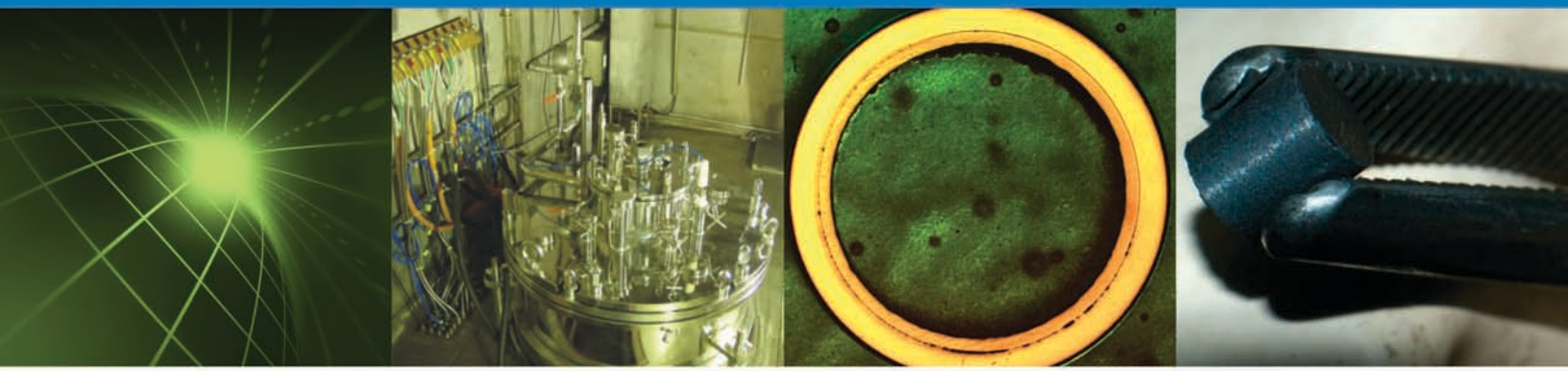
International Nuclear Energy Research Initiative



2009 Annual Report

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Foreword

The U.S. Department of Energy, Office of Nuclear Energy (DOE-NE), established the International Nuclear Energy Research Initiative (I-NERI) in fiscal year (FY) 2001 to conduct advanced nuclear energy systems research and development (R&D) in collaboration with international partners. This annual report provides an update on the FY 2009 accomplishments under the I-NERI collaborations.

I-NERI supports bilateral collaboration on scientific and engineering R&D linked to DOE-NE research programs such as the Generation IV Nuclear Energy Systems Initiative and the Fuel Cycle Research and Development Program. I-NERI is designed to foster international partnerships to address key issues affecting the future global use of nuclear energy. Through the I-NERI collaboration, DOE can effectively leverage its economic resources, more readily expand the knowledge base of nuclear science and engineering, and establish valuable intellectual relationships with researchers from other countries. Forging these partnerships enhances the United States' participation in the global nuclear community and helps build an international consensus on critically important issues such as increasing the safety and expanding the benefits of nuclear power and helping design proliferation resistance into advanced nuclear systems.

Current I-NERI collaborators include Canada, the European Union, France, Japan, the Republic of Korea, and the Republic of South Africa. In FY 2009, DOE initiated two additional projects with the Republic of Korea and completed eight projects involving collaborations with Euratom, France, Japan, and the Republic of Korea. This *I-NERI 2009 Annual Report* provides a description of the new projects, along with a comprehensive summary of the progress of each collaborative research project initiated since FY 2006.

Dr. Warren F. "Pete" Miller Jr.
Assistant Secretary, Office of Nuclear Energy
U.S. Department of Energy

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INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE

1.0 Overview and Program History

The International Nuclear Energy Research Initiative (I-NERI) is an international, research-oriented collaboration that supports advancement of nuclear science and technology in the United States and the world. I-NERI promotes bilateral 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 use 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 2009 Annual Report* serves to inform interested parties about the program's progress of current collaborative nuclear energy research projects. Following is an overview of each section:

- **Section 1** discusses background information on the events that led to the I-NERI program's creation.
- **Section 2** presents the countries and international organizations participating in current I-NERI collaborative agreements. This section also provides 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 3** provides a summary of recent programmatic accomplishments and a list of participating organizations.
- **Section 4** presents details of the R&D work scope for current I-NERI collaborative projects with Canada, the European Union (EU), France, Japan, and the Republic of Korea. For each participant, the report presents an index of projects and a summary of technical accomplishments achieved in fiscal year (FY) 2009.

History

DOE-NE created I-NERI in FY 2001 in response to recommendations provided 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 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 stated, "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."

The initial focus of the I-NERI program was on developing international collaborations, program planning, and project procurements. Since the inception of the program, 90 projects have been awarded, 60 of which have been completed. Both the number of awards and the consistency of project achievement demonstrate I-NERI's success in fostering international collaboration.

Table 1 presents a breakdown, by fiscal year, of the number of projects undertaken with each partnering country.

Table 1. Number of I-NERI projects awarded.

Collaborator	FY 01	FY 02	FY 03	FY 04	FY 05	FY 06	FY 07	FY 08	FY 09	Total
Brazil					1	1				**2**
Canada				7			2	3		**12**
EURATOM				8	2	5				**15**
France	4	1		11		3		2		**21**
Japan					1	1				**2**
OECD-NEA		1								**1**
Republic of Korea		6	5	6	4	4	7	3	2	**37**
Total	**4**	**8**	**5**	**32**	**8**	**14**	**9**	**8**	**2**	**90**

I-NERI is integrated into DOE-NE's principal research programs, including the Generation IV Nuclear Energy Systems Initiative (Gen IV), Fuel Cycle R&D Program (FCR&D), and Nuclear Hydrogen Initiative (NHI). Note that NHI became part of Gen IV after the time period of this report and is no longer a stand-alone program.

INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE

2.0 I-NERI Program Goals and Objectives

I-NERI's mission is to sponsor innovative scientific and engineering R&D in cooperation with international partnering countries. 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. 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 a nuclear science and engineering infrastructure to meet future technical challenges.

Through the I-NERI program, DOE-NE has coordinated wide-ranging scientific discussions among governments, industry, academia, and the worldwide research community regarding the development of Generation IV nuclear energy systems, advanced fuel cycles, and nuclear hydrogen technologies. Figure 1 graphically illustrates key features of the I-NERI program.

International Agreements

In order to initiate an international collaboration, a government-to-government agreement must first be in place. These agreements serve as the vehicles to conduct R&D under the various DOE programs. 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), predecessor to the Ministry of Education, Science and Technology (MEST)
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 (MST)
7) Agency of Natural Resources and Energy (ANRE) and the Ministry of Education, Culture, Sports, Science, and Technology (MEXT) of Japan
8) The Government of the Republic of South Africa

I-NERI bilateral agreements enable U.S. researchers to establish international R&D collaborations supporting the development of next-generation nuclear energy systems. DOE also supports multilateral R&D collaborations under the Generation IV International Forum (GIF) and the international Global Nuclear Energy Partnership. Please visit their websites for more information: *http://www.gen-4.org* and *http://gneppartnership.org*.

Funding

I-NERI is an important vehicle for enabling international R&D in nuclear technology on a leveraged, cost-shared basis. Each country in an I-NERI collaboration provides funding for its respective project participants. The United States funds I-NERI projects through its national laboratories, with the annual contribution 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 partnering country.

To date, I-NERI participants have committed a total R&D investment of $221.2 million: $118.5 million contributed by the United States and $102.7 million by international collaborators. International investment consists of:

- $2.3 million from Brazil
- $19.2 million from Canada
- $12.3 million from the European Union
- $30.2 million from France
- $2.7 million from Japan
- $36.0 million from the Republic of Korea

I-NERI projects typically last three years, with the U.S. portion funded annually by the DOE-NE R&D programs. Funding provided by the United States may be spent only by U.S. participants.

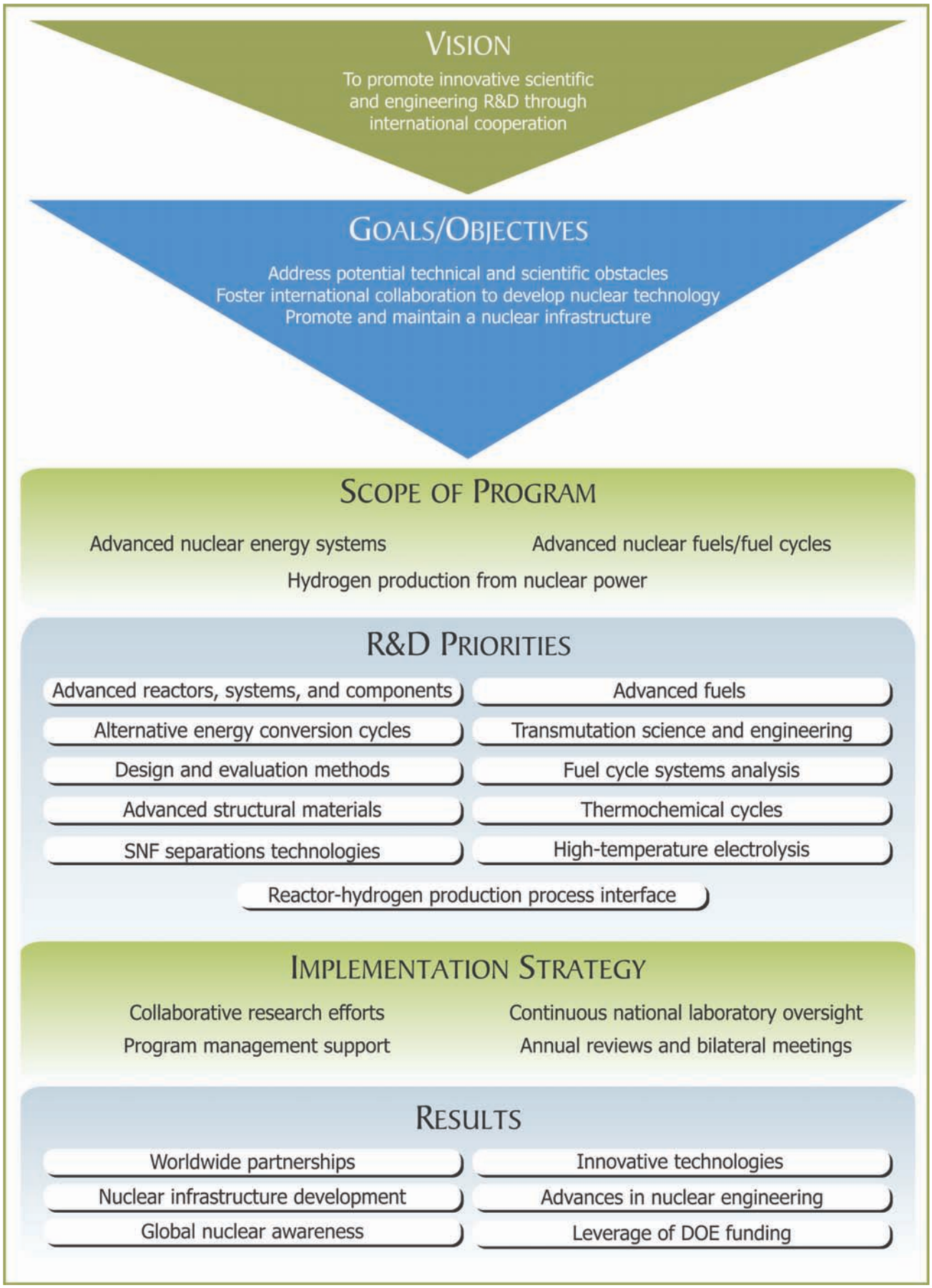


Figure 1. Graphical depiction of I-NERI program.

Work Scope

The work scope of all current I-NERI projects is directly linked to the scientific and engineering needs of the DOE-NE R&D programs. Project work scopes were jointly developed by the United States and the collaborating country based on conformance with the bilateral agreement and common R&D programmatic needs. Following is an overview of the individual work scopes for NE's three R&D programs.

Gen IV Nuclear Energy Systems Initiative. Gen IV 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: molten salt reactor, lead-cooled fast reactor (LFR), gas-cooled fast reactor (GFR), supercritical water-cooled reactor (SCWR), sodium-cooled fast reactor (SFR), and very high-temperature reactor (VHTR). Current U.S. research priorities are focused on the VHTR and SFR. The VHTR concept is being pursued as the Next Generation Nuclear Plant (NGNP), designed for economical and safe production of hydrogen and electricity with very low greenhouse gas emissions.

There are eight technology goals for the Gen IV program:

1) 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) 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) Increase the assurance that systems are a very unattractive and undesirable route for diversion or theft of weapons-usable materials.

4) Ensure that systems will excel in safety and reliability.

5) Design systems that have a very low likelihood and degree of reactor core damage.

6) Create reactor designs that minimize the need for offsite emergency response.

7) Ensure that systems have a clear life-cycle cost advantage over other energy sources.

8) Create systems that have a level of financial risk that is comparable to other energy projects.

Fuel Cycle Research and Development. The mission of FCR&D is to enable the safe, secure, economic, and sustainable expansion of nuclear energy by conducting research, development, and demonstration focused on nuclear fuel recycling and waste management to meet U.S. needs. In this way, nuclear energy can satisfy the growing energy needs of the United States while improving management of waste requiring geologic disposal. These technologies may also be of value to Gen IV.

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

Nuclear Hydrogen Initiative. The NHI program, which was integrated with Gen IV, was designed 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 with very low greenhouse gas emissions and without consuming fossil fuel resources.

Figure 2 shows the distribution of projects by each of the three major program areas since the program's inception. (Prior to 2004, all I-NERI projects were related strictly to Gen IV.)

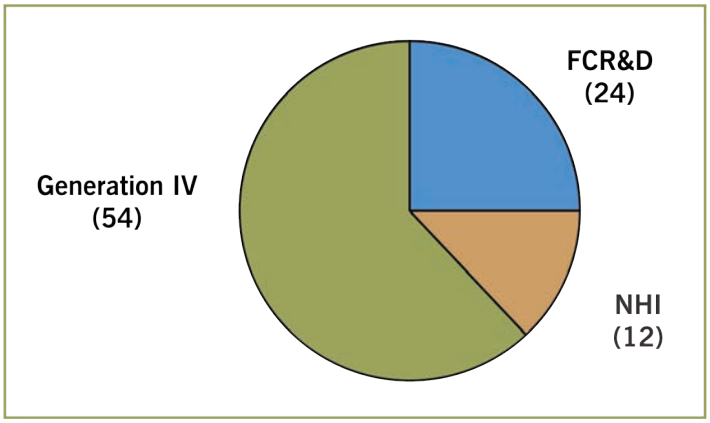


Figure 2. Project distribution by program area (FY 2001 to FY 2009).

INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE

3.0 I-NERI Program Accomplishments

Research Findings

I-NERI collaborative projects produce findings that would take the United States alone far more time and money to accomplish. The U.S. investment in I-NERI is matched nearly dollar for dollar. I-NERI has helped the U.S. role within the international community. Moreover, creating an international infrastructure brings multiple perspectives and priorities together to address shared obstacles. I-NERI collaborative efforts have contributed greatly to industry knowledge, furthering the ability to overcome technological challenges to nuclear energy's continued—and enhanced—role in meeting global energy requirements.

For example, Gen IV projects during FY 2009 focused on research associated with tristructural-isotropic (TRISO) fuel particles; crosscutting materials for treating spent nuclear fuel; the properties of and environmental effects on very high-temperature materials such as metallic alloys and ceramic composites; software and simulation models to calculate and assess by-pass flow measures, coupled neutronics, and thermo-fluid dynamics of VHTRs; and the potential of advanced power conversion technologies for use with advanced nuclear and fossil energy sources. Two recently completed Gen IV projects included experiments on TRISO particle layers in which researchers 1) successfully completed neutron irradiation of surrogate particle samples coated with zirconium carbide in different coating/heat treatment conditions and 2) measured a series of TRISO samples relevant to past and present fuel development programs spanning a broad range of anisotropies. In a third completed project, researchers selected and qualified high-temperature materials and developed a handbook of the mechanical properties of these metallic materials.

The thirteen active FCR&D projects during FY 2009 investigated multiple topics: one researched ways to improve fuel performance, thus increasing efficiency and reducing waste; two specifically focused on safety systems and safety validation; three researched transmutation and related fuels; one examined alloys for the SFR; and four examined fuel separations, fuel recycling, and new waste forms. All clearly met the agenda of finding new fuel resources and systems for proliferation-resistant nuclear power. The two FCR&D projects completed during this fiscal year developed core designs for sodium-cooled transuranic burner reactors and developed processes for selectively removing fission products from lithium chloride-potassium chloride eutectic salts used in the electrochemical processing of used nuclear fuel.

Two FY 2009 NHI research projects involved assessing methods for producing hydrogen with advanced reactors using 1) high-temperature solid-oxide electrolysis cells and 2) the sulfur-iodine (S-I) thermochemical cycle. During this fiscal year, researchers concluded the S-I experiment and determined that 1) the small scale of these experiments constrained the options for basic components; 2) the results from the hydrogen-iodide decomposition section operations showed that extractive distillation works, but it is complex and less energy-efficient than reactive distillation; and 3) the proposed multi-tube scaling approach on the SiC bayonet heat exchangers needs to be demonstrated in large scale. Two other current NHI projects involved developing the copper-chloride (Cu-Cl) cycle as part of the alternative thermochemical cycle program and developing a preliminary design for an integrated complex consisting of a nuclear reactor, electrolytic hydrogen plant, and bitumen upgrader.

International Growth

During FY 2009, the following activities were completed:

- Completion of eight I-NERI research projects that began in FY 2006 with four collaborating entities:
  - Euratom (2)
  - France (2)
  - Japan (1)
  - Republic of Korea (3)
- Initiation of two new collaborative research projects with the Republic of Korea.
- Annual project performance reviews and bilateral program planning meetings with the following partner countries:
  - Canada
  - Euratom
  - France
  - Republic of Korea

DOE plans the following international activities for FY 2010:

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

Program Participants

The map on the next page provides a complete listing of the I-NERI program participants and the locations of international collaborators.

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 3, a total of seven universities and colleges participated in I-NERI research projects during FY 2009. Approximately 58 students from these institutions worked on active I-NERI research projects during the year, distributed by degree level as shown in Figure 4.

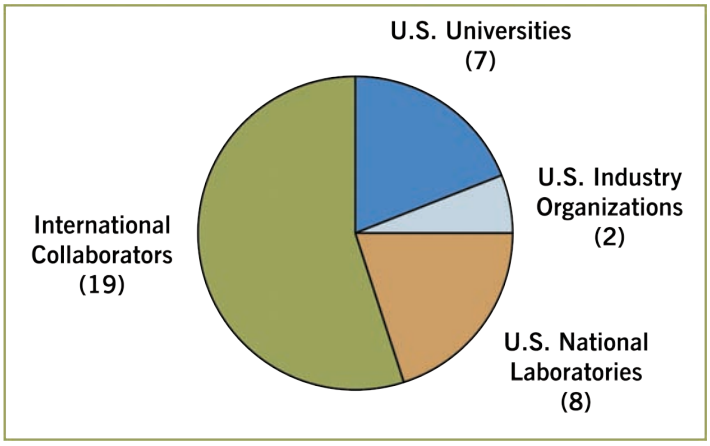


Figure 3. FY 2009 I-NERI participant profile.

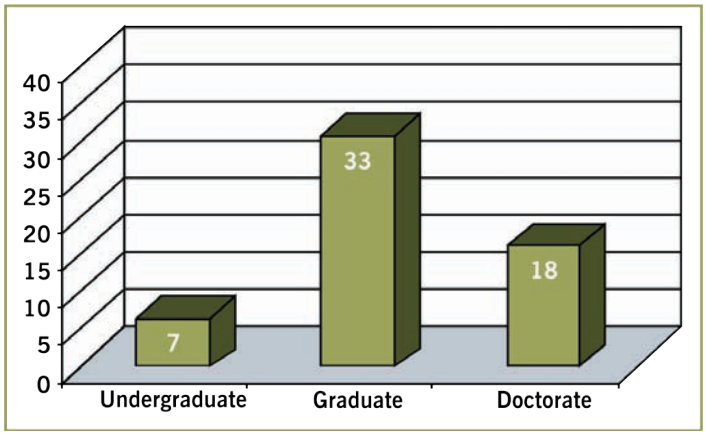


Figure 4. FY 2009 I-NERI student participation profile.

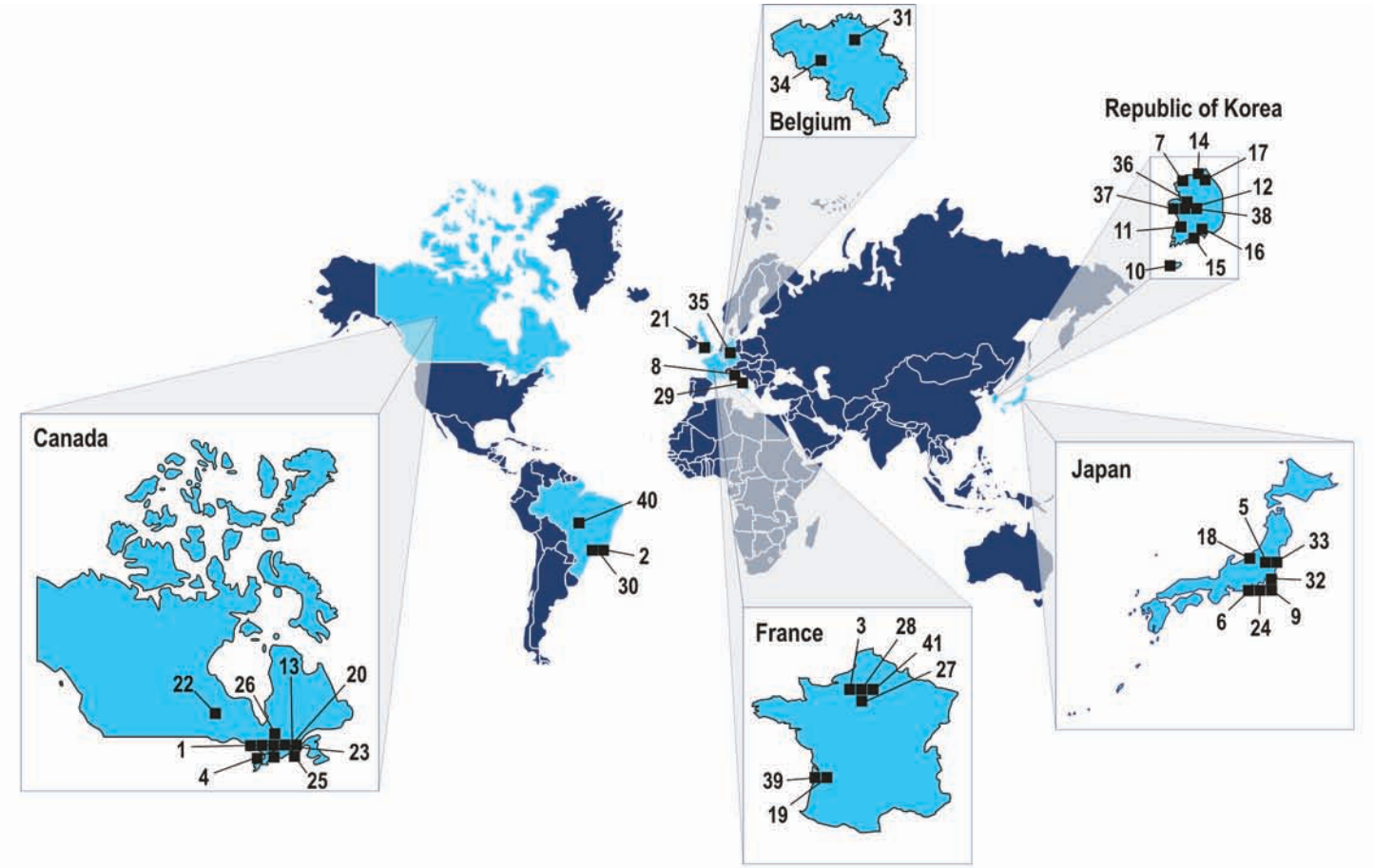
Completed I-NERI Projects

This year marked the completion of eight I-NERI research projects, all of which began in FY 2006:

- 2006-003-E – Development of Novel Transmutation Systems for Sustainable Nuclear Fuel Cycles
- 2006-004-E – Development of Oxide Fuels for Transmutation in Fast Reactors
- 2006-001-F – Sulfur-Iodine Integrated Laboratory-Scale Experiment, Sandia National Laboratories
- 2006-003-F – Comparison of Characterization Methods for Anisotropy and Microstructure of TRISO Particle Layers, Oak Ridge National Laboratory
- 2006-001-J – Assessment of Irradiation Performance of Zirconium Carbide (ZrC) TRISO Fuel Particles, Oak Ridge National Laboratory
- 2006-001-K – Core Design Studies for Sodium-Cooled TRU Burner Reactors, Argonne National Laboratory
- 2006-002-K – Separation of Fission Products from Molten LiCl-KCl Salt Used for Electrorefining of Metal Fuels, Idaho National Laboratory
- 2006-006-K – VHTR Environmental and Irradiation Effects on High-Temperature Materials, Oak Ridge National Laboratory

The following section provides detailed summaries of each of these projects.

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 United States and partnering international entities have resulted in significant scientific and technological enhancements in the global nuclear power arena. International collaborations have forged lasting ties that will continue promoting the strong infrastructure necessary to overcome future challenges to the expanded use of this vital source of clean and reliable power. The resulting technological and scientific advances will help ensure the United States remains competitive in both the global and domestic nuclear energy marketplaces.



U.S. Collaborators

National Laboratories

Argonne National Laboratory*
Brookhaven National Laboratory
Idaho National Laboratory*
Lawrence Livermore 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-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

1 Atomic Energy of Canada Limited (AECL)*
2 Eletronuclear
3 Framatome ANP
4 Gamma Engineering
5 Hitachi Works
6 Hitachi, LTD
7 Korea Hydro and Nuclear Power Company (KHNP)
8 Società Informazioni ed Esperienze Termoidrauliche (SIET)*
9 Toshiba Corporation

Universities

10 Cheju National University
11 Chosun University
12 Chungnam National University*
13 École Polytechnique de Montréal
14 Hanyang University
15 Korea Maritime University
16 Pusan National University
17 Seoul National University*
18 Tohoku University
19 Université Bordeaux*
20 Université de Sherbrooke
21 University of Manchester
22 University of Manitoba
23 University of Ontario Institute of Technology*
24 University of Tokyo

Governmental Organizations

25 CANMET Energy Technology Centre-Ottawa (CETC-O)*
26 Chalk River Laboratories (CRL)*
27 Commissariat à l'énergie atomique (CEA)*
28 Electricité de France (EDF)
29 Ente per le Nuove Tecnologie, l'Energia e l'Ambiente (ENEA)*
30 Instituto de Pesquisas Energéticas e Nucleares (IPEN)
31 Institute for Reference Materials and Measurements (IRMM)
32 Japan Atomic Energy Agency (JAEA)*
33 Japan Atomic Energy Research Institute (JAERI)
34 Joint Research Centre-Institute for Energy (JRC-IE)
35 Joint Research Centre-Institute for Transuranium Elements (JRC-ITU)*
36 Korea Advanced Institute of Science and Technology (KAIST)*
37 Korea Atomic Energy Research Institute (KAERI)*
38 Korea Electric Power Research Institute (KEPRI)
39 Laboratoire des Composites Thermostructuraux (LCTS)*
40 Ministério da Ciência e Tecnologia (MST)
41 Organisation for Economic Co-operation and Development-Nuclear Energy Agency (OECD/NEA)

* Indicates FY 2009 participant

Figure 5. I-NERI program participants.

INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE

4.0 Project Summaries and Abstracts

Sections 4.1 through 4.5 provide descriptions of the work scopes, listings of funded projects, and brief project status reports for I-NERI R&D projects undertaken with Canada, the European Union, France, Japan, and the Republic of Korea. The last project with Brazil concluded in FY 2008 and no new collaborations have been initiated since.

4.1 U.S./Canada Collaboration

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

Work Scope Areas

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

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

Project Summaries

A listing of the I-NERI U.S./Canada projects that are currently under way follows, along with summaries of the accomplishments achieved in FY 2009. No projects concluded during the past year and no new collaborations with Canada were initiated. A new collaboration agreement is currently being developed.

Directory of Project Summaries

2007-001-C Hydrogen Production Using High-Temperature Electrolysis 13

2007-002-C Thermochemical Hydrogen Production Process Analysis.....17

2008-001-C Upgrading of the Athabasca Oil Sands for the Production of Diesel and Gasoline 21

2008-002-C Evaluation and Demonstration of Advanced Power Conversion and Hydrogen Generation Technologies for Low-Emission Nuclear, Fossil, and Renewable Energy Sources.....23

2008-003-C Study of Burning Transuranic Elements in Heavy- and Light-Water Reactors.....27

INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE

Hydrogen Production Using High-Temperature Electrolysis

PI (U.S.): Stephen Herring, Idaho National Laboratory (INL)

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

Collaborators: None

Project Number: 2007-001-C

Program Area: NHI

Project Start Date: July 2007

Project End Date: March 2010

Research Objectives

This project has addressed research and scale-up issues associated with the use of energy-efficient, high-temperature solid-oxide electrolysis cells to produce hydrogen from steam. The project had 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. The HTE integrated laboratory scale (ILS) experiment operated in two phases: 1) from September through October 2007, with a single 240-cell module and 2) from September through October 2008, with three modules containing 720 cells. The ILS diluted the 850°C oxygen stream with air and cooled it to room temperature before it was exhausted outside the building. During the second phase, the incoming dilution air was heated to the required 800°C by the outgoing air-oxygen stream in heat recuperators. Researchers developed methods for safely handling the 2.8 Nm^3/hr of hot oxygen produced (4.0 kg O_2/hr), along with the 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—was critical to achieving high performance of the overall HTE process. In addition, recuperation of heat from the product gasses allowed a reduction in the reactor's required outlet temperature. In 2007 and 2008, researchers conducted a series of corrosion tests to analyze material performance in both the HTE cell and the balance-of-plant.

The following tasks comprise the main elements of this project:

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

Since this I-NERI project expires in March 2010, the following progress is a summary of the results of all four of these tasks.

Research Progress

Task 1. INL is working on a project to generate hydrogen by HTE. In this task, the team carried out a detailed economic analysis of nuclear hydrogen production, as outlined in Task 1 above. A summary of the results of that analysis is shown in the sensitivity diagram on the next page.

Tasks 2 and 3. In an HTE system, safety precautions need to be taken to handle high-temperature oxygen at ~830°C. During FY 2008, the INL published a report aimed at addressing Tasks 2 and 3, concerning the handling and cooling of oxygen in a HTE plant.

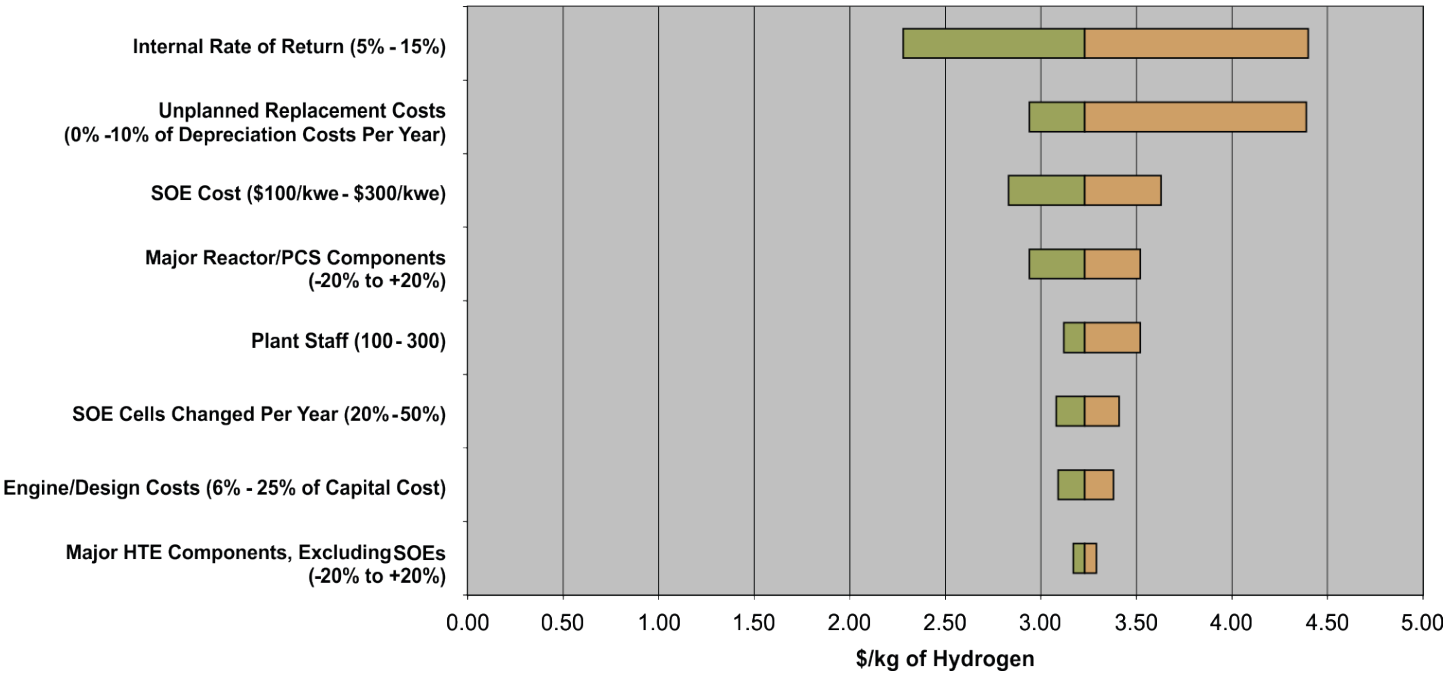


Figure 1. VHTR/HTE economic sensitivity analysis (for plant gate cost ~$3.23/kg hydrogen and 10 percent internal rate of return).

Though oxygen itself is not flammable, most engineering materials, including many gases and liquids, will burn in the presence of oxygen under some favorable physicochemical conditions. At present, an absolute set of rules does not exist that can cover all aspects of oxygen system design, material selection, and operating practices to avoid subtle hazards related to oxygen. Because most materials, including metals, will burn in an oxygen-enriched environment, hazards are always present when using oxygen. Most materials will ignite in an oxygen-enriched environment at a temperature lower than that in air; once ignited, combustion rates are greater in the oxygen-enriched environment. However, these hazards do not preclude the operations and systems involving oxygen. In fact, the incidence of oxygen system fires is reported to be low with a probability of about one in a million.

The team's report is a practical guide and tutorial for the safe operation and handling of gaseous oxygen in an HTE system. The intent was to provide safe, practical guidance that permits the accomplishment of experimental operations at INL, while being restrictive enough to prevent personnel endangerment and to provide reasonable facility protection. Adequate guidelines are provided to govern various aspects of oxygen handling associated with an HTE system to generate hydrogen. The intent here is to present acceptable oxygen standards and practices for minimum safety requirements. A summary of operational hazards, along with oxygen safety and emergency procedures, is provided.

When selecting a material for use with oxygen, the circumstances that cause oxygen to react with the material need to be understood. When the ignition energy input exceeds the configuration-dependent threshold, ignition and combustion may occur. Therefore, safe use of oxygen requires the control of potential ignition energy mechanisms within oxygen systems by judiciously selecting ignition-resistant materials. Ignition mechanisms that can result in pipeline failures include the following:

- Particle impact ignition caused by impingement of metallic or non-metallic materials with the pipeline's metal components.
- Adiabatic compression, acoustic resonance, and flow friction, which create temperature increase.
- Ignition initiated by the combustion of organic materials or contaminants entrained in the oxygen flow.
- Friction caused by rubbing, as in a valve between adjacent moving and stationary parts.
- Electric arcing between metallic components due to static electricity or lightning, which generates enough energy to ignite metallic or non-metallic materials.

When the ignition mechanism has started, the combustion can propagate through the kindling chain. Once ignited, the combustible material or component generates heat, which can, depending on many factors, ignite the bulk

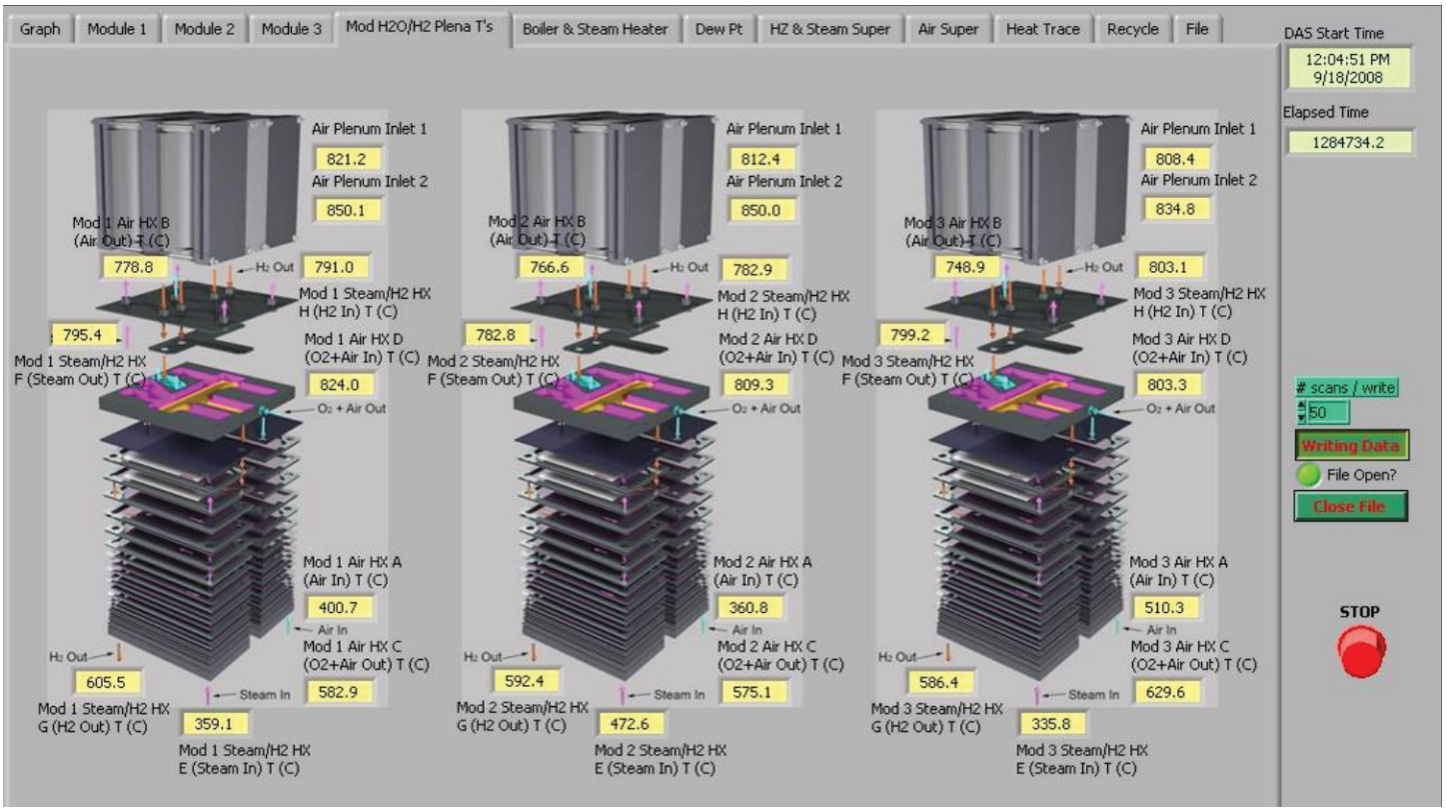


Figure 2. Operation of the heat recuperators during the ILS experiment, 2008.

material of the pressure envelope. The rate and extent of the propagation of the fire along the pressure envelope will depend on the material's thickness and flammability. The use of exempt materials will limit the combustion's propagation by interrupting the kindling chain. The rate and extent of the fire's propagation is also influenced by oxygen parameters such as pressure, purity, temperature, and the total oxygen inventory available to support combustion. For pressures below 30 psig (0.21 MPa), experimental data show that the combustion rates of potential materials used in oxygen pipeline components such as carbon steel are very low and decrease with decreasing pressure. This effect has contributed to the excellent service experience demonstrated by properly designed carbon steel components in selected very low-pressure oxygen applications. Many causes of fire can be avoided and their consequences reduced by good design practices. Other causes of fire may be due to unsuitable maintenance and operating practices, such as:

- Overheating due either to a process failure or to an oxygen leakage from the system resulting in an external fire adjacent to the pipeline.
- Accidental mixing with fuel, due to either a process failure or contaminant introduction during maintenance work.

Task 4. In support of Task 4, INL has performed tests of various metals in either steam-hydrogen or air-oxygen mixtures at temperatures up to 925°C and durations to 500 hours. Post-test examination has determined the rates of oxide growth and whether the oxide film was adherent to the base metal. In addition, the materials used in the ILS experiment during early FY 2009 are available for examination. The ILS included heat recuperators in which either a steam-hydrogen mixture or an air-oxygen mixture was cooled from ~820°C to ~450°C for the test duration of 1,080 hours. The configuration of the HTE ILS modules and their heat recuperators is shown in Figure 2, which is a screen image of the LabView experimental control software during the September-through-October 2008 ILS test.

Planned Activities

This project ended March 2010.

INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE

Thermochemical Hydrogen Production Process Analysis

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

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

Collaborators: McMaster University, Pennsylvania State University, Tulane University, University of Guelph, University of Ontario Institute of Technology (UOIT), University of South Carolina, University of Toronto, University of Waterloo, University of Western Ontario

Project Number: 2007-002-C

Program Area: NHI

Project Start Date: September 2008

Project End Date: FY 2009

Research Objectives

The development of the copper-chloride (Cu-Cl) cycle is ongoing as part of the NHI alternative thermochemical cycle program. The Cu-Cl cycle is a promising thermochemical cycle for heat sources that provide heat from 550°C to 600°C. The primary objective at AECL was to develop the electrolyzer for the electrochemical reaction. The effort at ANL has focused on meeting two objectives: 1) optimizing the yields in the hydrolysis reaction and 2) determining the efficiency and cost of hydrogen production. The objectives of the universities were to support the primary activities with special reference to advanced electrochemical technologies in the United States and to the development of engineering-scale equipment. Other related work has been ongoing, such as identifying corrosion-resistant materials for construction and analyzing the reliability and stability systems for distributed control for nuclear-based hydrogen generation.

Research Progress

Over this past fiscal year, the team at AECL worked on the version of the Cu-Cl cycle, shown in Table 1, where simplified reactions are used to describe the chemistry (i.e., water as an aqueous solution is ignored). The reactions are considered stoichiometric, and the hydrolysis and decomposition of Cu_2OCl_2 are thermal reactions.

One of the challenges associated with the electrolyzer's development is determining the best method to minimize, or preferably eliminate, copper crossover. AECL's approach has been to change the design of their cell and to replace the Nafion membrane to a cation exchange membrane with a coating selective to monovalent ions relative to divalent ions. The result is shown in Figure 1. The curve near the top represents the cell voltage as a function of time when the catholyte is recycled. Note that the cell voltage is fairly stable for about 3 hours but increases after that. The curve near the bottom of the figure shows that the cell voltage is stable for up to 19 hours when there is no recycle of the catholyte, suggesting that little or no copper crossover occurs.

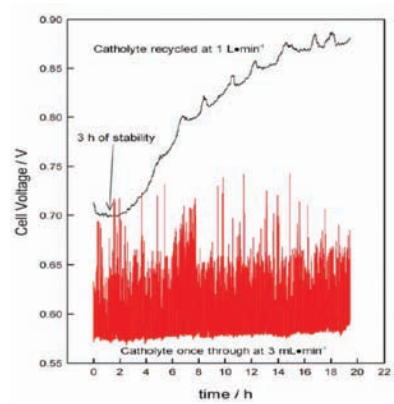


Figure 1. Cell voltage vs. time when the catholyte is recycled (top) and when the catholyte is not recycled (bottom).

Table 1. Reactions in the Cu-Cl cycle.

	Reaction	Temperature (°C)
(1) Hydrolysis	$2CuCl_2(s) + H_2O(g) \rightarrow Cu_2OCl_2(s) + 2HCl(g)$	350–400
(2) Decomposition	$Cu_2OCl_2(s) \rightarrow \frac{1}{2} O_2(g) + 2CuCl(s)$	400–530
(3) Electrolysis	$2CuCl(s) + 2HCl(aq) \rightarrow 2CuCl_2(s) + \frac{1}{2} H_2(g)$	~100

Table 2. The Cu-Cl cycle studied at UOIT.

Step	Reaction	Temp. Range (°C)	Press. (kPa)	Feed/Output (note: Q- *thermal energy*, V-*electrical energy*)	
1	$2CuCl_2(s)+H_2O(g) \rightarrow CuO*CuCl_2(s)+2HCl(g)$	400	101.325	Feed: Output:	Powder/granular $CuCl_2(s)+H_2O+Q$ Powder/granular $CuO*CuCl_2(s)+2HCl(g)$
2	$CuO*CuCl_2(s) \rightarrow 2CuCl(l)+\frac{1}{2}O_2(g)$	500	101.325	Feed: Output:	Powder/granular $CuO*CuCl_2(s)+Q$ Molten CuCl salt + O_2
3	$2CuCl(s)+2CuCl(aq) \rightarrow CuCl_2(aq)+Cu(s)$	Ambient (electrolysis)	101.325	Feed: Output:	Powder/granular CuCl and HCl + V Electrolytic Cu and slurry containing $HCl+CuCl_2$
4	$CuCl_2(aq) \rightarrow CuCl_2(s)$	>100	101.325	Feed: Output:	Slurry containing HCl+CuCl2+Q Powder/granular $CuCl_2+H_2O$/HCl vapors
5	$2Cu(s)+2HCl(g) \rightarrow CuCl(l)+H2(g)$	430-475	101.325	Feed: Output:	Electrolytic Cu + dry HCl + Q H_2 + CuCl(l) salt

UOIT researchers also contributed to the electrolyzer's development. They investigated ceramic-coated electrodes to enhance the transport of anionic Cu(I) (i.e., $[CuCl_2]^-$) and worked on developing new polymer exchange membranes (PEM) to minimize copper crossover. The extent of crossover was measured for commercially available PEMs and newly synthesized SPEEK (sulfonated polyetheretherketone). In addition, the team investigated methods, such as composites or coated membranes and/or the addition of pore fillers, to restrict the transport of larger ions such as Cu(I) and Cu(II) compared to H^+ as a means to minimize or eliminate copper crossover.

Other work at UOIT was concerned with a second version of the Cu-Cl cycle, described in Table 2. This version differs from the cycle described in Table 1 in the electrolysis reaction and the H_2 production reaction. One reason for studying the cycle in Table 2 is that it has a larger potential efficiency. The production of hydrogen by the reaction between HCl and Cu(0) is exothermic, and the heat released can be recovered and used elsewhere in the cycle. In addition, when the cycle is coupled with a CANDU reactor, a large amount of waste heat is available, and it can be used to dry the $CuCl_2$ solution from the spent anolyte.

Ideas for increasing the cycle's efficiency were also examined. For example, researchers investigated the use of cyclohexane for the bottoming cycle and biphenyl for the topping cycle. The coefficients of performance varied with temperature. Researchers also studied use of organic- and titanium tetrahalide-based working fluids for high-temperature heat pumps. They compiled thermophysical properties for each, including autoignition temperature, flash point, critical temperature, pressure, volume, compressibility, and accentric factor.

The kinetics and heat transfer of the hydrogen production reaction were calculated using the shrinking core model. The production of H_2 as a function of temperature and time were calculated. Also reported was the shrinkage of the particle as a function of time. However, this analysis did not include the effect of particle shape, which was an important parameter. Yields were near 100 percent when using very small dendritic particles (~3 microns) but <80 percent for larger dendritic and spherical particles.

Work on the fabrication of advanced materials during this time period focused on identifying a substrate and coating for the reactor used to solidify the molten CuCl. It has been assumed that the CuCl is molten and that the atmosphere is dry HCl. Inconel 625 and SS AL6XN were selected as possible substrate materials. The latter has outstanding resistance to chloride pitting and crevice corrosion and is resistant to acidic, oxidizing chloride (aqueous) solutions in lower temperature regions.

At UOIT, the scale is large, on the order of hundreds of grams. At ANL, experiments have been small in scale, where sample sizes have been about 0.3 g. The goal of university work is to run an integrated large laboratory-scale demonstration of the Cu-Cl cycle. Equipment was designed and built, and the various reactors are being cold-tested, where appropriate. Along with designing and fabricating the various reactors, researchers have been identifying the engineering challenges associated with each of these reactors and possible solutions.

Over this past year, ANL focused its research efforts on the thermal reactions on the hydrolysis and oxychloride decomposition reactions and the development of a process flowsheet with a mass and energy balance. This flowsheet was then used to estimate the cost of hydrogen production.

The Hydrolysis Reaction. The design that appears the most promising is a spray reactor. Several different versions of a laboratory-scale spray reactor have now been tested. The first design is shown in Figure 2. It uses a "pneumatic" nebulizer, in which the solution of $CuCl_2$ is injected through a capillary tube and is then atomized at its tip by an inert gas flowing through a concentric outer tube. The mist of $CuCl_2$ forms as a cone passes into the reactor heated by an external furnace. Superheated steam with an argon carrier gas was injected either co-currently or counter-currently to the $CuCl_2$ solution. After optimizing the operating variables, products that contained high yields of Cu_2OCl_2 were obtained only when the solution was injected counter-currently to the superheated steam. An X-ray diffraction (XRD) pattern of a typical product is shown in Figure 3.

Because the nebulizer was difficult to use and because it required a carrier gas for atomization, the nebulizer was replaced with an ultrasonic nozzle. After optimization of the experimental variables, very good products were obtained when the reactor was operated co-currently. A typical XRD pattern is shown in Figure 4. No lines characteristic of $CuCl_2$ or $CuCl_2{\cdot}2H_2O$ were visible. The line characteristic of CuCl was present.

The amount of CuCl was 3–5 wt% in most of the products obtained with the ultrasonic nozzle. The amount of Cu_2OCl_2 was assumed to be 95–97 wt% based on the absence of other phases in the XRD patterns. (Wet chemistry analysis of the three constituents was not possible because of funding limitations.) While XRD does not usually respond to phases present at 5 wt% or less, researchers did not believe that the products contained $CuCl_2$. The reason for this assertion is that solutions of $CuCl_2$ are intensely colored, a very pale blue for low concentrations and a deep green for high concentrations. When a small portion of the hydrolysis products was washed with water, an insoluble black material precipitated. The water remained clear. Upon standing, the precipitate turned blue/green, a characteristic color of one of the copper hydroxychlorides—a decomposition product of Cu_2OCl_2 and water.

The Oxychloride Decomposition. The most recent work involved heating a product from the hydrolysis reactor from room temperature to 700°C and following the evolution of the gaseous products with a mass spectrometry. The mass spectrum for O_2 is shown in Figure 5. Researchers calculated the peak area and, using a calibration curve, determined that the area corresponded to 100 percent of the theoretical amount if the sample contained 95 percent Cu_2OCl_2, which is consistent with the presence of some CuCl. Note that evolution of O_2 starts at about 400°C and is essentially completed around 525°C.

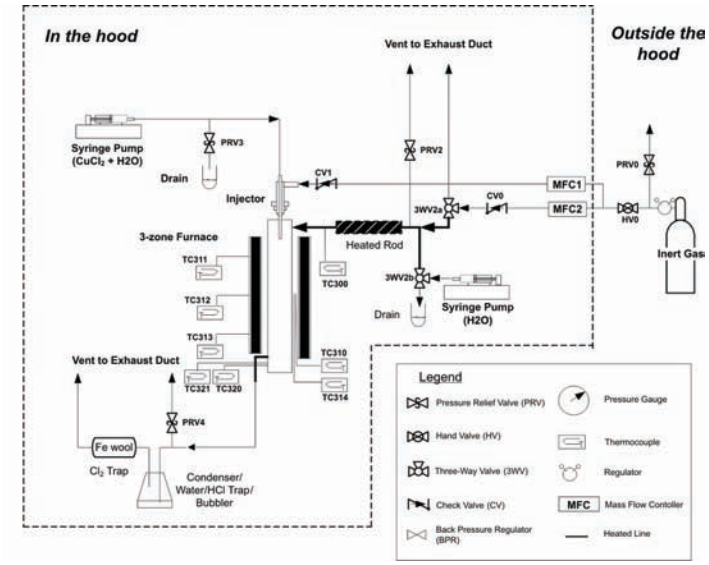


Figure 2. Schematic of the spray reactor apparatus with a pneumatic nebulizer.

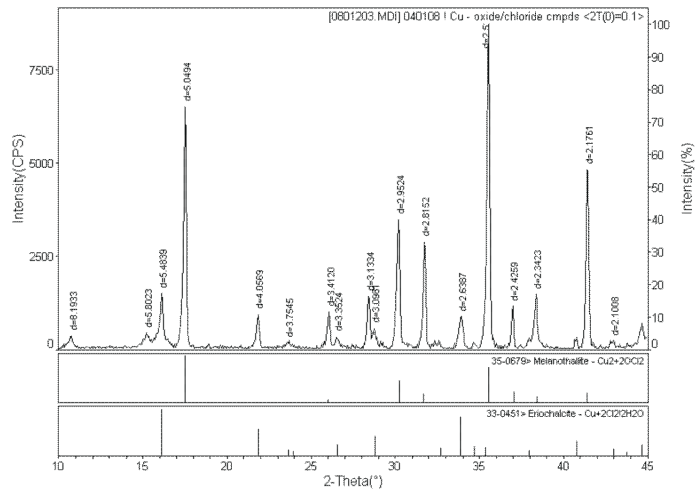


Figure 3. XRD pattern of a solid product produced with a nebulizer. The d-spacings are written above each line.

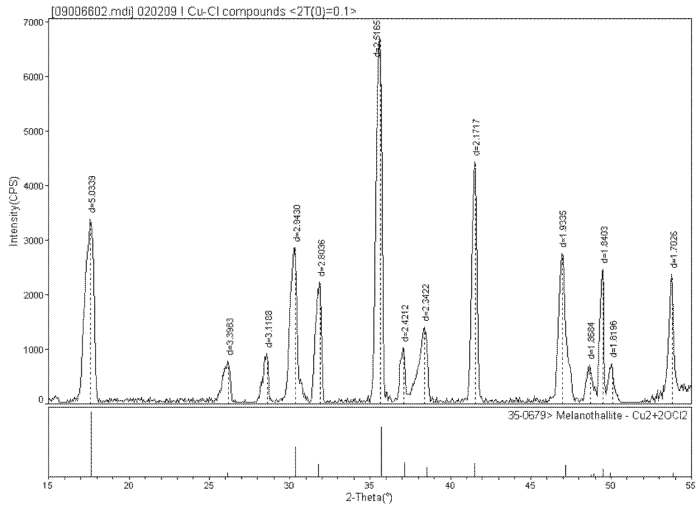


Figure 4. XRD pattern of the solid product produced with an ultrasonic nozzle.

Planned Activities

This project has been completed.

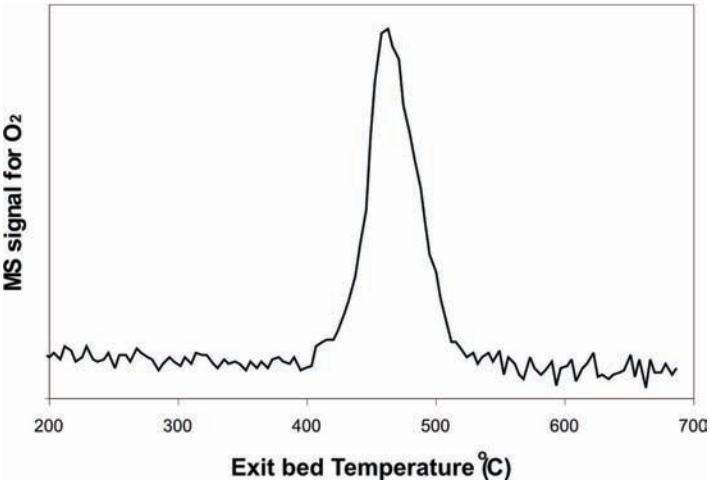


Figure 5. Mass spectrum of oxygen released during the heat-up of copper oxychloride.

INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE

Upgrading of the Athabasca Oil Sands for the Production of Diesel and Gasoline

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

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

Collaborators: Chalk River Laboratories

Project Number: 2008-001-C

Program Area: NHI

Project Start Date: June 2008

Project End Date: May 2011

Research Objectives

A three-year collaboration is ongoing between INL and AECL to develop a preliminary design for an integrated complex consisting of a nuclear reactor, electrolytic hydrogen plant, and bitumen upgrader. The complex will provide steam for bitumen extraction and electricity for the electrolytic hydrogen plant. The hydrogen produced will be sent to the bitumen upgrader to produce synthetic crude oil, gasoline, and diesel fuel. The integrated plant will be optimized to maximize thermal efficiency and minimize the use of water. This project builds on a previous I-NERI project that developed a preliminary design to integrate a nuclear reactor with a high-temperature electrolysis system.

The following is a list of project tasks:

- Task 1: Define requirements for the integrated complex.
- Task 2: Develop an integrated model of a nuclear reactor with electrolytic hydrogen plant, and bitumen upgrader.
- Task 3: Optimize integrated plant to maximize thermal management and minimize the use of water.

Research Progress

One technique chosen for large-scale in-situ recovery is the steam-assisted gravity drainage (SAGD) process. In the SAGD process, steam is injected to heat the reservoir to reduce the viscosity of the bitumen, allowing its production. At present, large quantities of natural gas are used to generate the steam, which produces large volumes of greenhouse gases and leaves operators exposed to volatile natural gas prices. Availability of natural gas is often cited as a barrier to growth in the oil sands.

Another issue that is emerging in Alberta is the availability of water for oil sands projects. Between 2 and 4.5 barrels of water are needed to produce each barrel of crude oil. Oil sands projects use water to generate steam for extracting the oil from the oil sand, as cooling water, and as feedwater for boilers and hydrogen production (in steam-methane reformers). However, Alberta has a very limited supply of fresh water, and there are concerns that water withdrawals in the northern part of the province may be limited in the future. Investors are starting to cite water risk as one of the potential barriers to growth in the oil sands.

An integrated facility containing a nuclear reactor, hydrogen plant, and bitumen upgrader is capable of utilizing the available heat and water resources most efficiently. For example, heat from the reactor, upgrader, and high-temperature electrolysis plant may be used to produce steam for SAGD. Water may be used for cooling before it is fed to boilers or electrolysis cells.

Task 1: Define Requirements for the Integrated Complex. In this task, which is being led by AECL, researchers are defining the requirements that the integrated complex will meet. These requirements include determination of the temperature, pressure, and quality of the steam required for the SAGD process. The team will select reference designs for the nuclear reactor, hydrogen production process, and process for bitumen upgrading from reference flowsheets developed by INL. The bitumen upgrading process selected will specify the requirements for the hydrogen stream (temperature, pressure, and quantity) produced by the electrolytic hydrogen plant.

Task 2: Develop an Integrated Model of a Nuclear Reactor with Electrolytic Hydrogen Plant and Bitumen Upgrader. In this task, researchers are developing the integrated model of the complex consisting of a nuclear

reactor, hydrogen plant, and bitumen upgrader. The model will be developed using the process simulator HYSYS from AspenTech, Inc. HYSYS was originally designed as a process simulator for oil and gas applications, so it is suitable for the simulation of a bitumen upgrading process. HYSYS was used by INL staff for their work on integrating a nuclear reactor with a high-temperature electrolysis plant.

Planned Activities

Task 3: Optimize Integrated Plant to Maximize Thermal Management and Minimize the Use of Water. In this final task, researchers will optimize the process model of the integrated complex developed in Task 2 to maximize the heat recovery in the process and minimize the amount of fresh water taken from the environment.

INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE

Evaluation and Demonstration of Advanced Power Conversion and Hydrogen Generation Technologies for Low-Emission Nuclear, Fossil and Renewable Energy Sources

PI (U.S.): Steven Wright, Sandia National Laboratories (SNL)

PI (Canada): Kourosh E. Zanganeh, CanmetENERGY Technology Centre-Ottowa (CanmetENERGY)

Collaborators: None

Project Number: 2008-002-C

Program Area: Gen IV

Project Start Date: June 2008

Project End Date: June 2011

Research Objectives

The objective of this collaborative research effort is to evaluate advanced power conversion technologies for application to low-carbon emission nuclear, fossil or renewable energy sources. The initial focus of the work will be on evaluating the potential of the supercritical CO_2 Brayton cycle for use with advanced nuclear and fossil energy sources. Based on the first-year evaluation, researchers will select a promising power conversion application. In the second year, they will develop a conceptual design for an appropriately sized, small-scale, integrated demonstration system.

Depending on the future availability of resources and programmatic priorities in the United States and Canada, options will be explored to expand the collaboration to include the construction and operation of a small-scale (5 to 20 MWt) integrated experiment to demonstrate system performance. Research to demonstrate the key technical features of the supercritical CO_2 cycle will continue to be conducted through Gen IV. Work on the key technical issues for the selected advanced heat source will be conducted at CanmetENERGY.

Coupling the advanced power conversion technology being developed by DOE for nuclear applications with the next-generation fossil fuel energy conversion technology being investigated by CanmetENERGY can accelerate development of both technologies. This collaboration will also consider advanced hydrogen production processes for use with these advanced nuclear and fossil energy systems at a later stage.

Research Progress

During FY 2009, research efforts focused on 1) evaluating candidate near-zero emission advanced energy conversion processes for use with the supercritical CO_2 Brayton cycle and 2) demonstrating the key technologies of the S-CO_2 cycle. Activities in the primary task areas for FY 2009 are summarized below.

Task 1. Evaluate Applicability of the Supercritical CO_2 Brayton Cycle Technology to Advanced Nuclear Energy Systems. The S-CO_2 cycle has been evaluated for use with advanced nuclear applications where the characteristics of the S-CO_2 cycle have potential cost or efficiency advantages. The S-CO_2 cycle is considered as a baseline power conversion approach for intermediate-temperature sodium- or lead-cooled nuclear reactors, since the highly recuperated S-CO_2 Brayton cycle allows high efficiency to be achieved at the intermediate outlet temperatures of these reactors. The very compact size of the S-CO_2 turbomachinery is also appropriate for many of the small modular advanced reactor systems currently under consideration at DOE. The high-temperature gas-cooled reactor is characterized by a relatively large temperature rise through the core, which requires a different configuration. For the gas-cooled reactor, a cascaded approach using two S-CO_2 cycles in series results in higher efficiency than Rankine or helium Brayton cycles for the same turbine inlet temperature.

SNL is developing the technology basis for the S-CO_2 cycle evaluation under the Gen IV program. Two supercritical CO_2 loops have been constructed at Barber Nichols Inc to demonstrate the key technologies of compression near the critical point of CO_2 and the control strategies necessary for stable operation for the high-efficiency dual compressor configuration. The Gen IV loop is sized for a maximum of one megawatt thermal (MWt) input power and will generate about 250 kilowatts electric (KWe) in the final configuration.

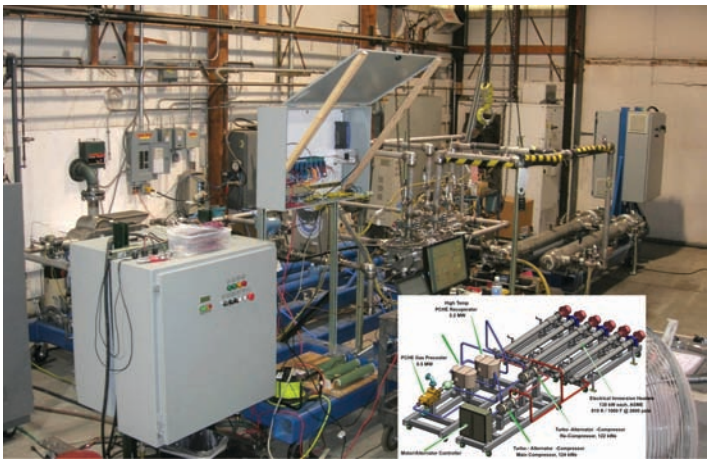


Figure 1. The dual compressor S-CO_2 Brayton cycle test loop as configured at Barber Nichols Inc.

Tests were conducted in FY 2009 to develop the bearing, seal, and motor alternator technologies necessary for the stable operation of these small, high-speed machines, which operate in the 50,000 rpm to 70,000 rpm range. The dual compressor loop was assembled in late FY 2009, and experiments are now under way to evaluate operation at higher temperatures with net electrical power production. FY 2010 tests will demonstrate the recuperated cycle at intermediate temperatures to confirm control algorithms and performance. The small-scale and high-rotational speeds of these machines (approximately one MWt) have required development of innovative bearing and seal approaches. However, as the size of the system is scaled up, most bearing, seal, generator, and related supporting technologies become more commercially available. A key issue, therefore, is to determine the optimum size of the next scale demonstration. Larger systems are more commercially relevant and supporting technologies are more available, but larger demonstrations are also more expensive to build and operate.

Task 2. Evaluate the Use of S-CO_2 for Fossil Fuel Applications. Researchers at CanmetENERGY examined near-term concepts for carbon-managed fossil-fired power conversion systems. SNL provided CanmetENERGY with their latest process modeling results on the mass, energy flow, and turbomachinery efficiencies associated with the SNL S-CO_2 closed Brayton cycle design. CanmetENERGY in turn developed ASPEN/HYSYS models of SNL's S-CO_2 Brayton cycle and validated the model's performance against the data provided by SNL. Using ASPEN/HYSYS thermodynamic modeling, CanmetENERGY analyzed a number of natural gas air combustion processes and cycle options for their respective thermal efficiency and effectiveness in integrating with the S-CO_2 closed Brayton power cycle. For the basic cycle, an overall maximum thermal-to-power efficiency of about 35 percent was identified, making this approach less attractive than conventional natural gas combined cycles (NGCC), which have a cycle thermal-to-power efficiency in excess of 50 percent. The basic S-CO_2 closed Brayton cycle was also investigated as a possible middle cycle in a conventional NGCC. Overall efficiencies were slightly lower than NGCC, offering little or no economic advantage over the conventional NGCC. Several advanced coal and natural gas combustion processes were then developed which, when integrated with the S-CO_2 closed Brayton cycle, showed efficiency and economic advantages over other technology options in a niche fossil-fired power market. This development is presently undergoing a patent application and is referred to as a "second generation" or "G2" clean fossil fuel technology for power production.

Task 3. Select the Most Promising Concept for an Integrated Small-Scale Demonstration of the S-CO_2 Brayton Cycle. Based on the range of candidate energy sources considered in Task 1, the objective of Task 3 was to select a promising option that could serve as the next-level scaling demonstration. The advanced coal and gas combustion cycles evaluated in Task 2 combined the advanced burner technologies with the S-CO_2 electrical power conversion system, providing distinct advantages for carbon management. In July 2009, under a non-disclosure agreement, DOE, SNL, CanmetENERGY, SaskPower, and Barber Nichols Inc met to discuss the options for a small-scale 5 MWt to 25 MWt demonstration of the natural gas-fired G2 technology. The discussion assumed that researchers at SNL would be responsible for providing the components for the S-CO_2 closed Brayton cycle process and researchers at CanmetENERGY would be responsible for providing the components for the integrated G2 combustion process. Discussions on this general approach are expected to continue in FY 2010.

Task 4. Develop Conceptual Designs for the Candidate System. Although most of the effort on this task is scheduled for FY 2010, a preliminary scoping analysis focused on the use of an advanced burner technology and a dual compressor (recompressor) S-CO_2 cycle with operation at high temperature (~650°C) to maximize efficiency. The size of the unit was considered to be in the 5 MWt to 20 MWt power level for the pilot demonstration. This activity will continue in FY 2010. To assist with the

design for the demonstration project, CanmetENERGY prepared an $11 million research and development proposal to develop proof-of-concept of the G2 combustion process and system components at the pilot scale. Funding considerations are presently under review by Natural Resources Canada.

Planned Activities

Activities in FY 2010 will focus on analyzing a 5 MWt to 20 MWt demonstration power conversion system coupled to an advanced burner heat source. The preliminary analyses done in FY 2009 will be refined in FY 2010 to provide the basis for further evaluation of the concept and to develop a test plan and schedule. The resulting information will be used to develop a more accurate cost estimate for the small-scale demonstration and definition of options for funding and the potential for broader collaboration.

INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE

Study of Burning Transuranic Elements in Heavy- and Light-Water Reactors

PI (U.S.): Jess C. Gehin, Oak Ridge National Laboratory (ORNL)

PI (Canada): Bronwyn Hyland, Atomic Energy of Canada Limited (AECL)

Collaborators: None

Project Number: 2008-003-C

Program Area: FCR&D

Project Start Date: May 2008

Project End Date: September 2009

Research Objectives

The objective of this project was to examine and compare the isotopic evolution of actinide-bearing fuel under irradiation in heavy-water reactors (HWRs) and light-water reactors (LWRs) to understand the physics involved and to provide a comparative analysis of the burning performance in the two reactor types. In this project, ORNL and AECL collaborated on evaluating the capability of LWRs and HWRs for burning transuranic elements, particularly minor actinides. The two teams focused on their respective technical strengths in a coordinated fashion: the ORNL team addressed the LWR aspects of this project, primarily pressurized water reactors (PWRs), while the AECL team placed their emphasis on the HWR Canada Deuterium Uranium (CANDU) technology. The work was arranged in four tasks: 1) to establish goals for an intermediate actinide burner and metrics for assessing repository benefits; 2) to determine a nominal composition for spent LWR fuel; 3) to calculate the isotopic evolutions in HWR and LWR cores with TRU-MOX fuel and targets, with considerations for use of recycled uranium (RU), as appropriate; and 4) to perform comparative analysis of results from LWR and HWR analyses. The results related to reducing the decay heat of high-level waste. ORNL led Tasks 1 and 2 and ORNL and AECL shared the lead on Tasks 3 and 4.

Research Progress

There is interest worldwide in reducing the burden on geological nuclear fuel disposal sites. In most disposal scenarios, the decay heat loading of the surrounding rock limits the capacity of these sites. In the long term, this decay heat is generated primarily by actinides; a major contributor 100 to 1,000 years after discharge from the reactor is americium (Am)-241. Am-241 is produced in the reactor during irradiation, but most of the Am-241 in spent nuclear fuel results from the decay of Pu-241 (half life of 14 years) after the fuel has been discharged from the reactor. Consequently, it is beneficial when investigating Am transmutation schemes to study fuel that has decayed for a significant length of time after discharge. This study used 10-year-cooled LWR fuel with a discharge burnup of 50 megawatt-days (MWd)/kg, which has a significant build-up of Am-241 to be transmuted.

One possible approach to reducing the decay heat burden is to reprocess spent reactor fuel and use thermal spectrum reactors to “burn” the Am nuclides. The viability of this approach is dependent on the detailed changes in chemical and isotopic composition of actinide-bearing fuels after irradiation in thermal reactor spectra. The currently available thermal spectrum reactor options include LWRs and HWRs such as the CANDU design. In addition, as a result of the recycle of spent LWR fuel, there would be a considerable amount of potential RU. One proposed solution for the RU is to use it as fuel in CANDU reactors; RU was considered in this work as a diluent for Am-241.

In order to investigate the potential of thermal reactors to transmute the Am nuclides, ORNL and AECL calculated the transmutation of Am added to fuel assemblies in PWRs and to fuel bundles in CANDU reactors. The CANDU scenarios study a full core fueled with homogeneous bundles of Am mixed with RU, while the PWR scenarios were performed with single assembly lattice calculations. The cases considered for both sets of scenarios are listed in Table 1. Parameters including the overall Am transmutation, decay head, and neutron and gamma sources were calculated and summarized.

Table 1. HWR and LWR cases considered in this study.

HWR1	Unenriched RU composition with 0.096% Am to obtain a burnup of 7.5 MWd/kg.
HWR2	0.28% Am, burnup of 7.5 MWd/kg.
HWR3	0.28% Am, burnup of 7.5 MWd/kg. Center pin of the bundle has 2.4% Dy in zirconia.
HWR4	0.28% Am, burnup of 7.5 MWd/kg. Center pin of the bundle has 7% Am in zirconia.
HWR5	0.28% Am, burnup of 21 MWd/kg.
HWR6	0.28% Am, burnup of 21 MWd/kg. Center pin of the bundle has 1.3% Dy in zirconia.
PWR1	Heterogeneous assembly, four fuel rods in the lattice composed of 5 wt% AmO_2 mixed with LEU, burnup of 42 MWd/kg.
PWR2	Heterogeneous assembly, four fuel rods in the lattice composed of 20 wt% AmO_2 mixed with LEU, burnup of 42 MWd/kg.
PWR3	Homogeneous assembly, all 248 standard fuel rods with 0.0807 wt% AmO_2 mixed with LEU, burnup of 42 MWd/kg.
PWR4	Homogeneous assembly, all 248 standard fuel rods with 0.15 wt% AmO_2 mixed with LEU, burnup of 42 MWd/kg.
PWR5	Reference case with no added AmO_2, burnup of 50 MWd/kg.

The CANDU reactor and the PWR can achieve similar fractions of transmutation of Am. The high burnup CANDU reactor cases achieve around 77 percent transmutation (HWR7), while the homogeneous PWR case (PWR4) is slightly lower, at 71 percent. It is likely that an AmO_2 mixture fraction much larger than 0.15 wt% would be viable. The highest mass transmutation cases burn 2.8 kg/MTIHE in CANDU (HWR7), versus 1.7 kg/MTIHE for the PWR (PWR2), though optimized PWR cases would be expected to be comparable to the current CANDU cases in terms of Am transmutation mass. While the rate of Am feed into the PWR is similar to that in the CANDU fuel cycle, the CANDU transmutes a higher percentage of its Am feed in the high feed cases (PWR2, HWR7). As a result, the net total destruction of Am is higher in the CANDU fuel cycles than the PWR fuel cycles.

As mentioned earlier, one parameter of interest is the decay heat generation. Figure 1 shows the effect on the decay heat in the Am transmutation fuel cycle from that which is generated in a reference fuel cycle for the two different reactors. The reduction in decay heat at 1,000 years is 63 percent for the CANDU case and 35 percent for the PWR case.

Planned Activities

This project has been completed.

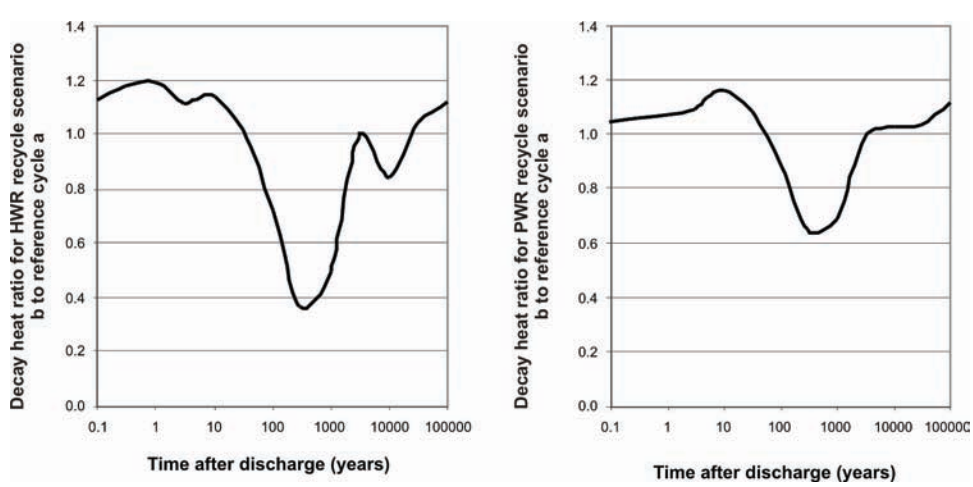


Figure 1. Comparison of the discharge fuel decay heat for the CANDU (HWR) Am-241 burning scenario (left) and the PWR Am-241 scenario (right) relative to the reference PWR case with typical 50 gigawatt-days per metric ton of uranium (GWd/MTU) discharge fuel.

INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE

4.2 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 (EU), 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 United States and Euratom selected the first ten projects for collaboration. A collaboration with Italy was established in 2006 under the Euratom agreement, resulting in two ongoing projects.

Work Scope Areas

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

- 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

Project Summaries

A listing of the I-NERI U.S./EU projects that are currently under way follows, along with summaries of the accomplishments achieved in FY 2009. Two of these projects concluded shortly after the end of the fiscal year, and no new collaborations with Euratom were initiated.

Directory of Project Summaries

2006-001-E Experimental Investigation of Small-Break LOCAs in Coupled Vessel/Containment Integral Reactors....31

2006-002-E Advanced Nuclear Fuel Properties Measurements and Fuel Performance Modeling....35

2006-003-E* Development of Novel Transmutation Systems for Sustainable Nuclear Fuel Cycles....39

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* 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 (ORNL), Società Informazioni ed Esperienze Termoidrauliche (SIET)

Project Number: 2006-001-E

Program Area: FCR&D

Project Start Date: May 2006

Project End Date: September 2011

Research Objectives

The purpose of this project is to verify, experimentally, the behavior of integral reactors during accident conditions. The Global Nuclear Energy Partnership (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 (LOCAs) that does not rely on dedicated safety systems for coolant injection. The integral configuration of IRIS (without the primary loop external-to-reactor vessel) also precludes the possibility of a large-break LOCA.

During a small-break LOCA, the reactor vessel depressurizes because of heat removal and condensation by the integral steam generators, while the pressure in the small, spherical containment increases from the steam release through the break. The two pressures equalize relatively quickly, nullifying the pressure differential that drives the coolant egress, thus terminating the LOCA without any coolant injection or 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 to SIET's existing AP600 test facility to represent the characteristics of an integral rather than a loop-type reactor and the vessel/containment coupling. Researchers will develop an analytical program to guide testing. The test results will support the analyses presented in the planned IRIS design certification application.

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 tests in the test matrix, including shakedown tests.
- Evaluate results and prepare a comprehensive report.

Research Progress

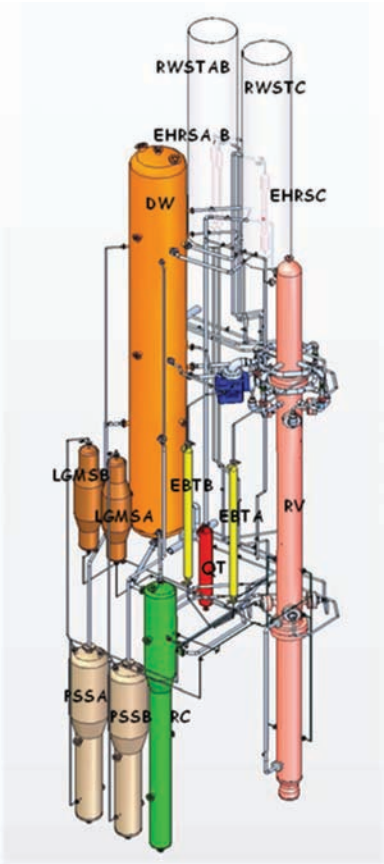
During FY 2006 and FY 2007, the team created a conceptual design for the small-break LOCA experimental facility (designated SPES-3). The team completed a stage gate review at SIET in Piacenza, Italy, on October 8, 2007. This was a major milestone, and its completion allows the team to move forward with equipment procurement. Twenty-six people from eleven organizations attended the review, including a representative from the U.S. Department of Energy.

Several organizations, including Westinghouse, SIET, and the University of Zagreb, formed a dedicated team to address scaling issues. Last year, the team "froze" an SPES-3 baseline design and an SPES-3 reference IRIS design. These reference designs became the basis for the IRIS/SPES-3 scaling analyses and for comparing SPES-3 and IRIS-calculated transient responses. Uncertainties in the IRIS plant dimensions now limit further improvement in the SPES-3 design.

This year, Westinghouse compared the simulation results and determined how well the SPES-3 facility scaled to the reference IRIS design. Although the results were encouraging, the team found some distortions, primarily in the containment pressure response. The main cause for this distortion is the larger area/volume ratio in SPES-3, which is a direct consequence of the full-height 1:100 volumetric scaling. Reducing the design pressure of the SPES-3 drywell allows thinner walls and a lower heat capacity, which improves the response. Throughout the year, the team identified several other reasons for the distortions and adjusted the models and design accordingly. Westinghouse compared the latest analyses of the adjusted facility design and IRIS. Many of the distortions have decreased, and the team now understands the causes and effects of most of those that remain.

The resulting design and analyses are for a full-height, 1:100 volumetric scale model of the following components:

- 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 system
- Containment system simulated by separate tanks and piping
  - Reactor cavity
  - Dry well
  - Pressure suppression system
  - Long-term gravity make-up system
  - Passive containment cooling system



The Italian partners continued procurement activities. SIET prepared valve specifications, tank specifications, instrumentation specifications, and data acquisition and control system specifications for procurement. SIET is working with their suppliers to develop detailed designs, including the drawings and documents needed to manufacture components. Suppliers identified the need for some minor changes to allow for easier manufacture, but none of these invalidated the basic design or the latest analyses.

Westinghouse issued a formal IRIS Integral System Test Specification, which ENEA agreed to use to supersede their earlier specification to SIET. Researchers are now updating the specification to reflect their latest agreed-to "frozen" dimensions. Last year, SIET issued a "Quality Plan of IRIS SPES-3 Project" to cover their design, testing, and other activities. This year, Westinghouse hired a quality assurance (QA) engineer to prepare an overall project-specific QA manual to cover activities and organizations associated with the IRIS program, including the SPES-3 program. Westinghouse reviewed and approved the SIET QA plan.

Planned Activities

The team will now procure components, assemble equipment, and then construct and commission the test facility. The Italian partners will have the main responsibility for this task, with consultation and advice from the U.S. partners. ENEA will bear the substantial cost of materials and refurbishment.

After commissioning, the main task will be to conduct the tests in the test matrix, including shakedown tests. The team will conduct some separate effects tests as part of the shakedown program, then follow with various aspects and phases of the integral tests. SIET will conduct the tests with advice from Westinghouse. ENEA and ORNL will provide consultation and perform ad hoc analyses during and after the tests to verify, interpret, and guide the tests.

The final task will be to prepare a comprehensive test report. This will be part of the safety documentation submitted to support the IRIS application for Final Design Approval with the U.S. Nuclear Regulatory Commission. Westinghouse will prepare the report with input from the other participating organizations.

Because of familiarity with the QA requirements, Westinghouse and SIET will continue preparing low-level procedures to address test planning and execution, as well as the configuration control of the theoretical studies (e.g., 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: Argonne National Laboratory, Arizona State University, Los Alamos National Laboratory, Oak Ridge National Laboratory, Pacific Northwest National Laboratory

Project Number: 2006-002-E

Program Area: FCR&D

Start Date: January 2007

End Date: December 2010

Research Objectives

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 is for mixed oxide fuel (MOX) performance code development and validation. MOX, together with metal alloy fuel, are under consideration in the Fuel Cycle Research and Development (FCR&D) program 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 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 FCR&D 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 of 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.

Research Progress

Experimental work in the United States during FY 2009 was primarily directed towards fabrication and characterization of advanced metal alloy transmutation fuels and fabrication of advanced MOX transmutation fuels intended for irradiation testing within the scope of the AFC-2 test series and with regard to achieving a more fundamental understanding of the fuel material. For the metal alloy fuels, the team initiated investigations on uranium-plutonium-zirconium (U-Pu-Zr) compositions related to the compositions of the U-Pu-Am-Np-Zr (with americium and neptunium) transmutation fuels. Phase compositions were analyzed from X-ray diffraction results using the Rietveld refinement technique and showed, as expected, that these alloys were composed of primarily three phases: ζ-phase arising out of the U-Pu binary, δ-MZr_2 phase arising out of the U-Zr binary (and maybe the Pu-Zr binary), and a small portion of Zr-rich precipitates.

Heating and cooling curves from ambient temperature to about 800°C obtained from differential thermal analysis (DTA)-type measurements on a transmutation test fuel are provided in Figure 1 for two compositions of a U-Pu-Zr alloy. The top set of curves is for a high-Zr-content fuel and the bottom set for a low-Zr-content fuel. The thermal

behavior between the two fuels is significantly different wherein two closely spaced transition signals can be deduced from the high-Zr-content fuel and at least three broader-spaced transition signals observed in the lower-Zr-content fuel. The traces are seen to be repeatable and reproducible for each alloy. Although there is reasonable consistency between the determined transition onset temperatures from the heating and cooling curves, the relative intensities of the peaks during heating and cooling vary considerably. This aspect is still under investigation.

The thermophysical property measurements on the metal alloys include determination of the thermal conductivity through measurements of the thermal expansion, density at room temperature, heat capacity, and thermal diffusivity. A thermal diffusivity plot for one of the U-Pu-Zr alloys is shown in Figure 2. It can be observed that although the general thermal behavior is consistent for the heating and cooling curves, the cooling curves appear to give a higher diffusivity than expressed during heating. This difference represents about a 100°C difference for and value of diffusivity, which is significantly less than any under-cooling observed in the differential scanning calorimetry (DSC)/DTA measurements (Figure 1). The source of the discrepancies is still under investigation.

DOE-sponsored work on converting FRAPCON modules from oxide to fast reactor metal fuel models was not funded in FY 2009; therefore, no progress can be reported in this area. It is still unclear whether this work effort will be supported in FY 2010.

DOE-sponsored activities during FY 2009 included work to secure required feedstock supplies of the minor actinides Np and Am for future studies. Part of this work was to continue electrochemical reduction of NpO_2 in a glovebox and begin testing a method and procurement equipment for Am oxide reduction in a hot cell environment. ITU researchers continued to provide valuable information through discussions and personal communications as well as archived documentation on past efforts performed at ITU on the reduction of Np and Am oxides.

A key feature to the collaboration, and perhaps one of the more logistically difficult tasks to institute, will be a combined characterization effort on irradiated fuel samples. Several irradiated and non-irradiated fuel materials have been discussed in this respect. In addition, separate discussions took place between U.S. researchers and individuals from ITU, CRIEPI, and CEA on the viability of sharing METAPHIX irradiation test samples with the United States. These discussions are continuing. The

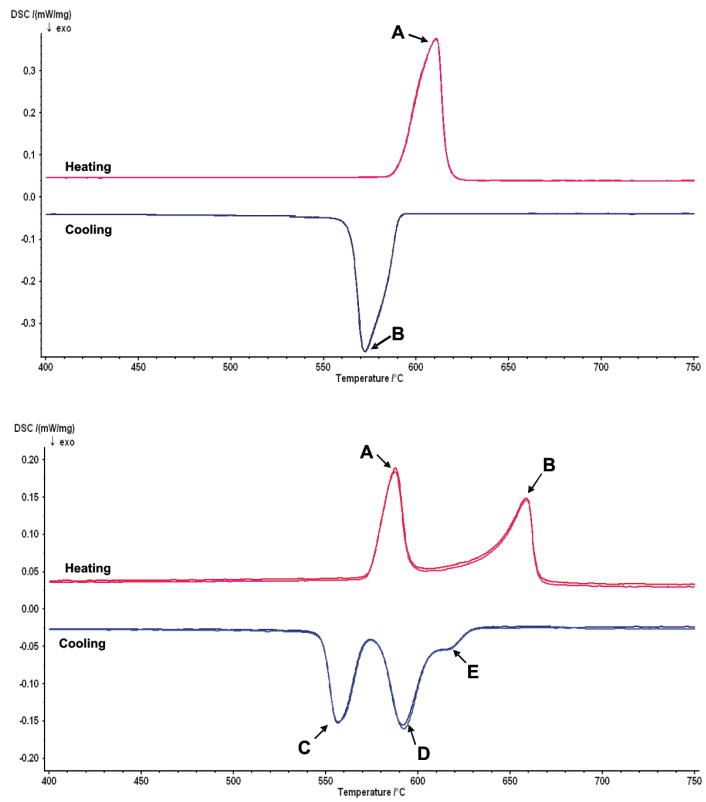


Figure 1. Heating and cooling traces obtained from the DSC/DTA measurements on a (top) high-Zr-content and a (bottom) low-Zr-content U-Pu-Zr fuel.

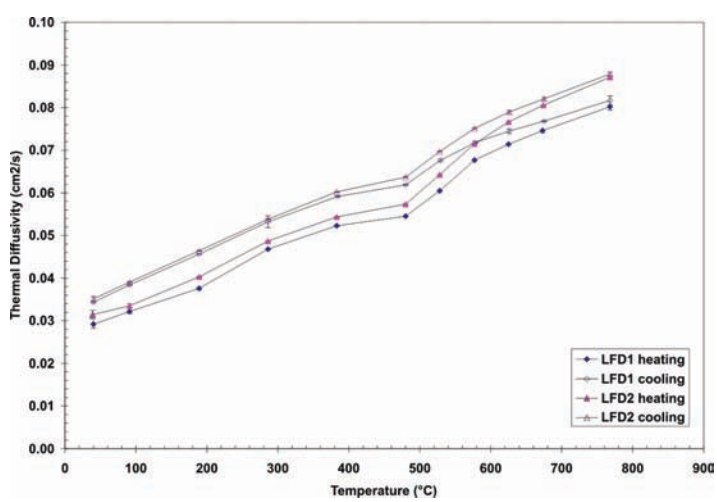


Figure 2. Heating and cooling traces obtained from laser flash thermal diffusivity measurements on a U-Pu-Zr fuel.

transport of samples between INL and ITU remains a difficult issue, but significant progress was achieved in 2009; the team identified several transport containers and began to analyze costs for transport. There is no current budget to support material transport.

Characterization of relevant properties of unirradiated nitride fuel at ITU, initiated in 2008, was completed in FY 2009. Fuels such as uranium nitride (UN) and (U,Pu)N and matrices like ZrN and (Zr,Pu)N were characterized in terms of thermophysical properties in general and compared to oxide fuels, with attention to radiation damage effects on the material properties due to the presence of Pu and other (minor) actinides. Such effects directly influence the evolution of properties during storage after irradiation. However, in the case of high specific activity fuel (e.g., for minor actinide-containing fuel) or in the case of long storage time after fabrication, possible alterations may occur also *before* irradiation.

Figure 3 illustrates a typical example of the studies on accumulated decay damage effects. The team studied unirradiated mixed ($U_{0.78}$ $Pu_{0.22}$)N samples stored for approximately 17 years. The oxygen content of the samples was in the range of 1,000 to 3,000 parts per million (ppm), and the carbon content was lower than 1,000 ppm. The density was 82 ±2 % TD.

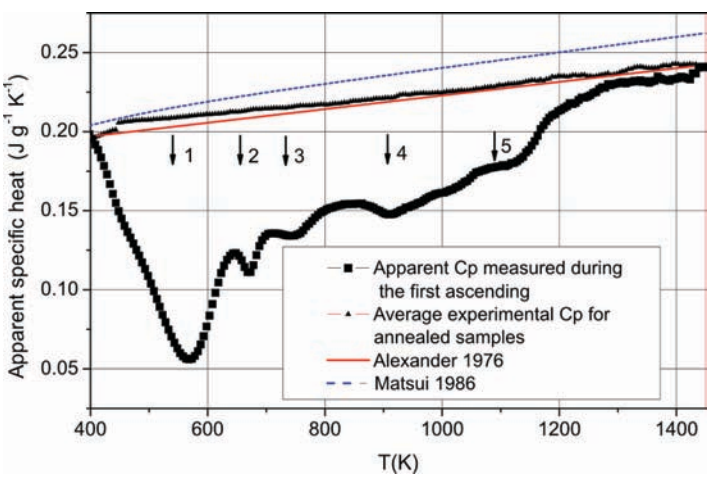


Figure 3. Experimental Cp of ($U_{0.78}Pu_{0.22}$)N compared to literature, and position of the five exothermic stages corresponding to annealing of families of defects in the material.

The specific heat (*Cp*) was measured by DSC (heating rate: 20 K/min) under high-purity argon at atmospheric pressure. The "true" *Cp(T)* was obtained by averaging the ascending and descending curves of undamaged or annealed samples. The average of the experimental *Cp* obtained for four samples was in good agreement with the data of Alexander, and was lower by about 8 percent compared to the Matsui values, as shown in Figure 3.

The deviation of the apparent *Cp*(T)* measured during the first heating run from the true heat capacity, *Cp(T)*, is related to the release of heat during annealing of the radiation damage accumulated during the long storage of the samples (Figure 1). The characteristic temperatures of the five observed annealing stages agree with the literature data available for UN, which found five annealing steps in UN measuring electrical resistivity, lattice parameter, and Knopp hardness (~400, 550, 750, 950 and 1,100 K for heating at 50 K/h), with characteristic temperatures strongly depending on the heating speed (especially for the first steps). As the heating rate used for these DSC runs is higher (20 K/min), slightly higher peak temperatures were observed.

Ab initio calculations were performed to assess the incorporation of helium in mixed (U,Pu)O_2 solid solutions. In addition to contributions from INL and ITU, the work involved contributions of a team from the Institute of Solid State Physics (ISSP) of Riga, Latvia, performed in the frame of a service contract with ITU.

Helium (He) is accumulated in MOX mostly as a result of actinide alpha-decay, which seriously affects fuel mechanical and thermal properties. Helium incorporation into MOX was studied using *ab initio* calculations. Researchers used the VASP 4.6 code for the simulations. They performed density functional theory (DFT) calculations for UO_2, PuO_2, and MOX containing He atoms in octahedral interstitial positions. The calculations were performed for He in the octahedral position only.

In particular, basic MOX properties and He incorporation energies were calculated as functions of Pu concentration within the spin-polarized, generalized gradient approximation (GGA) DFT calculations. The on-site electron correlation corrections were also included using the Hubbard model (in the framework of the so-called DFT+U approach). It was found that PuO_2 remains semiconducting with He in the octahedral position while UO_2 requires a specific lattice distortion. Both materials reveal a positive energy for He incorporation, which, therefore, is an exothermic process. PuO_2 reveals considerably larger He incorporation energy than UO_2. The He incorporation energy increases with the Pu concentration in the MOX fuel. The host crystalline symmetry reduction is particularly important for calculations of He incorporation energy into octahedral interstitial positions in actinide dioxides. In particular, properly calculated He incorporation energy in UO_2 turns out to be reduced in a comparison to high symmetry with conducting states. The calculations on MOX solid solutions require proper analysis of the ground state. Lifting the symmetry constraints resolves the problem but indicates complex lattice distortion. Future studies will include larger supercells and points defects on both the U and Pu MOX sublattices in order to analyze the role of defects in the He behavior.

Planned Activities

Researchers are working to resolve the issues associated with sample sharing transport. The identification of a viable transport solution to exchange samples between INL and ITU is essential for deploying the full scope of the present collaboration program. Until a solution is found, the scope of the joint work will remain forcibly limited.

The team will also continue interactions related to post-irradiation examinations with emphasis on operation of electron probe microanalysis as well as advanced technique development such as micro X-ray diffraction, focused ion beam, and thermal diffusivity measurements.

The results reported from ITU represent part of a larger ITU program of characterization, which includes studies on UN and solid solutions $(U_xPu_{1-x})N$ and will be extended to cover both (U, Pu, MA)N (where MA stands for minor actinides Np, Am, Cm) and irradiated nitride, carbide, and metallic alloy fuel samples. The team plans information exchange and discussions of the results with possible joint publication with U.S. researchers, particularly in the area of cooperation between the experimental measurements (ITU) and the modeling and simulation computations (U.S.) on the nitride system for heat and mass transfer.

The results reported from the United States as part of the FCR&D program will continue with a focus on metal alloy and oxide systems. Primary objectives will involve fundamental studies on basic actinide systems. Some emphasis will be given to high-temperature measurement development. The team plans information exchange and discussions of the results with possible joint publication with ITU researchers.

Modeling and simulation activities will involve information exchange on computational efforts and mechanism development related to fission gas release, microstructure development, elemental redistribution, phase stability, and thermophysical property behavior of irradiated fuels.

INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE

Development of Novel Transmutation Systems for Sustainable Nuclear Fuel Cycles

PI (US): J. Carmack, Idaho National Laboratory

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

Collaborators: Los Alamos National Laboratory

Project Number: 2006-003-E

Program Area: FCR&D

Project Start Date: January 2007

Project End Date: January 2010

Research Objectives

The goal of this project was 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 following areas: 1) nitride fuel development, 2) dispersion fuel development, 3) vibro-pac fuel development, 4) target system development, and 5) advanced claddings.

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

The baseline transmutation concept under the Fuel Cycle R&D program relies on a homogeneous core with group TRU included in the fuel. An alternative option may be to put a non-fertile minor actinide into the reactor using inert matrix materials. A longer-term option may be partitioning americium (Am) (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 actinide destruction. Oxide dispersion-strengthened (ODS) steels and advanced ferritic-martensitic materials are two such advanced claddings. R&D 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.

Research Progress

Nitride Fuels. The objective of the nitride fuel R&D is to develop fabrication processes that result in structurally stable fuel pellets and assess these fabrications through irradiation. The research focuses on fabrication process development at small scale and structural characterization of the resulting pellets. Fabrication tests will be conducted over a range of fabrication parameters (powder size, sintering temperature, press pressure, etc.). Nitride fuels may be a good alternative for the baseline transmutation fuels in the future.

Two nitride fuel compositions were irradiated in the Phenix fast spectrum test reactor during fiscal year (FY) 2009. These two fuel compositions were irradiated to 60 effective full power days and completed irradiation. The experiment is currently being returned to the United States for post-irradiation examination. Shipment of the fuel pins will be conducted early in FY 2011 with post-irradiation examination coordinated with researchers at JRC-ITU, who will conduct post-irradiation examination of two inert matrix fuel (IMF) compositions that were also irradiated in the Phenix fast spectrum test reactor.

The performance of the two nitride fuel pins irradiated in the Phenix fast spectrum test reactor will be compared with that of a nitride fuel pin irradiated in the U.S. Advanced Test Reactor in the AFC-1G experiment. The AFC-1G nitride fuel pin was irradiated to approximately 28 percent at burnup. The AFC-1G nitride fuel pin completed irradiation in FY 2008 and is currently undergoing post-irradiation examination.

JRC-ITU continues development of a facility for the fabrication of nitride and carbide fuels. In 2009, the research team conducted the first fabrication trials. Researchers continued post-irradiation examination

of nitride and carbide fuels from an earlier irradiation campaign, NIMPHE, in Phenix.

Inert Matrix Fuels. The irradiation programs on IMFs were completed in Phenix and the HFR Petten (FUTURIX and HELIOS).

Advanced Claddings. To burn fuels to greater than 20 percent burnup without changing the cladding requires development of materials to withstand doses greater than 200 displacements per atom (dpa). During FY 2008, researchers tested advanced ODS ferritic steels using proton irradiations at temperatures of 25°C, 300°C, and 550°C. The materials tested include MA-956 and MA-957, which were compared to HT-9. Nanohardness of the two materials was measured after irradiation and compared. In the area where a proton enters the samples, there is hardening in the HT-9 sample and no hardening (except in the peak dose location) in the MA-957 sample. The team also performed an atom probe and took small-angle neutron scattering measurements on these advanced materials to characterize the oxide dispersions at the atomic scale. The same advanced materials have also been included in the MATRIX SMI and MATRIX 2 irradiations presently under way in the Phenix reactor. Total dose expected is 17 and greater than 60 dpa from 400°C to 500°C while in contact with the sodium coolant.

Fast reactor cladding tubes have not been manufactured in the United States for more than a decade; it is important to revive this capability before the manufacturing knowledge is lost. To this end, a new 300-lb heat of the cladding alloy HT-9 was cast, and the tube manufacturing company Veridiam, Inc. (formerly Carpenter Technologies) has been contracted to fabricate the alloy into cladding tubes following NQA-1 standards. These cladding tubes will be used in future domestic and international fuels test irradiations that will be critical for optimizing fuel performance.

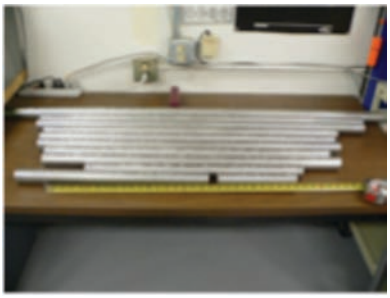


Figure 1. Bar stock drawn from 300-lb heat of HT-9 cladding.

A significant barrier to achieving high burnup levels in HT-9 clad metal fuel is the chemical interaction between the fuel and the cladding that occurs once the fuel swells and contacts the cladding. Both coating and liner technologies are being explored to reduce or eliminate fuel-clad chemical interaction (FCCI). By mitigating FCCI, burnup levels of metal fuel systems will be increased, dramatically reducing fuel handling and improving transmutation efficiencies. Coating the inner surface of a long narrow tube has proven challenging, but during the year researchers developed a novel technique to guide lasers into a cladding tube and deposit a titanium nitride coating using pulsed laser deposition technology. The team demonstrated successful fabrication of lined cladding with vanadium and zirconium liners through a plug-drawing process.

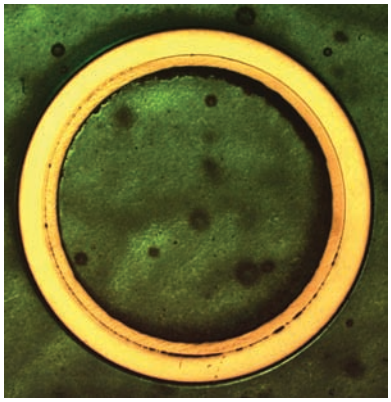


Figure 2. Zirconium-lined HT-9 cladding tube fabricated using a plug-drawing process.

The joining of advanced cladding such as T91 or ODS steel will no longer be possible using usual TIG methods, as the microstructure would be damaged. Possible post heating, even if effective, would be problematic for fuel pins, especially if they contain minor actinides and plutonium. A novel method to fabricate a cold joint is electromagnetic pulse technology (EMPT). With this method, the cladding tube is deformed around a central stop, which acts as the conventional end cap. This device has now been installed at JRC-ITU. First tests (see Figure 3) have shown that helium leak rates in the range of reactor specifications can be achieved. Further steps are necessary to improve the procedure and reliability.

Figure 3. End cap of T91 welded using EMPT method.

Planned Activities

This project has concluded. Further related activities and new collaborative activities related to the welding of advanced materials and fabrication of advanced ceramic materials will be defined for future agreements.

INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE

Development of Oxide Fuels for Transmutation in Fast Reactors

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

PI (EURATOM): J. Somers, Joint Research Center Institute for Transuranium Elements

Collaborators: Los Alamos National Laboratory (LANL)

Project Number: 2006-004-E

Program Area: FCR&D

Project Start Date: January 2007

Project End Date: January 2010

Research Objectives

This project established collaboration between the United States and EURATOM for minor-actinide-bearing mixed oxide fuel development. The purpose was to establish cooperative research and development on oxide fuels, including conduct of the AFC-2 series of irradiations carried out under the transuranic fuel development program in the U.S. Department of Energy Advanced Test Reactor (ATR) at INL. This project examined three main areas, described below: 1) oxide fuel selection and fabrication, 2) oxide fuel characterization and modeling, and 3) irradiation testing and post-irradiation examination.

Oxide Fuel Selection and Fabrication. This assessment included the analysis of neutronic behavior (plutonium [Pu] burn rate, reactivity swing), fuel thermal performance, fuel irradiation performance, and in-core corrosion resistance. Minor actinide mixed oxide fuel compositions were fabricated for inclusion in the AFC-2 test series.

Oxide Fuel Characterization and Modeling. Researchers conducted experimental characterization of oxide fuel microstructure, thermal properties, mechanical properties, corrosion testing, and ion-beam irradiation to satisfy requirements for in-reactor irradiation testing and to provide a basis for fuel behavior modeling. Special attention was paid to the characterization of fuels having varying oxygen-to-metal ratios. Fuel modeling was conducted using finite element analysis to determine the thermomechanical behavior of the fuel. Modeling of thermodynamic and transport properties was also conducted.

Irradiation Testing and Post-Irradiation Examination. An assessment of transmutation fuel feasibility requires in-reactor irradiation because of the extremely complex nature of the in-core fission environment and the inability to simulate this environment with out-of-pile testing. This project helped establish the basis of research for the AFC-2 irradiations.

Research Progress

The focus in fiscal year (FY) 2009 was on the irradiation of test pellets for the AFC-2 C/D irradiation experiment in the ATR.

Figure 1. AFC-2 C/D fuel pellet containing 2 wt% Np and 3 wt% Am.

Figure 1 shows an AFC-2 C/D fuel pellet containing 2 weight-percent (wt%) neptunium (Np) and 3 wt% americium (Am). These fuel pellets are the first of their kind to be fabricated in the United States. Procedures for stoichiometry adjustment to pellets were developed to meet the irradiation test requirements. Pellets were centerless and ground to meet the tight specification of 0.191 inches to 0.192 inches and density criteria of 88+-%TD. Microstructural and scanning electron microscope examinations found that the Am, Np, and Pu are evenly distributed throughout the matrix, showing good elemental homogeneity in the pellets. Dimensional stability during re-sintering was confirmed.

The oxide fuel pellets fabricated by LANL were placed in HT-9 cladding at INL and then in irradiation capsules for irradiation in the ATR. Two capsules (AFC-2C and -2D) were inserted in the ATR in September 2008. These capsules contain the program's first oxide fuels with compositions relevant to fast reactor transmutation of minor actinides uranium (U), Pu, Am, Np, and O_{2-x}. In these oxide fuels, compound stoichiometry is an important test variable that will be studied. Stoichiometry in oxide fuels often controls fuel-cladding chemical interaction in oxide fuel systems that use stainless steel cladding. A total of six fuel pins were fabricated for irradiation. Table 1 provides detailed compositions of the six fuel pins in the AFC-2 C/D experiment. This irradiation is expected to continue through FY 2010, followed by post-irradiation examination.

Table 1. Fuel compositions of the AFC-2 C/D experiment.

Rodlet	AFC-2C & -2D*
1	$(U_{0.75}, Pu_{0.2}, Am_{0.03}, Np_{0.02})\ O_{1.95}$
2	$(U_{0.8}, Pu_{0.2})\ O_{1.98}$
3	$(U_{0.75}, Pu_{0.2}, Am_{0.03}, Np_{0.02})\ O_{1.98}$
4	$(U_{0.8}, Pu_{0.2})\ O_{1.98}$
5	$(U_{0.75}, Pu_{0.2}, Am_{0.03}, Np_{0.02})\ O_{1.95}$
6	$(U_{0.75}, Pu_{0.2}, Am_{0.03}, Np_{0.02})\ O_{1.98}$

* Fuel composition expressed in mole percent.

Planned Activities

This activity has concluded. Further activities will be defined and new collaborative activities related to post-irradiation examination of oxide- and ceramic-based nuclear fuels will be developed for future collaboration.

INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE

Lead-Cooled Fast Reactor Concept Design and Evaluation

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

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

Collaborators: Lawrence Livermore National Laboratory

Project Number: 2006-005-E

Program Area: FCR&D

Project Start Date: May 2009

Project End Date: December 2011

Research Objectives

The purpose of this project is to conduct trade studies and develop a preconceptual design for a near-term deployable lead-cooled fast reactor (LFR) demonstrator. The demonstration reactor will implement innovative engineering features that exploit the characteristics of pure lead (Pb) 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 and to enable irradiations of advanced fuels and materials. The project is focused on defining the reactor core and primary system configuration with the following objectives:

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

The demonstration reactor will incorporate innovative features from the European lead-cooled system (ELSY) concept. The preconceptual design of the facility will be flexible enough to accommodate subsequent modifications to test new components and systems and 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 minor actinide-bearing 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.

Research Progress

Researchers investigated two key thermal-hydraulic features of the 1,500-megawatt (MWt) ELSY concept, developing an assumed 400-MWt demo concept that incorporates key ELSY features. The first thermal-hydraulic feature they examined was the combined spiral-tube steam generator/main Pb pump unit installed in the primary Pb hot leg. This feature provides the following advantages:

- Enables reducing the primary Pb coolant volume.
- Allows exposing the reactor vessel to only cold Pb during normal operation, thereby enabling the vessel to operate at temperatures below 425°C where control of the dissolved oxygen level in the Pb is not necessary to avoid corrosive attack on the vessel austenitic stainless steel.
- Eliminates thermal stress loadings resulting from temperature changes during startup and shutdown.
- Preserves a closed Pb coolant flow path for heat removal from the core in the event of postulated reactor vessel failure such that the coolant height decreases to the faulted level corresponding to Pb coolant entering the gap between the guard vessel and failed reactor vessel.

For a 200-MWt steam generator/Pb pump unit, the pump requirements can be met with a mechanical axial flow pump. Researchers determined the pump impeller diameter and rotational speed. The steam generator tube bundle dimensions were optimized by minimizing the total steel mass of the many steam generator spiral tubes—which is expected to be a major cost contributor for the steam generators. The team found that when the

maximum radial Pb velocity through the spiral-tube bundle is limited to 1 m/s to preclude removal of protective oxide layers at higher velocities, the optimal steam generator dimensions are a steam generator tube bundle outer diameter of about 2.5 m and a steam generator tube bundle height of about 4 m.

Each of the T91 or SS316 spiral tubes required was calculated to be about 70 meters (230 feet) long. An important question thus concerns the feasibility of precisely bending piping of this length to form the required spiral tubes. Another important question is whether aluminizing the steam generator tube outer surfaces can be carried out to further enhance resistance to corrosive attack by Pb.

The team created a CAD (computer-aided drawing) model to demonstrate the feasibility of connecting vertical feedwater and steam piping to all of the horizontal spiral tubes without significantly occluding the radial Pb flow. The current analysis indicated that the combined spiral-tube steam generator/main axial Pb pump unit is a viable concept in that the calculated axial pump impeller dimensions and steam generator tube-bundle dimensions are compact enough for the required number of units to be installed inside the reactor vessel, which is reduced in size as a result of using these units. The maximum Pb radial velocity through the tube bundle can be limited to 1 m/s to reduce the potential for removal of protective oxide layers on the outside of the steam generator tubes, although the effects on the heat exchanger dimensions of increasing the maximum Pb velocity from 1 to 3 m/s are small.

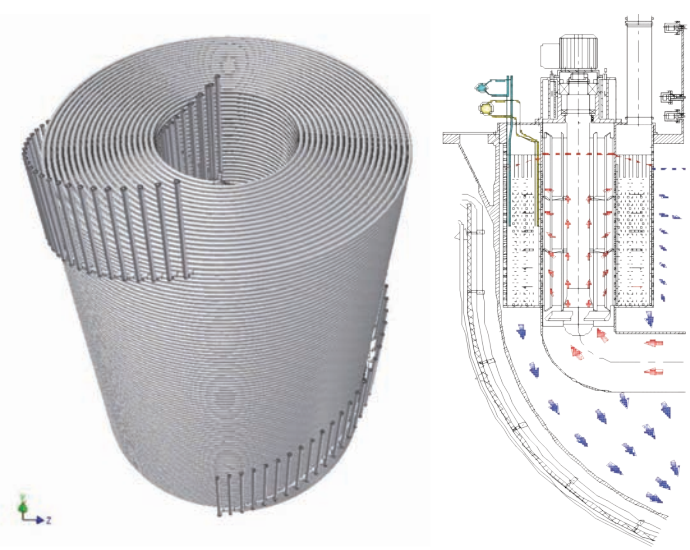


Figure 1. Spiral-tube intermediate heat exchanger tube bundle and illustration of combined spiral-tube intermediate heat exchanger and axial flow pump.

The second ELSY thermal-hydraulic feature investigated was the lack of a core support structure and inlet pressure plenum beneath the core and the open-lattice core. A core support structure below the core is not needed for structural support reasons because the fuel assemblies are buoyant in the dense Pb and may be supported from above. This enables elimination of support structure below the core which, together with elimination of below-core flow distribution structure, also reduces the required Pb coolant volume. A three-dimensional computational fluid dynamics (CFD) model was created, and calculations seeking steady-state velocity distribution solutions were run using the STAR-CCM+ computer code.

For the particular assumed core configuration, researchers installed a horizontal plate at the top of the fission gas plenum to preclude the development of recirculation flows inside the active core and to generate a mostly uniform flow field inside the ducts leading to the pump impellers. The team found crossflow within the active core to be very small, indicating that the open lower plenum configuration without an inlet pressure plenum can work in a fluid dynamics sense—provided that the horizontal plate is present at the top of the fission gas plenum and that the flows are steady.

The team calculated the pressure loss along the Pb flow through the model, which was 0.13 MPa and was consistent with the 0.15 MPa pressure drop quoted for ELSY. However, the assumed steady-state solution does not converge to a sufficient residual tolerance which, assuming that the physical modeling in STAR-CCM+ provides realistic predictions, indicates that the flow inside the lower plenum and the open-lattice core is inherently transient. This gives rise to concerns that the flow inside the core could oscillate or pulsate. Oscillations or pulsations in the flow would give rise to oscillations or pulsations in the core temperatures, which in turn might give rise to oscillations or pulsations in the core power due to the temperature-dependent reactivity feedbacks of the core.

To investigate whether such unsteady behavior can occur and to quantify its magnitude, transient three-dimensional CFD calculations of the lower plenum and open-lattice core regions should be explored. Scaled simulant experiments using scale models and a simulant fluid such as water should also be explored. For example, scale model tests utilizing water might reveal the presence of oscillations or pulsations in the flow that could be measured, making it possible to scale the results to predict the behavior in the full-size Pb system. If oscillations or pulsations of significantly large magnitude are predicted by transient three-dimensional CFD calculations or scale model experiments, the engineering solution may be to install an inlet pressure plenum providing a uniform pressure boundary condition across the bottom of the core. One

possible approach to such a plenum is to provide an elliptical flow distributor head incorporating circular flow hole perforations beneath the core. The flow distributor will incrementally increase the pressure drop around the primary coolant circuit and the required Pb coolant volume. It will need to be inspected or replaced if it is subjected to a neutron fluence that is too high. However, achieving good flow behavior in the core may require such tradeoffs.

Planned Activities

Future work will focus on mixed oxide- and metallic-fueled options for the demo core and determination of the tradeoff between core performance, including peak fast neutron flux, required fissile enrichment, and burnup versus power level and core size.

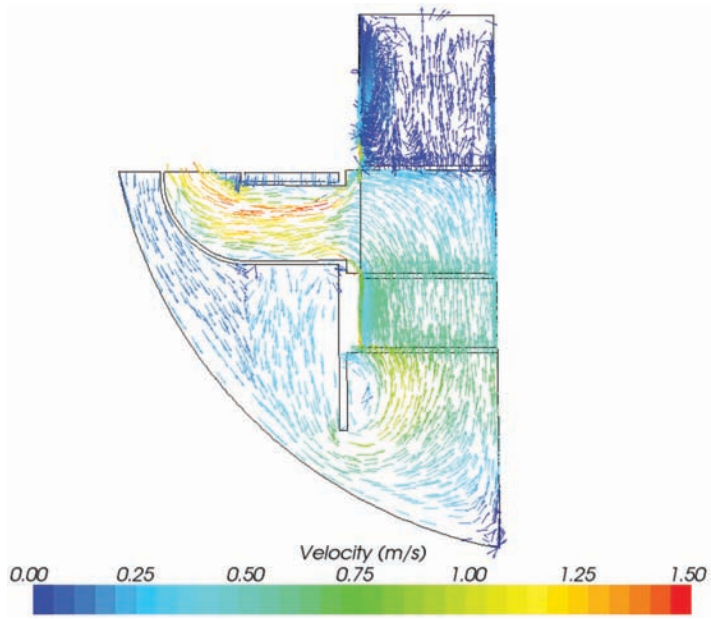


Figure 2. Calculated flow inside lower portion of cold pool, active core, fission gas plenum above core, and duct leading to axial flow pump/spiral-tube steam generator with a horizontal plate installed at the top of the fission gas plenum region.

INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE

4.3 U.S./France Collaboration

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; eighteen U.S./France collaborative research projects have been awarded since FY 2001.

Work Scope Areas

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

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

Project Summaries

Work has concluded on two collaborative projects initiated in FY 2006. Research continues on two other projects, one initiated in FY 2007 and one in FY 2008. No new collaborations with France were initiated during the past fiscal year. A listing of the I-NERI U.S./France projects follows, along with summaries of the accomplishments achieved in FY 2009.

Directory of Project Summaries

2006-001-F* Sulfur-Iodine Integrated Laboratory-Scale Experiment 49

2006-003-F* Comparison of Characterization Methods for Anisotropy and Microstructure of TRISO Particle Layers 53

2007-001-F Advanced Dispersion Strengthened Ferritic Alloys and F/M Steels with Fine Nanoscale Dispersions 55

2008-001-F SiC/SiC Composites for Gen-IV Reactors 59

* Completed in FY 2009

INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE

Sulfur-Iodine Integrated Laboratory-Scale Experiment

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

The U.S. Department of Energy (DOE) and the French CEA have investigated the sulfur-iodine (S-I) thermochemical cycle as a method of large-scale hydrogen (H_2) production for use with advanced reactors. The objective of this project was to develop an integrated laboratory-scale (ILS) experiment for the S-I cycle to investigate the key technical issues. The three major reaction sections of the S-I cycle were developed by three different organizations: CEA developed the primary (Bunsen) reaction section; General Atomics developed the H_2-generation section (hydrogen-iodide [HI] decomposition); and SNL developed and tested the sulfuric acid (H_2SO_4) decomposition section. The three sections were assembled at GA in San Diego, California, to conduct the integrated experiment. The first partially integrated tests were conducted in FY 2008 to provide information on system controls, interactions between sections, and corrosion's effects on component and catalyst performance. During FY 2009, the objectives were to complete the integrated test sequence to provide a technical basis for assessing the performance potential of this cycle.

Research Progress

Activities in FY 2009 focused on completing equipment modifications to the Bunsen section and conducting additional partially integrated testing. The Bunsen section was modified to improve iodine flow and level diagnostics. Several partially integrated tests were conducted. A sequence of tests was conducted on the high-temperature H_2SO_4 decomposer section to map sulfur dioxide (SO_2) production as a function of temperature, pressure, and flow rates. Researchers completed a series of tests on the HI decomposer section (H_2-generation stage) to define extractive distillation parameters and assess materials performance. They also conducted materials tests on tantalum (Ta) alloys used in the HI section to define temperature, pressure, and time limits for the HI section distillation vessel. During this final stage, several partially integrated runs were conducted including H_2-generation runs using the heavy acid phase materials generated in the CEA Bunsen section. Although only partially integrated tests were able to be conducted on the ILS system, these tests generated process chemistry, materials, and controls information to provide a basis for evaluating this cycle. The results of the tests conducted on each section are summarized in the following sections.

HI Decomposition Section. The HI decomposition section developed by GA for the ILS experiment is based on an extractive distillation approach that separates iodine (I_2) from the HI_x feed stream before decomposition. The basic steps involved in the extractive distillation process for HI are illustrated schematically in Figure 1(a). The HI section has been successfully operated to produce hydrogen using

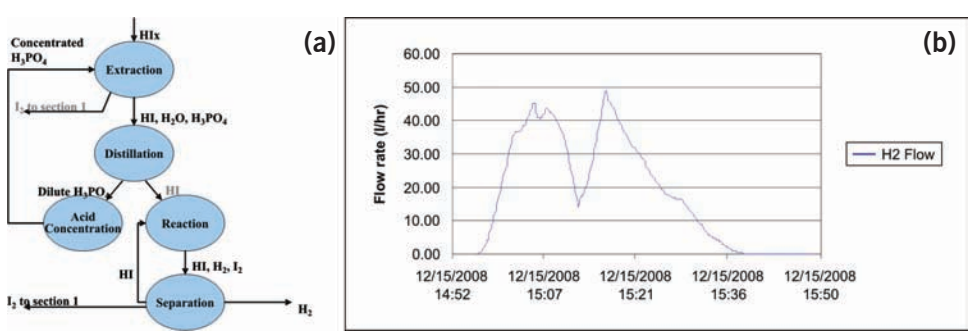


Figure 1. (a) Schematic of process sequence involved in extractive distillation and decomposition of HI; (b) H_2 production rate as a function of time using partial recycle of the under-composed HI to increase H_2 production.

the CEA Bunsen section feed material, with H_2 production of up to 50 l/hr from an operation with partial recycle, as shown in Figure 1(b).

The most important materials issue encountered in the initial ILS tests was the degradation of Ta alloy vessels in the HI distillation vessel. Ta-based materials performed well in corrosion screening tests. Ta vessels and Ta-coated components were used extensively in the HI section. The failures observed in Ta and tantalum tungsten (TaW) alloy vessels and tubing (localized, brittle fractures) were consistent with stress corrosion cracking (SSC) and H_2 embrittlement mechanisms. Both stress corrosion and H_2 embrittlement are known to occur in Ta and Ta alloys in a range of acid concentrations. These degradation mechanisms will preclude the routine use of Ta and Ta alloys as a primary boundary or pressure boundary, but they are effective as a coating or liner for other substrate materials for these applications. A Ta coating or liner provides significant corrosion resistance in the S-I environment, while the mechanical strength is provided by the Ta-protected substrate material. Ta-coated parts (on stainless steel fittings) performed well—except in those cases where damage to the coating allowed corrosion of the underlying material. The Ta alloy corrosion problems encountered in the HI distillation vessel due to hydrogen embrittlement of Ta materials placed limits on the time at temperature for the HI reaction vessel, but allowed sufficiently long runs to demonstrate the basic features of extractive distillation.

Bunsen Section. Based on initial Bunsen section runs conducted in FY 2008, modifications were made to improve iodine flow and level diagnostics. Raschig ring packing was added to improve SO_2-I_2-H_2O mixing, and differential pressure cells were installed to provide better indications of liquid levels in the reactor. With better control of I_2 flow rates, researchers conducted additional Bunsen runs with SO_2 supplied by the SNL H_2SO_4 decomposition section. In general, these initial tests produced a light H_2SO_4 phase that was too dilute, primarily because of the excess water that was necessary at startup for heat transfer reasons. Concentrations increased over the duration of the runs, but longer runs would be needed to reach equilibrium. The heavy HI acid phase produced was 57.8 wt% HI, which is on target. The separation of light and heavy acid phases was also not complete in these relatively short runs, which indicates the need for both longer runs to reach equilibrium and improved I_2 metering and control to achieve the proper phase composition and improved separation in this countercurrent design. The Bunsen section reactor was constructed using a glass-lined steel vessel, which performed well over the course of the Bunsen section runs.

Sulfuric Acid Decomposition Section. The SNL H_2SO_4 decomposer integrates the functions of acid vaporization, superheating, and catalytic decomposition to generate SO_2 in a single-tube SiC bayonet heat exchanger. After heat recuperation with the inlet acid stream, the product stream exits the bayonet at a relatively low temperature. The team ran approximately 30 experiments covering the parameter space of interest with reproducible results. No corrosion issues were identified during the relatively short-term operations. SO_2 production of up to 300 l/hr at 850°C has been achieved in a single-tube bayonet, with near-theoretical conversion at design flow rates. Figure 2 shows SO_2 production as a function of temperature and pressure. Significant SO_2 conversion can be obtained at temperatures as low as 700°C by recycling the unconverted acid. With effective heat recuperation, there is only a relatively small efficiency penalty caused by the increase in recycle. Since a significant fraction of the heat is required in the catalyst region to support the decomposition process, the achievable SO_2 production rate in the bayonet is limited by heat transfer to the catalyst region. The issues of catalyst stability will require additional study to confirm long-term performance. The product stream outlet temperature, which is an important design parameter for interfacing the acid decomposition section with the S-I or hybrid S cycles, is a function of recuperation effectiveness and system pressure. A series of experiments was run during this period to validate system design models for use in future design studies (Figure 2[b]).

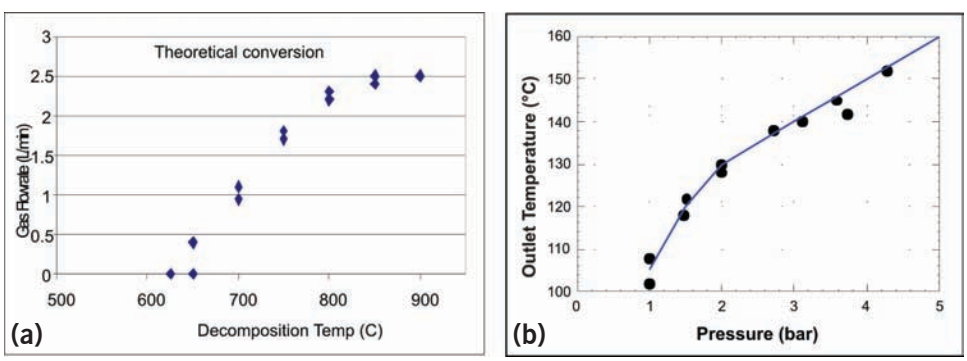


Figure 2. (a) SO_2 generation as a function of catalyst region temperature; (b) product stream outlet temperature as a function of system pressure.

Planned Activities

The DOE–CEA project on the S-I integrated laboratory-scale experiment was completed in April 2009. Documentation of the results has largely been completed, and the equipment is being cleaned and placed in safe shutdown condition until decommissioning later in FY 2010. Although the project has now been completed, several observations were noted that could be useful in future work. The small scale of these experiments constrained the options for basic components (pumps, valves, etc.) and required many components to be modified or created. Larger-scale experiments could possibly increase costs for some components but would allow a wider choice of component and material options that should reduce overall costs. The results from the HI decomposition section operations showed that extractive distillation works, but it is complex and is considered to be less energy-efficient than reactive distillation. Future work should focus on reactive distillation. Operations in the high-temperature H_2SO_4 decomposer based on the SiC bayonet heat exchanger were near-routine, with no material corrosion issues identified over the sequence of ILS runs, but the proposed multi-tube scaling approach for this concept to large sizes needs to be demonstrated.

INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE

Comparison of Characterization Methods for Anisotropy and Microstructure of TRISO Particle Layers

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

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

Collaborators: AREVA, Laboratoire des Composites Thermostructuraux, Université Bordeaux

Project Number: 2006-003-F

Program Area: Gen IV

Project Start Date: June 2007

Project End Date: September 2009

Research Objectives

The resurgence of coated particle fuel as a cornerstone technology for the next generation of nuclear reactors requires improved characterization methodology for process development and product qualification. This project consisted of two elements. Element 1 compared two systems for measuring the anisotropy of the pyrocarbon layers in tri-isotropic (TRISO) coated particle fuel. Pyrocarbon anisotropy is an important parameter related to fuel irradiation performance. High pyrocarbon anisotropy can lead to non-uniform shrinkage in the pyrocarbon layers that may result in cracking and failure of the coating layers. ORNL measured anisotropy using their newly developed, two-modulator generalized ellipsometry microscope (2-MGEM). CEA used a more traditional optical polarimeter technique called RAPAX. Element 2 investigated the use of X-ray analysis as a non-destructive technique for quality control.

Research Progress

A series of TRISO samples, relevant to past and present fuel development programs and spanning a broad range of anisotropies, has been measured using both optical instruments (i.e., 2-MGEM and RAPAX). Careful calibration using known standard materials has resulted in good agreement between the two sets of measurements. Transmission electron microscopy (TEM) analysis was also performed at the Centre d'Elaboration de Materiaux et d'Etudes Structurales (CEMES) and at Bordeaux University. The TEM analyses revealed trends in the microstructure that follow the relative levels of anisotropy determined using both optical methods.

X-ray radiographic imaging is a non-destructive technique that can obtain structural information without introducing artifacts induced by metallographic preparation. Moreover, this technique can offer three-dimensional (3D) information about layer thickness, density, interface structure, and coating defects (cracks, voids, etc.). ORNL and AREVA each utilize X-ray imaging for coated particle characterization. In addition to standard X-ray systems, ORNL has developed a new bench-top system capable of X-ray radiography and 3D tomography with a resolution down to one micrometer (μm). To shake down this new system and learn more about the relative capabilities of the various techniques, researchers examined a number of particles from each reference fuel lot. AREVA has also made great advances in the use of X-ray tomography for non-destructive determination of coating layer thickness and density. Both groups analyzed samples whose thickness and density had previously been determined by other methods and compared the results.

The team assembled a final report, which is a compilation of three summary reports. Element 1 of the collaboration involved the pyrocarbon anisotropy study. Part A of the final report for Element 1 discusses the results of the optical measurements of pyrocarbon anisotropy. The 2-MGEM and RAPAX methods are described, and data is presented and compared. Part B of the final report for Element 1 discusses the results of the TEM studies. The report describes two TEM methods, selected area electron diffraction (SAED) and dark field imaging. The same materials used for the optical analyses were studied via these TEM techniques. The TEM data is presented, and results from analyses used to quantify the observed anisotropy are compared to the optical results.

Element 2 of the collaboration involved the use of X-ray imaging to measure the coating thickness of a series of coated particle fuel samples. Some of the same samples used in the Element 1 study were used for this work. The final report for Element 2 describes the X-ray imaging methods used by AREVA and ORNL, and data is presented and compared.

Planned Activities

No further activity is planned under this project.

INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE

Advanced Dispersion Strengthened Ferritic Alloys and F/M Steels with Fine Nanoscale Dispersions

PI (U.S.): Dave Hoelzer, Oak Ridge National Laboratory, and Stuart Maloy, Los Alamos National Laboratory

PI (France): Yann de Carlan, Commissariat à l'énergie atomique (CEA), Saclay

Collaborators: None

Project Number: 2007-001-F

Program Area: FCR&D

Project Start Date: June 2008

Project End Date: June 2010

Research Objectives

Enhanced material performance is an important way to improve the economics of advanced nuclear reactor requirements by potentially allowing both 1) higher operating temperatures with enhanced thermal efficiency and power output and 2) longer lifetimes or burn-ups. The main objective of this project is to develop high-performance ferritic alloys that are strengthened by a dispersion of nano-sized particles for use in advanced high-temperature nuclear technologies such as cladding for sodium fast reactors. The key to developing these alloys will be to obtain 1) fundamental knowledge of the processing and fabrication methods for achieving the nano-sized particle dispersions in the microstructure, 2) the corresponding relationship between the microstructure and the deformation and fracture properties, and ultimately 3) the stability of the microstructure and mechanical properties during exposure to neutron irradiation. This project will focus on two ferritic alloy systems: 1) 9% chromium (9Cr) tempered martensitic steels (TMS) that are strengthened by dispersion of nano-size nitride, or nitro-carbide, particles and 2) advanced oxide dispersion-strengthened (ODS) ferritic alloys containing 13% Cr to 18% Cr and a dispersion of nano-sized oxide particles. The project will emphasize the knowledge that has been obtained with development of the 14YWT ferritic alloy at ORNL that contains a high concentration of titanium (Ti)-, yttrium (Y)-, and oxygen (O)-enriched nanoclusters.

Research Progress

The researchers on this project accomplished the following activities this past year:

- They used computational thermodynamics to investigate phase equilibria of four 9Cr TMS containing ~0.07% nitrogen (N) and variations in solute atoms and interstitial carbon (C) contents.
- The project initiated development and characterization of four experimental 9Cr TMS by applying different thermal mechanical treatments to form nano-size nitride, or nitro-carbide, dispersions.
- Researchers continued creep testing of 14YWT and used the results to update the calculated deformation map that identified the creep mechanism at 800°C.
- The team performed fracture toughness testing on 14YWT to investigate grain anisotropy's effects on the fracture behavior at low temperatures.
- The proposal for conducting ion irradiation experiments at the Joint Accelerators for Nano-Science and Nuclear Simulations (JANNuS) at CEA, Saclay, was approved.

Following is a summary of the research progress that has been made on this project to date.

9Cr TMS. The project team conducted research on four heats of 9Cr steels that contained ~0.07% N (wt%) and different concentrations of molybdenum (Mo), tungsten (W), manganese (Mn), tantalum (Ta), and C, as shown in Figure 1. Researchers performed thermodynamic calculations with Pandat software and an iron (Fe)-based database to assess the phase equilibria in the four steels. The calculations exhibited the equilibrium phase fraction as a function of temperature, which is very useful for guiding heat treatment and thermomechanical processing.

The calculations presented the mole/volume fraction and chemical composition of the expected precipitates at specific temperatures, which in summary showed the addition of Mo-promoted sigma phase formation; the addition of W-promoted Laves phase formation; the addition of Ta, which led to precipitation of TaN; and the addition of C, which caused the formation of $M_{23}C_6$ carbides. This information was helpful in guiding the microstructure analysis and interpreting the properties of the four 9Cr steels.

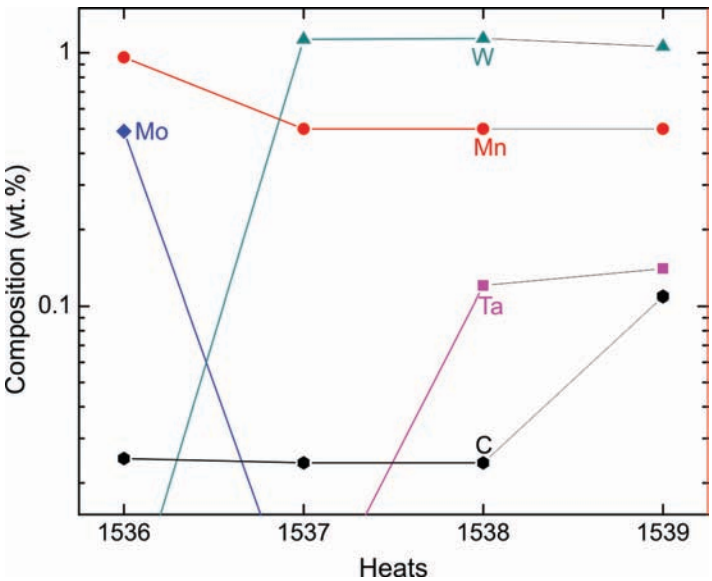


Figure 1. Chemical composition (wt%) of the alloying elements in the four heats (i.e., 1536, 1537, 1538, and 1539).

The four experimental 9Cr steels were developed using a standard heat treatment (ST) (austenitic annealing + tempering) and a thermomechanical processing treatment (TMP) (austenitic annealing + hot-rolling + tempering + cold-rolling + tempering). Optical microscopy showed that, because the four steels varied in composition, they differed in the propensity of martensite formation. Scanning electron microscopy (SEM)/energy dispersive spectroscopy (EDS) analysis confirmed the presence of Ta-rich and Cr-rich precipitates in the samples. The TMP samples showed smaller and denser distributions of precipitates compared to the ST samples. Thus, the results of the preliminary microstructural analysis are consistent with the thermodynamic calculations.

Assessment of the mechanical properties of the four 9Cr steels was obtained using Vickers microhardness (VH). In general, the TMP samples showed greater VH than ST samples, and the Ta-containing steels (1538 and 1539) showed greater VH than those without Ta (1536 and 1537). Similar VH was observed between 1536 and 1537 and between 1538 and 1539. Three general trends were observed in the data: 1) the 0.5 Mo in 1536 had a similar effect as the 1.0 W in 1537 on VH; 2) the Ta addition in 1538 and 1539 significantly increased the VH; and 3) the high fraction of $M_{23}C_6$ (as suggested by thermodynamic calculations), due to the high C content in 1539, did not show noticeable benefits to VH.

Recent modifications have been made to the ST (denoted STO) that resulted in formation of a mixture of ferrite and martensite/bainite in different volume fractions, which depended on the composition of the four 9Cr steels. The STO resulted in the highest VH and the largest variations in VH between the four heats. Interestingly, preliminary tensile tests performed on the 1536 and 1539 STO samples at room temperature showed extremely high strength for such category steels (e.g., yield strength > 0.9 GPa and ultimate tensile strength ~ 1.2 GPa).

NC-Strengthened 14YWT Ferritic Alloy. The research studies investigated the thermal creep and fracture toughness behavior of the 14YWT-SM10 heat, which was produced at ORNL in July 2007. This heat contained the smallest grain size (~136 +/-14 nm) and grain aspect ratio (GAR: length/width ~1.2-2) compared to the previous 14YWT heats produced at ORNL as well as the benchmark 12YWT and MA957 alloys.

The project developed an improved understanding of creep mechanisms for advanced NC-strengthened ferritic alloys through results obtained from the initial creep testing of 14YWT, combined with those from previous tests on 12YWT and MA957 during past projects at ORNL. The creep tests on 14YWT consisted of a constant load test at 800°C and 100 MPa that was started in April 2008 and is still in progress, as well as three new tests at 800°C using constant stresses of 200, 250, and 300 MPa. Figure 2 shows a plot showing the minimum creep rates (MCR) versus stress for the three constant stress tests on 14YWT plus those obtained from previous tests on 12YWT and MA957. The creep test on MA957 at 800°C and 100

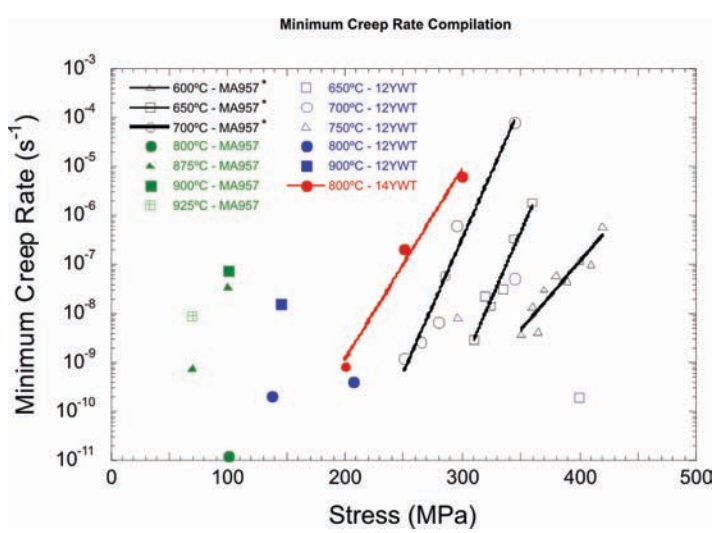


Figure 2. Minimum creep rates obtained from creep tests on 14YWT, 12YWT, and MA957.

MPa demonstrates the outstanding creep properties of the advanced NC-strengthened ferritic alloys. This test lasted 38,555 h (~4.4 years) with only ~0.361% strain occurring before the specimen failed, and an MCR of ~1.2 x 10^{-11} s^{-1} was the lowest value recorded. In addition, the exponential curve fit to the three data points for 14YWT indicates that stress exponent (n) and activation energy (Q) for creep at 800°C is similar to published values of n~35 and Q~815 kJ/mol obtained for creep tests on MA957 between 600°C and 700°C.[1] These high values are consistent with the threshold stress mechanism for creep at high temperatures.

The calculated deformation map for 14YWT, shown in Figure 3, was updated using the new creep results. The initial deformation map had been calculated using data from high strain rate (10^{-3} and 10^{-5} s^{-1}) tensile tests conducted in studies using earlier 14YWT heats. The deformation map showed very good agreement between the MCR values and the strain rates calculated using the back stress concept of the thermally activated dislocation detachment model, indicating that the dislocation creep mechanism is dominant for creep at 800°C. Interestingly, these calculations also showed that diffusional creep mechanisms overestimated the creep rate. However, further experiments covering a larger range of temperatures and stresses are required in order to more thoroughly explain the creep behavior of 14YWT since grain boundary, lattice, and pipe diffusion will likely have an effect due to the ultra-fine grain size and NC dispersion of this alloy.

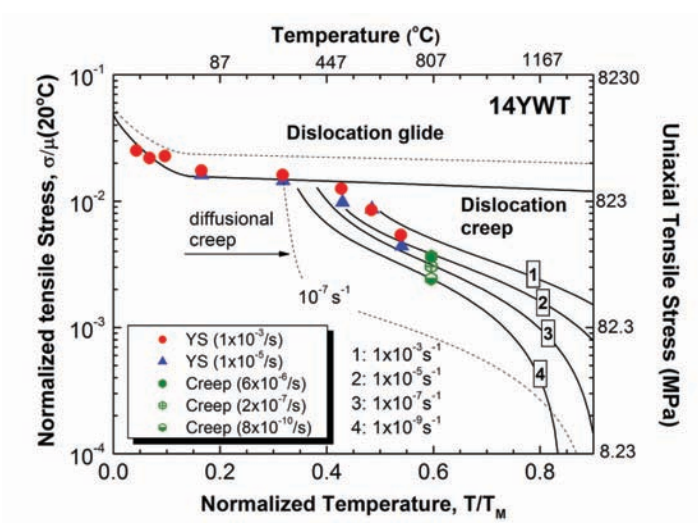


Figure 3. The calculated deformation map of 14YWT using data from tensile and creep tests.

Surprisingly, the fracture toughness results indicated that reducing the grain size and anisotropy (GAR) in 14YWT-SM10 did not have a beneficial effect on fracture toughness behavior. Compared to results obtained from a previous heat of 14YWT (SM6), the fracture toughness transition temperature (FTTT) for 14YWT-SM10 was much higher (-84°C) compared to that of 14YWT-SM6 (-150°C). In both cases, the fracture toughness was measured in the toughest L-T orientation, when the crack propagates in the direction that is transverse to the extrusion orientation. These results are shown in Figure 4 along with the fracture toughness results obtained in a previous project for 12YWT (also tested in the L-T orientation). The comparison shows that although the FTTT for 14YWT-SM10 was higher than that of 14YWT-SM6, it is still significantly lower than that measured for 12YWT, which possesses a larger grain size and higher GAR than the 14YWT heats.

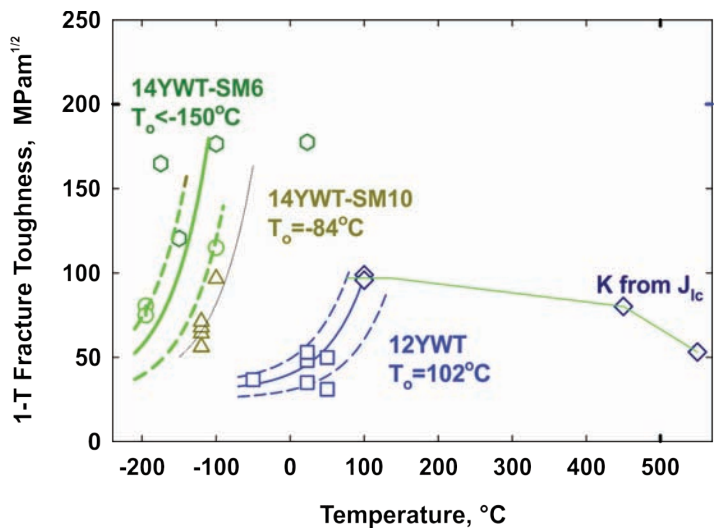


Figure 4. Fracture toughness of the two heats of 14YWT and 12YWT in the L-T orientation.

The fracture toughness of 14YWT-SM10 was also investigated in the weakest T-L orientation, when the crack propagates along the extrusion direction. Preliminary results indicated that the FTTT in the T-L orientation was ~18°C, which was much higher than that of the L-T orientation. In addition, the fracture toughness on the upper shelf was only ~60 MPa/m^2. These values are closer to those of 12YWT, which was tested with the more favorable L-T orientation. Thus, these results indicate that reducing the grain size to the nano-sized length scale is not the only factor in establishing good fracture toughness properties of NC-strengthened ferritic alloys.

Planned Activities

Researchers plan to conduct the following activities in FY 2010:

- Perform tensile tests on the 9Cr steels at room temperature and up to 650°C.
- Investigate the microstructure of the 9Cr steels using TEM and electron backscatter diffraction to correlate with the properties.
- Start long-term heat treatments (e.g., 10,000 hours aging at 650°C) and creep tests on the 9Cr steels to investigate the stability of their microstructures.
- Continue the investigation of creep mechanisms with further creep tests on 14YWT.
- Perform additional fracture toughness tests on 14YWT in the T-L orientation to help determine the source of statistical scatter in the data.
- Conduct fracture toughness tests on 14YWT at elevated temperatures up to 600°C.
- Conduct the first set of ion irradiation experiments on 14YWT at the JANNuS facility.

References

[1] *Materials Science and Engineering*: A, 386 (2004), 81.

INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE

SiC/SiC Composites for Gen-IV Reactors

PI (U.S.): Y. Katoh, Oak Ridge National Laboratory

PI (France): C. Colin, Commissariat à l'énergie atomique (CEA), DEN/DMN

Collaborators: Idaho National Laboratory

Project Number: 2008-001-F

Program Area: Gen IV

Project Start Date: April 2009

Project End Date: April 2012

Research Objectives

Ceramic composites are considered advanced materials for use in Gen IV nuclear systems at temperatures beyond the operating limits for metallic heat-resistant alloys. Two classes of ceramic composites are primary candidates for Gen IV applications: carbon fiber-reinforced carbon matrix (C/C) and silicon carbide fiber-reinforced silicon carbide matrix (SiC/SiC). C/C is appropriate for low neutron loading applications, mostly for the outside core in very high-temperature reactors (VHTRs); while SiC/SiC is required for high neutron loading and fast flux, some portions of VHTR control rods, and most core internals in gas-cooled fast reactors (GFRs).

The objective of this I-NERI collaboration is to address some of the most critical design and qualification issues for SiC/SiC composite components in VHTRs and GFRs: 1) to develop a reliable predictive capability of component strength and operational lifetime in a complex environment and 2) to analyze relations of the components' mechanical behavior in different geometries.

Research Progress

During FY 2009, the project team made significant progress: 1) characterizing the elastic behaviors of two kinds of SiC fibers (Hi-Nicalon™ Type-S [HNLS] and Tyranno™ SA3 [TSA3]) from room temperature to 1800°C; 2) developing the model of the first scale transition of the composites' elastic behaviors; and 3) developing a test technique for tensile properties of a uni-directional, single-fiber tow SiC/SiC composite.

Mechanical Behavior of Advanced SiC Fibers.
In collaboration with Laboratoire des Composites Thermostructuraux (LCTS), CEA developed a specific device (MecaSiC) for tensile tests at high temperature on single SiC fibers. Fibers with a gauge length of 25 mm could be tested in a secondary vacuum from room temperature to 1800°C. Researchers examined HNLS and TSA3 and estimated the elastic and electric behavior. The main results are given below:

- Both fibers' electrical resistivity exhibited the same sigmoid trend with regard to temperature, revealing an important decrease as the temperature rose to 1800°C. The electrical resistivity displayed irreversible behavior. Two different regimes of conduction could be evidenced.
- Elastic modulus also exhibited the same trend versus temperature for both fibers. The elastic modulus was quite constant below 1300°C but greatly decreased at higher temperatures. An empirical model was proposed to describe this evolution in the temperature range from room temperature to 1800°C, as shown in Figure 1.

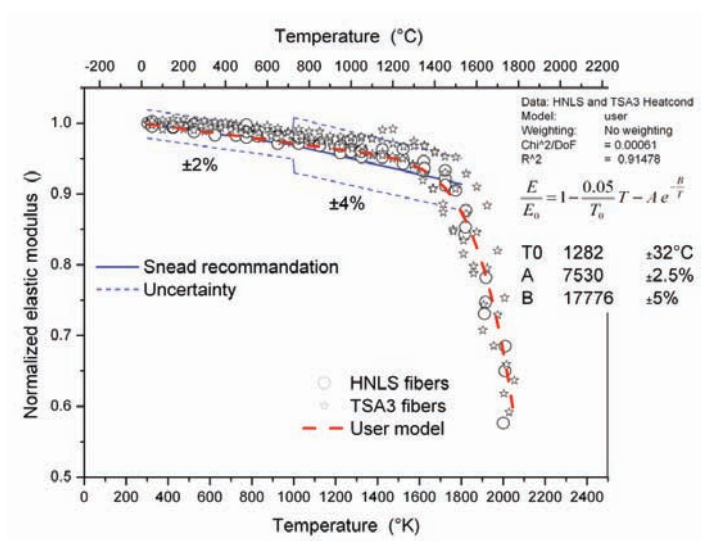


Figure 1. Elastic modulus thermal behavior of HNLS and TSA3 fibers.

Modeling Composite Elastic Behavior. Researchers performed a significant characterization of the microstructure based on the analysis of field emission gun scanning electron microscope images of several tows within a section of a woven SiC_f/SiC_m composite. Those characteristics are the foundation of the random generation of a virtual microstructure representative of the real material. Researchers used a numerical procedure to simulate a high number of volume elements (VEs) to deal with the representative volume element (RVE) question.

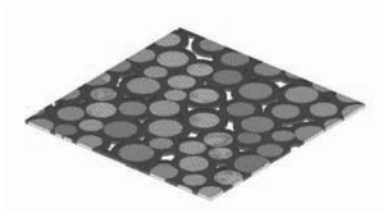


Figure 2. Example of a meshed VE.

The elastic problem of homogenization has been solved with mechanical calculations using a finite element code. Different kinds of boundary conditions have been applied on these porous random microstructures to compute the apparent stiffness tensor. The numerical results concerning four sizes of VEs led to the following conclusions:

- The apparent behavior is too sensitive to statistical variations if the VEs are too small, but the variations can be ignored only for overly large VEs. If the RVE is large compared to the tow, this microstructure does not satisfy the separability of the scales criterion; as a consequence, a homogeneous equivalent behavior cannot be strictly defined for the tow.
- In a first approximation, fluctuations are ignored, and researchers propose to evalulate homogeneous equivalent behavior by averaging the apparent behavior of several statistical volume elements (SVEs)—smaller than the RVE. An average of 5 SVEs, each comprising about 150 fibers, provides a satisfactory estimate of the homogeneous behavior thanks to its relative lacking dependence on the boundary conditions and the SVE size.
- The specific morphology of porosity is responsible for making the mechanical properties soft compared to the equivalent microstructure containing circular pores. This softening increases with variations of volume fraction of porosity and is not negligible at the scale of the tow. Finally, separability of scales issues are not only due to the heterogeneous voids distribution but also to the specific morphology of the pores. An increase of volume fraction of porosity also causes fluctuations in the apparent behavior, and differences between boundary conditions increase in importance.

Test Development for Uni-Directional Composite Mechanical Properties. Researchers developed an adequate test technique for evaluating the tensile properties of the uni-directional, single-fiber tow composites. This test is also adequate for determining the constituent mechanical properties that are useful in analyzing mechanical properties of the composite materials with more complex architectures. This test uses a model composite specimen consisting of single-fiber tow and the reinforced matrix. Moreover, the technique will be useful for future studies on environmental effects, including irradiation effects.

The test setup, shown in Figure 3, consists of an electromechanical load frame with a load capacity of 10 kilonewtons (kN), a pair of linear variable differential transformers (LVDT) for strain measurement, and the alignment fixture. The team adapted an alignment system typically used for single-fiber tensile tests (specified in ASTM C1557). A pair of aluminum tabs for gripping was attached to both ends of a composite sample. A V-notch shape fixture was prepared specifically for alignment of the tabs, as shown in Figure 4. The team effortlessly removed the specimens after testing using a polyethylene sheet and a silicone-based removal agent.

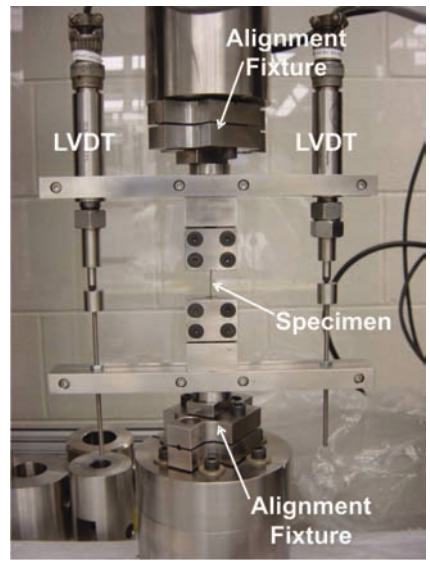


Figure 3. Single-fiber tow composite test setup developed in this study.

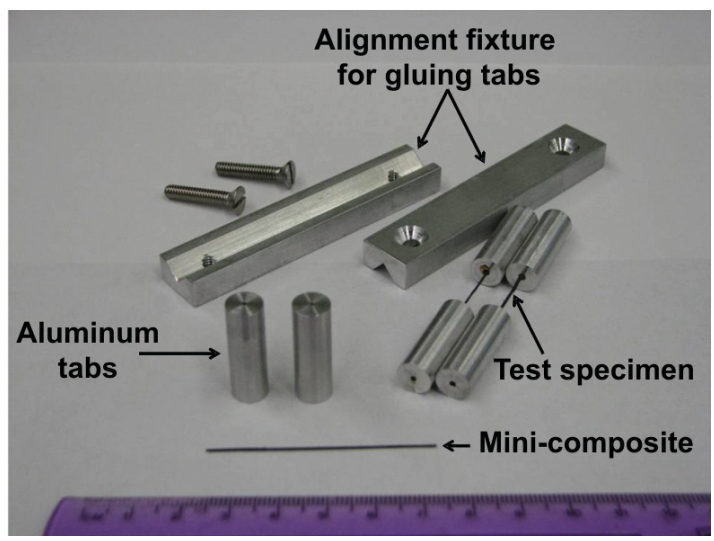


Figure 4. Composite sample with aluminum tabs attached and alignment fixture.

The basic test procedure is in accordance with ASTM C1275 for axial tensile testing of ceramic matrix composites at ambient temperature. In order to evaluate interfacial properties, the team may incorporate cyclic unloading/reloading. Researchers determined the fiber volume fraction (f) of the composites using a combination of optical microscopy and image analysis. The composite tensile stress (σ) is defined by $\sigma = P/(\pi r_f^2 N/f)$, with P being the load, r_f the average fiber radius, and N the number of filaments in a tow. The tensile properties were successfully evaluated for single-fiber tow composites with several different reinforcements. Figure 5 provides an example of the HNLS composite.

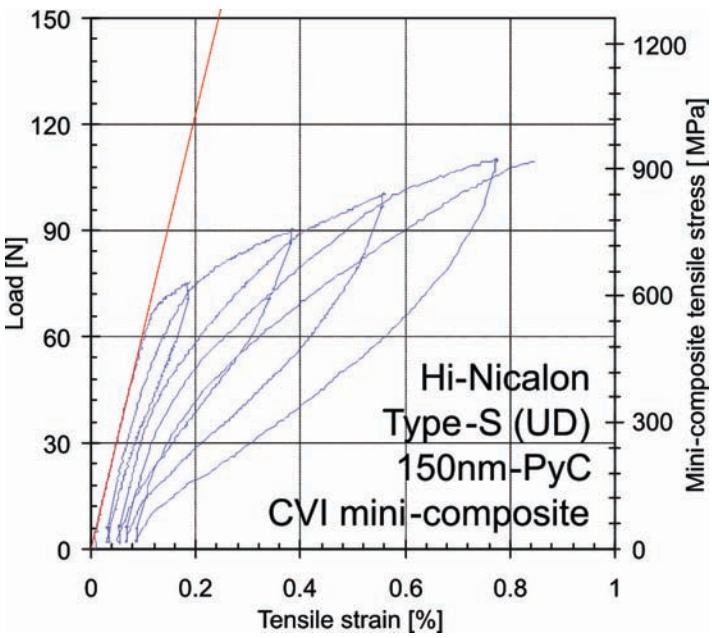


Figure 5. Example tensile load-strain behavior of HNLS composite.

Planned Activities

The team will continue the fiber mechanical properties evaluation, specifically emphasizing the plastic deformation at temperatures beyond ~1450°C. The team will also initiate mechanical properties evaluation of ion-irradiated fibers.

- The project will develop an alternative algorithm for the composite elastic properties modeling based on a rearrangement procedure to generate microstructures with higher surface fractions of fibers. Researchers need to verify their conclusions through stress and strain heterogeneity studies before applying the procedure to woven composite materials, at which time researchers will examine fluctuations and effects of volume fraction of porosity.
- The composite modeling study will be extended from linear elastic behavior to non-linear damage behavior. The team will use both experiment and modeling to investigate multiple cracking on single-fiber tow composites.
- The team will initiate evaluation of the mechanical properties of SiC/SiC test articles in a tubular geometry and then compare these properties with those of composites in uni-directional and two-dimensional architectures.

INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE

4.4 U.S./Japan Collaboration

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

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, DOE-NE's Deputy Director for Technology, and Mr. Shigeru Maeda, ANRE/METI's Nuclear Policy Division Director for Nuclear Energy Policy.

On February 8, 2005, the United States Department of Energy and Japan's MEXT signed the implementing arrangement concerning cooperation in the field of R&D of innovative nuclear energy technologies for I-NERI.

Work Scope Areas

Following are the R&D topical areas for the U.S./Japan collaboration with ANRE:

- Supercritical water-cooled reactor
- Innovative light-water technologies
- Oxide fuel processing for light-water reactors
- Fuel technologies using solvent extraction
- Radioactive waste processing

Following are the R&D topical areas for the U.S./Japan collaboration with MEXT:

- Innovative nuclear reactor technologies
- Innovative processing technologies
- Innovative fuel technologies using solvent extraction

Project Summary

Work was completed on the last U.S./Japan I-NERI project, which was awarded early 2006. A summary of the accomplishments achieved in FY 2009 follows. No new collaborations with Japan were initiated during the past fiscal year.

Directory of Project Summaries

2006-001-J* Assessment of Irradiation Performance of Zirconium Carbide (ZrC) TRISO Fuel Particles 65

* Completed in FY 2009

INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE

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

This project consists of the following tasks: 1) conduct post-irradiation examinations (PIEs) of zone-refined, high-purity ZrC samples to understand baseline irradiation effects, 2) characterize mechanical properties of unirradiated developmental ZrC coatings, 3) perform high-temperature neutron irradiation of ZrC-coated surrogate fuel particles and conduct a PIE, 4) develop fuel performance models and study the thermomechanical response of ZrC-coated particles, and 5) examine chemical interactions between ZrC and key fission products to identify potential degradation.

Research Progress

TEM Observation of Non-Irradiated ZrC Layers. Researchers observed surrogate samples for neutron irradiation with transmission electron microscopy (TEM). The ZrC coating layer has been deposited on a feed particle that is spherical inner pyrocarbon (IPyC)-coated stabilized ZrO_2 having an average diameter of around 0.8 mm. The surface of the feed particle is pyrolytic carbon (PyC) because the ZrC layer needs to be coated on the PyC in a practical process for the fuel particle. The ZrC-coated particles were annealed at about 1700°C for one hour to evaluate the effects of the fuel compact sintering process on microstructure evolution in the ZrC layers. The basic data of the batches are shown in Table 1.

Table 1. Basic data of the specimen.

Batch number (before heat treatment)	ZrC thickness (mm)	Nominal deposition temperature (K)	C/Zr ratio*1 (evaluated before heat treatment)	Density [g/cc]*2 (evaluated before heat treatment)
ZrC-06-2022	22	1632	1.11	6.45

*1: Evaluated by infrared light absorption during combustion in oxygen and by inductively coupled plasma-atomic emission spectrometry (ICP-AES).

*2: Evaluated by gas pycnometry.

Figures 1(a) and (b) show high-angle annular dark field (HAADF) images including the surface of a ZrC layer of a TEM specimen before and after heat treatment, respectively. The dark areas correspond to voids or free-carbon regions. Before the heat treatment, the voids or free-carbon regions had a thin (~10 nm) layered feature and were distributed like lines (Figure 1[a]). After the heat treatment, the voids or free-carbon regions changed, developing clod-like features with average sizes of 50 to 100 nm (Figure 1[b]). They were distributed not randomly but roughly like lines, although their intervals increased after heat treatment. Considerable crystal grain growth due to the heat treatment has occurred. The average grain size after the heat treatment is different along the deposition direction, where the average size was approximately the same as the above-mentioned interval between the lines

of the clods of the free carbons or voids (Figure 1[b]). The average grain size was found to be largest at the ZrC layer's surface area (Figure 1[b]).

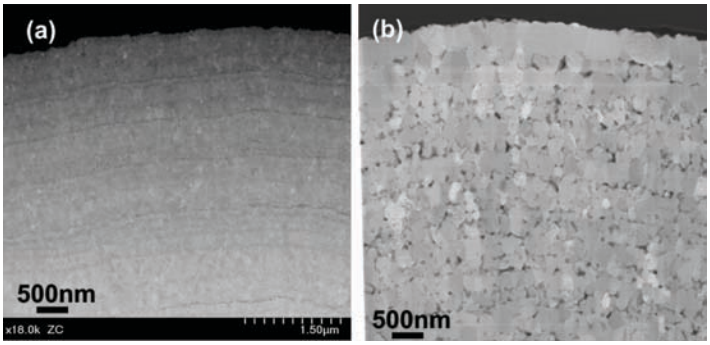


Figure 1. HAADF images of the cross section of the coating layer including the ZrC layer's surface (a) before heat treatment and (b) after heat treatment.

Neutron Irradiation of Surrogate Coated Particles. Researchers successfully completed neutron irradiation of surrogate particle samples coated with ZrC in six different coating/heat treatment conditions in the High Flux Isotope Reactor (HFIR) at ORNL. The team used capsules newly designed in the present program for neutron irradiation of these tiny particles at very high temperatures. These capsules were designed to hold approximately 50 particles, each of eight different lots, and to be accommodated in fixed rabbit tubes to be irradiated in the HFIR's flux trap. Three capsules were constructed and irradiated to 1) 2 x 10^{25} n/m^2 (E > 0.1 MeV, same hereafter) at 800°C, 2) 2 x 10^{25} n/m^2 at 1250°C, and 3) 6 x 10^{25} n/m^2 at 1250°C. The irradiated capsules were disassembled at the Irradiated Fuels and Materials Examination Facility at ORNL. Measurements showed a large increase in activity, which was attributed to Zr and Nb activation products, allowing only a small number of samples from each of the eight samples lots to be transferred for PIE in the Low Activation Materials Development and Analysis Laboratory (LAMDA) at ORNL.

Post-Irradiation Examination of Surrogate Coated Particles. The samples were ultrasonically cleaned and mounted in epoxy resin for further examination in LAMDA. The samples were metallographically prepared by grinding and polishing. The prepared samples were examined using a field emission gun scanning electron microscope available at ORNL.

Figures 2 (a–d) show the microstructure of heat-treated (1800°C for one hour) unirradiated and irradiated particles containing stoichiometric ZrC coating. In irradiated particles, the microstructural observations revealed significant grain coarsening due to irradiation, which in turn may suggest significant change in thermomechanical properties of the coating, warranting further detailed mechanical testing. Similar coarsening of the grain was observed in hyper-stoichiometric ZrC (C/Zr = 1.4), but to a lesser extent. No significant damage to the coatings was observed in the irradiated particles. Based on previous observation of irradiation effects on hot-pressed ZrC, it may be logical to expect minute defects such as frank and other prismatic loops; however, the overall damage to the microstructure in the case of CVD ZrC was minimal. Still, current available literature on the irradiation effects on CVD ZrC is lacking, so it may be premature to assume thermophysical and thermomechanical effects in CVD ZrC similar to previously studied hot-pressed material—especially with the former differing in its initial structure and fabrication conditions. This can only be determined by further detailed evaluation.

A comparison of the irradiated specimens indicated that the samples irradiated at six dpa had suffered less microstructural damage than the two-dpa samples. This could be attributed in part to the induced microstructural damage during handling of the two-dpa samples during capsule loading and unloading in the hot cells throughout the course of the irradiation experiment.

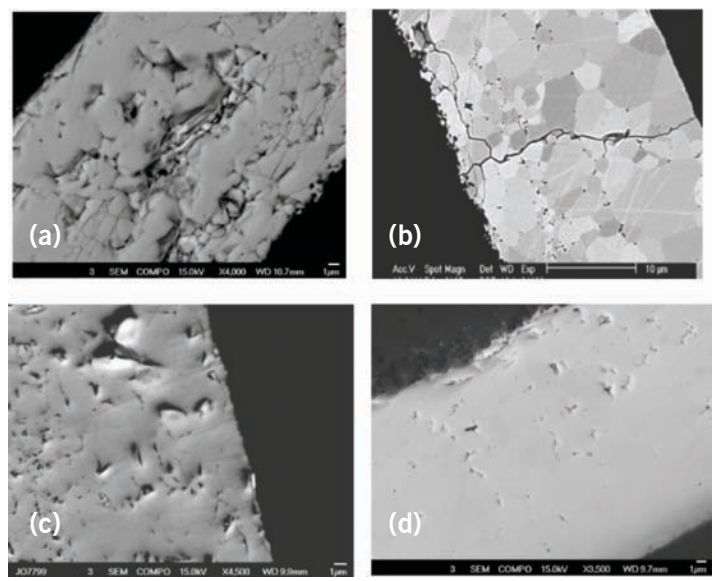


Figure 2. Heat-treated stoichiometric ZrC (a) unirradiated (b) irradiated to 2 dpa at 800°C; (c) irradiated at 1250°C to 2 dpa; (d) irradiated at 1250°C to 6 dpa.

Planned Activities

Researchers plan to accomplish the following activities during FY 2010:

- Complete post-irradiation characterization of the surrogate coated particles.
- Continue modeling and code development for fuel performance evaluation.

INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE

4.5 U.S./Republic of Korea Collaboration

William D. Magwood IV, Director of DOE-NE, signed the first bilateral I-NERI agreement on May 16, 2001, with Dr. Chung Won Cho, Director General of the Republic 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 35 projects awarded to date.

Work Scope Areas

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

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

2008 (3 projects):

- Advanced computational methods R&D
- Thermal-hydraulics
- Nuclear data/safeguards

2009 (2 projects):

- Physics data
- Advanced cladding and structural materials

Project Summaries

Three FY 2006 projects were completed during the past fiscal year. Research continued on ten other collaborative projects awarded to Republic of Korea partners from FY 2006 through FY 2008. Two new projects were awarded during the past year. In addition, Project 2007-005-K, researching small sodium fast reactors, terminated early.

Following is a listing of I-NERI U.S./Republic of Korea projects—those that are currently under way, those completed last year, and those newly awarded—along with summaries of FY 2009 accomplishments and abstracts of the new projects.

Directory of Project Summaries

2006-001-K* Core Design Studies for Sodium-Cooled TRU Burner Reactors 69

2006-002-K* Separation of Fission Products from Molten LiCl-KCl Salt Used for Electrorefining of Metal Fuels 73

2006-003-K Development of Crosscutting Materials for the Electrochemical Reduction of Actinide Oxides Used in Advanced Fast Burner Reactors....77

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2007-001-K Experimental Validation of Stratified Flow Phenomena, Graphite Oxidation, and Mitigation Strategies of Air Ingress Accidents....83

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2007-003-K Performance Evaluation of TRU-Bearing Metal Fuel for Sodium Fast Reactors to Achieve High Burnup Goal 89

2007-004-K Development and Characterization of New High-Level Waste Forms for Achieving Waste Minimization from Pyroprocessing 93

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2009-002-K Enhanced Radiation Resistance Through Interface Modification of Nano-Structured Steels for Gen IV In-Core Applications....113

* Completed in FY 2009

INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE

Core Design Studies for Sodium-Cooled TRU Burner Reactors

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

PI (ROK): Young-il Kim, Korea Atomic Energy Research Institute (KAERI)

Collaborators: None

Project Number: 2006-001-K

Program Area: FCR&D

Project Start Date: October 2006

Project End Date: September 2009

Research Objectives

The objective of this project was to 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. Specifically, the objectives were to 1) develop the core designs for TRU burner reactors, 2) perform verification and validation analyses, and 3) evaluate performance enhancements provided by innovative safety design features.

In the burner core design task, researchers examined various concepts to enhance the TRU transmutation rate under practical design constraints and developed optimum core designs for different conversion ratios and power levels. Researchers also investigated design options to minimize burnup reactivity loss and improve reactivity feedback coefficients.

The design method verification and validation task consisted of high-fidelity simulations and analyses of fast critical experiments. In particular, the team performed detailed benchmark analyses for the ZPPR-15, ZPPR-21, BFS-73-1, and BFS-75-1 critical experiments. Researchers analyzed the sensitivities of cross sections and computational methods on neutronics performance parameters and evaluated the uncertainties of performance parameters.

To evaluate safety performance enhancements, researchers analyzed 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 also demonstrated enhancements to safety margins provided by advanced modeling techniques.

Research Progress

Researchers developed metal and oxide fuel core concepts of an advanced burner reactor (ABR) at a reference power level of 1,000 megawatts (MWt). Using ternary metal and mixed oxide fuels, the team developed compact core concepts of medium TRU conversion ratio through a trade-off between the burnup reactivity loss and the TRU conversion ratio. Researchers used the same radial core layout (see Figure 1) and assembly dimensions for both metal and oxide fuel core concepts in order to allow the interchange between metal and oxide fuel assemblies. The resulting TRU conversion ratio for the recycled core was ~0.7, with corresponding burnup reactivity loss of ~2.0 percent Δk over a one-year cycle length. Calculated kinetics parameters and reactivity coefficients were adequate to provide sufficient negative reactivity feedback, and the control systems provided more than adequate shutdown margins.

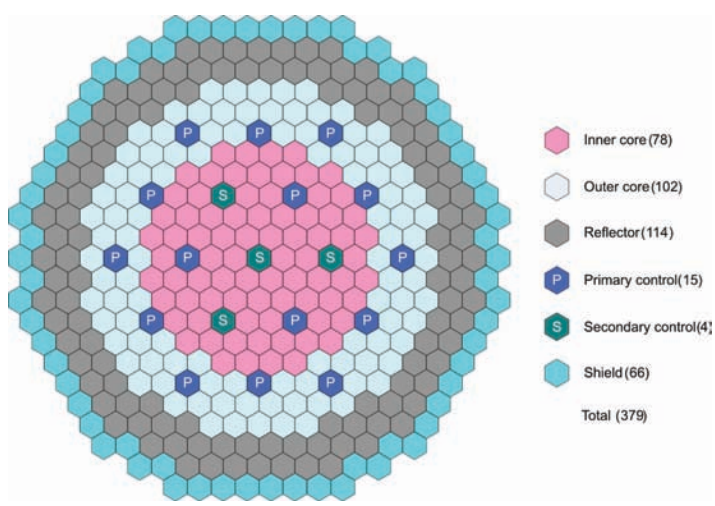


Figure 1. Radial core layout of the 1,000 MWt ABR concept.

In order to assess the impact of power on core performance and on safety characteristics, researchers developed core concepts for upgraded power levels of 2,000 MWt and 3,000 MWt, iteratively determining core size to yield power density and sodium void worth similar to those of the reference 1,000-MWt cores. The core performance parameters are generally comparable to those of the 1,000-MWt core, but the reduced core height to decrease the sodium void worth resulted in increased linear power density, even though the peak values remained within limits. The reactor kinetics parameters and reactivity feedback coefficients were comparable to those of the 1,000-MWt core.

To evaluate safety performance enhancements, the research team analyzed the performance characteristics of the ABR 1,000-MWt design, focusing on the ability to provide inherent protection against damaging consequences in low-probability accident sequences involving multiple equipment failures. 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 (ULOF) 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 ULOF 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 2,000-MWt and 3,000-MWt ABR concepts using the quasi-static reactivity balance method. The result indicated that the trends of the passive safety features are similar to the reference 1,000-MWt ABR core concepts; that is, the metal core concept has favorable passive safety features, while design fixes are required for the oxide core to meet the sufficient passive safety conditions.

The research team also performed a core design study for large monolithic reactors to identify the most limiting factor in scaling up transmutation core concepts. For this study, they adopted ternary metal fuel and employed a single enrichment approach to minimize the variation in power distribution over a burn cycle. They examined a wide range of design approaches for flatting the power distribution and enhancing the negative reactivity feedback, including variable cladding thickness, region-dependent fuel smeared density, and annular fuel pin designs with void or non-fuel materials in the center. By discovering appropriate design solutions to prevent the deterioration of reactivity feedback due to increased power level and reduced conversion ratio, researchers developed preliminary metal core designs of TRU conversion ratios of 0.75, 0.57, and 0.44 for each of three different power levels: 600 megawatts electric (MWe), 1,200 MWe, and 1,800 MWe. Researchers also performed an engineering feasibility study of fabrication and construction and developed pre-conceptual reactor layouts. They adopted passive decay heat removal circuits (PDHCs) to remove the decay heats passively for large monolithic reactors. As an example, the layout of a 1,800-MWe reactor is shown in Figure 2.

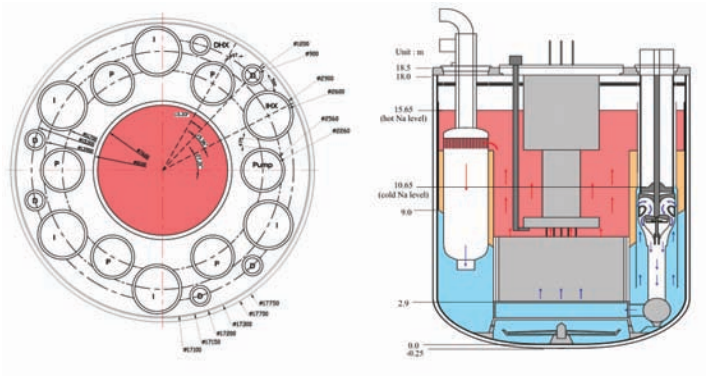


Figure 2. Pre-conceptual layout of a 1,800-MWe reactor.

The researchers performed safety analyses for these TRU burner reactor designs. The analysis results showed that these metallic fuel core designs had sufficient safety margins to coolant boiling and fuel damage in ULOF, unprotected loss-of-heat sink (ULOHS), and unprotected overpower transient (UTOP) accident sequences. The self-regulation of power without scram is mainly due to the inherent negative reactivity feedbacks in conjunction with the large thermal inertia of the primary heat transport system and the passive decay heat removal capability. The maximum allowable UTOP initiator was estimated to be about $1.07. It was also observed that the inherent safety features of the TRU burner reactors are highly dependent on the reactivity feedback coefficients, indicating that the calculated values should be validated with a sufficient confidence level.

In an effort to verify and validate the design analysis methods, the researchers performed detailed benchmark analyses for the ZPPR-21, ZPPR-15, BFS-73-1, BFS-75-1, and BFS-55-1 critical experiments. They analyzed criticality and various reaction rate ratios of major and minor actinide isotopes. They also investigated the impacts

of different evaluated cross-section libraries using the ENDF/B-VI, ENDF/B-VII, JEFF-3.1, JENDL-3.3, and JENDL-AC2008 libraries through high-fidelity Monte Carlo simulations. The effective multiplication factors calculated with the cross sections generated with the MC^2-3 code of ANL using the ENDF/B-VII data showed very good agreement with MCNP5 or VIM Monte Carlo solutions; within 141 pcm for six phases of ZPPR-21, within 46 pcm for three loadings of ZPPR-15A, and within 197 pcm for three BFS benchmark problems. No indication of systematic bias was observed. The analysis of ZPPR-15A experiments also showed that the cell-averaged cross sections generated with the new MC^2-3 code improved the criticality solutions by ~150 pcm, compared to those generated with the previous MC^2-2/SDX code system. The cross-section generation procedure of KAERI based on the NJOY and TRANSX codes showed good accuracies for homogenized benchmark problems, although it has a limited capability in reproducing local heterogeneity effects.

The comparison study of different evaluated cross-section libraries for analyzing metallic fuel-loaded critical experiments revealed that 1) uranium (U)-235 capture cross section was overestimated in JENDL-3.3, which was partially corrected in the new release of JENDL-AC2008; 2) zirconium capture cross section was underestimated in ENDF/B-VI; 3) U-238 inelastic scattering cross section was the main contributor to the observed variations in the multiplication factors and associated uncertainties of the three BFS critical assemblies calculated with different cross-section files; and 4) the revised sodium-23 scattering cross section of JEFF-3.1 improved the prediction accuracy of sodium void reactivity worth. This suggests that a combined set of basic cross-section data libraries would be useful in improving the prediction accuracy of various design parameters, although more extensive analyses are required to confirm the qualities of individual cross sections.

Planned Activities

This project has been completed as planned.

INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE

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

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

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

Collaborators: None

Project Number: 2006-002-K

Program Area: FCR&D

Project Start Date: January 2007

Project End Date: September 2009

Research Objectives

The objective of this INL/KAERI collaborative project was to develop processes for selectively removing fission products from lithium chloride–potassium chloride (LiCl-KCl) eutectic salts used in electrochemical processing of spent nuclear fuel. Separating fission products from the salt is critical for minimizing high-level waste generated during electrorefining of spent fuel. The current technology for disposing of fission products from the electrorefiner (ER) is simply to discard the LiCl-KCl salt along with the fission products as 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 ER salt. Separating accumulated fission products from ER salt would also allow the salt to be reused.

Two basic approaches have been studied during this project for selectively removing fission products from ER salt: 1) salt-zeolite ion exchange and 2) selective precipitation of fission products followed by salt purification by zone freezing and salt distillation. These methods were selected as the most promising technologies determined during the second year (FY 2008) of this project. Completing this research initiative in FY 2009 involved first demonstrating several separation methods using ER salt containing fission products from the treatment of spent nuclear fuel. Researchers then used these facility demonstrations and the data obtained from the experiments to assess and compare the separation options.

Project Conclusions

In the final year of this I-NERI project, the KAERI and INL research groups investigated the following four separation options:

1) Option A: ion exchange with zeolite-A to remove all radionuclides.
2) Option B: oxidative precipitation for rare earths followed by ion exchange with zeolite-A for Groups I and II radionuclides.
3) Option C: oxidative precipitation for rare earths followed by ion exchange with analcime for cesium (and possibly other Group I radionuclides) followed by zeolite-A for Group II radionuclides.
4) Option D: oxidative precipitation for rare earths followed by zone-freezing for Groups I and II radionuclides.

These options are illustrated in Figure 1; their functionality is summarized in Table 1. Options B and C are modifications of Option A, with oxygen sparging used to first remove rare earths. As oxygen sparging is extremely effective in removing rare earths, these modifications can be beneficial when the cleaned salt stream must have an extremely low rare earth concentration. But since

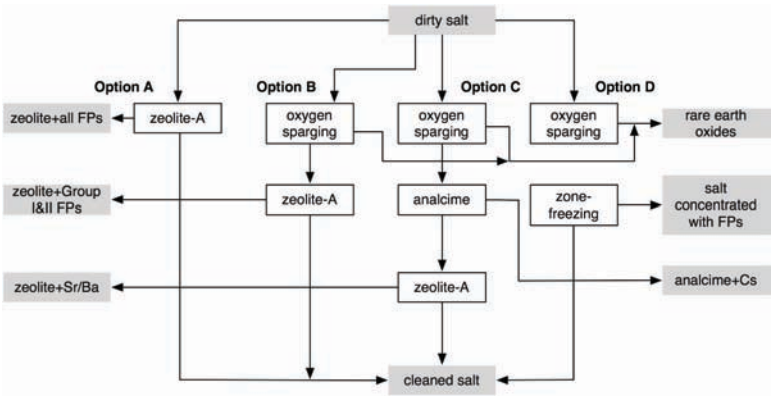


Figure 1. Schematic of the four most promising fission product separation options.

Table 1. Summary of functionality of proposed unit operations for fission product separations from salt.

Unit Operation	Target Fission Products	Waste Form Options	Comments
Zeolite-A Ion Exchange	All fission products	Glass-bonded sodalite	Relatively low fission product loading to the waste form and marginally clean salt, high selectivity for rare earths and actinides, leads to most mature waste form type, operation in a hot cell demonstrated
Analcime Ion Exchange	Cs	Pollucite	Efficient Cs removal at high concentration
Oxygen Sparging	Rare earths	Glass waste form, metal waste form	Extremely effective at rare earth removal, will react with residual actinides, operation in a hot cell demonstrated
Zone-Freezing	Groups I and II	Glass-bonded sodalite, SAP	Effective removal of Groups I and II, behavior in presence of rare earths viewed as unpredictable, operation in a hot cell may be problematic, discharged waste salt will include a considerable amount of LiCl-KCl, so a waste form capable of high salt loading such as SAP may be required

zeolite-A has a relatively high selectivity for rare earths, Options B and C introduce additional steps to the overall process without the added benefit of significant waste minimization. The bases for selecting an optimal process for fission product separation should include waste volume minimization and process simplicity. Option D is unique in that it completely departs from using zeolite or any ion exchange material in the process. The potential benefit of this approach is a dramatic improvement in waste volume minimization.

While none of the options can be eliminated at this time, it does appear that Options A and D should be considered as the preferred options for future investigation. Option A offers superior simplicity and demonstrated operability in a hot cell while being coupled to a mature waste form—the glass-bonded sodalite waste form developed for EBR-II spent fuel treatment. Meanwhile, Option D offers the greatest potential for waste volume reduction. Removal of rare earths via precipitation of their oxides offers outstanding waste volume reduction potential. It must be emphasized, however, that there is insufficient data at this time to eliminate any of the options. Several assumptions have been made in arriving at this recommendation. These include the following:

- Actinides can be sufficiently removed from the salt via a drawdown process so as to have little or no impact on the fission product separation unit operations or actinide loading to the waste form.
- A zone-freezing furnace can be effectively operated and controlled in a hot cell environment with high heat-emitting salt.
- In the case of sodium-bonded fuel, sodium chloride concentration in the ER salt can be regulated by using one or more of the separations unit operations.
- Iodine separation can be effectively removed by one of the separations unit operations, an assumption that has not been tested to date.
- Metal fuel has been assumed to be the feed into the process. If an oxide fuel is used, the Cs and Sr would likely be removed upstream of the electrorefiner, and it may be possible to achieve all of the needed separations via oxygen sparging.

Efficiently separating the fission product from ER salt to minimize high-level waste is a critical goal for the successful development of spent fuel treatment by the electrochemical/pyrometallurgical process. This project has been tremendously successful in producing a wealth of information on several viable options for achieving fission-product separation. Each institution has made outstanding contributions to the project. It is expected that these efforts will soon result in successful completion of the development and implementation of process unit operations that will greatly improve pyroprocessing technology.

Planned Activities

While this was the final year of this I-NERI project, favorable results from the project and the importance of waste minimization justify continued research into fission-product separation methods from ER salt at both KAERI and INL if future funding is available.

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 (ANL)

PI (Country): Hansoo Lee, Korean Atomic Energy Research Institute (KAERI)

Collaborators: Chungnam University at Daejon, University of Illinois at Chicago

Project Number: 2006-003-K

Program Area: FCR&D

Project Start Date: December 2007

Project End Date: September 2009

Research Objectives

The primary focus of this collaborative research between ANL and KAERI is to develop crosscutting materials for use in pyrochemical processing technology for the treatment of spent nuclear fuel (SNF). The high-temperature molten salts coupled with oxygen gas and convective flows generated as a result of the process cause corrosion in the most advanced nickel-based alloys. The project's objectives this year were 1) to examine the microstructural effects of heat treating the KAERI-fabricated alloys, N1 and N2, prior to corrosion testing and 2) to evaluate the corrosion resistance of a functionally graded material surface coating.

Research Progress

Corrosion Testing. Last year, KAERI developed two new alloys, N1 and N2, based on modifying the composition of Inconel 713LC by adding 2 wt% and 5 wt% Si, respectively, and eliminating the molybdenum. Corrosion testing was performed at 650°C in Li_2O/LiCl salt with an Ar/O_2 gas flow at 2 ml/min. This year, ANL examined the microstructure of alloys N1 and N2 in the as-cast (AC) and post-heat treatment (HT) condition before and after testing for a seven-day time interval in 1, 3, and 5 wt% Li_2O in LiCl. The heat treatment of half of the N1 and N2 coupons was undertaken to create a more homogeneous microstructure as the Si was observed to segregate in "islands" throughout the alloy matrix in the AC as-received condition. The heat treatment schedule appears in Table 1.

Table 1. Heat treatment parameters.

Alloy	Temperature (°C)	Time (Hours)	Cooling Method	Atmosphere
N1	1200	3	Air cool	Argon
N2	1200	6	Air cool	Argon

The alloy N2 AC showed the presence of a fairly uniform corrosion scale, but there appeared to be a slightly more advanced corrosion front in the areas where the Si-rich phase was adjacent to the scale at the scale/substrate interface. This is consistent with the findings from last year's work where the bulk matrix of the AC coupons consisted of Ti and Nb carbides and segregated areas of a Si-rich phase. A Cr-depleted zone was noted adjacent to a Cr_2O_3 inner scale, while an outer scale consisted of mostly NiO. As seen from the negativity of the Gibbs free energy calculation below, Al has the greatest affinity for combining with the available oxygen; however, the Al is combined with Ni to form the intermetallic Ni_3Al in the gamma prime phase for solid solution strengthening. This leaves the segregated Si free to react with the ionic oxygen and also accounts for the corrosion front advance in those areas of the adjacent Si phase.

$2Al + 1.5\ O_2\ (g) \rightarrow Al_2O_3$ $\Delta G = -1386.4$ kJ

$Si + O_2\ (g) \rightarrow SiO_2$ $\Delta G = -743.8$ kJ

$Ni + 0.5\ O_2\ (g) \rightarrow NiO$ $\Delta G = -155.3$ kJ

Coating Testing (NiCrAlY Bondcoat and 13 wt% YS-ZrO_2 Topcoat). Based on the results from a previous collaborative study with KAERI, performed in 2005, the high-density 13 wt% Y_2O_3 stabilized (13%YS)-ZrO_2 performed well as a topcoat. Since the coating was found to separate at the bond coat–substrate interface, a NiCoCrAlY bond coat was used. A commercial vendor applied the coating using a plasma spray technique. The micrographs of the coating before corrosion testing showed the coating microstructure to be very porous with many voids and cracks, as seen in Figures 1 and 2. The corrosion testing was performed at 675°C in 3 wt% Li_2O/LiCl salt with an Ar/O_2 gas flow at either 2 mL/min or 33 mL/min.

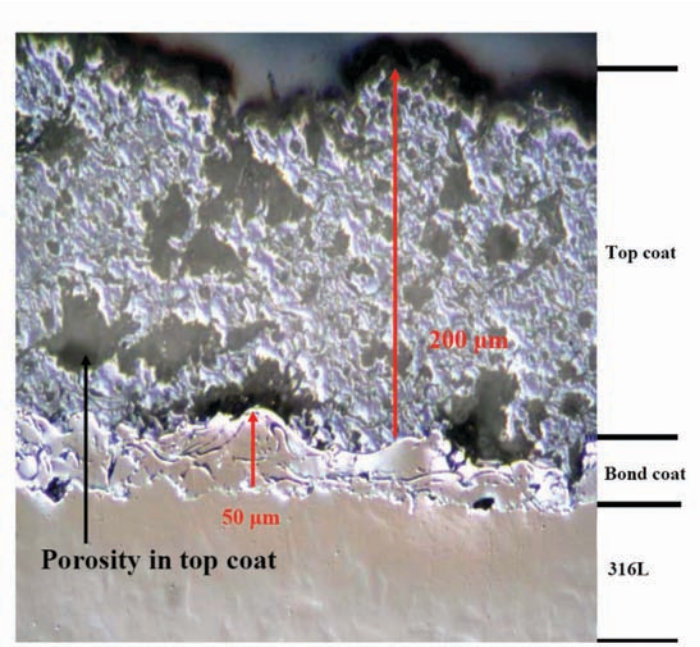


Figure 1. Coating layers, as-coated.

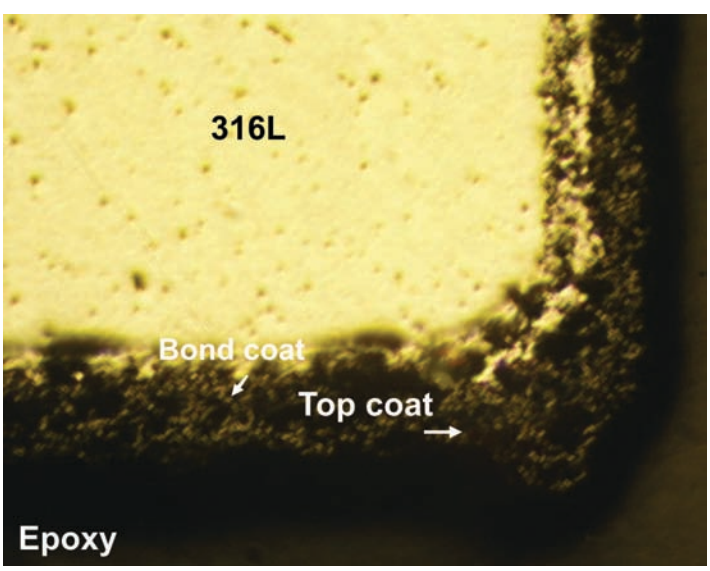


Figure 2. As-coated 316L coupon.

Initial analyses indicate the majority of the coating spalled either during immersion in the molten salt or during the post-corrosion cleaning procedure. Oxide formation of substrate metal components was observed. A heat treatment schedule for the as-coated samples was performed to increase the bond coat adherence, but the results showed no improvement after corrosion testing. Oxide formation of substrate metal components was observed with the formation of lithium zirconate, Li_2ZrO_3. Figure 3 shows the appearance of the coated coupons after corrosion testing and the X-ray diffraction pattern of the surface areas confirming the formation of Li_2ZrO_3.

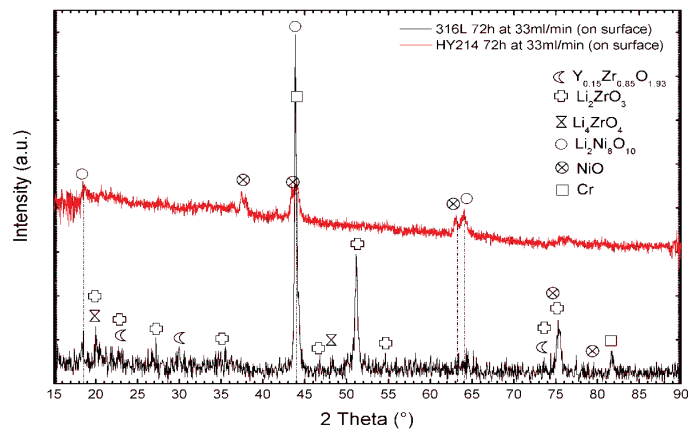


Figure 3. Haynes 214 and 316L SS XRD of surface area of coupons.

KAERI N1 Crucible Testing. A near-net-shaped crucible fabricated from KAERI alloy N1 was received in late August 2009 for use in a simulated oxide reduction experiment. The crucible was heat treated prior to testing, and the reduction experiment was performed on December 17–18, 2009.

Planned Activities

The near-net-shaped crucible fabricated from KAERI alloy N1 HT used in the simulated oxide reduction experiment will be analyzed and the results reported in a separate document.

INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE

VHTR Environmental and Irradiation Effects on High-Temperature Materials

PI (U.S.): Dane F. Wilson and William R. Corwin, Oak Ridge National Laboratory (ORNL)

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

Collaborators: Idaho National Laboratory (INL), Korea Advanced Institute of Science and Technology (KAIST)

Project Number: 2006-006-K

Program Area: Gen IV

Project Start Date: October 2006

Project End Date: September 2009

Research Objectives

This project selected and qualified high-temperature materials for the next generation of reactors. The collaboration included work to analyze irradiation and environmental effects and to develop a materials handbook containing the properties of high-temperature metallic materials. Researchers studied the effects of helium (He) environments on the mechanical properties of high-temperature metallic alloys proposed for use in the very high-temperature gas reactor (VHTR). The He 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 evaluated 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, provides the necessary design data and develops and confirms available margins.

Lastly, researchers developed the "Generation IV Materials Handbook," containing a database of high-temperature materials.

Research Progress

He Environmental Effects on High-Temperature Materials. The team evaluated the performance of materials in the proposed nominal impure He environment, which was inferred from past He-cooled reactor experience. In order to provide confidence in materials performance within the proposed nominal range, this range was extended to include (μ atm):

- H_2: 400 ± 100
- H_2O: 2 ± 1
- CO: 40 ± 10
- CO_2: 0.2 ± 0.15
- CH_4: 20 ± 5
- N_2: < 10

The proposed range allowed for significant changes in oxidation and carburization potentials. Testing in this expanded range was performed to evaluate the boundaries between the protective regime and the carburization and decarburization regimes. In addition, researchers designed and installed environmental creep testing chambers with a three-zone furnace, loading train, gas chromatograph, and control systems.

Recirculating He loops were designed, built, and used to evaluate the behavior of alloys 617 and 230. Tests were carried out with fixed gas chemistry for 500 hours or 1,000 hours at 1000°C, 900°C and 800°C. At 1000°C, under oxidizing conditions, Inconel 617 showed a relatively thick chromium oxide scale with significant formation of grain boundary aluminum oxides, whereas Haynes 230 showed less surface oxidation and a reduced tendency for formation of grain boundary oxides. Under carburizing conditions, Haynes 230 exhibited very little evidence of scale formation and a very large volume fraction of carbides along grain and twin boundaries. This

large volume fraction of carbides occurred throughout the cross section of the approximately three-millimeter thick coupon after 500 hours at 1000°C. Similar behavior was observed under carburizing conditions at 800°C and 900°C; however, the depth of carburization was reduced.

Alloy 617 and Haynes Alloy 230 were oxidized in the He-K1 (CO : 50 μ bar, CO_2 : 10 μ bar, CH_4 : 20 μ bar, O_2 : 0 μ bar, H_2O : 1.2 μ bar) and the He-K_2 (CO : 50 μ bar, CO_2 : 10 μ bar, CH_4 : <1 μ bar, O_2 : 0 μ bar, H_2O : 0.7 μ bar) environments. Oxidation resistance was dependent on the stability of the surface oxide layer, which can be affected by minor alloying elements such as titanium (Ti) and manganese (Mn). At 900°C, for Alloy 617, a mixture of an extensive NiO-Cr_2O_3 double layer and isolated NiO-$NiCr_2O_4$-Cr_2O_3 triple layer were observed under a steady-state condition. For Haynes 230, a $MnCr_2O_4$ layer formed on top of the Cr_2O_3 layer and resulted in a lower oxidation rate. At 1100°C, both alloys showed a double layer consisting of an inner Cr_2O_3 and outer $MnCr_2O_4$ or TiO_2. Spallation of the outer layer and subsequent volatilization of the Cr_2O_3 layer produced a rugged surface and interface as well as internal oxidation.

Thermal cycle experiments were performed with a peak-to-peak period of 2.5 hours at a 200°C temperature change. For cycling between 800°C and 1000°C in the presence of metallic surfaces, while there was some variability of the impurity composition around the set point, the impurity concentration remains within about 10 percent of the desired level. After thermal cycling of Alloy 617 and Alloy 230 under oxidizing conditions for a total of 500 hours, cross-sectional analysis showed no influence of thermal cycling, as compared to constant temperature exposure at 900°C, on the morphology or thickness of the oxide coating for this exposure time. No evidence of the oxide cracking was found that might be attributed to thermal cycling. In contrast, specimens thermally cycled at the same temperatures for 1,000 hours exhibited some evidence of spallation.

Tensile tests were performed, post-carburizing exposure, at room temperature and at 800°C. Haynes 230 specimens that were extensively carburized at exposure temperatures of 900°C (for 1,000 hours) and 1000°C (for 500 hours) had zero ductility at room temperature. At 800°C, the carburized Haynes 230 had reduction in area of about four percent after carburization at 900°C and seven percent after carburization at 1000°C. The fracture strength decreased from about 700 megapascals (MPa) at room temperature to 500 MPa at a test temperature of 800°C for both carburization temperatures. The results of these experiments once again reinforce the need for maintaining control of the He gas chemistry to avoid harmful effects on heat exchanger materials.

Researchers studied environmental degradation of Alloy 617 under constant tensile stress. A small punch test was also carried out to characterize the change in mechanical properties of carburized and decarburized (1000 hours) Alloy 617. In addition, creep tests in He atmosphere showed that time-to-rupture in He was longer than that in air at high stress, above about 30 MPa, but that at low stress, time-to-rupture in air was longer than that in the He environment. This behavior was attributed to oxide layer thickness formed during creep time.

Irradiation Effects on High-Temperature Materials. Tensile and V-notch Charpy specimens were machined from base materials and welded: hot-rolled Grade 91 Mod.9Cr-1Mo and forged heavy 9Cr-1Mo-1W steels. These materials were successfully irradiated in new design capsules for high-temperature materials testing in an OR test hole in HANARO. Mod.9Cr-1Mo and forged 9Cr-1Mo-1W specimens were irradiated for one cycle (24.62 days and 23.57 days, respectively) in the OR5 test hole of HANARO at a 30-megawatt (MW) thermal output. The irradiation temperature of the specimens was initially determined by the gamma heating and then finally adjusted to the desired value of 390±10°C by the He gas control system and electric heat. A fast neutron fluence was obtained in the range of 1.10~4.40×1019 n/cm^2 (E>1.0 megavolt [MeV]) and 0.27~1.04×1020 n/cm^2 (E>0.1 MeV) and in the range of 1.13~4.20×1019 n/cm^2 (E>1.0 MeV) and 2.85~9.9×1019 n/cm^2 (E>0.1 MeV) for Grade 91 Mod.9Cr-1Mo and forged heavy 9Cr-1Mo-1W steels, respectively. The dpa of the irradiated specimens was evaluated to be 0.034~0.07 using the SPECTER code.

Yield and tensile strength of the irradiated Mod.9Cr-1Mo base metal decreased with increasing test temperature, especially above 550°C, where the strength abruptly decreased. Yield and tensile strength of the irradiated Mod.9Cr-1Mo weldment showed similar trends as base metal. The strength of the weldment was higher than that of the base metal below 550°C. Above 550°C, the strength of the weldment was the same as or lower than that of the base metal. Tensile strengths of the base and weldment increased after irradiation by 18 percent and 10 percent, respectively. The elongation, however, decreased after irradiation by 25 percent in the base metal and 65 percent in the weldment. The hardening effect by irradiation was more severe in the weldment than the base metal.

The researchers tested Charpy V-notched Type A of ASTM Standard E23-07 (10x10x55 mm) specimens. The upper shelf energy (USE) of the irradiated Mod.9Cr-1Mo base metal was 285 joules (J). There was little USE change due to irradiation. The ductile-brittle transition temperature (DBTT) of the irradiated Mod.9Cr-1Mo steel was 0°C. The T41J and T68J index temperatures of the irradiated

Mod.9Cr-1Mo steel were measured as -44°C and -30°C, respectively. The USE of the weldment of Mod.9Cr-1Mo was 173 J after irradiation. The DBTT of the weldment was measured as 25°C. The T41J and T68J index temperatures of the irradiated weldment of Mod.9Cr-1Mo were measured as 0°C and 15°C, respectively. The USE of the unirradiated weldment of Mod.9Cr-1Mo was measured as 164 J, and the DBTT was measured as 1°C. The T41J and T68J index temperatures were measured as -16°C and -4°C, respectively.

The Mod.9Cr-1Mo (G91) and 9Cr-1Mo-1W (G911) base metal showed similar impact properties in the unirradiated condition. After irradiation, the G91 base metal showed similar impact properties to the G911 base metal. The USE of G91 base metal was higher than that of G911 base metal. The G911 weldment showed higher USE, but the DBTT, T41J, and T68J were less attractive than those of G91 weldment in the unirradiated condition. Neutron irradiation shifted the DBTT curve to the high temperature, and the degree of temperature shift was similar for G91 and G911. However, the DBTT of the G911 weldment was above room temperature after irradiation, while the DBTT for the irradiated G91 weldment was below room temperature.

High-Temperature Materials Handbook Development. The project team developed and deployed a database Gen IV materials handbook for international data collaboration associated with VHTR materials. In order to familiarize international users with its structure and features and provide feedback for structural modification and functionality enhancement to satisfy their needs, a demonstration version for the Gen IV International Forum (GIF), dubbed the Gen IV Materials Handbook GIF Demo, was released. Along with the release, a comprehensive handbook user guide and a step-by-step procedure for international user access were developed.

Researchers performed enhancements to the handbook's structure and functionality as well as its content expansion, base software upgrades, and data collection. The major functionality includes the following: querying tools using a combined select and search function; managing embedded equations and logic relations, which provides the ability to store and plot materials behavior models; and a data comparison chart, which allows quick comparison of a curve with stored raw data. Data for 283 creep test data records and 40 related pedigree information records for Haynes 230 alloy was digitized for upload to the handbook. The pedigree information includes information on the specific chemical compositions, product forms, and heat-treatment conditions (describing the heats that were used to generate the test data).

The team performed impact and tensile tests of a Mod.9Cr-1Mo hot-rolled plate and a thick-wall 9Cr-1Mo-1W forged steel to generate 17 data worksheets for use with the handbook.

Planned Activities

This project has been completed.

INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE

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

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

PI (International): Hee C. No, Korea Advanced Institute of Science & Technology (KAIST)

Collaborators: None

Project Number: 2007-001-K

Program Area: Gen IV

Project Start Date: October 2008

Project End Date: September 2009

Research Objectives

The major goal of this project is to perform air ingress-related analyses and experiments in order to reliably predict the consequences of air ingress in the Next Generation Nuclear Plant (NGNP) with a high degree of certainty. The research objectives are summarized below.

First, analyses will be performed to simulate air ingress accidents. The major objective of these analyses is to understand what will really happen in the air ingress accident and to evaluate various methods for mitigating the effects of the air ingress.

Second, experiments will be conducted to supply information needed to validate computer codes to model important phenomena during air ingress accidents. These experiments will measure the:

- Rate of air ingress into the reactor core because of density-driven, stratified flow.
- Variation in oxidation rate and density and their effects on burn-off in the bottom reflector.
- Effects of burn-off on the structural integrity of the core bottom structures.

Third, an important goal of this project is to develop coupled neutronics and thermal-hydraulic capability in the GAMMA code, which involves the following:

- Development of core neutronics models.
- Coupling of neutronic-thermal hydraulic tools.
- Verification and validation (V&V) of the coupled core model.

This will allow the simulation of recriticality in conducting cool-down without scram and accurate initial and transient power distribution, which are both essential for accurately estimating the safety margin.

Research Progress

This section highlights key FY 2009 accomplishments for all nine tasks.

Density Difference-Induced Stratified Flow Analysis by INL. Task 1 accomplished various theoretical and computational activities. First, the team established a new air ingress scenario based on the density gradient-driven flow, which divided the whole air ingress scenario into four steps: 1) depressurization, 2) Stage-1 stratified flow, 3) Stage-2 air moving upward to the reactor core, and 4) global natural circulation. Researchers carried out analytical investigations, including previous stratified flow research, to identify and compare important air ingress mechanisms (molecular diffusion vs. density gradient-driven flow). Theoretical estimations show that the density gradient-driven flow will dominate the whole air ingress process.

The team also performed a number of computational fluid dynamics (CFD) analyses, including a 3D CFD analysis with CFX-12 (ANSYS 2009) for the air ingress accident of the GT-MHR 600 MWth reactor under the assumption of a double-ended guillotine break (DEGB). The CFX results are shown in Figure 1. These calculations confirmed the current phenomenal understanding of air ingress based on density gradient-driven flow. The team also performed preliminary 2D CFD analyses, including the heterogeneous chemical reaction. Preliminary 2D CFD simulations were performed for a small-break accident, which is much more probable than the DEGB case. According to these

analyses, air ingress in the small-break situation is still very fast and highly dominated by density gradient flow rather than molecular diffusion.

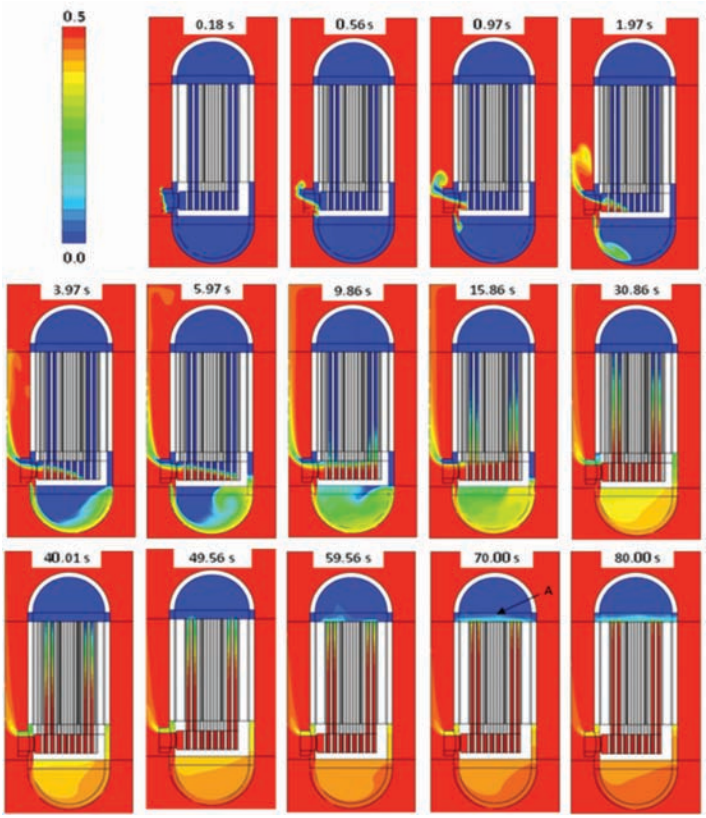


Figure 1. Variation of the air mass fraction (3D CFX model). Contours are plotted on the plane of y = 0.01 m. Symmetry plane is y = 0.0 m.

Experimental Study on the Stratified Flow by INL. Task 2 work established an experimental plan for the density gradient-driven stratified flow. The research team designed isothermal experiments to understand stratified flow phenomena in the DEGB situation. The DEGB experiment will focus on visualization of stratified flow in the horizontal pipe. This experiment is designed to measure local and global velocity through flow visualization, and the data will be used for code validation. For data comparisons, the team developed CFD models based on the final experimental design and completed some sensitivity studies. For validation, the team will compare the results of these CFD simulations to the experimental data.

Advanced Graphite Oxidation Study by INL. Task 3 work upgraded structural analyses of the graphite-supporting structure, performed in FY 2008, to the new air ingress scenario, established in FY 2009. It assumed that the flow is driven by density gradients in addition to molecular diffusion. This analysis estimated the collapse of the graphite structure for a simplified model of the reference VHTR using two computer codes: GAMMA code (system analysis code) and ABAQUS code (stress analysis code). The oxidation and corrosion of graphite were predicted by the GAMMA code, and the information was implemented in the ABAQUS code to estimate the core collapse. Aside from this analysis, the team also performed computations with MATLAB to conservatively estimate the maximum allowable burn-off to maintain graphite structural integrity. A graphite oxidation model and algorithm, including graphite corrosion and failure, were constructed and implemented in the GAMMA code. Based on the advanced model, VHTR air ingress analyses were carried out for two different graphite structural materials (IG-110, H-451) under conservative assumptions.

Air Ingress Mitigation Study by INL. In Task 4, researchers conducted some preliminary studies to investigate air ingress mitigation methods, thereby providing some idea of the air ingress mitigation or graphite oxidation mitigation methods proposed in this task.

Experimentation of Burn-off in the Bottom Structure by KAIST. As a continuation of FY 2008 work, Task 5 investigated various graphite oxidation and burn-off characteristics of IG-430 graphite. Researchers compared the experimental results to those of IG-110 graphite obtained in FY 2008. As a result, the kinetic parameters such as activation energy and order of reaction for IG-430 were experimentally measured to be 158.5 kJ/mol and 0.37, respectively, while the values for IG-110 are 218 kJ/mol and 0.75. The burn-off characteristics of IG-430 and IG-110 were also compared at Zone I dominated by kinetics. The maximum reaction rates for both IG-430 and IG-110 graphite appeared at about 40 percent burn-off, but the trends were somewhat different: IG-110 showed rapid increase of reaction in the beginning with large internal surface area changes, and IG-430 showed only small changes of reaction with burn-offs. One interesting observation was that the presence of moisture in the reacting gas does not affect the oxidation or burn-off history during the whole light-water reactor period.

Structural Tests on Burn-off Bottom Structure. In Task 6, researchers carried out mechanical tests of IG-430 graphite for various oxidized conditions. The test results were then compared to those of IG-110 obtained in FY 2008. Notably, the compressive strength of the original IG-430 was measured to be 88.99 MPa, about 13 percent higher than that of IG-110. The experiment for fracture of IG-430 showed that the graphite failure stress is a function of slenderness ratio only. Therefore, the failure strength of the graphite structure can be simply determined by the geometrical information and burn-off level. This experiment also showed that IG-430 is fractured by buckling at a slenderness ratio larger than 13.83, while IG-110 is fractured by buckling at a slenderness ratio larger than 11.76. These values can be considered the transition point of the slenderness ratio

showing different failure modes (compressive or buckling). The relationship between graphite burn-off and buckling strength for IG-430, expressed by a Knudsen-type formula, has been investigated by mechanical testing for the oxidized samples. This experiment showed that the mechanical degradation of IG-430 is quite a bit slower than that of IG-110, thus maintaining better structural integrity. Finally, the team performed mechanical testing of several graphite columns with different geometries and dimensions for various burn-off levels considering general geometries. Based on the experimental results, the strength of graphite columns oxidized in Zone I is dependent on the initial strength and burn-off level, even though the graphite columns have different dimensions with different failure modes.

Coupling Neutronics-Thermal Hydraulic Tools by KAIST. Task 7 consisted of analyzing VHTR thermal hydraulics behavior using KAIST-proposed coupling neutronics–thermal hydraulic tools. KAIST developed a multidimensional gas multicomponent mixture analysis code (GAMMA) and multigroup 3D hexagonal geometry neutronics code (COREDAX). These codes were coupled into a system code, known as GAMMA/COREDAX, so the VHTR core could be analyzed. The parameters for neutronic–thermal hydraulic coupling were set up between the GAMMA and COREDAX codes. Since the calculation nodes between the two codes are quite different, researchers developed and tested a node-mapping subroutine in the COREDAX code. By replacing the "point kinetics" subroutine in GAMMA code with the "COREDAX_coupling" subroutine, the rresearch team finally completed the coupling GAMMA and COREDAX codes. The researchers tested the GAMMA/COREDAX code coupling on a simplified GTMHR 600 MWth. This test showed that the calculation results between GAMMA with COREDAX results and GAMMA/COREDAX code are exactly matched.

Core Neutronic Model by KAIST. Task 8 work consisted of KAIST developing the COREDAX code based on the analytic function expansion nodal (AFEN) method in 3D hexagonal geometry. The AFEN method includes the use of node-interface flux moments as nodal unknowns. These nodal unknowns increase the accuracy of 3D calculation results. A multigroup extension based on matrix function theory and coarse group rebalance acceleration was applied to the COREDAX code. The COREDAX code implementing the AFEN method was verified by testing the VVER-440 benchmark problem, a simplified VVER-1000 benchmark problem, and the SNR-300 benchmark problem. The benchmark results showed that the COREDAX results are in good agreement with the reference results calculated by PARCS (for VVER-400), VENTURE (for VVER-1000), and DIF3D (for SNR-300).

Core Model Coupling and V&V. Task 9 work to be performed in FY 2010 will involve verification of the GAMMA code coupled with COREDAX code and validation of prediction results of thermal power distribution in the hexagonal reactor core.

Planned Activities

Following are the activities planned for FY 2010:

- Isothermal air ingress experiments by INL.
- Air ingress mitigation study by INL.
- Continuation of structural tests of oxidized graphite in the bottom reflector by KAIST.
- Coupled core model validation and verification by KAIST.

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: FCR&D

Start Date: October 2007

End Date: September 2010

Research Objective

This research project addresses 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, prepare fuel for subsequent processing, simplify downstream processing by removing volatile fission products prior to the pyroprocess, and safely trap the volatile fission products. 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.

The research project focuses on three primary areas: 1) evaluation of the effects of advanced voloxidation on pyroprocessing of spent oxide fuel, 2) optimization of the off-gas trapping capabilities for fission products with respect to pyroprocessing, and 3) development of operational parameters for advanced voloxidation with respect to pyroprocessing.

Research Progress

Researchers initiated experiments for particle size control with the rotary-type voloxidizer using SIMFUEL pellets. Particle aggregates up to twenty mm were successfully produced following the oxidation to U_3O_8 with rotational speeds of three rpm. Reduction of the agglomerated U_3O_8 particles to UO_2 with H_2 resulted in only a four percent shrinkage, on average, of the original particles. The team performed static tests with irradiated materials to 1200°C to assess the degree of particle agglomeration. Larger particles were generated by minimizing handling operations during the crucible unloading and sieving steps.

Based on co-collection of technetium and cesium during experimentation with irradiated fuels, the filter media were improved and tested with surrogate materials to verify separation of these volatile fission products. The calcia (CaO) content of the fly-ash filter media was adjusted, either up or down, depending on the filter's purpose (either technetium or cesium collection, respectively). The calcia levels of the fly-ash filters range from zero weight percent to thirty weight percent. Figure 1 shows the fly-ash filters used in testing with surrogate cesium and rhenium (technetium). Complete separation of the cesium and rhenium was achieved, as confirmed by X-ray diffraction analyses, scanning electron microscopy, and gravimetric analyses. In addition, the team performed parameter development testing with irradiated materials to verify equipment capabilities for future testing with the improved fly-ash filters.

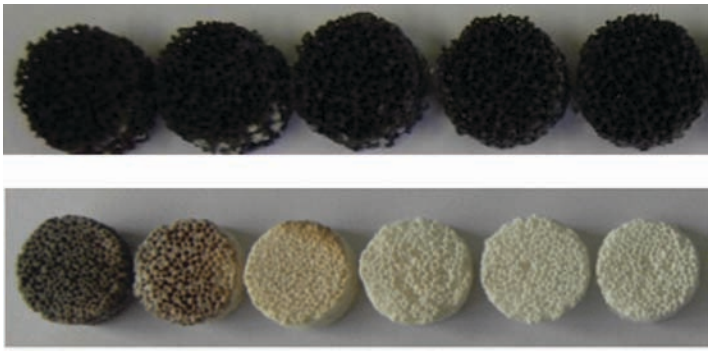


Figure 1. Photographs of fly-ash filters following testing: (a) for cesium and (b) for technetium.

Planned Activities

The team will continue experiments with both non-radioactive and irradiated materials to develop optimal operating conditions for off-gas trapping and particle size control. They will test improved fly-ash filters with irradiated spent oxide fuel to confirm the separation of radioactive cesium and technetium volatiles. Finally, the research team will improve the existing rotary voloxidizer to allow higher operating temperatures, and researchers will initiate a design for rotary equipment in a radioactive environment.

INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE

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

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

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

Collaborators: None

Project Number: 2007-003-K

Program Area: FCR&D

Project Start Date: November 2007

Project End Date: September 2010

Research Objectives

The objective of this project is to evaluate the high burnup performance of transuranic (TRU)-bearing metal fuel for the sodium fast reactor (SFR). The project includes the following tasks: 1) benchmarking a performance analysis code using AFC-1 test data, 2) fabricating barrier-cladding material, and 3) out-of-pile performance testing against metal fuel. The objectives of benchmarking the fuel performance code are to develop suitable models and to benchmark the code against irradiation data from AFC-1 tests. These codes make it possible to analyze the fuel's complicated irradiation behavior. The second task focuses on fabricating barrier cladding materials and tubes because one of the potential factors limiting alloy fuel burnup is fuel/cladding chemical interaction. The objective of the final task is to conduct the diffusion couple testing of metal fuel against barrier cladding. Using barrier-cladding material may increase fuel burnup performance.

Research Progress

Benchmarking of Fuel Performance Codes. INL and KAERI jointly decided the benchmarking area of the fuel performance code. INL provided the technical reports of the AFC-1 test in order to benchmark the KAERI models for thermal conductivity and fission gas release behavior. These models were selected as the benchmarking areas since temperature (which is a function of thermal conductivity) and pressure (which is a function of fission gas release) affect almost all other fuel behavior. The temperature and pressure can also be limiting factors for the high burnup potential of the fuel, so these models must be developed or upgraded for the TRU-bearing fuel.

Figure 1 shows the fuel radial temperature profile of U-Pu-10Zr and the AFC-1 fuel according to the established and the adjusted correlations, respectively. It was calculated that the fuel centerline temperature and the temperature gradient for the AFC-1 fuel were significantly higher than those of the conventional U-Pu-10Zr because the thermal conductivity of AFC-1 is much lower than that of U-Pu-10Zr. The maximum centerline temperature for the AFC-1 fuel was found to be about 880°C, and the temperature gradient was about 300°C. The centerline temperature for the conventional U-Pu-10Zr fuel was about 700°C, and the temperature gradient was about 150°C.

The AFC-1 fuels, with their lower thermal conductivity, are expected to require sensitivity studies regarding the fission gas release at high temperature and the fuel behavior at overpower in transient states.

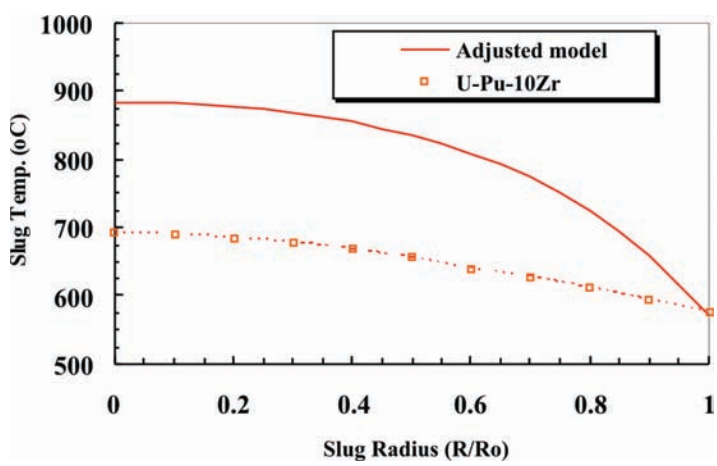


Figure 1. Fuel radial temperature profile.

Figure 2 shows the fission gas release for the AFC-1 fuel calculated by the existing model in MACSIS. In the case of the AFC-1 fuel, the fission gas release showed the same trend as the behavior during the transient state because of the high temperature and high-temperature gradient. It was calculated that the fission gas release at a burnup of 2 atomic percent (at%) was 100 percent.

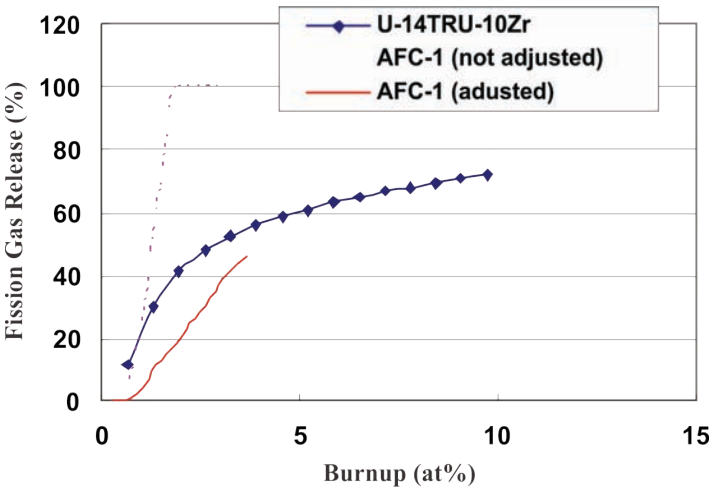


Figure 2. Calculated fission gas release.

In order to fit the existing model to coincide with the AFC-1 data, the number of atoms of gas at grain boundaries in the model was changed. Figure 2 also shows the fission gas release for AFC-1 in the case of changing the number of gas atoms. The fission gas release is observed to be significantly reduced but show a better correlation to the previously determined behavior with respect to at% burnup.

Figure 3 shows the fission gas release according to the fission density burnup. It was calculated that the fission gas release behavior of the AFC-1 fuel shows the same trend as the U-Pu-10Zr when correlated with fission density.

As shown in Figure 3, the fission gas release for AFC-1 can be calculated by adjusting the key variables in the existing model. The fission gas release model will be upgraded and redeveloped by considering such key factors as fuel power (temperature), burnup, fuel restraint, diffusion coefficient, bubble distribution, etc.

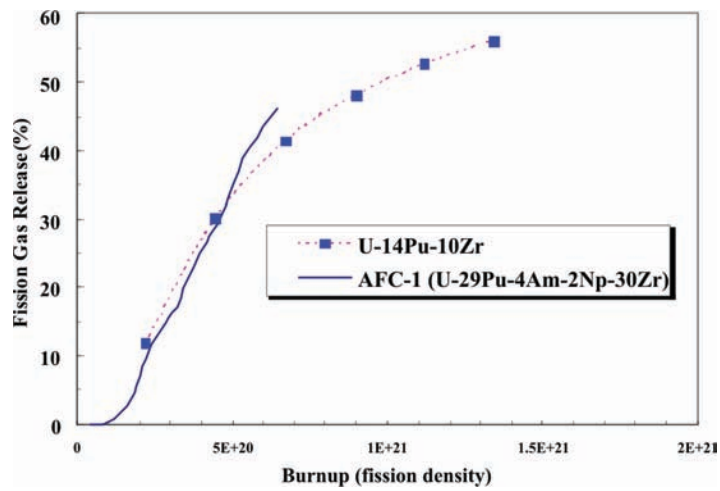


Figure 3. Fission gas release according to the fission density.

Fabrication of Barrier Cladding. Researchers at KAERI fabricated the chromium (Cr) barriers electroplated on ferritic/martensitic steel (FMS). In order to form the Cr layer on the surface of Gr. 91 steel, two plating methods were used that produced "hard Cr" and "crack-free Cr." The project team used a physical vapor deposition (PVD) to coat the disks with zirconium (Zr), tungsten (W), vanadium (V), titanium (Ti), vanadium nitride (VN), and zirconium nitride (ZrN). Four kinds of barrier cladding (VN, ZrN, V by PVD and Cr by electroplating) were selected for diffusion couple testing. Final application of a cladding barrier in an SFR will require the barrier coating to be applied to the inside surface of cladding tubes. The team evaluated fabrication processes used to coat cladding tubes.

Diffusion Testing of Metal Fuel Against Barrier Cladding. Diffusion couple tests were carried out at 750°C for 24 hours at INL; results are summarized in Table 1. INL

Table 1. Performances of the diffusion barrier.

Material	Type	Thickness (um)	Description
VN	PVD	1.3	- No eutectic or interdiffusion - VN adhered fuel side
ZrN	PVD	1.3	- No eutectic or interdiffusion - U, Pu locally diffused inside ZrN barrier
V	PVD	1.3	- No eutectic or interdiffusion - U, Pu, Zr locally penetrated inside V barrier
Cr	Electroplating	20	- No eutectic or interdiffusion - U, Pu locally penetrated along the crack

established an experimental schedule so that KAERI could physically participate in the entire process of the diffusion couple testing with TRU-bearing fuel. The results showed that neither the eutectic reaction nor inter-diffusion occurred in the case of the VN barrier. The other barriers also showed good results, but some local reaction on the fuel/barrier interface occurred because of the thin coating layer thickness. The diffusion couple tests with thicker VN, V, and ZrN cladding barriers are scheduled for later study.

Planned Activities

Following is a description of the planned activities for each of the three tasks described above.

Benchmarking of Fuel Performance Codes. The thermal conductivity correlation and the fission gas release behavior for AFC-1 should be evaluated continuously. Therefore, follow-on work will include confirmation of the thermal conductivity data and an upgrade of the fission gas release model through joint discussions between INL and KAERI.

Fabrication of Barrier Cladding. For this task, KAERI will prepare new thick barrier claddings and send them to INL for the next series of diffusion couple testing. KAERI will provide INL the cladding tubes whose inner surface has been barrier-coated for the interdiffusion tests.

Diffusion Testing of Metal Fuel Against Barrier Cladding. Researchers will analyze U-TRU-Zr diffusion couple tests against new barrier-cladding materials. KAERI may conduct the screening studies of the diffusion behavior of rare earth in contact with proposed barrier materials. INL will conduct the screening test with the cladding tube whose inner surface is coated with the barrier material prepared by KAERI.

INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE

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

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

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

Collaborators: None

Project Number: 2007-004-K

Program Area: FCR&D

Project Start Date: January 2008

Project End Date: September 2011

Research Objectives

The purpose of this project is to develop new high-level waste (HLW) forms and fabrication processes to dispose of active metal fission products recovered from electrorefiner (ER) salts during the electrochemical/pyrometallurgical processing of spent nuclear fuel. The current technology for disposing of fission products accumulated during electrochemical processing involves discarding the fission-product-loaded salt in a glass-bonded sodalite ceramic waste form (CWF). Selective removal of fission products from the ER salt would greatly reduce the quantity of HLW generated from electrochemical operations and allow the cleaned ER salt to be reused. Various novel methods are currently being developed to perform selective separation of fission products, but investigations to develop suitable waste forms to immobilize the separated fission products are still in the nascent stage. During the first year of this collaborative research project, KAERI focused primarily on developing immobilization methods for fission products isolated from ER salts using surrogate materials; at the same time, INL focused on producing and testing candidate waste forms using radioactive ER salt in their hot-cell facilities.

Research Progress

In the first year of this project, researchers at KAERI and INL investigated potential waste forms resulting from separation processes used to clean and recycle ER salt. Figure 1 illustrates several separation options being evaluated. An extensive literature review of immobilization technologies, either suggested or under development, indicated that dechlorination of the separated fission products followed by immobilization using a reactive agent would be the best option for overall waste minimization. The research team at KAERI performed a series of experiments to find effective materials for fission-product immobilization, which involved assembling processing equipment that integrated the dechlorination and solidification methods. The team performed the following activities to evaluate potential waste forms:

- Designed and synthesized new host matrix materials.
- Determined the reactivity of host materials to LiCl-KCl salt.
- Performed screening tests to find appropriate host materials for each waste.
- Determined the solidification procedure for each waste.
- Evaluated the solidification equipment for basic operating conditions.
- Fabricated ceramic or vitrified waste forms.
- Evaluated the ceramic or vitrified waste forms by compatibility and leach testing.

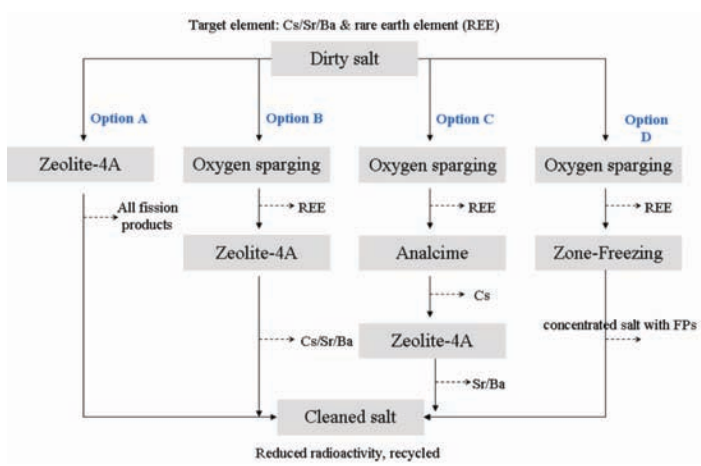


Figure 1. Separation options for removing fission products from ER salt.

Specific waste forms targeted for development from the separation process shown in Figure 1 include products produced from Options A and D. These options were chosen based on the maturity of the process and on the potential benefit of waste minimization. Option A is a modification of the EBR-II spent fuel treatment process—that is, instead of adding ER salt to zeolite and processing a low-loaded ceramic waste form (CWF), the zeolite is submerged in the salt and the cationic fission products ion exchange or occlude into the zeolite at high concentrations. This high-loaded zeolite is then fabricated into an advanced CWF material. In Option D, the primary component is oxygen sparging in the molten ER salt to chemically react the rare-earth fission products to produce oxychloride and oxide precipitates. Impurities in the salt are then concentrated into a small volume of electrolyte by a zone-freezing process. The highly concentrated salt containing fission-product impurities is then physically separated from the pure salt. The fission-product-concentrated salt is then distilled, separating the salt containing Group I and II fission products from the rare-earth fission product precipitates. The salt distillate is then immobilized in a silicon-aluminum-phosphate (SAP) host matrix, while the rare-earth precipitate is immobilized in a titinate host matrix. This process, which was developed by KAERI during the first year's research effort, is illustrated in Figure 2.

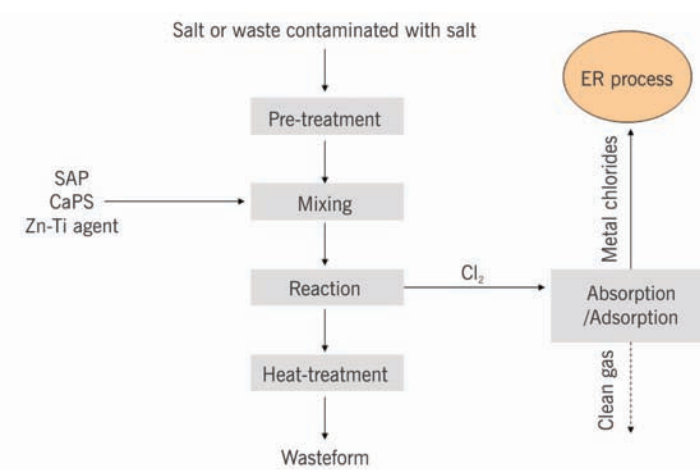


Figure 2. KAERI solidification procedure for each waste form material from the separation process.

The first year's efforts at INL involved setting up process equipment and methods for hot-testing advanced waste forms in the Hot Fuels Examination Facility (HFEF) hot cell. The two waste form feed materials produced were 1) a high-loaded zeolite to be used for fabricating a ceramic waste form and 2) a salt concentrate containing rare-earth precipitates and Group I and II fission products. These feed products will be processed into waste form products during Phase II of this I-NERI project (FY 2010).

Planned Activities

KAERI activities during FY 2010 will focus on optimization and initial characterization of favorable waste form products. The research team will study reaction characteristics for each waste form host composite to optimize the chemical routes and processing conditions. The compatibility between waste, composite, and glass materials will be tested to establish the solidification conditions, mixing ratio, glass composition, and processing temperatures. Finally, the operating conditions for the solidification equipment will be optimized. In addition, the team will perform an initial characterization of the most favorable waste form products. This activity will include the following tasks:

- Obtaining thermal stability and physical property data of HLW forms fabricated by using surrogate materials.
- Evaluating the chemical durability by static/dynamic leaching test methods.

FY 2010 activities at INL will include hot cell equipment fabrication and waste form characterization. During this time, the sample chamber and off-gas system for the salt distillation furnace will be modified for distillation of waste products. A high-loaded CWF will be processed and characterization initiated. The SAP waste form product containing the concentrated Group I and II fission-product distillate will be produced, as will the titanate waste form product produced from the rare-earth oxides. The team will confirm operability of waste characterization and testing equipment in HFEF for the following methods: chemical analysis of dissolved samples, X-ray diffraction, scanning electron microscopy, density, corrosion testing, and thermal testing.

INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE

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

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

I (ROK): K.R. Kim, Korea Atomic Energy Research Institute (KAERI)

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

Project Number: 2007-006-K

Program Area: FCR&D

Project Start Date: November 2007

Project End Date: September 2010

Research Objectives

This research aims to develop computational models of electrorefiners based on a fundamental understanding of the processes. The primary goal of this project is to develop rigorous yet flexible electrorefiner models. To support this objective, the team is focusing on two parallel approaches: 1) developing a computationally light and portable two-dimensional (2D) computational model and 2) investigating approaches for a computationally intensive, three-dimensional (3D) model for detailed and fine-tuned simulation of electrorefining processes.

Currently, spent driver fuel from the Experimental Breeder Reactor-II (EBR-II) at INL is being treated in electrorefiners. These electrorefiners are the crucial components of pyroprocessing, a process in which spent fuel is separated into four streams: 1) uranium metal, 2) U/TRU (transuranic) metal, 3) metallic high-level waste containing cladding hulls and noble metal fission products, and 4) ceramic high-level waste containing sodium and active metal fission products. Successful completion of the proposed research should be beneficial for improving operations of current electrorefiners and better understanding fundamental aspects of electrorefining.

Research Progress

Database Compilation and Experimental Measurements. Accurately assessing thermodynamic and electrokinetic data of the species involved in the electrorefining process is highly important for enhancing the computational models' reliability and credibility. Research participants compiled the required documentation for building an electrorefiner database and gathered the thermochemical and electrokinetic data to construct it. Some basic electrochemical properties, such as U^{3+}/U couple exchange current density, are not readily available in the open literature; therefore, researchers arranged experimental projects to measure the exchange current densities, charge transfer coefficients, and diffusion coefficients. They measured the exchange current density of the U^{3+}/U couple in a LiCl-KCl eutectic mixture at 500°C by applying a linear polarization resistance technique, which ranged from 202 to 584 A/m^2 depending on electrode materials. Researchers also calculated the charge transfer coefficients, which were in the range of 0.3 to 0.5. In addition, they measured the diffusion coefficient of U^{3+} in molten salt at 500°C via chronopotentiometry (CP) on a tungsten electrode and found the value to be 1.06×10^{-5} cm^2/sec.

2D Model Developments. Researchers continued their modeling efforts to further understand the fundamentals of electrorefining, shifting the focus to electrochemical-reducing behaviors at the cathode and electrolyte dynamics, while previous efforts were mostly focused on anodic oxidation behaviors. The optimization routine developed for describing anodic oxidization behaviors has been applied to the reduction reactions occurring at the cathode of the electrorefiner within the electrochemical processing of used nuclear fuel. The team made improvements to the anodic surface area calculations, including individual exposed surface areas for each species, and modeled the changing surface area as a function of fuel porosity. This updated routine has been used in developing a model of the Mark-IV electrorefiner at the Fuel Conditioning Facility (FCF) at INL. Results of the modeling process reveal that plutonium is rapidly exhausted from the anode fuel baskets. Throughout the remainder of the electrorefining process, uranium is the main species being both dissolved from the anode and

deposited on the cathode; however, as time progresses, zirconium dissolution and deposition start to become important.

The team has also examined 2D potential and current distributions within the bulk electrolyte of the Mark-IV electrorefiner. The electrical conductivity of the molten salt electrolyte depends upon composition and is shown to be around 199.8 mho/m throughout the electrorefining process. The potential distributions show that the highest potential gradients exist directly between the anodes and cathode. In general, the uranium, plutonium, and total potential gradients increase throughout the process, while zirconium potential gradients decrease. The result shows that as the electrorefining progresses, more zirconium and less uranium and plutonium is being electrotransported.

Researchers compared the total electrode potentials, as calculated by the latest model, to the potentials from a set of experiments run in the Mark-IV. The calculated and experimental potentials are shown to match, with root-mean-square errors of 2.54 percent and 13.9 percent for the anode and cathode, respectively. This compares to 2.82 percent for the earlier FY 2008 anode model.

3D Model Developments. Researchers proposed a computational electrofluid dynamics (CFD) model of a uranium electrorefiner, which they implemented on a CFD framework. The proposed model provides valuable information about the transport phenomena inside the electrolytic cell such as concentration, potential, and overpotential distribution. A unique feature of this model is the implementation of the potential-to-current algorithm that captures a more realistic spatial variation of the electrochemical kinetics.

The team developed an acceleration method for the proposed 3D simulation of electrorefining with multi-species. A key idea was to approximate multi-species ionic transport behaviors with uranium species only—as uranium is expected to be responsible for most ionic transportation behaviors in molten salt most of the time. This approximation approach significantly reduced computational time for calculating local concentration distribution and local multi-current density distribution. The current density distribution patterns were analyzed with respect to various rotating electrode speeds and applied current patterns. The results showed good agreement with the general principle of the electrorefining process.

The team noted that similar research of local mass transfer prediction had been previously studied in copper electroplating and flow accelerated corrosion fields. These studies showed that the local mass transfer effect in turbulent conditions is generally difficult to understand, mainly because turbulent flow shows irregularity and results in a very thin diffusion boundary layer. Regardless of these difficulties, the team believes that 3D modeling provides valuable insights into many complex electrochemical and transport phenomena that cannot be investigated experimentally.

Model Validation Preparations. During FY 2009, the team built the operational database for Mark-IV and Mark-V electrorefiners and collected geometry information for the Mark-V. In addition, the researchers built a rotating cylinder hull (RCH)-type cell to benchmark the proposed modeling approach. Preliminary experimental studies demonstrated that the constructed cell can be useful for verifying the developed models. The local overpotential and current density distributions along the rotating electrode were calculated and compared with experimental data.

Planned Activities

Continued Development of Models. A key parametric value, the exchange current density of uranium species, was measured during FY 2009. This value is arbitrarily assumed in the current version of the models and needs to be updated. Consequently, other parametric values will be adjusted in order to accommodate the measured exchange current density value.

Further work on the 2D electrorefiner model development will include increasing the number of species being examined. The ability of the model to accurately predict the exhaustion of uranium from the anode fuel baskets will be studied and improved. Other interfacial areas within the electrorefiner involved in electrochemical reactions (i.e., Cd pool/salt, vessel walls/salt) will be studied and included in the current model. Potential and current distributions will be calculated and examined from different planes to develop a 3D understanding of these distributions. As the software tool becomes mature, software manual documentation will be conducted.

Researchers will use the 3D computational platform to simulate and compute local overpotential, current density, and surface concentration in a type of rotating concentric anode–cathode module (ACM) resembling that of a Mark-V electrorefiner. Actual geometry information of a Mark-V ACM may be used, contingent upon DOE approval and release of the data. Otherwise, researchers will infer an approximate geometry from the open literature. They will also investigate the correlation between current density and potential. Further acceleration may be achieved with the identified correlation.

Continued Data Update and Model Validation Preparation. INL and KAERI will continue the coordination efforts for model validation. Upon the data release approval of DOE and export control during FY 2010, INL and KAERI will review a set of operational data from INL and determine the set of runs that may best serve for the model validations. The data will be fed into the developed models and drive the electrorefining simulation. The team will compare the resulting calculated outputs with the experimental measurement results from the electrorefiner operations. Researchers will use the comparison results to assess the capabilities and limitations of the models and perform cross validation between the 2D and 3D modeling approaches.

Using the constructed RCH cell, researchers will address the further experimental verification against the electrodeposition data in a copper electroplating system for model validation. Though experimental settings are different, the verification and validation efforts within this domain should add confidence to the overall modeling approach.

INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE

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: FCR&D

Start Date: November 2007

End Date: November 2010

Research Objectives

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). Efforts will ultimately be focused on SFRs with higher core outlet temperatures (550°C or greater) than traditional SFRs. At high temperatures and long 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.

Research Progress

During the first year of the project, researchers selected the advanced burner test reactor (ABTR) developed at ANL as the initial preconceptual reference design. The ABTR is a 250 megawatt thermal (MWt) metallic-fueled pool-type SFR with core outlet and inlet temperatures of 510°C and 355°C, respectively, and a 60-year service lifetime. To assure the structural integrity of the ABTR preconceptual design, it is necessary to show that the design can accommodate all of the specified duty cycle events corresponding to the service levels defined in the *ASME Boiler and Pressure Vessel Code Section III – Subsection-NH* (ASME-NH) with sufficient design margins. Researchers carried out a comprehensive review of the mechanical and thermophysical properties of materials available in the ASME-NH code and identified suitable materials for use in ABTR components.

For the elevated-temperature operating events, time-dependent structural damage such as creep rupture, creep ratchet strain, and creep fatigue have to be evaluated. Researchers developed a set of duty cycle events for Service Levels A, B, C, and D for the ABTR with a supercritical carbon dioxide (S-CO_2) Brayton cycle power converter. The project team carried out structural evaluations for three groups of duty cycle events: 1) heat-up from hot standby to full power and reverse operation with hold at full power, 2) heat-up from refueling to full power and reverse operation with hold at full power, and 3) daily load following. The evaluations were carried out using the SIE ASME-NH (Structural Integrity Evaluation ASME-NH) computer code developed at KAERI that implements the rules of ASME-NH and incorporates deterministic analyses of material inelastic behavior for use with results obtained from calculations assuming elastic behavior. Structural evaluations were performed on the following components: the reactor vessel, reactor support, redan, reactor upper head, core support, and intermediate heat transport system piping. Elastic analyses were performed using the ANSYS finite element computer code.

During the second year of the project, researchers conducted tests on Mod. 9Cr-1Mo steel to obtain data needed to develop modeling for inelastic material behavior suitable for use in high-temperature inelastic structural analyses. Data were measured to determine the values for 12 elastoplastic and viscoplastic parameters in Chaboche's inelastic constitutive equations at distinct temperatures of 500°C, 550°C, and 600°C. The tests were carried out according to the requirements of ASTM E466 using an Instron 8501 loading frame with an extensometer, 8-mm diameter test sections having a 16-mm gage length, strain rate of $1 \cdot 10^{-4}$/s, and a strain range of ± 0.6 percent. The parameters were validated by comparing the inelastic simulations of the tests and the test measurements. Good agreement with the test data was obtained for simulations of stabilized hysteresis loops, cyclic softening behavior, and viscosity/creep behavior.

Researchers used the Chaboche model and a nonlinear iso-hardening model for Type 316 stainless steel to investigate inelastic strain accumulation in the ABTR reactor vessel. The team used the ANSYS code for the same duty cycle loading events and calculated with ANSYS assuming elastic material behavior analyzed during the first year. According to the new inelastic analysis, progressive inelastic strain accumulation in the reactor vessel is negligible. A thermal ratcheting evaluation was carried out for the ABTR redan wall, conservatively assuming that the hot sodium pool free surface repetitively rises and falls by 0.76 m over 1,000 s (up or down) as a result of starting and stopping the primary sodium pumps, and assuming the Chaboche model and a bilinear iso-hardening model for 316 stainless steel.

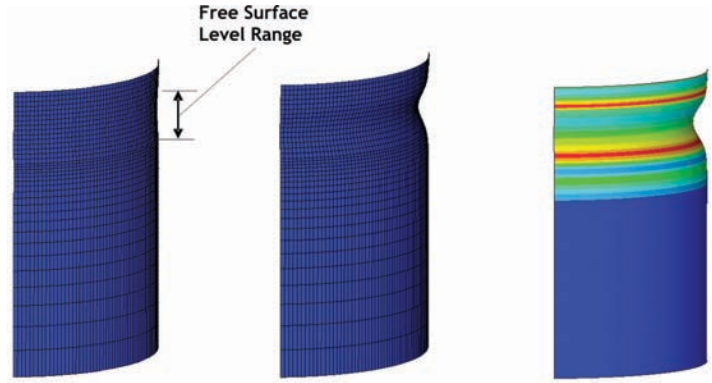


Figure 1. Calculated deflection of ABTR redan wall for inelastic analysis of ratcheting after 1, 10, and 20 cycles of hot sodium pool free surface level change; deflections are magnified by a factor of 20.

The team calculated progressive inelastic strain accumulation such that shakedown was nearly attained after 20 cycles (Figure 1). Researchers carried out inelastic design evaluations for the ABTR redan wall using the ANSYS code with the inelastic material model together with the inelastic analysis methods of the KAERI SIE ASME-NH computer code. The team calculated lower creep-fatigue values than those obtained previously based upon ANSYS results assuming elastic material behavior (Figure 2). Inelastic design evaluations were also performed for the 304 stainless steel ABTR intermediate heat transport system piping, and significant reductions were calculated for the accumulated inelastic strain and creep-fatigue values relative to the previous elastic analysis-based approach. The work carried out during the second year has established practical procedures for application of the inelastic analysis method of the ASME-NH rules.

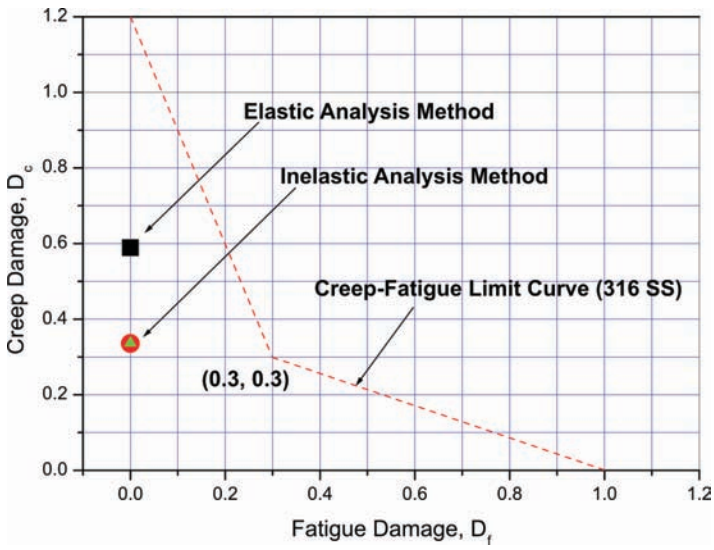


Figure 2. Results of inelastic creep-fatigue evaluation for ABTR redan wall and comparison with previous elastic analysis-based evaluation.

Planned Activities

In the third year of the project, the research team will carry out seismic and buckling instability evaluations. Third-year tasks include development of technology for high-temperature seismic evaluation, evaluations of short-term creep-fatigue damage, and development of methods for high-temperature seismic buckling analysis.

INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE

Advanced Multi-Physics Simulation Capability for Very High-Temperature Gas-Cooled Reactors (VHTRs)

PI (U.S.): Changho Lee, Argonne National Laboratory (ANL)

PI (ROK): Hyun Chul Lee, Korea Atomic Energy Research Institute (KAERI)

Collaborators: Seoul National University (SNU)

Project Number: 2008-001-K

Program Area: Gen IV

Project Start Date: October 2008

Project End Date: September 2011

Research Objectives

The objective of this project is to develop a suite of advanced multi-physics simulation methods and codes to be used for high-fidelity, spatially detailed analyses of the coupled neutronics and thermo-fluid behavior of VHTRs. The scope of this research, which focuses on prismatic block VHTRs, involves 1) developing neutronic and thermo-fluid models that can accurately account for important physical phenomena and 2) the consistent coupling of these models to obtain detailed, highly accurate multi-physics solutions.

The neutronics analysis capabilities will include methods for generating fine-level, multigroup cross sections that account for the double heterogeneity of coated particle fuels, plutonium resonances in the thermal energy range, and the thermal scattering kernel of graphite. The thermo-fluid analysis methods will be developed to accurately predict convective heat removal by the coolant flowing through the block and multi-dimensional conduction within the block. Bypass coolant flow effects in the gap regions between blocks and radiative heat transfer will also be taken into account. Researchers will investigate various turbulence models ranging from lower-order models to higher-order large eddy simulations. A practical multi-resolution approach will be studied by calculating the mean flow characteristics of sub-domains using higher-order models and providing them to lower-order model calculations for the larger solution domain.

Research Progress

High-Performance 3D Whole Core Transport Calculation. For Task 1, researchers at KAERI/SNU updated the DeCART v1.2 code for functionality extension, solution efficiency improvement, stability enhancement, and new cross-section data. The resulting version, v2.0, has the following features distinct from the previous version: 1) a new code structure with FORTRAN90 modules and derived types, 2) a more efficient ray tracing solver with the back tracing, 3) a more effective 3D CMFD transport solver, 4) a better depletion solver with the Krylov exponential method, and 5) a prototype VHTR-specific library (SNU version) using the NJOY and NRSC code systems.

For thermal feedback calculation, KAERI initiated the development of interface modules for coupling DeCART with GAMMA+. Since the modules were not ready for coupled calculations, SNU's task of improving resonance treatment for non-uniform conditions has been postponed until next year. Instead, the VHTR cross-section library generation work was carried out. Regarding the double-heterogeneity (DH) treatment capability of DeCART, a thorough evaluation of the DH effect was conducted with the newly generated VHTR cross-section library. To enhance the solution stability of the planar MOC solution method and the 3D transport solution scheme of DeCART, the team performed a preliminary development of the decoupled planar MOC solution method in which each planar MOC problem is independently solved as an eigenvalue problem.

Adaptation of the SHARP Modeling and Simulation Capability. The first step in Task 2 focused on coupled calculations with the 3D neutron transport code and fluid dynamics code. In this task, ANL updated the DeCART v1.2 to v1.2anl, which includes new features such as periodic boundary condition, anisotropic scattering capability, accurate modeling of large control holes, improvement of numerical stability, 3D CMFD formulation for solution acceleration, simplified VHTR thermo-fluid model, visualization edit, and new cross-section library structure. A detailed output visualization capability using

VisIt was originally planned for the third year, but it was partially accomplished this year for efficient code verification. Although a few updates overlapped with those in v2.0 developed by KAERI, most of them are new.

An initial version of multigroup cross-section libraries tailored for VHTR analysis were developed using the SCALE/CENTRM system of Oak Ridge National Laboratory (ORNL) since the existing 47-group and 190-group cross-section libraries of DeCART were originally prepared for light-water reactor (LWR) applications. With the ORNL system, three new libraries with different numbers of energy groups (49, 72, and 238 groups) were generated using the ENDF/B-VII data, in which the resonance energy range was extended to an energy interval from 0.1 eV to 500 keV.

For verification of the updated DeCART code, 2D and 3D VHTR benchmark problems were developed based on the NGNP/VHTR core concept that adopted the core radial layout and the prismatic block dimensions from the GT-MHR core design. The benchmark problems include four different core configurations such as cores with standard blocks only (STD), all control rods out (ARO), all control rods in (ARI), and operating control rods in (ORI). Figure 1 shows 3D group flux distributions resulting from 3D whole-core calculations. With the ORNL 70- and 238-group cross sections, core multiplication factors from DeCART agreed well with MCNP solutions.

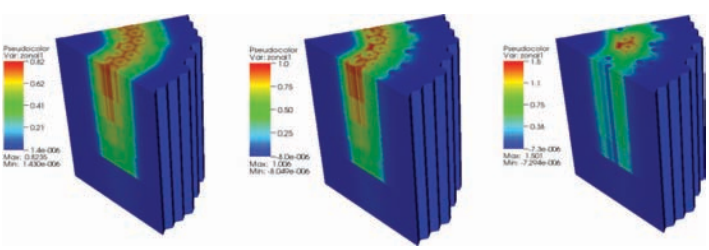
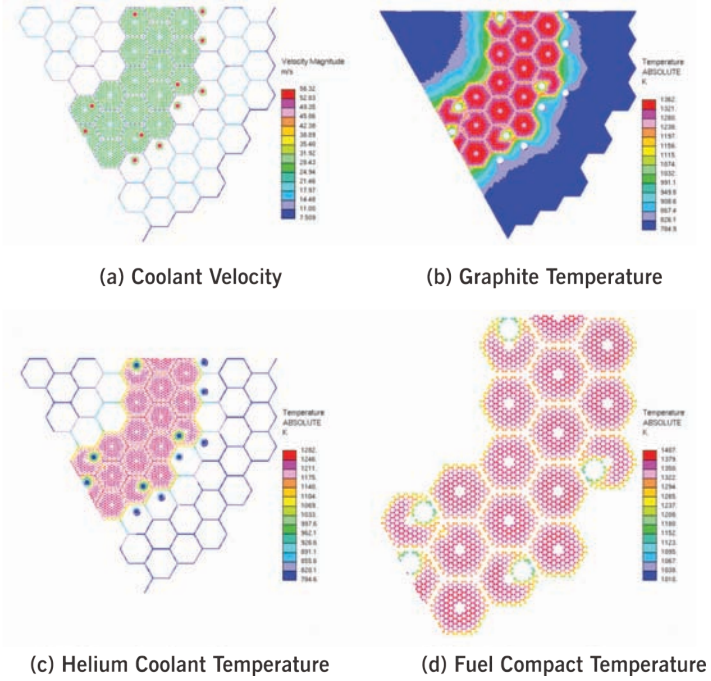


Figure 1. DeCART group fluxes of 3D VHTR cores at ~7 eV.

Development of Thermo-Fluid Methods. For Task 3, researchers at ANL and KAERI defined a preliminary benchmark problem in order to assess the turbulence model's applicability for predicting VHTR flow and temperature fields. Researchers at ANL and KAERI performed the 3D steady-state RANS simulations using the default STAR-CCM+ segregated flow solver, which was the SIMPLE algorithm with the Rhie-Chow interpolation for pressure-velocity coupling and algebraic multi-grid preconditioning. As an initial test of meshing and coupling strategy, a single fuel column model was developed that included 210 fuel compacts and 108 coolant channels within each block. The upper and lower plena are represented explicitly, although upper and lower shielding regions were neglected. Boundary faces of the hexagon were assumed to be reflective symmetry boundaries. The final computational mesh used 8.8 million polyhedral mesh elements. The team also investigated thermo-fluid models suitable for the coupled calculations.

Development of the Coupling Strategy. In Task 4, the DeCART neutron transport code was directly linked to the commercial CFD code, STAR-CD, for multi-physics core analysis applications so that data can be passed from STAR-CD to DeCART by arguments to special subroutines with no need for file input/output. The team at ANL conducted mapping of the STAR-CD and DeCART spatial meshes by assigning each CFD cell to the DeCART region in which its centroid lay, utilizing the native geometry description in DeCART. In initial STAR-CD simulations (Figure 2[a]), significant bypass flow velocities were observed in the gap regions as well as the control rod holes. The effects of these coolant flow paths were clearly visible in the predicted graphite temperature (Figure 2[b]). Little radial mixing occurred within the bypass gap flow, and the coolant temperature rise was primarily isolated to the fueled annulus (Figure 2[c]). Peak coolant temperatures occurred in the gap bypass flow. As a consequence of the large surface area, the much slower bypass flows had a non-negligible impact on local fuel temperatures (Figure 2[d]).



(a) Coolant Velocity (b) Graphite Temperature

(c) Helium Coolant Temperature (d) Fuel Compact Temperature

Figure 2. Predictions from CFD simulations of heat transfer in a 1/6th core model assuming a uniform power distribution.

Verification and Validation. For Task 5, ANL and KAERI developed the 2D and 3D benchmark models based on the NGNP/VHTR design at ANL and the PMR200 reactor design at KAERI. Monte Carlo calculations using MCNP5 were performed for the ANL benchmark models with different control rod conditions. For the KAERI benchmark models, depletion analysis to the equilibrium cycle was conducted using the McCARD Monte Carlo code.

Planned Activities

For Task 1, the team will implement more functionalities necessary for practical VHTR design, such as control rod treatment and fuel block shuffling for reload design, into the DeCART code. The team will also generate a cross-section library specific for the VHTR and initiate development of the transient capability. For Task 2, improvements will be made to DeCART to speed up convergence. The ORNL cross sections will be further examined to enhance accuracy of the DeCART solution. For Tasks 3 and 4, researchers will continue the coupled simulation of 3D VHTR cores with DeCART and STAR-CD. The effort will incorporate additional features of the core physics, including variable thermo-physical properties, buoyancy, and radiative heat transfer. At KAERI, coupling DeCART and GAMMA+ will be completed. For Task 5, researchers will refine the 3D benchmark models developed based on NGNP/VHTR or PMR200 designs. Prismatic-type high-temperature reactor experiments, such as the HTTR, will be analyzed using the coupled system developed.

INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE

Experimental and Analytic Study on the Core Bypass Flow in a Very High-Temperature Reactor

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

PI (Country): Min-Hwan Kim, Korea Atomic Energy Research Institute (KAERI)

Collaborators: Argonne National Laboratory (ANL), Seoul National University (SNU), Texas A&M University (TAMU)

Project Number: 2008-002-K

Program Area: Gen IV

Project Start Date: October 2008

Project End Date: September 2011

Research Objectives

The main objectives of this project are to generate experimental data for validating the software to be used to calculate the bypass flow in a prismatic very high-temperature reactor (VHTR) core, validate thermo-fluid analysis tools and their model improvements, and identify and assess measures to control the bypass flow to within a desired flow envelope. To achieve these objectives, the project team has defined five tasks:

- Design and construct experiments to generate validation data for software analysis tools.
- Determine the experimental conditions and define the measurement requirements and techniques.
- Generate and analyze the experimental data.
- Validate and improve the thermo-fluid analysis tools.
- Identify measures to control the bypass flow to within desired flow envelopes.

During the first year of this I-NERI project, the team addressed the first, second, and fourth tasks.

Research Progress

Design and Construction of Experiments. Researchers at KAERI focused on the construction and testing of a multi-block air test facility. This facility was designed to generate experimental data for validating thermal-fluid analysis tools such as GAMMA+ and GAS-NET. Data from the multi-block facility will include the flow rates in block columns and the associated local pressure distributions. The data sets will include the flow rates at the inlet and outlet of the block columns and the local static pressure of coolant holes and bypass gaps.

Researchers at INL and TAMU focused on the design of matched-index-of-refraction (MIR) experiments. Major objectives of the U.S.-directed bypass flow R&D are to 1) investigate flow behavior in the interstitial region between the prismatic block to gain a clear understanding of the dominant mechanisms that govern the flow in that region and 2) obtain data of sufficient quality to enable computational fluid dynamics (CFD) software to be validated. To achieve these objectives, there must be an intimate understanding of the potential flow behavior in the gaps themselves for a specified geometry—not considering the other variables that ultimately affect the gap flow (e.g., graphite distortion stemming from irradiation as a function of temperature).

To begin the investigation of flow behavior in the core bypass region and to provide support to the effort described below, a number of CFD calculations were undertaken. The CFD calculations were performed, in part, to assist in the design of MIR experiments and also to better understand the bypass flow behavior and its effect on the core temperature distributions. CFD calculations were performed using a one-twelfth section of a single prismatic block and a one-twelfth section of the entire core, plus the upper and lower reflector blocks. In addition to the experiment designed for the INL MIR facility, preliminary experimental designs were also initiated at TAMU.

Determination of Experimental Conditions and Development of Measurement Requirements and Techniques. Researchers at KAERI performed a preliminary estimation of the bypass flow gap sizes for a prismatic-core VHTR.

This work was accomplished by studying how to model the dimensional changes of the graphite blocks. The amount of dimensional change of the graphite can be assumed to be a function of the irradiation fluence and the irradiation temperature. Since the data accrued from the reactor operation and experiments are scattered, to simplify the evaluation procedure in the present study, the graphite is assumed to be isotropic, and a dimensional change at a fixed irradiation temperature can be represented by a single curve. For a given fluence and temperature, therefore, the dimensional changes of the graphite can be determined by a linear interpolation between the curves. Another major factor affecting the core bypass flow is the coefficient of the thermal expansion, which also depends on the neutron irradiation and temperature. Thus, four factors were considered for evaluating the local gap size: 1) installation tolerance of the graphite blocks, 2) thermal expansion of the metallic core support plate, 3) thermal expansion of the graphite blocks, and 4) irradiation shrinkage/swelling of the graphite blocks.

A preliminary analysis of the multi-block air test facility was carried out by researchers at SNU and KAERI. In order to determine the experimental conditions and the measurement requirements, researchers performed similitude analyses and preliminary CFD simulations.

Validation and Improvement of Thermo-Fluid Analysis Tools. The KAERI research team made progress on GAMMA+ modeling and pre-calculation for the core bypass-flow estimation. The team implemented a heat conduction model to calculate the temperature profile in TRISO particles dispersed within a fuel matrix. The implicit coupling method is used to consider heat exchange between a fuel matrix and coated TRISO particles. The multi-D heat conduction model was further improved to consider sub-meshes within hexagonal fuel and reflector blocks. The effect of cross diffusion by non-orthogonal geometry was also considered. The friction and heat transfer correlations for a rectangular flow channel were implemented and used for lateral and cross bypass flow paths. Researchers at KAERI performed the bypass flow and hot spot analysis to investigate the impacts of the variation of bypass gaps during core life cycle as well as the core restraint mechanism on the amount of bypass flow and thus maximum fuel temperature. The core thermo-fluid analysis was performed using the GAMMA+ code for the PMR200 block-core design.

The SNU unit-cell facility was designed to investigate the effect of bypass gap on flow distribution and consists of three coolant-hole blocks. The unit-cell is a regular triangle section that is formed by connecting the center points of three hexagonal blocks in the core. A Y-shaped bypass gap is controlled from 1 mm to 5 mm in the experiment. Since the discharge points of coolant flow and bypass flow are located at different positions, the researchers performing GAMMA+ modeling considered the additional frictional pressure drop through the flow measuring pipes of 120-mm diameter attached to each block.

ANL accomplished several tasks as part of overall project objectives. First, the GAS-NET thermo-fluid analysis tool was significantly improved to prepare for application to the design and optimization of bypass experiments that will be performed. Second, researchers identified and analyzed existing experiment datasets for their use in validating the GAS-NET code. They also selected an experiment and performed simulations that were compared to the corresponding data as a first step to validating the code. Third, computational benchmark problems were identified in collaboration with other project participants for inter-comparing the thermo-fluid analysis tools used in this project.

Planned Activities

The project is on schedule. Activities are shown in Table 1 below in the context of the entire three-year period of the project. For Year 2 (FY 2010), researchers will complete the design and construction of the experimental hardware and initiate testing. The team will use data from the experiments to continue the verification and validation effort.

Table 1. Project activity schedule.

Activity Description	Year 1	Year 2	Year 3
Design and Construction of Experimental Facility			
Experimental Conditions and Measurement Requirement/Techniques			
Testing and Interpretation of Data			
Validation and Improvement TF Analysis Tools			
Integration of System Level Analysis Tools			
Identification of Bypass Flow Measure			
Final Report			

INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE

Nuclear Data Uncertainty Analyses to Support Advanced Fuel Cycle Development

PI (U.S.): Michael D. Dunn, Oak Ridge National Laboratory (ORNL)

PI (ROK): Choong-Sup Gil, Korea Atomic Energy Research Institute (KAERI)

Collaborators: None

Project Number: 2008-003-K

Program Area: FCR&D

Project Start Date: November 2008

Project End Date: September 2011

Research Objectives

This three-year collaborative project will provide improved neutron cross-section data with uncertainty or covariance data for isotopes important for advanced fuel cycle (AFC) and nuclear safeguards applications. Additional goals are to assess uncertainties of the nuclear integral parameters due to the cross-section data, improve safety validation, and reduce capital cost through system design optimization for AFC developments.

To achieve these objectives, the research team will identify priority nuclides that need improved nuclear data with uncertainty information for AFC safeguards and design applications and then produce the improved nuclear data files with covariance data. They will demonstrate and test the improved evaluated data in sensitivity/uncertainty calculations for select benchmark problems. The tests will include, for example, uncertainty assessments due to the nuclear cross-section data of the integral parameters such as k_{eff}, reaction rates, reactivity worths, and spent fuel isotopic compositions for the AFC systems. These tests will also include validation of the cross-section data itself.

Potential benefits of the proposed activity include capital cost reductions through system design optimization, improved technical basis for licensing, improved safety validation, increased design flexibility/optimization, and potentially significant time and cost savings associated with the design and conduct of experimental programs. The final product of this work will be improved and tested nuclear data files with uncertainty information that will be submitted for inclusion in the U.S. Evaluated Nuclear Data File (ENDF/B) system.

Research Progress

In the first year of the project, researchers identified priority nuclides that need improved nuclear data with uncertainty information for AFC analyses and nuclear safeguards applications. Historically, nuclear data improvements have largely focused on major actinides that are important for AFC applications, such as ^{235}U, ^{238}U, and ^{239}Pu. There has been less effort on data improvements for other isotopes/nuclides important for AFC and nuclear safeguards applications (e.g., americium [Am], minor actinides, and other plutonium isotopes such as ^{240}Pu, ^{241}Pu, and ^{242}Pu). This collaboration provides an opportunity to assess the status of the minor actinides for supporting AFC and nuclear safeguards applications.

Because the ORNL work is funded by the U.S. nuclear safeguards program, their investigation largely focused on the data needs for supporting nuclear safeguards applications. The KAERI effort supports the larger AFC application needs for the development of an advanced nuclear fuel cycle.

During the first year, researchers met to discuss data improvements that would benefit both the U.S. and ROK in supporting AFC and nuclear safeguards applications. For nuclear safeguards applications, new measurement and analysis technologies will be needed to monitor and interrogate nuclear systems involving spent nuclear fuel (SNF). Researchers are developing measurement and analysis methods to safeguard SNF that contains significant quantities of uranium, plutonium, and neptunium isotopes. It is essential to simulate the radiation transport processes involving SNF for accurate safeguards analyses. Modeling and simulation is especially dependent

on the underlying cross-section data. Simulation of particle detector counters in material interrogation for nuclear safeguards applications depends on how well researchers understand the basic nuclear data and their uncertainties. For example, although ^{237}Np is a primary candidate isotope for AFC safeguards applications, which will be coupled with plutonium primarily to enable neptunium recycle, ^{237}Np also has a safeguards benefit. Protactinium-233, the decay product of ^{237}Np, emits an easily detected gamma ray that can facilitate detection and monitoring as well as impede attempts to divert material from a nuclear facility.

For active interrogation techniques, detailed nuclear data in the neutron resonance region are needed for key isotopes such as ^{237}Np, ^{244}Cm, plutonium isotopes, americium isotopes, and other minor actinides. In addition, cross-section uncertainty or covariance data are not available for ^{237}Np or ^{244}Cm, as well as many other key isotopes such as ^{241}Pu, ^{242}Pu, and ^{241}Am. Improved nuclear data with covariance data are needed to facilitate radiation transport modeling to support nuclear fuel cycle analyses including safeguards analyses of SNF (e.g., active interrogation). Based on the application needs and ongoing investigations, the team presented the basic nuclear data needs that would be of interest to their respective countries; both countries agreed that data assessment and improvement efforts would be needed for ^{237}Np, ^{240}Pu, and curium isotopes because these cross-section data are important for nuclear safeguards applications and general AFC analyses.

In the first year of the project, researchers performed a detailed assessment and initiated preliminary evaluation work of the cross-section data of these nuclides. As part of the data assessment effort, the teams performed inter-comparisons of cross-section data from recently released nuclear data libraries such as ENDF/B-VII.0, JEFF-3.1, and JENDL-3.3 as well as the EXFOR experimental data from KAERI. A detailed assessment of the latest evaluations of ^{237}Np and ^{240}Pu from these libraries has shown that improvements can be made with available differential data from cross-section measurements. The team also conducted a survey of available covariance data for all nuclides to identify the status of uncertainty information in cross-section data.

The team has initiated efforts to re-evaluate cross-section data for ^{237}Np and ^{240}Pu in the resonance energy region and to produce cross-section data with covariance data for ^{237}Np, ^{240}Pu, and all curium isotopes at energies above the resonance region. The initial evaluation efforts have resulted in the generation of preliminary ^{240}Pu covariance data, which were combined and processed using the latest cross-section processing software. Figure 1 provides a plot of the preliminary covariance data produced during the first year of the project.

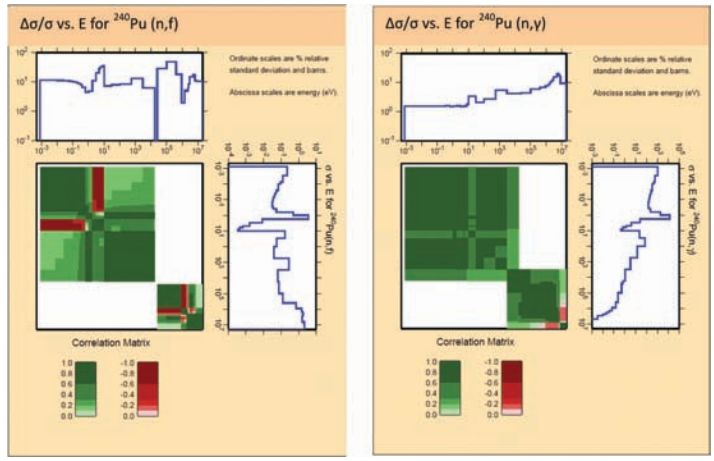


Figure 1. Covariance data of ^{240}Pu (n,f) and ^{240}Pu (n,γ) reactions.

The preliminary covariance data evaluation in Figure 1 will enable the team to perform two key tasks during the project:

1) To facilitate the selection of experimental datasets to use in the cross-section re-evaluation effort that will be performed in the second year of the project.

2) To help demonstrate a process to combine ORNL-resolved resonance data with KAERI's high-energy cross-section data.

In addition to the nuclear data evaluation efforts, KAERI and ORNL have performed benchmark calculations and sensitivity/uncertainty analyses using the latest cross-section data (prior to this work) and covariance data. These initial computation studies have been performed in preparation for testing the new cross-section and covariance data files that will be produced through this collaboration. Moreover, the results from these initial sensitivity/uncertainty analyses will be used in comparison studies with the improved nuclear data files produced as part of the I-NERI project.

The collaborative effort has produced preliminary cross-section and covariance data for ^{237}Np, ^{240}Pu, and $^{240\text{-}250}$Cm in the fast energy region. In summary, the researchers have accomplished the following during the first year of the project:

- Completed an assessment of the latest nuclear data files for AFC applications and determined that nuclear data improvements are needed for ^{237}Np, ^{240}Pu, and curium isotopes.

- Initiated a cross-section evaluation effort for ^{237}Np, ^{240}Pu, and curium isotopes and produced preliminary cross-section covariance data files in the neutron resonance region and high-energy region by ORNL and KAERI, respectively.

- Completed sensitivity/uncertainty analyses using the latest cross-section and covariance data files, which will serve as the basis for testing the new nuclear data files produced in the second year of the project.

Planned Activities

In FY 2010, the researchers will continue producing improved nuclear data files for ^{237}Np, ^{240}Pu, and curium isotopes. The ORNL effort will largely focus on developing new cross-section evaluations for ^{237}Np, ^{240}Pu, and curium isotopes with cross-section covariance data. In the later part of FY 2010 and throughout FY 2011, both labs will perform data-testing activities with the new nuclear data files in radiation transport and sensitivity/uncertainty analyses of AFC systems. At the conclusion of the collaboration in FY 2011, new cross-section evaluations for ^{237}Np, ^{240}Pu, and the curium isotopes will be distributed to the U.S. National Nuclear Data Center for release as ENDF/B files, thereby making the data files readily available to support nuclear safeguards modeling and simulation efforts. In addition, KAERI will be able to utilize the improved cross-section data evaluations for Korean AFC analyses.

INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE

ZPPR-15 and BFS Critical Experiments Analysis for Generation of Physics Validation Database of Metallic-Fueled Fast Reactor Systems

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

PI (ROK): Sang Ji Kim, Korea Atomic Energy Research Institute (KAERI)

Collaborators: None

Project Number: 2009-001-K

Program Area: Gen IV

Project Start Date: October 2009

Project End Date: September 2012

Project Abstract

A three-year collaboration is proposed between ANL and KAERI to generate a physics validation database of integral-type experiments for metallic-fueled fast reactor systems. The objective of the proposed project is to archive and evaluate the integral experiment data, analyze the experiments, and prepare detailed computational models to be used for validating the modern suites of fast reactor design analysis tools that are under development at ANL and KAERI.

A series of mockup experiments for a 330-MWe Integral Fast Reactor (IFR) at ANL under the ZPPR-15 Program, also known as the IFR Benchmark Physics Test Program, will be retrieved and analyzed in this project. The research team will also compile the BFS-73-1 and BFS-75-1 experiments of KAERI carried out as the mockup experiment of KALIMER-150 at the Russian BFS-1 facility.

ANL will participate in KAERI's transuranics (TRU) burner physics experiments, which researchers plan to perform at the BFS-2 facility in FY 2010 to validate the physics performance of its metal-fueled TRU burner reactor design and design analysis tools. This burner reactor physics experiment data will be a valuable addition to the existing experiment database for breeder reactors.

The principal outcomes of this project for the ZPPR-15, BFS-73-1, BFS-75-1 experiments will include:

- Compiled sets of measured physics parameters and uncertainties
- Consistent sets of as-built Monte Carlo models
- A mock-up of a 300-MWe-rated TRU burner reactor

Since both the United States and the Republic of Korea are interested in a TRU-burning option using metal-fueled fast reactors, it would be mutually beneficial to generate the physics validation database of metal-fueled fast reactors using the ZPPR-15 experiments and the planned TRU burner physics experiments at the BFS-2 facility.

INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE

Enhanced Radiation Resistance Through Interface Modification of Nano-Structured Steels for Gen IV In-Core Applications

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

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

Collaborators: Texas A&M University (TAMU), Seoul National University (SNU)

Project Number: 2009-002-K

Program Area: FCR&D

Project Start Date: October 2009

Project End Date: September 2012

Project Abstract

The project team will conduct three years of research through international collaboration between UW, TAMU, KAERI, and SNU to investigate enhancement of radiation resistance of in-core candidate materials for Generation IV reactors. Austenitic stainless steels have good high-temperature mechanical properties, fair compatibility with various coolants, and abundant field experience that is captured in material databases. However, they are restricted from applications as certain high-dose in-core components of future nuclear reactors because of their poor radiation resistance, resulting specifically in swelling and possibly in brittle failure at lower temperatures.

This project suggests nanocrystalline austenitic steels created by equal channel angular pressing (ECAP) of an Fe-14Cr-16Ni alloy to improve radiation resistance of austenitic steels. The radiation resistance would be attributed to a reduced grain size and drastically increased grain boundary area, with high-angle grain boundaries acting as sinks for radiation-induced point defects. The project team will also pursue grain boundary engineering to modify and improve the grain boundary character distribution to reduce radiation-induced cavity growth as well as to improve the interface-related properties, such as high-temperature mechanical properties. To estimate the radiation resistance of the ECAP-processed and grain boundary-engineered alloys, researchers will conduct helium and proton irradiation tests for temperatures up to 600°C. The team will also investigate the effects of nano-sized precipitates in HT-UPS steels and nano-sized oxide dispersoids in oxide dispersion-strengthened alloys with the nanocrystalline and grain boundary-engineered austenitic steels.

For the characterization of the nanocrystalline, grain boundary-engineered, and ion-irradiated samples, researchers will utilize x-ray diffraction, differential scanning calorimetry, various electron microscopy techniques (including field emission scanning electron microscopy, high-resolution transmission electron microscopy, and scanning transmission electron microscopy/energy dispersive spectroscopy/electron energy loss spectroscopy), and analytical techniques with neutron sources such as high-resolution powder diffraction and small-angle neutron scattering. Nanoindentation techniques, bulge tests, and high-temperature tensile tests will be also conducted to evaluate the samples' mechanical behavior.

For these studies, participating organizations from the United States and the Republic of Korea will contribute relevant expertise and facilities to the research team to investigate the fundamental mechanisms of grain boundary-enabled radiation tolerance. The optimal conditions of material and processing will be established for enhanced radiation resistance without compromising mechanical properties or compatibilities. After screening with ion irradiation tests of nanocrystalline alloy samples with optimum performance, the team will submit a proposal to the Advanced Test Reactor National Scientific User Facility program to evaluate the neutron irradiation resistance of the materials.

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