it may also support observations about PR&PP at the global system architecture level, with further analysis.

The baseline ESFR nuclear energy system design operates in a net actinide burning mode that requires an external source of actinides for make-up. Make-up is ensured by a continual supply of light water reactor (LWR) spent fuel elements delivered to the ESFR site for processing.

The ESFR consists of the following system elements:

- LWR spent fuel receipt and storage
- A co-located pyroprocessing facility
- ESFR spent fuel and new fuel storage cell

- Staging/washing area
- Four identical ESFR reactors with in-vessel storage baskets
- Waste storage

With input from designers, the group generated a list of interesting design options for consideration in the case study, including:

- Remote vs. on-site reprocessing of the LWR make-up feed
- Remote vs. on-site reprocessing of ESFR spent fuel
- Breeder vs. self-sustaining vs. burner conversion ratio
- Blended vs. unblended low burn-up fuel material
- Start-up phase options to generate the first fuel load
- Physical arrangement
- Passive access control

The first year of the case study focused on generating the necessary design information and assumptions for the baseline design, along with a notional safeguards and physical protection system. Researchers have initiated an evaluation of the baseline design with respect to a suitable threat space. The threat space considered for the case study includes the following categories:

- Concealed diversion of material
- Concealed misuse of the facility
- Abrogation and overt diversion or misuse
- Theft of weapons-usable material

For the first three threat categories, the actor is a host state with defined objectives and capabilities. For the fourth threat category, the actor is a sub-national group, also with defined objectives and capabilities. The researchers created four subgroups with international composition from within the PR&PP Working Group in order to address the respective threats. Specifically, they were charged to do qualitative evaluations of the outcomes of scenarios that would ensue from the threats.

Planned Activities

A limited number of alternative design options developed by the four subgroups will be selected for evaluation. The researchers will demonstrate how each of the alternatives impacts PR and PP evaluations, thus demonstrating the benefit of performing PR&PP evaluations at an early stage of the system design. Four key questions must be addressed: 1) Would the differences in results impact decisions? 2) Are the differences significant or insignificant? 3) Are there new, unexpected insights? 4) Overall, what is needed to demonstrate to potential users that the PR&PP systematic methodology can be of value to them in defining a problem and drawing logical and informative conclusions from the results?

This I-NERI project will continue developing the PR&PP evaluation methodology. Further updates of the methodology will be based on insights of the work done on the case study. It will be very important to further disseminate the results and to promote the use of the methodology among potential users, mainly among designers of advanced nuclear energy systems so that PR and PP concepts will be considered in the early design phases.

Toward that goal, the PR&PP Working Group is pursuing interactions with the Generation IV System Steering Committee (SSC). SSC representatives have been invited to participate in a January 2008 working group meeting in Marcoule, France. In addition, a PR&PP/SSC workshop will develop a Program Plan to identify scope, schedule, team composition, and resources for future joint evaluations. The overall objective is to identify design features and operating conditions that enhance PR&PP performance. The outcomes of these design studies would include:

- Generic PR&PP conclusions
- Insights from specific studies
- Integration of safety and PR&PP analysis
- Training of designers in PR&PP methods
- Insights for useful application of PR&PP methods

Characterization of Nuclear Waste Forms and Their Corrosion Products

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

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

Collaborators: Pacific Northwest National Laboratory

Project Number: 2004-005-E

Program Area: AFCI

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:

1) 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)
2) Investigate recovery mechanisms and study the accumulation/release behavior of helium (He) in different materials, comparing irradiated fuels and alpha-doped fuels/matrices
3) 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)
4) Examine corrosion mechanisms and characterize solid corrosion products formed during the aqueous alteration of spent nuclear fuel using x-ray photoelectron spectroscopy, TEM/energy-loss spectroscopy (TEMEELS), scanning electron microscopy/energy dispersive spectrometry (SEM-EDS), and x-ray diffraction (XRD)
5) 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 European Union collaborators.

INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE

Nitride Fuel Fabrication Research

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

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

Collaborators: None

Project Number: 2004-006-E

Program Area: AFCI

Project Start Date: October 2004

Project End Date: September 2007

Research Objectives

The objective of this research was to compare fabrication and property data of nitride fuels, focusing on fast-spectrum fuels that can be used in liquid-metal reactors or accelerator driven systems (ADS). Researchers investigated two fuel forms: 1) non-fertile fuel designated for ADS and 2) low-fertile fuel for liquid-metal fast reactors. They chose a non-fertile composition of 36 weight percent (wt%) $ZrN(Pu_{0.50}Am_{0.50})N$ for initial development and a low-fertile 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 the low-fertile composition reduced the americium boil-off. The data show only a 1.7 weight-percent deviation from the specification value, with an average theoretical density of 85.4.

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	Ar	Pressing Pressure	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.

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. %
C	≤3,000 ppm	11,800 ppm
O	≤3,000 ppm	2,620 ppm
N	8.3 wt. %	8.41 wt. %

Table 2. Summary of low-fertile chemistry results.

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.

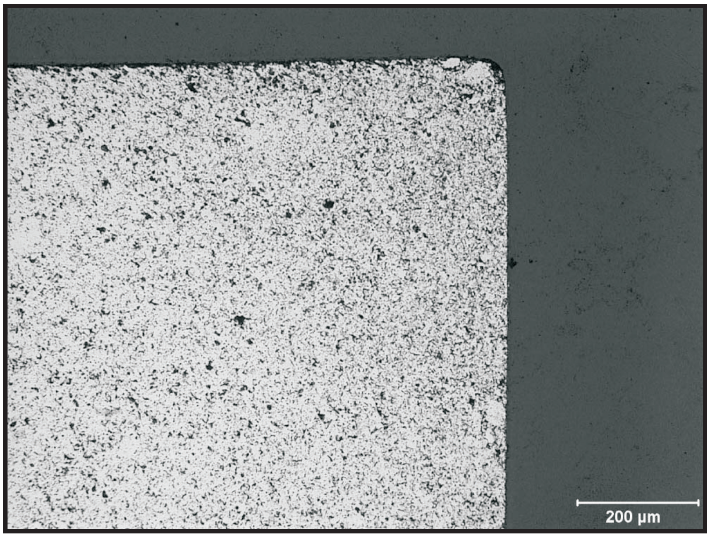


Figure 1. Low-fertile optical micrograph of axial pellet section.

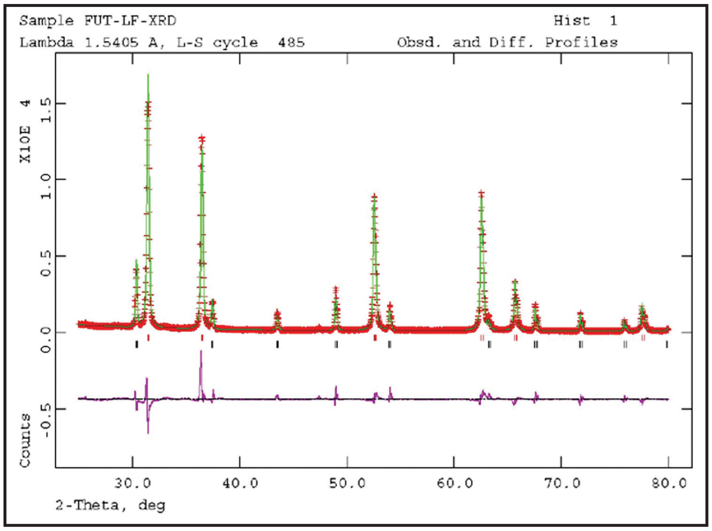


Figure 2. Low-fertile XRD pattern.

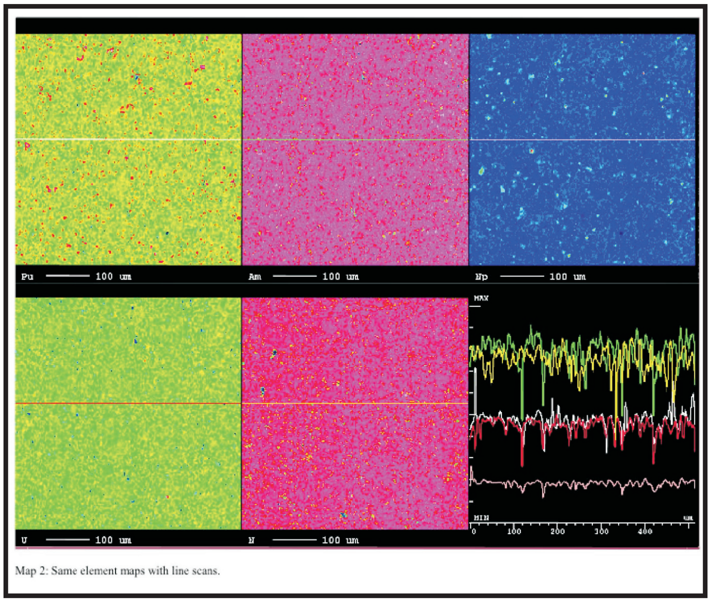


Figure 3. Low-fertile SEM microprobe element maps with line scan. Upper: plutonium (Pu), americium (Am), and neptunium (Np); Lower: uranium (U), nitrogen (N), and an elemental line scan.

Scanning electron microscopy (SEM) microprobe data (Figure 3) show some heterogeneity on the microscale (10–20 percent variation in actinide concentration over 5–10 µm), 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.

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.

Element	Specification Value	Actual Result
Pu	23.7 wt.%	24.43 wt.%
U	47.1 wt.%	47.23 wt.%
Am	14.3 wt.%	12.6 wt.%
Np	9.4 wt.%	7.7 wt.%
C	≤3,000 ppm	1,275 ppm
O	≤3,000 ppm	1,130 ppm
N	5.5 wt.%	5.63 wt.%

Table 4. Summary of non-fertile chemistry results.

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.

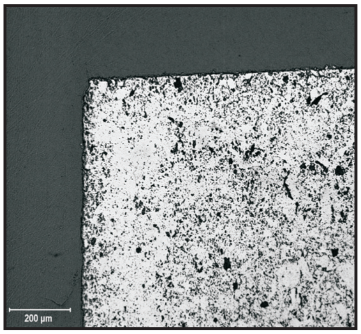


Figure 4. Non-fertile optical micrograph.

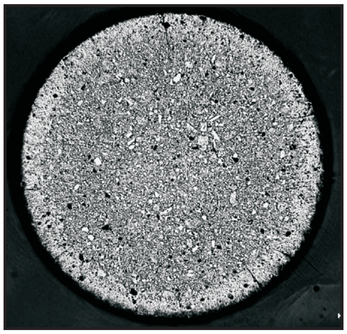


Figure 5. Non-fertile optical macrograph.

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 μm), 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 μm into the pellet.

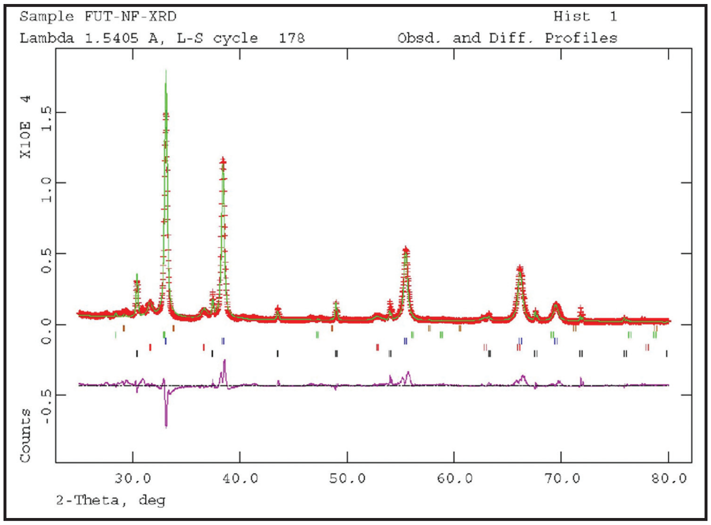


Figure 6. Non-fertile XRD pattern.

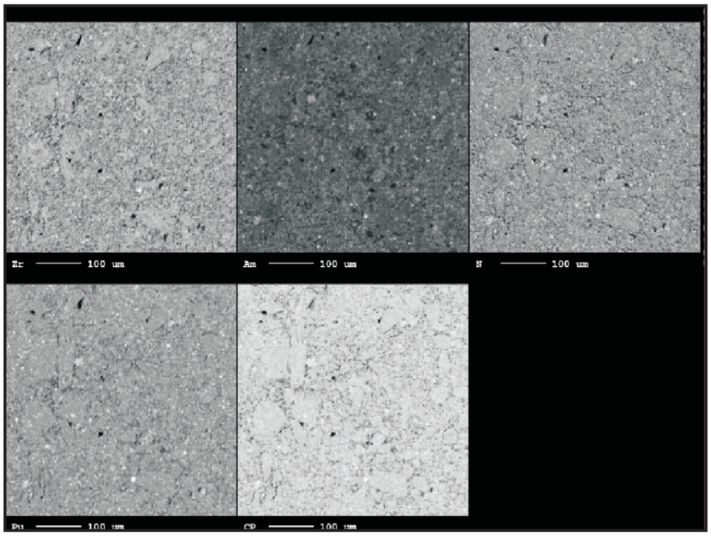


Figure 7. Non-fertile SEM microprobe element maps.

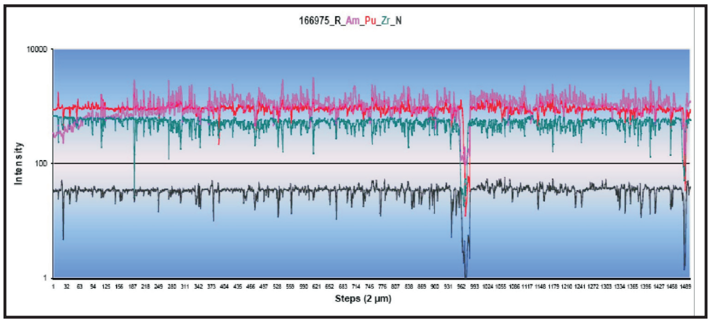


Figure 8. Non-fertile SEM microprobe line scan.

Planned Activities

This project has been completed.

INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE

Use of an Ionization Chamber in Fission Cross-Section Measurements

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

PI (Europe): P. Rullhusen, Institute for Reference Materials and Measurement, (IRMM)

Collaborators: None

Project Number: 2004-009-E

Program Area: AFCI

Project Start Date: October 2004

Project End Date: October 2007

Research Objectives

The aim of this collaboration is to pool resources and competencies in the area of 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 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

The protactinium ^{231}Pa(n, f) cross-section measurements in the energy range E_n = 15 to 21 MeV, performed earlier, suffered from the fact that neutron production based on the T(d, n)^{4}He reaction using a solid TiT target is not mono-energetic. Although researchers took special care to correct the measured fission cross-sections using data from a similar investigation performed at IRMM, the estimate of systematic uncertainties remained somewhat arbitrary. Therefore, they repeated the measurements (for E_n = 16, 19, 20, and 21 MeV) with simultaneous measurement of the neutron beam profile to unfold the cross-section data with the incident neutron spectrum. The neutron detectors have been characterized for pulse heights corresponding to neutron energies up to 21 MeV by simulating the response function starting from the californium ^{252}Cf(SF) fission neutron spectrum. Cross-section data analysis is ongoing.

Because the Van de Graaff accelerator facility at IRMM was overbooked, researchers postponed measuring the ^{233}U 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. Instead, they followed a recommendation of the Working Party on International Evaluation Cooperation (WPEC) and investigated the prompt fission-neutron spectrum from the reaction ^{235}U(n, f) at incident neutron energy E_n = 0.5 MeV. Spectral measurements by the time-of-flight method with a pulsed neutron beam achieved a time resolution of about 1.5 ns. Neutrons were produced via the ^{7}Li(p, n)^{7}Be reaction. The uranium sample, with a total weight of 161.3 g, was mounted at 0-degrees relative to the neutron beam. Three neutron detectors, shielded against direct and room-scattered neutrons, were placed at distances of 2.24 ± 0.01 m and 90-, 120-, and 150-degrees. Researchers applied traditional pulse-shape analysis for efficient gamma-ray suppression and used a small pilot-U scintillation detector for proton-pulse shape monitoring. They measured detector efficiencies relative to the ^{252}Cf(SF) spectrum and corrected data for multiple scattering and attenuation by means of the Monte-Carlo code MCNP.

The fission cross section measurement program at Los Alamos Neutron Science Center (LANSCE) continued this year, focusing on ^{242}Pu and ^{240}Pu. Figures 1 and 2 show the new data along with evaluations from ENDF/B-VII and JENDL3.3. The low-energy measurements carried out last year are included in the figures to show the entire range of the measurement. The Transmutation Physics Working Group under the Advanced Fuel Cycle Initiative (AFCI) determined these isotopes to be high priority due to large discrepancies in the data libraries in the fast region.

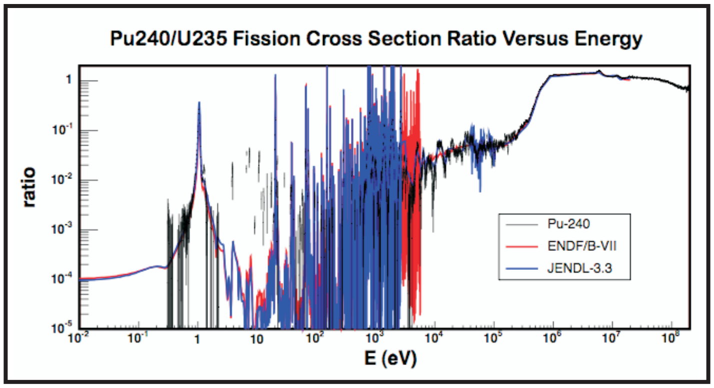


Figure 1. The ^{240}Pu(n,f)/^{235}U(n,f) cross section ratio as a function of incident neutron energy. The data (black crosses) were taken at the Lujan and WNR facilities at LANSCE. The ENDF/B-VII and JENDL3.3 evaluations are shown to demonstrate the discrepancy in these libraries in the fast region. The data and evaluations include the contribution of a 10^{-4} Pu-241 impurity in the sample, which has little impact in the fast region.

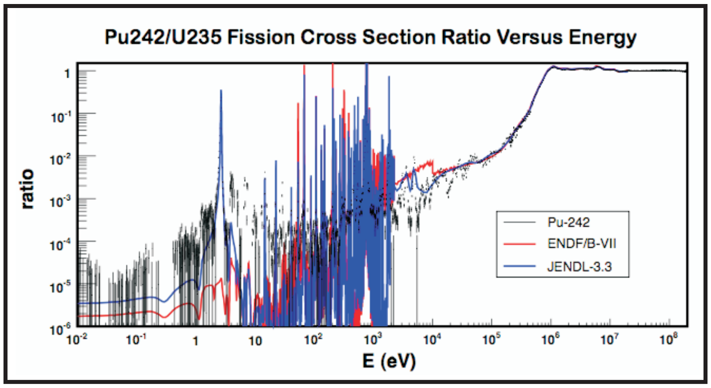


Figure 2. The ^{242}Pu(n,f)/^{235}U(n,f) cross section ratio as a function of incident neutron energy. The data (black crosses) were taken at the Lujan and WNR facilities at LANSCE. The ENDF/B-VII and JENDL3.3 evaluations are shown to demonstrate the discrepancy in these libraries in the fast region. The data and evaluations include the contribution of a 10^{-4} Pu-241 impurity in the sample, which has little impact in the fast region.

Planned Activities

Researches plan to conduct prompt fission-neutron spectrum measurements at thermal incident neutron energy at the Budapest Reactor in early 2008. They will also carry out measurements of ^{239}Pu and ^{241}Pu at LANSCE over the entire energy range.

INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE

Nuclear-Assisted Hydrogen Storage and Safety Issues

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

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

Collaborators: None

Project Number: 2004-010-E

Program Area: NHI

Project Start Date: May 2005

Project End Date: September 2007

Research Objectives

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

1) Perform analytical research on the safety aspects of hydrogen production, storage, and transport
2) Compare the efficiency, complexity, reliability/availability, and economy of different hydrogen production and distribution systems
3) Conduct 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

This project began with an investigation of low-temperature hydrogen production cycles and their potential to provide a more cost-effective means for generating hydrogen compared to high-temperature thermochemical cycles. The fundamental idea is that the structural capabilities of materials decrease exponentially with temperature while thermodynamic efficiency increases much more slowly (linearly). As a consequence, standard reactor materials severely constrain performance with increasing temperature giving rise to the need for more exotic materials, but at added cost. Using numerical simulations, researchers investigated the advantages of a low-temperature thermochemical process coupled to a direct-cycle gas reactor for hydrogen production. They reached a preliminary conclusion that the hydrogen product cost in a cogenerating plant would be less using a low-temperature cycle. This result, however, is contingent on moving the development of low-temperature cycles and the supporting experimental database from laboratory-scale demonstration to engineering-scale.

With this caveat, the researchers focused on a specific low-temperature cycle, i.e., the copper-chloride cycle (Cu-Cl), which they had identified as particularly promising based on laboratory experiments. They designed a combined plant layout and performed computer simulations to analyze efficiency and operability at full power and partial power for the combined plant. A schematic of the plant equipment layout is shown in Figure 1. This work showed several advantages compared to the representative sulfur-iodine high-temperature thermochemical cycle,

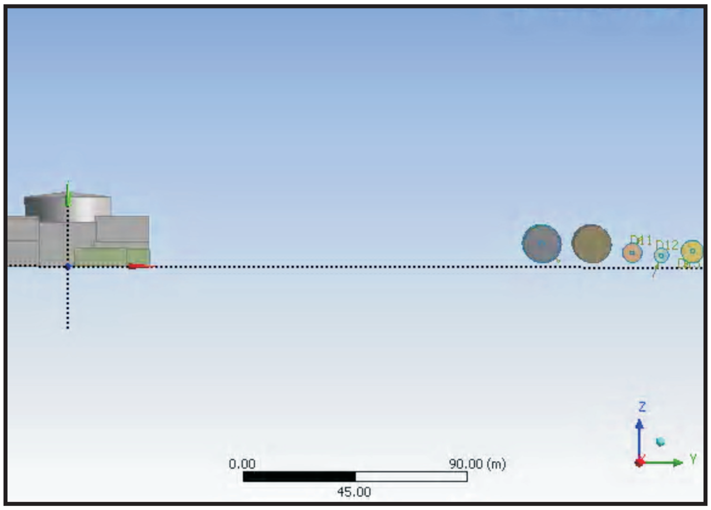


Figure 1. Configuration of building and storage vessels used in chemical release study.

including greater combined electricity/hydrogen production efficiency, reduced chemical plant complexity, less challenging materials requirements as a consequence of operation at reduced temperature, and less noxious intermediate products. Partly as a consequence of these findings, the development of the copper-chloride cycle is continuing in the U.S. and internationally.

In the second phase of this project, researchers investigated a competing high-temperature cycle with the goal of comparing performance to the low-temperature system. They focused on the high temperature steam electrolysis (HTSE) plant (see Figure 2) coupled to a very high temperature reactor (VHTR)—one of two DOE reference plant concepts for hydrogen production. They identified the components in the plant, developed models for the components, and integrated the models to obtain a working steady-state simulation for the combined plant. Using computer simulations, they investigated the sensitivity of overall plant efficiency to equipment configuration and they investigated how best to operate such a plant. Such an understanding is required before a prototype plant can be built.

A significant finding was that the high-temperature process heat exchanger as it appears in the reference design on the nuclear side of the plant might be eliminated. A plant configuration was developed that eliminates this heat exchanger and as a consequence significantly improves safety and maintainability of the nuclear side at the same time—potentially reducing capital costs. This simplification is based on the observation that 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 conventional heat exchanger technology is applicable, the HTSE process could be engineered to require minimal high-quality reactor heat. Because a relatively small amount of high-temperature heat is needed, this heat might be added by electrical heaters in the chemical plant—without a significant efficiency penalty.

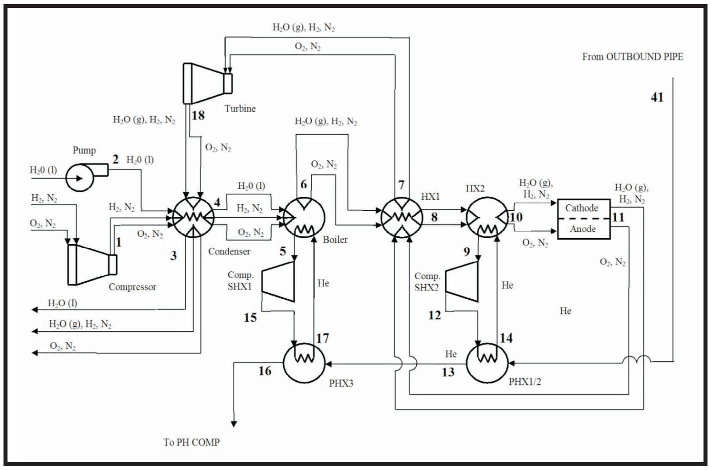


Figure 2. High temperature steam electrolysis plant equipment configuration.

The operability of this coupled nuclear-hydrogen plant was also characterized. Researchers developed a control strategy for meeting changes in hydrogen demand at the plant fence, which they evaluated with the GPASS dynamic simulation code. The goal was to maintain near-constant temperatures on the hot side of the plant using an inventory-control strategy in the VHTR and flow-control in the HTSE plant. Active electrolyzer cell area fluctuated such that electrolyzer joule heat varied linearly with hydrogen production rate. An initial simulation in which material flow rates varied linearly with hydrogen production rate showed that hot-side temperatures can likely be maintained near constant as shown in Figure 3.

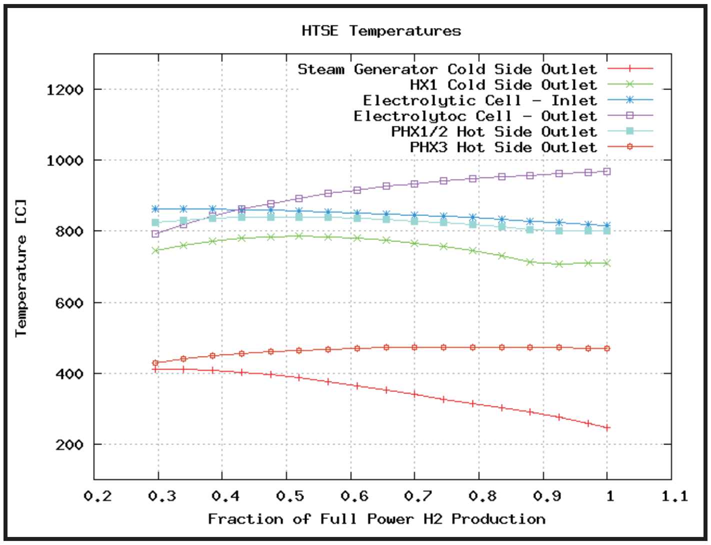


Figure 3. Temperatures in high temperature steam electrolysis plant.

There was also a finding that might guide future systems integration studies. It became clear in the course of this research that requirements to meet hydrogen production demand at the plant fence will influence the control strategy for operating the coupled plant. Thus, a good understanding of the hydrogen market that the plant will serve is needed. Future systems integration studies should attempt to define a set of demand requirements that a plant would need to meet. These would be based on projections for hydrogen demand on a daily, weekly, yearly, and geographic basis and on the role of local storage in mitigating the impact of temporal swings in hydrogen demand on plant operation.

Planned Activities

This I-NERI project has concluded.

INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE

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

PI (U.S.): Milorad Dzodzo, Westinghouse Electric Corp.

PI (Italy): Fosco Bianchi, Ente per le Nuove Tecnologie, l'Energia e l'Ambiente (ENEA)

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

Project Number: 2006-001-I

Program Area: AFCI

Project Start Date: May 2006

Project End Date: March 2010

Research Objectives

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 (i.e., with the primary loop entirely internal to the 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 pressure within the small spherical containment increases from the steam release through the break. These two pressures equalize relatively quickly, nullifying the pressure differential that drives the coolant egress, thus terminating the LOCA without any coolant injection or 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 the SIET's existing AP600 test facility to represent the characteristics of an integral, rather than a loop-type, reactor and the associated vessel–containment coupling. Researchers will develop an analytical program to guide testing. The four-year project duration supports the IRIS goal of submitting the Design Certification application in 2012.

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 quality assurance (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

Research Progress

The primary effort to date was to establish requirements and a conceptual design for the small break LOCA experimental facility, designated SPES-3. The resulting design is for a full height, 1:100 volumetric scale model of the following components, as shown in Figure 1:

- Primary circuit
    - Reactor vessel and internals
    - Core
    - Reactor coolant pump

    - Circulation paths
    - Pressurizer
- Secondary circuits
    - Steam generators
    - Feed line
    - Steam line
- Emergency systems components and piping
    - Emergency boration tanks
    - Emergency heat removal systems
    - Refueling water storage tank
    - Automatic depressurization systems
- Containment system simulated by separate tanks and piping
    - Reactor cavity
    - Dry well
    - Pressure suppression system
    - Long-term gravity make-up system
    - Passive containment cooling

The team scaled the design to preserve key parameters, including fluid thermodynamic conditions (e.g., temperature, pressure, and enthalpy), power-to-volume ratio during transients (not fully preserved during full-power steady state due to limits on the power available to the SIET facility), power-to-flow ratio, fluid transit times, heat flux, and differential pressures.

Researchers used RELAP models of the IRIS design to perform pre-test analyses to develop a test matrix and to guide the test facility design. Now that the preliminary design is complete, they are developing a RELAP model of the facility to compare results with those of the full IRIS model. The team will use the comparison results to confirm and finalize the facility design before starting equipment procurement in 2008.

They also completed a stage gate review, a major milestone that allows moving forward with equipment procurement.

Planned Activities

The team will use the IRIS and RELAP facility models to refine the facility design. They will then procure components, assemble equipment, and construct and commission the test facility. The Italian partners will have the main responsibility for this task, with the very substantial cost of materials and refurbishing borne by ENEA.

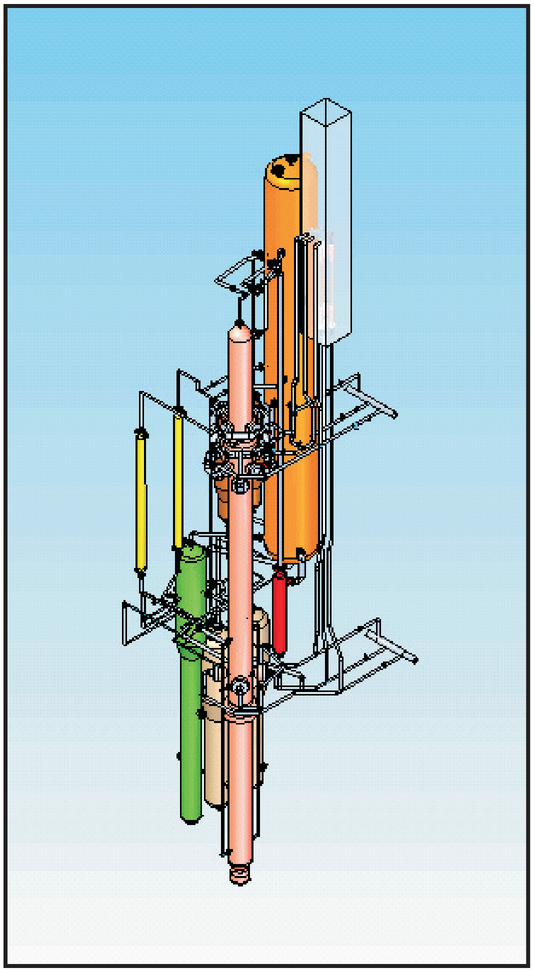


Figure 1. SPES-3 scale model.

After commissioning, the main task will entail performing the test matrix, including shakedown tests. The team will conduct separate effects tests as part of the shakedown program, and then follow with various aspects and phases of the integral tests. Analyses conducted during and after the tests will verify, interpret, and guide the testing.

The final task will be to prepare a comprehensive test report which will be part of the safety documentation package submitted to the NRC for final design approval.

The team will review and update existing QA plans as necessary to satisfy the IRIS integral testing needs, and then prepare a project-specific QA manual. The plan will address test planning and execution, as well as the configuration control of theoretical studies such as pre- and post-test analyses.

INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE

Advanced Nuclear Fuel Properties Measurements and Fuel Performance Modeling

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

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

Collaborators: Los Alamos National Laboratory, Pacific Northwest National Laboratory, Oak Ridge National Laboratory, Argonne National Laboratory

Project Number: 2006-002-E

Program Area: AFCI

Start Date: January 2007

End Date: December 2010

Project Abstract

The behavior characteristics of various advanced fuel forms must be effectively modeled to determine in-pile performance. Considerable effort has been extended to model uranium oxide (UO_2) fuels, including recent studies of the thermal conductivity of irradiated UO_2 as a function of burnup and irradiation temperature. The next step in fuel performance code development and validation is for mixed oxide fuel (MOX), which, together with metal alloy fuel, are under consideration in the Global Nuclear Energy Partnership (GNEP) and act as a bridge to other advanced fuels. These include minor actinide (MA) transmutation fuel types such as advanced MOX, advanced metal alloy, and inert matrix fuel (IMF), in addition to nitride and carbide ceramic fuels that may also have application to space energy systems. The purpose of this project is to promote 1) the effective use of international resources for the fabrication and characterization of fuel types and 2) development and validation of fuel performance codes.

The U.S. Nuclear Regulatory Commission (NRC) relies on the FRAPCON and FRAPTRAN codes for fuel licensing. Further development is required to support MOX and metal alloy fuels, in view of the anticipated fuel definition and licensing process for GNEP and the framework of research and development activities. One of the main European nuclear fuel performance codes is TRANSURANUS. ITU has begun applying this code to MOX fuel. Further update with the thermo-physical properties of MOX and metal alloy fuels with specific microscopic features is necessary, along with models for helium generation and release.

In order to improve understanding of the basic mechanisms behind materials properties and fuel performance and to predict the behavior of fuel material, researchers will undertake a multiscale modeling approach covering a spectrum of systems from ab initio atomistic calculations to macroscopic models.

Development of Novel Transmutation Systems for Sustainable Nuclear Fuel Cycles

PI (U.S.): Jon Carmack, Idaho National Laboratory (INL)

PI (EURATOM): D. Haas, Joint Research Centre Institute for Transuranium Elements (JRC-ITU)

Project Number: 2006-003-E

Program Area: AFCI

Start Date: January 2007

End Date: January 2010

Project Abstract

This project seeks to develop transmutation fuel systems to support future nuclear reactor systems. Economic sustainability and increases in operating efficiencies may be empowered by novel developments in fuel matrices and fuel system configurations. This project provides the mechanism for research collaboration in the areas of nitride fuel development, dispersion fuel development, vibro-pac fuel development, target system development, and advanced claddings. The collaboration will be focused on the following areas: fabrication, characterization, and irradiation performance of fuel and fuel systems. Irradiation testing of these fuel compositions may be accomplished following the initial three year period under follow-on agreements.

Nitride fuel R&D will develop fabrication processes that result in structurally stable fuel pellets. The research will focus on fabrication process development at small scale and structural characterization of the resulting pellets. Low temperature or low heat fabrication processes will be developed. Research on innovative processes (microwave processing, combustion synthesis, etc) will continue.

High-temperature dispersion fuels research will focus on design concept and laboratory-scale fabrication tests. Some initial irradiation of the fuel samples in ATR will also be conducted to assess the design options. Assessment of the candidate matrix materials for dispersion fuels will continue and the results of the FUTURIX-MI tests in the Phenix reactor will be incorporated into the fuel design considerations.

Vibrationally compacted fuels are amenable to remote fuel fabrication. Sphere-pac fabrication may offer considerable simplification in fabrication. Performance data on sphere-pac fuels, especially for transmutation applications are quite limited. Small fuel samples will be fabricated and characterized.

The baseline transmutation concept under GNEP relies on homogeneous core with group TRU included in the fuel. An alternative option may be to put non-fertile minor actinide into the reactor using inert matrix materials. A longer-term option may also be partitioning americium (possibly with curium) and transmuting Am targets in a moderated pin.

Advanced nuclear fuel claddings have the potential to extend fuel life and burnup maximizing fuel resources and maximizing actinide destruction. ODS and advanced ferritic-martensitic materials are two such advanced claddings. Research and development of advanced cladding will include fabrication and joining development as well as material property characterization. Material from in-process material irradiations will be used to begin characterization of irradiated material properties.

Development of Oxide Fuels for Transmutation in Fast Reactors

PI (U.S.): Jon Carmack, Idaho National Laboratory (INL)

PI (EURATOM): D. Haas, Joint Research Centre Institute for Transuranium Elements (JRC-ITU)

Collaborators: None

Project Number: 2006-004-E

Program Area: AFCI

Start Date: January 2007

End Date: January 2010

Project Abstract

This project establishes the mechanism for minor actinide-bearing mixed oxide fuel development collaboration between the United States and EURATOM. The purpose is to provide oxide fuels for testing in the AFC-2 series of irradiations to be carried out under the transuranic fuel development program in the Advanced Test Reactor at INL. This project will examine three main areas:

Oxide Fuel Selection and Fabrication. Assessment will include analysis of neutronic behavior (plutonium burn rate, reactivity swing), fuel thermal performance, fuel irradiation performance, and in-core corrosion resistance. Minor actinide mixed oxide fuel compositions will be fabricated for inclusion in the AFC-2 test series.

Oxide Fuel Characterization and Modeling. Experimental characterization of oxide fuel microstructure, thermal properties, and mechanical properties, corrosion testing, and ion-beam irradiation will be carried out to satisfy requirements for in-reactor irradiation testing and provide a basis for fuel behavior modeling. Special attention will be paid to the characterization of fuels having varying oxygen-to-metal (O/M) ratios. Fuel modeling will be conducted using finite element analysis to determine the thermo-mechanical behavior of the fuel. Modeling of thermodynamic and transport properties will also be conducted.

Irradiation Testing and Post-Irradiation Examination. An assessment of transmutation fuel feasibility requires in-reactor irradiation due to the extremely complex nature of the in-core fission environment and the inability to simulate this environment with out-of-pile testing. This project will establish the basis of research for the AFC-2 irradiations to be conducted following the initial three-year period.

INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE

Lead-Cooled Fast Reactor Concept Design and Evaluation

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

PI (EURATOM): Stefano Monti, Ente per le Nuove Tecnologie, l'Energia e l'Ambiente (ENEA)

Collaborators: Lawrence Livermore National Laboratory (LLNL), Del Fungo Giera Energia S.p.A.

Project Number: 2006-005-E

Program Area: Generation IV

Start Date: January 2007

End Date: December 2009

Project Abstract

The purpose of this project to conduct trade studies for a near-term deployable demonstration lead fast reactor (LFR). The demonstration reactor will implement innovative engineering features that exploit the characteristics of pure lead as the primary coolant to show its economic potential and industrial attractiveness. Researchers will select a suitable power level to confirm the key features of the prospective LFR fleet. The reactor will be initially loaded with either standard enriched uranium or U-Pu fuels. However, it will be designed to accept different fuels, especially minor actinide (MA)-bearing fuels, as soon as they are available. The project is focused on defining the core and primary system configuration, with the following objectives:

- To identify the most promising design features of the future LFR to demonstrate its feasibility
- To create conditions and incentives for construction of commercial LFR prototypes
- To guide the selection of options based on experimental results

The demonstration facility will be flexible to accommodate subsequent modifications and improvements in order to allow testing of new components and systems, as well as to serve as a test facility for the continuous improvement of prospective industrial/commercial reactors. The core layout will feature geometrical and functional flexibility in order to test different fuels and fuel assemblies, particularly cores with MA assemblies. In addition, easy access to the core and core instrumentation will allow it to serve as a fast-spectrum fuels and materials irradiation facility.

8.0 U.S./France Collaboration

Then-U.S. Secretary of Energy, Spencer Abraham, and 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; 16 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 that began in FY 2004 have been completed during the past year. Work on three collaborative projects initiated in FY 2006 is continuing.

A listing of the I-NERI U.S./France projects that are currently underway follows, along with summaries of the accomplishments achieved in FY 2007.

Directory of Project Summaries

INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE

Hydrogen Process to High-Temperature Heat Source Coupling Technology

PI (U.S.): Mike Patterson, Idaho National Laboratory

PI (France): Dominique Barbier, Commissariat à l'énergie atomique (CEA) Cadarache

Collaborators: University of Nevada Las Vegas (UNLV)

Project Number: 2004-001-F

Program Area: NHI

Project Start Date: August 2004

Project End Date: July 2007

Research Objectives

This project was designed to develop the technology necessary to couple a high-temperature heat source to hydrogen production processes for the Nuclear Hydrogen Initiative. The primary focus of this research is to develop the Sulfur-Iodine (S-I) cycle process technology. Specific objectives include:

- Propose design solution schemes, including component connections for the following interfaces Compare characteristics and select component arrangements to reduce heat loss.

Reactor → Intermediate Heat Exchanger →

High Temperature Step → Medium-Temperature Step →

Low-Temperature

- Evaluate heat transmission and exchangers, including technical and industrial feasibility, flexibility of coupling schemes, and conversion energy losses for the design solutions. Develop potential scheme designs.
- Develop and utilize a model to evaluate the complete heat balances for the different schemes, analyzing the use of energy (electricity and/or heat) in relation to hydrogen production. This model will improve understanding of the process behavior during normal operation, transient, and accident conditions. The information from the models and simulations will provide the data needed to perform availability and safety analysis, as well as information and interface data for the economic evaluation and cost estimation work teams.

Research Progress

Researchers performed a thermal/mechanical analysis of conventional and advanced heat exchanger designs, with emphasis on the following subjects:

- kinetics, catalysts , and corrosion behavior
- design and modeling of a shell and tube heat exchanger
- technological realization of a compact silicon carbide (SiC) plate heat exchanger

The selection of liquid salt was based on several criteria, including temperature, heat transport capacity, viscosity, thermal conductivity, chemical behavior, cost, and French regulations. Three lithium, sodium, and potassium salts were selected for study, focusing on their application in the intermediate circuits of the very high temperature reactor (VHTR) and the gas-cooled fast reactor (GFR): LiF-NaF-KF, NaF-KF-ZrF_4, and KF-ZrF_4.

Intermediate circuit studies were performed to understand how components are connected and assembled in the overall plant. Researchers examined configurations for effects on energy efficiency and overall facility safety, including increasing helium (He) pressure from 27 bar to 47 bar in order to limit the reactor outlet temperature to 900°C. They also examined the He intermediate loop pipe design to limit heat losses below 1 MWth/km.

CEA developed the CYCLOP/COPERNIC modeling tools to permit multi-criteria optimization of power generation and cogeneration designs, including hydrogen facilities, conventional industries, and desalination plants.

Researchers have also performed initial safety studies based on a defense-in-depth approach, addressing normal operation, abnormal operation, accidents, and severe accidents. They evaluated inherent safety features, operational controls, and procedures to mitigate hypothesized events and initiated transients calculations.

For coupling studies related to sulfuric acid decomposer development, researchers continued evaluating material corrosion in sulfuric acid S-I environments. They also completed a design of a materials and component test flow loop for long-term testing in hazardous chemical environments (e.g., H_2SO_4, SO_2, O_2). For the technical data on candidate liquid salts, they completed measurements of liquid salt properties and studies on redox corrosion control methods. The design selection of a dedicated test loop included comparisons of intermediate circuit design studies and collaboration on modeling approaches and shared results on safety analyses. In addition, they refined the steady-state simulation tools for the combined facility and incorporated transient response elements for the purposes of developing safety and control elements.

For high-temperature electrolysis (HTE) coupling studies, researchers completed the conceptual design of the coupling circuits based on the electrolyzer technological design. They also compared the safety approaches between S-I and HTE.

The 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). This FY2007 research included:

NHI Materials and Component Development Plan. The NHI Materials and Component Development Plan describes a decision process for evaluating the technical readiness or maturity of components (e.g., heat exchangers, chemical reactors, valves) for use by the Nuclear Hydrogen Initiative.

Measurement of Alloy Mechanical Properties. Results of the tensile data measurements showed that the austenitic alloys Waspaloy, C-22, and C-276 all had sufficient tensile strength at 800°C to be used for high-temperature nuclear hydrogen production applications, although Waspaloy exhibits reduced ductility in relation to the other metals at that temperature.

Corrosion and Crack Growth Studies in HIx Solutions. In an initial round of materials corrosion testing, samples composed of various materials were exposed to the fluids and environments typical of the hydrogen iodide (HI) decomposition section of an S-I process using extractive distillation. The results of these tests showed that various forms of tantalum (Ta, Ta-2.5W, and Ta-10W) and SiC are appropriate for use in the iodine separator and phosphoric acid concentration processes. Also, Hastelloy B-2, C-22, and C-276 may be used for gaseous HI decomposition environments.

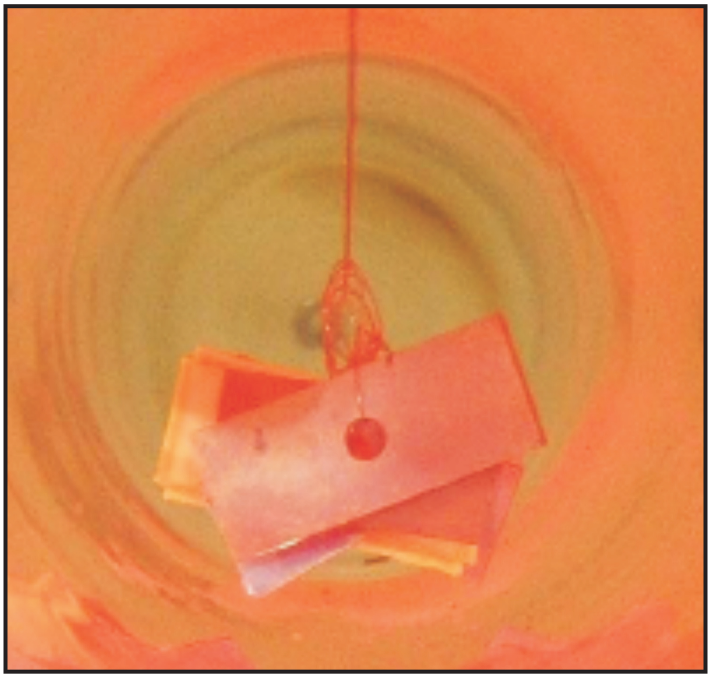


Figure 1. Coupon air oxidation tests at 850˚C.

Numerical Analysis of Advanced Heat Exchanger Concepts. Researchers analyzed three basic sulfuric acid decomposer types: the Sandia bayonet, the Ceramatec ceramic compact decomposer, and the shell-and-tube heat exchanger. Each of these device concepts use SiC as the structural material for the flow channel(s) and assume the use of a platinum (Pt)-based catalyst on either a solid support or as part of a coating. The numerical analyses of these concepts included examinations of thermal and mechanical stresses and chemical reaction conversion based on assumed reaction kinetics.

C-SiC Materials for High-Temperature Heat Exchangers. A two-dimensional transient porous media model was developed for studying ceramic compact heat exchanger transient flow and heat transfer which is being applied to examine a C-SiC heat exchanger design. The interest in this particular technology is for application to sulfuric acid decomposition, liquid salt heat transfer, and, as a corollary, development of chemical vapor deposition (CVD) techniques for applying pyrolytic carbon coatings for the purposes of sealing heat exchanger surfaces against porous gas transfer through the relatively porous C-SiC heat exchanger plates. Heat exchanger fabrication work is underway employing the new CVD-coated plates.

Ceramatec Heat Exchanger Development for Application to NHI Hydrogen Production Processes. Research on this topic in FY2007 concerned five areas: 1) further examination of materials corrosion due to exposure to sulfuric acid at elevated temperature, 2) development of porous ceramic structures for catalyst support, 3) further design revisions of the Ceramatec decomposer, 4) fabrication of test wafers, and 5) joining processes for application to metal/ceramic joints and individual heat exchanger plates. Some key findings of this work are as follows.

- Team members designed and performed experimental tests under scaled conditions similar to the working conditions of the prototype intermediate heat exchanger (IHX). Dimensionless analysis, computational fluid dynamics simulations, and bench test results compared well with the design parameters.

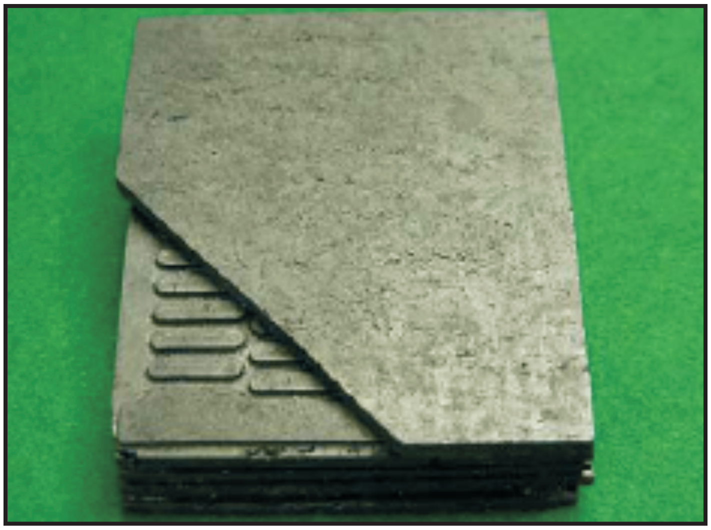


Figure 2. SiC heat exchanger test specimen.

- Researchers developed self-catalytic materials for thermochemical water splitting using the S-I process. They also examined the microstructure of a second set of Pt-added alloy heats (Alloy 800H+1% Pt, Alloy 800H+2%Pt, Alloy 800H+5% Pt, Alloy 617+1%Pt, and Alloy 617+2%Pt, all measured by weight) using scanning electron microscopy (SEM). Preliminary results show that the new heats have much lower carbon contents than the first heat and are much more representative of what would be achievable in commercial practice.

Planned Activities

This I-NERI collaboration was completed in 2007, but additional work is progressing under different initiatives.

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

PI (U.S.): R. Klann, Argonne National Laboratory (ANL)

PI (France): A. Lyoussi, Commissariat à l'énergie atomique (CEA) – Cadarache Research Center

Collaborator: CEA-Valrho

Project Number: 2004-002-F

Program Area: AFCI

Project Start Date: October 2004

Project End Date: September 2007

Research Objectives

The goal of the OSMOSE program (Oscillation in Minerve of Isotopes in "Eupraxic" Spectra) was 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 measured 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. These measurements provided the experimental data that is needed for improving basic nuclear data files.

The OSMOSE program is generic; that is, it measures 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 provides precise experimental, integral, absorption cross-section data for a majority of the heavy nuclides important to reactor and nuclear fuel cycle physics, as shown in Table 1.

Element	Isotope
Thorium	^{232}Th
Uranium	^{233}U ^{234}U ^{235}U ^{236}U ^{238}U
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.

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 FY2007, researchers made technical progress on the OSMOSE project in the areas of reactor modeling, sample fabrication, experimental measurements, and data analysis. Following is an overview of accomplishments in each area.

Reactor modeling and analysis for pre-experiment planning was performed for two reactor configurations: 1) the R2-UO$_2$ configuration (representative of an over-moderated thermal spectrum) and 2) the MORGANE-R configuration (representative of a thermal spectrum with a large epithermal component). These are the next two configurations scheduled for experimental measurements in the MINERVE facility in 2008.

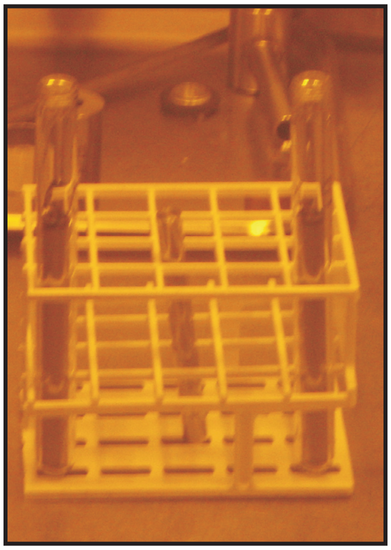


Figure 1. Pins containing a curium sample (a, b) and reference pin (c).

Researchers completed OSMOSE sample pellet fabrication and analysis (isotopic composition vs. mass ratio) for the first two sample batches, except for ThO_2. The last set of pellets is comprised of five samples: 1) UO_2 + $^{233}UO_2$, 2) UO_2 + $^{243}AmO_2$, 3) U_3O_8 + $^{244}CmO_2$, 4) U_3O_8 + $^{244+245}CmO_2$, and 5) the U_3O_8 reference. The OSMOSE furnace and the uniaxial three-part die were used during the fabrication of the third set of samples. For the samples containing curium (Cm), researchers decided not to sinter the pellets because of the lack of radioprotection of the OSMOSE furnace. As a consequence, a new press was installed in a hot cell of the ATALANTE C10 line which is suitable for the fabrication and the study of high neutron activity compounds. The green pellets for two samples (^{244}Cm and $^{244+245}$Cm doped compound) and a set of uranium sesquioxide for calibration were fabricated.

To help reduce uncertainties associated with the experimental results, ANL is providing post-fabrication destructive analysis of selected OSMOSE sample pellets for isotopic composition and impurities. The sample analysis activity involves providing corroborative compositional data for one pellet composed of isotopically natural UO_2, and eight pellets composed of the natural uranium matrix doped with specific minor-actinide isotopes (U-234, Th-232, Pu-240, Pu-242, Np-237 [2 levels] and Am-241 [2 levels]).

After a shipping delay while authorizations were pending from Safety Authorities in France, the OSMOSE sample pellets were delivered to ANL in December 2006 and the shipping container was returned to CEA.

The analysis of the natural uranium pellet to determine the uranium mass fraction, uranium isotopic composition, and select impurity elements has been completed. Impurity results are shown in Table 2. Measurements with the Th-232 and U-234 doped pellets have also been completed and the Pu-240 and Pu-242 doped pellets have been processed and analyzed. In general, the results were in agreement with the CEA reported values; however, there were a few significant findings that need to be resolved, such as: 1) the isotopic composition of the Pu-240 doped pellet sample differs from the CEA listed values and 2) the mass values of the Pu-242 doped pellet do not agree.

The schedule for conducting OSMOSE measurements in the MINERVE facility is dictated by commitments for facility operations for several programs at CEA-Cadarache. The R1-MOX configuration was previously loaded in MINERVE in July 2006 and calibration samples were oscillated in early FY 2007. The first and second batches of OSMOSE samples were then oscillated in the R1-MOX configuration from January 2007 to March 2007, followed by the third

	Element	Analysis Result, µg/gU
Absorber Impurities	B	< 0.5
	Cd	< 0.02
	Li	0.44
	Gd	0.94
	Sm	< 0.02
	Eu	< 0.02
	Dy	< 0.02
Banal Impurities	Ag	NR
	Al	11.5
	Bi	< 0.02
	Ca	12.0
	Co	0.09
	Cr	2.3
	Cu	1.7
	Fe	18.5
	In	< 0.02
	Mg	4.7
	Mn	0.15
	Mo	0.25
	Ni	5.7
	Pb	0.47
	Si	3.3
	Sn	0.19
	Ta	NR
	Ti	4.6
	Th	NR
	V	0.15
	W	20.8
	Zn	2.4
	Zr	NR
	Ba	< 0.5
	K	< 10
	Na	< 10
	Sr	0.04

Table 2. Impurity results.

NR = Not reported due to low recovery in separations

NC = Not calculated; spike recovery not calculated because element present in sample batch in December 2007. Preliminary comparisons of calculated values to the measured results for the R1-MOX configuration are presented in Table 3. As noted from the chemical analysis, the pellet mass for the Pu-242 sample did not agree with the CEA value, which might be the source of the large difference in the Pu-242 result.

In order to qualify the relevant experimental configurations for achieving the proposed goals, the researchers performed calculations to investigate the similarity of the flux spectra of the OSMOSE configurations with the neutron energy distributions characterizing existing thermal and fast reactors proposed under the Department of Energy's advanced reactor programs. From the direct comparison of the flux spectra calculated at the core center, the team observed that for fast reactors, the fraction of neutrons below 1 keV is practically negligible and the peak of the distribution is about 100–200 keV. In the case of the OSMOSE configurations, the flux spectra below 1 keV is still relevant and the peak of the distributions is about 1 MeV (becoming more pronounced in the case of R1-MOX and MORGANE/R configurations). The researchers concluded that the OSMOSE flux spectra look much more similar to pressurized water reactor (PWR) spectra than to the spectra typical of fast systems.

Direct comparison of the flux spectra is not completely appropriate to quantify the relevance of the proposed experimental configurations for their similarity with actual reactors. For this purpose, researchers carried out a representativity study based on the comparison of sensitivity profiles associated with the integral parameters of interest. In addition to k_{eff}, they performed a representativity analysis with respect to the parameter $\eta = \nu\Sigma_f\Phi/\Sigma_a\Phi$ calculated at the sample location of the experimental configurations and in relevant core positions of the actual thermal and fast reactors. The parameter η is calculated with the sample cross-sections $\nu\Sigma_f$ and Σ_a. Table 4 shows representativity factors for R1-MOX compared to other reactor types.

OSMOSE Samples	Calculated Reactivity-Worth (pcm)	Experimental Reactivity-Worth (pcm)	(C-E)/E in %
Np237-1	-18.42	-17.33	6.31
Np237-2	-103.30	-97.57	5.88
Pu239	65.72	64.46	1.95
Pu242	-29.14	-24.20	20.39
U234	-24.73	-22.93	7.84
U-Th232	-17.22	-17.23	-0.05
URE	96.70	97.98	-1.32
Am241-1	-33.84	-34.16	-0.93
Am241-2	-102.70	-96.31	6.63

Table 3. C/E comparison of the OSMOSE samples in the R1-MOX configuration.

Sample	ABTR	SFR	EFR	GFR	LFR	ADS	PWR
Th-232	0.1976	0.0748	0.2421	0.2104	0.1659	0.0290	0.9302
U_{th}	0.0950	0.0357	0.1154	0.0993	0.0791	0.0132	0.9117
U-234	0.0974	0.0368	0.1188	0.1022	0.0809	0.0135	0.9131
URE	0.0281	0.0103	0.0342	0.0290	0.0226	0.0039	0.7990
UO_2	0.0925	0.0347	0.1123	0.0960	0.0769	0.0129	0.9158
Np-237_1	0.0972	0.0366	0.1183	0.1015	0.0808	0.0135	0.9103
Np-237_2	0.1152	0.0440	0.1415	0.1231	0.0957	0.0159	0.8918
Pu-238	0.0859	0.0324	0.1058	0.0918	0.0705	0.0119	0.9218
Pu-239	0.0315	0.0117	0.0384	0.0330	0.0255	0.0044	0.8302
Pu-240	0.1134	0.0427	0.1380	0.1185	0.0943	0.0157	0.6938
Pu-241	0.0625	0.0234	0.0759	0.0645	0.0517	0.0087	0.8783
Pu-242	0.0921	0.0349	0.1125	0.0972	0.0765	0.0127	0.8929
Am-241_1	0.1013	0.0381	0.1231	0.1054	0.0842	0.0141	0.9092
Am-241_2	0.1183	0.0447	0.1440	0.1240	0.0985	0.0163	0.8866

Table 4. Representativity factors between OSMOSE R1-MOX and other reactors for η.

Low representativity factors (<0.2 on a scale from 0 to 1) were obtained in general with respect to k_{eff} and the parameter η between the OSMOSE configurations and all fast reactors, while a high representativity is shown as expected between the R1-UO_2 or R2-UO_2 configuration and the PWR. The obtained results lead to the conclusion that the reactivity worth measurements obtained from the R1-MOX and MORGANE/R configurations are not to be intended as measured in flux spectra characterizing actual fast reactors. It is expected that the additional configurations possible in MINERVE would be much more representative of fast reactors as they were designed to address harder neutron spectra. However, representativity calculations for these configurations were not performed as they were not part of the OSMOSE program.

Planned Activities

CEA will continue the measurement campaign on the OSMOSE program in FY 2008 with the completion of measurements for the Batch 3 samples in the R1-MOX and R1-UO_2 configurations. French researchers will complete measurements of the OSMOSE samples for the R2-UO_2 configuration and the CARMEN configuration. CARMEN is a new lattice that produces an epithermal spectrum to replace the MORGANE/R configuration.

U.S. participation in the OSMOSE program will not continue in FY 2008. The only planned activities are to issue a final report for the sample analysis and a final report for the DOE/CEA OSMOSE collaboration.

INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE

Thermal-Hydraulic Analyses and Experiments for GCR Safety

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

PI (France): Denis Tenchine, Commissariat à l’énergie atomique (CEA)

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

Project Number: 2004-003-F

Program Area: Generation IV

Project Start Date: January 2005

Project End Date: January 2008

Research Objectives

The objective of this collaborative research project was 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 for 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 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.

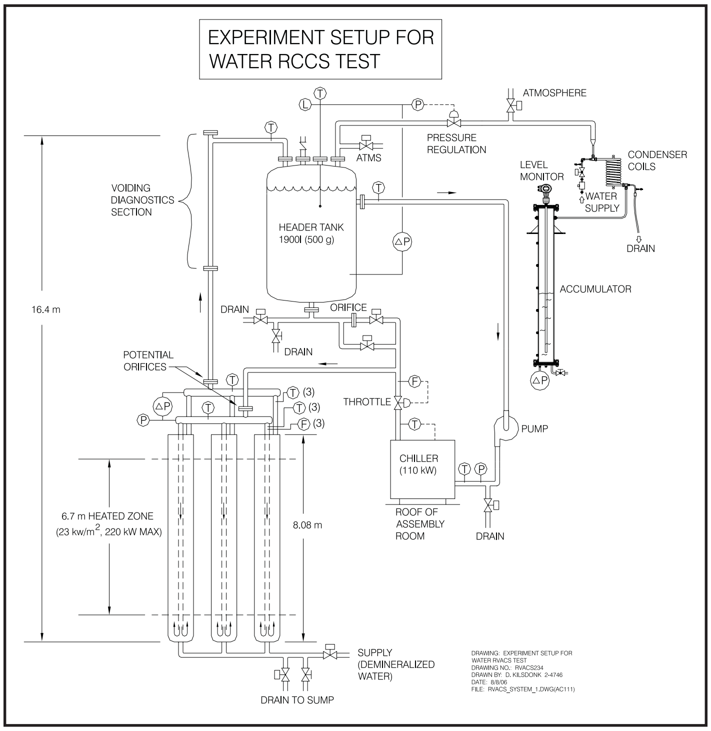


Figure 1. Sketch of NSTF system modifications for water-based RCCS testing.

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; a volumetric thermal source term passes data from the CFD approach to the porous media.

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.

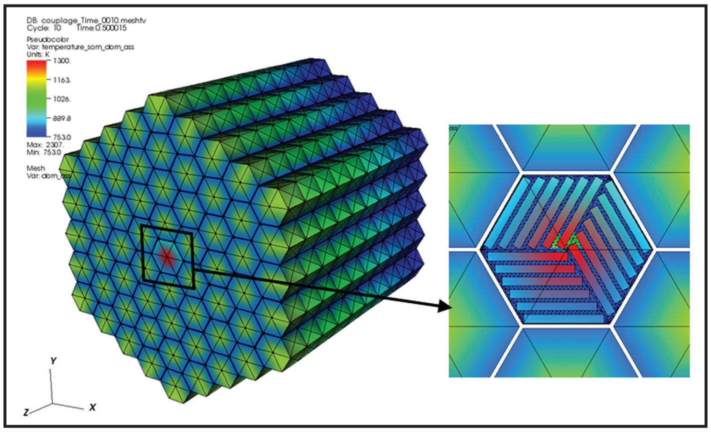


Figure 2. Porous media domain with coarse mesh, showing details in the central subassembly.

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/cm^3. Table 1 shows the maximum temperatures inside the pebbles.

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, was maintained by controlling temperature in the test section to within 0.1°C.

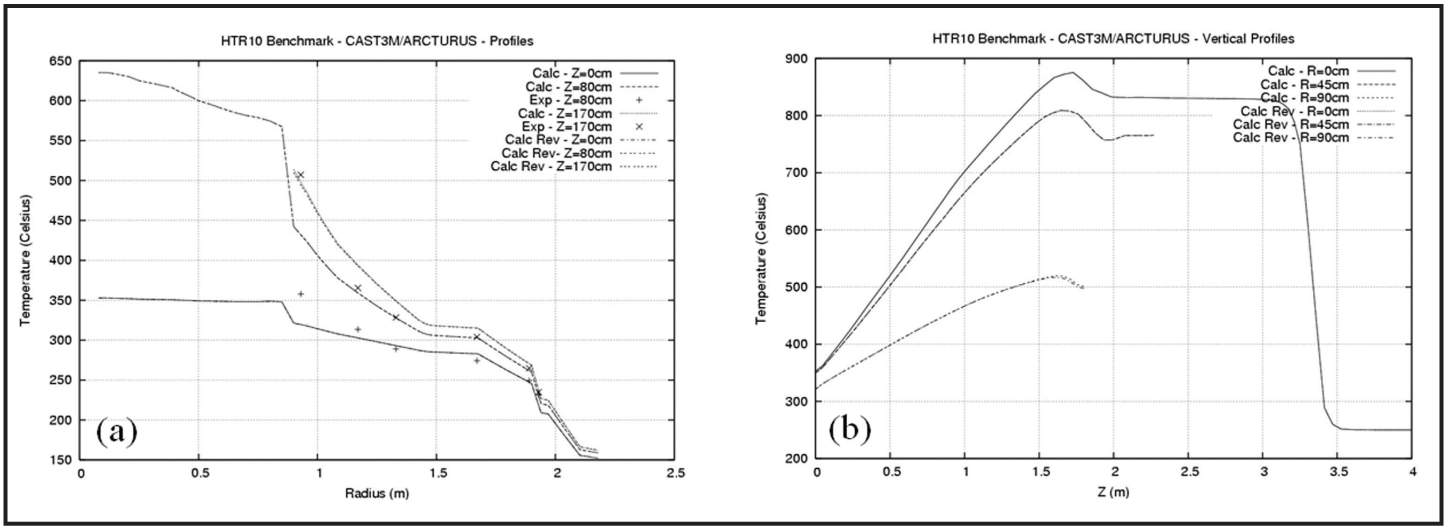


Figure 3. HTR-10–CAST3M/ARCTURUS benchmark results: (a) radial and (b) axial temperatures.

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 cross-flow 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.

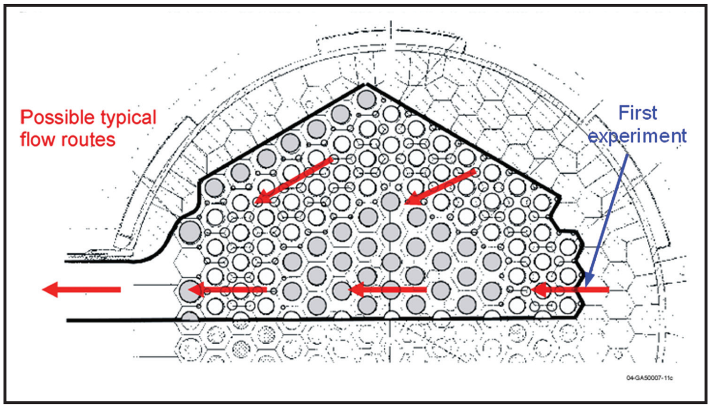


Figure 4. Examples of possible flow paths in the lower plenum of a typical prismatic VHTR concept.

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 refractive-index 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).

Planned Activities

There was no U.S. activity in this I-NERI project in fiscal year 2007 due to a decrease in funding. The project has terminated.

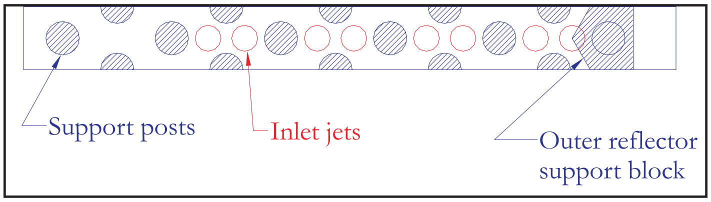


Figure 5. Plan view of scale model for the lower plenum experiment.

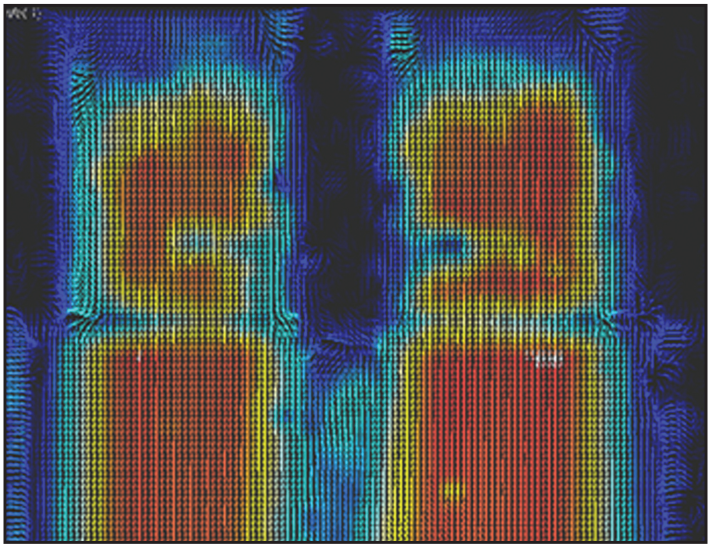


Figure 6. Mean velocity vectors of two inlet jets.

INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE

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'énergie 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. There has been an insufficient amount of physical property data to qualify candidate materials for gas-cooled fast reactor (GFR) designs. The goals of this project were 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 was to improve high-temperature 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 focused 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 was 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, radiation response of these alloys were be characterized 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

The researchers successfully performed GBE of Inconel Alloy 800H 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 (Σ3) and twin variants (Σ9 and Σ27) 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 Σ3 boundaries (0.64), which is about 4.4 times that of the as-received sample and approaches the theoretical limit for the Σ3 fraction (2/3).

Thermal stability of the GBE-treated samples was evaluated by annealing the samples at 850°C and 1,000°C for 4 and 6 weeks. Researchers selected two GBE-treated samples with 2- and 5-cycle TMP due to their special GBCD characteristics, i.e., the highest fraction of Σ1 and low-Σ CSLBs. They also tested as-received samples at the same time for comparison.

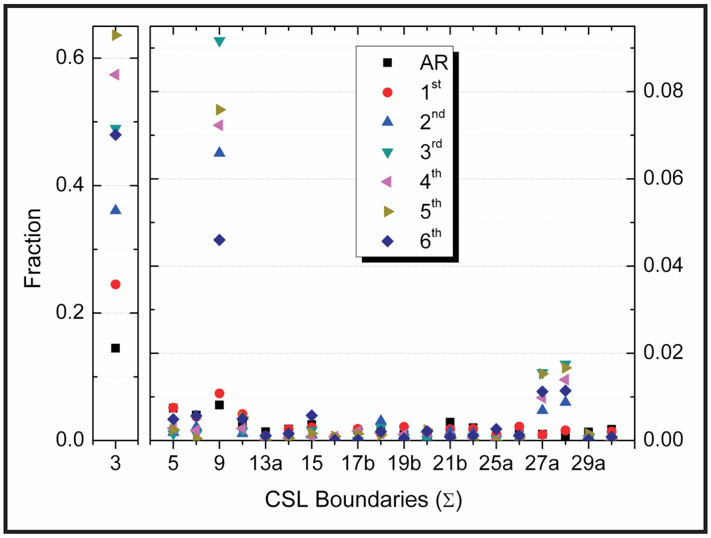


Figure 1. Fraction of low-Σ CSLBs of the as-received (AR) and GBE-treated samples with 1 to 6 cycles of thermomechanical processing.

Figure 2 shows the GBCD results of the annealed samples. Annealing slightly increases the fraction of random boundaries, but the induced GBCD variation is very small for samples with 5-cycle TMP compared to the other samples. This limited-time annealing test indicates that the use of GBE-treated sample with a low fraction of Σ1 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.

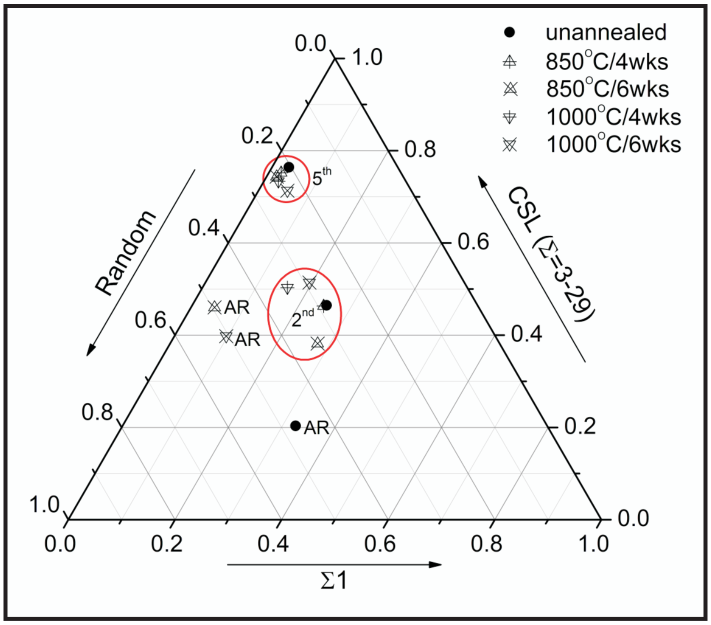


Figure 2. Thermal stability of the GBCD of as-received and GBE-treated samples with 2- and 5-cycle TMP (2nd and 5th) after exposure to air at 850°C and 1,000°C for 4 and 6 weeks.

In continuation of the work conducted earlier on the microstructural analysis of as-received, crept, and optimized alloys T91, additional transmission electron microscopy (TEM) analysis was performed. The results are very similar to those obtained by scanning electron microscopy (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.

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 Σ1 boundaries with misorientation angle <3° in the CSLE condition as compared to the AR condition. This increase results from the introduction of new subgrains as confirmed by OIM analysis, which shows a higher fraction of these boundaries in the CSLE condition and by TEM analysis. Figure 3 shows the TEM images of both the AR and the CSLE conditions showing the typical tempered lath structure with subgrains forming within these laths. There is a higher volume fraction of subgrains in the CSLE condition or conversely a higher volume fraction of regions in the AR condition free of any subgrains. There is enough evidence in the literature to show the presence of long-range internal stresses in subgrain structures, which have been proven experimentally by observing strongly curved/bowed dislocations in the vicinity of these boundaries, by X-ray line profiles and of HOLZ lines in convergent beam electron microscopy. Further TEM analysis

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

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

Sample designation scheme: crept samples (condition-temperature-stress)

* Test conducted to a strain of 2%; 'NA' not applicable

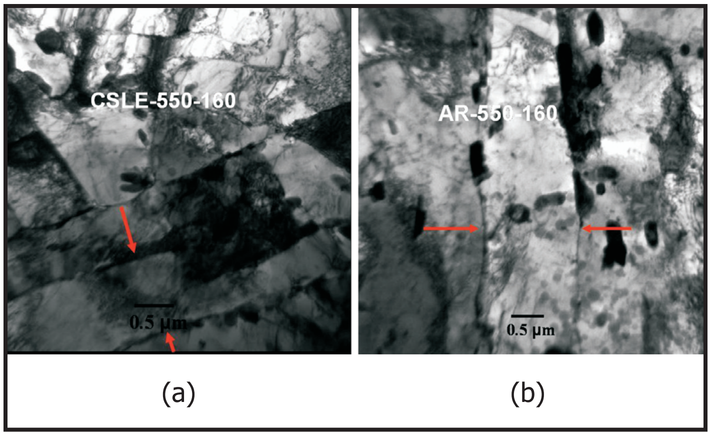


Figure 3. TEM images showing tempered lath sizes after creep test in a) CSLE-550-160 and b) AR-550-160.

also confirmed that the average subgrain size in the CSLE condition is smaller compared to the AR condition. There is also evidence that hardening due to long-range 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 CSLE condition by inducing long-range internal stresses which opposes the applied stress and thus reduces the effective stress for creep.

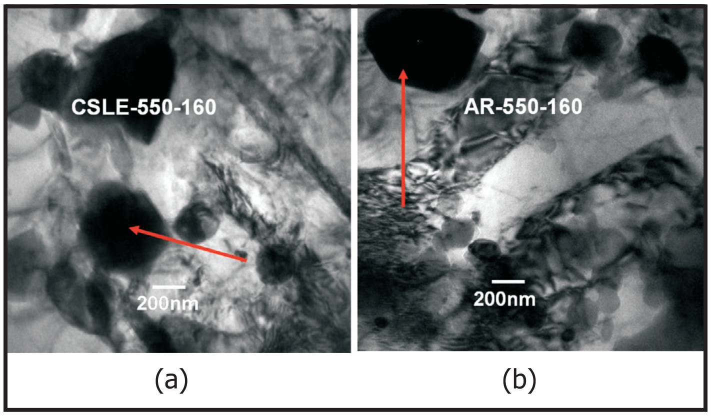


Figure 4. TEM images showing carbide sizes after creep test in a) CSLE-550-160 and b) AR-550-160.

Planned Activities

This project has concluded.

INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE

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

PI (U.S.): G.E. Apostolakis, Massachusetts Institute of Technology (MIT)

PI (France): F. Bertrand, Commissariat à l'énergie atomique (CEA)

Collaborators: None

Project Number: 2004-010-F

Program Area: Generation IV

Project Start Date: April 2005

Project End Date: April 2008

Research Objectives

The objective of this project was 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. Researchers evaluated the system under a range of scenarios, including loss-of-coolant accidents (LOCA), station blackout, and anticipated transients without scram (ATWS).

For GFRs to meet the high standards for safety assurance expected of new reactor designs, 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: 1) PRA models for passive systems must be developed and 2) 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 Nuclear Regulatory Commission (NRC) and International Atomic Energy Agency (IAEA) are working to establish probabilistic goals. It is uncertain whether a new plant design would need to satisfy both deterministic and probabilistic criteria, particularly if a design option meets probabilistic goals but fails the deterministic criteria.

Research Progress

According to an IAEA definition, passive systems are either composed of passive components and structures, or they use active components in a very limited manner to initiate subsequent passive operation. Of great importance are thermal-hydraulic (T-H) systems that fall under IAEA Categories B and C. These are characterized by moving working fluids and can contain moving mechanical parts such as check or relief valves.

Overall plant reliability should increase by replacing certain active systems with passive, as the external energy source is a primary contributor to unreliability of active systems. Passive system functionality does not rely on an external source of energy, but on an intelligent use of natural phenomena, such as gravity, conduction and radiation, which are always present. Although natural driving forces are always available for a passive system, they are normally weak, which is particularly significant for systems that depend on natural circulation of fluids. In addition, operators cannot control passive systems the way they can control the performance of active 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 functional failure" describes failure in terms of exceeding a system's load capacity 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 exceed the corresponding values of the capacity vector. The load vector reflects the epistemic uncertainties in the calculations while the capacity vector is fixed at the regulatory limits, i.e., the maximum temperature in the cladding and in the hot leg of decay heat removal (DHR) loops.

The reliability assessment of passive systems focuses on the thermal-hydraulic aspects for several reasons. The most important involve the uncertainties surrounding model predictions and system performance and the variety of possible failure mechanisms that may affect these systems.

The GFR design considered consists of a 600 MWth helium-cooled fast reactor, a Brayton cycle power conversion unit (PCU), and two 50 percent DHR loops (Figure 1). The core is designed to have very low pressure drop to maximize the natural circulation capability. A guard containment building with a design pressure of 2 MPa (20 bar) ensures that, after primary system depressurization, the backpressure would be high enough to accomplish DHR by natural circulation of helium.

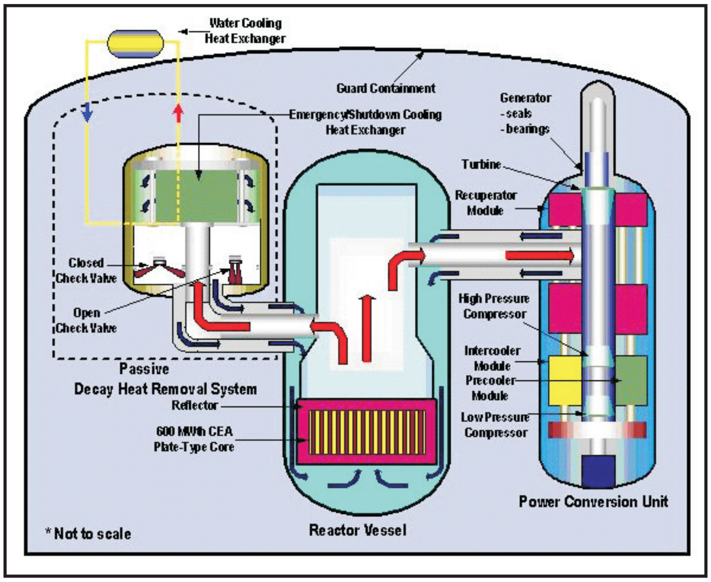


Figure 1. Schematic GFR system layout.

Each DHR loop consists of two separate circuits. The first connects to the pressure vessel through a coaxial pipe, with the outer portion the cold leg. Heat is delivered to a second water-filled circuit through a heat exchanger that discharges the heat to a pool outside the containment building. The Brayton power cycle PCU, with recuperator, precooler, and intercooler, provides energy to the generator and to the low- and high-pressure compressors.

Researchers have completed two studies. The first performed a time-dependent reliability evaluation of a two-loop passive DHR system as part of the iterative design process for the GFR. The system was modeled using RELAP5-3D and uncertainties in input parameters were assessed and propagated through the model using Latin Hypercube Sampling. An important finding was that flow would bypass the core through one DHR loop, if two loops operated in parallel, due to the smaller pressure loss through the DHR heat exchanger. This finding is a warning against modeling only a single lumped DHR loop and assuming that multiple loops will remove n times the decay heat. The initial design consisted of two 50 percent DHR loops working in parallel; this configuration was modified because of the core bypass possibility. Each DHR loop is now designed to extract 2 percent of the reactor nominal power (i.e., 12 MWth) at a back-pressure of 1.3 MPa.

Sensitivity analyses revealed that there are values of some input parameters for which failures are very unlikely. Figure 2 shows such a plot for the two leakages associated with each Monte Carlo realization (one for each DHR loop) for the maximum temperature in the cladding. On the horizontal axis, researchers plotted the maximum of the two leakages and on the vertical axis the minimum. The plus signs indicate a realization and the square around a plus sign indicates a failure of the cladding. Because no failures occurred below the dashed line (lower left corner), the researchers concluded that maximum leakage in the loops less than about 0.8 kg/s and minimum leakage less than about 0.5 kg/s preclude clad failure.

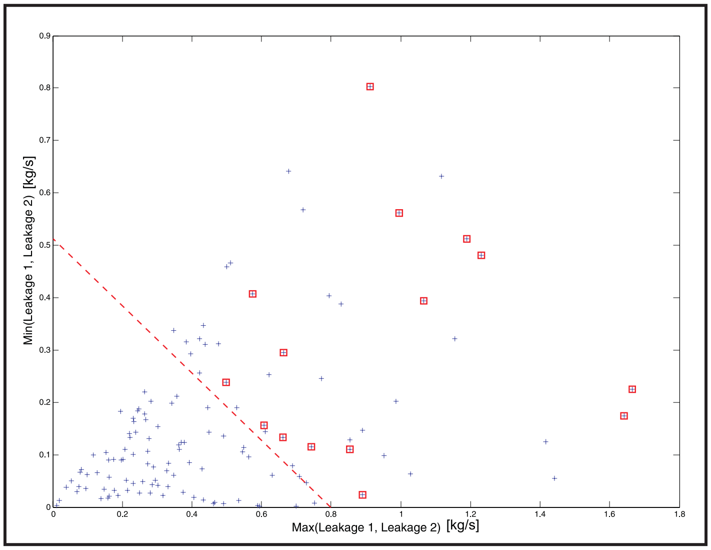


Figure 2. Leakage Threshold Effect (cladding failure).

Following is the explanation for this behavior. After the shaft stops, the system attempts to open the check valves by overcoming the leakage. The valve with the smallest leakage is the one that eventually opens. The higher the minimum leakage, the later the check valve opens. Such a delay means that more heat has been deposited in the core with no extraction and, therefore, the temperature has increased.

Once the valve opens, natural circulation develops through the corresponding DHR loop, but part of the fluid in the lower plenum is taken by the other DHR loop's leakage and transferred directly to the upper plenum, thus bypassing the core. The higher the maximum leakage, the smaller mass flow is available to the core for heat removal. Failure is the combined effect of the two quantities and, as Figure 2 shows, only values outside the lower triangle can lead to failure.

The calculated conditional failure probability (i.e., given the LOCA) was deemed too high, which led to the identification of several design changes to improve system reliability. A sensitivity analysis showed that check valve leakages were most important. An adiabatic boundary condition between the two coaxial DHR pipes was assumed meaning that, for the purpose of that study, they were actually separated. The adiabatic assumption was introduced for model simplification to reduce computational time, as nearly all heat transfer is through the heat exchangers and heat losses from the hot leg to the cold leg are small.

Some improvements to the system were suggested, such as relaxing the perfect insulation assumption between the two pipes by inserting insulation material on the inner side of the hot leg, combined with an increase in containment backup pressure in order to increase the mass flow rate in the DHR loop. This study is an example of the kinds of insights that can be obtained by including a reliability assessment in the design process. It is different from the usual use of PRA in design, which compares different system configurations, because it focuses on the thermal-hydraulic performance of a safety function.

The second study (Patalano et al.) investigated a number of design options aimed at reducing the conditional failure probability. The design chosen for detailed investigation placed insulation material on the outer surface of the hot pipe, covered by a 2.7 mm stainless steel foil. This foil prevents small particles from being detached and brought along in the loop by the gas causing damage to the structures or occlusions in the core channels.

The parameter and model uncertainties were propagated through the RELAP5-3D code, using a sample size of 128. As stated earlier, two failure limits were considered: one for the fuel and one for the DHR structure. The purpose of these calculations was to evaluate possible improvements with respect to the basic design and previous findings on the conditional failure probability of the passive system. Figure 3 shows the temperature profiles for 128 cases in the steel structure of both DHR loop hot pipes. The DHR system is composed of two 100 percent loops, only one of which operates while the other remains closed. Consequently, Figure 3 has 256 curves: the always-decreasing temperature profiles are those for the closed loop, in which the temperature decreases because the hot helium is passing through only the open loop. The curves with a local minimum point between 2,000 and 3,000 seconds are the temperature evolutions of the hot leg pipe in the open loop. There are 14 realizations that exceed the failure criterion of 1,123.15 K and would lead to structural failure of the DHR pipes, giving a total failure probability of 0.125.

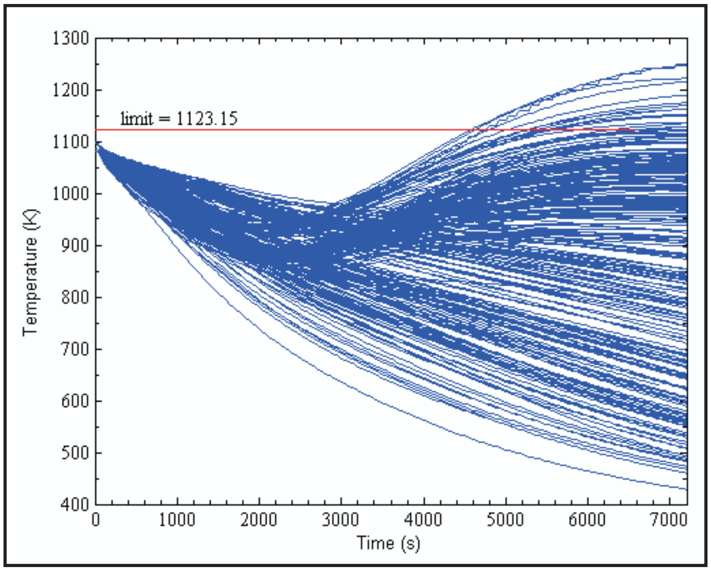


Figure 3. Temperature evolutions in the DHR loop hot pipes for 128 samples.

Extensive sensitivity analyses were conducted by Patalano et al. It was concluded that the parameter whose reduction in uncertainty would reduce the output uncertainty the most is the thermal conductivity of the insulation. The second most important parameter is the heat transfer coefficient in the containment. All the parameters have an impact on the mean maximum temperature. It appears that the roughness is an irrelevant parameter with respect to the output.

An important conclusion of this work was that a purely passive GFR system had a high conditional probability of failure that was difficult to reduce significantly. With design changes, researchers obtained a reduction by roughly a factor of five in the failure probability. However, the conditional probability was still deemed to be high. Therefore, a reliable active system appeared to be better suited for this particular application. Results led the MIT design team to adopt an active DHR as the main mode of decay heat removal for the GFR.

Planned Activities

In 2008, a final report will be prepared by MIT and CEA. It will contain the major findings produced by this project and recommendations as to how the unreliability of passive cooling safety systems can be evaluated.

INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE

Thermochemical Hydrogen Production Process Analysis

PI (U.S.): M. Lewis, Argonne National Laboratory

PI (France): P. Carles, P. Anziew, and J.M. Borgard, Commissariat à l'énergie atomique (CEA)

Collaborators: None

Project Number: 2004-011-F

Program Area: NHI

Project Start: October 2004

Project End: September 2007

Research Objectives

This I-NERI project had two objectives: 1) to develop a consistent methodology for evaluating the potential of a given thermochemical cycle to produce hydrogen with 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, the researchers would be able to compare all of the cycles on a consistent basis.

Research Progress

Researchers identified alternative thermochemical cycles and developed a consistent evaluation methodology based on the CEA concept of cycle efficiency changing as the level of knowledge increases.

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 in greater detail. The Level 1 screening only considers heat and work inputs and assumes stoichiometric reactions, resulting in the maximum theoretical efficiency. The Level 2 analysis requires the same heat and work inputs, but also includes equilibrium data to identify unrealistic process conditions, problematic separations, unexpected by-products, and similar such issues. Researchers adjusted 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 also be used to 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 two levels.

The researchers evaluated several promising alternative cycles, including copper-sulfate (Cu-SO$_4$), zinc-sulfate (Zn-SO$_4$), magnesium-chloride (Mg-Cl), and copper-chloride (Cu-Cl). Table 1 shows the calculated efficiencies. The data for the Cu-SO$_4$ cycle show a wide spread in calculated values, indicating a lack of optimization. More 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 indicated that required reaction temperatures are too

Cycle	Level	Efficiency % (LHV)	Maximum Temperature, °C	Other Conditions
Cu-SO$_4$	1	46.0	850	1 mol water
	2	38.1	1,100	1 mol water
	2	30.7	1,200	10 mols water
Cu-Cl	1	45.0	550	1 mol water
	2	43.9 From Aspen-Plus®	550	Excess water and HCl
Zn-SO$_4$	1	40.5	850	1 mol water
	2	40.8	1,400	2.7 mols water
Mg-Cl	1	35.2	600	1 mol water
	2	30.0–33.1	600	2 mols water

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

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 also investigated a fifth cycle, cerium-chloride (Ce-Cl).

Researchers conducted proof-of-principle work for the Cu-Cl, Cu-SO_4, 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 producing hydrogen through high-temperature processes such as thermochemical cycles and electrolysis. For thermochemical cycles, raw material investment, maintenance, heat exchanger costs, and energy recovery are the important parameters. For high-temperature electrolysis, the important parameters are the heat exchanger and investment cost for the electrolyzer, as well as the assumed electrolyzer lifetime and its ability to handle shutdowns and recycle events. Researchers quantified general screening criteria for potential thermochemical cycles as part of this effort.

Planned Activities

This I-NERI project is inactive except for periodic information exchange. No significant collaboration is taking place.

INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE

Sulfur-Iodine Integrated Laboratory-Scale Experiment

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

PI (France): Philippe Carles, Commissariat à l'énergie atomique (CEA)

Collaborator: General Atomics (GA)

Project Number: 2006-001-F

Program Area: NHI

Project Start Date: April 2006

Project End Date: April 2009

Research Objectives

This project focuses on evaluating the potential of the sulfur-iodine (S-I) thermochemical cycle for large-scale hydrogen production using high-temperature advanced reactors. S-I is one of the leading thermochemical cycle candidates due to potential high efficiency and a relatively advanced state of development. The ultimate objective of this project is to develop an integrated laboratory-scale experiment for the S-I process that can be used as the technical basis for future technology selection and scale-up decisions. Initial objectives include developing and testing the three major reaction sections of the S-I cycle as stand-alone units. The CEA has developed the primary (Bunsen) reaction section; General Atomics developed the hydrogen-iodide (HI) decomposition section; and Sandia National Laboratories developed and tested the sulfuric acid (H_2SO_4) decomposition section. Stand-alone tests on these three sections established approaches for process chemistry, materials of construction, controls, and diagnostics. The integrated laboratory-scale (ILS) experiment has now been assembled, and after completion of initial testing, the researchers will conduct fully integrated experiments to demonstrate closed-loop operation of the S-I cycle. These experiments will provide information on overall system controls, the effects of cross-talk between sections, and potential corrosion product contamination effects on catalysts and process chemistry.

Research Progress

The objective of this project is to develop an ILS experiment that will provide the technical basis for evaluating the S-I process for large-scale hydrogen production using high-temperature nuclear reactors. The key issues being addressed include high-temperature materials in highly corrosive environments, process chemistry and thermophysical data uncertainties, innovative heat exchanger designs and materials to couple to the nuclear heat source, and evaluation of the potential performance and costs of these cycles for large-scale hydrogen production. The status of the integration of the three major reaction sections is summarized below.

Sulfuric acid decomposition section. Researchers completed sulfuric acid decomposition experiments at temperatures up to 875°C using the silicon-carbide (SiC) bayonet heat exchanger concept developed at SNL (see Figure 1). This approach integrates the vaporizer, superheater, catalytic decomposer, and recuperator into a single ceramic unit that eliminates all high-temperature connections, allowing the use of glass or Teflon-lined steel in lower temperature areas.

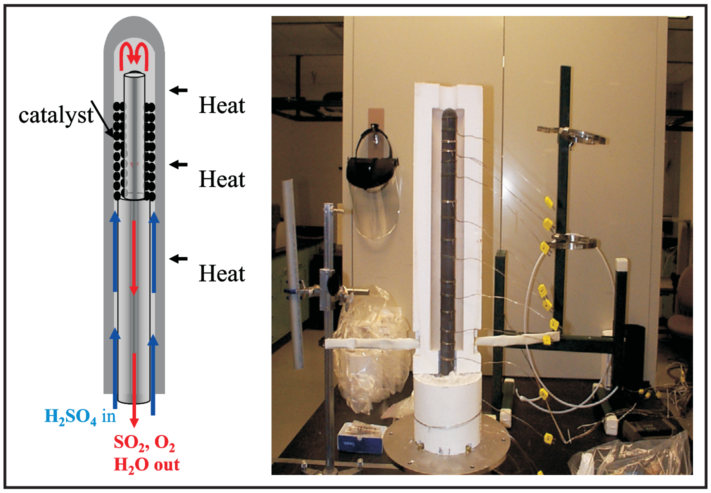


Figure 1. SiC bayonet acid decomposer.

The SiC decomposer has been tested to confirm operation in the range of 200–300 l/hr of SO_2 production, as required for the ILS experiment. Tests to date have confirmed that the SiC bayonet design minimizes corrosion issues, simplifies the operation of the acid decomposer, and provides a more compact and robust acid decomposer system for the S-I cycle. The unit is in operation as an integrated unit at the GA experiment site.

HI decomposition section. The HI decomposition section is based on an extractive distillation approach that separates iodine (I_2) from the HIx feed stream before decomposition. Experiments during the past year have quantified the liquid-liquid extraction step where phosphoric acid (H_3PO_4) strips the HI and water from the HIx feed solution and established the desired operating conditions for effective distillation and extraction (see Figure 2).

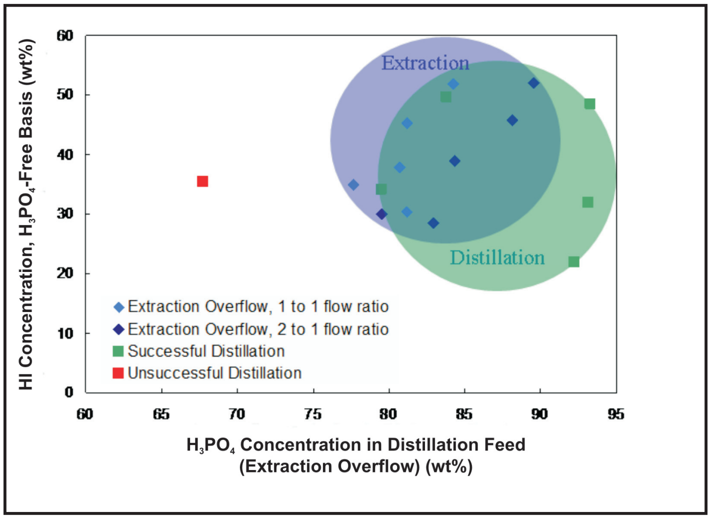


Figure 2. H_3PO_4 operating space for the extraction and distillation for the HI decomposition section.

Bunsen section. The team has completed reassembly of the Bunsen section at the GA experiment site, as shown in Figure 3. Chemical testing is underway to establish operation data for the sulfur dioxide–oxygen (SO_2–O_2) separation section and the primary Bunsen reactor.

Figure 3. The Bunsen section for the S-I ILS experiment (CEA).

Planned Activities

Planned activities for FY 2008 are to complete testing of the individual sections, and initiate integrated closed-loop, ILS experiments. Several types of experiments will be conducted including control verification tests, transient response propagation tests, and evaluation of chemical cross-contamination effects. The first tests will demonstrate closed-loop operations including startup and shutdown and continuous steady state operations. The results of this series will establish the stable parameter space for the entire cycle. Subsequent testing will also address transient response and control strategies for off-normal conditions that may propagate through the cycle. Chemical cross-talk between sections and the effects of corrosion or contamination on cycle performance are also of concern. In particular, the sulfuric acid and HI decomposition sections should be able to continue operating with iodine and sulfuric acid contamination. The data from these tests will be compared to model predictions and improved models will be developed to provide projections of cycle performance for large-scale systems.

INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE

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

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

PI (France): P. Billot, Commissariat à l'énergie atomique (CEA)

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

Project Number: 2006-002-F

Project Start Date: January 2006

Project End Date: December 2008

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.

INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE

Characterization Methods for Anisotropy and Microstructure of TRISO Particle Ceramic Layers

PI (U.S.): J. Hunn, Oak Ridge National Laboratory (ORNL)

PI (France): O. Dugne, Commissariat à l'énergie atomique (CEA)

Collaborators: Université Bordeaux, AREVA, Laboratoire des Composites Thermostructuraux (LCTS)

Project Number: 2006-003-F

Program Area: Generation IV

Start Date: June 2007

End Date: May 2009

Research Objectives

The resurgence of coated particle fuel as a cornerstone technology for the next generation of nuclear reactors requires characterization methodology for process development and product qualification. This I-NERI project compares two systems for measuring the anisotropy of the pyrocarbon layers in tri-isotropic (TRISO) coated particle fuel, which is an important parameter related to fuel performance: 1) the newly developed two-modulator generalized ellipsometry microscope (2-MGEM) and 2) the more traditional optical polarimeter (RAPAX) technique.

Evaluation of Pyrocarbon Anisotropy. The structural stability of pyrolytic carbon under irradiation is sensitive to the degree of preferred orientation of the carbon basal planes. For this reason, limits on the level of pyrocarbon anisotropy are included in the criteria for coated particle fuel acceptance. However, full understanding of the relationship between fuel performance and pyrocarbon anisotropy has not been achieved due to limitations in the available technology. Current advances in material science analytical techniques have opened the door to further study.

The researchers have each developed a methodology for determining pyrocarbon anisotropy on a micrometer-scale by reflecting polarized light from the surface of a polished cross-section. CEA's RAPAX technique is based on the use of an optical polarimeter, which has historically been used to determine pyrocarbon anisotropy in coated particle fuel. The ORNL 2-MGEM approach goes beyond a simple optical polarimeter by using generalized ellipsometry to fully determine the effect of reflecting polarized light from the pyrocarbon surface. There is a need to connect the results of the ellipsometer to historically reported values for anisotropy obtained using various polarimeter systems. Careful comparison of the two techniques using identical samples and calibration standards will be of great value in making this connection and in validating the two techniques.

In addition to the optical measurements discussed above, the CEA researchers have applied a selected area electron diffraction technique (SEAD) using a transmission electron microscope (TEM). TEM-SEAD can evaluate pyrocarbon anisotropy on a nanometer-scale to obtain the microstructural resolution necessary for better understanding of the phenomenon. This will help to strengthen the connection between the measured optical anisotropy and the underlying crystallographic anisotropy in the pyrocarbon.

Examination using X-ray imaging techniques. X-ray radiography is a non-destructive technique which can obtain structural information without introducing artifacts induced by metallographic preparation. Moreover, this technique can offer 3-D information about layer thickness, density, interface structure, and coating defects (cracks, voids, etc.).

ORNL and the CEA each utilize x-ray radiography for coated particle characterization. In addition to standard x-ray systems, ORNL has developed a new system capable of x-ray radiography and 3 dimensional tomography with a resolution down to 1 µm. To shake down this new system and learn more about the relative capabilities of the various techniques, a number of particles from each reference fuel lot will be examined.

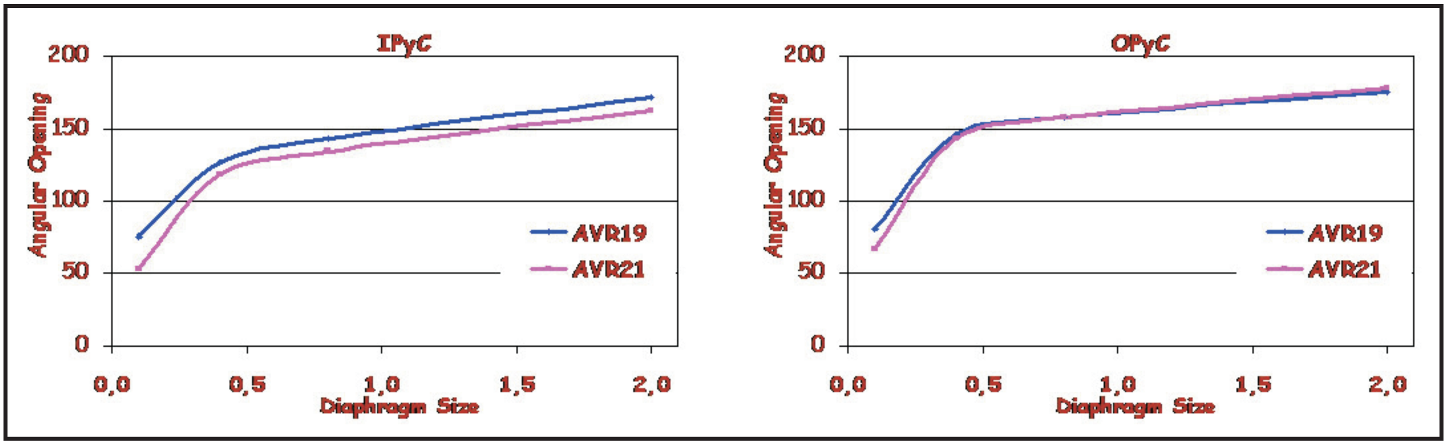


Figure 1. SEAD determination of pyrocarbon anisotropy on AVR19 (CEA German reference fuel) and AVR21 (ORNL German reference fuel).

Research Progress

Work on pyrocarbon anisotropy commenced in June 2007 and is nearing completion. Work on X-ray tomography is scheduled to commence in Summer of 2008. Tables 1 and 2 list the samples included in the anisotropy study along with a brief reason for their inclusion. The samples provide a range of anisotropy that covers the typical range observed in these materials. Comparison of the 2-MGEM and RAPAX techniques over this extended range will well establish the correlation between the two methods.

Inclusion of the calibration standards listed at the bottom of Table 1 was key to the success of this project. The aluminum sample allowed for the identification and quantification of the polarization effects in the optics of the two measurement instruments. It is vital that these effects be accounted for in order to obtain the most accurate results. After making calibration corrections based on the aluminum reference sample, good agreement was obtained between the RAPAX and 2-MGEM measurements on the other two standard materials.

Early in this project, samples were prepared at ORNL and shipped to the CEA for TEM analysis and SEAD measurement of anisotropy at the nano-meter scale. SEAD provides a semi-quantitative analysis of pyrocarbon anisotropy using electron diffraction to determine the width of the 002 diffraction spots in terms of an angular opening value. Lower values of angular opening correspond to higher anisotropy. This angular opening is determined for varying aperture size to provide information on the localization of the anisotropy. Figure 1 shows the SEAD plots for the two German reference materials. It can be seen that the IPyC layers have a slightly higher anisotropy than the OPyC layers in these two samples, as expected. TEM and SEM study of the microstructure of the various pyrocarbon samples was also performed in the hopes of identifying correlations between the observed microstructure and the variations in the optical anisotropy.

Planned Activities

ORNL and the CEA have each completed preparation and measurement of the optical anisotropy on polished cross-sections of the samples in Tables 1 and 2. These polished particles have been exchanged and are being analyzed by the other organization. Comparison of the results is scheduled to occur in the next several months.

A meeting at the CEA in Cadarche is scheduled for June 2008. At this meeting final results and analysis of the anisotropy study will be discussed and the final report for this portion of the task will be drafted. Planning for the next element involving X-ray tomography will also occur at this meeting. In addition, opportunities for further work in the area of pyrocarbon anisotropy will be considered, as a follow up to this valuable collaboration.

Sample description	IPyC diattenuation	OPyC diattenuation	Reason for inclusion in study
German reference fuel EUO 2358-2365 composite (AGR-06-05-03)	0.0141	0.0076	Primary sample for this study. This fuel demonstrated acceptable performance and is considered to be a starting point for modern coated particle fuel programs.
AGR-1 baseline fuel – pre compacting (LEU01-46T-B01)	0.0074	0.0061	This is the baseline US fuel design and is currently being irradiated. It will provide the first available irradiation performance data on modern fuel and will be available for post-irradiation examination.
AGR-1 baseline fuel – post compacting (LEU01-46T-Z06)	0.0091	0.0107	The pre-compacting AGR-1 baseline fuel can be compared more directly with the pre-compacting German reference fuels and is the fuel fabrication stage at which QC measurements are made. However, heat treatment during compacting increases anisotropy of the pyrocarbon layers. Measurements after compacting provide the true pre-irradiation data.
Heat treated ORNL German reference fuel (AGR-06-05-03-01)	0.0188	0.0122	Heat treatment at 1,800°C for 1 hour simulates the effect of compact fabrication on the coated particles. Heat treatment is expected to result in an increase in pyrocarbon diattenuation and will provide data more relevant to the actually pre-irradiated fuel condition.
AGR-1 variant 1 fuel – pre-compacting (LEU01-47T-B01)	0.0054	0.0056	A high temperature pyrocarbon variant also currently being irradiated. It possesses the lowest available pyrocarbon diattenuation in a relevant coated particle sample.
HRB-21 reference fuel (AGR-10-H06)	0.0192	0.0126	Reference fuel produced for the HRB-21 irradiation test (1988-1995). It possessed high pyrocarbon diattenuation and did not perform well under irradiation.
Heat treated HRB-21 reference fuel (AGR-10-H07)	0.0189	0.0187	Heat treatment at 1,800°C for 1 hour will simulate the effect of compact fabrication on the coated particles. Heat treatment is expected to result in an increase in pyrocarbon diattenuation.
Aluminum mirror	0		Normalization standard. Allows for the first order cancellation of polarization effects in the system optics.
Rutile (single crystal TiO2) polished with c-axis in plane	0.087		Calibration standard. Provides a definitive check on the accuracy of the measurement system.
HOPG (Highly oriented pyrolytic graphite) polished with c-axis in plane	0.572 at 546 nm		This is the closest thing available to single crystal graphite.

Table 1. ORNL samples for I-NERI collaboration.

Sample description	IPyC RAPAX	OPyC RAPAX	Reason for inclusion in study
CEA German reference fuel AVR-19	1.11	1.07	Primary sample for this study. Demonstrated acceptable performance and is a starting point for modern coated particle fuel programs.
PyC on plane substrate Sample ID: PyC-CEA-10	1.25	-	PyC deposited on plane substrate with a rate of 1μm/min–the highest pyrocarbon anisotropy.
PyC on plane substrate Sample ID: PyC-CEA-9	1.11	-	PyC deposited on plane substrate with a rate of 2μm/min–intermediate pyrocarbon anisotropy.
PyC on plane substrate Sample ID: PyC-CEA-6	1.11	-	This is PyC deposited on plane substrate with a rate of 5μm/min–intermediate pyrocarbon anisotropy.
PyC on plane substrate Sample ID: PyC-CEA-5	1.09	-	This is PyC deposited on plane substrate with a rate of 11μm/min–lowest pyrocarbon anisotropy of this set of PyC on plane substrate.
CEA baseline Sample ID: UA19	?	?	This sample is the baseline CEA fuel design similar to AGR2. It will provide the first available irradiation performance data on modern fuel. To be available for post-irradiation examination.

Table 2. CEA samples for I-NERI collaboration.

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, Director of DOE-NE, and Mr. Kusaka, Director-General of ANRE. An annex regarding 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
- Innovative fuel technologies using solvent extraction

9.2 Project Summaries

The first project awarded under the cooperative agreement with ANRE in FY 2005 was completed during the past year. Work continues on the second project in collaboration with MEXT, 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 2007.

Directory of Project Summaries

INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE

Development of Materials for Supercritical-Water-Cooled Reactors

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

PI (Japan): Hideki Matsui, Tohoku University

Collaborators: Idaho National Laboratory, University of Wisconsin–Madison, University of Tokyo, Toshiba Corporation, Hitachi, Ltd.

Project Number: 2005-001-J

Program Area: Generation IV

Project Start Date: October 2004

Project End Date: September 2007

Research Objectives

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

Material development is a critical issue for the SCWR. 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 representative conditions, along with a comprehensive materials database.

In this work, researchers subjected selected commercial alloys and several newly developed alloys to micro-analysis and corrosion tests in supercritical water. They evaluated long-term reliability via phase stability tests and long-duration corrosion tests. Through these experimental studies, they gained an understanding of the materials' behavior under SCWR conditions. This knowledge enabled 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 completed a series of stress corrosion tests on 21 irradiated samples of alloy JPCA in deaerated supercritical water (SCW) at 400°C. 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. Samples were tested in argon (Ar) and SCW at 400°C and 23.4 MPa; at 385°C and 27.6 MPa; and at 500°C and 24.0 MPa.

Samples tested in argon failed at about 3 percent plastic strain and exhibited approximately 40 percent reduction in area. No intergranular cracking was observed on the fracture surfaces. No cracks were observed on the gage surfaces far from the fracture surface, although some were visible near the fracture surface where deformation was high. A typical stress-strain curve is shown in Figure 1a and the resulting fracture surface is shown in Figure 1b.

A total of eight samples were strained in 400°C deaerated SCW. All samples exhibited a lower strain-to-failure than those in argon, with some failing before the yield point, as shown in Figure 1a. Three samples failed in the pinhole in a mode that will be described in a subsequent section. The samples exhibited between 7 and 45 percent intergranular (IG) cracking on the fracture surface, as shown in Figure 1c. IG cracks were observed on the gage surface, mainly in the vicinity of the fracture surface, but some were found away from the fracture surface. No correlation between the percent IG and cracking on the gage surface was observed. However, there were significantly more cracks on the sample tested in SCW than on the samples tested in argon.

Researchers tested four samples in pure, deaerated SCW at 385°C, 27.6 MPa and 400˚C, 23.4 MPa to determine the relative roles of water density and temperature in stress corrosion cracking 1) solution annealed (SA) at 407°C, 41.1dpa; 2) SA at 427°C, 43.9dpa; 3) cold worked (CW) at 407°C, 41.1dpa; and 4) CW at 427°C, 43.9dpa. All sample conditions exhibited a higher percentage of IG cracking on the fracture surface at 400°C, as shown in Figure 2. While cracking should depend on both solvent density and

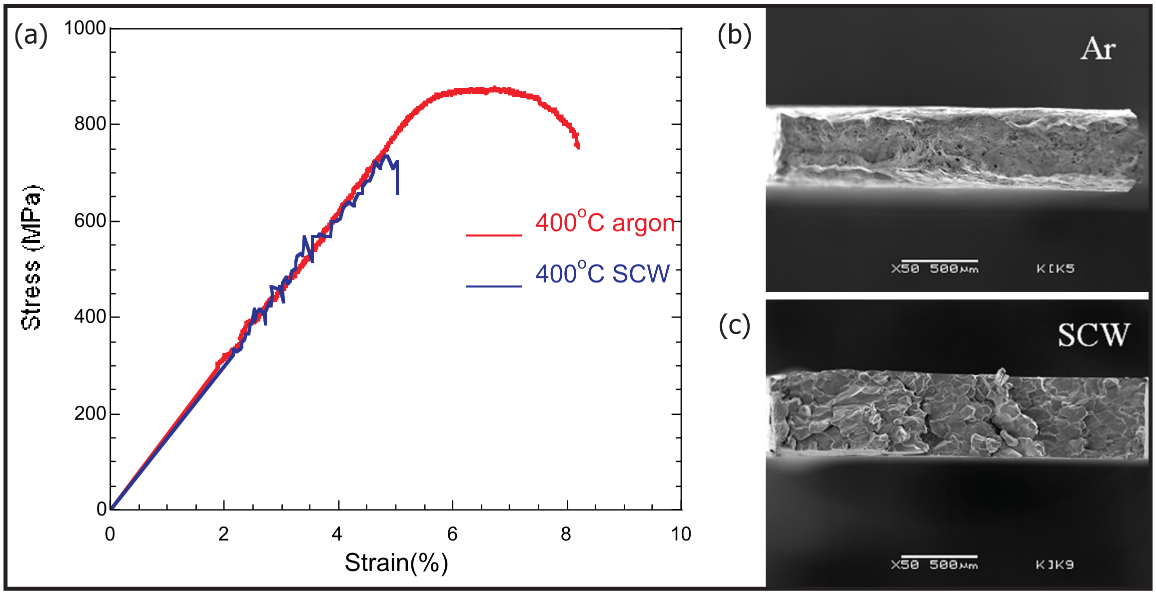


Figure 1. (a) stress-strain curves for irradiated JPCA tested at 400°C in argon and SCW and resulting fracture surfaces from (b) testing in argon and (c) testing in SCW.

temperature, it should decrease with density and increase with temperature. The results show that the effect of temperature strongly overshadows the effect of water density on cracking of the irradiated alloys.

The influence of hardening on cracking was assessed by comparing the cracking susceptibility of SA:427°C:43.9dpa and CW:427°C:43.9dpa strained in three different types of environments (400°C deaerated SCW, 385°C deaerated SCW, and 400°C hydrogenated SCW) and by comparing SA:427°C:43.9 dpa and SA:417°C:41.1dpa in the same environment (400°C deaerated SCW). In the first set of tests, the difference in hardening is due to the cold work performed before the irradiation; in the second, the difference in hardening is due to the different irradiation condition. In all cases, the harder samples exhibited higher stress corrosion cracking (SCC) susceptibility (see Figure 3.) Results are consistent with the data collected for LWRs, where hardening due to cold work is known to cause an increase of crack growth rate in austenitic stainless steel strained in 288°C boiling water reactor (BWR) normal water. Increased yield strength due to irradiation also correlates with an increase of susceptibility to irradiation-assisted stress corrosion cracking (IASCC).

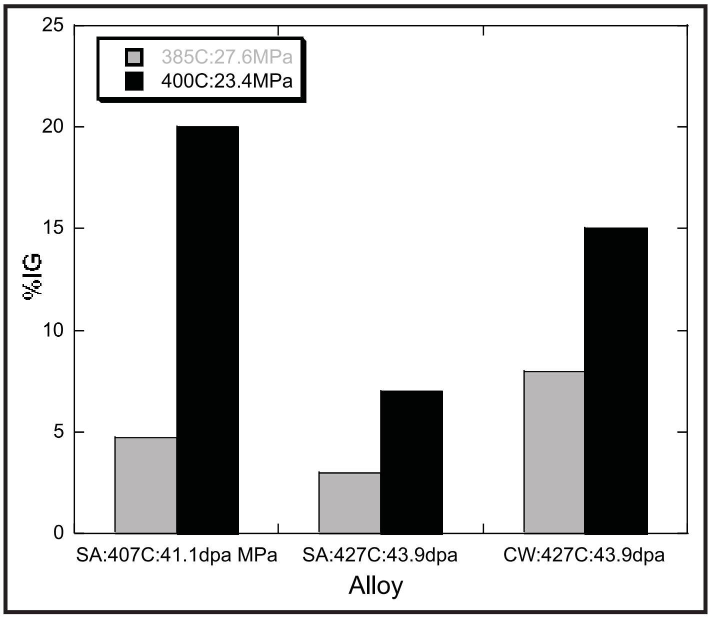


Figure 2. Effect of water density and temperature on cracking in SCW at 385°C:27.6 MPa and 400°C:23.4 MPa.

Corrosion testing was conducted on candidate alloys, including F/M steels, austenitic steels, and nickel-based alloys in both subcritical and supercritical water at temperatures of 360°C, 500°C, and 600°C for times up to 1,026 hours. The effect of oxygen concentration on the corrosion behavior was also investigated. Researchers also applied surface modification and grain boundary engineering (GBE) treatment on selected materials and investigated the corrosion response. In addition to testing bulk samples, evaluation of surface modified steels using techniques including shot peening and thermal spray coating are being conducted.

For materials tested to roughly 1,000 hours, weight gain due to oxidation in SCW is typically smaller but less predictable in austenitic stainless steels as compared to ferritic-martensitic steels. Nickel (Ni)-based alloys show good corrosion resistance in SCW environment but pitting occurs in most tested materials. Oxygen concentration is

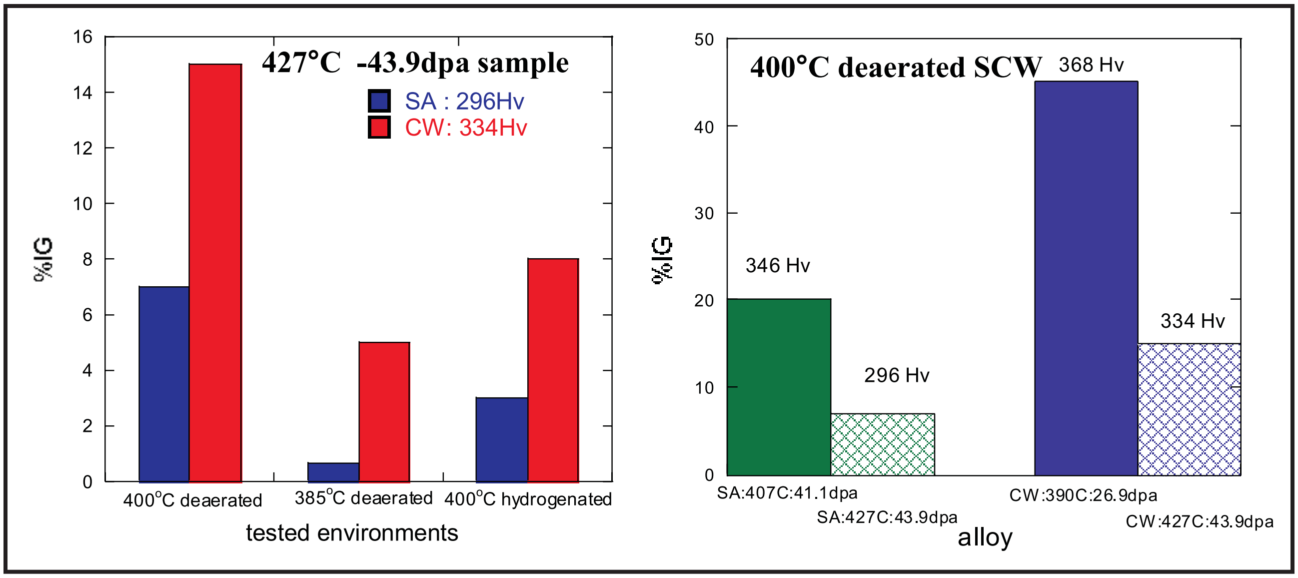


Figure 3. Influence of hardening on cracking. (a) Effect of cold-work on sample irradiated at 427°C to 43.9 dpa and tested in different environments, and (b) effect of irradiation.

the one of the most important factors that influences or changes the corrosion behavior of materials in the SCW environment. Figures 4 (a) and (b) present the cross sectional morphologies of austenitic alloy D9 after exposure to 500°C SCW with inlet oxygen concentration of 25 ppb and 2,000 ppb dissolved oxygen contents, respectively. By comparing the cross sectional scanning electron microscope (SEM) images, we observe that increasing the oxygen content to 2,000 ppb alters the microstructure and corrosion mechanism.

For low-oxygen concentration exposure, the formed oxide scale on D9 is composed of an outer magnetite layer and an inner $(Fe, Cr, Ni)_3O_4$ spinel layer. Depending on the local microstructure and compositions of the bulk grains, the oxygen penetration depth in different areas varied, resulting in a nodular structure of the oxide scale and an uneven metallic/substrate interface. The region between two nodules was typically a Ni-enriched region. For high oxygen concentration exposure, however, the outward growth of the oxide scale and inward oxygen diffusion rate are homogenous and the scale thickness is uniform.

Figures 5 and 6 present cross sectional EDS and TEM examination results for D9 exposed to higher oxygen concentration. This data, along with the associated x-ray diffraction (XRD) and electron backscatter diffraction (EBSD) results (not shown), indicate that the oxide scale of D9 is a four-layer structure containing an outer hematite layer; an Fe, Cr, Ni oxide spinel layer; an approximately 400 nm amorphous layer with oxygen depletion in this layer; and an innermost layer with increased oxygen. The oxide scale developed on D9 in the high oxygen level environment was not stable and was prone to spallation.

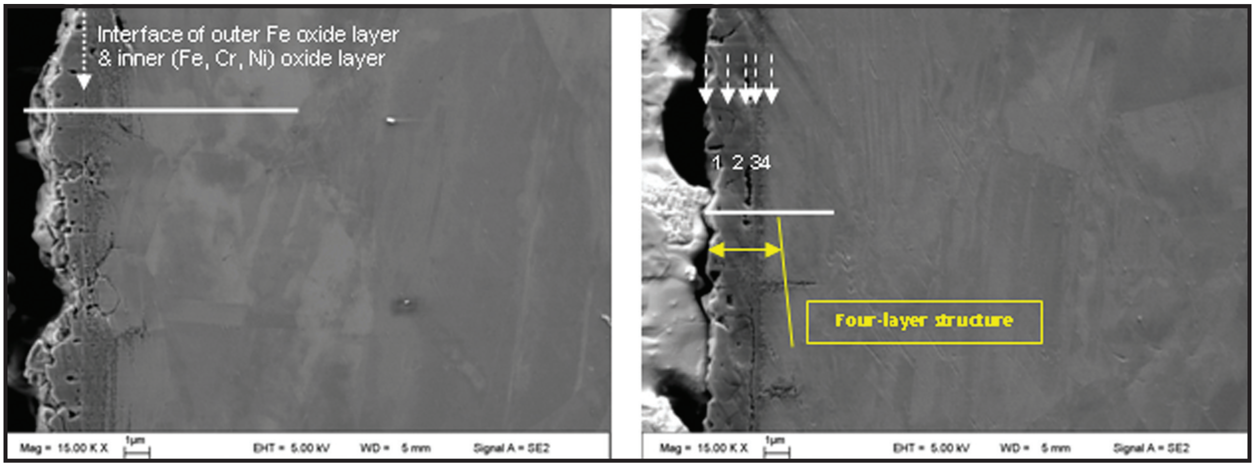


Figure 4. Comparison of cross-sectional morphologies of austenitic alloy D9 after exposure to 500°C supercritical water with (a) 25 ppb dissolved oxygen content and (b) 2,000 ppb dissolved oxygen content for approximately 505 hours.

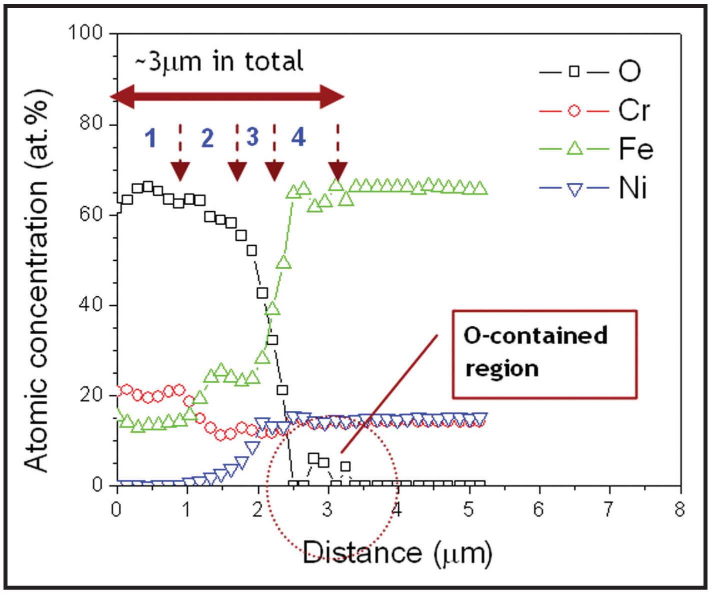


Figure 5. Composition profile across the oxide thickness for D9 after exposure to SCW with 2,000 ppb oxygen content.

As shown in Figure 3, the weak interface appears to be the hematite/amorphous interface where long cracking and delamination were found.

Loop temperature significantly influences the oxidation behavior of the tested alloys. Figure 7 shows the cross sectional morphology of D9 after exposure to an inlet oxygen concentration of 25 ppb at 600°C for 1,000 h. A non-uniform scale (with 10-40 µm in thickness) was formed. The oxide scale bonded well at the interface of the outer/inner oxide layers. Micron-sized cavities developed within the magnetite layer.

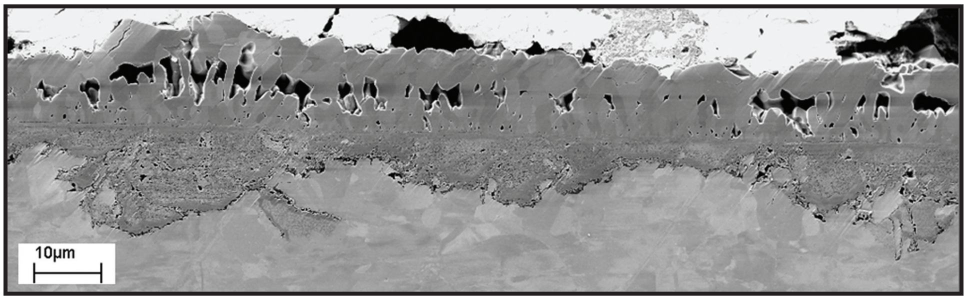


Figure 7. Cross-sectional SEM image of D9 alloy after exposure to 25 ppb oxygen concentration supercritical water at 600°C for 1,000 hours.

Thermomechanical processing (TMP) was employed on INCONEL alloy 617 for GBE purposes. The idea is to control the oxide development by changing the diffusion pathways in the bulk metal, purposely creating what are known as low-Σ coincidence site lattice boundaries, or CSLBs. As-received samples were cold rolled with an approximately 5 percent thickness reduction followed by annealing at 1,100°C for a variety of times. Two GBE-treated samples (labeled GBE1 and GBE2) were investigated. Both possess a similar low fraction of random boundaries (approximately 27 percent). The major difference is that sample GBE2 has a 13 percent smaller fraction of Σ1 boundaries (or a higher fraction of low-Σ CSLBs) compared to sample GBE1.

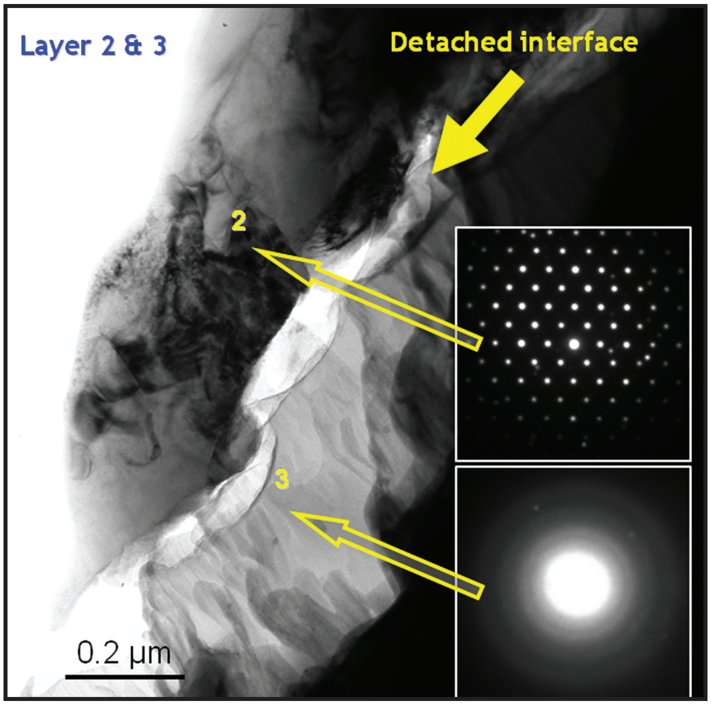


Figure 6. TEM/SAD examinations on the oxide layers 2 and 3 as indicated in Figure 1(b) in D9 after exposure to SCW with 2,000 ppb oxygen.

The effect of GBE on oxidation behavior of Alloy 617 was studied by exposing the as-received and GBE-treated (GBE1 and GBE2) samples to SCW at 500°C and 25 MPa. The fractions of $\Sigma 3^n$ (Σ3, Σ9 and Σ27) boundaries were greatly enhanced for GBE-treated samples GBE1 and GBE2 (Σ3 boundaries increased to 36 and 49 percent, respectively, compared to 11 percent for the as-received). The fractions of Σ9 and Σ27 boundaries of GBE1 and GBE2 were about 5–9 and 7–10 times of the as-received sample, respectively. The increased fractions of $\Sigma 3^n$ boundaries indicate that significant twinning and multiple twinning events occurred during annealing after cold rolling to 5 percent. This is desirable for property improvement of materials.

Material evaluation tests were carried out using un-irradiated specimens to understand basic properties of test materials. For mechanical properties, high-temperature tensile tests, uni-axial creep tests, and pressurized tube creep tests were conducted. For phase stability, microstructure analysis was conducted after heating to 700°C. For helium (He) embrittlement, He yielding calculations and ion injection tests were conducted. Figure 8 shows the temperature dependence of 0.2 percent proof stress of the test materials. Grain refined 310S (F-310S) exhibited extremely good performance at room temperature, although the advantage decreases with increasing temperature and turned negative at 800°C.

The basic properties of mechanical strength, phase stability, and He embrittlement will be utilized to evaluate the effect of neutron irradiation by comparing these data with those of irradiated materials.

The complex effect of irradiation and corrosion was evaluated in terms of general corrosion in SCW with the specimens irradiated in JOYO at 500°C and 600°C in He-bond capsules. The neutron irradiated ODS (oxide dispersed steel) was included as test material in addition to the materials prepared in this project, since corrosion behavior is the greatest concern for ODS use in SCWR.

In parallel with neutron irradiation tests, general corrosion tests were carried out with non-irradiated specimens. In contrast to the oxygenated condition in the previous project, which was tested in 8 ppm dissolved oxygen (DO), researchers focused on the deaerated condition with DO lower than 10 ppb. Type 316L and Type 316L+Zr exhibited a linear increase of weight gain until 2,000 hours, while Ni-based alloys and Type 310S series showed small weight change and no increase of weight after 500 hours. The net corrosion rate was finally evaluated after the oxide film removal.

The SCC susceptibility evaluated with SSRT tests focused on the performance of modified alloys and the effects of aqueous impurities and dissolved oxygen. Micro-cracks on the side surface of specimens tested under the SCW condition, which was found in the previous project, is another concern in this study. To consider the mechanism of their occurrence, SSRT tests were performed in vacuum at the same temperature as SCW. Figure 9 shows the SEM micrograph of the side surface and the optical micrograph of the cross-section after SSRT at 550°C in vacuum. Micro-cracking occurred not only on the surface but also on the inside of the specimen. This result suggests that the micro-cracks were generated primarily due to creep deformation. Figure 10 shows the comparison of the maximum stress in tensile tests in air and SSRT tests in SCW and vacuum at 550°C for Type 304 SS and Type 316L. This figure indicates that the decrease in maximum stress in SCW is comparable to that in vacuum.

Planned Activities

This I-NERI project has concluded.

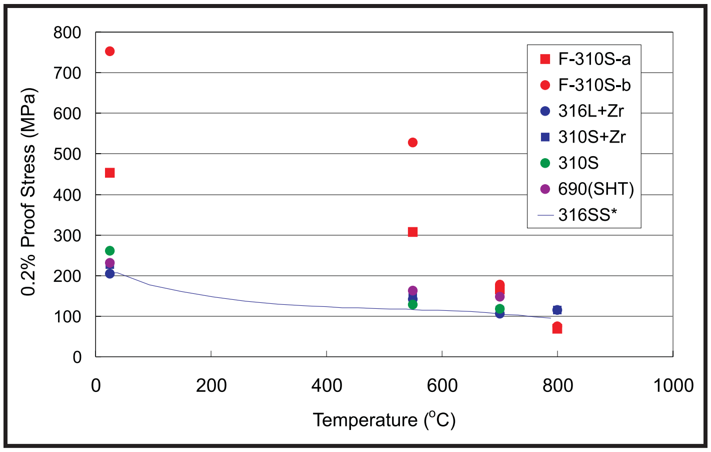


Figure 8. Temperature dependence of 0.2 percent proof strength of test materials.

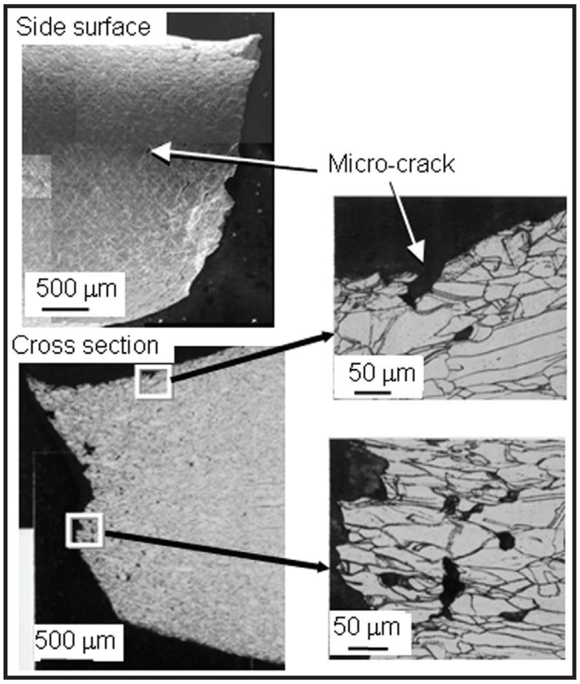


Figure 9. SEM and optical micrograph after SSRT tests at 550°C in vacuum.

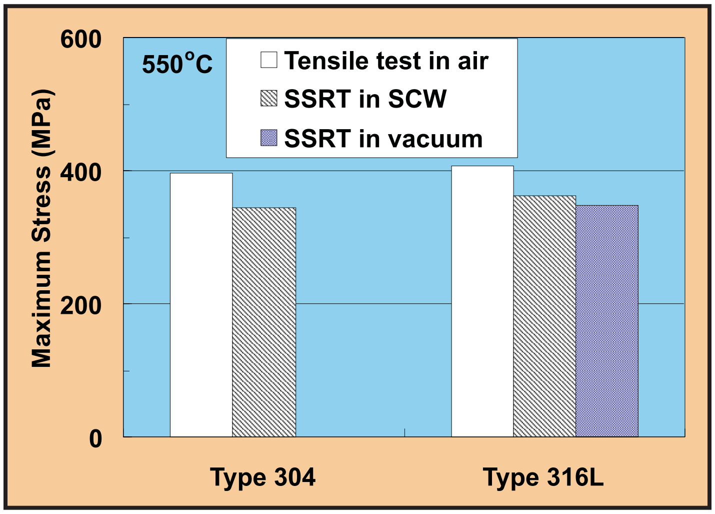


Figure 10. Comparison of maximum stress between tensile tests and SSRT tests at 550°C.

INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE

Assessment of Irradiation Performance of ZrC TRISO Fuel Particles

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

PI (Japan): K. Sawa, Japan Atomic Energy Agency (JAEA)

Collaborators: Idaho National Laboratory (INL)

Project Number: 2006-001-J

Program Area: Generation IV

Project Start Date: March 2006

Project End Date: February 2009

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:

- Conduct post-irradiation examinations (PIE) of zone-refined, high-purity ZrC samples to understand baseline irradiation effects
- Characterize mechanical properties of unirradiated developmental ZrC coatings
- Perform high-temperature neutron irradiation of ZrC-coated surrogate fuel particles and conduct a PIE
- Study the thermo-mechanical response of ZrC-coated particles using the PARFUME model
- Examine chemical interactions between ZrC and key fission products to identify potential degradation
- Irradiate fueled UO_2-ZrC TRISO-coated particles

Research Progress

Optimization of ZrC coating process. The researchers performed coating tests on surrogate kernels to investigate the influence of coating parameters on stoichiometry and density of the ZrC. In the test runs, the coating temperature was varied parametrically in the range of 1,300 to 1,550°C. The composition ratio of $CH_4/ZrBr_4$ and the batch size were chosen to be 1.4 and 50g, respectively. As a result, a systematic dependence of the properties of ZrC on the coating temperature was found. As coating temperature decreased from 1,550 to 1,350°C, the atomic C/Zr ratio decreased from 1.4 to 1.0; deposition rate similarly decreased with the decreasing temperature. Coating tests with the particle batch size enlarged from 50 to 100g were also performed. Neither density nor stoichiometry of ZrC were dependent on the particle batch size for the coating temperature range and the source gas composition used in this test. From these results, the optimum coating temperature to obtain the stoichiometric ZrC was determined to be approximately 1,350°C.

Based on these results, researchers successfully fabricated the ZrC-coated surrogate particles for irradiation testing in the High Flux Isotope Reactor (HFIR) at Oak Ridge National Laboratory. The appearance and the cross-sectional images of the particles are shown in Figure 1.

Characterization techniques. Measurement techniques for the density and stoichiometry of the ZrC-coating layer were developed. Before the measurement, researchers removed the ZrC-coating layer from the IPyC layer by means of mechanical grinding with quartz and chemically separated the material using tetrabromoethane. For the determination of mass density of the ZrC-coating layer, gas-pycnometry using helium as the substitution gas was determined adequate for the volume measurement. Researchers found that the device works very well with sample weights 100mg or greater. For the stoichiometry of the ZrC-coating layer, which can be described as the atomic

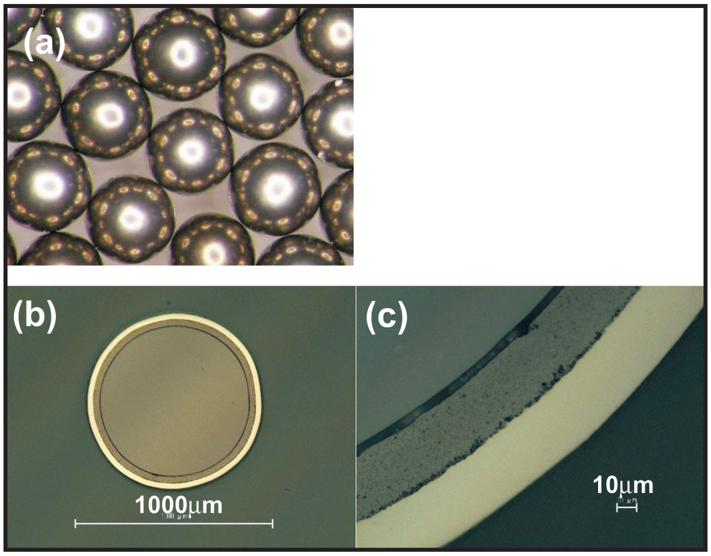


Figure 1. (a) Appearance and (b,c) cross sections of ZrC-coated particles (ZrC-06-2048).

Figure 2. ZrC-coated particles with ZrO_2 kernel after heat treatment (Lot ZrC-06-2048HT).

ratio of C and Zr, researchers developed a technique that combines applications of inductively coupled plasma (ICP) mass spectrometry and oxidation methods.

Non-irradiated characterization of the developmental coatings. Fabrication of surrogate samples for neutron irradiation has been completed in this reporting period and the samples listed in Table 1 are being considered for study. Researchers prepared three sets of ZrC-coated particles with a ZrO_2 kernel to investigate the effect of stoichiometry on irradiation behavior: C/Zr ratios were chosen to be 1.0, 1.1, and 1.3 in the ZrC layer. The as-fabricated ZrC samples were heat-treated at 1,800°C for 1 hour in order to simulate conditions during the actual fuel compact process.

By observing the outer surfaces of ZrC-coated particles after the heat treatment, neither cracks nor other specific problems were found, as shown in Figure 2. X-ray diffraction of the ZrC layer after the heat treatment indicated that heat-treated ZrC also showed a single phase, unchanged from the as-fabricated state.

Neutron irradiation. Researchers tentatively agreed to irradiate the coated surrogate particles at temperatures of 900 – 1,200°C and fast neutron fluence between 2 – 6 x 10^{25} $n\text{-}m^{-2}$ using the target holder rabbit facility at HFIR. Irradiation capsules employing the standard refractory metal sleeves for ceramic bend bar specimens and graphite specimen holders with cylindrical slots were designed specifically for irradiation of particle specimens.

Type of Particle	Particle Diameter (μm)	Material	Size or Thickness (μm)	Density (g/cm3)	C/Zr Ratio of ZrC Layer	Lot ID
ZrO_2+PyC+ZrC (ZrO_2 kernel with 4.82% Y_2O_3)	843	ZrO_2	720 (diameter)	6.07	1.11	ZrC-06-2022HT
		PyC	35	1.9		
		ZrC	22	6.45		
	841	ZrO_2	720 (diameter)	6.07	1.03	ZrC-06-2048HT
		PyC	35	1.9		
		ZrC	28	6.50		
	858	ZrO_2	720 (diameter)	6.07	1.35	ZrC-06-2003HT
		PyC	35	1.9		
		ZrC	21	6.01		

Table 1. Irradiation samples.

ZrC coated fuel particle modeling. JAEA has developed a pressure vessel failure fraction calculation code for SiC-coated TRISO-coated fuel particles based on the rigid SiC model. However, plastic deformation of the 3rd layer (ZrC) has to be considered for ZrC-TRISO coated fuel particles. Researchers modified the existing code to incorporate a pressure vessel failure fraction calculation which can treat the plastic deformation of the 3rd layer. A finite element method (FEM) code is employed in the modified model. It was confirmed that the modified model can treat the elastic deformation of each layer and plastic deformation of the 3rd layer. A benchmark calculation is in progress.

Planned Activities

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

- 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
- Continue post-irradiation examination of the zone-refined ZrC samples that were previously irradiated
- Plan irradiation study of ZrC-TRISO fuel particles

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 (6 projects):

- Instrumentation, controls, and diagnostics
- Advanced light water reactors (LWR)
- Advanced LWR fuels and materials technology
- LWR safety technology
- Advanced LWR Computational methods

2003 (5 projects):

- Next generation reactor/fuel cycle technology
- Innovative nuclear plant design
- Advanced nuclear fuels and materials

2004 (6 projects):

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

2005 (4 projects):

- Hydrogen production by nuclear systems
- Advanced fuels and materials development

2006 (4 projects):

- Sodium-cooled fast reactor
- Advanced fuels and materials development

2007 (7 projects):

- Advanced gas-cooled fast reactor
- Sodium-cooled fast reactor
- Advanced fuels and materials development
- Next-generation reactor and fuel-cycle technology

10.2 Project Summaries

The final three remaining FY 2004 projects were completed during the past fiscal year, along with one started in FY 2005. Research continued on three other collaborative projects awarded to Korean partners in FY 2005 and four awarded in FY 2006. Seven new projects were initiated in FY 2007.

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 2007 accomplishments and abstracts of the new projects.

Directory of Project Summaries

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

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

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

Collaborators: Idaho National Laboratory

Project Number: 2004-001-K

Program Area: Generation IV

Project Start Date: June 2004

Project End Date: May 2007

Research Objectives

This research project supported 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 addressed the thermal-hydraulic and safety analysis capabilities that will be used in reactor plant analysis and auditing. Specific objectives of this research were 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 thermal-hydraulic and safety analyses.

Research Progress

This research addressed the thermal-hydraulic and safety analysis capabilities that are to be used in the reactor plant analysis/auditing work scope. Specific objectives of this research were 1) development of a formal qualification framework, 2) initial filtering of the existing databases, and 3) preliminary screening of tools for use in thermal-hydraulics and safety analyses.

The work completed in this project is an essential step in the development of codes and selection of experiments that must be performed to support eventual reactor demonstration. These tools will establish the thermal-hydraulics conditions and safety margins and will be a basis for setting reactor technical specifications. It is imperative that the nuclear systems codes used to compute thermal-hydraulic margins have been properly developed to ensure that the relevant physical phenomena are modeled and that the uncertainties in code predictions are known.

Both the PBR and the PMR have been included in the work. The research in this project identified the systems models to be used and the model development needs and supporting experiment requirements for code qualification through the generation of phenomena identification and ranking tables (PIRTs). In the absence of specific reactor plant designs, preliminary generic PIRTs have been developed for three high-priority accident sequences: low pressure conduction cooling (LPCC), high pressure conduction cooling (HPCC), and load change. An additional three transient scenarios were then selected for the generation of PIRTs: the water ingress accident, intermediate heat exchanger (IHX)/hydrogen plant side upsets, and a rod withdrawal anticipated transient without scram (ATWS). Subsequent to this joint work, researchers undertook the task of developing the PIRT for the water-cooled reactor cavity cooling system (RCCS). Details of the scenarios were developed.

To further explore the modeling needs for these phenomena and proceed further with the code scaling, applicability, and uncertainty (CSAU) process, the work proceeded to the point of identifying underlying “sub-phenomena” and performing initial scaling analysis and sensitivity calculations to evaluate the relative importance of these sub-phenomena to the safety criteria. For the events that had been screened, the RCCS heat removal and the core flow distribution were identified as highly ranked phenomena in importance for the safety criteria. The steady-state core flow and the bypass at normal operation set the core initial peaking conditions for the accidents and transient scenarios.

Part of this project's focus has been on these phenomena, for which the researchers have performed calculations. To aid in this evaluation, they produced prototypic HPCC, LPCC, and LC conditions for the generic reference plants using the MARS-GCR and GAMMA codes and a scale-down analysis started for the PMR option. The results were used to identify the range of parameters of importance to the phenomena which occur in these sequences.

The project identified the thermal-hydraulic and safety systems tools to be used and a preliminary top-level filtering has been performed, but task work remains to be done due to the funding situation on the U.S. side. The filtering of the thermal-hydraulic and safety tools which were selected was performed with a review of their applicability relative to the PIRTs and scaling parameters application ranges. Areas of improvement continue to be identified.

A first-iteration review of the publicly available experimental data relevant to the identified phenomena has also been carried out. In screening this data, the researchers generated tables linking the selected accidents and identifying relevant top-level phenomena, along with the facility at which the data originated. Both the PBR and the PMR were included in this work.

In addition, key scoping type benchmark analyses were performed to identify the range of parameters of importance to the phenomena which occur in these sequences. Scoping type steady-state analyses were performed using MARS-GCR and GAMMA to compare with the International Atomic Energy Agency's (IAEA) PBMM and HTR-10 benchmarks. Comparisons were also made with air-ingress and computational fluid dynamics (CFD) benchmark experiment data. Researchers also included CFD code-to-code benchmarking with these codes for the lower plenum region and a set of reference plant transient analysis relevant to the PIRT development.

Planned Activities

This I-NERI project has concluded.

INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE

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

PI (Korea): Y.Y. Bae, Korea Atomic Energy Research Institute (KAERI)

Collaborators: Rensselaer Polytechnic Institute

Project Number: 2004-002-K

Program Area: Generation IV

Project Start Date: October 2004

Project End Date: September 2007

Research Objectives

Because of the lack of phase change in the supercritical water reactor (SCWR) core, these reactors cannot use light water reactor (LWR) 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, little information exists about the type of modeling needed to accurately predict the system behavior of a supercritical reactor.

The objective of this project is twofold: 1) to address the issues associated with measuring heat transfer to supercritical water at prototypical SCWR conditions and 2) to develop tools to predict SCWR thermal transients. In addition to using supercritical water as an experimental medium, researchers will also use surrogate fluids such as CO_2 at supercritical conditions. These alternative fluids will provide valuable insight into the physical phenomena that may be present. Because of their lower critical temperatures and pressures, they can significantly reduce the cost and time to complete the experimental program.

Research Progress

Research this year concentrated on joint analysis of supercritical CO_2 data from the KAERI test loop. Data was analyzed using both the NPHASE computational fluid dynamics code and the FLUENT code, using the same set of experiments in order to draw comparisons between the two analytical methods. A schematic of the test loop is shown in Figure 1.

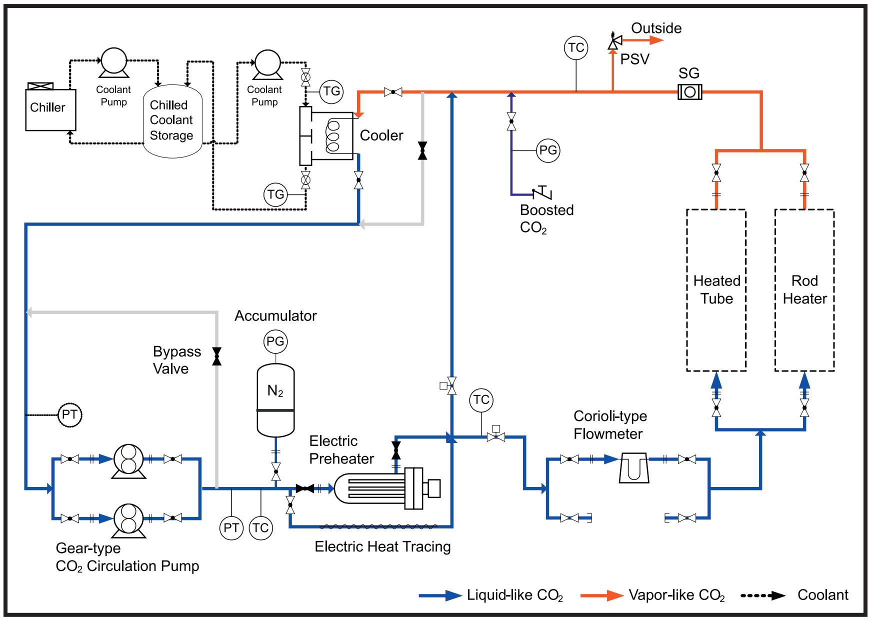


Figure 1. KAERI supercritical CO_2 test loop.

Overall, both NPHASE and FLUENT performed acceptably in analyzing a wide range of test conditions. As an example of this agreement, calculated temperature distribution and heat transfer coefficients in the test section are compared to measured values in Figures 2 and 3, respectively, for one specific test.

Researchers have reached the following conclusions as a result of this collaboration:

- Demonstrated that local flow and heat transfer effects are very important in the evaluation of heated wall temperature, explaining why standard 1-D correlations fail to predict the peak heat transfer coefficient

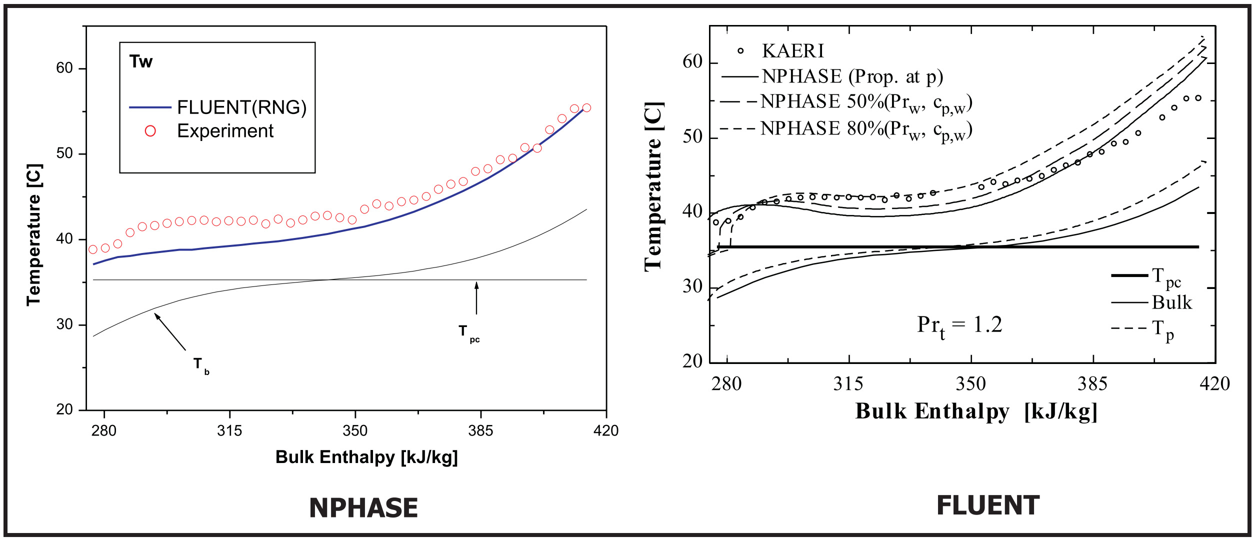


Figure 2. Temperature distribution predictions compared with experimental data.

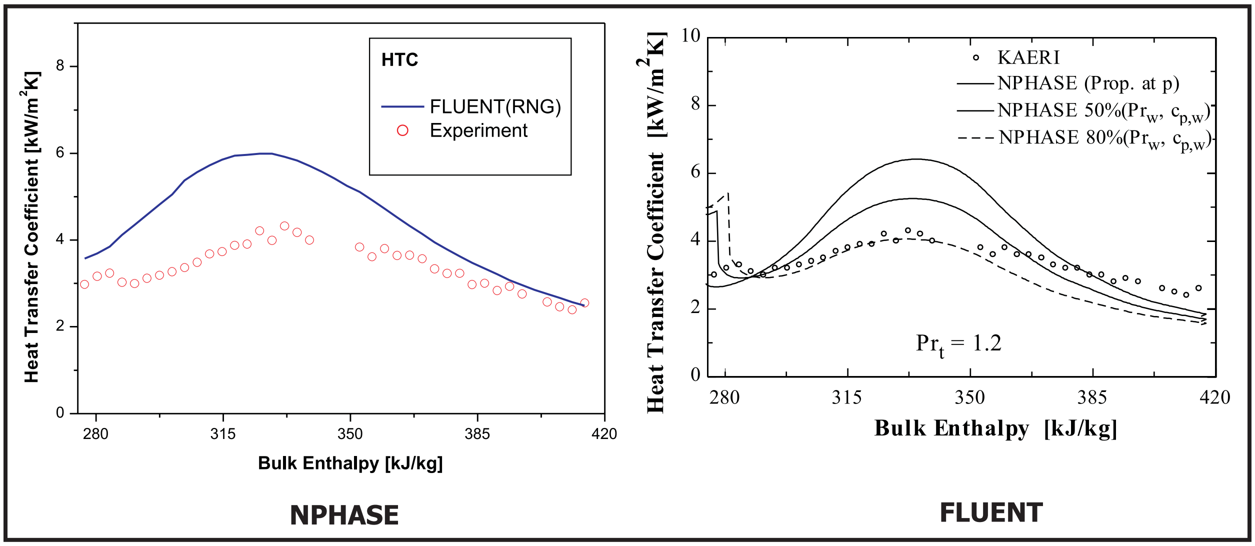


Figure 3. Calculated heat transfer coefficients compared with experimental data.

- Proven that results of multidimensional predictions depend on the modeling details of both kinematic and thermal aspects of turbulence
- Identified the use of a proper approach to the modeling of local changes in the fluid properties across the turbulent boundary layer as the key factor affecting the accuracy of predictions of the peak heat transfer coefficient

Planned Activities

This project has been completed and no further activities are planned.

INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE

Development of Voloxidation Process for Treatment of LWR Spent Fuel

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

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

Collaborators: Oak Ridge National Laboratory (ORNL)

Project Number: 2004-004-K

Program Area: AFC R&D

Project Start Date: June 2004

Project End Date: April 2007

Research Objectives

This research project developed a head-end process, known as voloxidation, for aqueous or pyrochemical treatment of spent nuclear fuel. Voloxidation, involving the oxidation of uranium oxide fuel at high temperature using either air or oxygen, provides three important advantages. First, it may be used to separate the fuel from the cladding, which would simplify process flow sheets by excluding the cladding constituents from the process stream. Segregation of cladding may also generate less high-level waste. Second, voloxidation decreases fuel particle size as a result of the oxidation cycle, which may improve the efficiencies of downstream dissolution processes. Third, voloxidation treatment may remove problematic constituents from the fuel prior to downstream treatment operations. Gaseous fission products such as cesium, krypton, xenon, technetium, iodine, carbon, and tritium (^3H) may be removed prior to fuel dissolution, thus simplifying the process flow sheets and yielding more flexible waste treatment operations.

The objective of this research is to develop a voloxidation process for light water reactor (LWR) spent fuel, which provides a means of separating fuel from the cladding, reduces particle size, and removes volatile fission products. 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

Research Progress

As a culmination of the project, researchers applied those conditions developed to maximize release and capture of the volatile and semi-volatile fission product gases to the design of an off-gas treatment system (OTS). Although jointly designed, the researchers fabricated separate systems (Figure 1) in order to perform collaborative experimentation. Both systems include four distinct 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, technetium, and iodine to be collected on fly-ash, calcium-based, and zeolite filters (Figure 2), respectively. Other secondary fission products to be trapped include rubidium, molybdenum, ruthenium, rhodium, and tellurium.

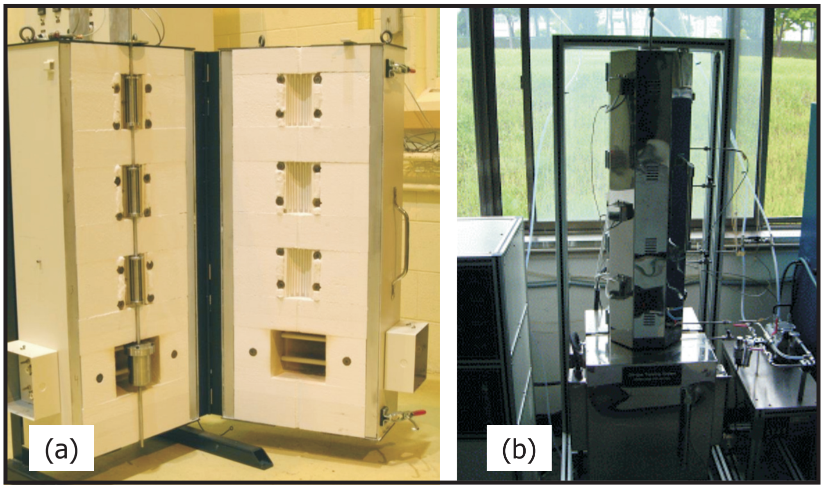


Figure 1. Photographs of off-gas treatment system at (a) INL and (b) KAERI.

Figure 2. Off-gas filter media, L to R: calcium-based, zeolite, and fly-ash.

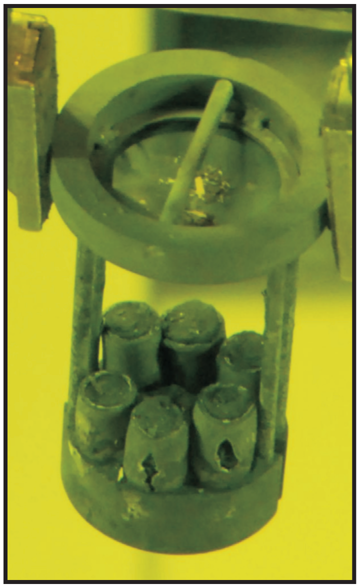


Figure 3. Oxidized fuel segments following irradiation testing.

During the past year, surrogate testing with non-radioactive materials was performed at the KAERI facilities while experimentation with irradiated LWR fuels was conducted in the Hot Fuel Examination Facility at INL. This complementary research program proved invaluable after researchers encountered challenges with the operating conditions during testing of the spent oxide fuels. Fundamental data were readily obtained with surrogates so that testing could be continued with the spent fuels. A total of 11 tests, eight surrogate and three irradiated, were conducted in the two facilities to evaluate the trapping efficiency of the filters and optimize the operating parameters with respect to the removal efficiency of volatile fission products.

Analytical chemistry results from samples taken of the oxidized fuel (Figure 3) and filter media following the tests with spent LWR fuel revealed that iodine can be completely removed and collected on the zeolite filter media. In addition, significant quantities of technetium and cesium (as high as 99 percent) can be removed and trapped on the calcium-based and fly-ash filters, respectively.

Planned Activities

This project has concluded. However, research to optimize the process in terms of the removal and collection of volatile fission products will continue via a follow-on I-NERI project.

INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE

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

PI (U.S.): N. Li, Los Alamos National Laboratory (LANL)

PI (ROK): T. Song, Korea Atomic Energy Research Institute (KAERI)

Collaborators: Seoul National University (SNU)

Project Number: 2004-005-K

Project Start Date: June 2004

Project End Date: May 2007

Research Objectives

The objectives of this research project were 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 explored both subcritical and critical systems in support of the Advanced Fuel Cycle Initiative (AFCI) and Generation IV lead-alloy-cooled fast reactor (LFR) programs. They tested 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 expected to develop internationally acceptable procedures for measuring corrosion rates by conducting tests with a set of controlled materials.

This project consisted 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 defined the materials and test conditions. The research team incorporated improved oxygen sensors, accurate flow meters, and coolant chemistry monitoring techniques into the process. They systematically constructed 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

The researchers achieved the following results after the third year of the project:

Loop Corrosion Measurement and Benchmarking. The objective of this task was to obtain corrosion data for steel samples immersed in the lead-bismuth (Pb-Bi) and Pb test loops developed by the collaborating laboratories. Researchers tested conventional reactor materials and selected new cladding materials, including silicon (Si) and chromium (Cr)-containing alloys, to identify suitable candidates for further development.

They conducted Pb-Bi and Pb corrosion measurements at high temperature, testing 316L, HT-9, and T91 alloys in 450°C Pb-Bi for up to 1,200 hours at flow velocity of 1.1 – 1.8 m/s. Oxygen content of the 333 and 666 hour samples was $9\text{x}10^{-7}$ weight-percent and $2.8\text{x}10^{-7}$– $9\text{x}10^{-7}$ weight-percent for the 1,200 hour samples. No firm oxide layer was formed for any sample. Researchers could see the corrosion attack when testing 316L. This unexpected result may come from the failure of the oxygen control due to malfunctioning of the oxygen sensor.

Researchers also tested the same samples in 570°C Pb-Bi for 300 hours. The velocity and oxygen content were 1.1–1.8 m/s and $1.0\text{x}10^{-7}$ weight-percent, respectively. In the case of HT-9 and T91, a partially formed oxide layer was observed. The 316L sample suffered a severe corrosion attack.

Testing of 316L and T91 alloys in 450°C Pb for 850 hours at flow velocity of 1.2 – 4.8 m/s resulted in no observable oxide layer on the 316L sample using scanning electron microscopy (SEM). However, about 9 microns of Fe_3O_4 and 1 micron of Cr spinel were found on the T91 samples. The T91 oxide layer looks similar to that of HT-9, but a broken Fe_3O_4 layer was observed in both samples.

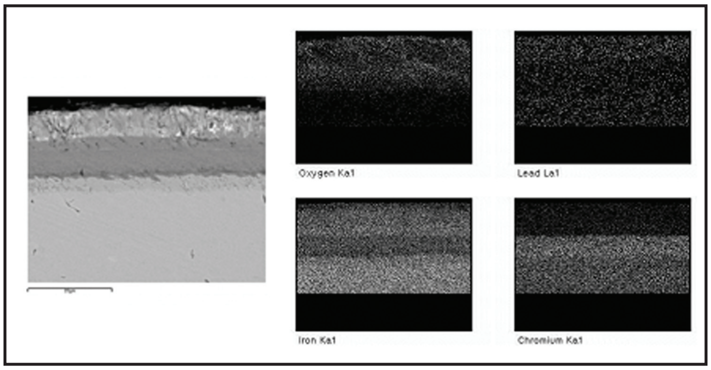


Figure 1. Results from energy dispersive X-ray (EDX) analysis for the T91 sample (570°C, Pb).

Researchers conducted Pb-Bi loop tests at low temperature using four types of steel: SS316L, HT-9, T-91, and EP-823. All specimens were put into a specimen cage and loaded into the test loop without additional surface preparation. Experiments were carried out at 450°C for 333 hours, 666 hours, and 1,000 hours with a specimen size of 35mm x 8mm x 1mm. The lead-bismuth eutectic (LBE) flow velocity at the test section was about 0.35 m/sec and the oxygen concentration in LBE was about 10^{-6} weight-percent. Figure 2 shows the test specimens after being discharged from the LBE.

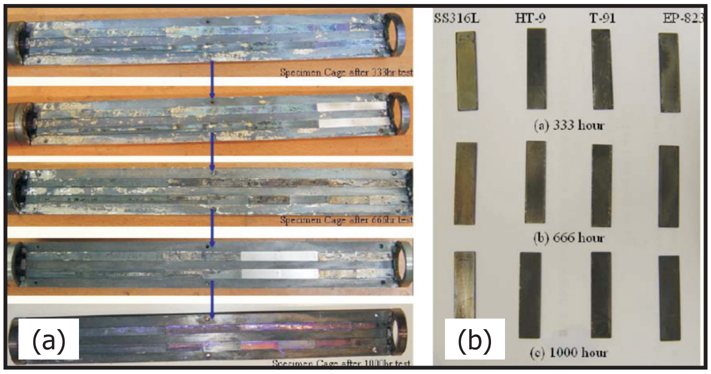


Figure 2. (a) Specimen cage discharged after exposure to lead-bismuth eutectic (LBE). (b) Test specimens washed in hot glycerin.

The researchers conducted online corrosion monitoring in the HELIOS test loop. Due to significantly reduced funding to operate the DELTA Loop, LANL analyzed the microstructure, mechanical and transport properties of oxide on the previous LBE-tested specimens (in DELTA or IPPE Loops) in much greater detail using atomic/magnetic force microscope (AFM/MFM), conductivity-AFM (C-AFM), and nano-indentation. The C-AFM measurement provides a basis for detailed understanding of oxide conductivity on a nano-scale, and will be coupled to macro electric impedance spectroscopy (EIS) measurement to establish online corrosion monitoring. Selected results from HELIOS and EIS measurements are shown in Figure 3.

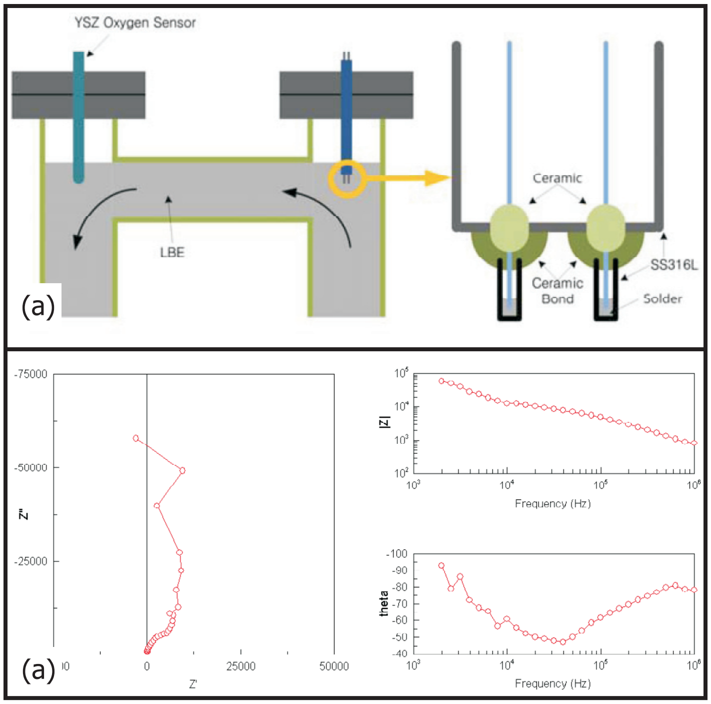


Figure 3. (a) Online corrosion measurement experimental setup in HELIOS; (b) EIS (electric impedance spectroscopy) measurement results showing evidence of oxide growth at the end of corrosion test.

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

The team constructed oxygen sensors for the material tests at KAERI and for static cell tests and material corrosion tests at SNU. Many of the sensors were damaged during or after calibration, mainly by thermal shock that caused breakage of the YSZ tube. One sensor was damaged due to oxidation of the feed-through lead-wire at high temperature during long-time material testing, indicating that the lead wire needs further protection.

The researchers developed and tested an oxygen control system (OCS), using the reaction between hydrogen gas and water vapor, and found it to have excellent stability. However, the predicted oxygen concentration based on the mass flow ratio of water to hydrogen $m(H_2O)/m(H_2)$ revealed a non-negligible discrepancy with measured oxygen potential. Therefore, the researchers recommend

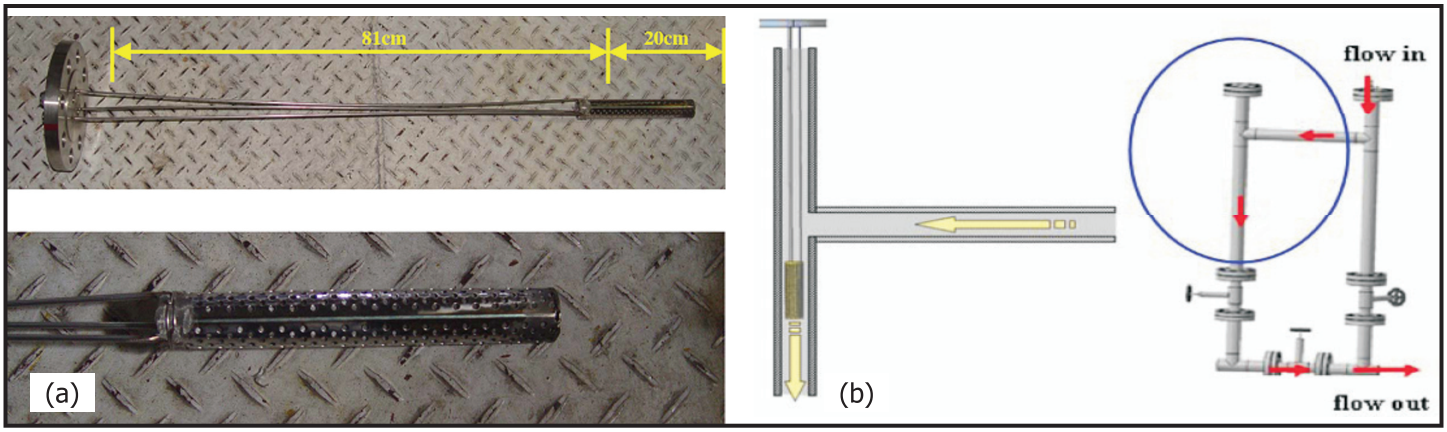


Figure 4. (a) Photos of the corrosion product filter; (b) Location of filter installation in HELIOS.

against relying on this mass ratio control method for accurate oxygen activity management over a wide activity range. The OCS was used in the calibration of the YSZ sensor at several discrete oxygen levels, which is a good option for confirming sensor accuracy in the desired range of oxygen concentration.

Corrosion Product Transport and Low-Temperature Embrittlement. A systematic framework was developed to construct a set of system corrosion kinetics models by incorporating interface oxidation and scale removal with hydrodynamic transport and redistribution of corrosion products. The team collected lead-alloy samples during operation of the lead-alloy loop and analyzed the constituent elements for comparison with the theoretical prediction. They postulated a mechanism of low-temperature embrittlement and examined a means of accelerating and measuring the embrittlement. Upon completion, they verified and tested the mechanism and conducted fractographical analysis.

Corrosion product measurement were made using HELIOS. Researchers designed a filter for installation in a downward flow location after the heat exchanger. Cylindrical stainless steel screens with a closed bottom end (Figure 4) used as filtering elements failed to collect corrosion products. Further experiments are necessary to identify the cause.

KAERI installed a bucket containing a known amount of Pb in the sump tank of their KPAL-II Pb loop. After performing the Pb corrosion tests at 450°C and 570°C, the Pb was drained to the sump tank and a sample collected for later elemental analysis. Inductively coupled plasma atomic emission spectroscopy (ICP-AES) analysis showed that Cr, Fe, and Ni contents in the lead sample were less than 100 µg/g and the oxygen content was 0.005 weight-percent.

More experiments and theoretical research are needed to fully understand the behavior of liquid metal eutectic (LME) at low temperatures. It is necessary to test many kinds of steel for embrittlement under flowing or static LBE. The team has designed and fabricated LME testing equipment for use in the flowing condition test (Figure 5).

Experiments were conducted both in air and LBE at various conditions ranging from 200–400°C and strain rates from $3x10^{-5}$ – 10^{-4} /s, with a low oxygen concentration maintained by injecting 5 percent H_2/Ar cover gas into the test chamber. The test results showed negligible effects of LME on 316L SS in LBE under the test condition.

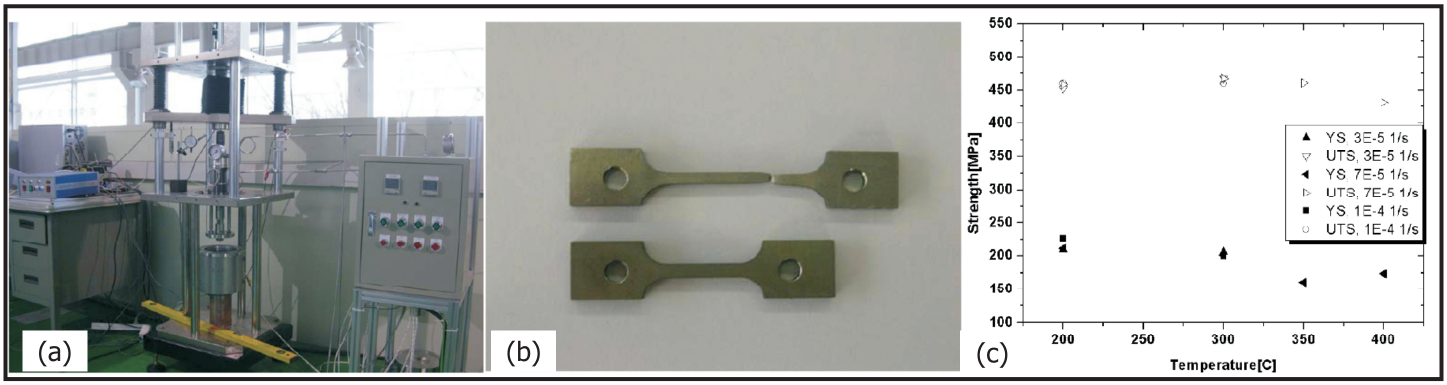


Figure 5. (a) SNU LME experimental setup; (b) Tested specimens; (c) Yield and ultimate strength at various temperatures.

Planned Activities

This project is complete. The international collaboration has achieved the majority of its objectives and laid a good scientific and technological foundation for further development. The mid-project funding reduction in the U.S. was a result of DOE programmatic priority re-alignment with the launch of Global Nuclear Energy Partnership. The ROK partners completed all tasks. Collaborations in other forms continued.

INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE

Supercritical Carbon Dioxide Brayton Cycle Energy Conversion

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

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

Collaborators: None

Project Number: 2005-001-K

Program Area: Generation IV

Project Start Date: October 2005

Project End Date: September 2008

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-CO_2 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:

- Development of design concepts for S-CO_2 Brayton power conversion systems
- Evaluation of turbine and compressor performance
- Development of design concepts for primary-to-secondary heat exchangers and recuperators
- Analysis of plant transient behavior for an SFR coupled to an S-CO_2 Brayton cycle power converter
- Development of a plant dynamics code for the S-CO_2 Brayton cycle energy converter coupled to an LFR
- Investigation and development of control strategies for the S-CO_2 Brayton cycle
- Dynamic analyses of plant transient behavior for specific control strategies
- Improvement of S-CO_2 Brayton cycle turbomachinery modeling based upon interactions with a commercial turbomachinery vendor

Research Progress

All objectives for the first two years of the project have been met. The following is a summary of progress during the second year.

Researchers developed the RECOBA and DENOP system design codes for an S-CO_2 Brayton cycle energy converter and used them to develop a layout (Figure 1) for the 600 MWe (1,523 MWt) KALIMER-600 design, one of three reference SFR plants under the Generation IV International Forum (GIF).

The one-dimensional TUBDEP and COMP1D turbine and centrifugal compressor design codes were used to develop design concepts for the KALIMER-600 S-CO_2 Brayton cycle. The researchers evaluated turbine performance with three-dimensional computational fluid dynamics (CFD) calculations. They also developed a one-dimensional computer code and applied it to develop concepts for diffusion-bonded primary-to-secondary heat exchangers and recuperators with single-banked, zigzagged semicircular channels (similar to a Printed Circuit Heat Exchanger™ [PCHE™]). The STASCOR computer code was developed for analyzing sodium-carbon dioxide (Na-CO_2) chemical reactions coupled to the system thermal hydraulic behavior and an analysis carried out for a potential interfacial boundary failure accident in a shell-and-tube Na-to-CO_2 heat exchanger.

Researchers continued developing a systems-level plant dynamics code to model the S-CO_2 Brayton cycle coupled to a natural circulation LFR plant. The code was significantly modified and improved to incorporate a realistic cooler

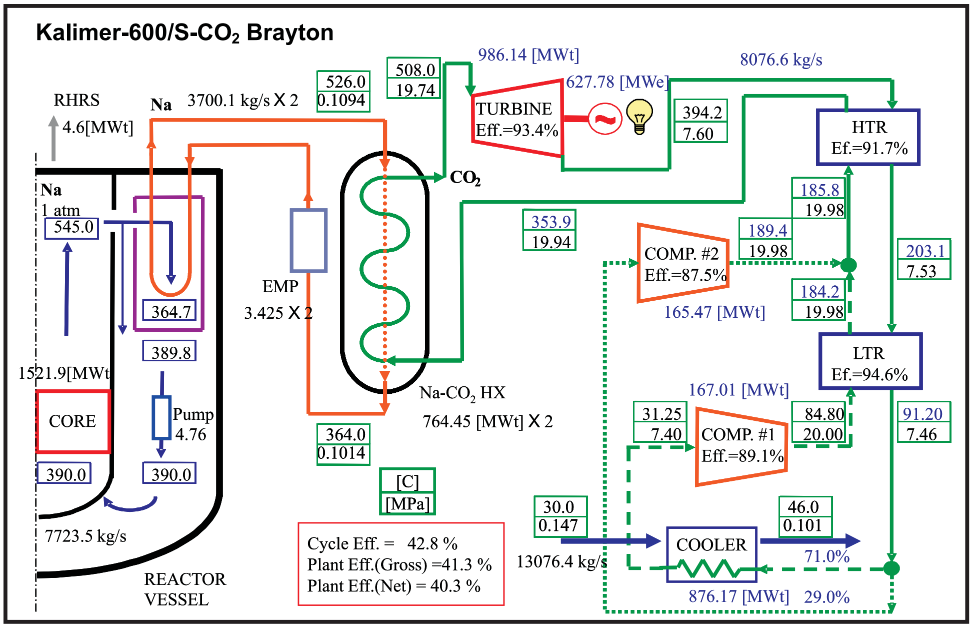


Figure 1. Schematic of S-CO_2 Brayton cycle coupled to KALIMER-600.

model that enables simulating specific postulated accident initiators that rapidly affect conditions near the critical point. Using the modified code, it is possible to demonstrate that the temperature at the inlet to the first stage of the main compressor does not fall below the critical temperature, thereby avoiding the onset of two-phase flow that potentially degrades operation or damages compressor blades. The code was also improved to support specification of the configuration of components and piping through the input file facilitating easy investigation of changes in cycle layout/configuration.

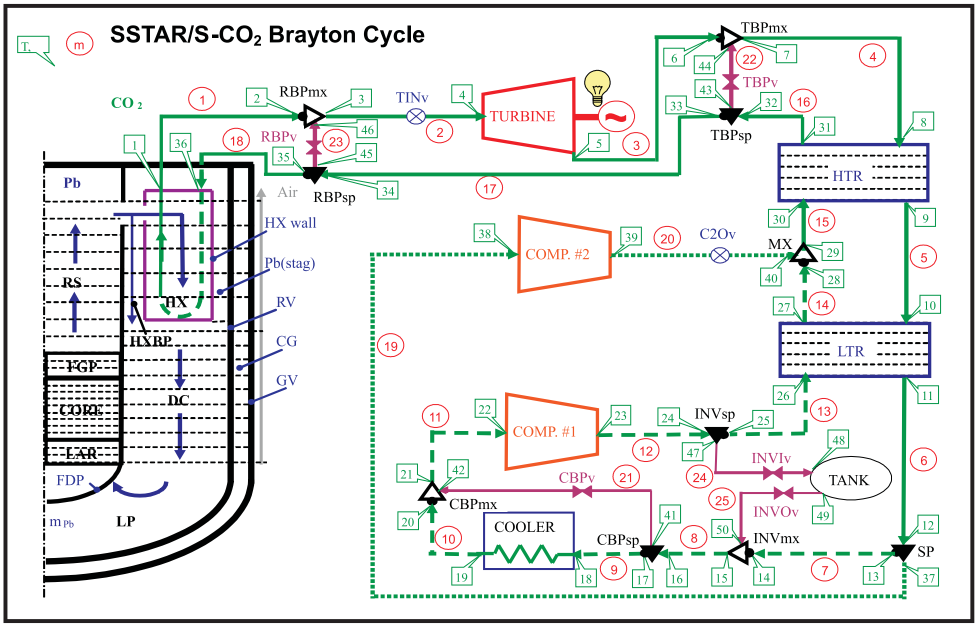


Figure 2. Plant Dynamics Code nodalization of 20 MWe (45 MWt) SSTAR LFR plant.

An improved control strategy was developed for the 20 MWe (45 MWt) Small Secure Transportable Autonomous Reactor (SSTAR) LFR S-CO_2 Brayton cycle involving: 1) turbine bypass control for fast changes in heat removal at all load levels, 2) additional inventory control between 90 and 35 percent nominal load (limited by the inventory control tank volume) to maintain cycle temperatures, 3) cooler bypass control to maintain CO_2 temperature at the main compressor first stage inlet (i.e., the lowest temperature in the cycle) above the 30.98°C critical temperature, avoiding the onset of two-phase flow, 4) cooling water flow rate control that prevents full opening or closing of the cooler bypass valve, and 5) compressor surge prevention systems to preclude compressor damage during postulated accidents.

Researchers carried out plant dynamics code calculations to simulate the behavior of the S-CO_2 Brayton cycle, including the automatic control system response and the coupled reactor–autonomous load following response. Figure 2 shows the nodalization for the SSTAR plant.

The team continued interacting with the custom-designed turbomachinery manufacturer that is fabricating a small-scale compressor for testing with CO_2 near the critical point; testing is expected to begin about April 2008.

Planned Activities

Researchers will continue developing and evaluating major components for the S-CO_2 Brayton cycle. They will compare compressor modeling incorporated in the plant dynamics code with available data from the small-scale S-CO_2 compressor tests.

INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE

Development of HyPEP, A Hydrogen Production Plant Efficiency Calculation Program

PI (U.S.): C. Oh, Idaho National Laboratory

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

Collaborators: R. Vilim, Argonne National Laboratory

Project Number: 2005-002-K

Program Area: NHI

Project Start Date: October 2005

Project End Date: September 2007

Research Objectives

This research project evaluated and optimized cycle efficiencies for producing hydrogen and electricity in a very high-temperature reactor (VHTR). Such systems are complex and the calculations associated with optimizing them are intensive, involving a large number of operating parameter variations and many different system configurations. This research project produced 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 were 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 was fully developed and validated, researchers used it to execute calculations on candidate system configurations.

Research Progress

The researchers improved the graphical user interface (GUI) of the alpha version of HyPEP by fixing program bugs, enhancing graphics, and adding component models. They further developed the object classes TNode, TLink, and THeatBlock and coded for the Node, Link, and HeatBlock basic components. The object classes for other components are derived from the base components by object inheritance or object polymorphism and these have been developed and coded using the Delphi programming language. The object class tree developed for HyPEP is shown in Figure 1.

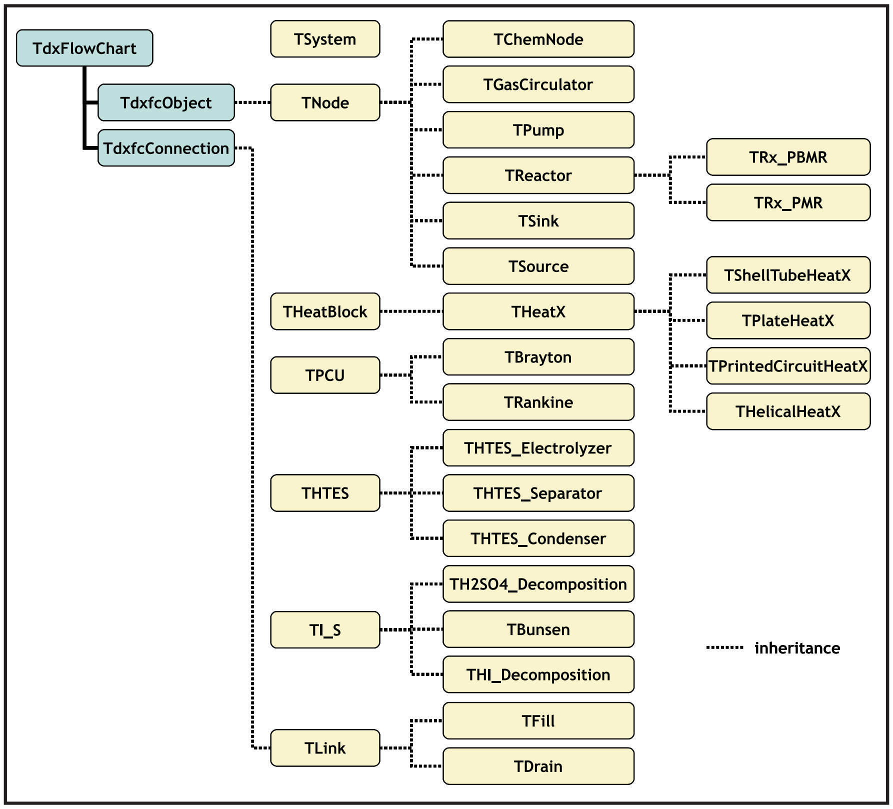
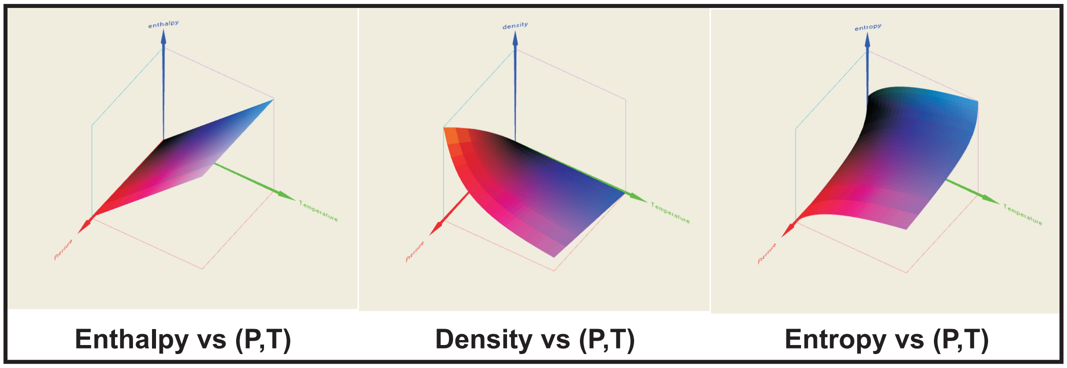


Figure 1. HyPEP object class definition tree.

Researchers developed various routines for the creation and deletion of components. Simplified thermodynamic models for specialized components such as the HTES, Brayton Cycle, and S-I process models have been established and are being coded into HyPEP.

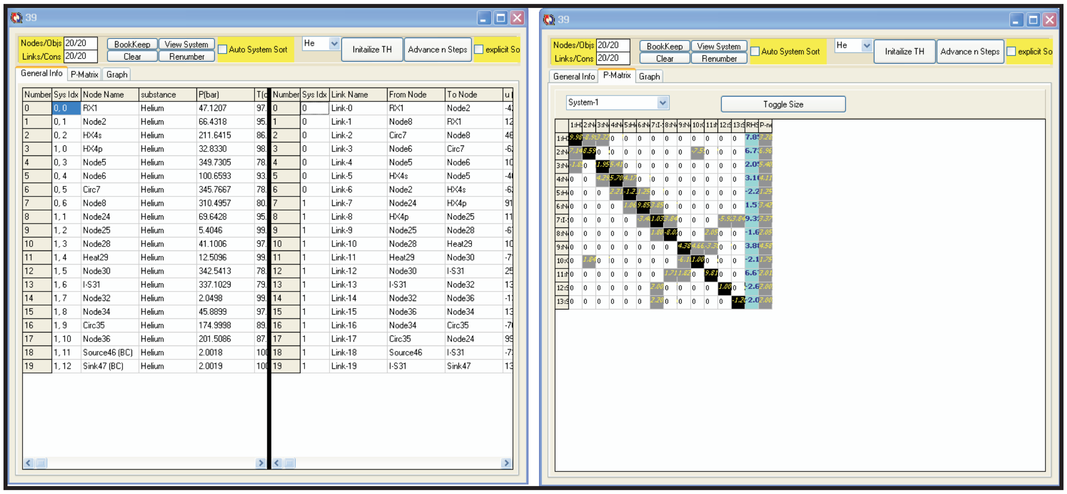
In the second year, researchers developed the following programs and code segments to facilitate simpler problem correction during the program qualification phase:

1) A stand-alone gas property testing program to graphically view the thermodynamic property surfaces of six substances in 3-dimensional surface, as shown in Figure 2.
2) Code segments to display the values of the elements of the pressure matrices of the system. This can be useful in examining the system topology and in viewing the current status of the matrix. An example is shown in Figure 3.
3) Code segments to display the current status of the system components.



Enthalpy vs (P,T) **Density vs (P,T)** **Entropy vs (P,T)**

Figure 2. Gas property surfaces (helium) from stand-alone program.



Number	Sys Idx	Node Name	substance	P(bar)	T(c
0	0, 0	RX1	Helium	47.1207	97.
1	0, 1	Node2	Helium	66.4318	95.
2	0, 2	HX4s	Helium	211.6415	86.
3	1, 0	HX4p	Helium	32.8330	98.
4	0, 3	Node5	Helium	349.7305	78.
5	0, 4	Node6	Helium	100.6593	93.
6	0, 5	Circ7	Helium	345.7667	78.
7	0, 6	Node8	Helium	310.4957	80.
8	1, 1	Node24	Helium	69.6428	95.
9	1, 2	Node25	Helium	5.4046	99.
10	1, 3	Node28	Helium	41.1006	97.
11	1, 4	Heat29	Helium	12.5096	99.
12	1, 5	Node30	Helium	342.5413	78.
13	1, 6	I-S31	Helium	337.1029	79.
14	1, 7	Node32	Helium	2.0498	99.
15	1, 8	Node34	Helium	45.8899	97.
16	1, 9	Circ35	Helium	174.9998	89.
17	1, 10	Node36	Helium	201.5086	87.
18	1, 11	Source46 (BC)	Helium	2.0018	10
19	1, 12	Sink47 (BC)	Helium	2.0019	10

Number	Sys Idx	Link Name	From Node	To Node	u
0	0	Link-0	RX1	Node2	-4
1	0	Link-1	Node8	RX1	12
2	0	Link-2	Circ7	Node8	46
3	0	Link-3	Node6	Circ7	-6
4	0	Link-4	Node5	Node6	10
5	0	Link-5	HX4s	Node5	-4
6	0	Link-6	Node2	HX4s	-6
7	1	Link-7	Node24	HX4p	91
8	1	Link-8	HX4p	Node25	11
9	1	Link-9	Node25	Node28	-6
10	1	Link-10	Node28	Heat29	10
11	1	Link-11	Heat29	Node30	-7
12	1	Link-12	Node30	I-S31	25
13	1	Link-13	I-S31	Node32	13
14	1	Link-14	Node32	Node36	-1
15	1	Link-15	Node36	Node34	13
16	1	Link-16	Node34	Circ35	-7
17	1	Link-17	Circ35	Node24	99
18	1	Link-18	Source46	I-S31	-7
19	1	Link-19	I-S31	Sink47	13

Figure 3. An example of SolverDisplay window showing the general information and the system matrix.

Work in the final year focused on integrating the VHTR and high-temperature steam electrolysis (HTSE).

In this system, the heat generated in the reactor core is transferred to the HTSE system through various heat exchangers that increase the steam temperature to more than 800°C. A solid oxide fuel cell electrolyzer converts the heated steam into hydrogen and oxygen molecules. Approximately 10 percent of the total energy is transferred as process heat and the rest as electricity generated in the power conversion unit (PCU). The PCU is based on the widely used closed-loop gas Brayton cycle, which shows high efficiency, especially at higher temperatures. The reactor core and PCU can be integrated in two different configurations: the direct cycle and the indirect cycle. In the direct cycle, the PCU receives high temperature gas directly from the primary system, in series with the reactor core. The indirect cycle incorporates intermediate heat exchangers, resulting in a more complicated configuration and reduced efficiency; however, separating the reactor core and PCU results in easier maintenance and should reduce the strong dynamic coupling between the two systems, which may improve safety.

In the current study, the efficiency of VHTR/HTSE system was estimated using HYSYS, a commercial process flowsheet analysis code that has been used in the chemical and petroleum industry. The HYSYS code supports models for many basic system components and has process optimization capability. Typical HYSYS components used in this study are the reactor, pump, compressor, heat exchanger, turbine, cooler, and vessel. VHTR system modules of various configurations are developed with these basic components. Since HYSYS does not model electro-chemical processes, researchers developed an electrolyzer model which they integrated into HYSYS for the balance of the HTSE process. They combined a HYSYS model for the VHTR system with the HTSE system to obtain the VHTR/HTSE plant simulation.

As part of the system integration, researchers evaluated five different configurations of the integrated system in terms of overall hydrogen production efficiency. Figure 4 illustrates the indirect parallel system. The flow in the secondary coolant system is divided, with most going towards the PCU and the remainder through a secondary heat exchanger (SHX) that directs heat towards the HTSE plant. The flow through the hot side of the SHX is then mixed with the flow from the PCU to feed the cold side of the intermediate heat exchanger (IHX). However, some flow is diverted away from the PCU, which decreases cycle efficiency.

There are three coolant loops:

1) The primary coolant system contains the nuclear reactor, the hot side of the IHX, and a compressor.
2) The secondary coolant system contains the cold side of the IHX, the hot side of the SHX, the PCU, and connecting piping, which is assumed to be short.
3) The intermediate heat transport loop connects the secondary coolant system to the HTSE plant through several process heat exchangers (PHXs).

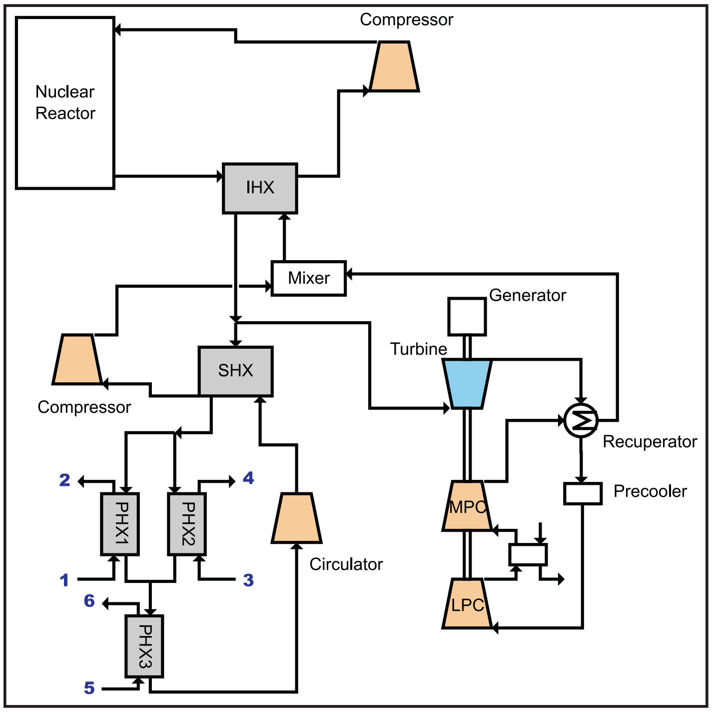


Figure 4. Configuration 1—indirect parallel cycle.

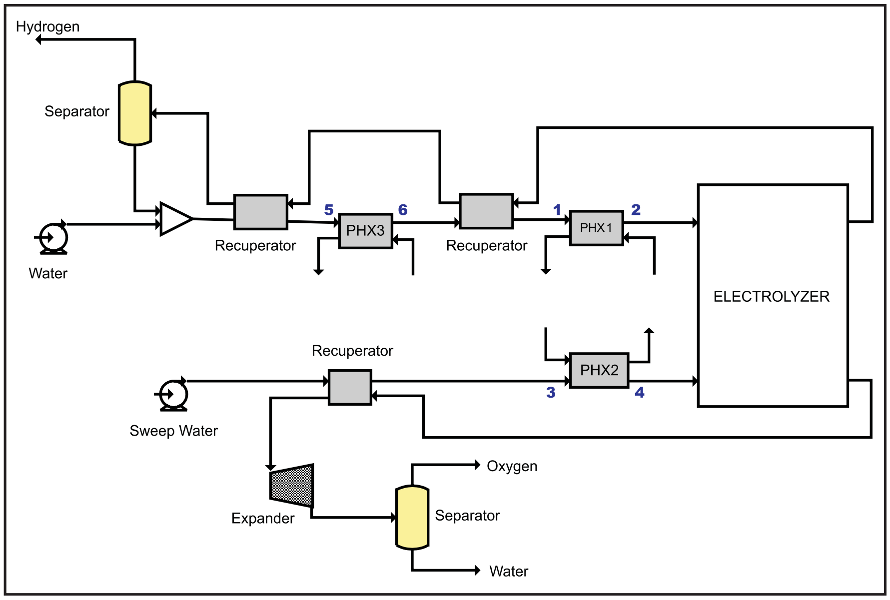


Figure 5. HTSE system.

For electrolysis, steam is heated above 800°C with the SHX, then converted into hydrogen and oxygen in the electrolyzer and discharged through their respective outlets, as shown in Figure 5. Three recuperators recover heat from the discharged gases. As the fuel product stream also contains steam, it discharges through a separator to collect the hydrogen gas and recycle the steam back to the inlet, as shown in Figure 5. In the oxidizer outlet stream, heat is first recuperated before passing through an expander to recover work. The oxygen and water components of the stream are then separated.

As part of the system integration, researchers developed an optimizing heat exchanger model and performed quasi-static and dynamic analysis of the VHTR/HTSE.

Planned Activities

This project has concluded.

INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE

Development of Head-End Pyrochemical Reduction Process for Advanced Oxide Fuels

PI (U.S.): S. D. Herrmann, Idaho National Laboratory (INL)

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

Collaborators: None

Project Number: 2005-004-K

Program Area: AFCI

Project Start Date: October 2005

Project End Date: September 2008

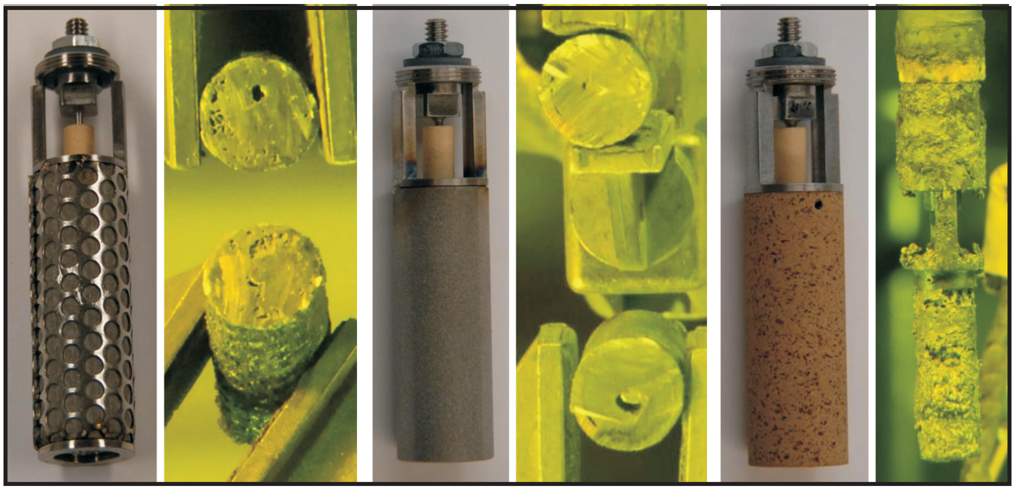
Research Objectives

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 Republic of Korea (ROK) have been actively developing pyrochemical conversion methods for producing feed material that is compatible with pyroprocessing. Both countries favor an 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 four bench-scale pyrochemical processing experiments with spent oxide fuel. These experiments involved the electrolytic reduction of irradiated uranium dioxide (UO_2) fuel that had been subjected to a voloxidation process to form U_3O_8. The primary variables in each of the four electrolytic reduction runs were 1) fuel basket containment material and 2) the concentration of lithium oxide (Li_2O) in the lithium chloride (LiCl) reduction salt. The basket materials were stainless steel mesh for run 1, sintered stainless steel for run 2, and porous magnesia for runs 3 and 4. The concentration of Li_2O was 1 weight-percent for runs 1 through 3 and 3 weight-percent for run 4. Researchers performed the four runs with a single loading of salt to assess the accumulation and potential effect of fission products on the process. Specifically, they submerged each of the baskets, loaded with 20 to 40 grams of spent fuel, into 500 milliliters of 650°C molten lithium chloride salt containing the prescribed amount of lithium oxide. An electric current applied between the basket (cathode) and a platinum wire (anode) reduced the ceramic fuel to metal in the basket, while simultaneously forming oxygen gas on the surface of the platinum wire. After applying a sufficient charge, researchers removed the fuel baskets from the reduction apparatus and sectioned them to obtain the fuel samples. As the porous magnesia cathode containment failed after runs 3 and 4, researchers were unable to obtain a representative post-test fuel sample from run 4, although they were able to do so for run 3. Figure 1 shows the original fuel baskets and the post-test baskets and fuel forms for runs 1 through 3. This series of runs demonstrated a 90 to 99.96 percent reduction of uranium oxide to uranium metal. Analysis of the pre- and post-test salt samples for each of the runs identified the diffusion and accumulation of cesium, barium, strontium, rubidium, and tellurium from the fuel phase into the salt phase.



Run 1 - stainless steel mesh **Run 2 - sintered stainless steel** **Run 3 - porous magnesia**

Figure 1. Original cathode baskets (left) and post-test baskets and fuel forms (right) for runs 1 through 3, including stainless steel wire mesh, sintered stainless steel, and porous magnesia.

After processing sufficient fuel through electrolytic reduction, the fission product concentration rises to a point where either the salt must be discarded or the fission products removed. Researchers performed experiments to investigate the removal of fission products from reduction salt via ion exchange with zeolite. They prepared a nonradioactive LiCl salt mixture containing 1 weight-percent Li_2O, 2 weight-percent CsCl, and 2 weight-percent $SrCl_2$ and contacted it with zeolite under two different conditions: 1) with the salt mixture molten at 650°C and 2) with the salt mixture crushed and solid at 500°C. Researchers observed a limited uptake of cesium and strontium into the zeolite at 650°C, due to a transformation of the zeolite compound at this temperature which restricts the ion exchange. On the other hand, more than 99 percent of the cesium and strontium transferred from the salt mixture to the zeolite at 500°C.

During this fiscal year, researchers completed the post-test analysis of salt and fuel samples from electrolytic reduction runs at an engineering scale in KAERI's Advanced Spent Fuel Conditioning Process (ACP) facility. The engineering scale process operates with a porous magnesia fuel containment basket (configured as the cathode) and platinum anodes in LiCl with 3 weight-percent Li_2O at 650°C. Figure 2 depicts the engineering-scale system configuration in the ACP facility. The porous magnesia is capable of holding up to 20 kg of U_3O_8 particulate. Researchers sampled the post-test fuel product at various radial and axial positions within the porous magnesia containment and measured uranium reduction yields greater than 99.9 percent at all positions. They also observed elevated concentrations of lithium oxide in the reduced uranium product compared to the lithium oxide concentration in the bulk salt. Results of the engineering-scale runs prompted a redesign of the reduction vessel to improve the performance of the engineering-scale process.

Researchers also performed bench-scale experiments to assess the kinetics of the electrolytic reduction process. Specifically, they conducted a matrix of 16 electrolytic reduction tests by varying the applied charge from 20, 40, 60, and 80 percent of theoretical for 10 g loadings of U_3O_8 in porous magnesia baskets versus constant applied currents of 0.1, 0.2, 0.4, and 0.8A. By sampling the fuel following each run, researchers determined the extent of uranium reduction. These experiments demonstrated the direct reduction of uranium oxide without lithium metal formation at the 0.1, 0.2, and 0.4A conditions. However, accumulation of oxygen ions in the integrated cathode prohibited further reduction, causing current efficiencies to decrease. At the 0.8A condition, the cathode reached the lithium reduction

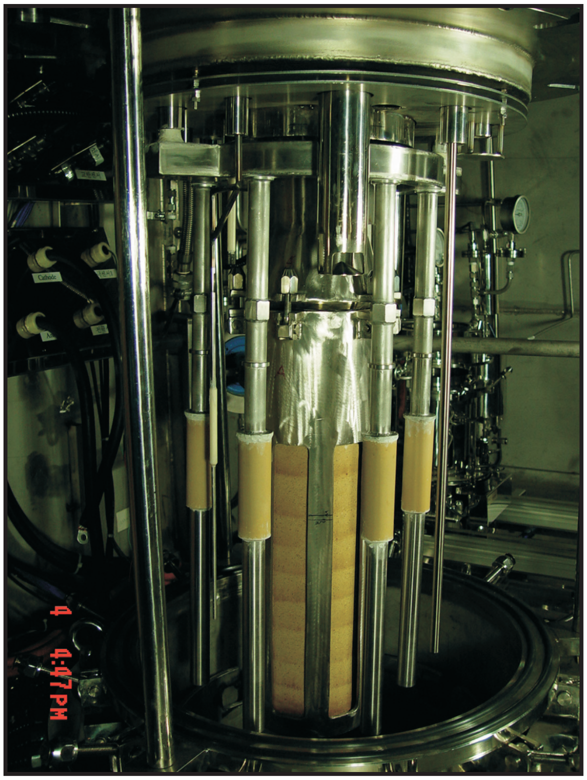


Figure 2. Engineering-scale reduction equipment in KAERI's Advanced Spent Fuel Conditioning Process (ACP) facility with vessel head in raised position.

potential at the onset of the run and the reduced fraction increased almost linearly with the supplied charge. The results of these bench-scale experiments will provide input for the development of a kinetics model for the electrolytic reduction process.

Planned Activities

Over the next period, the research team plans to conduct a series of electrolytic reduction runs with a single salt loading using high burn-up (18 percent) fast reactor mixed oxide (MOX) fuel in the hot-cell bench-scale electrochemical apparatus at INL. The oxide fuel for the tests will consist of crushed MOX fuel (as UO_2 / PuO_2), as well as the same fuel that will be subjected to a voloxidation process to form U_3O_8 for the uranium oxide portion of the mixed oxide. The purpose of the tests is to assess 1) the extent of metal oxide reduction, 2) the distribution of fission products between the salt and fuel phases, and 3) the possible effect that the accumulation of fission products may have on the electrolytic reduction process. The use of high burn-up fast reactor MOX fuel, as opposed to light water reactor fuel used in previous tests, allows for a closer examination of the extent of reduction for transuranic constituents and the potential impact that higher fission product concentrations may have on the process. The removal of fission products from reduction salt via zeolite ion exchange will be further investigated.

Researchers will perform three electrolytic reduction runs at engineering scale in KAERI’s modified reactor in the ACP facility. These runs will involve the electrolytic reduction of SIMFUEL, which is U_3O_8 loaded with nonradioactive fission products in concentrations that are representative of spent light water reactor fuel. Researchers will also collaborate in the development of a kinetics model for the electrolytic reduction process.

Core Design Studies for Sodium-Cooled TRU Burner Reactors

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

PI (Korea): Yeong-Il Kim, Korea Atomic Energy Research Institute (KAERI)

Collaborators: None

Project Number: 2006-001-K

Program Area: AFC R&D

Project Start Date: January 2007

Project End Date: September 2009

Research Objectives

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, long-term 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 loss 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, BFS-73-1, and BFS-75-1 critical experiments. Researchers will analyze the sensitivities of cross sections and computational methods on neutronics performance parameters and evaluate the uncertainties of performance parameters.

To evaluate 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.

Research Progress

Researchers developed metal and oxide fuel core concepts for a 1000 MWt advanced burner reactor (ABR). Using ternary metal and mixed oxide fuels, they developed compact core concepts of medium TRU conversion ratio through a trade-off between the burnup reactivity loss and TRU conversion ratio. The same radial core layout (see Figure 1) and assembly dimensions were used for both metal and oxide fuel core concepts in order to allow the interchange between metal and oxide fuel assemblies. As shown in Table 1, the average linear power of the metal core was 23.3 kW/m and 18.8 kW/m for the oxide core. High-purity plutonium (Pu) was assumed for the startup core, and TRU from light water reactor (LWR) spent fuel was used for the recycled core. For the metal fuel, the average TRU fraction was 15.5 percent for the startup core and 22.1 percent for the recycled core, with corresponding TRU conversion ratios of 0.81 and 0.73, respectively. The burnup reactivity loss over a cycle length of one-year was 3.6 %Δk for the startup core and 2.2 %Δk for the recycled core. The lower heavy metal density of the oxide fuel increased the TRU fraction to 18.2 percent for the startup core and 27.1 percent for the recycled core. The burnup reactivity loss and TRU conversion ratios for oxide fuel were comparable to those of the metal cores. For both metal and oxide cores, calculated kinetics parameters and reactivity coefficients were adequate to provide sufficient negative reactivity feedback, and the control systems provided more than adequate shutdown margins.

Parameter	Metal Fuel		Oxide Fuel	
	Startup Core	Recycle Core	Startup Core	Recycle Core
Linear power	23.3 kW/m		18.8 kW/m	
TRU fraction	15.5 %	22.1 %	18.2 %	27.1 %
TRU conversion ratio	0.81	0.73	0.83	0.71
Burnup reactivity loss	3.6 %Δk	2.2 %Δk	3.5 %Δk	2.0 %Δk

Table 1. Metal and oxide fuel parameters.

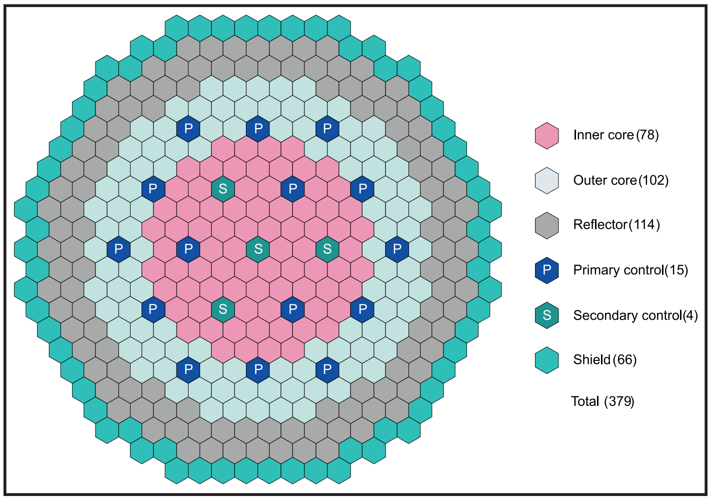


Figure 1. Radial core layout of 1000 MWt ABR concept.

The research team also performed a core design study for large monolithic reactors in order to identify the most limiting factor in scaling up transmutation core concepts. Two design approaches were examined: 1) fuel pin designs with region-dependent cladding thickness to flatten the power distribution with single fuel enrichment and 2) a pancake core to reduce the sodium void worth and achieve a high TRU transmutation rate. With each design approach, preliminary core designs were developed with metal fuel at three power levels: 600, 900, and 1,200 MWe. The cycle length was 332 effective full power days (EFPD) for the region-dependent cladding thickness designs and 220 EFPD for the pancake designs. The TRU enrichment was about 30 percent for all six core designs. The TRU conversion ratio was in the range of 0.55 to 0.59, and the burnup reactivity loss was in the range of 2.4 %Δk to 3.0 %Δk. For the same power rating, the pancake design approach yielded a smaller core volume and thus a smaller fuel inventory. The calculated reactivity coefficients were adequate to provide negative reactivity feedback, although the sodium density coefficient became more positive and the axial and radial expansion coefficients became less negative with increasing power rating. Design refinements are in progress to ensure the capability to provide inherent protection against unprotected accident scenarios.

In an effort to verify and validate the design analysis methods, the researchers analyzed the six benchmark problems for the ZPPR-21 critical experiments phases A through F specified in the *Handbook of Evaluated Criticality Safety Benchmark Experiments*. For all six configurations, the core multiplication factor determined with deterministic design tools agreed with Monte Carlo solution within 0.20 %Δk, and there was no indication of any systematic bias. The statistics of differences between calculated values and specified benchmark experimental values showed similar agreement (-0.28 %Δk to 0.33 %Δk) for deterministic and Monte Carlo methods. The research team also collected and reviewed the BFS-73-1 and BFS-75-1 experiment data, including criticality, reaction rate ratios, fission rate distributions, and control rod and sodium void worth. Benchmark specifications were developed for the homogenized sub-assembly models of BFS-73-1 and BFS-75-1 critical assemblies and the problems were analyzed with ENDF/B-VI.6, ENDF/B-VII.0, JEFF-3.1, and JENDL-3.3 libraries. The calculated multiplication factor agreed well with the measured values within measurement uncertainties, except for the cases calculated with the JENDL-3.3 library. The reaction rate ratios predicted with three latest nuclear data libraries (ENDF/B-VII.0, JEFF-3.1, and JENDL-3.3) were in good agreement within 2 percent for plutonium (Pu-239) and uranium (U-238) fission reactions. These latest libraries significantly improved the prediction accuracy for U-238, Pu-240, and neptunium (Np-237) fission reactions. However, the calculated values still showed large discrepancies from measured data for americium (Am-241 and Am-243) and curium (Cm-246) fission reactions. These results suggest that americium cross sections need to be improved further, since Am-241 and Am-243 are the dominant minor actinides in the equilibrium cycle core of TRU burners.

To evaluate safety performance enhancements, the research team analyzed the performance characteristics of the ABR 1000 MWt design, focusing on the ability to provide inherent protection against damaging consequences in low probability accident sequences involving multiple equipment failures. The primary significance of the analysis results is that both metal and oxide core designs demonstrated significant safety margins to coolant boiling and fuel damage in *protected* loss of flow accident sequences. The metal core design also exhibited significant safety margins in the *unprotected* loss of flow accident sequence. This is a direct consequence of the high thermal conductivity and low operating temperature of metal fuel, and the favorable negative feedback due to thermal expansion. However, the unprotected loss of flow analyses for the oxide core design indicated that margins to coolant boiling would not be adequate without additional accident mitigation features. Additional enhancements, such as a self-actuating shutdown device, would be required for the oxide core to increase safety margins so that no fuel damage or cladding failures would occur, even when multiple safety systems are assumed to malfunction. The researchers also carried out preliminary estimation of safety characteristics of the six core concepts of large monolithic transmutation reactors using the quasi-static reactivity balance method. For all six core concepts, calculated integral reactivity parameters generally satisfied three sufficient conditions for acceptable asymptotic core outlet temperatures for possible unprotected accident scenarios. However, the 900 and 1,200 MWe core design with region-dependent cladding thickness did not satisfy the sufficient condition for unprotected transient overpower scenarios.

Planned Activities

Researchers will conduct the design optimization study for the large monolithic core concepts with iterations between core design and safety analysis. The dependence of core performance and safety parameters on the TRU conversion ratio will also be studied. Additionally, they will develop innovative fuel rod design concepts and evaluate the impacts on core performance parameters. ANL will develop ABR core designs with increased power level (2,000 and 3,000 MWt) and they will analyze a selected set of ZPPR-15 experiments. The research team will also analyze the sensitivities of cross sections on neutronics performance parameters and evaluate the uncertainties of performance parameters for the 1000 MWt ABR design.

Researchers at KAERI will analyze the ZPPR-21 experiments using design analysis tools they have developed. They will also perform detailed safety analyses for the large monolithic core concepts with the SSC-K safety analysis code.

INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE

Separation of Fission Products from Molten LiCl–KCl Salt Used for Electrorefining of Metal Fuels

PI (U.S.): Michael Simpson, Idaho National Laboratory

PI (Korea): In-Tae Kim, Korea Atomic Energy Research Institute (KAERI)

Collaborators: None

Project Number: 2006-002-K

Program Area: AFC R&D

Project Start Date: January 2007

Project End Date: September 2009

Research Objectives

The objective of this project is to develop a process for selectively removing fission products from the lithium-chloride–potassium-chloride (LiCl–KCl) eutectic salts used in electrochemical processing of spent nuclear fuel. Separating fission products from the salts is critical to minimizing the waste volume generated from electrorefining processing of spent nuclear fuel. The current method for disposing of fission products from the electrorefiner is simply to discard the LiCl–KCl salts along with the fission products in a waste form. This disposal option creates a larger quantity of high-level radioactive waste than if the fission products could be removed from the electrorefiner salt and disposed of separately. This would allow the electrorefiner salt to be reused and would significantly reduce the waste volume of the fission products.

Researchers are considering two basic approaches for the selective removal of fission products from the electrorefiner salt: 1) salt-zeolite ion exchange and 2) selective precipitation of fission products. Experiments are currently being performed involving both methods and the data obtained will be used to perform a complete assessment and comparison of the two separation options. This will allow determination of which technology or mix of technologies is most effective at fission product separation from electrorefiner salts. The final research objective will involve both laboratories performing an integrated experimental demonstration using electrorefiner salt containing fission products. From these results, systems analysis will determine if one or multiple technologies should be implemented into the reference electrorefining flowsheet.

Research Progress

Salt-Zeolite Ion Exchange. A major effort for the past year was directed at expanding the fission product-zeolite equilibrium studies. Ternary-salt mixtures ($M^{+x}Cl_x$–LiCl–KCl) investigated in 2007 were barium (Ba), cerium (Ce), lanthanum (La), praseodymium (Pr), and yttrium (Y) with results from these studies added to the existing

Cation	Published Values		Constrained Li and K		Unconstrained Li and K (best current fit)	
	K^{IX}	K^{OCC}	K^{IX}	K^{OCC}	K^{IX}	K^{OCC}
Cs^+	0.057	4.9	0.057	4.9	0.15	2.46
Na^+	0.32	0.92	0.32	0.92	0.29	1.15
Li^+	1	1	1	1	1	1
K^+	0.26	0.62	0.26	0.62	0.31	0.53
Rb^+	0.024	2.3	0.024	2.3	0.053	1.97
Sr^{2+}	1.78	0	1.78	0	1.96	0
U^{3+}	0.89	0.04	0.89	0.04	0.69	0.004
Nd^{3+}	N/A	N/A	0.5827	0.0009	0.76	0
Ce^{3+}	N/A	N/A	0.8175	0	0.92	0
Ba^{2+}	N/A	N/A	3.344	0	3.64	0

Table 1. The current equilibrium constant model parameters for fission product ion exchange (IX) and fission product occlusion (OCC) into the zeolite structure.

fission product chloride database. Determination of the total fission product equivalent loading that is both ion exchange and occlusion of fission product chlorides into the zeolite provides information on the salt-zeolite exchange mechanism and is used to validate the current equilibrium model. The current best-fit fission product equilibrium model parameters for ion exchange and occlusion are shown in Table 1. The plotted comparison of experimental data to model prediction is shown in Figure 2.

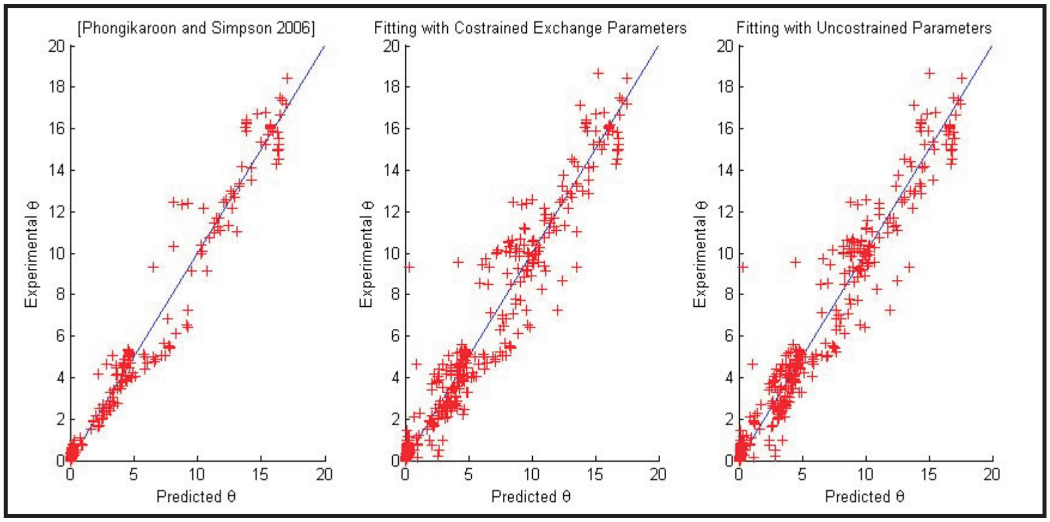


Figure 1. Scatter plots comparing experimentally determined total fission product equivalence loading (Θ) in zeolite to the predicted value of the exchange model for parameters in Table 1.

T(°C) RECl$_3$	400	450	500	550	650
EuCl$_3$	0.440	0.650	0.755	0.888	> 0.999
NdCl$_3$	0.200	0.649	0.709	0.818	> 0.999
CeCl$_3$	0.984	> 0.999	> 0.999	>0.999	> 0.999
PrCl$_3$	0.365	0.524	0.717	0.823	> 0.999

Table 2. Conversion ratio of some rare-earth chlorides to precipitates in LiCl–KCl molten salt by using oxygen sparging method.

Fission Product Precipitation. Primary activities conducted in 2007 involved rare-earth (RE) precipitation in LiCl–KCl eutectic salts by oxygen sparging and salt removal from precipitates by distillation. Oxygen sparging experiments were conducted in molten, eutectic salt with added chlorides of europium (Eu), neodymium (Nd), Ce, and Pr representing rare-earth fission products. The experiments were conducted at different molten-salt temperatures and oxygen bubble sizes and rates. Precipitated products were identified as rare-earth oxychlorides (REOCl) or as oxides. Table 2 shows the conversion ratio of the RE chlorides to the precipitate at different molten salt temperatures.

Researchers concluded that a small hole (about 1 mm) sparger with slow bubble formation and rise resulted in more favorable hydrodynamic characteristics and better precipitate product formation. Recovery of the salt after precipitate formation was performed by distillation with greatest efficiencies (distillation rates) at reduced pressure. This also allowed a lower distillation temperature, as shown in Figure 2.

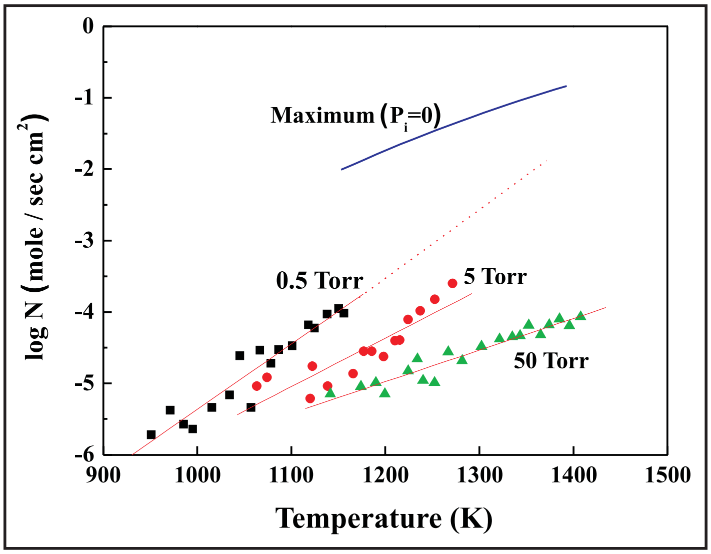


Figure 2. Salt distillation rates under different vacuums.

Planned Activities

Salt-Zeolite Ion Exchange. Activities for 2008 will involve kinetic studies of the fission product-zeolite ion exchange/occlusion process. This information will allow process modeling and equipment scale-up determination. Equilibrium studies of multi-cation fission product eutectic salt will continue to validate the salt-zeolite ion exchange predictive model.

Fission Product Precipitation. On the basis of results obtained in FY 2007, activities for 2008 will include experiments to determine optimum conditions such as sparging time and rate and molten salt temperature. In addition, rare-earth precipitates will be characterized for particle size, chemical form, and settling time. These experiments will be performed in 3 kg-salt/batch scale apparatus.

INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE

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

PIs (Korea): Eung-Ho Kim and Jong-Hyeon Lee, Korea Atomic Energy Research Institute (KAERI)

Collaborator: University of Illinois at Chicago

Project Number: 2006-003-K

Program Area: Generation IV

Project Start Date: January 2007

Project End Date: September 2010

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 are among 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 reduce oxides that are components of the coating material, resulting in degradation, and 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.

INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE

VHTR Environmental and Irradiation Effects on High-Temperature Materials

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

PI (Korea): Woo-Seog Ryu, Korea Atomic Energy Research Institute (KAERI)

Collaborators: None

Project Number: 2006-006-K

Program Area: Generation IV

Project Start Date: October 2006

Project End Date: September 2009

Research Objectives

This project will select and qualify high-temperature materials for the next generation of reactors. The collaboration will include work to analyze irradiation and environmental effects and to develop a materials handbook containing the properties of high-temperature metallic materials.

Specifically, researchers will study the effects of helium environments on the mechanical properties of high-temperature metallic alloys proposed for use in the very high-temperature gas reactor (VHTR).The helium primary coolant in an operating VHTR is expected to be contaminated by small amounts of gaseous impurities from a variety of sources. Corrosion of structural alloys by these impurities at elevated temperatures can be significant. Researchers will also evaluate the effects of irradiation on these alloys using the Republic of Korea's High-Flux Advanced Neutron Application Reactor (HANARO) and the Irradiated Materials Evaluation Facility (IMEF). Reactor operating conditions are very challenging for these materials and they require qualification against the effects of irradiation. Materials testing of key components, such as the reactor pressure vessel, will provide the necessary design data and develop and confirm available margins. Lastly, researchers will develop the *Generation IV Materials Handbook* containing a database of high-temperature materials.

Research Progress

No progress was reported for FY 2007.

INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE

Experimental Validation of Stratified Flow Phenomena, Graphite Oxidation, and Mitigation Strategies of Air Ingress Accidents

PI (U.S.): Chang H. Oh, Idaho National Laboratory (INL)

PI (ROK): Hee Cheon No, Korea Advanced Institute of Science and Technology (KAIST)

Collaborators: None

Project Number: 2007-001-K

Program Area: Generation IV

Start Date: November 2007

End Date: November 2010

Project Abstract

Previous studies have identified an air ingress event following reactor depressurization as an important accident scenario for the very high temperature reactor (VHTR). Consequently, the development of validated advanced air ingress-related models is a high priority for the Next Generation Nuclear Plant (NGNP), which will incorporate VHTR technology.

The major goal of this project is to upgrade the GAMMA code to reliably predict the consequences of air-ingress in the NGNP. To accomplish that, researchers will conduct experiments to understand important phenomena that occur during air-ingress accidents. These experiments will measure the following: 1) the effects of air ingress induced by a density-driven stratified flow into the reactor core, 2) the internal pore area density of nuclear grade graphite—which is an important parameter for determining the oxidation rate, 3) the oxidation and density variation in terms of graphite oxidation in the bottom reflector, and 4) the effects of the graphite oxidation on the structural integrity of the bottom reflector.

Accurately estimating safety margins in the VHTR design requires precise experimental data and tools, including accurate multi-dimensional thermal-hydraulic and neutronic models, a graphite oxidation model, and a fracture model. Researchers will develop a coupled neutronics and thermal-hydraulic capability in the GAMMA code, which will allow for high-pressure conduction cooldown transient simulations as well as accurate determination of initial and transient power distributions. Both of these are essential to accurately calculate safety margins.

Researchers will use the upgraded code to simulate air ingress accidents in order to evaluate various methods for mitigating the effects of air ingress. Air ingress leads to oxidation of the in-core graphite structure and fuel, which accelerates heat-up of the bottom reflector and the reactor core and eventually causes the release of fission products. The potential collapse of the bottom reflector due to graphite oxidation and the release of carbon monoxide would lead to serious safety problems.

INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE

Development of an Advanced Voloxidation Process for Treatment of Spent Fuel

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

PI (ROK): Jang Jin Park, Korea Atomic Energy Research Institute (KAERI)

Collaborators: None

Project Number: 2007-002-K

Program Area: AFC R&D

Start Date: October 2007

End Date: September 2010

Project Abstract

This project will address the head-end treatment of spent nuclear fuel undergoing pyroprocessing. The objective is to develop an advanced voloxidation process that provides a means to recover fuel from the cladding, prepares fuel for subsequent processing, simplifies downstream processing by removing volatile fission products prior to the pyroprocess, and safely traps the volatile fission products. Advanced voloxidation will also address fuel types other than light water reactor fuels, such as fast reactor oxide fuels.

The basis of the voloxidation process is to oxidize spent oxide fuel at low temperatures (about 500°C) in order to remove tritium and increase the dissolution rate during aqueous processing. Advanced voloxidation not only performs these functions but also removes and collects specific fission products such as krypton, xenon, cesium, rubidium, iodine, technetium, molybdenum, and ruthenium.

This new advanced voloxidation project encompasses three primary tasks related to pyroprocessing: 1) evaluation of the effects on downstream processes including feed requirements, equipment durability, waste generation, process efficiency, facility layout, economics, and proliferation concerns; 2) optimization of the capabilities of the off-gas trapping system for fission products; and 3) development of specific operational parameters for advanced voloxidation, such as temperature, hold times, and mechanical agitation.

INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE

Performance Evaluation of TRU-Bearing Metal Fuel for Sodium Fast Reactors to Achieve High Burnup Goal

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

PI (ROK): Byoung-Oon Lee, Korea Atomic Energy Research Institute (KAERI)

Collaborators: None

Project Number: 2007-003-K

Program Area: AFCR&D

Start Date: November 2007

End Date: November 2010

Project Abstract

This project will evaluate the high burnup performance of transuranic (TRU)-bearing metal fuel for the Sodium-Cooled Fast Reactor (SFR). A potential factor limiting alloy fuel burnup is fuel/cladding chemical interaction (FCCI), which causes cladding wall thinning and increased stress. One method proposed to solve this issue is the use of a barrier on the cladding's inner surface. Researchers will evaluate two types of barriers: 1) a metal liner and 2) an inert compound coating the inner surface.

Specifically, this project will involve the fabrication and performance testing of the barrier and cladding materials against the metal fuel. Current fuel performance codes do not address the performance of fuels with high TRU and rare earth element content. Therefore, the project also includes the development of models and benchmarking of the fuel performance codes to enable a comprehensive analysis of the metallic fuel performance.

The scope of the proposed work consists of three major tasks: 1) benchmarking of fuel performance codes, 2) fabrication of barrier cladding, and 3) diffusion testing of metal fuel against barrier cladding. The proposed work forms the basis for establishing key technologies that will enable the development of higher burnup TRU-bearing fuel and contribute to a more complete understanding of the behavior of metal fuel for very high burnup applications.

INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE

Development and Characterization of New High-Level Waste Forms for Achieving Waste Minimization from Pyroprocessing

PI (U.S.): Terry Battisti, Idaho National Laboratory (INL)

PI (ROK): Yung-Zun Cho, Korea Atomic Energy Research Institute (KAERI)

Collaborators: None

Project Number: 2007-004-K

Program Area: AFC R&D

Start Date: November 2007

End Date: November 2010

Project Abstract

The objective of this project is to develop new high-level waste (HLW) forms and fabrication processes to dispose of active metal fission products that are chemically separated from electrorefiner salts in the pyroprocessing-based fuel cycle. The current disposal technology involves discarding the fission product-loaded salts in a glass-bonded sodalite ceramic waste form. By selectively removing fission products from the molten salt, researchers would minimize the amount of HLW generated. Novel methods, such as chemical precipitation, are currently being developed to achieve this selective separation of fission products, but no investigation has yet to be performed regarding suitable waste forms for the separated fission products. The precipitates formed would likely consist of oxides, sulfates, and/or phosphates that are incompatible with the sodalite ceramic waste form. Thus, a completely novel approach to waste form synthesis is required.

During the first year, researchers will assess the characteristics of fission product waste streams and screen candidate solid matrices for waste forms. In preparation for producing transuranic (TRU)-contaminated waste forms, they will procure or build small-scale waste processing equipment, test its operation, and then install the equipment in a hot cell. In the second year, the team will conduct more rigorous testing of promising waste forms using non-radioactive surrogates and will test waste form fabrication techniques in a hot cell using TRU-contaminated radioactive salt. In the final year, researchers will characterize the TRU-contaminated waste forms and optimize the waste form fabrication process. The team will produce the final set of TRU-contaminated waste forms based on the optimized methods they have developed.

INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE

Development of Technology for Viable International Deployment of Small Sodium-Cooled Fast Reactors

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

PI (ROK): Kune Y. Suh, Seoul National University (SNU)

Collaborators: None

Project Number: 2007-005-K

Program Area: Generation IV

Start Date: November 2007

End Date: November 2010

Project Abstract

Small, modular, transportable sodium-cooled fast reactors (SFRs) can play a major role in helping to meet the significant projected growth in worldwide nuclear capacity, while reducing the risk of proliferation. Particular applications for which such reactors are well suited include powering industrial enterprises such as mining in remote regions and providing electricity to consumers in developing nations that do not have major grid connections.

This project will investigate and enhance the viability of deploying small modular transportable SFRs at remote sites. Researchers will identify approaches that minimize the construction time, labor, and costs and will develop component modularization techniques, transportation, and site installation sequences that reduce onsite assembly time and labor.

As these small modular reactors lack economies of scale, they must take advantage of innovations and simplifications that reduce capital and generation costs. A key contributor to cost reduction is through manufacturing and assembly processes that streamline nuclear plant construction. One approach is to pre-assemble components into modules in a factory setting. Applying this approach to fast reactor plants requires taking full advantage of factory mass production techniques and incorporating innovations that increase plant efficiency in order to reduce costs. Modules must be readily transportable to remote sites and based on a standardized design suitable for a worldwide market that meets regulatory requirements in various nations.

It is initially planned to utilize the 50 MWe (125 MWt) Small Modular Fast Reactor (SMFR) concept developed by an international team including ANL as the reference design. This reactor incorporates a supercritical carbon dioxide Brayton cycle power converter to further reduce costs through a simpler and less expensive balance-of-plant design and higher plant efficiency. Researchers will use time dependent, three-dimensional computer aided design models to simulate component fabrication, modularization, transportation, and installation sequences that are suited for deployment at remote sites and to determine optimal sequences for reducing construction time and cost.

Development of Computational Models for Pyrochemical Electrorefiners of Nuclear Waste Transmutation Systems

PI (U.S.): Michael Simpson, Idaho National Laboratory (INL)

PI (ROK): Kwang-Rag Kim, Korea Atomic Energy Research Institute (KAERI)

Collaborators: Seoul National University (SNU), University of Idaho

Project Number: 2007-006-K

Program Area: AFC R&D

Start Date: November 2007

End Date: November 2010

Project Abstract

Electrorefiners are central elements in pyroprocessing technology and are currently used to treat driver and blanket fuels from the Experimental Breeder Reactor-II (EBR-II) at INL. Despite successful test results, the multi-dimensional computational models essential for design and operational analysis of advanced processors can be greatly improved. The currently available electrorefiner model, General Purpose Electrochemical Simulator (GPEC), suffers from severe limitations in predictive capability due to its extensive reliance on empirical parameters.

The goal of this project is to develop two multi-dimensional computational models in order to improve the operation of electrorefiners currently used in pyroprocessing technology. These 2-D and 3-D mathematical models will be based on the fundamental physical and chemical properties of the electrorefiner processes. Researchers will use the models to validate compiled and evaluated experimental data to provide better information for developing advanced electrorefiners for actinide recovery.

In this project, each organization will separately develop their computational models, then share data and work jointly to validate the models against experimental data sets. In the first phase, researchers will plan and assess a common computational platform for the modeling work and identify spatial characterization requirements. The second phase will focus on developing a 3-D electro-fluid dynamic model and a 2-D electrorefiner model. In the last phase, researchers will validate and benchmark the two models with compiled experimental data sets.

Sodium-Cooled Fast Reactor Structural Design for High Temperatures and Long Core Lifetimes/Refueling Intervals

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

PI (ROK): Gyeong-Hoi Koo, Korea Atomic Energy Research Institute (KAERI)

Collaborators: None

Project Number: 2007-007-K

Program Area: Generation IV

Start Date: November 2007

End Date: November 2010

Project Abstract

The objective of this project is to develop state-of-the-art analysis capabilities for the high-temperature structural design of sodium-cooled fast reactors (SFRs). The research will focus on SFRs with higher core outlet temperatures than traditional SFRs (550°C or greater). These are desired attributes of the small, modular fast reactors intended for international deployment that will help meet the significant projected growth in nuclear capacity while reducing the risks of proliferation. However, at higher temperatures and longer service lifetimes, the reactor structures may undergo creep and creep-fatigue and accumulate inelastic strain as the reactor goes through operational cycles and transients. Structural stability of the reactor vessel and in-vessel components must also be assured during seismic events.

This project will investigate the design of core, vessel, and in-vessel structures using the SIE ASME-NH computer code developed at KAERI, that implements the rules of ASME Boiler and Pressure Vessel Code Section III, Subsection-NH and incorporates deterministic analyses of material inelastic behavior. Researchers will also carry out seismic and buckling evaluations at high temperatures.

The approach represents an advanced development in engineering structural design technology that enables researchers to investigate ways of reducing the conservatism inherent in the elevated temperature design rules. It also facilitates comparisons of the simplified elastic analysis and the detailed inelastic analysis methods. Researchers will specify thermal loadings for operational cycles and transients expected during the plant lifetime. It is initially planned to use the 50 MWe (125 MWt) Small Modular Fast Reactor (SMFR) concept developed by an international team including ANL as the reference SFR design. The capabilities the team develops will also be applicable to other high-temperature Generation IV reactors.

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