

2008

University of Nevada, Las Vegas Transmutation Research Program Annual Progress Report Academic Year 2007-2008

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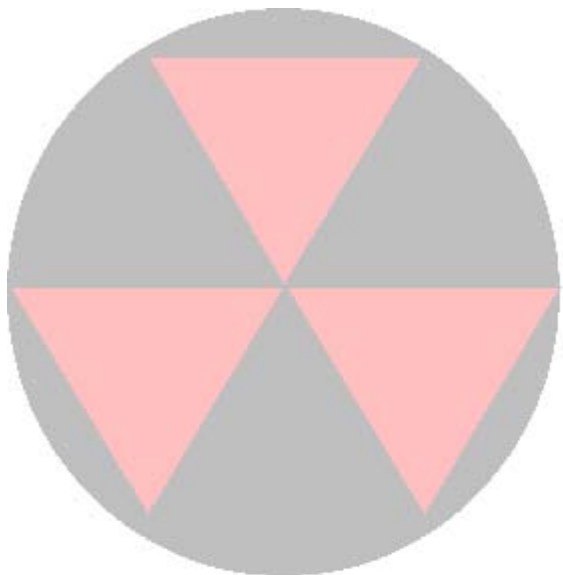
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University of Nevada, Las Vegas
Transmutation Research Program
Annual Progress Report
Academic Year 2007-2008

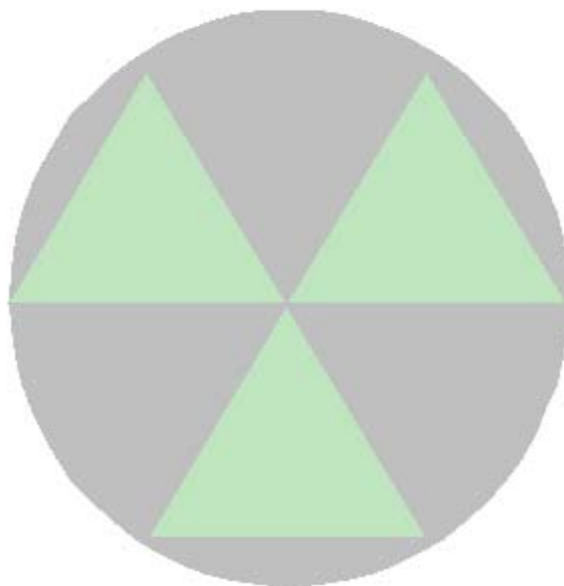


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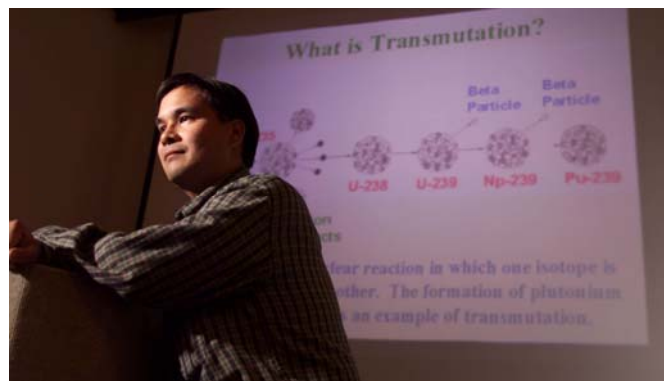
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Letter from the Director



Anthony E. Hechanova, Ph.D.
Director, UNLV Transmutation Research Program



It is my pleasure to present the UNLV Transmutation Research Program's seventh annual report that highlights the academic year 2007-2008. Supporting this document are the many technical reports and theses that have been generated over the past seven years, which can be found on our program's website at <http://aaa.nevada.edu>.

In the seventh year of our program, we continued to see growth in the Radiochemistry Ph.D. program with a total of 20 students in the fourth year of the program (we anticipated twelve in the program proposal). Since our inception, the program has sponsored to their conclusion 48 M.S. and 6 Ph.D. degrees. The program supported 53 graduate students, 11 undergraduates, and eight post-doctoral scholars in eight academic departments across the UNLV scientific and engineering communities in the academic year 2007-2008.

Our research tasks span the range of technology areas for transmutation, including separation of actinides from spent nuclear fuel, methods of fuel fabrication, reactor-accelerator coupled experiments, corrosion of materials exposed to lead-bismuth eutectic, and special nuclear materials protection and accountability.

We continued our emphasis on molten metal technology and actinide chemistry in our enhancements to UNLV this year to build a foundation in areas that are in line with UNLV's strategic growth and our ability to address student-appropriate research in the transmutation program.

Finally, I believe that through the Transmutation Research Program, UNLV has established itself as the core university participant in transmutation research with active collaboration with national laboratories, universities, and international institutions.

I congratulate our students and faculty on a job well done and I look forward to the opportunities and challenges of this exciting research endeavor.

Sincerely,

A handwritten signature in black ink that reads "Anthony E. Hechanova". The signature is written in a cursive, flowing style.



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UNLV Transmutation Research

The UNLV Transmutation Research Program was established in March 2001 as part of the national transmutation program (currently called the Global Nuclear Energy Partnership of the U.S. Department of Energy, Office of Nuclear Energy) to develop the technologies necessary for the ecological and economical treatment of spent nuclear fuel.

The goal of the UNLV program is to develop a sustainable and robust research community at UNLV that is able to contribute to and support the national effort through faculty-supervised, student-conducted research. In establishing the UNLV program, four basic principles were followed: peer review, program focus, integration with the national program, and commitment to the long-term goals of the university.

The primary role of the UNLV program in the national effort is the training of post-doctoral fellows and graduate and undergraduate students in nuclear engineering and other related fields to support the augmentation of the human infrastructure for transmutation technologies. However, the research conducted by UNLV faculty and students is also an integral part of the national research effort. This is accomplished through the direct involvement of students in collaborative research supporting the national transmutation research and development program.

Twenty-seven independent student research tasks, supporting 53 graduate students and 11 undergraduates, were initiated, ongoing, or concluding this academic year. The UNLV program includes the involvement of 30 faculty in eight academic departments.

UNLV research tasks span the range of technology areas for transmutation: separations of used fuel materials, fuel fabrication, accelerator design, materials corrosion and performance, neutron multiplicity and transport simulations and experiments, lead-bismuth eutectic technology development, material protection, accounting, and control, and waste forms.

Student research and training is the primary focus of the UNLV Transmutation Program. To support this focus, the student re-



Post-doctoral scholar Frederic Poineau (left) and Los Alamos National Laboratory scientist Efrain Rodriguez (right) prepare glassware to be used in experiments with technetium.

search program is augmented by the remaining components of the UNLV Transmutation Research Program: Research Infrastructure Augmentation, International Collaboration, and Program Support.

These components are responsible for all aspects of the program that are not directly related to the student research projects, including the expansion of the research faculty, bringing new research equipment to campus, assisting in the development of new student research projects, coordination with the national and international programs, as well as handling the administrative details required for a program of this size.

The primary goals of these components are to support the current UNLV research efforts in transmutation technologies, to augment faculty and staff, and to increase research infrastructure necessary for the strategic growth of the UNLV research mission.

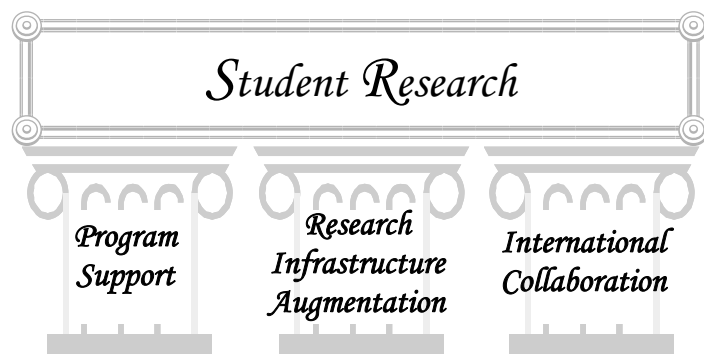


UNLV hosted the Nuclear Science Merit Badge Day at UNLV and the Atomic Testing Museum on November 3, 2007.



UNLV Radiochemistry Ph.D. graduate student (and Eagle Scout) Troy Robinson explains atomic and nuclear science to Boy Scouts at the Nuclear Science Merit Badge Day at UNLV and the Atomic Testing Museum on November 3, 2007.

Program Overview



UNLV Transmutation Research Program Components



UNLV Radiochemistry Ph.D. graduate student Rich Gostic presents his research at the Second Integrated Symposium between UNLV and NSTec (the M&O contractor to the Nevada Test Site) on February 22, 2008.



UNLV students and faculty and other participants from Nevada preparing to tour the San Onofre Nuclear Generating Station in Southern California on August 9, 2007.



UNLV Radiochemistry Ph.D. graduate student Narek Gharibyan, displays a model of a used nuclear fuel canister-containing truck at the Town of Pahrump Fall Festival held October 5-7, 2007.

The program support component sponsored a number of workshops and meetings this year between UNLV researchers and scientists from our national laboratory partners and the international community for the development of transmutation technologies, as well as sponsoring the participation of UNLV students in the American Nuclear Society Student Conference at Texas A&M in College Station, TX, and the American Chemical Society's National Meeting in New Orleans, LA.

The 2007-2008 academic year was a busy year that included extracurricular activities organized by the students such as a field trip to three nuclear facilities in the San Diego, CA, area: the San Onofre Nuclear Generating Station, the General Atomics DIII-D Fusion Facility, and the USS Topeka Nuclear Submarine, August 9-11, 2007; an annual outreach at the Town of Pahrump Fall Festival from October 5-6, 2007; and, a Radiochemistry Recruiting event February 28-29, 2008.



UNLV graduate students touring the DIII-D Fusion Facility at the General Atomics site in La Jolla, CA, August 10, 2007.

Transmutation at a Glance

Introduction

Over 20% of the electricity generated in the U.S. is provided by nuclear power reactors. It is estimated that the amount of used nuclear fuel in the U.S. will reach 140,000 tons by the end of the operational period of current reactors.

Many countries take different approaches to the management and recycling of used nuclear reactor fuel. The U.S. is pursuing a strategy of waste management that would place used nuclear fuel in deep geologic repositories for a long time, separating it from the biosphere and allowing the radioactive isotopes of the waste to decay to more stable progeny.

Transmutation is an alternative waste management strategy undergoing research and development in the U.S. The new national program was authorized by Congress to begin in fiscal year 2001, as the Advanced Accelerator Applications program. In fiscal year 2002, the national program was continued and expanded by Congress, and renamed the Advanced Fuel Cycle Initiative (AFCI). In February 2006, the Bush administration announced a broad program named the Global Nuclear Energy Partnership (GNEP) that effectively expanded the AFCI program (which was renamed Advanced Fuel Cycle Research and Development (R&D)) and accelerated the development of commercial-scale facilities. The goals of the GNEP program are to:

- Provide abundant energy without generating carbon emissions or greenhouse gases.
- Recycle used nuclear fuel to minimize waste and reduce proliferation concerns.
- Safely and securely allow developing nations to deploy nuclear power to meet energy needs.



The Global Nuclear Energy Partnership logo. More information about the national program can be found at www.gnep.energy.gov.

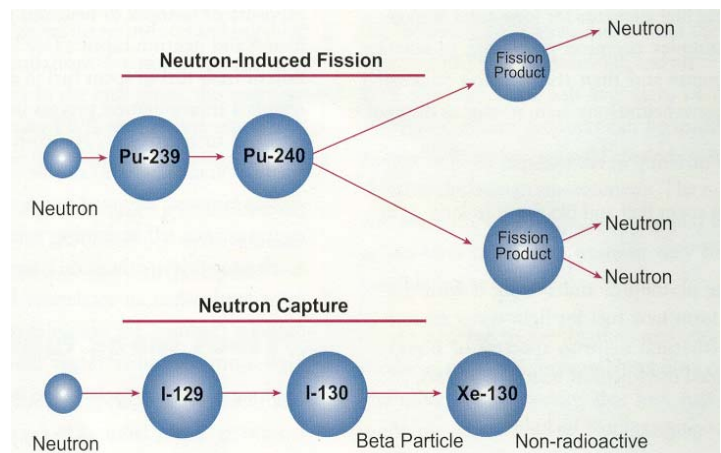
- Assure maximum energy recovery from still-valuable used nuclear fuel and reduce the number of required U.S. geologic waste repositories to one for the remainder of this century.

This concept could provide a dramatic shift in the U.S. waste management strategy by lessening the inventory of residual radioactivity, allowing recycling of fuel, providing optimization of final waste forms, and potentially removing much of the material from Nevada.

What is Nuclear Transmutation?

For centuries, alchemists have been trying to transform elements into other elements, primarily lead into gold. With modern nuclear science, this can finally be accomplished. Scientists are using nuclear transmutation to change one isotope into another more favorable isotope by changing its nuclear structure. This process for waste management is aimed at plutonium, other actinides, and long-lived fission products, with the ultimate goal of converting them into short-lived isotopes that can be managed over a reasonable timeframe.

Transmutation can be done with two different processes: neutron-induced fission and neutron capture. Both processes start with the target nucleus absorbing an incident neutron. After the neutron is absorbed by the target nucleus, the nucleus can either fission (splitting of the nucleus) or go through another nuclear transmutation process such as radioactive decay. Both processes lead to the same final result: transmutation of waste. These processes are illustrated in the figure below.

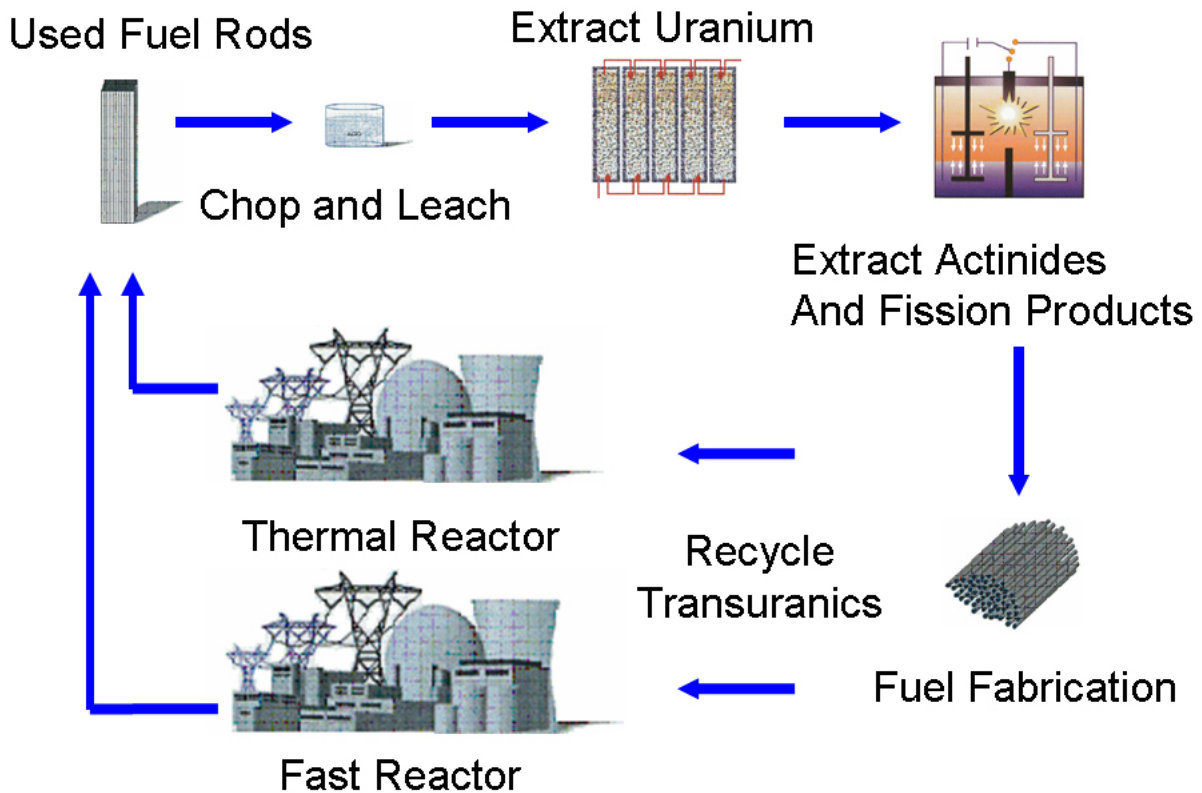


This picture illustrates how neutrons are used to transmute actinides and fission products.

Neutron-Induced Fission (top): a neutron is captured by a fissile actinide (e.g. Plutonium-239) and is induced to split (or fission).

Neutron Capture (bottom): a neutron is captured by a nucleus (e.g. Iodine-129). A new nucleus is produced, Iodine-130, which decays into Xenon-130 which is a stable isotope (i.e., not radioactive). This decay process is complete within a few days.

Multi-tier Transmutation Concept



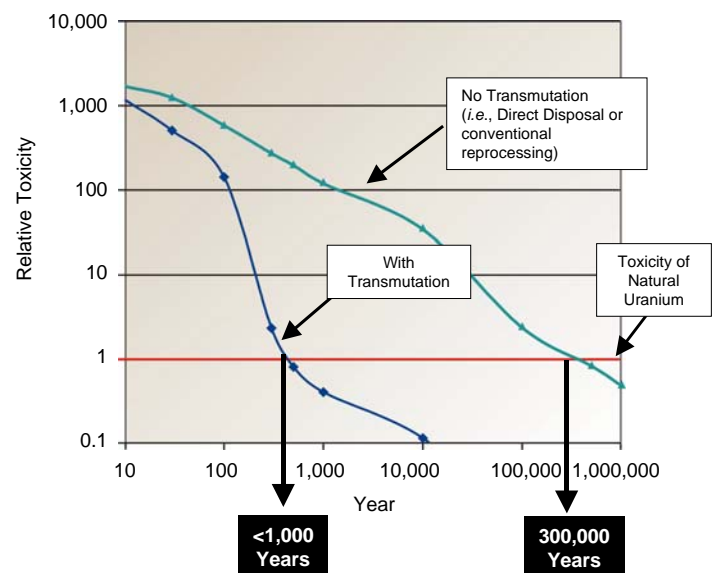
The above figure shows how used nuclear fuel could be managed under a multi-tiered transmutation strategy. In the first tier, plutonium (and other fissile actinides) would be recycled from used fuel and transmuted in an advanced thermal reactor. In the second tier, remaining long-lived radioactive isotopes would be transmuted using a fast spectrum reactor. The GNEP program is currently focused on sodium-cooled fast burner reactors to transmute the recycled transuranic actinides (such as plutonium and americium).

RECYCLE, REDUCE, RESEARCH

Benefits from Transmuting Nuclear Waste

Many benefits are obtained from transmutation of nuclear waste. Some of them are:

- (1) The initial amount of high-level radioactive waste will be reduced by virtue of separation (only two percent of used nuclear fuel is highly radioactive, the rest, if separated, can be classified as low-level or non-radioactive).
- (2) The radiotoxicity of the residual waste will be reduced such that it could be less radiotoxic after 300 years than direct disposal of used nuclear fuel after 100,000 years.
- (3) Usable energy is produced by destroying hazardous components of used nuclear fuel. Plutonium and other isotopes can be continuously recycled.
- (4) Someday other used nuclear fuel materials, such as uranium, may be reused. It is possible that other isotopes separated from nuclear waste could have a useful purpose in medicine and industry.



Incentives for recycling used nuclear fuel and “closing” the nuclear fuel cycle include lowering the radiotoxicity of the residual waste, shortening the time frame of concern, smaller amounts of radioactive waste, the elimination of plutonium, and the ability to make optimized waste forms.

Task 1

Design and Analysis of a Process for Melt Casting Metallic Fuel Pins Incorporating Volatile Actinides

Y. Chen, D.W. Pepper, and R. Clarksean

BACKGROUND

The incorporation of non-fertile actinides into a fuel matrix for a transmuter blanket is of interest to the Advanced Fuel Cycle Initiative. One of three proposed candidate matrices for the transmuter is a metallic alloy fuel matrix. Metallic fuels are an outstanding candidate for a transmutation fuel due to excellent irradiation performance and ease of fabrication. However, incorporating a volatile constituent during fabrication of these or other fuel pins presents a challenge.

Volatile actinides, particularly americium, are susceptible to rapid vaporization during the traditional metal fuel casting processes. The actinide vapors boil off, and flow out of the system into the off-gas recovery system, resulting in only a fraction of the volatile actinide charge being incorporated into the fuel pins. The loss of these actinides from the fuel greatly complicates the task of preparing them for transmutation, requiring additional recovery and fuel fabrication steps to try to incorporate the volatile actinides into the transmuter fuel.

RESEARCH OBJECTIVES AND METHODS

The goal of this project was to investigate the casting processes for metallic fuels to help design a process that minimizes the loss of the volatile actinide elements from the fuel. The research effort centered on the development of advanced numerical models to assess conditions that significantly impact the transport of volatile actinides during the melt casting process and represents a joint effort between researchers at UNLV and Argonne National Laboratory (ANL). Assessing critical equipment and process variables is required to build a successful system that will operate efficiently.

RESEARCH ACCOMPLISHMENTS

Development of the induction-heating model

Modeling efforts centered on the development of the governing equations, incorporating these equations into computer codes, setting up a test problem, and making preliminary calculations for the geometry of interest. Detailed analyses were conducted for an induction skull melter previously built and tested by ANL.

Modeling of casting process

Efforts continued to improve a model for the casting of fuel pins. Work considered the flow of the melt into the mold and heat transfer into the mold during solidification (after flow has stopped). Results from an energy balance model indicate that the thermal mass would typically be greater than needed to solidify

the melt within the mold. The results of this simple model have aided in designing a mold to hold and solidify the fuel pins. Detailed flow and heat transfer models show how the melt flows into the mold and ultimately solidifies.

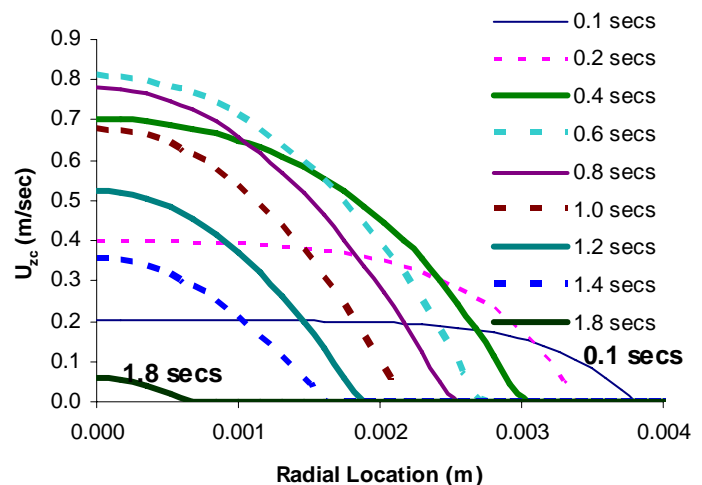
In order to test the impact of process parameters (temperature, pressure, alloying elements, etc.) on the casting process, parametric studies were carried on different processing parameters. These studies centered around model development and analysis of the impact of mold preheating on heat transfer into the model. Results will assist with determining which process parameters are critical in manufacturing a suitable metallic fuel pin.

Americium Transport Models

A model that analyzes the transport of americium from the melt to the vapor phases above the crucible was also developed. The model considers mass transport in the melt, vaporization at the surface, and transport through the vapor phase. Parametric studies were performed to evaluate the impact of different properties or situations on the transport of americium from the melt.

Develop Prototype Furnace Design

A preliminary furnace design that can be built and tested with surrogate materials is critical in order to assess the viability of metal fuels. A preliminary analysis of potential surrogate materials was completed. Manganese appears to be an acceptable surrogate material. Discussions will be held with Argonne National Laboratory staff members to insure that no health and safety issues prevent manganese from being used.



Axial velocity profile for constant inlet pressure of 20 kPa (Mold temperature = 400°C, initial melt temperature = 1500°C, interfacial heat transfer coefficient = 2,000 W/m²-K).

TASK 1 PROFILE

Start Date: June 2001

Completion Date: June 2004

Theses Generated:

Xiaolong Wu, M.S., Mechanical Engineering, "Design and Analysis for Melt Casting Metallic Fuel Pins Incorporating Volatile Actinides," August 2002.

Taide Tan, M.S., Mechanical Engineering, "Design and Simulation of an Induction Skull Melting System," May 2004.

Journal Article:

T. Tan, R. Clarksean, Y. Chen, S. Hsieh, and M. Meyer, 2004, "Simulation of the Casting Process for Melt Casting a Metallic Fuel Pin," submitted to *Nuclear Engineering and Design*.

Conference Proceedings:

T. Tan, R. Clarksean, Y. Chen, S. Hsieh, and M. Meyer, "Analyzing for Casting Volatile Actinides," 6th International Topical Meeting on Nuclear Reactor Thermal Hydraulics, Operations and Safety, Nara, Japan, October 4-8, 2004.

T. Tan, R. Clarksean, Y. Chen, S. Hsieh, S., and M. Meyer, "Numerical Simulation Induction Heating Processing an

Induction Skull Melting Furnace," International Symposium on Advances in Computational Heat Transfer, Norway, April 19-24, 2004.

T. Tan, R. Clarksean, Y. Chen, S. Hsieh, and M. Meyer, "Simulating the Casting Process for Melt Casting a Metallic Fuel Pin," 12th International Conference on Nuclear Engineering (ICONE-12), Arlington, VA, 25-29, 2004.

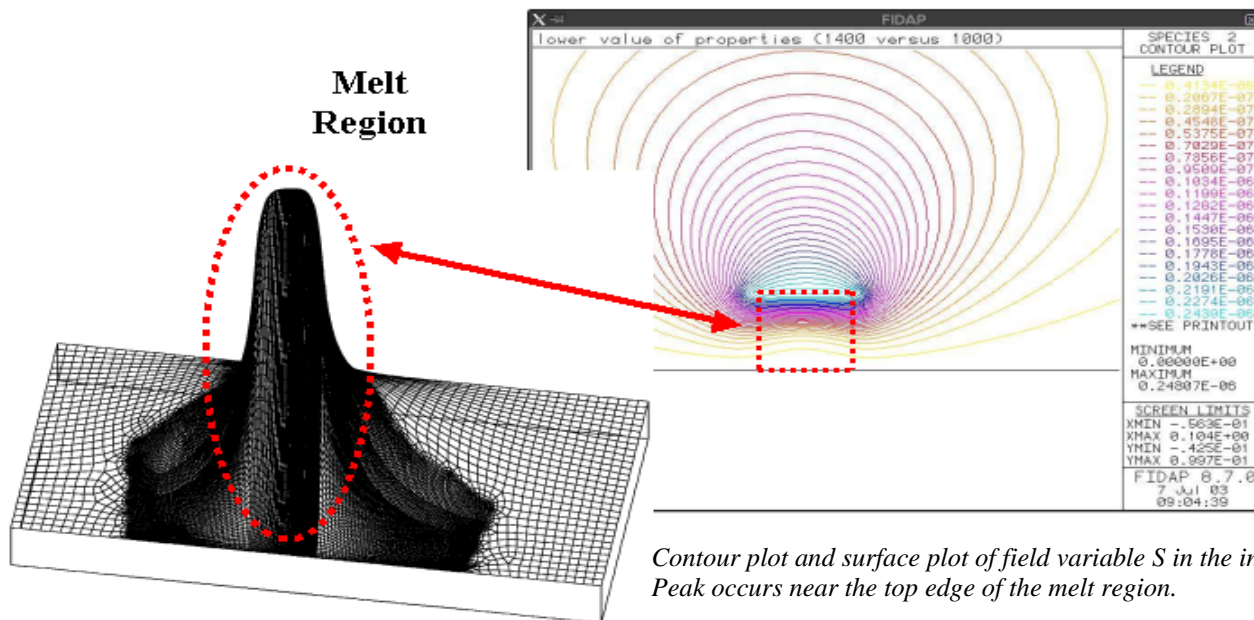
T. Tan, R. Clarksean, Y. Chen, S. Hsieh, and M. Meyer, "Simulating the Induction Heating Process in an Induction Skull Melter for Casting Metallic Fuel," ICONE-12, Arlington, VA, April 25-29, 2004.

T. Tan, R. Clarksean, Y. Chen, D. Pepper, and M. Meyer, "Simulation and Analysis for Melt Casting a Metallic Fuel Pin Incorporating Volatile Actinides" International Mechanical Engineering Congress and Exposition, Washington, DC, November 16-21, 2003.

X. Wu, R. Clarksean, Y. Chen, D. Pepper, and M.K. Meyer, "An Analysis of the Melt Casting of Metallic Fuel Pins," International Mechanical Engineering Congress and Exposition, New Orleans, LA, November 17-22, 2002.

X. Wu, R. Clarksean, Y. Chen, and D. Pepper, "Design and Analysis for Melt Casting Metallic Fuel Pins Incorporating Volatile Actinides," International Youth Nuclear Congress, Daejeon, Korea, April 16-20, 2002.

X. Wu, R. Clarksean, and Y. Chen, "Design and Analysis for Melt Casting Metallic Fuel Pins Incorporating Volatile Actinides," International Congress on Advanced Nuclear Power Plants, Hollywood, FL, June 9-13, 2002.



Research Staff

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Darrell W. Pepper, Professor, Mechanical Engineering Department; Director, Nevada Center for Advanced Computational Methods

Randy Clarksean, Adjunct Professor, Mechanical Engineering Department

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Collaborators

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Steven L. Hayes, Manager of Fuels & Reactor Materials Section, Nuclear Technology Division, Argonne National Laboratory

Task 2

Modeling, Fabrication, and Optimization of Niobium Cavities

R.A. Schill, Jr., M.B. Trabia, and W. Culbreth

BACKGROUND

One of the key technologies for the deployment of accelerator-driven transmutation systems is the accelerator itself. Elliptical superconducting niobium cavities are used to increase the efficiency of the high-power accelerators needed to support the transmutation mission.

One of the major sources of energy loss from a superconducting accelerator cavity is a process known as multiple impacting (or “multipacting”) of electrons. This phenomenon limits the maximum amount of energy and power that the niobium cavity can store. As a result, the maximum power available for accelerating the desired charge, as well as the overall performance of the accelerator is reduced. Furthermore, the energy absorbed as a result of multipacting eventually turns into heat. This negatively impacts the performance of both the superconducting cavity and the accelerator.

RESEARCH OBJECTIVES AND METHODS

This project was tasked with examining the impacts of the design and fabrication technologies for these elliptical niobium cavities on their performance. Niobium was selected primarily due to its behavior at low temperatures.

One objective of this study was to experimentally model the fluid flow resulting in the chemical etching of niobium cavities with the aid of a baffle. Numerical analyses tend to show that the current etching process with a baffle does not uniformly etch the cavity surface. Multiple cavity cell geometries were investigated. Optimization techniques were applied in search of the chemical etching processes, which would lead to cavity walls with near

ideal properties.

A Monte Carlo secondary electron emission (SEE) code was modified to provide a statistical study of electron impact from Los Alamos National Laboratory (LANL) cleaned niobium samples at near cryogenic temperatures in an ultra high vacuum environment. A one-of-a-kind secondary electron emission test stand was developed to study, in part, the dynamics of the emitted particles subjected to an electron beam. Coordinating code studies with experiments offers a wealth of knowledge regarding the surface physics of the material that can enhance modeling codes at LANL.

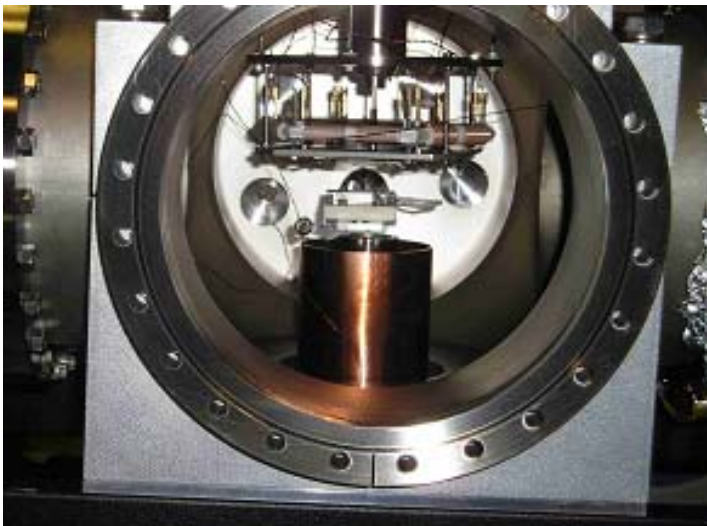
Modeling codes, optimization techniques, and experimentation provided UNLV researchers with a well-rounded study to examine existing and novel niobium cavity designs and cleaning processes for the superconducting radio frequency high-current accelerator.

RESEARCH ACCOMPLISHMENTS

Monte Carlo SEE Code Development and Modifications: A Monte Carlo secondary electron emission code based on the “Single Scatter” approach was obtained from Dr. David Joy of both Oak Ridge National Laboratory and the University of Tennessee. The “Single Scatter” approach follows the primary electron and each generation of secondary electron through the entire cascade. The code was modified to allow for surface layer contaminants in the hopes to verify experimental studies. The code only offers approximate results since the inherent mechanisms internal to the code are valid at higher primary beam energies. A SEE code was used to study the initial particle trajectory conditions of the secondary electron released by the primary electron impacted niobium sample. Results were compared with experiments.

SEE Studies: Niobium samples were cleaned both at LANL and Cornell University and used for experimentation. Two types of cleaning procedures have been used: electro-polishing and buffered chemical polishing. *In situ* pictures of the sample surface were taken with a long-range light microscope for future comparisons.

The experimental setup is unique offering further insights to the SEE process not attained elsewhere. Individual electrons are detected and mapped over the detector surface. With the aid of particle tracking codes, knowledge of the final condition of the electron suggests a family of initial particle trajectory conditions of the emitted electron. Using the Monte Carlo SEE code, the family of initial conditions were evaluated. By March 2004, many of the components of the experimental study reached UNLV with some assembly accomplished. The first SEE measurement was made from the surface of a Faraday cup in September 2004. In December of 2004, the software for the particle positioning detector was finally up and running. The integrity of the code and detector were fine-tuned and initial

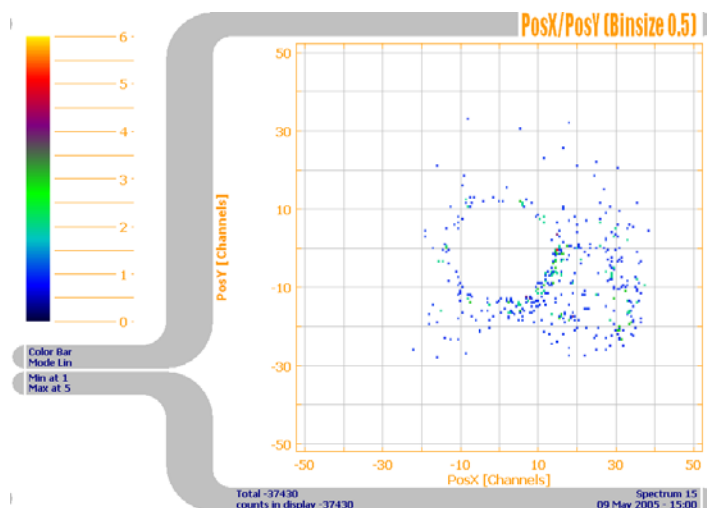


Side view of the secondary electron emission system. The components from top to bottom are: electron gun end, electron beam tube, particle position detector, micro-channel plate stack, business end of the manipulator arm, top of cryostat, and cryostat thermal guard.

experiments were completed by April 2005. Experiments on the surface cleaned samples were completed in May of 2005 experimental studies were completed on the samples provided by LANL.

Revised Etching Process: The current etching method, which uses a baffle to direct the etching fluid toward the surface of the cavity, partially succeeded in achieving its task. However, flow was restricted to the right half of the cavity with very limited circulation in the left half. An alternative design was proposed and modeled. The exit flow is now parallel to flow inlet. Results show that flow circulation was eliminated. The flow is now closer to the surface of the cavity. Optimization techniques were used to improve this design.

Developed Flow Visualization System: To confirm the predictions from the fluid flow models used to analyze the etching process, the UNLV team developed and deployed a flow visualization system. A complete experimental setup, including a computer-controlled x-y traverse and digital camera, was assembled. Flow visualization experiments using a plastic prototype of the niobium cavity used dye injection. Dye injection provided quantitative verification that laminar flow exists within the niobium cavities during etching. Additionally, it verified the absence of re-circulation pockets within the cavities.



Typical secondary electron emission detected when a 100 ms pulse, 1 keV electron beam impinges on a 30 degree incline, virgin surface of niobium before in situ sample baking. Each pixel represents a spatial bin on the detector. The color of the pixel corresponds to the number of electrons detected at that position. The sample was buffered chemically polished. Single count events have been removed to enhance the scattering tendency.

TASK 2 PROFILE

Start Date: June 2001

Completion Date: July 2005

Theses Generated:

Qin Xue, M.S., Mechanical Engineering, "Modeling and Simulation of the Chemical Etching Process in Niobium Cavities," August 2002.

Satish Subramanian, M.S., Mechanical Engineering, "Modeling, Optimization, and Flow Visualization of Chemical Etching Process in Niobium Cavities," May 2004.

Anoop George, M.S. Electrical and Computer Engineering, "Study of Secondary Electron Emission from Niobium at Cryogenic Temperatures," August 2005.

Conference Proceedings:

A. George, R.A. Schill, Jr., R. Kant, and S. Goldfarb, "Secondary Electron Emission from Niobium at Cryogenic Temperatures," International Conference on Plasma Science 2005, Monterey, CA, June 20-24, 2005.

M.B. Trabia, W. Culbreth, S. Subramanian, and T. Tajima, "Optimization of Chemical Etching Process in Niobium Cavities," *Proceedings, American Society of Mechanical Engineers 2004 Design Engineering Technical Conferences and Computers and Information in Engineering Conference*, Salt Lake City, UT, September 28 – October 2, 2004.

M. Holl, M. Trabia, and R.A. Schill, Jr., "Optimization of a Five-Cell Niobium Cavity," Sixth International Topical Meeting on the Nuclear Applications of Accelerator Technology, San Diego, CA, June 1-5, 2003.

Q. Xue, S. Subramanian, M. Trabia, Y.T. Chen, and R.A. Schill, Jr., "Modeling and Optimization of the Chemical Etching Process in Niobium Cavities," International Congress on Advanced Nuclear Power Plants, Hollywood, FL, June 9-13, 2002.

Special Commendation:

A. George and R.A. Schill, "Preparation Studies for Secondary Electron Emission Experiments on Superconducting Niobium," American Nuclear Society Student Conference, April 1-4, 2004, Madison, WI. (Outstanding Student Paper Award)

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William Culbreth, Associate Dean for Research, College of Engineering

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Collaborator

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Task 3

Corrosion of Steel by Lead Bismuth Eutectic

J.W. Farley, A. Johnson, and D.L. Perry

BACKGROUND

There is an active international interest in lead-bismuth eutectic and similar liquid lead systems because of the relevance to the transmutation of nuclear waste, fast reactors, and spallation neutron sources.

Materials in these systems must be able to tolerate high neutron fluxes, high temperatures, and chemical corrosion. For lead-bismuth eutectic (LBE) systems, there is an additional challenge because the corrosive behaviors of materials in LBE are not well understood. Most of the available information on LBE systems has come from the Russians, who have over 80 reactor-years experience with LBE coolant in their Alpha-class submarine reactors. The Russians found that the presence of small amounts of oxygen (on the order of parts per million) in the LBE significantly reduced corrosion. However, a fundamental understanding and verification of its role in the corrosion of steels is incomplete.

RESEARCH OBJECTIVES AND METHODS

This research program has analyzed various steel samples that have been exposed to lead bismuth eutectic as part of the national program to develop LBE and allied technologies. The goal of this research was to understand the basic science of corrosion in the steel/LBE system. This information will be paramount in developing engineering efforts to control, avoid, and/or minimize the effect of corrosion of steels by LBE in transmuter and LBE systems. Additionally, this program provided UNLV researchers with hands-on experience that will be crucial in developing the UNLV molten metal program.

Investigators performed post-experiment testing and analysis on steel samples that have been exposed to lead bismuth eutectic. Surface analysis techniques were employed that included scanning electron microscopy, energy dispersive X-ray

spectroscopy, X-ray photoelectron spectrometry, and laser Raman spectrometry.

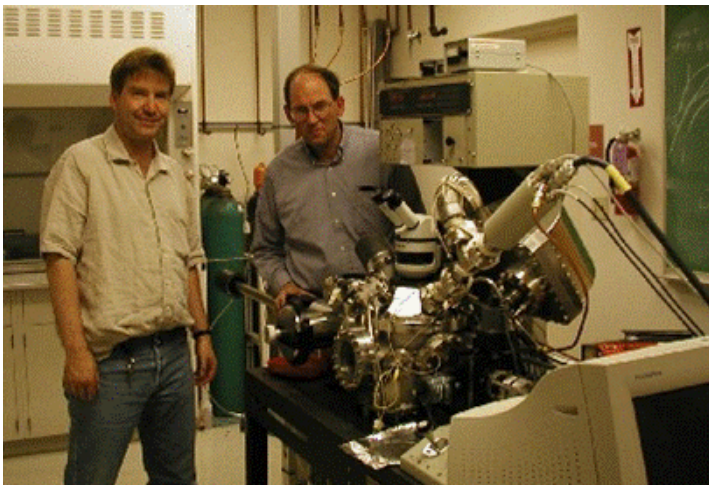
These techniques, applied to the steel surface, have probed the surface morphology, elemental composition, and oxidation states as a function of position. Chemical alterations and resulting chemical species were studied at the steel surface. Additionally, the experimental facilities at the Advanced Light Source (Lawrence Berkeley National Laboratory) and the Advanced Photon Source (Argonne National Laboratory) were used to characterize the systems. This allowed spectroscopic characterization of the stainless steel before and after interaction with LBE to determine its composition.

RESEARCH ACCOMPLISHMENTS

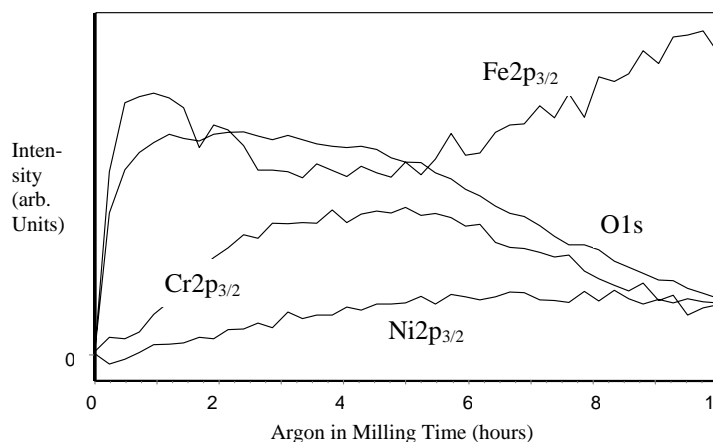
Several samples were compared having the same or similar compositions (standard nuclear grade 316/316L) but different surface preparation, including cold-rolled, annealed and a special low-corrosion treatment, "D-9." The cold-rolled sample had an order of magnitude less corrosion (i.e., both lower oxidation and less weight change) than the annealed sample.

Sputter depth profiling of the exposed annealed sample and cold-rolled sample showed a marked difference in oxide layer composition between the annealed and cold-rolled sample. The annealed sample showed a complex oxide structure (iron oxide over chromium/iron oxide mixtures) of tens of microns thickness, while the cold-rolled sample was covered with a simple, primarily chromium oxide layer approximately one micron thick. Interestingly, the D-9 material behaved more like the annealed than the more resistant cold-rolled material. In addition to these studies of 316/316L steel, other compositions of steel such as HT9 were also studied. These studies show the importance of surface preparation in resistance to corrosion by LBE.

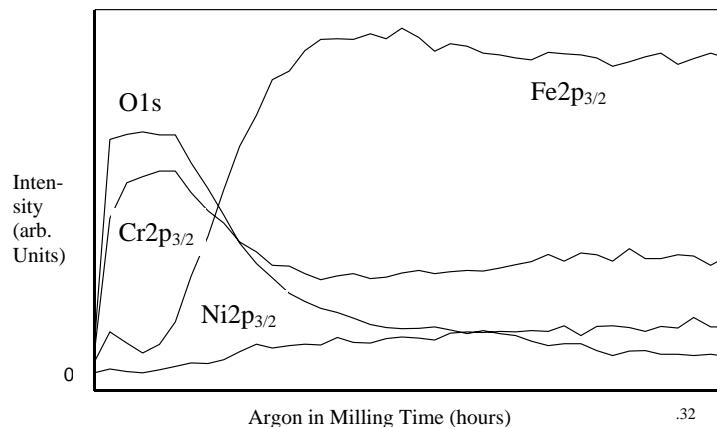
Another alloy studied in the Institute for Physics and Power Engineering in Obninsk, Russia series was EP823, an alloy similar to HT9 but with added silicon to improve corrosion resistance. A study of silicon in iron was undertaken to investigate the role of silicon in steels for LBE service. A series of silicon-containing iron samples were examined that were exposed to LBE at Idaho National Laboratory. Several unique features were observed: at low silicon concentrations in the starting alloy, the silicon was found as a silicate (SiO_3^{2-}). As the concentration of silicon increased in the starting alloy, a layer containing silica (SiO_2) was found also. Under the oxide layer, the silicon concentration in the metal was found to be decreased by approximately 30%. These were model systems, and the oxide layers that formed were not very protective. In all cases, pockets of LBE were found underneath the oxide layer, potentially corroding the steel.



Professors Allen Johnson and John Farley next to an X-ray Photoelectron Spectrometer.



Sputter Depth Profile of annealed 316/316L stainless steel sample. Note iron oxide at the surface.



Sputter Depth Profile of cold-rolled 316/316L stainless steel sample. Note chromium oxide at the surface.

TASK 3 PROFILE

Start Date: June 2001

Completion Date: May 2004

(This work continued under Task 18)

Thesis Generated:

Daniel Koury, M.S., Physics Department, "Investigation of the Corrosion of Steel by Lead-Bismuth Eutectic Using Scanning Electron Microscopy And X-ray Photoelectron Spectroscopy," December 2002.

Journal Article:

A.L. Johnson, D. Parsons, J. Manzerova, D.L. Perry, D. Koury, B. Hosterman, and J.W. Farley, "Spectroscopic and Microscopic Investigation of the Corrosion of 316/316L Stainless Steel by Lead-Bismuth Eutectic at Elevated Temperatures: Importance of Surface Preparation," *J. Nuclear Materials*, 328 (2004) 88-96.

Conference Proceedings:

"Mechanisms of Lead-Bismuth Eutectic Corrosion Chemistry involving Stainless Steels in Nuclear Reactor Cooling Systems," Annual Meeting of the American Chemical Society, Anaheim, CA, March 29-31, 2004.

A. Johnson, D. Parsons, J. Manzerova, D. Koury, B. Hosterman, D.L. Perry, and J.W. Farley, "Surface Studies of the High Temperature Corrosion of Stainless Steel by Oxygen Controlled Lead-Bismuth Eutectic," American Vacuum Society, Baltimore, MD, Nov. 2-7, 2003.

"Chemical Species Involved in High Temperature Corrosion Resulting from the Interaction Between Stainless Steel and Lead-Bismuth Eutectic," 38th Western Regional Meeting of the American Chemical Society, Long Beach, CA, October 15-18, 2003.

A.L. Johnson, D. Parsons, J. Manzerova, D.L. Perry, D. Koury, B. Hosterman, and J.W. Farley, "Surface Studies of the Corrosion of Stainless Steel by Lead-Bismuth Eutectic: Surface Preparation Effects on 316 Stainless Steels," American Nuclear Society Summer Meeting, San Diego, CA, June 1-5, 2003.

D. Koury, A.L. Johnson, D.L. Perry, and J.W. Farley, "Investigation of Corrosion of Steel by Lead-Bismuth Eutectic," American Nuclear Society Topical Meeting on Accelerator-Driven Transmutation Technologies and Applications, Reno, NV, November, 2001.

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Collaborators

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Task 4

Environment-Induced Degradation and Crack-Growth Studies of Candidate Target Materials

A.K. Roy

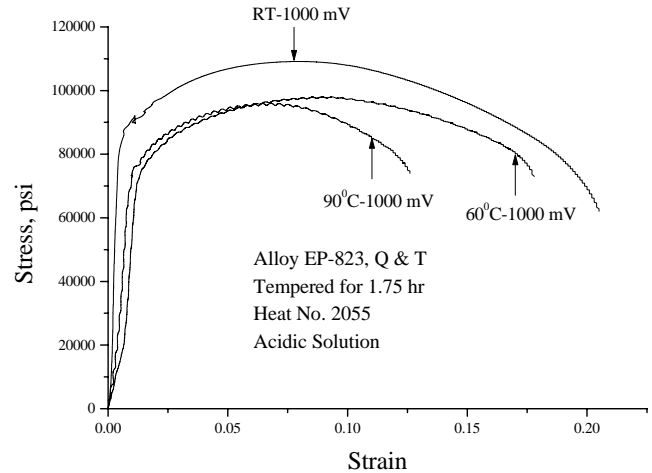
BACKGROUND

The primary objective of this task was to evaluate the effects of environmental and mechanical parameters on environment-induced degradations of candidate target structural materials for applications in spallation-neutron-target systems. The materials selected for evaluation and characterization were martensitic stainless steels including Alloys HT-9, EP-823, and 422.

Accelerator-driven transmutation systems involve bombarding a target material such as molten lead-bismuth eutectic (LBE) by a proton beam, thereby producing neutrons. The molten LBE target will be contained in a subsystem structural container made of a suitable material such as Alloys HT-9, EP-823, and 422. During the transmutation process, the target structural material may become susceptible to different types of environment-induced degradations such as stress corrosion cracking (SCC), hydrogen embrittlement (HE), and localized (pitting and crevice) corrosion. While the performance of these candidate materials in the presence of a molten LBE is yet to be evaluated, substantial work has been performed in this task to evaluate the corrosion behavior of these alloys in aqueous environments of interest. These baseline data can eventually be utilized to compare them to those yet to be generated in the molten LBE environment.

RESEARCH OBJECTIVES AND METHODS

This investigation was focused on the evaluation of the susceptibility of all three martensitic alloys to SCC, HE, and localized corrosion in neutral and acidic aqueous environments at ambient and elevated temperatures. State-of-the-art experimental techniques including constant-load (CL), slow-strain-rate (SSR), and cyclic potentiodynamic polarization were employed to evaluate these corrosion phenomena. The susceptibility of these alloys to HE was evaluated by applying cathodic potential while the test specimens were loaded in tension by the SSR method.



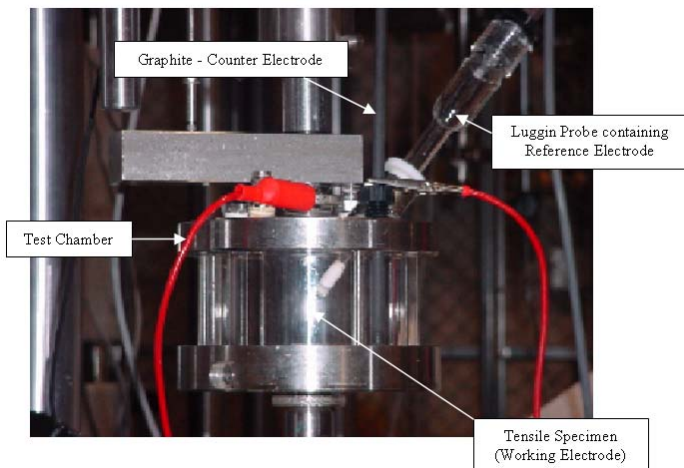
Stress vs Strain under Controlled Potential.

Optical microscopy and scanning electron microscopy (SEM) were used to analyze the metallurgical microstructures and fractography, respectively of the tested specimens.

RESEARCH ACCOMPLISHMENTS

The significant results derived from this task are summarized as follows:

- No failures were observed in smooth specimens of Alloys EP-823 and 422 in the neutral solution when tested at CL. However, Alloy HT-9 showed failure in the 90°C neutral solution at an applied stress (σ_a) of 112 ksi.
- All three alloys exhibited failure in the 90°C acidic solution at 95% of their yield strength (YS) values. Alloys HT-9 and 422 also showed failures at σ_a of 90 and 85% of their YS values, but no failure was observed with Alloy EP-823 at stresses below 0.95 YS.
- The magnitudes of the threshold stress (σ_{th}) for cracking for Alloys EP-823, HT-9 and 422 were 100, 95, and 98 ksi (689, 655, and 676 MPa), respectively based on CL testing in the 90 °C acidic solution. The presence of a notch in the test specimen reduced the σ_{th} values in all three alloys.
- The results of SSR testing in the acidic solution involving smooth specimens showed gradual reduction in ductility parameters (percent elongation - %El and percent reduction in area-%RA), time-to-failure (TTF), and true failure stress (σ_f) with increasing temperature, indicating a synergistic effect of pH and temperature in enhancing the cracking susceptibility. The presence of a notch in the specimen produced enhanced SCC susceptibility due to the stress concentration. However, the σ_f value was increased due to plastic constraint resulting from triaxial stress field at the notch.
- The magnitude of %El, %RA, TTF, and σ_f was reduced under



Controlled Potential Test Setup

an applied potential of -1,000 mV (Ag/AgCl) compared to those obtained without an applied potential.

- The failure mode at the primary fracture face of the specimen tested in the neutral solution, determined by SEM, was characterized by dimpled microstructure, indicating ductile failures. However, intergranular and/or transgranular brittle failures were observed in the acidic environment.
- Secondary cracks with branching were observed by optical microscopy on all three tested materials along the gage section of the specimens tested in the acidic solution.

TASK 4 PROFILE

Start Date: June 2001

Completion Date: December 2004

Theses Generated:

Ramprashad Prabhakaran, M.S., "Environment-Induced Degradations in a Target Structural Material for Transmutation Applications," August 2004.

Sudheer Sama, M.S., "Embrittlement and Localized Corrosion in Alloy HT-9," August 2004.

Phani P. Gudipati, M.S., "Stress Corrosion Cracking Resistance of Martensitic Stainless Steels for Transmutation Applications," December 2004.

Mohammad K. Hossain, Ph.D., "Stress Corrosion Cracking and Hydrogen Embrittlement of Martensitic Alloy EP-823," December 2004.

Venkataramakrishnan Selvaraj, M.S., "Environment Assisted Cracking of Target Structural Materials under Different Loading Conditions," December 2004.

Journal Articles:

A.K. Roy and M.K. Hossain, "Cracking of Martensitic Alloy EP-823 under Controlled Potential," *Journal of Materials Engineering and Performance*, American Society of Metals International, Vol. 15, No. 3, June 2006, pp. 336-344.

A.K. Roy, M.K. Hossain, R. Prabhakaran, and S. Sama, "Environment-Assisted Cracking of Structural Materials under Different Loading Conditions," *Corrosion*, National Association of Corrosion Engineers (NACE) International, April 2005, Vol. 61, No. 4, pp. 364-370.

A.K. Roy, R. Prabhakaran, M.K. Hossain, and S. Sama, "Stress Corrosion Cracking of Nuclear Transmutation Structural Materials," *Materials Performance*, NACE International, September 2004, Vol. 43, No. 9, pp. 52-56.

Conference Proceedings:

A.K. Roy, and M.K. Hossain, "Environment Induced Embrittlement of Martensitic Stainless Steel for Transmutation Applications," Corrosion 2005, NACE International, Houston, TX, April 2005.

A.K. Roy, Phani P. Gudipati, Venkataramakrishnan Selvaraj, "Environment Degradation of Martensitic Stainless Steels for Transmutation Applications," Materials Science and Technology (MS&T) 2004, New Orleans, LA, September 26-29, 2004.

A.K. Roy, R. Prabhakaran, "Stress Corrosion Cracking of Type 422 Stainless Steel for Transmutation Applications," International Youth Nuclear Congress, Toronto, Canada, May 9-13, 2004.

R. Prabhakaran and A.K. Roy, "The Effect of Environmental and Mechanical Variables on Stress Corrosion Cracking of Martensitic Stainless Steels for Transmutation Applications," International Conference on Nuclear Engineering, ICONE-12, Student Paper Competition, Arlington, VA, April 25-29, 2004.

A.K. Roy, M.K. Hossain, R. Prabhakaran, S. Sama, V. Selvaraj, P.P. Gudipati, "Stress Corrosion Cracking of Target Materials for Transmutation Applications," Corrosion 2004, NACE International, New Orleans, LA, March 28-April 1, 2004.

A.K. Roy, R. Prabhakaran, "Characterization of Environment-Induced Degradation in Type 422 Stainless Steel," Minerals, Metals and Materials Society, Charlotte, NC, March 14-18, 2004.

A.K. Roy, R. Prabhakaran, M.K. Hossain, S. Sama, V. Selvaraj, P.P. Gudipati, "Effect of Environmental Variables on Cracking of Martensitic Stainless Steels under Different Loading Conditions," American Nuclear Society Meeting, Global 2003, New Orleans, LA, November 16-20, 2003.

A.K. Roy, R. Prabhakaran, M.K. Hossain, S. Sama, V. Selvaraj, P.P. Gudipati, "Environmental Effects on Materials For Nuclear Applications," MS&T 2003, Chicago, IL, November 9-12, 2003.

A.K. Roy, S. Sama, R. Prabhakaran, M.K. Hossain, "Cracking of Martensitic Stainless Steels under Applied Electrochemical Potential," 204th Meeting of the Electrochemical Society, Abstract No. 1255, Orlando, FL, October 12-17, 2003.

A.K. Roy, R. Prabhakaran, M.K. Hossain, S. Sama, B. J. O'Toole, "Environment-Induced Degradation of Spallation Target Materials," American Nuclear Society Meeting, AccApp'03, San Diego, CA, June 1-5, 2003.

A.K. Roy, M.K. Hossain, B. J. O'Toole, "Stress Corrosion Cracking of Martensitic Stainless Steel For Transmutation Applications," The 10th International High-Level Radioactive Waste Management Conference, Las Vegas, NV, March 30-April 3, 2003.

R. Prabhakaran, "Stress Corrosion Cracking of Type 422 Stainless Steel For Applications in Spallation-Neutron-Target Systems," Spallation Neutron Source, Joint Institute for Neutron Sciences, Neutrons in Solid-State Chemistry and the Earth Sciences Today and Tomorrow, Oak Ridge, TN, March 12, 2003.

A.K. Roy, M.K. Hossain, and B.J. O'Toole, "Stress Corrosion Cracking of Martensitic Stainless Steel for Transmutation Applications," International High-Level Radioactive Waste Management Conference, Las Vegas, NV, Sept. 2002.

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Stuart A. Maloy, Ph.D., AFCI Materials Team Leader, Los Alamos National Laboratory

Task 5

Modeling Corrosion in Oxygen Controlled LBE Systems with Coupling of Chemical Kinetics and Hydrodynamics

S. Moujaes and Y. Chen

BACKGROUND

The corrosion of structural materials is a major concern for the use of lead-bismuth eutectic (LBE) systems for nuclear applications such as in transmuter targets or fast reactors. Corrosion in liquid metal systems can occur through various processes, including, for example, dissolution, formation of inter-metallic compounds at the interface, and penetration of liquid metal along grain boundaries. Predicting the rate of these processes depends on numerous system operational factors: temperature, system geometry, thermal gradients, solid and liquid compositions, and velocity of the liquid metal, to name a few. Corrosion, along with mechanical and/or hydraulic factors, often contributes to component failure.

The goal of this project was to develop a corrosion model that combines the chemical kinetics and hydrodynamics in the system to predict corrosion rates. In this effort, these models were developed for the Delta test loop at Los Alamos National Laboratory (LANL) and a theoretical LBE accelerator target system. The resulting models are predictive tools that can be validated with corrosion test data and used to systematically design tests, interpret the results, and provide guidance for optimization in LBE system designs.

RESEARCH OBJECTIVES AND METHODS

There were two subtasks to this research. The first subtask developed the necessary tools to predict the levels of oxygen and corrosion products close to the boundary layer using Computational Fluid Dynamics (CFD) modeling. The second subtask predicted the corrosion process kinetics between the LBE and structural materials by incorporating pertinent information from the first subtask.



Concentration profile for T-joint at $Re = 200,000$.

STAR-CD software was used to model the corrosion and precipitation rates in the LBE loop. This allowed researchers to compare the theoretical analysis with available experimental results. The surface corrosion analysis is being developed as a user supplied subroutine to STAR-CD. The information obtained from this analysis theoretically predicts likely locations for corrosion and precipitation along the axial lengths of the test loop.

The first subtask involved performing a series of parametric runs. Models prepared from the earlier work were used as guides for the parametric studies. Variables investigated included the average eutectic flow velocity, average mean bulk eutectic flow, inlet temperatures, and average inlet oxygen concentrations in the three geometries: a straight flow section, an elbow bend, and a tee section. The thermal-hydraulics study involved using a 3-D CFD code simulation to obtain averaged values of stream-wise velocity, temperatures, and oxygen and corrosion product concentrations at various axial locations close to the walls of several partial loop sections within the LBE loop. The oxygen and corrosion products inside the test loop were simulated to participate in chemical reactions with the eutectic fluid as it diffused towards the walls. Details of the geometry of these loops were obtained from scientists at LANL. These values acted as a set of starting boundary conditions for the second task.

The second subtask focused on the kinetics of the dissolution/deposition process as a function of temperature, flow velocities, dissolved metal concentrations, oxygen potentials of the system, the kinetics of film formations in the presence of oxygen, and the kinetics of metal transport through the oxidized surface film.

RESEARCH ACCOMPLISHMENTS

Geometries and flow conditions similar to experimental results in the literature were set up and used to benchmark the models assembled using the STAR-CD software. Test case studies indicated that the outcome from STAR-CD was correct and that numerical modeling is applicable to the research in this problem.

Using these models, concentration flux profiles were obtained for both laminar and turbulent profiles in a straight pipe. This information was then used for the chemical kinetics analysis for corrosion on the inside walls of the LBE loop.

Several CFD runs were made to simulate flow in pipe fittings. These include sudden contraction, sudden expansion and a T-joint. Testing of a new turbulent model was also performed using the $k-\epsilon$ Chen model which works better with high Re number flows and predicts some of the peculiar flow features relevant to sudden expansions where eventually vortex generation is expected at the backward step.

The k- ϵ Chen model which works better with high Re number flows and predicts some of the peculiar flow features relevant to sudden expansions where eventually vortex generation is expected at the backward step.

A user subroutine was developed that simulated the corrosion/precipitation processes and predicted their maximum/minimum location in a typical LBE loop. A chemical reaction subroutine dealt with surface chemistry. The Arrhenius Equation was used to calculate the reactant reaction rate but some constants and coefficients used in calculating chemical reaction rate were updated using some empirical data found in chemical reaction research papers and textbooks.

The location in the loop with the highest concentration occurred in the baffle cell which is adjacent to the iron surface while the area with lowest concentration occurs around the central line of the pipe. It can be inferred that after Fe_3O_4 is formed, it diffuses to the fluid area close to the central line of pipe. But, overall, the concentration value was very low.

Similar to the concentration gradient, the area with the highest temperature occurs in the baffle cell which is adjacent to iron surface while the area with lowest temperature occurs around the central line of the pipe. It is obvious, based on the model, that heat was transferred from inner wall to central area of the pipe due to the setting of constant heat flux boundary conditions in the baffle cells.

The chemical reaction model subroutine worked well and this simple CFD model roughly simulated the chemical reaction, diffusion, and heat transfer condition inside the LBE piping. Since the subroutine works well, it can be used in much more complex modeling.

TASK 5 PROFILE

Start Date: August 2001

Completion Date: August 2004

Theses Generated:

Chao Wu, M.S., Mechanical Engineering, "Study of Geometry Effects on Local Corrosion Rates for LBE Loop," July 2003.

Kanthi Dasika, M.S., Mechanical Engineering, "Chemical Kinetics and Thermal Hydraulics of Lead Bismuth Flow Loops," December 2003.

Narain Armbya, M.S., Mechanical Engineering, "CFD Analysis of 3-D Thermal Hydraulics Flow Effects on Wall Concentration Gradient Profiles for LBE Loop Fittings," December 2004.

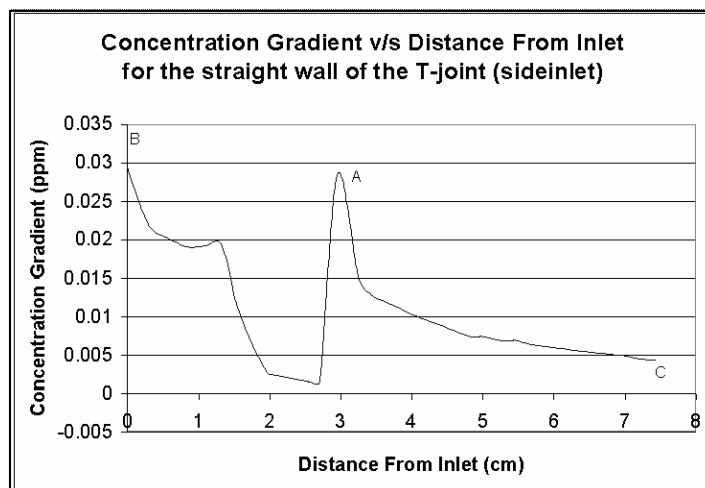
Conference Proceedings:

C. Wu, K. Dasika, Y. Chen, S. Moujaes, J. Zhang and N. Li, "Study of Geometry Effects on Local Corrosion Rates for LBE Loop," American Nuclear Society Summer Meeting, San Diego, CA, June 1-5, 2003.

S. Moujaes, Y. Chen, K. Dasika, C. Wu, N. Li, and J. Zhang, "Simulation Considerations in Lead-Bismuth Transmutation Loops: Corrosion Concentration, velocity and Temperature Profiles of LBE Loops," 10th International Conference on Nuclear Reactor Thermal Hydraulics, South Korea, October 5-9, 2003.

K. Dasika, C. Wu, S. Moujaes, and Y. Chen, "Modeling of Oxygen Controlled Surface Reaction Kinetics and Hydrodynamics," International Youth Nuclear Congress, Seoul, South Korea, April 2002.

K. Dasika, C. Wu, S. Moujaes, and Y. Chen, "Numerical Modeling of Lead Oxidation in Controlled Lead Bismuth Eutectic Systems: Chemical Kinetics and Hydrodynamic Effects," International Congress on Advances in Nuclear Power Plants, American Nuclear Society, Hollywood, FL, June 2002.



Concentration gradient versus distance from the inlet for the straight wall.

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Task 6

Neutron Multiplicity Measurements of Target/Blanket Materials

D. Beller

BACKGROUND

The U.S. Advanced Fuel Cycle Initiative (AFCI) is a program to develop economic and environmental methods to reduce the impact of waste from commercial nuclear fuel cycles. One concept for near-complete destruction of waste isotopes from used nuclear fuel is accelerator-driven transmutation. High-power accelerators would be used to produce high-energy charged particles, which then collide with heavy metal targets to create a cascade of neutrons. These neutrons then cause nuclear reactions in subcritical systems.

To design these systems, complex reactor physics computer codes and highly detailed data libraries are used to compute the reactivity of systems, reaction rates, destruction rates, and nuclear-induced damage rates to materials. In this project, a Russian-built detector system was used to make measurements of neutrons generated in a central target by a variety of accelerators.

RESEARCH OBJECTIVES AND METHODS

A prototype modular Neutron Multiplicity Detector System (NMDS) with 64 ^3He gas counting tubes was developed to measure the neutron multiplicity of scaled lead accelerator targets. Its modularity allows it to be configured for a variety of experiments to measure neutron multiplicity from different sources: protons, electrons, high-energy neutrons, or even cosmic particles such as muons.

This system may be used to measure neutron production in a variety of configurations, on a variety of targets, with a variety of source particles, and over a range of energies (10 to 800 MeV) to produce a large database that may be used to validate neutron multiplicity predictions. This should enable the quantification of systematic errors in the latest version of MCNPX and its accompanying data libraries. Time-dependent measurements of neutron production in the NMDS should provide a systematic set of precise data that will enable direct comparison with code calculations.

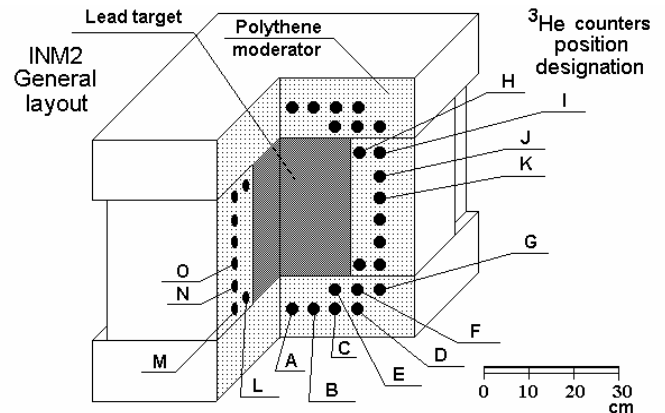
Comparison of results from the NMDS may decrease uncertainties and allow the derivation of relative measurements in the few percent range at the 95 percent confidence level. In addition, discrepancies that are discovered with this system can contribute to the improvement of the codes and data libraries. Improved models of beam line experiments, accelerator targets, and detector designs will result from these code improvements.

RESEARCH ACCOMPLISHMENTS

Initial work in this project included the development of geometric models of the target-detector assemblies for use with the MCNPX

radiation transport code to optimize the design of the NMDS. Radiation transport calculations of neutron detection efficiency were completed and interpreted prior to developing designs of the neutron detection systems needed to perform multiplicity measurements. Following completion of the modeling, a fabrication effort was initiated. A series of MCNPX models were developed at UNLV for a cylindrical target. Another generic model was created to examine response times, collection efficiencies, and escape probabilities. In addition to modeling at UNLV, colleagues at the V.G. Khlopin Radium Institute (KRI) completed preliminary radiation transport modeling using the CONTROL code developed by KRI researchers.

The 64-element ^3He -detector-based NMDS was fabricated by KRI and shipped to UNLV. It was assembled at UNLV in the “CUBE” geometry (30 cm x 30 cm x 30 cm of lead inside and 8 to 12 detectors on each of the 6 sides of the cube. Detection efficiencies in ^3He as well as fractional capture in Pb and polyethylene were calculated. A ^{252}Cf source was used to calibrate the NMDS. Measured efficiencies were comparable to calculations and measurements were completed at KRI.



NMDS in the INM2 (“CUBE”) geometry.

ISU-IAC Experiments

To evaluate its usefulness for measurement of spallation neutrons, the NMDS was transported to the Idaho Accelerator Center at Idaho State University where it was used in conjunction with a linear accelerator to determine its performance.

An americium-beryllium (AmBe) source with a neutron production rate of $2500 \text{ n/s} \pm 10\%$ was placed against the face of the Pb in all 5 configurations to measure the overall efficiency of the system. This weak source exceeded the counting capacity of the NMDS, such that its efficiency was reduced to 3.5% compared to earlier values at UNLV of 20-30% with source strength of $\sim 600 \text{ n/s}$. This was the first indication of a severe count-rate limitation of the NMDS.

To accommodate this limitation, the accelerator was “de-tuned” to

reduce the neutron production rate within the boundaries of the detectors. All results from the ISU-IAC accelerator-driven experiments were influenced by a count-rate limitation that is inherent in the NMDS hardware and software.

Accelerator-based testing commenced with the electron beam and several parameters and results were examined and compared to radiation transport predictions in these studies. These parameters include the neutron absorption time or lifetime, efficiencies of the systems and individual detectors, and multiplicity distributions.

NTS-RSL Experiments

In an effort to determine system contributions to dead-time, a dead-time measurement experiment was conducted at the Remote Sensing Laboratory (RSL) of the Nevada Test Site (NTS). Results indicated that the performance of individual detectors depends upon the number of detectors operating as well as the source strength. The individual detectors were determined to have a dead-time coefficient of a few ms, which would indicate a capability of the total system to count several thousand neutrons per second. However, the system has never counted more than 200 n/s, even with strong neutron sources. Each system component contributes to dead-time. In the final analysis, however, these experiments at ISU-IAC and NTS-RSL demonstrated that the performance of the system is critically limited by the Russian hardware and, as a consequence of these experiments, a modern data acquisition system was acquired that will support the maximum performance of each detector and all the detectors combined.



Graduate student Shruti Patil checks signals on one of the 8-detector control boxes of the NMDS.

TASK 6 PROFILE

Start Date: August 2001

Completion Date: December 2005

Conference Proceedings:

- T. Beller, D. Curtis, D. Beller, A. Rimsky-Korsakov, and T. Ward, "The UNLV Neutron Multiplicity Detector System," *Proceedings of the Eighth Information Exchange Meeting on Actinide and Fission Product Partitioning & Transmutation*, Organization for Economic Cooperation and Development, Nuclear Energy Agency, Paris, France, pp 687-698, 2005.
- T. Ward, A. Rimski-Korsakov, N. Kudryashev and D. Beller, "Integral Neutron Multiplicity Measurements from Cosmic Ray Interactions in Lead," *Proceedings, XVII Particles and Nuclei International Conference*, Santa Fe, NM, October 24-28, 2005.
- D. Curtis, D. Beller, C. Hull, A. Rimsky-Korsakov, and T. Ward, "Modeling Neutron Multiplicities in a 60-element ³He Detector System," *Proceedings of the Sixth International Meeting on Nuclear Applications of Accelerator Technology*, American Nuclear Society, pp. 190-194, 2004.
- T. Beller, D. Curtis, D. Beller, A. Rimsky-Korsakov and T. Ward, "The UNLV Neutron Multiplicity Detector System," Eighth Information Exchange Meeting on Actinide and Fission Product Partitioning and Transmutation, Organization for Economic Cooperation and Development, Nuclear Energy Agency, Las Vegas, NV, November 9-11, 2004.
- D. Curtis, D. Beller, C. Hull, A. Rimsky-Korsakov, and T. Ward, "Modeling Neutron Multiplicities in a 60-Element ³He Detector System," Sixth International Meeting on Nuclear Applications of Accelerator Technology (AccApp'03), American Nuclear Society, San Diego, CA, June 1-5, 2003. (Awarded Best Overall Poster for AccApp'03)
- T. Ward, A. Rimski-Korsakov, M. Todosow, G. Greene, M. Divadeenam, C.L. Snead, Jr., A. Hanson, E. Pitcher, D. Beller, C. Hull, W. Culbreth, A. Hechanova, and G. Cerefice, "Note on the Benchmark and Validation of the LAHET Code System," American Nuclear Society Winter Meeting, Reno, NV, November 11-15, 2001.

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Thomas Ward, AFCE Science Adviser to the U.S. Department of Energy

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Task 7

Development of Dose Coefficients for Radionuclides Produced in Spallation Targets

P.W. Patton and M.J. Rudin

BACKGROUND

Ensuring the safety of workers at accelerator-driven nuclear facilities is paramount before these systems can be deployed for nuclear transmutation or any other mission. Spallation neutron sources produce as many as 660 rare radionuclides in either the target or blanket during the spallation process. No data exists for many of these radionuclides in the current radiation protection guidelines and standards. This research program seeks to address this problem through generating internal and external dose coefficients (DCs) for these “new” isotopes.

Dose coefficients permit simple determination of radiation dose associated with various exposure scenarios, and ultimately permit radiation safety personnel to assess the health risks to workers in a nuclear facility. Specifically, radiation safety personnel use dose coefficients to determine the radiation dose incurred to a tissue or organ system from a given exposure. These parameters are often expressed in terms of Annual Limits on Intake (ALIs) and Derived Air Concentrations (DACs).

RESEARCH OBJECTIVES AND METHODS

Results from this study will be used to produce ALIs and DACs for these rare radionuclides created by spallation target systems that are not included in Federal Guidance Report (FGR) No. 11. Additionally, DCs developed will augment the radiological data in Publications 68 and 72 of the International Commission on Radiological Protection (ICRP), contributing to the safe operation of accelerator-driven nuclear systems.

A Dose Coefficient Working Group was established in 2001 (the first year of the project) to direct and oversee consortium activities. Representatives from the Dose Coefficient Working Group developed and verified a methodology to determine internal and external dose for select radionuclides. The first step involved obtaining radiological data from the ENSDF nuclear physics database developed at Brookhaven National Laboratory. Data collected included decay modes, decay energy levels, and radiation energies and intensities.

The DC working group prioritized a list of radionuclides projected to be released via air emissions or in the inventory of a mercury target following a lengthy irradiation period. Only radionuclides with a half-life greater than one minute were considered. These 81 radionuclides were then categorized into three distinct categories, based on half-life, available information, and other technical factors.

All Category 2 radionuclides were investigated to determine which database was most current. However, this task was not straight forward and thus both databases were used to calculate all radionuclides that had complete data. Dose coefficients were then

generated for the Category 2 radionuclides using both ENSDF and NUBASE. The results were compared and showed good agreement. Metabolic models and data from ICRP publications (30 and 66) were applied in order to use the best technology available and to maintain consistency with current standards. In accordance with FGR No. 11, dose coefficients were evaluated for an adult male with the target tissues of gonads, breast, lung, red marrow, bone surface (endosteum), thyroid, remainder, and total committed effective dose equivalent (this considers total dose incurred to specific organs or tissues with respect to radiation type over a period of 50 years). Following determination of these variables, values of ALIs and DACs were then calculated for each radionuclide.

The consortium investigated the competing databases to determine the most appropriate one to use for dose coefficient calculations. However, conflicting data made it unclear which was better suited for this task, therefore the previously calculated coefficients were reported. Additionally, time was devoted to trying to acquire missing nuclear data for radionuclides in category three. The consortium believes large scale accelerator driven experiments are needed to accomplish this goal.

RESEARCH ACCOMPLISHMENTS

The research consortium comprised of representatives from several universities and national laboratories has successfully generated internal and external dose conversion coefficients for twenty radionuclides produced in spallation neutron sources. These dose coefficients fill data gaps exist in Federal Guide Report No. 11 and in Publications 68 and 72 of the International Commission on Radiological Protection, and two articles containing the data have been accepted for publication in the *Journal of Health Physics*. Currently, more nuclear data is needed for the rare radionuclides produced from a mercury target. While attempting to develop a workable plan to acquire this missing data, Q-value discrepancies were investigated and reported. A detailed plan was developed to start a research effort at Idaho State University to produce rare radionuclides. This process will allow for the investigation of the missing nuclear data needed to complete dose coefficients.

TASK PROFILE

Start Date: June 2002

Completion Date: April 2006

Theses Generated:

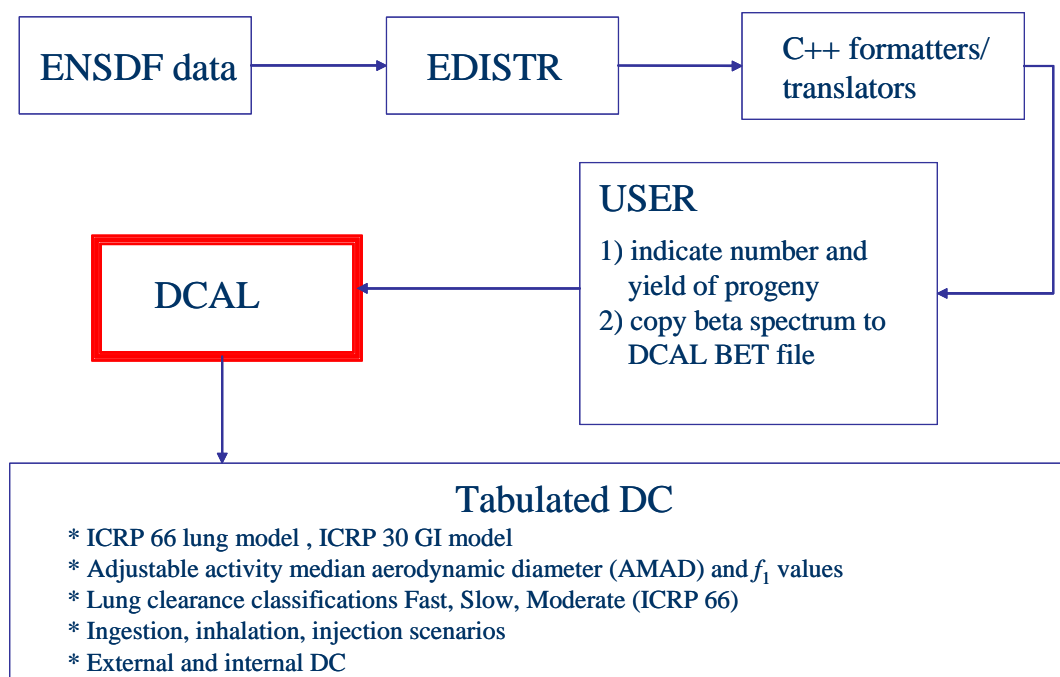
John P. Shanahan, M.S., Health Physics Department, “Dose Coefficients for Radionuclides Produced in a Spallation Neutron Source,” August 2003.

Yayun Song, M.S., Health Physics Department, “Investigation and

Calculations of Dose Coefficients for Radionuclides Produced in a Spallation Neutron Source Using the ENSDF and NUBASE Nuclear Databases,” May 2004.

Publications:

- J. Shanahan, K. Eckerman, A. Arndt, C. Gold, P. Patton, M. Rudin, R. Brey, T. Gesell, V. Rusetski, and S. Pagava, “Calculation of Dose Coefficients for Radionuclides Produced in a Spallation Neutron Source Utilizing NUBASE and the Evaluated Nuclear Structure Data File Databases.” Health Phys 90:56-65, 2006.
- A. Arndt, J. Shanahan, C. Gold, R. Brey, T. Gesell, P. Patton, M. Rudin, K. Eckerman, V. Rusetski, and S. Pagava “Procedures and Quality Assurance for Calculating Dose Coefficients Using DCAL Software.” Health Phys 90:74-80, 2006.
- J. Shanahan, K. Eckerman, A. Arndt, C. Gold, P. Patton, M. Rudin, R. Brey, T. Gesell, V. Rusetski, and S. Pagava, “Calculation of Dose Coefficients for Radionuclides Produced in a Spallation Neutron Source Utilizing NUBASE and the Evaluated Nuclear Structure Data File Databases,” accepted August 16, 2005 for publication in the Health Physics Journal, Health Phys. 89(6), 2005.
- A. Arndt, J. Shanahan, C. Gold, R. Brey, T. Gesell, P. Patton, M. Rudin, K. Eckerman, V. Rusetski, and S. Pagava, “The Procedure for Determining and Quality Assurance Program for the Calculation of Dose Coefficients Using DCAL Software,” accepted April 2005 for publication in the Health Physics Journal.
- J. Shanahan, K. Eckerman, A. Arndt, C. Gold, P. Patton, M. Rudin, R. Brey, T. Gesell, V. Rusetski, and S. Pagava, “An Interdatabase Comparison of Nuclear Decay and Structure Data Utilized in the Calculation of Dose Coefficients for Radionuclides Produced in a Spallation Neutron Source,” accepted March 2005 for publication in the Health Physics Journal.
- J. Shanahan, A. Arndt, C. Campbell, R. Brey, M. Rudin, K. Eckerman, and P. Patton, “Calculation of Dose Coefficients for Radionuclides Produced in Spallation Neutron Sources,” Health Phys. 84 (6S): S252 (2003).
- J. Shanahan, P. Patton, and M. Rudin, “Dose Coefficient Methodology Report,” Health Physics Department, University of Nevada, Las Vegas, NV, Aug. 2002.



Dose Coefficient Working Group Methodology Flow Sheet. The ENSDF code is used to obtain nuclear physics data. The EDISTR code prepares the data for input into the dose calculation code DCAL.

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Task 8

Development of a Systems Engineering Model of the Chemical Separations Process

Y. Chen, D.W. Pepper, and S. Hsieh

BACKGROUND

The chemical processing of used nuclear fuel is an integral component of any strategy for the transmutation of nuclear waste. Due to the large volume of material that must be handled in this first step of the transmutation process, the efficiency of the separations process is a key factor in the potential economic viability of transmutation strategies. The ability to optimize the chemical separation systems is vital to ensure the feasibility of the transmutation program.

Systems analysis, or total systems modeling, is one of the strongest tools available to researchers for understanding and optimizing complex systems such as chemical separations processes. Systems analyses permit researchers to present decision-makers concise evaluations of system options and their characteristic features. The primary goal of this project was to develop a systems model that can be used to parameterize and optimize chemical separations processes.

RESEARCH OBJECTIVES AND METHODS

This work includes reviewing and analyzing the Argonne Model for Universal Solvent Extraction (AMUSE) code structure, examining other possible implementations, defining software activities, developing a verification plan, and modifying and improving the software. This work also involves redefining the graphical user interface (GUI) to increase the utility of the AMUSE code suite as a stand-alone analytical package.

Developing a systems engineering model required discussions with Argonne National Laboratory personnel to identify pertinent

components of the chemical separations process. Each step required model development to establish its significance with regards to the overall process. Comprehensive model development involved defining the inputs and outputs from individual models and establishing how each connected to the other within in the chemical separations process.

RESEARCH ACCOMPLISHMENTS

This project developed software for a general-purpose systems engineering model named Transmutation Research Program System Engineering Model Program (TRPSEMPro) that will be used to improve productivity in the design process. The system model also includes various numerical optimization technologies and “Design of Experiments” study technologies.

Object-Oriented Analysis and Design was used for developing and implementing the TRPSEMPro system. A graphical notation, Unified Modeling Language, was employed to express object-oriented designs. Microsoft.Net architecture was used for system development and Visual Basic.NET was the major programming language behind the system. XML (Extensible Markup Language) was used widely to describe data and sets of elements and attributes that can be defined by researchers. XML Schema was used for describing the structure of the system engineering model. XML Database was used to store all the run-time data for the AMUSE module. Since significant experimental data will be generated and require systematic analysis, MS SQL Server 2000 database was selected for housing all run-time parameters and simulation results.

The model is combined with commercial software packages MATLAB OPTIMIZATION toolbox and SIMULINK module from Mathworks.

The system model, TRPSEMPro, considers input simulation modules from multiple disciplines with inconsistent input/output handles. The package with the aid of middleware can communicate with various simulation modules developed by other research groups and create an XML-based model description file. The critical components for the system engineering modeling include System Manager, Model Integration, Study Plan, and Solution Viewer.

The introduction of the middleware design provides flexibility to interface to other simulation modules without significant program modifi-

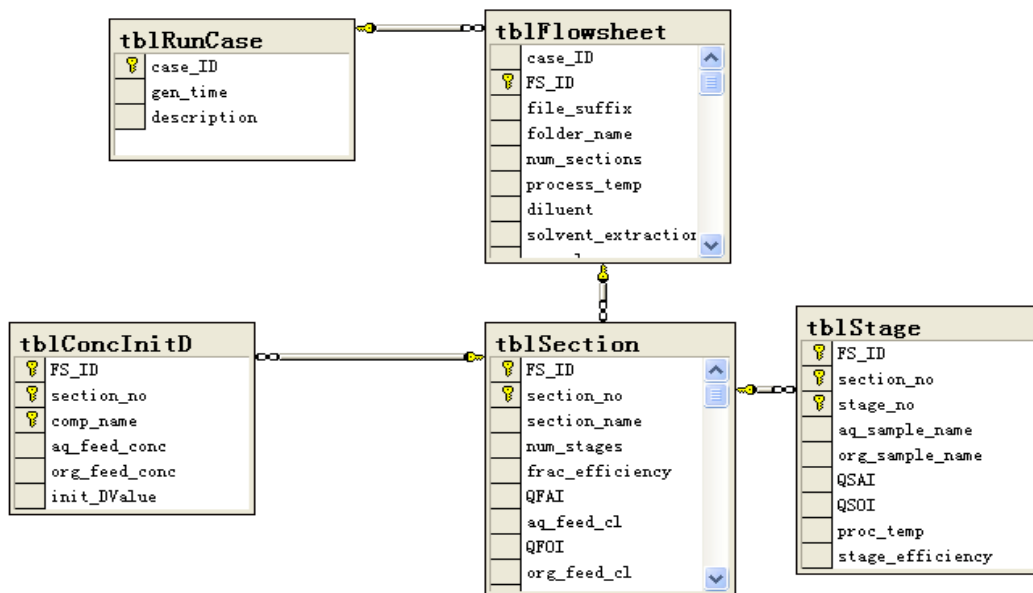


Diagram for the Structure of the Database Design.

cation. The demonstration code from AMUSE macros is kept intact during all system development stages. AMUSESimulator, is the middleware software package which was designed and implemented to serve as a bridge between the AMUSE code, and the systems engineering model, TRPSEMPPro. Such an approach can reduce the time-consuming modification on the system model side and keep flexibility on the simulation modules development side.

Further system enhancement allowed the user to select various process types. An interface for conducting multiple runs was created. The GUI included a list of variables, a range for those variables, all of which provide an envelope of end results.

TASK 8 PROFILE

Start Date: August 2001

Completion Date: August 2004

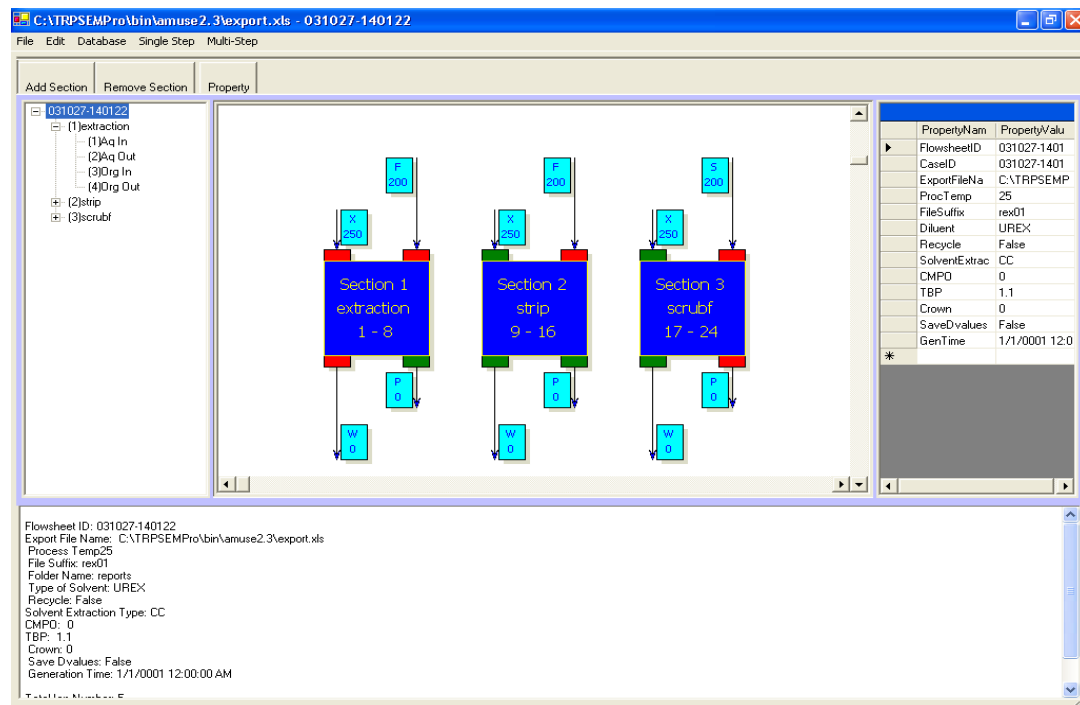
(This work continued under Task 24)

Thesis Generated:

Lijian Sun, M.S., Mechanical Engineering, "Development of a Systems Engineering Model for Chemical Separation Process," December 2003.

Conference Proceedings:

- L. Sun, H. Royyuru, S. Hsieh, Y. Chen, R. Clarksean, G. Vandegrift, J. Copple, and J. Laidler, "Development of Systems Engineering Model for Spent Fuel Extraction Process," International Mechanical Engineering Congress and Exposition Conference, Anaheim, CA, November 13-19, 2004.
- L. Sun, H. Royyuru, S. Hsieh, Y. Chen, R. Clarksean, G. Vandegrift, J. Copple, and J. Laidler, "Development of Systems Engineering Model for Spent Fuel Extraction Process," International Congress on Advanced Nuclear Power Plants, Pittsburgh, PA, June 13-17, 2004.
- H. Royyuru, L. Sun, Y. Chen, S. Hsieh, R. Clarksean, D. Pepper, G. Vandegrift, J. Copple, and J. Laidler, "Development of Systems Engineering Model For UREX Process," International Mechanical Engineering Congress and Exposition, Washington, DC, November 16-21, 2003.
- H. Royyuru, L. Sun, Y. Chen, S. Hsieh, R. Clarksean, D. Pepper, G. Vandegrift, J. Copple, and J. Laidler, "Development of Systems Engineering Model For UREX Process," Accelerator Applications in a Nuclear Renaissance, San Diego, CA, June 1-5, 2003.
- L. Sun, Y. Chen, R. Clarksean, and J. Laidler, "The Development of a Systems Engineering Model for the DOE Advanced Accelerator Applications Program," International Congress on Advanced Nuclear Power Plants, Hollywood, FL, June 9-13, 2002.



The main GUI for AMUSESimulator includes five parts as shown here. Menu and toolbar (Top Part) – where user can invoke all the available commands. Flowsheet contents displayed in tree view format (Left Part) – where user can select different section, stream. Flowsheet contents displayed in way of drawing blocks (Middle Part) – where user can select different sections, streams by clicking mouse at corresponding area.

Research Staff

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Task 9

Design and Evaluation of Processes for Fuel Fabrication

G. Mauer

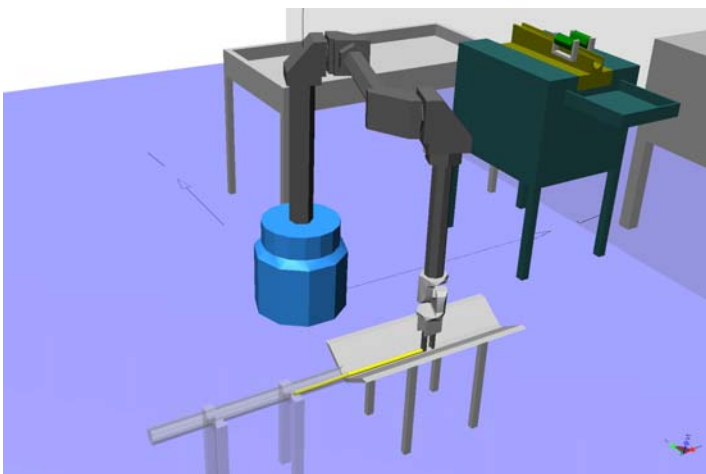
BACKGROUND

The safe and effective manufacturing of actinide-bearing fuels for any transmutation strategy requires that the entire manufacturing process be contained within a shielded hot cell environment. To ensure that the fabrication process is feasible, the entire process must be designed for remote operation. The equipment must be reliable enough to perform over several decades, and also easy to maintain or repair remotely. The facility must also be designed to facilitate its own decontamination and decommissioning. In addition to these design factors, the potential viability of any fuel fabrication process will also be impacted by a number of variables, such as the current state of technology, potential problem areas, deployment scaling, facility safety, and cost.

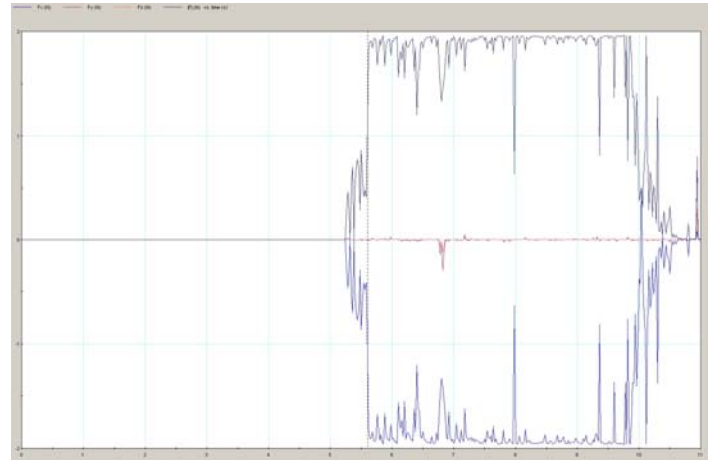
RESEARCH OBJECTIVES AND METHODS

The goal of this research project was to provide technical support to process designers working on the development of the fuel cycles for transmutation applications. Detailed process models were developed to better define the impact of fuel choice on the transmuter fuel cycle, including relative process losses, waste generation, and plant capital cost. These process models provide insight regarding required plant size and number of plants needed to mesh with the fuel recycling line. They also determine requirements for automation.

Manufacturing models for large-scale production in a hot cell environment were also developed. Combined, these two models allow the assessment of plant layout, and provide the framework for estimation of plant capital and operating cost estimates, and for feasibility in general. The need for development in the areas of robotic and sensor technology was assessed. The manufacturing technology developed for hot cell applications was also applicable to other, more general uses, where occupational hazards prevent human presence near processes.



The Waelischmiller robot inserts the fuel pins in the cladding tube.



Friction Force (N) between Second Pin and Cladding Tube vs. Time (sec) during Insertion.

The research work was divided into several tasks and subtasks:

Methods and Processes – A literature survey and detailed analysis of the research and development pertaining to candidate processes for transmuter fuel manufacture was performed. Industry standards were used to refine equipment, instrumentation, and control specifications, and assessed the reliability and safety of operations.

Simulations – This task modeled manufacturing processes to generate a realistic assessment of plant layout, size, feasibility, and technology development required for large-scale remote fabrication of fuel. Modeling of the candidate fuel manufacturing processes was conducted using the MSC.visualNastran and ProEngineer simulation software tools. The modeling of powder-processed fuels was completed, and the modeling of other fuel types (metallic, TRISO etc.) was initiated.

Process and Equipment for Autonomous Manufacturing – This task developed an understanding of the cost and capability of current generation remotely operated equipment suitable for use in radiation environments. Monitoring of the market for equipment and components with regard to suitability for automated manufacturing under hot cell conditions was conducted.

Sensors, Controls, and Operational Safety – This task determined the adequacy of current technology and the need for suitable sensor technology development for deployment in hard radiation environments. A means to identify the precise location and spatial orientation of all parts in the robot's work envelope were implemented. The ability to position and handle materials along with trouble shooting techniques were evaluated. Radiation hardened vision systems appear to be promising technologies.

Cost, Feasibility, and Large Scale Deployment – This task developed the database necessary to provide cost estimates and differential cost for various fuel manufacturing options. Efforts were initiated to tabulate and quantify estimates regarding projected cost, reliability, and plant life.

RESEARCH ACCOMPLISHMENTS

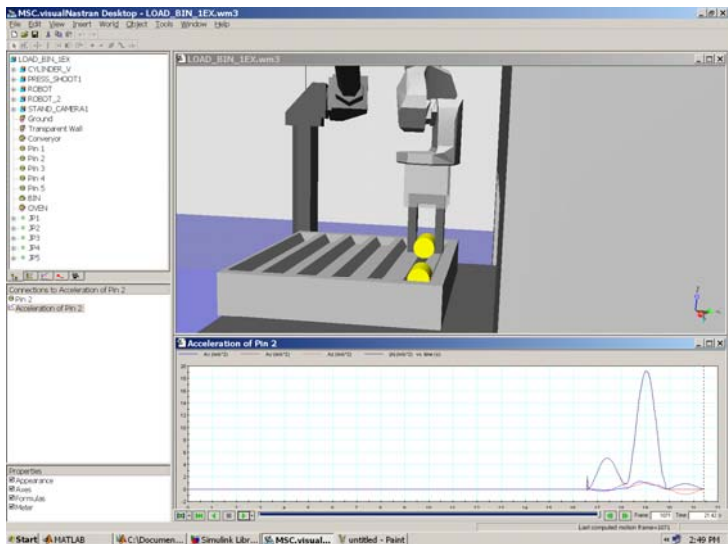
A special simulation model with a Waelischmiller hot cell robot was developed and coupled with MatLab control software. MatLab provides the interface with the robot and is used to control the system. This renders a realistic simulation of the forces and torques present during robot motion. A 3-D manufacturing process simulation using computer aided design models and the Newtonian dynamics of the moving components was developed.

Results exist in the form of movies, data sets, and images. Simulations for several robot types were developed and their proper kinematic configuration was verified. The simulations permit the detailed analysis of forces and torques in any modeled part or component.

While the simulation process generally worked flawlessly, the simulation time rose considerably as more details were added to the simulation. The speed of the simulation has been increased about 100-fold by moving to fast dual-processor computers.

Efforts to develop a vision-based methodology for locating and identifying objects within the robot's workspace were conducted using the Artificial Intelligence algorithm for object identification.

Another accomplishment involved the development of algorithms for knowledge based pattern recognition using IF (a set of conditions is satisfied) THEN (a set of consequences can be executed) routines. Other simulation variables established included pattern matching using clustered indexing vectors containing information about an object and feature vector indexing, where a 3-D object is segmented into a set of simple geometric features. Each feature is stored with its vector segmentation and geometry information (magnitude, inner angle, etc.).



TASK 9 PROFILE

Start Date: August 2001

Completion Date: August 2004

(This work continued under Task 22)

Thesis Generated:

Jae-Kyu Lee, Ph.D., Department of Mechanical Engineering, "Three Dimensional Pattern Recognition using Feature-based Indexing and Rule-based Search," December 2003.

Conference Proceedings:

J.K. Lee and G. Mauer, "Feature-Based Pattern Recognition and Object Identification for Telerobotics," International Conference on Mechatronics, Institute of Electrical and Electronics Engineers, Seoul, South Korea, July 10-12, 2005.

G.F. Mauer and J. Renno, "Virtual Testing of Robotic Assembly Processes for Hot Cells," *Proceedings*, 10th International Conference on Robotics and Remote Systems for Hazardous Environments, March 2004.

G.F. Mauer and J. Renno, "Design and Analysis of Robotic Manufacturing Processes," *Proceedings*, American Nuclear Society Winter Annual Meeting, New Orleans, LA, November 2003.

G.F. Mauer, "Design and Evaluation of Processes for Transmuter Fuel Fabrication," *Proceedings*, American Nuclear Society Winter Annual Meeting, November 2002.

G.F. Mauer, "Object Recognition Over An Expanded Range Of Viewing Angles Using Indexing Methods," *Proceedings*, International Conference on Computer Applications in Industry and Engineering, San Diego, CA, November 2002.

Visual Nastran Analysis of Pellet accelerations. The simulation verified that pellet's acceleration did not exceed a specified maximum of 20 m/s².

Research Staff

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Task 10

Development of a Mechanistic Understanding of High-Temperature Deformation of Alloy EP-823

A. K. Roy and B.J. O'Toole

BACKGROUND

During the transmutation process, a significant amount of heat can be generated in a molten lead-bismuth eutectic (LBE) target, which will be contained in a subsystem structural container made of a suitable martensitic iron-chromium-molybdenum (Fe-Cr-Mo) stainless steel such as Alloys EP-823, HT-9 and 422. These materials will be subjected to high tensile stresses while they are in contact with the molten LBE at temperatures ranging between 400 and 600°C. Therefore, a research program was conducted to evaluate the deformation characteristics of all three alloys in properly heat-treated conditions at temperatures relevant to the operating conditions.

RESEARCH OBJECTIVES AND METHODS

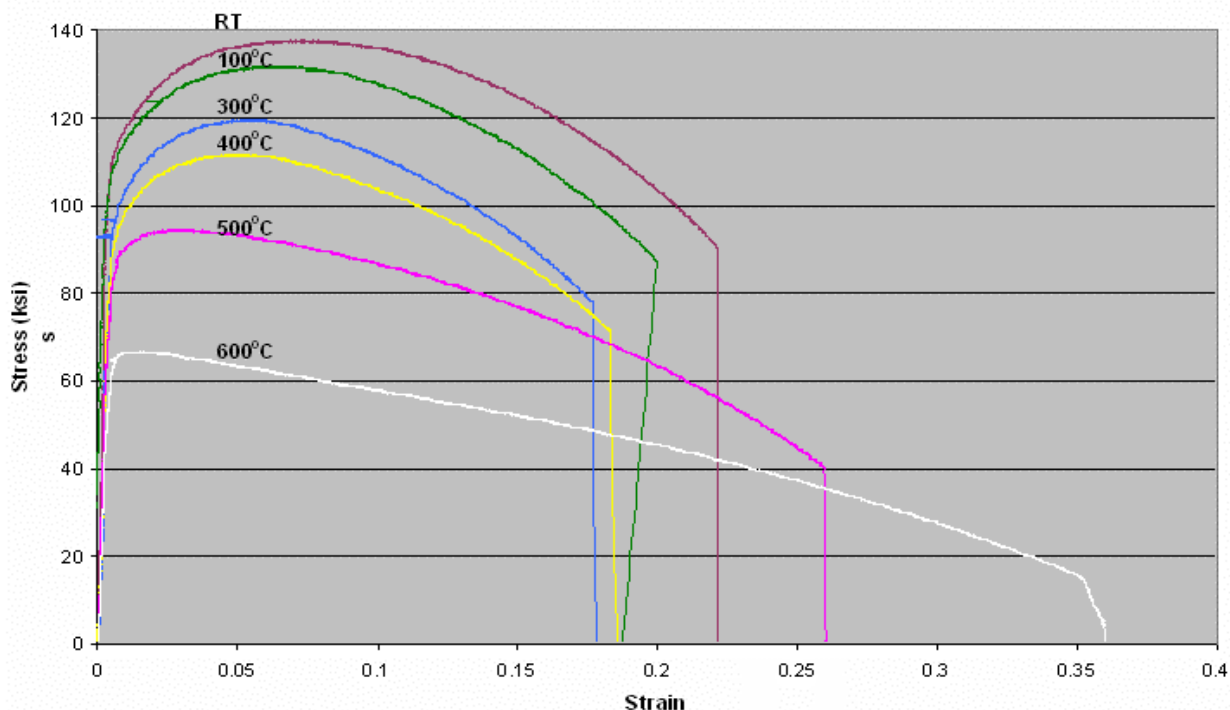
Experimental heats of Alloys EP-823, HT-9 and 422 were vacuum-induction-melted, followed by forging and hot rolling. The hot-rolled bars were subsequently thermally treated to produce a fully tempered and fine-grained martensitic microstructure without any retained austenite. They were austenitized at 1010°C, oil-quenched, and tempered at 621°C. The tempering operation was performed for 1.25, 1.75, and 2.25 hours, respectively, to study the effect of tempering time on the tensile properties. The hardness of all materials, before and after tempering, was measured using the Rockwell hardness scale. The tensile properties were determined at temperatures ranging from ambient to 600°C using smooth cylindrical specimens at a strain rate of 10^{-3} sec^{-1} according to the American Society for Testing and Materials (ASTM) Designation

E 8. A minimum of two specimens were tested under each condition, with the average value recorded. The metallurgical microstructures of the tested specimens were evaluated by optical microscopy. The primary fracture surface of the tensile specimens was analyzed by scanning electron microscopy (SEM) to determine the extent and morphology of failure. The resultant data include the percentage elongation (%El), percentage reduction in area (%RA), yield strength, and ultimate tensile strength as functions of the testing temperature and thermal-treatments. At least two specimens per material were tested under each of the three metallurgical conditions at the desired temperatures.

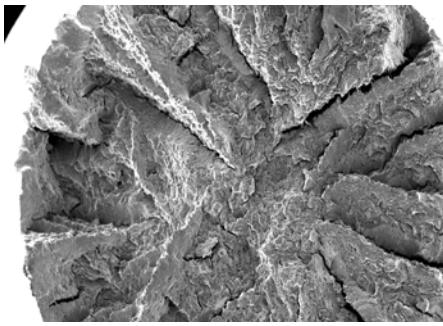
RESEARCH ACCOMPLISHMENTS

The significant conclusions drawn from this investigation are summarized below:

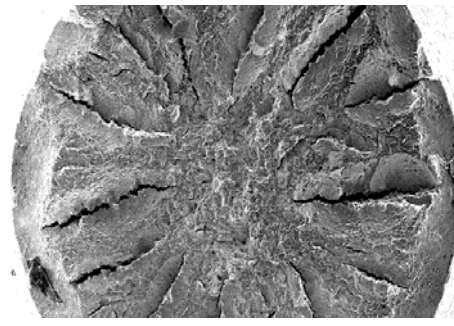
- The hardness of all three austenitized and quenched alloys were significantly reduced due to tempering, showing a gradual reduction with increasing tempering time.
- The magnitude of the yield, ultimate and failure stress were gradually reduced with increasing temperature, showing significant reductions at temperatures above 400°C.
- The extent of ductility in terms of %El and %RA was reduced to some extent in the temperature regime of ambient to 300°C due to strain hardening. However, beyond 300°C, the magnitude of these parameters was enhanced due to increased plastic flow.



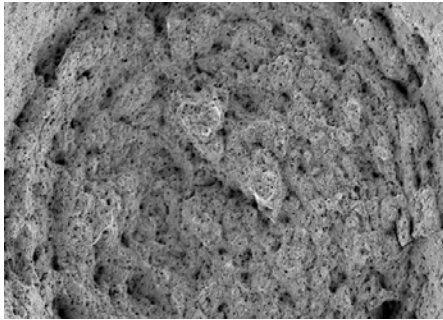
Stress-strain curves for Alloy HT-9 at different temperatures.



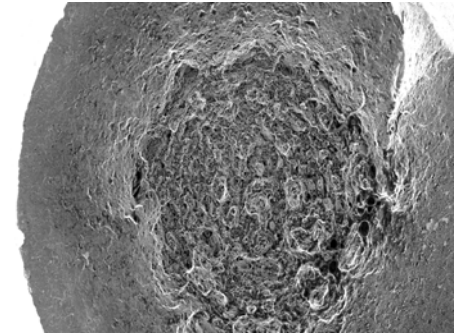
Room Temperature (RT)



100°C



300°C



600°C

Scanning Electron Microscope micrographs of Alloy HT-9 at various temperatures (35X).

- The morphology of failure was characterized by increased plastic deformation at elevated temperatures. Reduced cracking and dimpled microstructures were observed on the fracture surfaces indicating improved ductility at higher temperatures.
- The tempering time did not influence the metallurgical microstructure and the resultant tensile properties to any great extent irrespective of the testing temperature.

Bhagath Yarlagadda, M.S., Department of Mechanical Engineering, "Elevated Temperature Mechanical Properties and Corrosion Characteristics Evaluation of Alloy HT-9," August 2004.

Journal Article:

A.K. Roy, S. R. Kukatla, B. Yarlagadda, V. N. Potluri, M. Lewis, M. Jones and B. J. O'Toole, "Tensile Properties of Martensitic Stainless Steels at Elevated Temperatures," *Journal of Materials Engineering and Performance*, American Society of Metals International, April 2005, Vol. 14, No. 2, pp. 212-218.

Conference Proceedings:

A.K. Roy, S.R. Kukatla, and B. Yarlagadda, "High Temperature Deformation Characteristics of Martensitic Stainless Steels," Society for the Advancement of Material and Process Engineering, International Technical Conference, Long Beach, CA, May 16-20, 2004.

M. Lewis, M. Jones, A.K. Roy, and B.J. O'Toole, "High-Temperature Deformation of Alloy EP-823 for Transmutation Applications," American Nuclear Society Conference, San Diego, CA, June 1-5, 2003.

TASK 10 PROFILE

Start Date: August 2001

Completion Date: August 2004

Theses Generated:

Srinivasa R. Kukatla, M.S., Department of Mechanical Engineering, "Corrosion and High-Temperature Deformation Characteristics of a Target Structural Material for Transmutation Applications," June 2004.

Venkata Nagarjuna Potluri, M.S., Department of Mechanical Engineering, "Effect of Heat Treatment on Deformation and Corrosion Behavior of Type 422 Stainless Steel," August 2004.

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Task 11

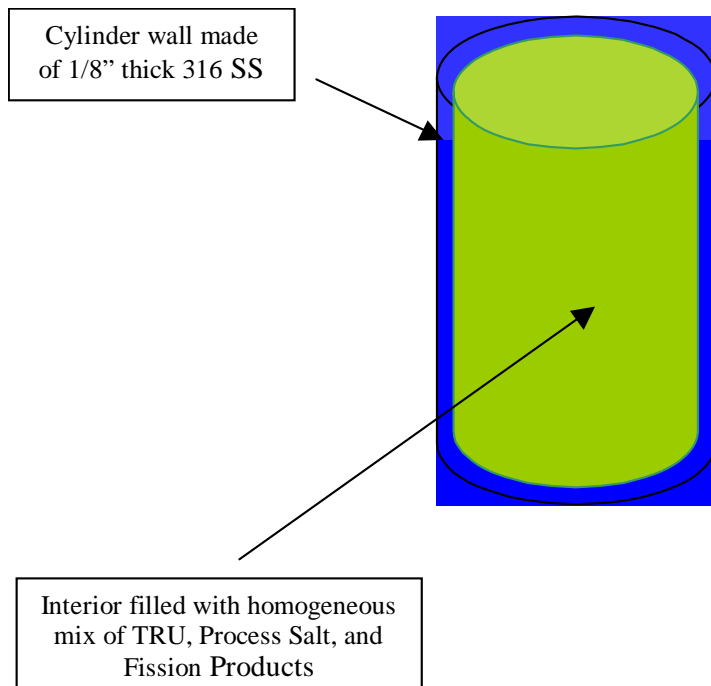
Nuclear Criticality, Shielding, and Thermal Analyses of Separations Processes for the Transmutation Fuel Cycle

W. Culbreth and D. Beller

BACKGROUND

The first step in any transmutation strategy is the separation of radionuclides in used nuclear fuel. The current separation strategy supporting the Advanced Fuel Cycle Initiative (AFCI) program is based on the use of a solvent extraction separation process to separate the actinides, fission products, and uranium from used commercial nuclear fuel, and on the use of pyrochemical separation technologies to process used transmuter fuels. To separate the fission products and transuranic elements from the uranium in used fuel, the national program is developing a new solvent extraction process, the Uranium Extraction Plus, or UREX+, process based on the traditional solvent extraction reprocessing technologies.

Preparing fuel for possible burn up in light water reactors, fast reactors, or accelerator-driven systems involves various chemical processes to partition the transuranics (neptunium, americium, plutonium, and curium) from the fission products. This results in waste streams that are highly radioactive and require radiation shielding for safety. These transuranic elements pose varied criticality, thermal, and radiation risks during storage and handling. Additionally, the radioactive decay of strontium and cesium waste products of the UREX+ technique produce roughly half of the thermal products and gamma radiation emissions in spent fuel. These radioisotopes require storage for approximately 300 years before heat and radiation hazards decrease to a safe level.



Cylindrical problem with TRU, process salt, and fission products.

As the volume of waste requiring treatment increases, a higher probability exists that fissionable isotopes of plutonium, neptunium, and curium can accumulate and form a critical mass. Criticality concerns warrant an assessment of the effective neutron multiplication factor, or k_{eff} , to prevent a possible sustained fission reaction. Maintaining k_{eff} below a safe level (<0.95) prevents criticality events. This parameter can be computed for any combination of fuel and geometry using Monte Carlo neutron transport codes. Monte Carlo simulations establish the best means of examining the criticality safety of the proposed separation processes, and allow engineers to develop proper safety measures for the reprocessing and fabrication of actinide fuels.

Candidate storage containers also require analysis to assess the need for radiation shielding. Since minor actinides generate significant amounts of heat through radioactive decay, proposed containment measures must be designed to avoid excessive temperatures. Radioactive decay also generates heat that can lead to melting of the fuel during storage and handling.

RESEARCH OBJECTIVES AND METHODS

The primary goal of this research program was to provide the nuclear and thermal modeling support for the development of this new separation process. The assessments of nuclear criticality, radiation for shielding, and thermal analyses of wastes in the Cs/Sr, Pu/Np, and Cm/Am waste streams will assist in designing the UREX+ process. This project was identified as a critical R&D need of the Chemical Technology Division (CTD) at the Argonne National Laboratory (ANL) as safety concerns associated with criticality, shielding, and heat buildup must be addressed prior to further development of the UREX+ process.

UNLV students used nuclear analysis codes to perform assessments of k_{eff} at different points in the separation processes that have been identified by the project collaborators at ANL-CTD. They also worked on problems to assess the need for radiation shielding and to develop software to assess the possibility of excessive temperatures due to radioactive decay in separated wastes. ANL-CTD has provided sample fuel process geometries and compositions for calculation of k_{eff} as a function of the relative concentrations of process salt, transuranics, and fission products.

RESEARCH ACCOMPLISHMENTS

An investigation and analysis of criticality and thermal effects for the safe storage of curium was completed. The assessment involved determining k_{eff} as a function of fuel burnup, initial enrichment, and time since irradiation. Additionally, since curium generates a substantial quantity of decay heat, an analysis was com-

pleted to determine the mass of curium that will lead to temperatures high enough to melt the metal. A spherical geometry was used in the analysis. Heat removal from the sphere was assumed to be a combination of natural convection and radiation heat transfer. This heat transfer analysis was also modified and resulted in an analysis that utilized a more sophisticated and suitable cylindrical container. A report developed for Dr. Laidler at ANL described the in-depth investigation regarding the criticality and thermal properties of curium. The heat transfer spreadsheet was used for other combinations of minor actinides as indicated by ANL.

Investigators also investigated the properties of the other minor actinides, including plutonium and americium. A study was conducted on the values of k_{eff} and ultimate temperature for varying combinations of plutonium, americium, and the remaining minor actinides.

UNLV students used nuclear analysis codes SCALE 4.4 and/or MCNPX to perform assessments of k_{eff} at different points in separation processes that have been identified by ANL-CTD. They also worked on problems to assess the needs for radiation shielding and develop software to assess the possibility of excessive temperatures due to radioactive decay in separated wastes. ANL-CTD has provided sample fuel process geometry and composition for calculation of k_{eff} as a function of the relative concentrations of process salt, transuranic actinides (TRU), and fission products. The research team analyzed the cesium/strontium waste stream, the plutonium/neptunium waste stream, and the americium/curium waste stream.

TASK 11 PROFILE

Start Date: August 2001

Completion Date: August 2004

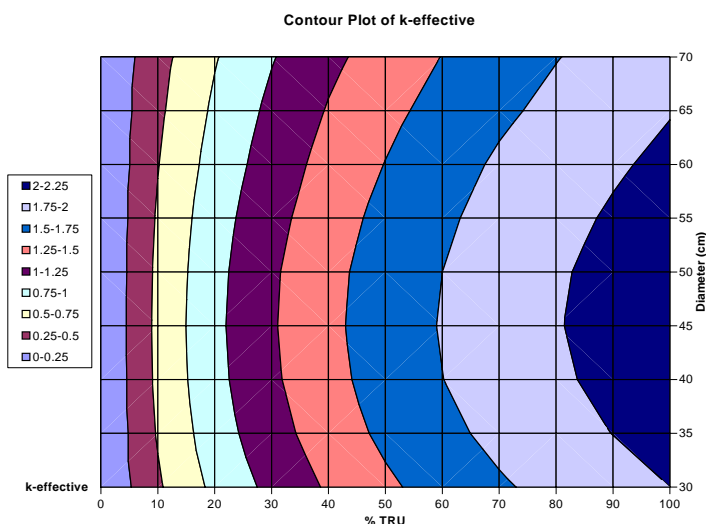
Thesis Generated:

Elizabeth Bakker, M.S., Mechanical Engineering Department, "Criticality and Thermal Analysis of Separated Actinides in Transmutation," November 2003.

Reports:

B. Culbreth, E. Bakker, and J. Viggato, "The Fission Properties of Curium Separated from Spent Nuclear Fuel," Department of Mechanical Engineering, University of Nevada, Las Vegas, April 2002.

B. Culbreth, D. Lowe, and J. Viggato, "Assessment of Criticality Safety for Cylindrical Containers to be Used In the Processing of Spent Fuel," Department of Mechanical Engineering, University of Nevada, Las Vegas, October 28, 2001.



Contour Plot of the Effective Neutron Multiplication Factor as a function of cylinder diameter and % TRU in the mixture.

| Cm Metal Cylinder | | Criticality | | Heat Transfer | |
|---|-------|-------------|-----------|---------------|-----------|
| | | rad (cm) | mass (kg) | rad (cm) | mass (kg) |
| | bare | 4.6 | 82.52 | 1.1 | 1.129 |
| | water | 2.7 | 16.69 | 3.3 | 30.48 |
| Cm ₂ O ₃ Cylinder | | Criticality | | Heat Transfer | |
| | | rad (cm) | mass (kg) | rad (cm) | mass (kg) |
| | bare | 5.4 | 125.59 | 1.55 | 2.74 |
| | water | 3 | 21.53 | 2.25 | 8.37 |
| Am ₂ O ₃ Cylinder | | Criticality | | Heat Transfer | |
| | | rad (cm) | mass (kg) | rad (cm) | mass (kg) |
| | bare | 11.2 | 1032.28 | 1.6 | 3.01 |
| | water | 10.5 | 850.58 | 1.9 | 5.04 |
| Am ₂ O ₃ +Cm ₂ O ₃ Cylinder | | Criticality | | Heat Transfer | |
| | | rad (cm) | mass (kg) | rad (cm) | mass (kg) |
| | bare | 10 | 737.9 | 1.4 | 2.02 |
| | water | 9.3 | 593.54 | 2 | 5.88 |

Recommendations for Cm and Am oxide inventories based on criticality and melting temperature.

Research Staff

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 George F. Vandergriff, III, Senior Scientist, Chemical Technology Division, Argonne National Laboratory

Task 12

Radiation Transport Modeling using Parallel Computational Techniques

W. Culbreth and D. Beller

BACKGROUND

One of the most significant tools available for the design and analysis of accelerator-driven systems, such as the systems proposed for transmutation, is the high-energy particle transport code MCNPX. The MCNPX code suite, developed by the national laboratories, allows researchers and engineers to model the complex interactions of high-energy particles with the target and related systems, including the spallation reaction and subsequent neutron multiplication expected in the accelerator targets.

The next stage in the development of the MCNPX code suite is to validate the code by comparing the theoretical predictions from the models with experimental observations. Additionally, the nuclear database, particularly the cross sections (i.e., reaction probabilities) for high-energy particle interactions, needs to be revisited to reduce the uncertainties associated with key nuclear properties.

The Department of Energy, through its national laboratories, has initiated several experiments geared towards removing uncertainties in the MCNPX libraries, with more in the planning stages. These experiments utilize the proton and neutron beam lines at the LANSCE proton accelerator at the Los Alamos National Laboratory (LANL) to irradiate a target, producing a pulse of neutrons which are observed by the experimenters. The results of these experiments are then compared against the predictions from the MCNPX models of the system. By comparing the predicted system behavior to the data acquired from the experiments, the experimenters will be able to validate the MCNPX code and its nuclear data libraries.

Through this project, UNLV researchers were involved in support of these experiments by developing the system models in MCNPX and benchmarking/validating the models against the experimental results. UNLV students were also involved in conducting experiments at LANL and in assisting researchers in designing new experiments.

RESEARCH OBJECTIVES AND METHODS

This project involved modeling several aspects of the LANSCE beam experiments:

- Modeling targets of varying diameter in air, in a vacuum, and in the presence of humid air;
- Modeling various proton beam profiles;
- Modeling the effects of off-axis proton beam impingement on the target;
- Modeling the asymmetry introduced by the steel table below the target;
- Modeling the effect of varying ratios of Pb to Bi and the effect of impurities; and
- Modeling the system, including other structures within the test room.

With the experience gained through modeling these systems, the UNLV researchers developed, with the assistance of their national laboratory collaborators, a benchmark program for the neutron leakage tests and other tests related to transmuter development. A comprehensive three-dimensional computer-aided design (CAD) image of the LANSCE experiments was prepared using ProEngineer to help benchmark the experiments and provide accurate geometric data for MCNPX modeling.

RESEARCH ACCOMPLISHMENTS

Undergraduate student Daniel Lowe worked on neutron spallation tests at the LANSCE facility (Summer 2002). He performed MCNPX runs and worked on calculations for initial benchmarking data. His early MCNPX calculations helped the experimenters determine where foil packets should go and what types of neutron flux to expect from these foils. He also prepared foils to determine neutron flux from the experiment and assisted in radiation counting of the foils. Mr. Lowe completed Solid Works CAD models of the Blue Room at LANSCE and conducted MCNPX simulations of the summer experiments when he returned back to UNLV. His MCNPX runs included estimations of the effect of the proton beam striking the target at positions slightly off of the centerline. He also estimated the neutron energy spectra expected from the time-of-flight neutron detectors.

Through MCNPX simulations of the neutron leakage from lead-



Robert O'Brien (left of center) and Daniel Lowe (far right) are both undergraduate Mechanical Engineering students working on the project.

bismuth targets, the UNLV team was able to assist in the design of the experimental configurations for the LANCSE experiments. These models were also used to predict the results for the experiments, and assist in positioning detectors for measuring the leaking. Similar computational support was also provided for proton activation experiments in sodium coolant.

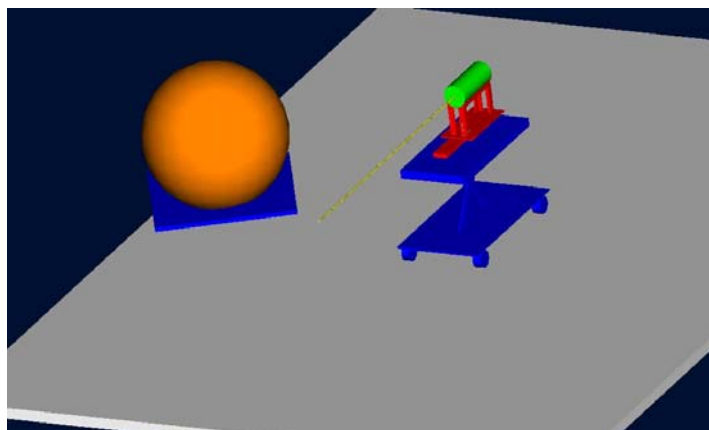
Extensive studies on how MCNPX performs with respect to MPI (Message Pass Interface) and PVM (Parallel Virtual Machine) have been run. PVM was no longer supported by the LANL team after 2005, hence more emphasis was placed on how MCNPX runs with MPI on a Beowulf system.

Parallelization of MCNPX for a Parallel Virtual Machine was completed. MPI bugs and compiling problems were resolved.

Analysis of linearization characteristics on a Beowulf cluster was completed. Work was then focused on characteristics of the Supercomputing Center and the linearization of criticality studies.

Benchmarking and optimization of MCNPX to run on multiple platforms was performed. This insured that the user will not be limited to a specific system type when running simulations. In addition, the MCNPX simulations of experiments was performed.

User guides were developed for future users. These will describe how to implement an optimized version of MCNPX on a heterogeneous cluster using a Message Passing Interface. Efforts to increase the speed of MCNPX on parallel clusters of computers was conducted and a preliminary graphical user interface (GUI) for MCNPX using open source code and tools was developed. The GUI interface is written in Visual Basic allowing easy integration into Excel codes that contain databases of results from criticality and heat transfer studies of waste storage containers.



Schematic of the experimental facility at LANSCE used for modeling.

With the help of Trevor Wilcox, a doctoral student in mechanical engineering, the MCNPX software was ported to a Beowulf cluster located in the Tiberti Laboratory at UNLV. The cluster has between 32 and 50 processors available to run a single MCNPX job.

TASK 12 PROFILE

Start Date: August 2001

Completion Date: August 2004

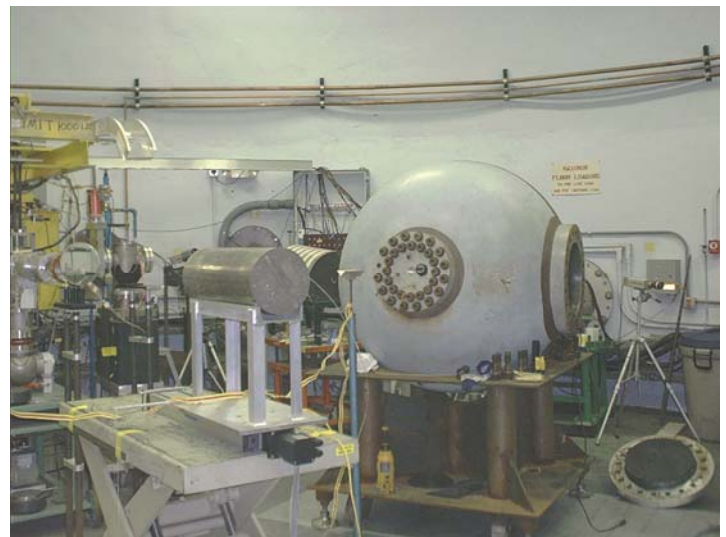
Thesis Generated:

Suresh B. Sadineni, M.S., Mechanical Engineering Department, "Benchmarking Photoneutron Production of MCNPX Simulations with Experimental Results," December 2002.

Conference Proceedings:

M.A. Reda, J.F. Harmon, and S.B. Sadineni, "Properties of Photo-Neutron Sources for Accelerator Driven Sub-Critical Systems," American Nuclear Society Conference, June 1-5, 2003, San Diego, CA.

M.R. James, R.T. Klann, G.L. Morgan, E.J. Pitcher, M.A. Paciotti, J.M. Oostens, J.E. Platte, and D.R. Lowe, "Measurements from Activation Foils of a Proton Irradiated Lead-Bismuth Target," American Nuclear Society Conference, June 1-5, 2003, San Diego, CA .



Experimental facility at LANSCE, Los Alamos, NM.

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Laurie S. Waters, Los Alamos National Laboratory

Task 13

Developing a Sensing System for the Measurement of Oxygen Concentration in Liquid Pb-Bi Eutectic

Y. Jiang and B. Fu

BACKGROUND

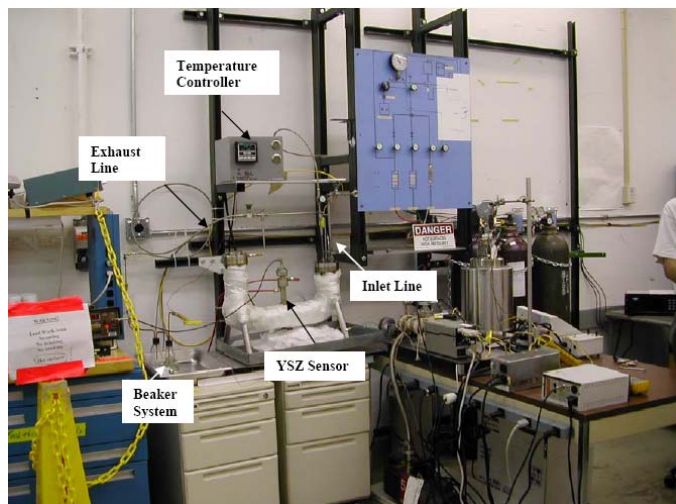
Although liquid lead-bismuth eutectic (LBE) is a good candidate for the coolant that can be employed in a subcritical transmutation blanket, it is known to be very corrosive to stainless steel, the material used in the containment structure. To mitigate this problem, trace levels of oxygen can be introduced into the system, causing the formation of a protective oxide layer at the interface between the LBE and steel. The proper formation of this oxide layer largely depends on the accurate measurement and subsequent control of the oxygen concentration in liquid LBE.

Yttria Stabilized Zirconia (YSZ) oxygen sensors, using molten bismuth saturated with oxygen as the reference, have been utilized to accurately measure the concentration of oxygen dissolved in LBE. By measuring the voltage difference across the YSZ sensor, the oxygen concentration in test solutions can be determined relative to that in the reference solutions (the potentiometric method). The theoretical model for calculating oxygen concentration based on voltage measurements from YSZ sensors in static conditions is well understood. The real world performance of these systems, however, is less predictable.

RESEARCH OBJECTIVES AND METHODS

The research objectives of this project were as follows:

- To generate calibration curves of voltage versus oxygen concentration for the YSZ oxygen sensor system under various temperatures in liquid LBE.
- To determine the sensor characteristics of the YSZ sensor system.
- To determine oxygen dissolving rates in LBE under different temperatures in vitro.
- To study the effects of unwanted electrical conductivity, contributed by the mobility of the electrons at high temperatures, for more accurate oxygen measurement.



The first sensor calibration experimental setup.

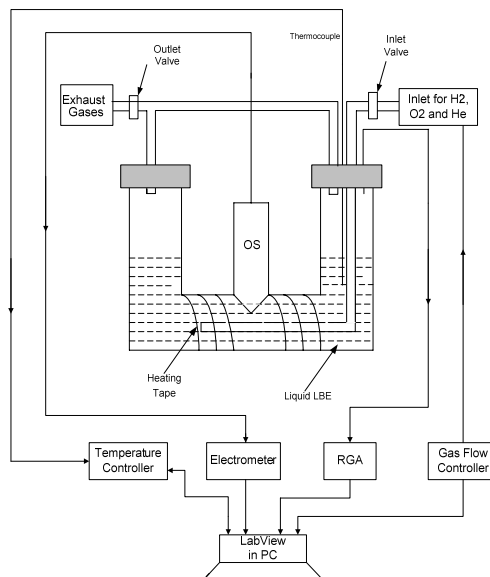
- To study alternative and promising oxygen measuring methods.

RESEARCH ACCOMPLISHMENTS

The first experimental set up consisted of a temperature controlled U-shape container, gas supplies and exhaust, a residual gas analyzer (RGA), a high-impedance electrometer, and a computer for data acquisition. The container is tightly sealed from the outside atmosphere using a conflat flange except for gas inlet and outlet, and openings for insertion of thermocouple or RGA signal wire. Flexible heating tapes around the container heated the liquid metal to the required temperature under the control of a temperature controller. Meanwhile, the thermally insulated container is placed on a rocker to provide fluid motion that promotes mixing and homogenization of oxygen concentration in LBE.

Although some calibration results were obtained using this setup, it had several shortcomings. For instance, it was impossible to heat the system to reach the desired high temperature (up to 750 °C). In addition, direct injection of O₂/H₂ method adopted in this system is unlikely to produce a required extremely low oxygen level (ppm to tens of ppb) in liquid LBE. These problems in part were solved by developing a new apparatus. Some noticeable features of this apparatus were:

- Instead of using a steel tube, this apparatus incorporates a cylindrical crucible made of Magnesia Stabilized Zirconia (MSZ), and the liquid LBE is contained in this MSZ crucible. MSZ was chosen because of its desirable material properties.
- The MSZ crucible sits on a stainless steel beaker. The beaker acts as a pressure boundary, and it can distribute the weight of the molten metal and crucible to the outer support. Back-up materials are used to fill the gap between the inner crucible and beaker.



- The crucible is tightly sealed with a metallic flange, with only a few openings left for gas inlets and outlets.
- A stirring unit is employed in order to mix the gases with the molten metal. The stirrer is made of Silicon Nitride (Si₃N₄) ceramic. Silicon Nitride has high temperature strength, creep resistance, oxidation resistance, and it is not wetted by any molten metal. On top of those, compared to

many other ceramic materials, Si_3N_4 has good mechanical strength.

Oxygen Sensor Calibration Results

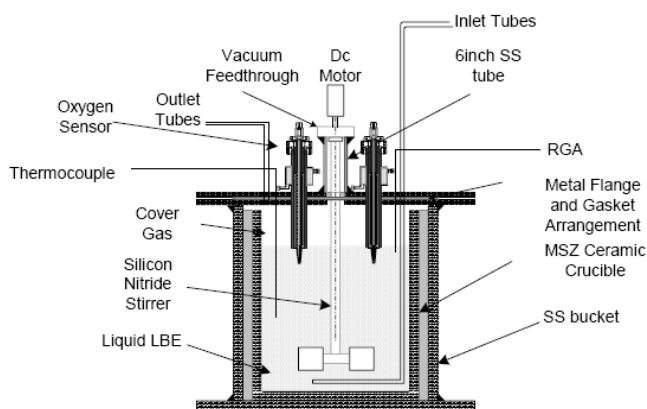
Calibration curves were obtained from two oxygen sensors. After some initial transients, voltage-temperature curves first follow constant oxygen concentration lines according to the Nernst equation, and then turn to the concentration saturation line.

The slopes of experimental and theoretical curves are almost identical, ranging from 0.33 to 0.5. This indicates that the YSZ sensors are of high sensing quality. Overlapping of the calibration curves for the two sensors indicates consistency in the sensors of the same design.

Experimental results show that tantalum oxidizes at high temperature around 480°C after running for a certain amount of time. This directly causes tantalum wires to lose electrical conductivity, and consequently, the sensor fails to provide meaningful signal output. This effect was demonstrated. In the first trial experiment cycle, molybdenum, stainless steel (SS) and tungsten were tested for this purpose. When introducing hydrogen and helium mixture into the system to clean excessive oxygen in the LBE container, it was found that Mo responds a little faster than SS.

It had been suspected that different combinations of Bi or Bi_2O_3 may have affected the sensor response. During Phase III of this project, several experiments were performed and it was determined that there are no evident effects on the sensor response. Experiment results further confirmed that even a small amount of residual oxygen inside sensor tubes will be sufficient for Bi to be oxidized.

Commercial FEM (finite element method) software, FEMLAB



Schematic of the second sensor calibration experimental setup.

was used to simulate the oxygen dissolving process and the distribution in liquid LBE for the new system under three different boundary conditions. Simulation results show that the oxygen concentration distributes evenly in the whole flow field due to the strong convection flow in LBE. These simulation results with the experimental measurements help to not only determine the oxygen dissolving rate and the diffusion coefficient under different temperatures, but also provide suggestions for better experimental design.

TASK 13 PROFILE

Start Date: May 2002

Completion Date: May 2005

Theses Generated:

Ramkumar Sivaraman, M.S., Department of Electrical and Computer Engineering, "Calibration of YSZ Sensor for the Measurement of Oxygen Concentration in Liquid LBE," December 2003.

Xiaolong Wu, M.S., Department of Electrical and Computer Engineering, "Instrumentation of YSZ Oxygen Sensor Calibration in Lead-Bismuth Eutectic," December 2004.

Journal Article:

J. Ma, P. Guo, J. Zhang, N. Li, B. Fu, "Enhancement of Oxygen Transfer in Liquid Lead and Lead-Bismuth Eutectic by Natural Convection" *International Journal of Heat and Mass Transfer*, Vol. 48 (13), p. 2601-2612, January 2005.

Conference Proceedings:

X. Wu, J. Ma, Y. Jiang, B. M. Fu, W. Hang, J. S. Zhang, and N. Li, "Instrumentation of YSZ Oxygen Sensor Calibration in Liquid Lead-Bismuth Eutectic," *Proceedings of 2005 IEEE International Symposium on Circuits and Systems*, May, 2005.

X. Wu, R. Sivaraman, N. Li, W. Hang, T. W. Darling, Y. Jiang, W. Yim, B. M. Fu, "Design of an Oxygen Sensor Calibration/Measurement Apparatus for the Liquid Lead-Bismuth Eutectic System," *Proceedings of the 2003 Annual Meeting*, San Diego, CA, June 1-5, 2003.

X. Wu, R. Sivaraman, N. Li, W. Hang, T. W. Darling, Y. Jiang, W. Yim, B. M. Fu, "Calibration of YSZ Sensors for the Measurement of Oxygen Concentration in Liquid Pb-Bi Eutectic," *Proceedings of the 10th International High-Level Waste Management Conference*, Las Vegas, March 30-April 3, 2003.

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Collaborators

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Task 14

Use of Positron Annihilation Spectroscopy for Stress-Strain Measurements

A. K. Roy

BACKGROUND

Engineering metals and alloys, when subjected to tensile loading beyond a limiting value, undergo plastic deformation resulting in lattice defects such as voids and dislocations. These imperfections interact with the crystal lattice, producing a higher state of internal stress, also known as residual stress, which can be associated with reduced ductility. Residual stresses are also generated in welded structures due to rapid solidification and resultant dissimilar metallurgical microstructures between the weld and the base metals. Development of these internal stresses is often influenced by incompatible permanent strain resulting from thermal and mechanical operations associated with welding and plastic deformation. These types of operations can cause premature failures in structural materials unless these stresses are relieved by thermal treatments, which are commonly known as stress-relief operations.

During the past academic year, this project was focused on the characterization of residual stress in welded specimens consisting of austenitic and martensitic stainless steels using an activation technique based on the Positron Annihilation Spectroscopic (PAS) method. The extent of residual stress was expressed in terms of three line-shape parameters (S-, W- and T-). Further, efforts were made to characterize linear lattice defects such as dislocations in the vicinity of Fusion-Line (FL), Heat-Affected-Zone (HAZ), and the base material of the welded specimens using Transmission Electron Microscopy (TEM). The metallurgical microstructures at these three regions have also been evaluated by optical microscopy.

RESEARCH OBJECTIVES AND METHODS

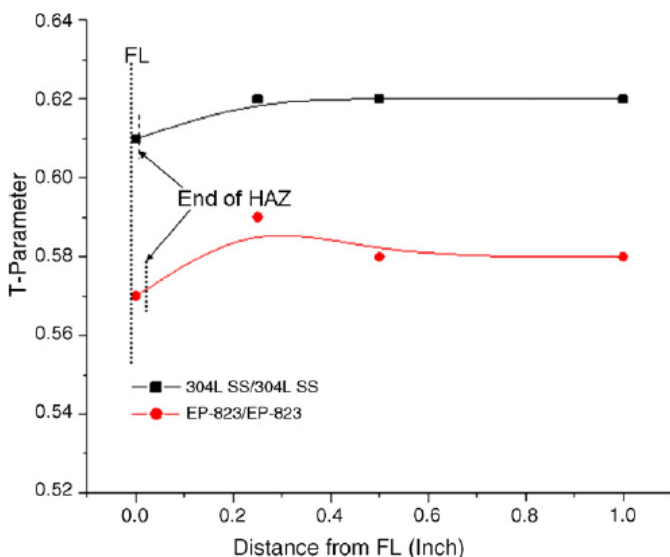
The primary objective of this task was to evaluate the feasibility of the characterization of residual stresses in plastically-deformed and welded structural materials using a new nondestructive technique based on PAS. The residual stresses measured by a modified PAS method have been compared to those measured by three other techniques, namely the Ring-Core (destructive), X-Ray Diffraction (non-destructive), and Neutron Diffraction (non-destructive).

All four techniques have been used to evaluate residual stresses in cold-worked, plastically deformed and welded specimens of austenitic Type 304L Stainless Steel (SS), and martensitic Alloys EP-823 and HT-9. Alloy EP-823 is a leading target structural material to contain the molten lead-bismuth eutectic nuclear coolant needed for fast spectrum operations of an Accelerator-Driven Transmutation System. Type 304L SS is a universally-known corrosion resistant low-carbon iron-nickel-chrome alloy having optimum formability and weldability. Alloy HT-9 is known for its superior high temperature tensile properties. The metallurgical microstructures and the nature of defects have been analyzed by optical microscopy, scanning electron microscopy, and TEM.

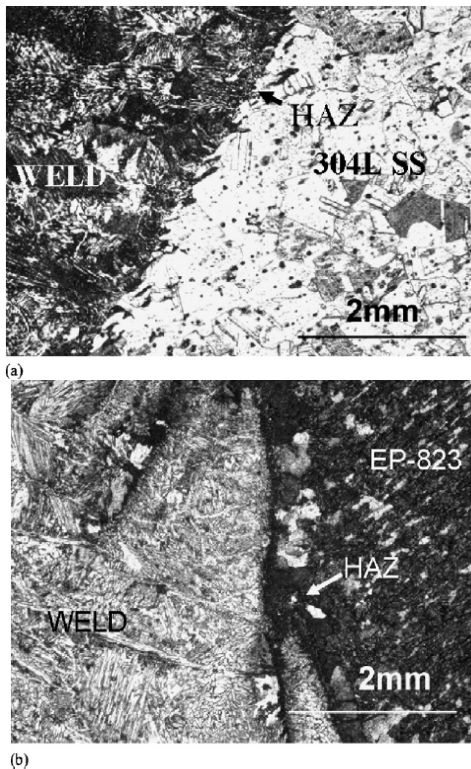
RESEARCH ACCOMPLISHMENTS

The significant results obtained from this investigation during the past academic year are summarized below.

- For welded specimens consisting of similar materials on both sides, the residual stress in terms of the S-, W-, and T-parameters was maximum at the FL. A gradual drop in residual stress was observed with these specimens at locations away from the FL.
- The extent of residual stress was higher in martensitic Alloy EP-823 compared to that of austenitic Type 304L SS, irrespective of the weld configuration.
- Compressive residual stresses were observed in Alloy EP-823, when welded to Type 304L SS.
- The magnitude of dislocation density (ρ) was substantially higher at the HAZ compared to that of the base material of the welded specimens consisting of similar materials (Type 304L SS or Alloy EP-823) on both sides.
- In the case of the welded specimen of dissimilar materials (Type 304L SS and Alloy EP-823) on the opposite side, the concentration of dislocation in terms of ρ was greater at the HAZ on the Alloy EP-823 side of the weld.
- The enhanced value of ρ at the HAZ on the Alloy EP-823 side of the weld may be attributed to the faster rate of solidification of this alloy compared to that of the austenitic SS.
- The sizes of the HAZ on the Alloy EP-823 sides of the welded specimens were relatively larger, irrespective of the weld configuration.



T-Parameter versus Distance from FL for Welded Specimens of Similar Materials



Optical micrographs of welded specimens of similar materials. (a) Type 304L SS/Type 304L SS, $\text{HNO}_3 + \text{CH}_3\text{COOH} + \text{C}_3\text{H}_5(\text{OH})_3$; (b) Alloy EP-823/Alloy EP-823, Fry's reagent.

TASK 14 PROFILE

Start Date: May 2002
Completion Date: December 2006

Theses Generated

- Srinivas Chanda, M.S., Mechanical Engineering, "Characterization of Residual Stress and Defects in Welded Specimens," December 2006.
- Silpa B. Suresh, M.S., Mechanical Engineering, "Use of Neutron Diffraction and Microscopy for Characterization of Residual Stresses and Defects," December 2005.
- Subhra Bandyopadhyay, M.S., Mechanical Engineering, "Residual Stress Characterization and Defects Analyses by Microscopy," December 2005.
- Anand Venkatesh, M.S., Mechanical Engineering, "Comparative Analyses of Residual Stresses in Target Sub-System Materials," August 2004.
- Vikram Marthandam, M.S., Mechanical Engineering, "Metallurgical Characterization and Residual Stress Measurements of Target Structural Materials," August 2004.

Satish B. Dronavalli, M.S., Mechanical Engineering, "Residual Stress Measurements and Analyses by Destructive and Nondestructive Techniques," August 2004.

Journal Articles

- A.K. Roy, S. Chanda, A. Ghosh, P. Kumar, and L. Ma, "Defects Characterization of Welded Specimens by Transmission Electron Microscopy," *Materials Science and Engineering A*, Elsevier Science, Vol. 464/1-2, pp. 274-280, 2007.
- A.K. Roy, S. Chanda, D.P. Wells, A. Ghosh, and C.K. Mukhopadhyay, "Residual Stress Characterization of Welded Specimens by Nondestructive Activation Technique," *Materials Science and Engineering A*, Elsevier Science, Vol. 464/1-2, pp. 281-287, 2007.
- D.P. Wells, et al., "Gamma-induced Positron Annihilation Spectroscopy and Application to Radiation-damaged Alloys," *Nuclear Instruments and Methods in Physics Research A*, Elsevier Science, Vol. 562, pp. 688-691, 2006.
- A.K. Roy, et al., "Comparison of Residual Stress in Martensitic Alloys by Nondestructive Techniques," *Materials Science and Engineering A*, Elsevier Science, Vol. 419, pp. 372-380, 2006.
- A.K. Roy, et al., "Relationship of Residual Stress to Dislocation Density in Cold-Worked Martensitic Alloy," *Materials Science and Engineering A*, Elsevier Science, Vol. 416, pp. 134-138, 2006.
- A.K. Roy, et al., "Residual Stress Measurements in Welded and Plastically Deformed Target Structural Materials," *Journal of ASTM International*, Vol. 2, No. 6, pp. 1-13, June 2005.
- A.K. Roy, et al., "Residual Stress Characterization in Structural Materials by Destructive and Nondestructive Techniques," *Journal of Materials Engineering and Performance*, ASM International, Vol. 14, No. 2, pp. 203-211, April 2005.
- F.A. Selim, et al., "Stress Analysis using Bremsstrahlung Radiation," *Advances in X-ray Analysis*, Joint Committee on Powder Diffraction Standards, Vol. 46, pp. 106-111, 2003.

Conference Proceedings

Ten conference papers were also published. See TRP website at <http://aaa.nevada.edu> for more information.

Dislocation density (ρ) in welded specimens of different configurations

| Weld configuration | ρ (no./m ²) | |
|--------------------------------|------------------------------|----------------------|
| | Base material | HAZ |
| 304L SS/304L SS | 5.1×10^{13} | 2.4×10^{14} |
| EP-823/EP-823 | 7.6×10^{13} | 1.3×10^{14} |
| 304L SS side of 304L SS/EP-823 | 1.7×10^{13} | 3.2×10^{13} |
| EP-823 side of 304L SS/EP-823 | 6.5×10^{13} | 2.2×10^{14} |

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Task 15

Immobilization of Fission Iodine by Reaction with a Fullerene Containing Carbon Compound and Insoluble Natural Matrix

S.M. Steinberg, G.S. Cerefice, and D.W. Emerson

BACKGROUND

The recovery of iodine released during the processing of used nuclear fuel poses a significant challenge to the transmutation of nuclear waste. Iodine-129, a long-lived fission product formed by both commercial nuclear power generation and nuclear weapons production, is released when reprocessing nuclear fuel. Since iodine can be concentrated in the human thyroid, any uncontrolled release of iodine may result in an increased rate of thyroid cancer in the exposed population. For this reason, recovery of iodine is important for implementing any nuclear transmutation strategy.

The first step in any transmutation strategy is the processing of the used nuclear fuel. This step involves separating the used fuel into its constituent elemental components, allowing the recovery of the uranium, transuranic actinides, long-lived fission products, and other components, depending on the strategy and processes involved.

When used fuel rods are dissolved in concentrated nitric acid in preparation for actinide recovery, iodine is released from the fuel. A significant fraction of the iodine is lost to the vapor phase during this process, where it may potentially become a fugitive emission and be released from the plant. To avoid this, specialized filtration systems are used to try to trap and sequester the released iodine (and other fission product gases).

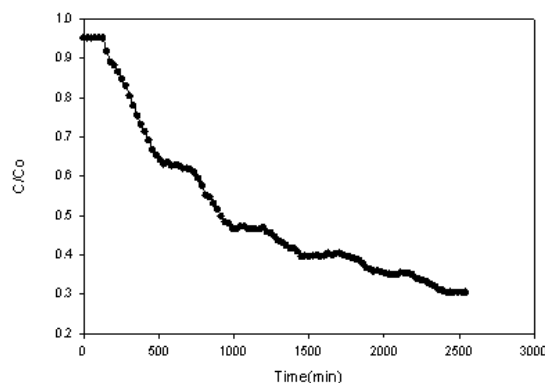
The primary goal of this research is to capture and immobilize the iodine released from these processes in a form that can easily be converted to a suitable target for neutron-induced transmutation. The investigators believe that iodine released during fuel reprocessing can be immobilized in a Fullerene Containing Carbon (FCC) compound or a Natural Organic Matter (NOM) matrix.

NOM (such as sphagnum moss, peat or brown coal) is an inexpensive and a renewable resource. Further processing of the trapped iodine using simple desorption or combustion processes should be able to produce iodine in a form suitable for transmutation. Furthermore, collaborators at the Khlopin Radium Institute (KRI) in St. Petersburg, Russia, have proposed that the iodine-loaded FCC material, when combined with ceramics, is stable enough for use as a long-term storage form, and may be usable as a transmuter target matrix.

RESEARCH OBJECTIVES AND METHODS

The stability of the association of iodine with FCC and NOM products were studied. Product distributions for the various matrices under various reaction conditions were examined in order to maximize the binding of iodine. The recovery of the iodine from the sequestration matrices was also examined, along with the conversion of the iodine to matrices more suitable for geological storage and/or use as transmutation targets.

Iodide Sequestration at pH 4
Continuous Flow method



Sequestration of iodide ($10^{-4}M$) in the presence of sphagnum peat and MnO_2 . The solution was circulated through a column at 5 mL/min.

The following are the specific research objectives and goals:

- Develop bench-scale experimental set-up and procedures for simulating plutonium extraction process (PUREX) head-end vapor phase.
- Develop experimental procedures for evaluating iodine sequestering methods using bench-scale procedures.
- Develop FCC bearing material as potential iodine sequestration matrix.
- Determine binding of iodine to FCC and NOM.
- Examine alternate iodine sequestration matrices using techniques developed for FCC and NOM studies.
- Examine the effect of reaction conditions on binding.
- Elucidate the nature of the reaction products (volatile, hydrophobic, soluble, insoluble).
- Develop methodology and host matrix for converting sequestered iodine to solid matrix for evaluation as transmutation target and/or disposal matrix.
- Examine recovery of iodine from sequestration matrices.

The FCC compounds were developed and prepared by the KRI Research Industrial Enterprise (KIRSI). The KRI-KIRSI team researched the impacts of process parameters on sorption of iodine, and examined the material properties, such as how iodine attaches to the FCC compounds. The KRI-KIRSI team also examined the conversion of the iodine loaded FCC compound to a stabilized matrix (similar to ceramic) for potential use as a disposal form, acceptable transportation material, or potential target material.

RESEARCH ACCOMPLISHMENTS

Observations related to the oxidation of iodide to iodine (I_2) or hypoiodic acid (HIO) by MnO_2 were continued. The formation of triiodide presumably involves the adsorption of iodide onto the MnO_2 surface (perhaps displacing a surface hydroxyl group). The

iodide should be subsequently oxidized and released back into solution as IOH or I₂, which rapidly forms I₃⁻. The kinetic data has been modeled as a first order process. First order rate constants have been obtained for the formation of iodine in the presence of MnO₂. The increase in iodide oxidation rates with MnO₂ concentration is evident in the data. The reaction rate increases with iodide concentration although the dependence is not first order (an order of 1.4 appear to fit the data). The oxidation rate also increases with temperature and has a apparent activation energy of 16.2 kJ/mol.

The total yield of iodine from these materials was compared with the resulting Mn (II) concentration to estimate the oxygen to manganese ratio (MnO_x) for the starting material. This ratio (x) is a function of the average oxidation state of the material and is given by:

$$x = 1 + \frac{I_3^-}{Mn^{+2}}$$

The rate of iodide oxidation can be seen to vary significantly for the various preparations.

It is clear that some manganese oxide can oxidize iodide to iodine under mild pH and temperature conditions. By comparison, laboratory grade MnO₂ reacted sluggishly under these conditions. Because of the wide distribution of this mineral in nature, it is believed that the oxidation of iodide by manganese oxide may result in the formation of organic iodine bonds in sedimentary and soil organic matter.

TASK 15 PROFILE

Start Date: August 2002

Completion Date: October 2007

Theses Generated:

Gregory Tye Schmett, M.S., Department of Chemistry, "Immobilization of Fission Iodine by Reaction with Fullerene Containing Carbon Compounds or Insoluble Natural Organic Matter," August 2005.

James W. Dorman, M.S., Department of Chemistry, "Analysis of Iodide and Iodate in Soil and Water," December 2007.

Ginger Marie Kimble, M.S., Department of Chemistry, "Reactions of Iodine and Iodate with Sphagnum Peat: An Examination of Kinetics and pH Dependence," August 2008.

Journal Articles:

M.A. Cheney, N.R. Birkner, L. Ma, T. Hartmann, P.K. Bhowmik, V.F. Hodge, and S.M. Steinberg, "Synthesis and characterization of inor-

ganic double helices of cryptomelane nanomaterials," *Colloids and Surfaces A: Physicochemical Engineering Aspects* 289, pp 185–192, 2006.

S.M. Steinberg, B. Buck, J. Morton, and J. Dorman, "The speciation of iodine in the salt impacted Black Butte Soil Series along the Virgin River, Nevada, USA" Accepted for publication with minor revision.

Conference Proceedings:

G.T. Schmett, G.M. Kimble, S.M. Steinberg, D.W. Emerson and G.S. Cerefice. "Immobilization of Fission Iodine by Reaction with Insoluble Natural Organic Matter" Proceedings of the Actinide and Fission Product Partitioning & Transmutation, 8th Information Exchange Meeting, OCED/NEA: Las Vegas, Nevada. November 9-11, 2004.

S.M. Steinberg, G. Kimble, G.T. Schmett, and D.W. Emerson. "Abiotic Reaction of Iodine and Iodate with Sphagnum Peat," abstract and poster, American Chemical Society, Division of Environmental Chemistry, Vol 4, No 1, Spring 2005.

S.M. Steinberg, G.M. Kimble, G.T. Schmett, and D.W. Emerson, "Abiotic Reaction of Iodate and Iodine with Natural Organic Matter," American Chemical Society, 40th Western Regional Meeting, Anaheim, CA, January 22-25, 2006.

S.M. Steinberg, G.M. Kimble, G.T. Schmett, D.W. Emerson, M.F. Turner, and M. Rudin, "Immobilization of Fission Iodine by Reaction with Insoluble Natural Organic Matter," MARC VII, Honolulu, HI, April 3-7, 2006.

S.M. Steinberg, G.M. Kimble, G.T. Schmett, and D.W. Emerson, "Abiotic Reaction of Iodate with Sphagnum Peat and Other Natural Organic Matter," MARC VII, Honolulu, HI, April 3-7, 2006.

S.M. Steinberg, B. Buck, J. Morton, and J. Dorman, "Speciation of iodine in the salt-impacted Black Butte soil series along the Virgin River, NV," 234th American Chemical Society National Meeting, Boston, MA, August 19-23, 2007.

S.M. Steinberg and N.R. Birkner, "Formation of organic iodine in soil and sediment by reaction of manganese oxide with iodide," 234th American Chemical Society National Meeting, Boston, MA, August 19-23, 2007.

| Mineral | O/Mn Ratio | K(sec ⁻¹) |
|---------------------------------|------------|-----------------------|
| Cryptomelane Type II | 1.39 | 0.0328 |
| Sodium Manganese Oxide Hydrate | 1.87 | 0.0149 |
| Black Birnessite + Cryptomelane | 1.74 | 0.0137 |
| Cryptomelane Type II | 2.2 | 7.97 E-03 |
| Cryptomelane Type II | 2.47 | 1.49 E-03 |
| Cryptomelane Type II | 1.92 | 3.27 E-03 |
| Manganes Oxide Hydrate | 1.83 | 1.27 E-02 |

Rate constants for iodide oxidation (at pH 4 and solid concentration of 1 mg/mL) are tabulated along with the measured oxygen to manganese ratio of the manganese oxide. The mineral was identified by powder x-ray analysis.

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Task 16

Evaluation of Fluorapatite as a Waste-Form Material

D.W. Lindle, O. Hemmers and D.L. Perry

BACKGROUND

High Temperature Gas-cooled Reactor (HTGR) systems are being designed and evaluated as part of the Advanced Fuel Cycle Initiative. HTGR designs use a TRISO-coated fuel (a silicon-carbide and pyrocarbon composite coating) to provide much of the passive containment for radionuclides.

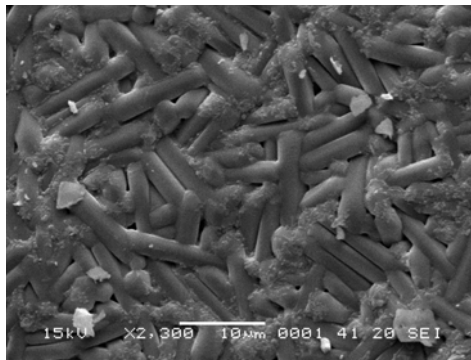
Argonne National Laboratory has proposed a new extraction procedure to handle TRISO-coated fuels, the Fluoride Extraction Process (FLEX). The FLEX process is designed to separate the uranium in the fuel from the actinides and most fission products by taking advantage of the unique properties of uranium hexafluoride (UF_6). In the FLEX process, the used TRISO fuel is reacted with zirconium fluoride salt, forming UF_6 and the fluoride salts of the actinides and fission products. At process temperatures, the UF_6 volatilizes into a gas, and is released from the molten salt mixture. This leaves behind the actinides and most fission products in a fluoride salt, which is subsequently processed using pyrochemical techniques to recover the actinides and other long-lived fission products for transmutation. The UF_6 is then cooled, causing it to sublime into solid form, which is then further processed for disposal or reuse.

The primary waste stream from the FLEX process is the fission products from the fuel, which are in a zirconium fluoride salt at the end of the process. Due to the fluorine in this waste stream, the fluoride salts are unsuitable for conversion into the traditional borosilicate waste glass. Therefore, without a suitable disposal form, this process can not be deployed.

RESEARCH OBJECTIVES AND METHODS

The following were the specific research objectives:

- To develop a waste matrix for the disposal of the fission product waste stream from the FLEX process;
- To develop a process to make synthetic fluorapatite that incorporates the FP-bearing ZrF_4 salt;
- To develop a fundamental understanding of the chemistry of this new waste form in order to better predict its long term behavior in a repository environment; and,



SEM micrograph of Ca_4SrFap (annealed sample).

- To develop a fundamental understanding of natural, fluoride-bearing mineral phases to use as natural analogs to bound the predicted behavior of the FLEX fission product waste stream.

The research effort was divided along two parallel paths: the *Fabrication Path*, led by collaborators at the Khlopin Radium Institute (KRI) in St. Petersburg, Russia; and the *Characterization Path*, led by researchers from UNLV.

The Fabrication Path focused on examining and evaluating various techniques for fabricating synthetic fluorapatite; synthesizing synthetic fluorapatite; and examining the impacts of waste loading and other fabrication process factors on the performance of the synthetic fluorapatite as a potential waste form.

The Characterization Path focused on adapting and refining the X-ray spectroscopy techniques currently used to characterize borosilicate glass for use in examining the fluorapatite system. This path also encompassed the examination of the ceramic and synthetic mineral waste forms created at KRI, with subsequent examination of these techniques to develop a molecular-level understanding of natural fluorapatite and other fluorine-bearing natural phases as natural analogs for the waste form. These techniques were also be used to examine the changes in surface chemistry caused by environmental degradation of these materials.

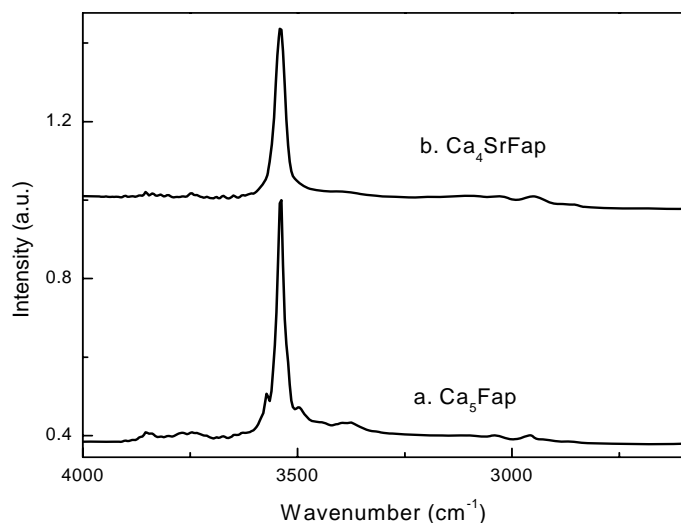
RESEARCH ACCOMPLISHMENTS

Microscopy Techniques:

- The microstructure of as-synthesized fluorapatite samples were characterized by transmission electron microscopy (TEM). The phase purity and crystallographic structure of these samples were first verified by X-ray powder diffraction with the help of Rietveld analysis.
- Different methods (solution-drop and microtome cutting) were used to make the samples for TEM analysis. The solution-drop method was suitable to analyze the crystallographic structure of the fluorapatite by high resolution TEM. Furthermore, the solution-drop method was successfully used to study the particle morphology of fluorapatite.
- The microtome cutting method allowed studying both particle shape and cross-sectional morphology of fluorapatite crystals. Microtome cutting, however, introduced periodical fringe artifacts in high resolution TEM imaging due to deformation effects on the material. Therefore, in order to completely characterize the microstructure and morphology of fluorapatite-type particles a combination of both sample preparation techniques was necessary.

Calcium Substitution by Zinc:

- Substitution of calcium in fluorapatite by zinc is possible and was demonstrated to be concentration dependent. The phase purity of fluorapatite in the samples decreases with the increase in zinc substitution.
- A 25 mol % of zinc substitution leads to a single-phased fluorapatite, but the formation of a secondary phase was



Infrared spectra of Ca_5Fap and Ca_4SrFap after annealing.

identified when the zinc level increased to 50 mol %. A fluorapatite phase could not be identified in samples with 75 mol % and 100 mol % zinc substitution.

- The two samples synthesized using 25 mol % and 50 mol % of zinc showed some similar morphological and microstructural properties to that of phase-pure fluorapatite. However, the closest similarities to pure fluorapatite were identified in the 25 mol % sample.
- The 50 mol % sample shows a lower thermal stability that decreases with increasing zinc level. The low thermal stability of the 50 mol % sample is most likely due to the identified secondary chemical phase.
- The sintering properties of the 25 mol % sample are similar to pure fluorapatite. However, complete decomposition of the zinc-incorporated fluorapatite chemical phase in the calcined 25 mol % sample results in lower thermal stability than for phase-pure fluorapatite.

Calcium Substitution by Strontium:

- The synthesis of foreign cation-incorporated fluorapatite, in this case strontium ions, was achieved successfully. XRD powder patterns of fluorapatite and Sr-fluorapatite showed identical peaks except for slight peak shifts and peak merging.
- Increasing strontium content showed a linear relationship with unit cell dimensions, the relationship of which can allow one to measure unknown percentages of strontium in fluorapatite.
- It has been shown that Sr occupies only Ca sites and not P or O sites.
- Scanning electron microscope images of strontium-incorporated fluorapatite showed the formation of hexagonal crystals on the surfaces of most samples. Formation of

fluorapatite crystals has not been observed in the same samples before annealing at 1200 °C.

- Re-synthesis of apatite using natural fluorapatite as a starting material was achieved. The final product contained fluorapatite, chlorapatite and hydroxyapatite phases and it is inconclusive as to which was the major component.

TASK 16 PROFILE

Start Date: August 2002

Completion Date: August 2005

Theses Generated:

G.W. Chinthaka Silva, M.S., Chemistry, "Evaluation of Apatites as a Matrix for the Waste Stream of the Flex Process," June 2005.

Chirantha P. Rodrigo, M.S., Chemistry, "Synthesis and Characterization of Strontium Fluorapatite," August 2005.

Journal Articles:

G.W.C. Silva, L. Ma, O.A. Hemmers, and D.W. Lindle, "Micro-Structural characterization of precipitation-synthesized fluorapatite nano-material by transmission electron microscopy using different sample preparation techniques," *Micron*, 39/3, 269-274, 2007.

G. W. Chinthaka Silva, Oliver Hemmers, Ken R. Czerwinski, and Dennis W. Lindle, "Investigation of microstructure and thermal behavior of zinc-substituted fluorapatite" submitted to *Journal of Inorganic Chemistry*, 2008.

Conference Proceedings:

C.P. Rodrigo, G.W.C. Silva, O. Hemmers, D.L. Perry, and D.W. Lindle, "Evaluation of Fluorapatite as a Waste Form," ANS Student Conference, UW at Madison, WI, April 1-3, 2004.

G.W.C. Silva, D.L. Perry, A.L. Johnson, O. Hemmers, and D.W. Lindle, "Characterization of the thermal stability of apatites containing different cations," ANS Student Conference, University of Wisconsin at Madison, WI, April 1-3, 2004.

G.W.C. Silva, D.L. Perry, A.L. Johnson, O. Hemmers, and D.W. Lindle, "Characterization of the thermal stability of apatites containing different cations," Graduate and Professional Student Research Forum, UNLV, Las Vegas, NV, April 17, 2004.

G.W.C. Silva, O.A. Hemmers, and D.W. Lindle, "Characterization of the Thermal Stability of Zinc-containing Fluorapatite," ANS Student Conference, Columbus, OH, April 2005.

C.P. Rodrigo, O.A. Hemmers, and D.W. Lindle, "Characterization of Fluorapatite as a Waste Form," ANS Student Conference, Columbus, OH, April 2005.

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Task 17

Interaction between Metal Fission Products and TRISO Coating Materials

C. Heske

BACKGROUND

This project focuses on the chemical bonding and interface formation of metal fission products with the coating materials used in tri-isotropic (TRISO) fuel particles for gas-cooled reactors. By combining surface- and bulk-sensitive spectroscopic and microscopic methods, intermediate chemical phases at the interface, intermixing/diffusion behavior, and the electronic interface structure for different coating materials and metals are examined.

In detail, the project studies the interface formation of Pd, Cs, and Ag with SiC and pyrolytic carbon. Using SiC single crystals and highly-ordered pyrolytic carbon (HOPG) as substrates, interfaces are prepared under controlled conditions in an ultra-high vacuum environment and are studied with a combination of experimental methods, including Photoelectron Spectroscopy, Auger Electron Spectroscopy, X-Ray Emission Spectroscopy, and X-Ray Absorption Spectroscopy. Furthermore, microscopic techniques (in particular Atomic Force Microscopy) are being employed.

RESEARCH OBJECTIVES AND METHODS

By combining results for the electronic and chemical structure of the fission product/TRISO layer interfaces, the research results give information about failure mechanisms of TRISO particles and fission product transport.

Furthermore, they can be used to derive strategies to tailor the interface properties for an optimization of TRISO particles in terms of, e.g., chemical and long-term stability.

The experiments use two different experimental set-ups. First, a multi-chamber ultra-high vacuum system at UNLV houses the majority of surface-sensitive methods. Second, the soft X-ray fluorescence (SXF) endstation at Beamline 8.0 at the Advanced Light Source, Lawrence Berkeley National Laboratory is used to perform X-ray absorption and emission spectroscopy. The apparatus at UNLV consists of two different surface spectroscopy chambers, a preparation chamber, and an ultra-high vacuum (UHV) scanning probe microscope. Samples can be introduced into the machine through a glove box with integrated load-lock and then transferred to each of the chambers via a distribution chamber. The analysis chambers are each

equipped with a state-of-the-art electron analyzer, an X-ray source, an ultraviolet (UV) source, and an inverse photoemission setup consisting of a low-energy high-flux electron gun and a UV detector. The preparation chamber is used for cleaning samples with an ion source and for the deposition of metal films with an electron-beam evaporator (Pd, Ag) or a dispenser element evaporator (Cs).

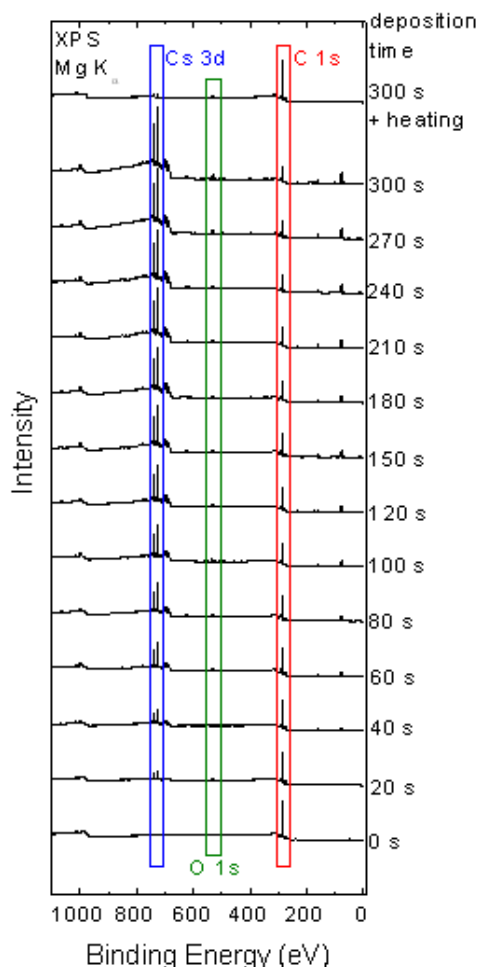
RESEARCH ACCOMPLISHMENTS

In the past year, emphasis was placed on a detailed analysis and description of the Cs/SiC and Cs/HOPG interface formation processes. Here, some of the results for Cs/HOPG will be briefly described.

A series with Cs films of different thickness on HOPG were prepared. UV photoemission (UPS), x-ray photoemission (XPS), and in-vacuum atomic force microscopy (AFM) experiments were performed for each Cs film thickness. HOPG substrates were prepared by cleaving under nitrogen in the glove box directly connected to the surface analysis system at UNLV and then directly introduced into the UHV apparatus without any air exposure.

In the figure to the left, the XPS survey spectra for different Cs evaporation times (and hence different Cs film thicknesses) are shown. The cleaving of the sample under nitrogen clearly results in a very clean and nearly adsorbate-free surface, as can be seen from the weak O 1s line in the bottom-most spectrum. After Cs evaporation, all expected Cs lines can be found in the spectra. The intensity of the lines, which can be related to the film thickness, increases for increasing deposition time, as expected. In parallel, the lines of the HOPG substrate (i.e., the C 1s photoemission line and the C KVV Auger line) decrease in intensity due to the attenuation by the covering Cs layer.

Focus was also placed on understanding the HOPG surface morphology changes when annealing the Cs/HOPG interface at 600 °C. A series of stepwise Cs depositions was performed on a cleaved HOPG surface at room temperature under ultra-high vacuum conditions and monitored with photoelectron spectroscopy. After 300 seconds of Cs deposition, the surface was imaged by UHV-AFM. The AFM image clearly shows Cs atoms forming clusters



XPS survey spectra of an HOPG substrate, as well as after stepwise deposition of Cs and subsequent heating under inert conditions.

on the HOPG surface, which is corroborated by a detailed intensity analysis of X-ray photoelectron spectroscopy (XPS) data. Next, the sample was removed from UHV, annealed in a furnace in air (600 °C) for 60 minutes, and reintroduced into UHV. The center image (below figure) shows the AFM image after annealing. As again corroborated by the XPS data, most of the Cs atoms/clusters are removed from the surface. Furthermore, a significant formation of craters and voids were found that can act as diffusion pathways for various metallic species (e.g., for fission products diffusing through pyrolytic carbon layers in TRISO fuel). To verify whether the crater formation is induced by the presence of Cs clusters on the surface, a second HOPG sample was prepared that was cleaved, characterized, and annealed in the exact same fashion as the first sample (but not exposed to Cs). The right image (see figure below) shows a corresponding AFM image of the HOPG surface after annealing. Similar cracks are observed as in the Cs/HOPG samples, but no evidence of crater formation could be found (in any of the AFM images). This supports the conclusions (a) that Cs grows in a cluster-like fashion on HOPG (at room temperature), and (b) that the presence of Cs clusters on the surface of pyrolytic graphite can lead to the formation of crater- and crack-like diffusion pathways during annealing at 600 °C, while the annealing of pyrolytic graphite only leads to the formation of cracks, but not craters.

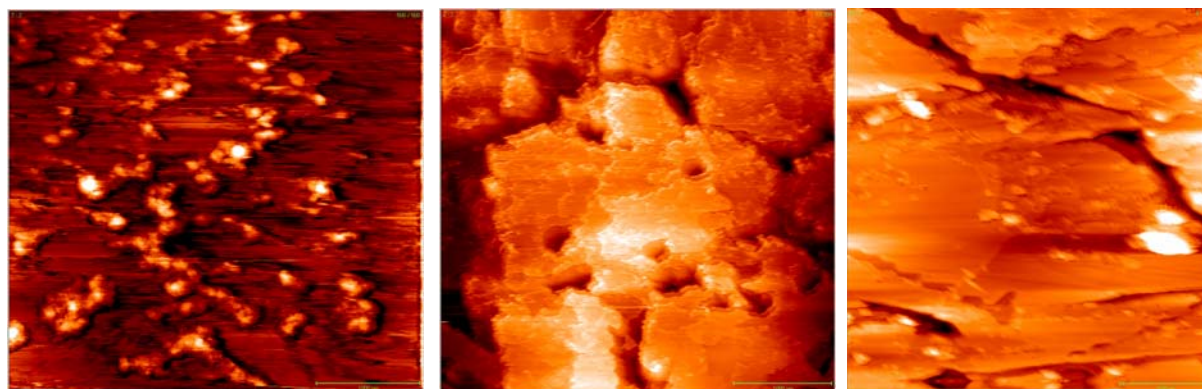
FUTURE WORK

The experiments on Cs/SiC and Cs/HOPG will be completed, in particular by varying the substrate temperature during deposition (up to approximately 1200 °C). These experiments will allow the study of both the temperature dependence as well as the influence of air (in particular oxygen) on the high-temperature behavior and the crater- and crack-formation results of the present study. In

ACADEMIC YEAR HIGHLIGHTS

- ◆ Participation of graduate and undergraduate students in experimental campaigns at the Advanced Light Source, Lawrence Berkeley National Laboratory, Berkeley, CA (September 2007, January 2008, May 2008).
- ◆ Sharath Sudarshanam passed his M.S. prospectus presentation meeting in March 2008.
- ◆ C. Heske, “Using XES and RIXS to investigate the chemical and electronic structure of materials for energy conversion,” RIXS 08 Workshop, Uppsala, Sweden, June 13-14, 2008.
- ◆ C. Heske, “Soft X-rays and the electronic structure of buried things – interfaces, liquids, and below dirty surfaces”, Materials Science Institute, University of Oregon, Eugene, February 26, 2008.
- ◆ C. Heske, “Investigating the electronic and chemical properties of surfaces, interfaces, and other buried things with soft X-ray spectroscopy,” Chemical Engineering Department, University of California at Santa Barbara, October 25, 2007.
- ◆ C. Heske, “Surface and Interface Science on Devices for Energy Conversion,” X-ray and Neutron Scattering Contractor’s Meeting, Division of Materials Sciences and Engineering (DMS&E), DOE Office of Basic Energy Sciences (BES), Warrenton, VA, October 18, 2007.
- ◆ C. Heske, “Investigating the electronic and chemical properties of surfaces, interfaces, and other buried things with soft X-ray spectroscopy,” Cornell University, Materials Science and Engineering Department Seminar, October 11, 2007.

addition, the experiments will simulate the “natural” high temperature environment present in the nuclear reactor. Furthermore, it is planned to initiate first interface studies involving ZrC layers.



Left: AFM picture of a highly ordered pyrolytic graphite (HOPG) substrate after deposition of Cs for 300 seconds. Center: AFM picture Cs/HOPG substrate after annealing at 600 °C for 60 minutes in a furnace. Right: AFM picture of HOPG without cesium deposition, annealed at 600 °C for 60 minutes in a furnace. All image scales are 4 x 4 μm².

Research Staff

Clemens Heske, Principal Investigator, Associate Professor, Department of Chemistry
 Marcus Bar, Associate Research Professor, Department of Chemistry
 Lothar Winhardt, Post-doctoral Fellow, Department of Chemistry

Students

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 Alessio Luinetti, Sunil Subramanyam, and Kyle George, Undergraduate Students, Department of Chemistry

Collaborators

David Williams, Oak Ridge National Laboratory

Task 18

Fundamental and Applied Experimental Investigations of Corrosion of Steel by LBE under Controlled Conditions: Kinetics, Chemistry Morphology, and Surface Preparation

J.W. Farley and A.L. Johnson

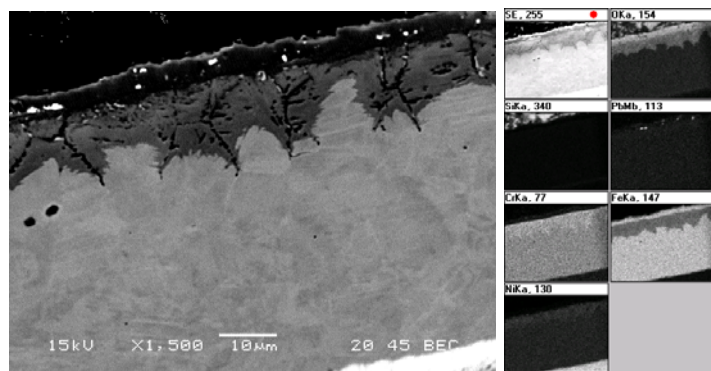
BACKGROUND

Advanced nuclear processes and facilities (e.g., transmutation of nuclear waste, fast reactors, and spallation neutron sources) impose special demands on materials, which must withstand high temperatures, high radiation fields, and chemical corrosion. Proposed schemes for transmuting nuclear waste require a non-moderating coolant such as lead-bismuth eutectic (LBE) or liquid sodium. While LBE corrodes most steels, small amounts of oxygen in the LBE greatly reduces the corrosion rate, and could ideally re-grow a damaged oxide layer in situ. The protective oxide layer would thus be self-healing. However, a fundamental understanding of the role of oxygen and passivating oxide layers is presently incomplete.

RESEARCH OBJECTIVES AND METHODS

The overall goal is to obtain scientific insight into the fundamental processes involved in the corrosion of steel by LBE. The experimental methods include characterization of steel samples before and after exposure to LBE using different techniques including: Scanning Electron Microscopy (SEM), Energy Dispersive X-ray analysis (EDX), Wavelength Dispersed X-ray (WDX), X-ray Photoelectron Spectrometry (XPS), Sputter Depth Profiling (SDP), X-ray Diffraction (XRD) and laser microRaman spectroscopy. These techniques have been previously used by this research group to probe the effect of surface preparation on corrosion-resistance of samples with the same chemical composition, and to examine the role of silicon in silicon-containing steel.

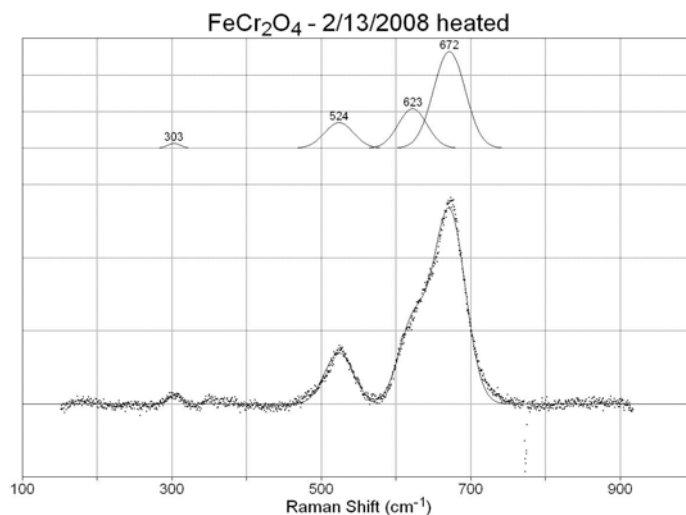
During the past year, gas-phase experiments were conducted in which steel samples were oxidized in glass capsules at elevated



Back Scattered Image

Elemental Map

Transverse section of D9, a 316 class stainless steel. Note the large anisotropy in the growth of the thick inner oxide layer, showing obvious pathways for migration of oxidation reactants affecting the growth rate and morphology of the oxide layer.



Raman Spectrum of FeCr_2O_4

temperatures in a tube furnace. Corroded steel samples were analyzed from a variety of sources, including the Delta loop at LANL and samples corroded at UNLV in the gas phase experiments.

RESEARCH ACCOMPLISHMENTS

Gas-phase experimental results

Measurements have been made of the gas-phase oxidation of steel, using the tube furnace in the High Temperature Materials Exposure Facility in the UNLV Chemistry building. The goal is to clarify the differences between gas phase corrosion and corrosion by LBE. Oxidation of metal surfaces is carried out by the residual gas, which was characterized using a residual gas analyzer. The residual gas was determined to consist of much water vapor but little oxygen or hydrogen. The role of water as an oxygen carrier may be an underestimated factor in corrosion. These experiments are part of the doctoral dissertation of Thao Ho.

In other experiments, investigations were performed to determine whether alloys undergoing sputtering are chemically altered by the sputtering process. Such an artifact would reveal itself as a change from stoichiometric to non-stoichiometric composition during the sputtering process. Chromia (Cr_2O_3) was found to be affected minimally if at all, but hematite (Fe_3O_4) was significantly affected.

Facility development

Progress continued on the construction of the apparatus for the Liquid Metal Corrosion Experiment, which will be able to expose steel samples to either LBE or sodium.

The Oxygen Control System (OCS) from Germany's Karlsruhe Lead Laboratory was used for initial experiments, and very low oxygen concentrations were recorded.

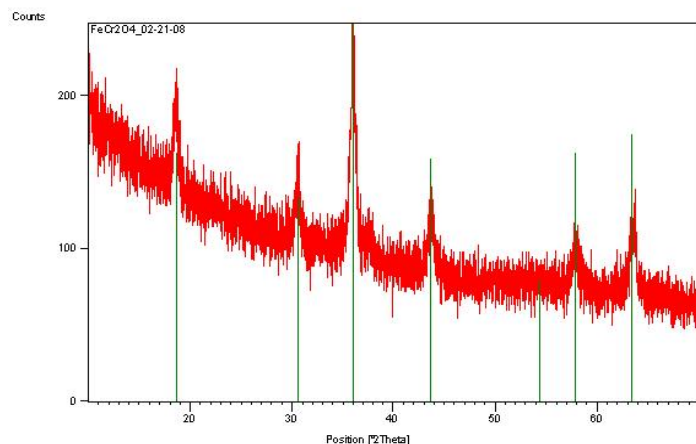
Characterization of samples

Oxide layers were studied using high-resolution XPS with sputter depth profiling, revealing the extent of oxidation as a function of position within the oxide layer.

A new capability in microRaman spectroscopy was built by physics graduate student Brian Hosterman in order to examine the Raman spectra of compounds relevant to corrosion. Raman studies can distinguish between different chemical species by their characteristic vibrational spectra. For example, Fe_2O_3 can be distinguished from Fe_3O_4 by its Raman spectrum. The Raman microscope gives a lateral spatial resolution of the order of a few microns.

Synthesis of standards

Chemical characterization of the species involved in corrosion can be revealed by microRaman studies, in which the spectrum of an unknown compound is matched to a library of spectra of standards. Standards have been purchased when possible, but some standards have been synthesized in the laboratory. Spinel (compounds with formula XY_2O_4) are of special interest because they are believed to form part of the metal oxides under investigation. Spectra of standards have been measured, and are in good



XRD Spectrum of FeCr_2O_4

ACADEMIC YEAR HIGHLIGHTS

- ◆ Dan Koury, doctoral dissertation, "Characterization of the Mechanism of Duplex Oxide Growth on Austenitic Stainless Steels 316 and D9 in Oxygen-Controlled Lead-Bismuth Eutectic (LBE)," August 2008 (expected).
- ◆ Brian Hosterman, doctoral dissertation, "Studies of corrosion of steel by laser microRaman spectroscopy," August 2009 (expected).
- ◆ Thao Ho, doctoral dissertation, "XPS studies of Si in Fe/Si alloy steel in corrosion of steel by LBE," August 2009 (expected).
- ◆ A.L. Johnson, D. Koury, J. Welch, T. Ho, S. Sidle, C. Harland, B. Hosterman, U. Younas, L. Ma, and J.W. Farley. "Spectroscopic and microscopic investigation of the corrosion of D-9 stainless steel by lead-bismuth eutectic (LBE) at elevated temperatures. Initiation of thick oxide formation," *Journal of Nuclear Materials* (in press, 2008).
- ◆ A. Johnson, "Heavy Liquid Metal efforts at UNLV, USA: LBE Corrosion of D9 and other 316-group Steels," presented at the IV Workshop on Materials for Heavy Liquid Metal Cooled Reactors and Related Technologies, Rome, Italy, May 21-23, 2007.

agreement with literature values. The crystal structure of the oxide layers can be determined by X-ray diffraction. The XRD spectrum of FeCr_2O_4 is illustrated (below, right). The lattice spacings derived from analysis of this spectrum agree with literature values.

Outcomes

A model of the LBE corrosion process is emerging, incorporating theoretical and experimental results from UNLV laboratory results and elsewhere. Several of the presentations at the May 2007 Heavy Liquid Metals Workshop in Rome, Italy were based on the new insights. UNLV has contributed the observation that localized failure in the initial oxide layer leads to formation of duplex/complex oxide structures. Scientific collaboration with Los Alamos National Laboratory has continued. Steel samples exposed to LBE at LANL have been analyzed by the UNLV group.

FUTURE WORK

Future work includes expansion of this program to studies covering the effect of liquid sodium on structural materials.

Faculty

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John Farley, Professor, Department of Physics

Students

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Dan Koury and Brian Hosterman, Graduate Students, Department of Physics and Astronomy
Jenny Welch and Tim Lane, Undergraduate Students, Department of Physics and Astronomy
Noah Kapley (Undergraduate; Western Kentucky University) and Zara Fewsmith (Local high school volunteer), Summer 2007 Students

Collaborators

Ning Li, LBE Project Leader, Los Alamos National Laboratory
Peter Hosemann, Visiting Scientist, Los Alamos National Laboratory

Task 19

Dissolution, Reactor, and Environmental Behavior of ZrO₂-MgO Inert Fuel Matrix

K. Czerwinski

BACKGROUND

There has been a recent resurgence of interest in different oxide fuel types (e.g., Th, inert matrix, and Pu fuels) as potential advanced fuels that can be operated to relatively high burnups at lower costs than current UO₂ fuels. These fuels can also be formed to incorporate transuranics elements in the matrix. Inert fuel matrices have the advantage of burning Pu and other transuranic elements from the fuel cycle without the production of other actinide elements. Of the possible materials for use in an inert matrix, ZrO₂ has been examined. The inclusion of ZrO₂ is expected to increase chemical stability and radiation resistance. The natural analogue of zirconia, baddeleyite ((Zr,M)O₂), where M is a tetravalent ion such as hafnium), contains up to 3000 parts per million U or Th. This supports the durability of inert matrix fuels using ZrO₂ in reactor conditions and repository conditions. However, fuels appropriate for the advanced fuel cycle applications should have desirable reprocessing properties, namely ease of dissolution for separations. An additional oxide which is somewhat soluble may need to be added to the ZrO₂ matrix to achieve desirable reprocessing properties. A candidate oxide is MgO.

RESEARCH OBJECTIVES AND METHODS

This project will examine inert fuels containing ZrO₂ and MgO as the inert matrix. Ceramics with this inert matrix, Ce, U and eventually Pu will be synthesized and examined. While the Advanced Fuel Cycle Initiative focus is on inert fuels with Pu as the fissile component, this task will perform initial laboratory experiments with Ce and U. The initial work with Ce will be performed early in the project with results used as a basis for U studies. Reactor physics calculations will be used to examine suitable quantities of burnable poisons from the candidate elements Gd, Er, or Hf. Most fuels use Gd or Er, but the chemical properties of Hf lend themselves to formation of solid solutions with Zr and the tetravalent actinides and will therefore be investigated. This project will provide the necessary data for evaluating the performance, reprocessing, and waste behavior of the MgO-ZrO₂ fuels from a quantified, chemical perspective.

Reactor physics calculations are used to examine suitable quantities of burnable poisons from the candidate elements Gd, Er, or Hf with reactor grade Pu providing the fissile component, with up to

Metal Concentrations for Uranium Containing Ceramics.

| Sample # | Zr % | Mg % | U % | Er % |
|----------|------|------|-----|------|
| 1 | 92.5 | 0 | 5 | 2.5 |
| 2 | 87.5 | 5 | 5 | 2.5 |
| 3 | 82.5 | 10 | 5 | 2.5 |
| 4 | 77.5 | 15 | 5 | 2.5 |
| 5 | 72.5 | 20 | 5 | 2.5 |
| 6 | 62.5 | 30 | 5 | 2.5 |
| 7 | 47.5 | 45 | 5 | 2.5 |
| 8 | 32.5 | 60 | 5 | 2.5 |
| 9 | 17.5 | 75 | 5 | 2.5 |
| 10 | 0 | 92.5 | 5 | 2.5 |

10% of ²³⁹Pu. Ceramics are synthesized and characterized based on the reactor physics results. The solubility of the fuel ceramics, in reactor conditions, reprocessing conditions, and repository conditions, are investigated in a manner to provide thermodynamic data necessary for modeling.

The research objectives of this project are as follows:

- To examine the neutronic behavior of MgO-ZrO₂ inert fuels. Variation of MgO and ZrO₂ composition ranges from 30% to 70% MgO in ZrO₂. Analysis of Gd, Er, and Hf for reactivity control ranging from 5-10% lanthanides. Analysis of reactor grade Pu as fissile component ranging from 5-10% Pu. Results will be used as parameters for fuel composition.
- To synthesize and characterize MgO-ZrO₂ ceramics containing burnable poison and fissile composition. Synthesis is based on a precipitation method. Range of MgO in ZrO₂, fissile component concentration, and burnable poison concentration based on results of neutronic calculations. Characterization of ceramics will include density, X-ray diffraction (XRD), surface area analysis, X-ray absorption fine structure, and chemical composition. Results will be applied to behavior in high temperature water, acid, and environmental conditions.
- To describe the chemical behavior of synthesized ceramics. Chemical thermodynamic and kinetic analysis will use equilibrium data, kinetic data, and surface area normalized dissolution. Different conditions will include reactor conditions (high temperature and high pressure water) and reprocessing conditions (nitric acid and elevated temperature). Environmental conditions will be near neutral solution conditions.
- To utilize project data in kinetic and thermodynamic modeling codes to evaluate the speciation of the elements in the ceramics under reactor, reprocessing, and repository conditions.

RESEARCH ACCOMPLISHMENTS

Development of X-ray fluorescence characterization method

A reliable method for X-ray fluorescence (XRF) was developed involving ashing the individual oxides, then preparing standards through the dry synthesis route. It was believed that the samples that showed heterogeneous Ce distribution were due to insufficient sintering times. Therefore, one such sample was removed from resin, resintered, and elemental mapping was performed a second time. This second mapping showed a homogenous distribution of cerium demonstrating the suitability of the method.

Synthesis and characterization of U-containing ceramics

Ceramics were synthesized using MgO-ZrO₂ as the inert matrix and Er₂O₃ as a burnable poison as in previous studies; however, UO₂ is now being used as a more accurate PuO₂ analog than the previous CeO₂ containing ceramics.

A soxhlet experiment was initiated to determine the corrosion resistance of the ceramics. The results demonstrated the corrosion resistance enhancement from the inclusion of ZrO_2 . A thermogravimetry and differential scanning calorimetry scan was performed on the precipitated material to examine calcining and sintering behavior. The material appears to change from the oxy-hydroxide precipitate to the oxide at the same temperature as the Ce-containing ceramics (260°C). The phase change also starts at the same temperature (510°C), but it does not proceed as quickly and therefore shows a broader peak.

XRD patterns were taken of all samples under investigation to qualitatively determine the phases present within the sintered sample. With no MgO present the UO_2 and Er_2O_3 only partially stabilizes the ZrO_2 resulting in a mixture of monoclinic and tetragonal Zr oxide phases. With as little as 5% wt/wt MgO , the material fully stabilizes to form a pure cubic zirconium phase. Over 10% MgO results in a MgO phase, in addition to the cubic zirconia. This pure MgO phase increases with total MgO content. When there is no longer ZrO_2 in the sample a MgO phase dominates, while there is a minor cubic uranium erbium oxide phase. Quantitative analysis will be performed once TOPAZ software is obtained.

Oxide waste forms based on project results

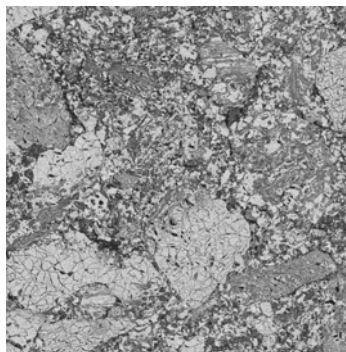
UY_6O_{12} pellets were synthesized for Los Alamos National Laboratory for initial waste form studies complementary to the inert fuel research. Composition was confirmed to be delta phase by XRD.

Other progress

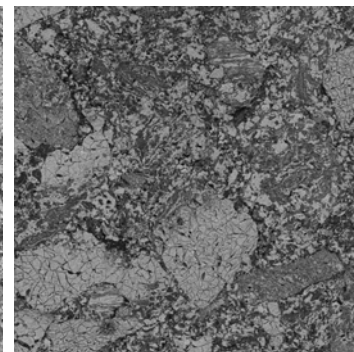
- The Soxhlet has concluded with samples ($\text{Zr}_{0.885}\text{Mg}_{0.05}\text{U}_{0.05}\text{Er}_{0.025}\text{O}_{1.96}$, $\text{Zr}_{0.725}\text{Mg}_{0.2}\text{U}_{0.05}\text{Er}_{0.025}\text{O}_{1.79}$, and $\text{Mg}_{0.925}\text{U}_{0.05}\text{Er}_{0.025}\text{O}_{1.06}$). The data has been analyzed and compared with previous experiments.
- Pure water, silicate water, and brine solutions were used to test fluoride leachability in polytetrafluoroethylene (PTFE) vessels for environmental dissolution studies.
- Environmental dissolution study was conducted with $\text{Zr}_{0.625}\text{Mg}_{0.3}\text{U}_{0.05}\text{Er}_{0.025}\text{O}_{1.6875}$, $\text{Zr}_{0.475}\text{Mg}_{0.45}\text{U}_{0.05}\text{Er}_{0.025}\text{O}_{1.5375}$, and $\text{Zr}_{0.325}\text{Mg}_{0.6}\text{U}_{0.05}\text{Er}_{0.025}\text{O}_{1.3875}$ in water, silicate water, and brine each in triplicate.
- Scanning Electron Microscopy images were taken of corrosion damage from Soxhlet study.
- UY_6O_{12} was synthesized for the LANL group. Delta phase was confirmed by XRD. UY_6O_{12} pellets were characterized by SEM, microprobe, XRD, optical microscopy, and TEM.
- HNO_3 dissolution studies were conducted, residue was analyzed by XRD.

ACADEMIC YEAR HIGHLIGHTS

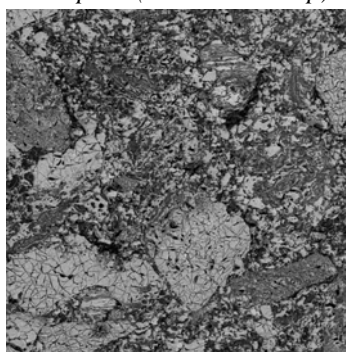
- ♦ K. Czerwinski was the U.S. delegate to the International Atomic Energy Agency Minor Actinide Inert Fuel Matrices working committee from May 2004 to present.
- ♦ K. Holliday, T. Hartmann, and K. Czerwinski, "Characterization and dissolution of Zr-Mg ceramics for inert matrix fuel," Advanced Fuel Cycle Topics, American Chemical Society Regional Meeting, Boise, ID, June 2007.
- ♦ K. Holliday and K. Czerwinski, "Dissolution behavior of actinide containing inert matrix fuel," 236th ACS National Meeting in Philadelphia, PA, August 17-21, 2008.
- ♦ K. Holliday and K. Czerwinski, "Characterization of actinide containing inert matrix fuels," 236th ACS National Meeting in Philadelphia, PA, August 17-21, 2008.
- XAFS data taken at the Advanced Photon Source (ANL) was analyzed to evaluate the local environment of uranium and zirconium in a cubic zirconia matrix.
- Inert Matrix Fuel material was synthesized and analyzed with laser ablation mass spectrometry.



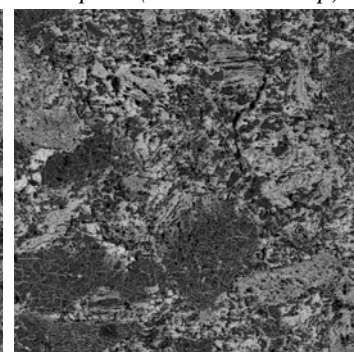
Sample 6 (3 x 3 mm U map)



Sample 6 (3 x 3 mm Er map)



Sample 6 (3 x 3 mm Zr map)



Sample 6 (3 x 3 mm Mg map)

Elemental Scanning by Microprobe.

Research Staff

Ken Czerwinski, Principal Investigator, Associate Professor, Department of Chemistry
Thomas Hartmann, Staff Scientist, Harry Reid Center
Frederic Poineau, Post-Doctoral Researcher, Harry Reid Center

Students

Kiel Holliday, Graduate Student, Department of Chemistry (Radiochemistry Ph.D. Program)

Collaborators

Mitchell Meyer, Idaho National Laboratory

Task 20

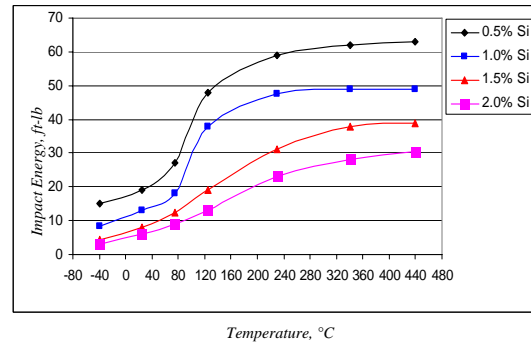
Effect of Silicon Content on the Corrosion Resistance and Radiation-Induced Embrittlement of Materials for Advanced Heavy Liquid Metal Nuclear Systems

A.K. Roy

BACKGROUND

This task is focused on the evaluation of the effects of silicon content on both the corrosion behavior and radiation-induced embrittlement of martensitic stainless steels having compositions similar to that of modified 9Cr-1Mo steel, also known as T91 grade steel. T91 grade steel was selected to be a candidate structural material to contain molten lead-bismuth eutectic (LBE), which can act both as a target material and a coolant during the spallation process in an accelerator-driven system. The operating temperature during this process may range from 420-550 °C. Thus, moderate tensile strength of the containment material (T91) is a major requirement.

The beneficial effects of Si on both the metallurgical and corrosion properties of Cr-Mo steels have previously been demonstrated at UNLV. Therefore, additions of Si ranging from 0.5-2.0 weight percent (wt%) was attempted in this investigation to explore Si effect on both the high temperature tensile properties and corrosion behavior of T91 grade steel. Corrosion studies in the presence of molten LBE could not be performed due to a lack of proper experimental facilities at UNLV. Therefore, detailed corro-



Impact Energy (ft-lb) vs. Temperature (°C)/

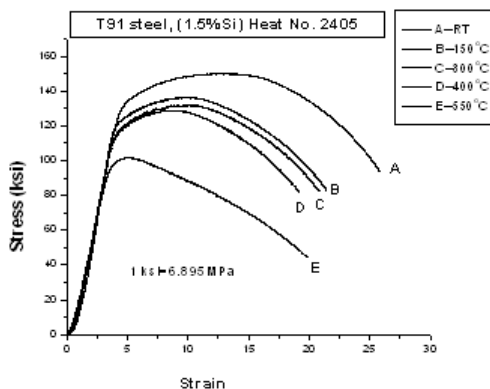
sion studies involving Si-containing T91 grade steels were performed in an aggressive aqueous solution of acidic pH. Further, significant efforts have been made to determine both the impact and fracture toughness of the tested materials as a function of Si content.

RESEARCH OBJECTIVES AND METHODS

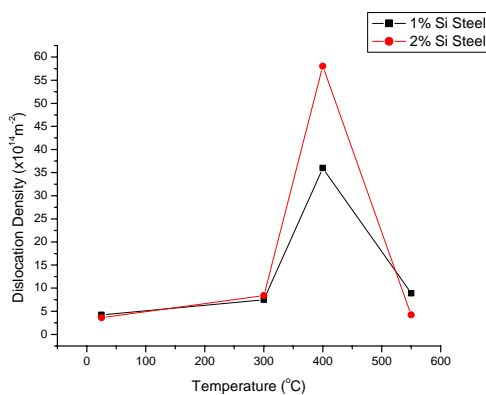
Four heats of T91 grade steel having Si levels of 0.5, 1.0, 1.5 and 2.0 wt% were custom-melted by a vacuum-induction-melting practice. They were subsequently processed and thermally treated to achieve fully-tempered martensitic microstructures. These materials were then machined to fabricate desired types of specimens for evaluation of tensile properties, impact toughness, fracture toughness, resistance to localized corrosion, stress corrosion cracking (SCC), and crack propagation behavior. Limited tensile testing was also performed on T91 grade steels under different levels of radiation. Numerous state-of-the-art experimental techniques were employed to evaluate the desired properties. The relevant experimental techniques have been described in previous annual reports. The significant results obtained during the past year are summarized below.

RESEARCH ACCOMPLISHMENTS

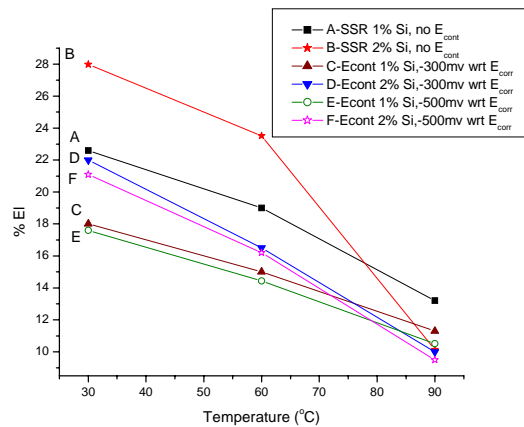
- The results of tensile testing indicate that the magnitude of failure strain (ϵ_f) was gradually reduced with increasing temperature within a susceptible temperature range, irrespective of the Si content. Serrations were noted in the engineering stress vs. strain diagrams.
- The reduced ϵ_f values and the occurrence of serrations within a specific temperature regime can be attributed to a metallurgical phenomenon known as Dynamic Strain Ageing (DSA). Irrespective of the Si content, the susceptibility to DSA was predominant at temperatures ranging from ambient to 400 °C.
- The DSA phenomenon, as seen in this investigation, is commonly associated with the diffusion of solute elements into the matrix of the material at elevated temperatures, thus impeding the movement of dislocations through the matrix and grain boundaries. Such reduction in dislocation mobilities can



Stress-Strain Diagrams of T91 Steel with 1.5 wt% Si.



Dislocation Density vs. Temperature (°C).



Variation of %El with temperature for steels (1 and 2 wt% Si), with and without cathodic E_{cont} .

significantly impair the plastic deformation, which is manifested by reduced e_f values, as seen in this study.

- A maximum dislocation density (ρ), determined from transmission electron micrographs, was seen at 400 °C, irrespective of Si content.
- The work hardening index (n) and activation energy for diffusion are two key parameters influencing the DSA behavior of engineering materials. Simultaneously, both temperature and strain rate can influence the DSA behavior. The variation of n with temperature at a strain rate of $5 \times 10^{-4} \text{ sec}^{-1}$ is shown in the table (opposite page). A gradual increase in the n value was observed at temperatures up to 400 °C, as expected.
- A longer activation time using an electron beam resulted in reduced ductility in terms of e_f .
- The presence of higher Si content in T91 grade steel resulted in reduced impact energy and higher ductile-brittle transition temperature, indicating reduced impact resistance.
- The susceptibility to SCC was enhanced at more cathodic (active) controlled potentials (E_{cont}) and higher testing temperature, implying a synergistic effect of cathodic potential and temperature on SCC.
- The morphology of failure in double-cantilever-beam specimen used in SCC testing was characterized by a combination

Variation of n with Temperature (RT: Room Temperature).

| Heat no. / Temp.(°C) | RT | 150 | 300 | 400 | 550 |
|----------------------|--------|--------|--------|--------|--------|
| 2403 (0.5%Si) | 0.1643 | 0.1698 | 0.1784 | 0.1984 | 0.1432 |
| 2404 (1.0%Si) | 0.1716 | 0.1794 | 0.1945 | 0.2143 | 0.1564 |
| 2405 (1.5%Si) | 0.1586 | 0.1702 | 0.1794 | 0.1802 | 0.1642 |
| 2406 (2.0%Si) | 0.1669 | 0.1709 | 0.1772 | 0.1997 | 0.0994 |

of striations (fatigue), SCC (intergranular/transgranular), and fast fracture (dimples).

TASK 20 PROFILE

Start Date: July 2004

Completion Date: December 2007

Thesis Generated:

Pankaj Kumar, Ph.D. Dissertation, Mechanical Engineering, "Effects of Temperature, Strain Rate and Si Content on Dynamic Strain Ageing of Modified 9Cr-1Mo Steel," December 2007.

Debajyoti Maitra, Ph.D. Dissertation, Mechanical Engineering, "Tensile Deformation and Environmental Degradation of T91 Grade Steels with Different Silicon Content," August 2007.

Sreenivas Kohir, M.S. Thesis, Mechanical Engineering, "Characterization of Martensitic Stainless Steels with High Silicon Content," December 2007.

Harish Krishnamurthy, M.S. Thesis, Mechanical Engineering, "Metallurgical and Corrosion Characterization of a Martensitic Stainless Steel as a Function of Silicon Content," December 2005.

Journal Articles:

A.K. Roy, D. Maitra, and P. Kumar, "The Role of Silicon Content on Environmental Degradations of T91 Steels," Journal of Materials Engineering and Performance, ASM International, in press.

A.K. Roy, P. Kumar, and D. Maitra, "The Effect of Silicon Content on Impact Toughness of T91 Grade Steels," Journal of Materials Engineering and Performance, ASM International, in press.

Conference Proceedings:

S. Kohir, "Tensile Properties and Environmental Degradation of martensitic and Austenitic Stainless Steels," ISRS-2006, Madras, India, December 18-20, 2006.

P. Kumar, D. Maitra, and A. Roy, "Temperature and Silicon Content Effects on Tensile deformation of T91 Grade Steel," Society for the Advancement of Materials and Process Engineering, Fall meeting, Dallas, TX, November 2006.

D. Maitra, P. Kumar, and A. Roy, "The Role of Silicon Content on Tensile and Corrosion Properties of T91 Grade Steels," MS&T, Fall meeting, Cincinnati, OH, October 2006.

D. Maitra, A. Roy, "Environmental Cracking and Localized Corrosion of T91 Grade Steel with Different Silicon Content," AMPT 2006, Las Vegas, NV, July 2006.

P. Kumar, A. Roy, "Tensile Properties of T91 Grade Steel as a Function of Silicon Content," AMPT-2006, Advances in Materials and Processing Technology, Las Vegas, NV, July 2006.

P. Kumar, D. Maitra, and A. Roy, "Metallurgical and Corrosion Studies of Modified T91 Grade Steel," Materials Research Symposium Proceedings, v 929, pp 149-154, 2006.

Research Staff

Ajit K. Roy, Principal Investigator, Professor, Mechanical Engineering Department

Arindam Ghosh, Research Scholar, Mechanical Engineering Department

Students

Debajyoti Maitra and Pakaj Kumar, Ph.D. Graduate Students, Mechanical Engineering Department

Srinivas Kohir, Harish Krishnamurthy and Vinay Virupaksha, M.S. Graduate Students, Mechanical Engineering Dept.

Collaborators

Stuart A. Maloy, Ph.D., AFCEI Materials Team Leader, Los Alamos National Laboratory

Ning Li, Ph.D., LBE Technology Development, AFCEI, Los Alamos National Laboratory

Doug Wells, Ph.D., Associate Professor and Chair, Department of Physics, Idaho State University

Task 21

Theoretical Modeling of Protective Oxide Layer Growth in Non-isothermal Lead Alloy Coolant Systems

Y. Chen, T. Tan, J. Zhang, and J. Li

BACKGROUND

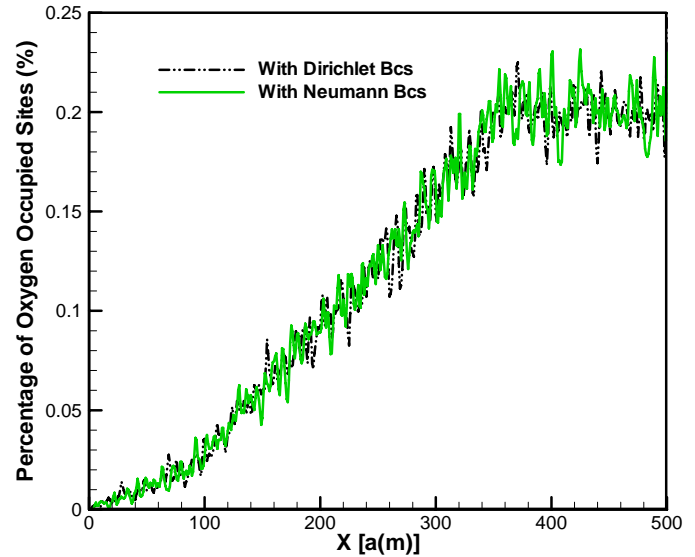
In advanced nuclear energy systems, lead alloys emerge as strong candidates for transmutation and advanced reactor systems as nuclear coolants and spallation neutron targets. However, it is widely recognized that corrosion of materials caused by lead alloys presents a critical barrier to their industrial use. A few experimental research and development projects have been set up by different groups such as at Los Alamos National Laboratory to study the corrosion phenomena in their test facilities and to develop mitigation techniques and materials. One of the central or main techniques under development is to use active control of oxygen thermodynamic activity (OTA) to provide protective oxide layers.

Setting OTA in flowing lead alloys makes corrosion highly dependent upon the oxygen concentration and the oxidation processes at materials surfaces. The active oxygen control technique exploits the fact that lead and bismuth are chemically less active than the major components of steels, such as Fe, Ni, and Cr. By carefully controlling the oxygen concentration in lead-bismuth eutectic (LBE), it is possible to maintain an iron- and chrome-based oxide film on the surfaces of structural steels, while keeping lead and bismuth from excessive oxidization that can lead to precipitation contamination. Thermal analysis has given an ideal oxygen level range in a non-isothermal lead alloy coolant system. However, in a practical coolant loop, the proper oxygen level depends not only on thermal factors but also on hydraulic factors (system operating temperature, temperature profile, flow velocity, etc.). In addition, the oxygen distribution in a non-isothermal lead alloy coolant system is still unclear. The optimal oxygen levels still need to be investigated.

RESEARCH OBJECTIVES AND METHODS

The goals of this research project are to provide a basic understanding of protective oxide layer behaviors and to develop oxide layer growth models of steels in non-isothermal lead alloys coolant systems, in particular:

- To elucidate the mechanism of the protective oxide layer growth of steels in static, non-isothermal flowing lead alloy coolant systems with oxygen concentration level control.
- To elucidate the mechanism of mass transport of oxygen and corrosion products in the multi-phase system.
- To develop oxidation growth models of steels in lead alloy coolant systems.
- To clarify the dependence of the oxidation process on the hydraulic factors and the oxygen concentration distribution and level.
- To clarify the optimal oxygen concentration levels in practical coolant system scales.
- To interpret the experimental results from test loops and to apply them to the design of practical nuclear coolant systems.



The percentage of the oxygen occupied sites for $K_d=4$, $C_{oxy}=0.2$ and $P_{act}=0.0005$ at $N_t=200,000$ with Neumann and Dirichlet boundary conditions at $y=0$.

RESEARCH ACCOMPLISHMENTS

Boundary conditions study of the self-coded cellular automata (CA) oxidation model coupled with inward oxygen diffusion

In a previous study of the CA model, the oxide layer growth of steel in a liquid lead alloy environment was studied considering the inward oxygen diffusion. The boundary condition of the oxygen concentration at the far away conditions was set as a Dirichlet boundary condition. The oxygen sites were simply eliminated whenever they diffused to the far boundary. This, however, is not a proper physics model. The code of the improved CA oxide layer growth model has been modified with a Neumann boundary condition of oxygen concentration at the far end of the specimen (at $y=0$). The self-coded CA model considers eight Moore neighborhood lattice sites and oxygen diffuses along the boundary of the lattices. The result shows that the oxygen concentration at the far end satisfies the Dirichlet boundary condition as well if the oxygen diffusion rates in the oxide layer and the metal are relatively small compared to the reaction rate. This is true in most of the real cases and the specimen is usually relatively large compared to the oxide layer thickness.

A simple scale removal model based on the improved CA oxidation model

A stochastic CA oxidation model was developed to consider the scale removal effect. In the simple scale removal model, it is assumed that the oxide layer sites which are close to oxidant sites have a probability to move away. An average removing probability \overline{P}_{kr} of a lattice oxide cell was assumed, if the oxide cell is contacting with the LBE flow. The simulation results show that the oxide layer growth following the parabolic law, with a thinner

thickness than the case without scale removal.

A new scale removal model considering the hydraulic effect (the flow direction)

In the new scale removal model, the scale removing probability considers the influence of more neighboring cells (the eastern, western, northern, northeastern, and northwestern sites) which surround the objective oxide layer site. The effect of the neighboring cells on the objective oxide cell is based on the estimation of the hydraulic effect (the flow direction). The proposed scale removal model can lay a basis for a future study coupling with alloy components added in the stainless steel.

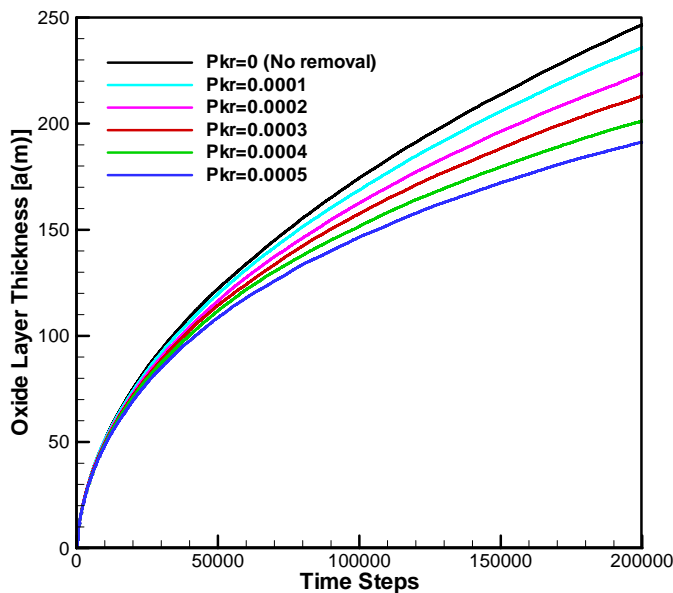
For example, if the flow direction is assumed to be from left flowing to right, more solid sites at the left hand side will make the objective oxide cell more difficult to be removed. Basically, the scale removing probability averaged from the total possible cases should equal to \bar{P}_{kr} as in the simple model. However, the advantage is that the new \bar{P}_{kr} model may predict a different roughness on the oxide surface, especially when alloy components are considered, for each cell,

$$P_{kr}^i = f_i \bar{P}_{kr}$$

Where f_i is a factor reflects the difficulty by which the objective oxide cell will be removed. The more difficult a cell will be to remove, the lower the factor value will be. Thus,

$$\frac{1}{M} \sum_i^M f_i = 1$$

where M is the total number of the possible cases. The scale removal effect on the oxide layer growth has been studied extensively using the self coded CA model. The oxide layer growth was



The oxide layer thickness vs. time steps for cases with $P_{act}=0.005$ with $K_d=2$, $C_{oxy}=0.2$.

ACADEMIC YEAR HIGHLIGHTS

- ◆ Taide Tan successfully defended his Ph.D. dissertation titled “Modeling of Protective Oxide Layer Growth in Non-isothermal Lead-Alloys Coolant Systems” on June 20, 2007.
- ◆ T. Tan, Y. Chen, H. Chen, and S. Hsieh, “Modeling of a Diffusion Controlling Oxidation Process with Scale Removal in Oxygen Contained Liquid Flow,” *Proceedings, IMECE*, Seattle, WA, November 11-15, 2007.
- ◆ T. Tan, Y. Chen, and H. Chen, “An Improved Mesoscopic Oxidation Model of Metals in Lead Bismuth Eutectic,” *Computational Material Science*, 43 (2) pp 251-267, August 2008.
- ◆ H. Chen, Y. Chen, J. Zhang, and H.-T. Hsieh, “A Lattice Boltzmann Modeling of Oxygen Transport and Corrosion Behavior in Natural Convection Lead-alloy Flow,” *Nuclear Engineering and Design*, Vol. 237, pp 1987-1998, 2007.
- ◆ H. Chen and Y. Chen, “Cellular automaton modeling on the corrosion/oxidation mechanism of steel in liquid metal environment,” *Progress in Nuclear Energy*, Vol. 50, pp 587-593, 2008.
- ◆ T. Tan and Y. Chen, “Simulations of Metal Oxidation in LBE at a Mesoscopic Level,” *Proceedings, ASME ICONE16*, Orlando, FL, May 11-15, 2008.
- ◆ Y. Chen, T. Tan, and H. Chen, “Oxidation Companioned by Scale Removal: Initial and Asymptotical Kinetics,” *Journal of Nuclear Science and Technology*, 45 (7) pp 662-667, 2008.

found to obey Tedmon’s theory.

Optimization approach in variable charge potential for metal/metal oxide systems in the molecular dynamics (MD) simulations

A new direct approach for finding charge distributions among ions has been proposed in the MD study. Earlier attempts on minimizing the total system energy of metal/metal oxide systems with given charge constraints appear to be indirect and unnecessarily complicated. The energy minimization problem is in fact an optimization problem and hence can be solved by an optimization method. The approach is based on an optimization algorithm, called the Generalized Reduced Gradient (GRG) method. This efficient approach can be readily employed in molecular dynamic simulations for metal/metal oxide systems.

FUTURE WORK

The next phase of the project involves accomplishing the following tasks:

- To interpret the macroscopic corrosion/oxidation process with the mesoscopic CA model, including scale removal and erosion to the base material.
- To benchmark the CA model with the available experimental corrosion data.
- To perform molecular dynamic simulations of lead/lead-bismuth and iron under varied oxygen concentrations.

Research Staff

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Collaborators

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Task 22

Design Concepts and Process Analysis for Transmuter Fuel Manufacturing

G. Mauer

BACKGROUND

The safe and effective manufacturing of actinide-bearing fuels for any transmutation strategy requires that the entire manufacturing process be contained within a shielded hot cell environment. To ensure that the fabrication process is feasible, the entire process must be designed for remote operation. The equipment must be reliable enough to perform over several decades, and also easy to maintain or repair remotely. The facility must also be designed to facilitate its own decontamination and decommissioning. In addition to these design factors, the potential viability of any fuel fabrication process will also be impacted by a number of variables, such as the current state of technology, potential problem areas, deployment scaling, facility safety, and cost.

RESEARCH OBJECTIVES AND METHODS

The goal of this project was to provide technical support to process designers working on the development of the fuel cycles for transmutation applications. Detailed process models were developed to better define the impact of fuel choice on the transmuter fuel cycle, including relative process losses, waste generation, and plant capital cost. These process models provide insight regarding required plant size and the number of plants needed to mesh with the fuel recycling line. They also determine requirements for automation.

Manufacturing models for large-scale production in a hot cell environment were also developed. Combined, these two models allow the assessment of plant layout, and provide the framework for estimation of plant capital and operating cost estimates, and for feasibility in general. The operations of robotic equipment and the sensor technology required for safe and reliable robot control have been evaluated through simulations in three-dimensional space. The manufacturing technology developed for hot cell applications is also applicable to other, more general uses, where occupational hazards prevent human presence near processes.

Simulations: This task modeled manufacturing processes to generate a realistic assessment of plant layout, size, feasibility, and technology development required for large-scale remote fabrication of fuel. Modeling of the candidate fuel manufacturing processes was performed using the MSC.visualNastran and ProEngineer simulation software tools. The modeling of dispersion and TRISO fuels were completed.

Cost, Feasibility, and Large Scale Deployment: This task developed the database necessary to provide cost estimates and differential cost for various fuel manufacturing options. Cost estimates regarding projected capital cost, reliability, and plant life were developed and should be refined as additional knowledge is developed.

Automated Vision-Based Image Acquisition and Robot Control: This task explored and demonstrated strategies for the reliable and

Robot grasping a cylindrical pellet



flexible control of the material handling robots inside the hot cell by means of automated vision systems. Since the cameras can be positioned outside the hot cell, such systems would have significant advantages over sensors inside the hot cell, resulting in potentially reduced system maintenance and increased system reliability.

RESEARCH ACCOMPLISHMENTS

Hot Cell robot control: Visual servoing was demonstrated. For this purpose, the stereo vision system was set up in a laboratory next to a Fanuc M-16iB industrial robot.

Sensor-Robot Integration: Driver software was written, tested and implemented for the transmission of sensor data, i.e., the target object's position and orientation, to the Fanuc robot controller.

Accelerated Object Extraction: The contour extraction algorithm can require long durations during image feature extraction (Canny Edge Detection, up to one second per image). Since the cylindrical targets each cover only a portion of the image, there is no need to apply the feature extraction to the whole image. By fragmenting each image into sub-images containing the desired objects, the duration of the feature extraction can be significantly reduced.

Targets are found by under sampling the image and testing the neighborhood of the respective pixels for their color. In application, black pellets were used for black pixels in the neighborhood. By merging all black neighboring locations, the rectangular boundaries of the pellets can be determined by ascertaining their respective contours.

To avoid the issue that corners of targets in certain angles are not collected by the sampling, and possibly eliminated, the search algorithm doubles the number of search points at each of the four sides. The duration of the edge detection process depends on the number of objects in the scene, but is usually a fraction of the amount a complete detailed search.

Steering and Rotation of the Robot End Effector, Gripping Operations: For gripping horizontal cylindrical targets, the orientation of the cylinder is required. The orientation can easily be computed as the angle between the two extracted surface points and the x-

axis. To align the gripper with the target's orientation, the last joint of the robot has to be rotated. For this purpose, the orientation of the cylinder in the bin (in world coordinates) is transformed to robot-specific joint coordinates

After accomplishing the process of picking up targets with a given position and orientation, the first autonomous tests were conducted. In these tests, a single target was randomly placed in the scene, picked up by the robot, and dropped in a box. In later test scenarios, the robot placed the target at randomly generated positions and picked it up again. This procedure was tested iteratively.

Multiple Objects: To extract multiple objects, the existing algorithm, which so far extracted only the coordinates of one object, was extended. The extended algorithm returns a list of points representing the two cylinder surface points for each identified target in the corresponding image. When these lists are created for each of the stereo image pairs, the correspondence matching between targets in both lists is performed using epipolar geometry.

To organize the search for correspondences uniquely, additional information may have to be included. These could be the locations of the image edges, for example, the bottom of the first image matches with the top of the second image.

Upright and Horizontal Targets: In general, there is no need to differentiate between upright and horizontal objects during the target extraction phase. The objects' orientations have no bearing on the appearance of the cylinder in the image (usually the mantle and one end surface). The determination of a cylinder's orienta-

tion is made after the two surface end points of each detected object have been triangulated. If the resulting 3D points are located at the same vertical elevation, the object is horizontal. Otherwise a standing object is assumed.

Graphical User Interface: After the completion of the basic operations for cylinder grasping, a demonstration application was developed. The program offers a graphical map of the scene contents (such as position and orientation of the objects), and tracks the manipulation of single objects. To perform picking and placing of targets, the user selects a specific target by mouse click in order to have the robot execute a set of user-defined operations.

TASK 22 PROFILE

Start Date: July 2004

Completion Date: November 2007

Theses Generated:

Jamil Renno, "Virtual Design and Modeling of Various Manufacturing Processes for Remote Fuel Fabrication of Transmuter Fuel," M.S. Thesis, Department of Mechanical Engineering, UNLV, March, 2005.

Andreas Roderburg, "Design Optimization Of Robotic Manufacturing Work Cell For Transmuter Fuel – Analysis And Simulation Of Robotic Manufacturing In A Hot Cell Environment," Diploma Thesis, Department of Mechanical Engineering, RWTH, Aachen, Germany. January 2006.

Conference Proceedings:

G.F. Mauer, and J. Renno, "Conceptual Workcell Design and Throughput Analysis for Robotic Transmuter Fuel Fabrication," *Proceedings*, ANS Winter Annual Meeting, November 2004.

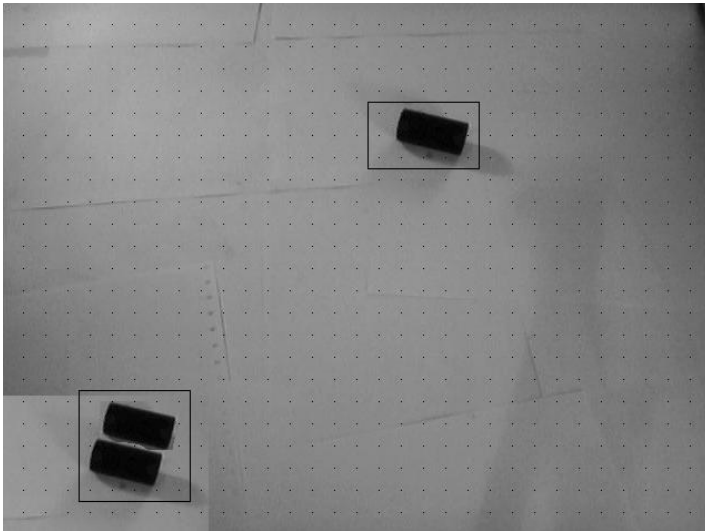
G.F. Mauer, "Design Concepts and Process Analysis for Transmuter Fuel Manufacturing," *Proceedings*, 8th Partitioning and Transmutation Information Exchange Meeting, OECD Nuclear Energy Agency, Las Vegas, NV, November 2004.

G.F. Mauer and J. Renno, "Reliability Analysis of a Robotic Workcell for Transmuter Fuel Fabrication," *Proceedings*, ANS Winter Annual Meeting, November 2005.

G.F. Mauer, "Equipment Redundancy and Plant Reliability in Robotic Hot Cells for Fuel Fabrication," *Proceedings*, American Nuclear Society Winter Annual Meeting, November 2006.

G. Mauer, "Design Concepts and Process Analysis for Reliable, Automated Transmuter Fuel Manufacture," AccApp'07, Pocatello, Idaho, July 2007.

G. Mauer, "Accuracy Analysis of a Robotic Radionuclide Inspection and Mapping System for Surface Contamination," American Nuclear Society Topical Meeting on Decommissioning, Decontamination and Reutilization, September 2007.



Finding the regions containing the targets by undersampling: The black spots represent the sample points, the black rectangles are associated with the extracted regions.

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Collaborators

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Task 23

Development of Nanostructure Based Corrosion-Barrier Coatings on Steel for Transmutation Applications

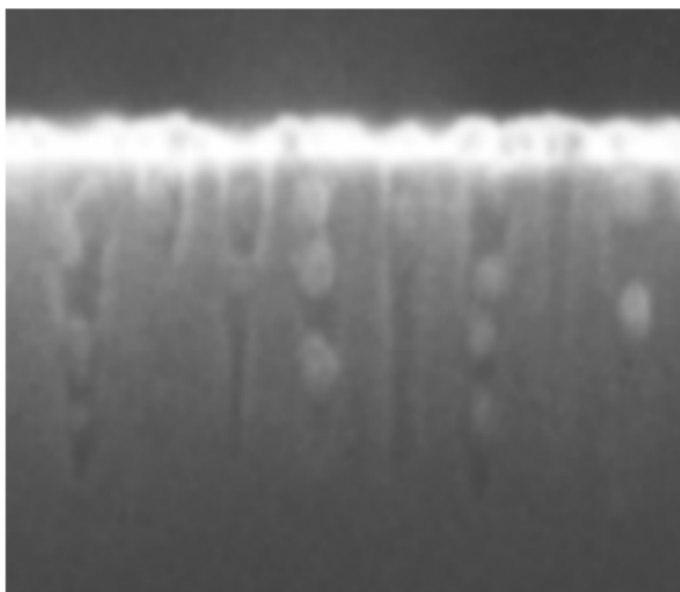
B.J. Das

BACKGROUND

Advanced transmutation systems require structural materials that are able to withstand high neutron fluxes, high thermal cycling, and high resistance to chemical corrosion. The current candidate materials for such structures are ferritic and ferritic-martensitic steels due to their strong resistance to swelling, good microstructural stability under irradiation, and the retention of adequate ductility at typical reactor operating temperatures.

In parallel, lead-bismuth eutectic (LBE) has emerged as a potential spallation target material for efficient production of neutrons, as well as a coolant in the accelerator system. While LBE has excellent properties as a nuclear coolant, it is also highly corrosive to stainless steel. The corrosion is due to relatively high solubilities of the base and major alloying components of steel, such as Ni, Fe, Cr, etc. in LBE at elevated temperatures. Without some protection, the steel structures rapidly corrode in LBE through dissolution and leaching of these materials.

Thus, for long term reliability of the structures, it is necessary to provide some protection of the steel surface from corrosion, without affecting the bulk properties of the steel. One such technique that has been well investigated is the use of oxygen control at the surface of the steel, which maintains a coating of oxide layer that protects the steel surface. The protective layer forms due to the higher affinities of the steel alloying components to oxygen compared to lead and bismuth. However, once a continuous film of oxide is formed, a competing process takes place; the oxide layer interacts with the LBE causing reduction of the oxide layer at higher temperatures.



Cross-sectional image of electro-phoretically deposited nanoparticles inside nanoporous alumina.

It is thus critical to maintain an optimum flow of oxygen at the LBE/steel interface, which is made challenging by the non-uniform temperature distribution in the transmutation systems. In addition, while the oxygen control technique works effectively at lower temperatures, it is not appropriate for higher operational temperatures (500-600 °C), which is becoming increasingly important. Thus, it is necessary to develop alternative techniques for corrosion protection of steel that will perform reliably at elevated temperatures and under thermal cycling in LBE.

RESEARCH OBJECTIVES AND METHODS

The objective of this project was to develop a novel nanostructure based coating technology that will provide significantly improved corrosion resistance for steel in LBE at elevated temperatures (500-600 °C), as well as provide long-term reliability under thermal cycling. The nanostructure based coatings consisted of a layer of nanoporous alumina with the pores filled with an oxidizing metal such as Cr, followed by a capping layer of alumina. Alumina, which is a robust anti-corrosion material, provides corrosion resistance at elevated temperatures. The Cr serves two purposes: (1) it acts as a solid filler material for the pores in the alumina, enhancing its mechanical and chemical integrity, and (2) it acts as a second layer of defense against corrosion by providing a replenishable source of Cr (for the formation of a chromium oxide protective layer) in case the alumina layer is compromised.

The innovation of this project was the use of a nanoporous alumina layer for the coating, which is mechanically flexible and can expand and contract with the underneath steel surface. As a result, the mechanical integrity of the coating is preserved under thermal cycling. In addition to their usefulness at higher temperatures, the proposed coatings can also provide increased reliability at lower temperatures by complementing the oxygen control technique. The nanostructure based coatings developed in this project will significantly enhance the long-term reliability of steel structures in LBE at elevated temperatures and under thermal cycling.

Working with the national laboratory collaborator, the stainless steel alloys HT-9 and EP-823 were chosen as the candidate materials for investigation at this time. The above project objective was achieved in three phases; each phase will be carried out over a one-year period.

Phase I developed the fabrication technology for the coatings on steel, and studied their structural integrity at elevated temperatures and under thermal cycling.

Phase II performed corrosion studies of the structures in LBE at elevated temperatures.

Phase III used the data from Phases I and II to develop an

optimized coating technology for improved structural integrity under thermal cycling, and improved corrosion resistance in LBE at elevated temperatures. If necessary, multiple layers of such coating structures were used for increased resistance to corrosion.

During Year 3 of the project (2006-2007), extensive investigation was carried out on the deposition of longer and improved quality metal nanowires inside the pores using a new technique as well as the establishment of the experimental setup for characterization of the coatings.

RESEARCH ACCOMPLISHMENTS

As reported previously, during Phases I and II of the project, a significant problem was encountered with the synthesis of Cr nanowires inside the alumina pores, the uniformity of coverage was very poor. As a result, alternative metals to form nanowires were looked into and nickel was selected primarily due to its established electrochemical synthesis procedure.

A number of recipes were investigated for their suitability for synthesis inside nanoscale pores, and the process parameters were experimentally optimized for the synthesis of nanowires. The deposition of the Ni nanowire was performed in a two-electrode arrangement with a mixture of $\text{NiSO}_4 \cdot 6\text{H}_2\text{O}$, $\text{NiCl}_2 \cdot 6\text{H}_2\text{O}$ and H_3BO_3 as the source of Ni. The wires were deposited at a potential difference of 1 V between the anode and the cathode and at ambient temperatures. The Ni nanowire samples were characterized by Field Emission Scanning Electron Microscopy (SEM), which showed excellent uniformity and coverage. However, while Ni can provide very good structural integrity to the porous alumina, one potential problem is its higher dissolution rate in LBE.

In addition, it was noted that the heights of the Ni nanowires were limited, and even with extended deposition times, the heights were limited to a few tenths of a micron. This limited height was believed to be arising due to the dynamics of fluid flow inside the nanoporous alumina.

To address the above issues, two different approaches were taken. A systematic study was continued during the past year to improve the deposition uniformity of the Cr nanowires, and some improvement was achieved. The uniformity of coverage showed improvement and experimental optimization of process parameters was continued for further improvement of the coverage uniformity. In addition, a new and promising approach was initiated for the deposition of metal nanowires inside the nanoporous alumina as described below.

During the third year, a new technique was initiated and investigated for the deposition of metal nanowires inside the nanoporous alumina. This technique involves the electro-phoretic deposition of nanoparticles inside the pores followed by recrystallization into nanowires. The major advantage of this technique is that the chemistry of nanoparticle synthesis is separated from the formation of nanowires, as a result it has the potential to eliminate the limitations encountered in nanowire synthesis. Nanowire deposition inside the alumina pores is made challenging by the nanoscale fluid flow and chemical reactions inside the nanoscale volume of nanoporous alumina pores. In this new technique, nanoparticles of the desired metal are first synthesized by an established technique in an emulsion form, which are then incorporated inside the nanoporous alumina pores. Since nanoparticles are relatively less complex to incorporate inside pores, it provides an excellent means for the synthesis of nanowires of good uniformity and desired heights. The nanoparticles can be synthesized using an established technique such as sol gel technique. The nanoparticles can be then annealed to form the nanowires or sonication can be used during electro-phoretic incorporation to form the nanowires.

Deposition of Cr nanowires on steel substrates will be achieved by using the following procedure. A new batch of steel samples will be coated with metallic aluminum which will be anodized using constant current anodization. Since it is not possible to obtain cross-sectional images of the structures on steel samples, the voltage-time characteristics will be measured to monitor the quality of the anodized alumina structures. A typical voltage-time characteristic obtained during the anodization of aluminum on steel samples, when compared to data from silicon samples, confirms the good quality of the nanoporous alumina. Cr nanoparticles will be synthesized using the sol-gel technique and will be then electro-phoretically incorporated inside the nanoporous alumina. Sonication will be used during deposition to fuse the nanoparticles to form nanowires. A sonication apparatus was obtained for this purpose. This will be followed by the deposition of a thick film of dense alumina, the process technology for which has already been developed during the previous phase of the research.

TASK 23 PROFILE

Start Date: July 2004

Completion Date: November 2007

Conference Proceedings:

B. Das, "Nonlithographic Nanoscale Devices," invited paper, International Conference on MEMS and Semiconductor Nanotechnology, Kharagpur, India, December, 2006.

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Task 24

Development of Integrated Process Simulation System Model for Spent Fuel Treatment Facility Design

Y. Chen and S. Hsieh

BACKGROUND

Integrating and enhancing the Argonne Model for Universal Solvent Extraction (AMUSE) code, which contains a great deal of chemical separations processing, was part of TRP Task 8. Simulating the Light Water Reactor (LWR) Spent Fuel Treatment Facility (SFTF) processes is the major focus for this project. This approach combines commercial process simulation software (ASPEN-Plus) with the chemical separation calculation from the AMUSE code. Based on the current Integrated System Optimization Program, ISOPro (developed by UNLV) this project aims to create a system framework that interacts with both programs and provides analyzed results useful for a SFTF design that provides the functionality of receiving, temporarily storing, and preparing spent nuclear fuel for leaching.

A spent fuel treatment facility has many individual processes that make up the overall separation. Each block in the overall process flow sheet represents a unique process that carries out an individual chemical separation, and each block contains numerous operations that are responsible for the chemical separation.

The Uranium Extraction Process (UREX) is often the first removal process in the overall scheme of spent fuel recycling. After extracting U and Tc from the spent fuel, the washed and rinsed effluents (Cs/Sr raffinate) move on to the next separation process.

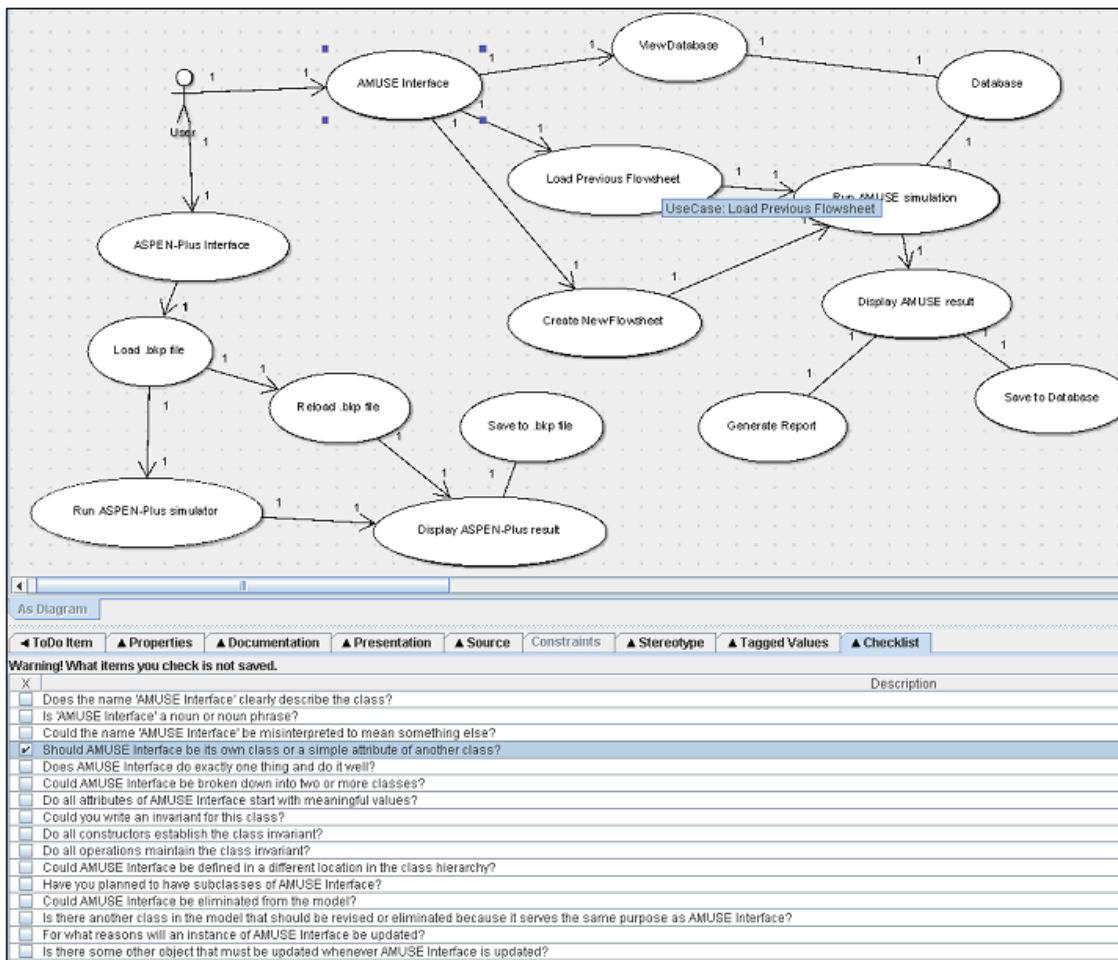
A key concept in the SFTF plant design is the recycling of nitric acid. The purpose of the nitric acid recycling system is to concentrate the spent nitric acid to a desired molarity which, in turn, can be recycled back into the process. The spent nitric acid streams from many processes are collected and sent to a distillation column where it is separated from the impurities collected in the various separation processes.

RESEARCH OBJECTIVES AND METHODS

The major objectives will lead to the creation of a framework that combines all the strengths of AMUSE's complicated calculations, well-established commercial system process package, and ISO-Pro's flexible parameter optimization modules. Development of the process simulation code can be done using the solvent extraction

process at Argonne National Laboratory in collaboration with the research team from the Mechanical Engineering Department at UNLV. The objectives are as follows:

- To develop a framework for simulating the Spent Fuel Treatment Facility process using the AMUSE code, ASPEN-Plus commercial process package and ISOPro system engineering model.
- To develop middle-ware interfaces that can communicate between the AMUSE code and ASPEN-Plus packages.
- To extend the existing system engineering model for the optimization process that includes process simulation results.
- To include a scenario-based database system that efficiently reports required information as chart output using web-based programming, and Microsoft Visual Basic (MS VB).



Use Case for the updated ISOPro package.

Task 25

Electrochemical Separation of Curium and Americium

D.W. Hatchett and K. Czerwinski

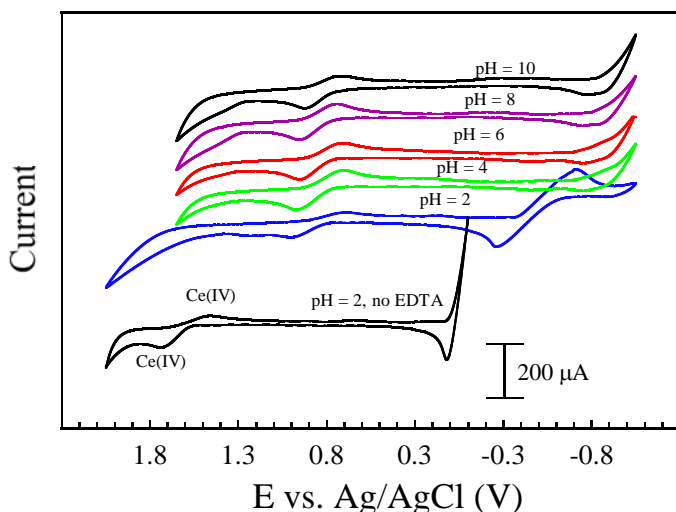
BACKGROUND

In the Advanced Fuel Cycle Initiative program, the separation of the trivalent actinides Am and Cm from the trivalent lanthanides, and even each other, has been identified as an area of particular interest. The removal of the lanthanides from the trivalent actinides is necessitated for neutron economy of a fuel containing Am and Cm. Furthermore, a fuel containing only Am would ease the demands on a reactor for transmutation.

The destruction rate of Cm isotopes in a reactor is comparable to their decay rate. Separation and storage of the Cm, and reintroduction into the fuel as the Pu daughters is a transmutation option that can be explored if a suitable method for the separation of Cm from Am is obtained. Furthermore, removal of Cm would reduce difficulties in fuel fabrication.

Electrochemical methods can be used to effectively separate actinide and lanthanide species from complex mixtures. This is based on the unique electrochemical properties of each specific target species. It has been found through study that, with the exception of Ce, aqueous solutions provide unsuitable electrochemical windows to effectively evaluate the thermodynamic properties that are useful for chemical separation. Therefore a more novel approach was examined which eliminated the aqueous solution with a room temperature ionic liquid (RTIL) solution.

RTIL solutions do not suffer from the side reactions that are prominent in aqueous environments. In addition, the potential window is much larger for the RTIL solutions. The RTIL solutions are a new starting point for the electrochemical separation of individual species from a mixture.



Electrochemistry of the Ce(III)/Ce(IV) redox couple at a Platinum electrode uncomplexed and complexed with EDTA as a function of pH. [EDTA] = 12 mM [Ce(III)] = 6 mM, $v = 100$ mV/s. Free [Ce(III)] = 6 mM is shown on the bottom of the figure for comparison.

RESEARCH OBJECTIVES AND METHODS

The objective was to use electrochemical techniques to develop a thermodynamic understanding of actinide and lanthanide species in RTIL solutions, and use this data to effectively separate species with very similar chemical properties.

In consultation with a DOE collaborator, electrochemical methods and materials were evaluated and used to exploit the thermodynamic differences between similar chemical species enhancing the ability to selectively target and sequester individual species from mixtures. This project, in its third year, successfully completed phases 1, 2. Phase 3 was partially completed. The project expanded to include phase 4.

Phase 1

Evaluated thermodynamic oxidation/reduction properties of Ce using electrochemical methods.

Phase 2

Evaluated the thermodynamic properties of chelated Ce, Sm, and Eu at carbon, platinum and gold electrodes.

Phase 3

Examined the use of conductive polymer membranes for the uptake and expulsion of complexed and uncomplexed actinide and lanthanide species. The uptake and selective adsorption and separation of individual actinide and lanthanide species, including the isolation of Cm from Am, still needs to be examined using conductive polymer/metal composite membranes containing bound chelates. Follow-on work was conducted under a new TRP project (Task 38).

Phase 4

Prepare and characterize RTIL solutions. Examine the electrochemical window and evaluate the electrochemical properties of lanthanide and actinide species in the non-aqueous ionic environment.

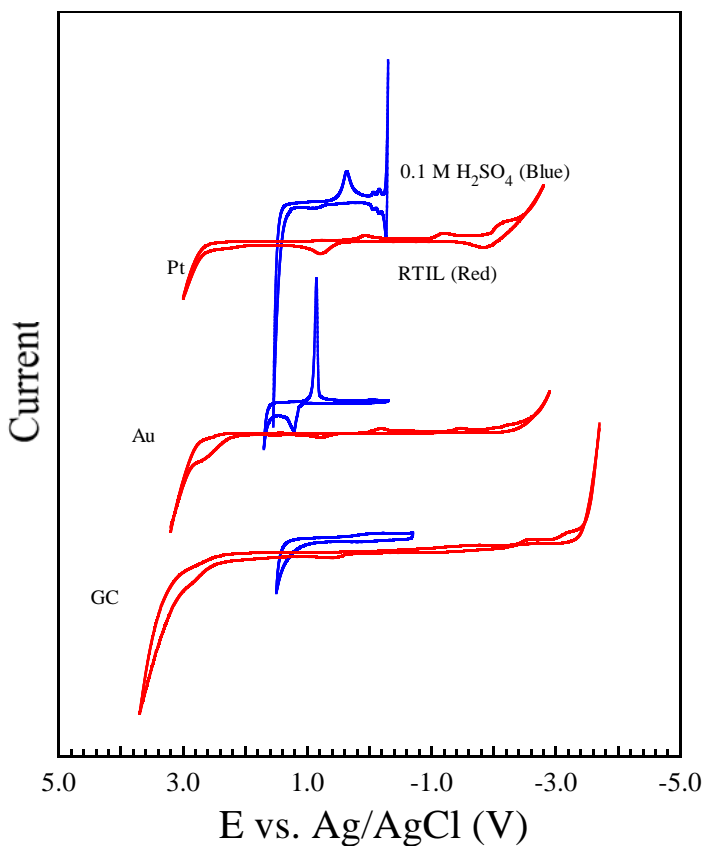
The following were specific goals for 2007-2008:

- To prepare RTIL solutions.
- To evaluate the potential window of RTIL solutions.
- To develop a fundamental understanding of the thermodynamic properties of actinide and lanthanide species such as Cm, Am, Ce, Nd, Eu, and Sm in RTIL solutions.

Follow-on work was conducted under a TRP Task 38.

RESEARCH ACCOMPLISHMENTS

The research accomplishments can be separated into aqueous and non-aqueous systems. The initial goal of the research was to examine the thermodynamic properties of lanthanide and actinide species in aqueous solutions. It was apparent from the initial studies that the ability to examine lanthanide and actinide species was limited in aqueous solution. However, there was success in re-



Electrochemistry of Gc, Au, and Pt electrodes in RTIL ([MeBu₃N][NTf₂]), (Red) and 1.0 M H₂SO₄, (Blue).

solving the Ce redox couple in aqueous solution. In addition, complexation with EDTA provided increased stability of the Ce redox couple at pH values that would typically lead to the precipitation of Ce(OH)_x (x = 2 or 3). The voltammetric response of the Ce redox couple before and after EDTA complex formation is illustrated (see opposite page).

The advantage to eliminating aqueous side reactions and expanding the potential window is that previously inaccessible electrochemical reactions may be observed. These systems provide a huge potential window for the observation of oxidation and reduction of lanthanides and actinides.

In addition, the potential mediated deposition of f-elements is possible indicating the controlled separation of chemical species is thermodynamically feasible. For example, the reduction of Pu³⁺, Am²⁺, Am³⁺, Cm³⁺, and U³⁺ all occur before E = -1.90 V vs. Ag/AgCl, well within the negative potential limit for the RTIL at platinum (-2.6 V vs. Ag/AgCl), gold (-2.2 V vs. Ag/AgCl), and

glassy carbon electrode (-3.4 V vs. Ag/AgCl).

A second advantage is the increased solubility of lanthanide and actinide species in the RTIL. RTIL solutions have been utilized in the waste cycle for extraction of species. These solutions could be useful in the potential dependent separations of lanthanide and actinide species. The preliminary evaluation of RTIL solutions relative to normal aqueous conditions is also illustrated.

TASK 25 PROFILE

Start Date: July 2004

Completion Date: December 2007

Follow-on Task: "f-Element Electrochemistry in RTIL Solutions: Electrochemical Separation of Lanthanides and Actinides," TRP Task 38.

Theses Generated:

Sandra Elkouz, M.S. Thesis, Department of Chemistry, "Complexation and Electrode Effects on the Electrochemical Behavior of the Cerium(IV)/Cerium(III) Redox Couple," April, 2007.

Journal Articles:

S. Elkouz, P. Houlihan, K. Czerwinski, and D.W. Hatchett, "The influence of electrode composition, pH, and ionic strength on the redox properties of Ce(IV)/Ce(III)," submitted to *Electrochimica Acta*, April 2007 (Ms. Ref. No.: EA-S-07-00835).

D.W. Hatchett, S. Elkouz, P. Houlihan, K. Czerwinski, "Electrochemical properties of Ce/EDTA complex in alkaline solutions," *Electrochimica Acta*, submitted 2008.

S. Elkouz, K. Czerwinski, and D.W. Hatchett, "The Electrochemical Characterization of Ce at Au, Pt and Glassy Carbon Electrodes: Influence of pH and Ionic Strength," manuscript submitted, 2006.

Conference Proceedings:

S. Elkouz, K. Czerwinski, and D.W. Hatchett, "The Electrochemical Characterization of Ce at Au, Pt and Glassy Carbon Electrodes: Influence of pH and Ionic Strength," 209th Electrochemical Society Meeting, Denver, CO, May 9 – 13, 2006.

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Patrick Houlihan, Undergraduate Student, Department of Chemistry

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Task 26

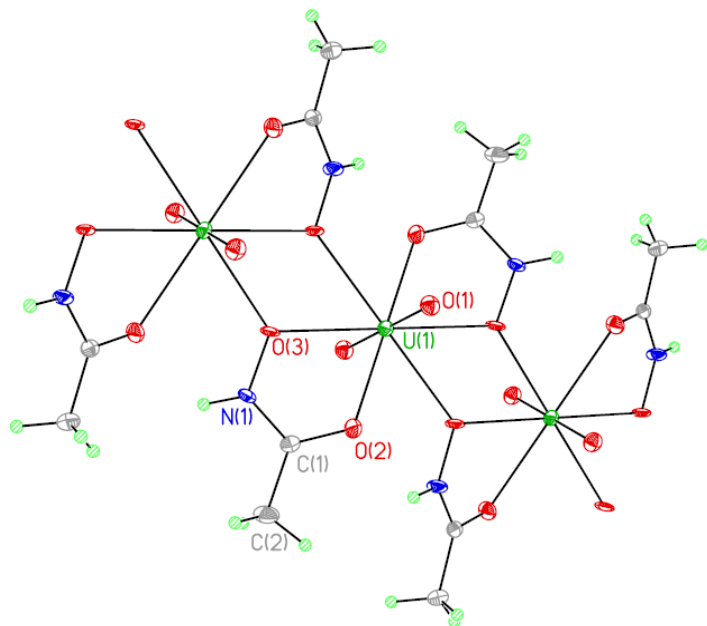
Fundamental Chemistry of U and Pu in the TBP-Dodecane-Nitric Acid System

K. Czerwinski

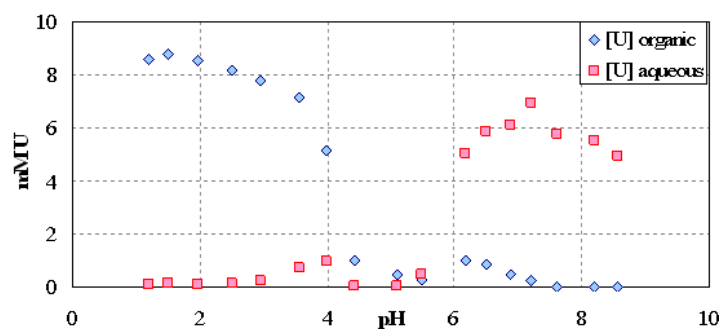
BACKGROUND

The extraction of tetravalent Pu and hexavalent uranyl in nitric acid solution by tributylphosphate (TBP), based on the formation of organic phase neutral complexes such as $\text{Pu}(\text{NO}_3)_4 \cdot 2\text{TBP}$ and $\text{UO}_2(\text{NO}_3)_2 \cdot 2\text{TBP}$, has been the foundation of actinide purification for a number of decades. Upon reduction of Pu(IV), Pu(III) is back extracted into an aqueous phase. However, the data needed for detailed modeling of Pu extraction is not available for all conditions of concern to the Advanced Fuel Cycle Research and Development program. This proposal was developed with Dr. George Vandegrift of Argonne National Laboratory to obtain the necessary data for Pu extraction modeling.

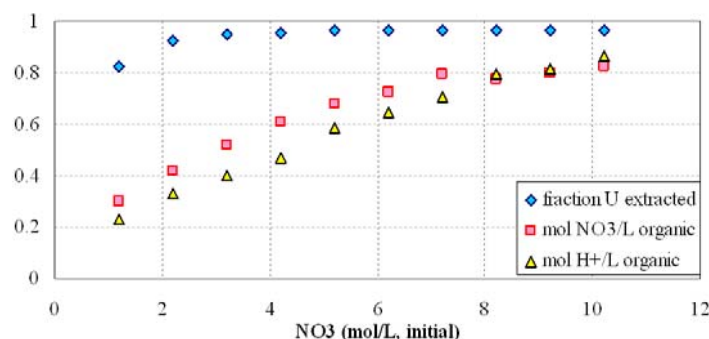
Understanding the role of nitrate in actinide speciation is important for determining the necessary data for extraction modeling. Difficulties in modeling Pu extraction under some nitrate conditions may be due to ill-defined constants of formation for the dintrate and trintrate species of U and Pu. The influence of pertechnetate on the speciation of U and Pu in the TBP-dodecane-nitric



Crystal Structure of Uranyl-AHA.



The measured concentrations in the organic and aqueous phases of 10 mM uranyl, 100 mM AHA extracted with TBP.



The measured concentrations of uranyl, nitrate, and acid in the organic phase extracted from 0.02 M U, 1.0 M HNO_3 , and varied initial nitrate.

acid system is still poorly understood. Since pertechnetate is capable of reoxidizing reduced Pu species, it may have a profound impact on the extraction of Pu.

In the uranium extraction process, acetohydroxamic acid (AHA) is expected to be used. AHA can complex and reduce Pu, decreasing its extraction into the organic phase; the effect of uranyl-AHA on U extraction has not been quantified. AHA can reduce pertechnetate as well, initiating a redox cycle with both U and Pu. In a solvent extraction system using AHA, it is necessary to determine the complexation kinetics, redox reactions, and thermodynamics of AHA interactions with these elements.

RESEARCH OBJECTIVES AND METHODS

The research objective is to experimentally evaluate the fundamental speciation of Pu and U in the TBP-dodecane-nitric acid-AHA system and the effect of pertechnetate, specifically:

- To determine the influence of nitrate on the speciation of U and Pu in the TBP-dodecane-nitric acid system. The aqueous and organic speciation of U and Pu are examined as a function of the nitric acid concentration, nitrate concentration, actinide ion concentration, temperature, and time.
- To determine the speciation of U and Pu with AHA in the presence and absence of TBP-dodecane organic phase. The aqueous and organic speciation of U and Pu are evaluated as a function of AHA concentration, metal ion concentration, metal ion redox state, pH, and temperature.
- To determine the interaction of AHA with pertechnetate, and the effect on the interaction of AHA and pertechnetate with U and Pu.
- To incorporate thermodynamic and kinetic data into existing modeling codes.

All of the initial experiments were performed with uranyl, UO_2^{2+} . The results obtained from U are the basis for further experiments with Pu. In extraction experiments, the aqueous and organic phases are contacted in equal volumes from 0.3 to 5.0 mL.

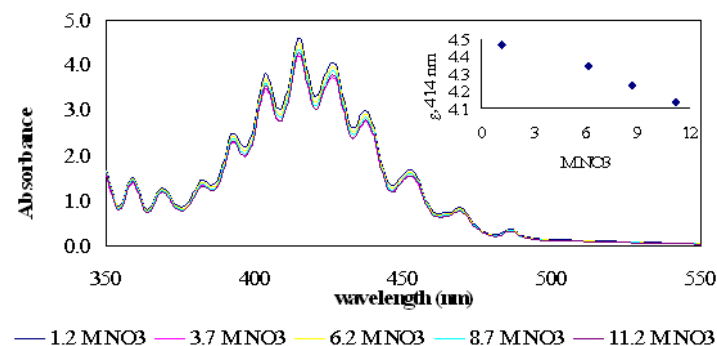
RESEARCH ACCOMPLISHMENTS

Uranyl Nitrate. The complexation of uranyl (0.01 to 0.1 M) with nitrate was studied at 1 M HNO₃ with excess nitrate (from LiNO₃) varied from 0 to 10 M. It was found that at higher nitrate concentrations, more uranyl was present in the organic phase, and, in the absence of uranyl, more HNO₃ extracted.

Uranyl-AHA. The complexation of uranyl with AHA and its effect on U extraction into TBP-dodecane was studied via ultraviolet-visible spectroscopy (UV-Vis), nuclear magnetic resonance (NMR), and Inductively Coupled Plasma-Atomic Emission Spectroscopy in perchlorate and nitrate media. The concentrations of uranyl (0.1 to 10 mM) and AHA (1 to 700 mM) and the pH (1 to 10) were systematically varied. It was determined via NMR that there are at least two species of uranyl-AHA, that the species formed is pH-dependent, and that there is little hydrolysis at 25 times or higher molar excess AHA. The low-pH species extracts from perchlorate media as well as free uranyl, and the high-pH species remains in aqueous solution. In nitrate media, the nitrate competes with and oxidizes the AHA under acidic conditions; the exact interactions remain to be explored.

In addition, methods for synthesis of the uranyl-AHA solid were developed; the solid was characterized by UV-Vis, Fourier Transform Infrared spectroscopy, NMR, laser fluorescence, melting point, and X-ray Absorption Fine-structure Spectroscopy (XAFS), and a crystal structure was obtained. The uranyl-AHA is present as a highly symmetrical polymer, in which each equatorial atom is exactly 180° from its opposite and the ligand planes are twisted within and between unit cells.

Technetium-AHA. Previous work failed to observe pertechnetate reduction by AHA; however, in the presence of catalytic acid (0.5 to 4 M HNO₃ or HClO₄) and a large excess of AHA (0.5 to 4 M),



The UV-Vis spectrum of uranyl nitrate in 30% TBP varies with nitrate concentration.

pertechnetate (0.1 - 20 mM) will be reduced to the Tc^{II}NO(AHA)₂ species in aqueous media. This species is highly water soluble and does not extract into 30% TBP. The presence of uranyl has no effect on the formation or extraction of this species, and up to hundredfold excess UO₂²⁺ does not affect the rate of formation. There is significant evidence that the reaction is instantaneous and the change in the Tc-AHA UV-Vis spectrum is due to the increasing pH as AHA hydrolyzes to acetic acid. However, the mechanism of formation is yet to be elucidated.

TASK 26 PROFILE

Start Date: August 2004

Completion Date: December 2007

(This work continued under Task 40, see pages 84-85.)

Journal Articles:

C.S. Gong, W.W. Lukens, F. Poineau, and K.R. Czerwinski, "Reduction of pertechnetate by acetohydroxamic acid: Formation of [TcNO(AHA)₂(H₂O)]⁺ and implications for the UREX process," *Inorg. Chem.*, 47(15), 6674-6680, 2008.

C. Gong, F. Poineau, and K. Czerwinski, "Synthesis and characterization of the solid uranium(VI) dioxo-diacetohydroxamate complex," *Radiochim. Acta*, 95(8), 439-450, 2007.

Conference Proceedings:

A.D. Wright and K. Czerwinski, "Nitrate effects on uranium and plutonium extractions," 234th ACS National Meeting, Boston, MA, August 2007.

C.S. Gong, F. Poineau, and K. Czerwinski, "Reduction of Technetium by Acetohydroxamic Acid in Nitric and Perchloric Acids and its Effects on Extraction into Tributylphosphate-dodecane," Actinide Separations Conference, Las Vegas, NV, June 2007.

A. Wright and K. Czerwinski, "Nitrate effects on uranium and plutonium extractions," 233rd ACS National Meeting, Chicago, IL, March 2007.

C. Gong, F. Poineau, and K. Czerwinski, "Synthesis and characterization of the solid uranyl-acetohydroxamic acid complex," 233rd ACS National Meeting, Chicago, IL, March 2007.

K. Czerwinski, A. Wright, N. Smith, L. Wright, and G. Cerefice, "Fundamental Chemistry of Uranium in Extraction for use in Enhanced Separations and Proliferation Resistance," MRS Scientific Basis for Nuclear Waste Management XXX, Boston, MA, November-December 2006.

A. Wright, N. Smith, F. Poineau, and K. Czerwinski, "Nitrate effects on uranium and plutonium extractions in the tributylphosphate-dodecane system," 232nd ACS National Meeting, San Francisco, CA, September 2006.

A.D. Wright and K. Czerwinski, "Nitrate effects on uranium and plutonium extractions in the tributylphosphate-dodecane system," 231st ACS National Meeting, Atlanta, GA, March 26-30, 2006.

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Task 27

Reactor Physics Studies for the AFCI Reactor-Accelerator Coupling Experiments Project

D. Beller

BACKGROUND

Investigation of transmutation using accelerator-driven subcritical systems (ADS) was an original component of the Advanced Fuel Cycle Initiative. The Reactor Accelerator Coupling Experiments (RACE) represented the nations efforts in ADS experimental research. The RACE project was a university-based research project lead by the Idaho Accelerator Center of Idaho State University (ISU). The RACE project was initiated in 2003 at ISU and experiments were conducted there and at the University of Texas (UT) at Austin from 2004 to 2006. In these experiments, source neutrons were generated by using electron accelerators to produce high-energy bremsstrahlung photons that then induced photon-neutron reactions in heavy-metal targets. These compact, transportable accelerator/target systems produced a source of about 10^{10} - 10^{12} n/s, which then initiated fission reactions in the subcritical systems.

RESEARCH OBJECTIVES AND METHODS

The specific research objective of this three-year project was to design and conduct accelerator driven experiments, to help demonstrate the ability to design, compute, and conduct ADS experiments and to predict and measure source importance, coupling efficiency, sub-critical reactor kinetics and source-driven transients. In addition, databases were created for both steady state and transient ADS experiments for the nuclear community to develop and test new computational codes and methods. The importance of a driving neutron source in various regions of different subcritical assemblies was mapped. Experiments were conducted and compared to calculations with radiation transport and thermal-hydraulics codes such as MCNPX and RELAP.

RESEARCH ACCOMPLISHMENTS

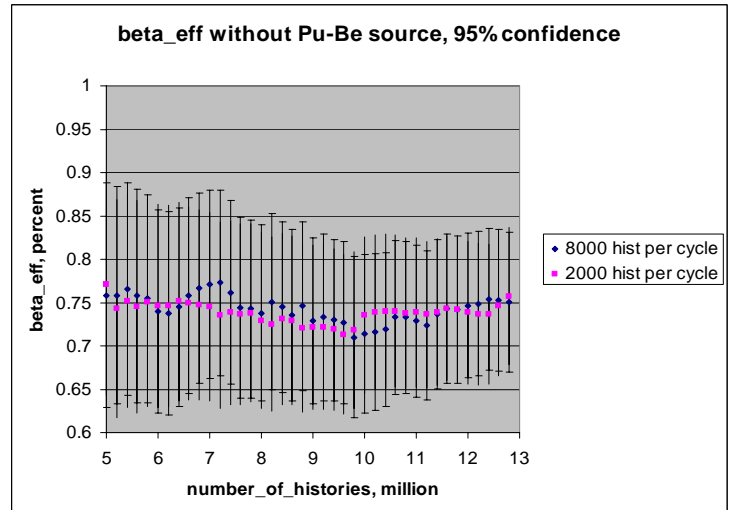
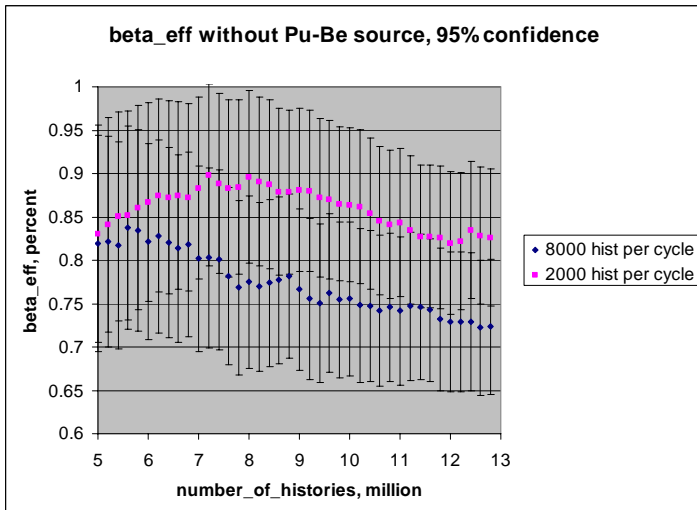
Full-core ADS experiments were conducted at ISU in a compact, subcritical assembly that was constructed at the ISU and fueled with a modular core. Accelerator-coupled experiments with the

TRIGA reactor at the UT-Austin were also conducted beginning summer 2005, and were completed in March 2006. The most extensive experimental program was conducted at ISU, where accelerators were assembled, the beam-target neutron generators were developed, a subcritical assembly was licensed and constructed, and RACE tests were conducted for more than two years. These experiments are described in a wide array of papers.

An accelerator was constructed at ISU, shipped to Texas, and installed in the summer of 2005. It was assembled in a cave at the floor level of the UT Nuclear Engineering Teaching Laboratory (NETL), adjacent to the NETL TRIGA reactor. This reactor can operate at 1 MW_{th}, and can produce pulses up to 1000 MW_{th}. The accelerator target was placed immediately adjacent to one side of the core, centered on that side.

The first UT experiments were conducted with the reactor completely shut down with a criticality of about 0.92, similar to the criticality at ISU experiments. Follow-on experiments were conducted with k_{eff} between 0.92 and 0.95 to possibly 0.97, 0.98, or even critical. Measurements were made with flux wires, fission chambers, and other detectors. In addition, beam diagnostics and monitoring techniques were developed at UT to improve the performance of the linac/target system.

In the early phases of the RACE project, modeling studies were also conducted at both the University of Michigan and Texas A&M University. Texas A&M examined different experiments: (1) use of the TAMU Nuclear Science Center (NSC) TRIGA, which is fueled with 70%-enriched "FLIP" fuel, and has a capability to be pulsed to 1000 MW_{th}; and, (2) assembly of an existing used-fuel, 20%-enriched core around the accelerator target in a different part of the NSC pool (the Texas Transmutation System). The University of Michigan supported the RACE project with computational reactor physics studies of the kinetics of subcritical systems. They studied dynamic response and optimized the ERANOS transport theory model for a RACE configuration by using



Calculated effective delayed neutron fraction for a subcritical (left side) versus a critical (right side); without a PuBe source.



UNLV graduate students Timothy Beller and Ryan LeCounte couple the UNLV High-Power RACE Target to a linear electron accelerator at the Idaho Accelerator Center.

several different energy group structures and orders of polynomial expansion for the neutron flux. In addition, they worked to resolve difficulties encountered with the time-dependent VARIANT transport theory solution.

A low-power phase of the RACE project was conducted in collaboration with the European Community (EC) at ISU with the participation of a member of the EC, ISU, and UNLV. Participants investigated the potential to increase neutron generation by one to two orders of magnitude, which would allow for high-average-power experiments with significant, and easily measured, thermal feedback. In a separate effort, ISU investigated connecting a high-power modulator and klystrons to existing 20 and 25-MeV linacs. This new configuration would provide 20 to 30 kW of electron current, compared to 1 to 2 kW with the existing ISU and Texas accelerators. In addition, incorporating depleted or natural uranium into existing heavy metal targets was investigated to increase the photon-neutron yield. A high-power RACE target was designed and constructed at UNLV and tested for thermal and neutron generation performance at ISU. Simultaneously, an EC Target Working Group examined several alternate high-power target designs. The combination of these two enhancements, if successful, would generate 5×10^{13} to 10^{14} n/s in the center of one of the Texas reactors.

TASK 27 PROFILE

Start Date: September 2004

Completion Date: September 2007

The RACE project produced M.S. and Ph.D. graduates from five universities and more than 90 publications, presentations, and posters directly linked to faculty and students at UNLV. Selected publications are listed below.

Theses Generated:

Evgeny Stankovskiy, Ph.D., Department of Mechanical Engineering, UNLV, "Reactor Physics Studies for the Advanced Fuel Cycle Initiative Reactor-Accelerator Coupling Experiments Project," May 2008.

Timothy Beller, M.S., Department of Mechanical Engineering, UNLV, "Neutronics Analysis of the High-Power Race Target," December 2007.

Ryan LeCounte, Department of Mechanical Engineering, UNLV, "Thermal Hydraulic Analysis of the High-Power Race Target," December 2007.

Journal Articles:

C.O. Maidana, A.W. Hunt, D. Beller, and K. Folkman, "Design, Modeling and Simulations in the RACE Project: First Study for the Development of a Transport Line," *Nuclear Instruments and Methods-A*, 562, pp 892-895, 2006.

V. Kulik, J. Lee, and D. Beller, "Dynamic Analysis of Space-Time Effects in the ISU RACE Configuration," *Nuclear Instruments and Methods-A*, 562, p 838, 2006.

Conference Proceedings:

R. LeCounte, T. Beller, and D. Beller, "Thermal Analysis of the High-Power RACE Target," *Proceedings*, Eighth International Topical Meeting on Nuclear Applications and Utilization of Accelerators (AccApp'07), Pocatello, ID, July 30-August 2, 2007.

D. Beller, Frank Harmon, Thomas E. Ward, and Frank Goldner, "Overview of the U.S. Reactor-Accelerator Coupling Experiments (RACE) Project," *Proceedings*, Eighth International Topical Meeting on Nuclear Applications and Utilization of Accelerators (AccApp'07), Pocatello, ID, July 30-August 2, 2007.

T. Beller, R. LeCounte, B. Howard, and D. Beller, "High-Power Accelerator Target Design for the AFCI RACE Project," *Proceedings*, Eighth International Topical Meeting on Nuclear Applications and Utilization of Accelerators (AccApp'07), Pocatello, ID, July 30-August 2, 2007.

T. Beller, R. LeCounte, and D. Beller, "Analysis of Neutron Production in the High-Power RACE Target," *Proceedings*, Eighth International Topical Meeting on Nuclear Applications and Utilization of Accelerators (AccApp'07), Pocatello, ID, July 30-August 2, 2007.

D. Beller, Frank Harmon, Thomas E. Ward, and Frank Goldner, "Overview of the U.S. Reactor-Accelerator Coupling Experiments (RACE) Project," *Proceedings*, Eighth International Topical Meeting on Nuclear Applications and Utilization of Accelerators (AccApp'07), Pocatello, ID, July 30-August 2, 2007.

D. Beller, "Update on the Reactor-Accelerator Coupling Experiments (RACE) Project," *Transactions of the ANS*, 95, Washington, DC, pp. 943-944 2006.

R. LeCounte, T. Beller, and D. Beller, "Thermal Analysis of Neutron Production in the High-Powered RACE Target," *Transactions of the ANS*, 95, Albuquerque, NM, 2006.

T. Beller, B. Howard, R. LeCounte, and D. Beller, "High-Power Accelerator Target Design for the AFCI RACE Project," *Proceedings*, 2006 International High Level Radioactive Waste Management Conference, Las Vegas, NV, April 30-May 4, 2006.

T. Beller, B. Howard, R. LeCounte, and D. Beller, "Design of the High-Powered RACE Target," 2006 Winter Meeting of the ANS, Student Design Competition.

D. Beller and J. Knebel, "Phase IV of the RACE Project: European Collaborations," *Transactions of the ANS*, 93, pp. 901-902, Washington, DC, 2005.

D. Beller, "Overview of the AFCI Reactor-Accelerator Coupling Experiments (RACE) Project," *Proceedings*, Eighth Information Exchange Meeting on Actinide and Fission Product Partitioning & Transmutation, OECD/NEA, pp 495-504, Paris, France, 2005.

D. Beller, A. Hunt, J. Bennion, M. Reda, K. Sabourov, R. Spaulding, and K. Folkman, "Initial Results from the AFCI Reactor-Accelerator Coupling Experiments (RACE) Project," *Proceedings*, Eighth Information Exchange Meeting on Actinide and Fission Product Partitioning & Transmutation, OECD/NEA, pp 699-710, Paris, France, 2005.

D. Beller, "The Need for Accelerator-Driven Transmutation of Nuclear Waste," *Proceedings*, 3rd Annual Idaho ADS Experiments Workshop, Pocatello, ID, June 2005.

D. Beller, A. Hunt, J. Bennion, M. Garfield, K. Folkman, and M. Reda, "Initial Results from the AFCI Reactor-Accelerator Coupling Experiments (RACE) Project," *Transactions of the ANS*, 91, 446-447, 2004.

D. Beller, "Overview of the AFCI Reactor-Accelerator Coupling Experiments (RACE) Project," *Transactions of the ANS*, 90, 2004.

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Task 28

Impact of the Synthesis Process on Structure Properties for AFCI Fuel Candidates

K. Czerwinski

BACKGROUND

Synthesis of actinium mononitrides using carbothermic reduction of the corresponding oxides has a few outstanding issues, including the formation of secondary phases such as oxides and carbides and low densities of the final product. Furthermore the requirement of a high process temperature at 1700°C, for more than 12 hours is also a drawback particularly for Americium-bearing samples. Therefore, it is important to explore the use of other possible routes to synthesize actinide mononitrides.

A low temperature process is used in this research to produce actinide mononitrides using a fluoride route in which the first step is to mix the actinide oxide with NH_4HF_2 . The second step involves the heat-treatment of the resulting ammonium actinide fluoride salts in ammonia atmosphere. Using different analytical techniques available, the experimental conditions can be studied and optimized to synthesize the required materials with high phase purity. Such available techniques are X-ray Powder Diffraction (XRD), Thermogravimetry and Differential Scanning Calorimetry (TG/DSC), and microscopic techniques such as Scanning Electron Microscopy (SEM) and Transmission Electron Microscopy (TEM). Once the experimental conditions are studied and optimized, a number of actinide nitride systems (uranium, thorium, and neptunium) will be synthesized and characterized to provide knowledge on the chemistry of the systems. Characterization of these nitride systems will include chemical phase identification, lattice parameter refinements, morphological studies, microstructural verifications, thermal behavior, reaction mechanism, and reaction kinetics.

RESEARCH OBJECTIVES AND METHODS

The research objectives are:

- To explore a low-temperature fluoride route to synthesize actinide nitrides.
- To characterize actinide nitrides structurally and thermally.

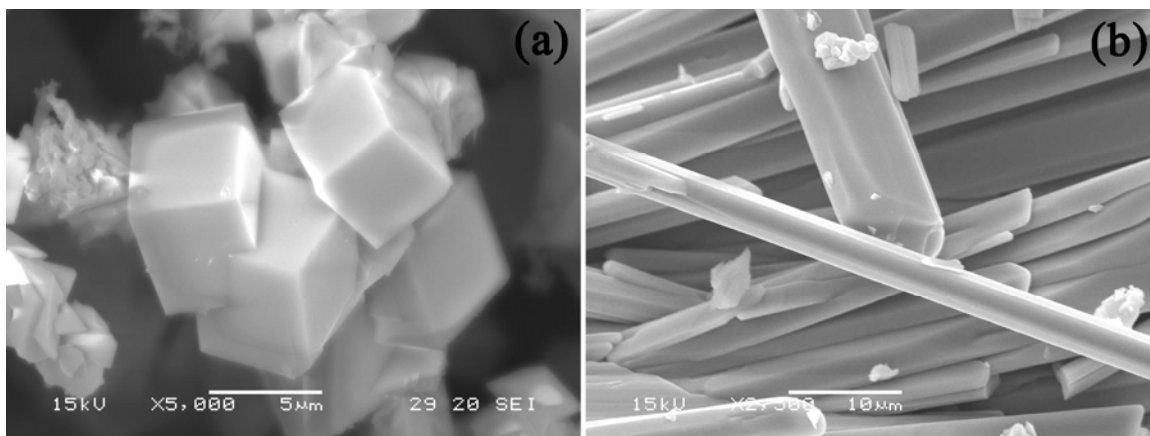
- To use high resolution TEM techniques to explore the microstructure of the radioactive samples.

RESEARCH ACCOMPLISHMENTS

Uranium based nitride synthesis and characterization

The fluoride route was successfully used to synthesize three uranium nitride samples with different stoichiometry (UN_2 , U_2N_3 , and UN). Experimental conditions were optimized to synthesize high phase purity UN (97 wt.%). Thermal decomposition of UN_2 under different atmospheric conditions was also studied, and ultra high purity argon could successfully be used to reduce the sample completely to UN at 1100°C, see graph on the opposite page. UN_2 and U_2N_3 decomposition kinetics into UN were studied under argon at three different temperatures (1000, 1050, and 1100°C).

Optical microscopy and SEM were used to explore the morphology of uranium nitride samples. Bright Field Transmission Electron Microscopy was also used to confirm the morphological observations. Microstructural studies of the samples were carried out using high resolution (HR) TEM with the help of selected area diffraction (SAD) patterns. X-ray energy dispersive spectrometry of TEM was utilized to characterize the elemental distribution and to verify the phase purity of the samples. Powder XRD patterns of the as-synthesized uranium nitrides, UN_2 , U_2N_3 , and UN were collected and analyzed. Optical microscopic studies showed that the particle sizes of these uranium nitride samples range from 100 to 5000 nm. The microstructure of the UN sample shows the presence of UO_2 as a secondary phase on the surface of the sample. In this region, the lattice fringes correspond to the (222) interplanar d -spacing of UO_2 . X-ray Energy Dispersive Spectrometry (XEDS) demonstrated that U was prominent, but it is difficult to identify the presence of N due to overlaps with peaks from O and C. However, the magnified XEDS spectra verifies the presence of N in samples, and this figure also displays the presence of O only in the UN sample. Thus, the XEDS verifies the phase purity of the



Scanning Electron Microscopic images of the (a) $7\text{NH}_4\text{F} \cdot 6\text{UF}_4$ and (b) $(\text{NH}_4)_4\text{ThF}_8$ samples. (a) $7\text{NH}_4\text{F} \cdot 6\text{UF}_4$ particles are well-crystallized (hexagonal unit cell with a rhomb-centered, $a(b) = 15.40 \text{ \AA}$ and $c = 10.49 \text{ \AA}$ and UN_2 is cubic (fcc) with $a = 5.310 \text{ \AA}$) (b) Well-crystallized $(\text{NH}_4)_4\text{ThF}_8$ acicular-shaped particles (triclinic unit cell with lattice parameters $a = 8.477$, $b = 8.364$, and $c = 7.308 \text{ \AA}$).

synthesized sample.

Thorium based nitride synthesis and characterization

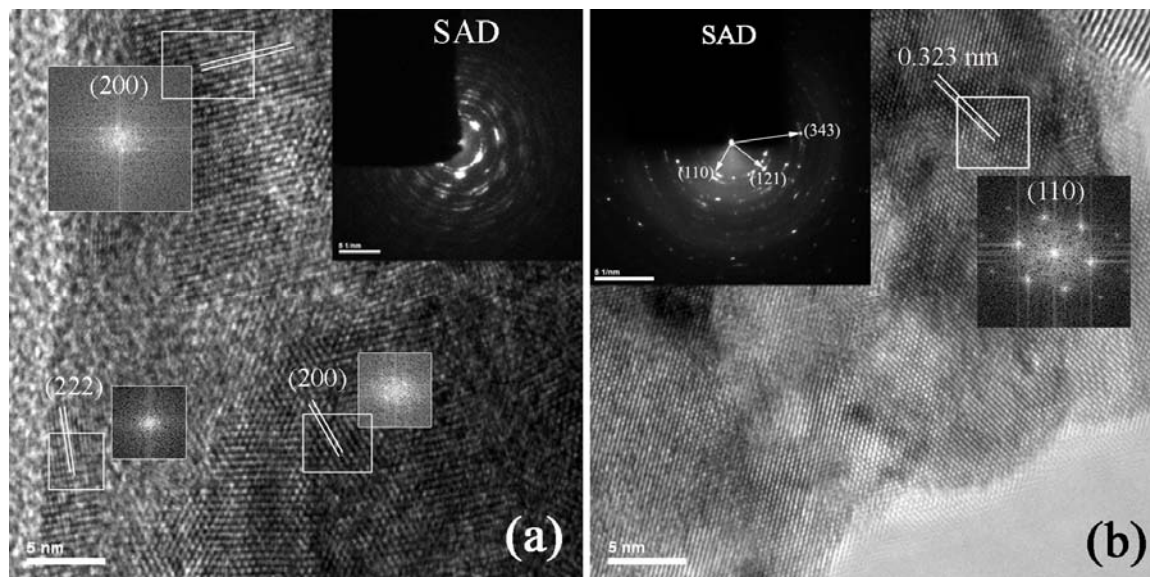
Use of the fluoride route was successful only up to the formation of ThNF. The removal of fluorine, which should have lead to the formation of thorium nitrides was unsuccessful at different experimental conditions. However, the characterization of ammonium thorium fluoride and ThNF was done using the above mentioned techniques.

Other progress

- $(\text{NH}_4)_4\text{NpF}_8$ was found to be formed at room temperature for the reaction of NpO_2 with NH_4HF_2 after two days.
- Polycrystallinity of $(\text{NH}_4)_2\text{NpF}_6$ could be explained by observing the nanostructure of the compound by high-resolution transmission electron microscopy.
- Amorphous characteristics and disturbances in the lattice fringe formation were seen in the nanostructure of $(\text{NH}_4)_2\text{NpF}_6$.
- First NpN sample was synthesized by heating $(\text{NH}_4)_4\text{NpF}_8$ under NH_3 and subsequently under argon.
- The NpN sample was gold/yellow color. A secondary NpO_2 phase was also observed in the sample. DFT calculations of the thermodynamics properties of NpN.

ACADEMIC YEAR HIGHLIGHTS

- ♦ Low temperature synthesis of Np nitride (NpN) confirmed June 2008. First transuranic synthesis of the nitride at UNLV.
- ♦ G.W.C. Silva, C.B. Yeaman, L. Ma, G.S. Cerefice, K.R. Czerwinski, and A.P. Sattelberger, "Microscopic Characterization of Uranium Nitrides Synthesized by Oxidative Ammonolysis of Uranium Tetrafluoride," *Chem. Mater.*, 20(9), 3076-3084, 2008.
- ♦ G.W.C. Silva, F. Poineau, A. Sattelberger, and K.R. Czerwinski, "Low-temperature fluoride route for actinide ceramic synthesis," European Research Materials Society, Strasbourg, France, May 2008.
- ♦ P.F. Weck, E. Kim, N. Balakrishnan, F. Poineau, C.B. Yeaman, and K.R. Czerwinski, "First-principles study of single-crystal uranium mono- and dinitride," *Chemical Physics Letters*, 443(1-3), 82-86, 2007.
- ♦ G.W.C. Silva, C.B. Yeaman, G.S. Cerefice, F. Poineau, and K. Czerwinski, "Evaluation of a low temperature fluoride route to synthesize actinide mononitrides," 234th ACS National Meeting, Boston, MA, August 2007.
- ♦ C. Silva, T. Sullens, P. Weck, F. Poineau, T. Hartmann, C. Yeaman, A. Sattelberger, G. Jarvinen, D. Clark, and K. Czerwinski, "Novel Routes for the Synthesis of Actinide Nitrides," ACE Workshop on AFCI Materials, Idaho State University, Boise, ID, May 2007.



High resolution TEM images of (a) UN and (b) ThNF samples. (a) Crystallography of UN was confirmed using the lattice fringes of HRTEM image, and the secondary oxide phase was only identified at the surface of the particle edge. (b) ThNF crystal structure which is determined using XRD was confirmed by the HRTEM and SAD pattern.

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Task 29

Investigation of Optical Spectroscopy Techniques for On-Line Materials Accountability in the Solvent Extraction Process

G. Cerefice and K. Czerwinski

BACKGROUND

Increasing the proliferation resistance of the nuclear fuel cycle is one of the primary goals for the U.S. Department of Energy's Global Nuclear Energy Partnership (GNEP) program. As part of the GNEP "safeguards-by-design" concept, new separations facilities will incorporate integral systems capable of providing in-line, real-time analyses to verify that the process is being operated as designed (process monitoring) and that fissile material has not been diverted (materials accountability). The goal of this project is to evaluate optical spectroscopy techniques, such as Ultraviolet-Visible Absorbance Spectroscopy (UV/Vis) and Laser Fluorescence Spectroscopy (LFS), to meet these needs.

In UV/Vis spectroscopy, the sample is illuminated by a continuous spectrum (from the UV through the Visible wavelengths). The transmitted light is measured, allowing the determination of the absorbance of the light as a function of wavelength. The wavelength of the absorbance is dependant on the electronic structure of the absorbing atom, and is proportional to the concentration of the absorbing element in the sample. For LFS, the sample is illuminated at a single wavelength, which is absorbed by the target atoms in the sample. The energy absorbed is re-emitted through fluorescence. The wavelength of the absorbance, and the fluorescence-response, is again dependent on the electronic structure of the absorbing atom, and is proportional to the concentration of the absorbing element in the sample.

Both UV/Vis and LFS are quantitative analytical techniques that have been used for measuring the concentration of the actinides under laboratory conditions. These techniques are strongly dependent on the chemical speciation of the elements to be measured, providing a tool for not only the determination of material concentrations for mass balances, but also providing inspectors

and plant operators with a tool to examine the process chemistry itself. As optical techniques, both of these methods can be adapted for fiber optics, allowing the instrumentation to be placed in shielded areas of the plant to minimize the impact of the radiation fields on the detectors and increase the accessibility of the systems for maintenance and inspection.



Laser induced fluorescence of a uranium sample in nitric acid

RESEARCH OBJECTIVES AND METHODS

The goal of this project is to evaluate the application of optical spectroscopy techniques to the on-line, real-time measurement of the actinide elements and "cold" chemicals in the process streams of a solvent extraction process. Based on the experience gained through this effort, engineers will have the information necessary to decide if these technologies should be advanced to the prototype stage and tested at the pilot plant level. Through the experimental work planned as part of this effort, researchers will also develop a better understanding of the chemical interactions of the actinide elements, providing additional data for the development of first-principles based models of the solvent extraction process, expanding current thermochemical databases for the solvent extraction process, particularly in the off-normal operating regimes.

The research objectives are:

- To evaluate the potential for utilizing optical spectroscopy techniques to determine actinide concentrations under process conditions.
- To examine what process chemistry information can be extracted from the spectroscopic signals along with the actinide metal concentrations.
- To examine the fundamental chemistry underlying the spectroscopic behavior under process conditions in support of process chemistry modeling activities.

RESEARCH ACCOMPLISHMENTS

Recent efforts have focused on reducing measurement uncertainty in the determination of uranium concentrations under process conditions and on measuring the nitrate concentration spectroscopically for process monitoring applications. Previous work exploring the application of UV-Vis spectroscopy in the UREX and PUREX separation processes demonstrated that the nitrate concentration in the sample significantly impacted the measured absorption spectra, requiring that any spectrometer system be calibrated for the expected process conditions. Deviations in nitrate concentration from the expected process chemistry result in an increase in uncertainty in the measured uranium concentration. Not only do these deviations in process chemistry increase measurement uncertainty, but they also reveal a systemic bias on the measurement that could be exploitable in a potential diversion scenario.

To address this concern, the research team developed a new analytical method, the Peak Ratio Method (PRM) to simultaneously determine the nitrate concentration and uranium concentration in the system. Taking advantage of modern solid-state detectors to measure the full uranium absorption spectrum (instead of a single wavelength), the PRM utilizes the ratio of two secondary absorbance peaks to determine the nitrate concentration, which is then used to correct the uranium concentration measurement determined from the primary absorbance peak. By simultaneously determining the nitrate concentration and uranium concentration in

the process stream, the systemic bias introduced into the uranium determination by deviations in process chemistry is eliminated and the uncertainty in the measurement itself is reduced.

Through the evaluation of the PRM as process conditions varied, it was observed that the uranium absorbance spectrum was unexpectedly dependant on acid concentration. As proposed chemical models for uranyl ion speciation in nitric acid systems do not include any acid dependant species, this observation further elucidates the need to better understand the fundamental chemistry underlying the solvent extraction processes. While the root cause of this effect still needs to be determined, an empirical relationship has been developed to correct the nitrate concentrations determined by the peak ratio method. Using this correction to the PRM, the bias introduced by acid concentration in the process stream can be greatly reduced. However, it does require the use of some other technique to determine acid concentration in parallel to the UV-Vis system.

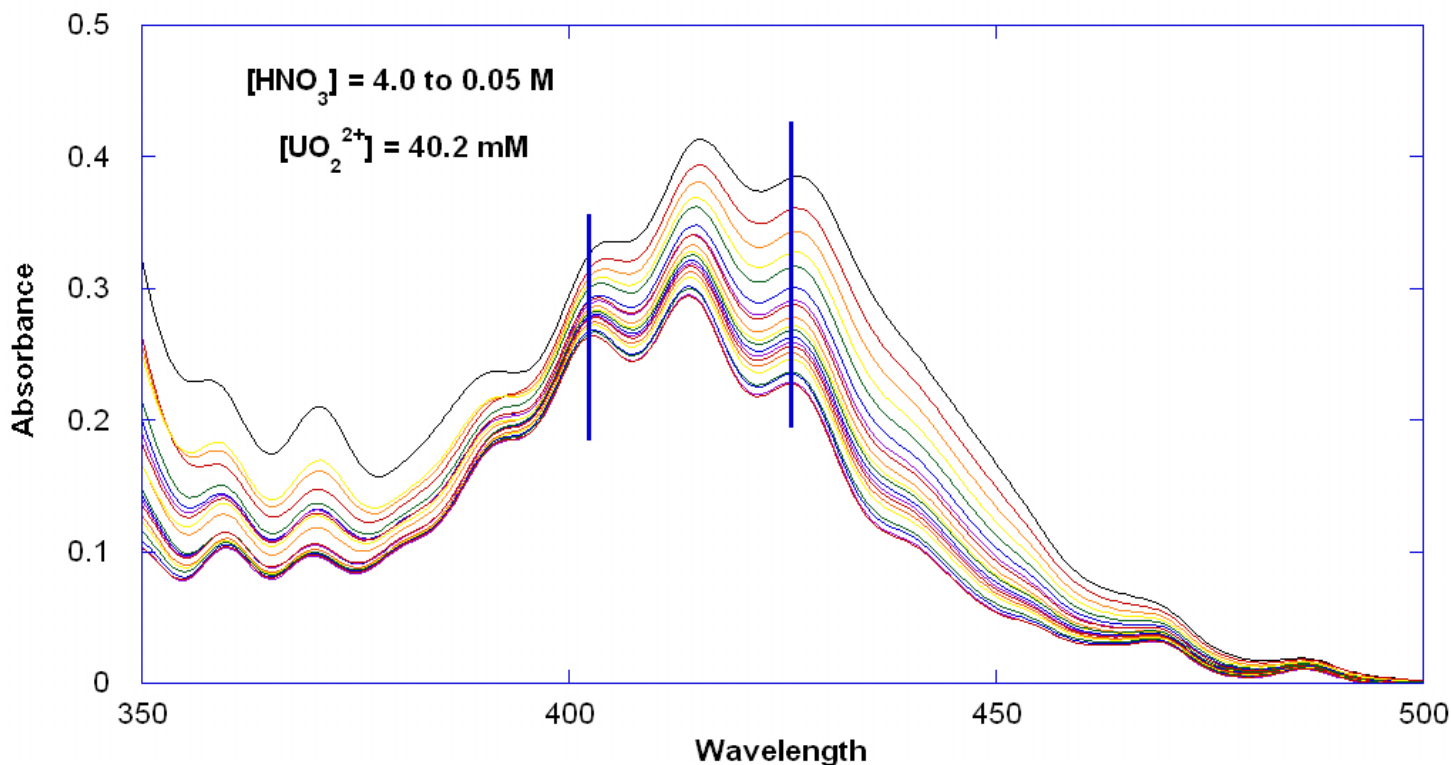
FUTURE WORK

Going forward, researchers on this project will continue to examine the development and adaptation of these optical spectroscopy

ACADEMIC YEAR HIGHLIGHTS

- ◆ N.A. Smith, G.S. Cerefice, and K.R. Czerwinski, "UV-Visible Spectroscopy for Process Monitoring and MC&A," chapter in Bench Scale Experiments for Advanced Safeguards, P. Santi, (editor), prepared for U.S. Department of Energy Safeguards Campaign, March 29, 2008.

techniques for safeguards and process monitoring applications. Continuing efforts are planned to examine the potential for determining the acid concentration directly from the spectral data to try to reduce or eliminate the need for a separate measurement technique. Through collaborations with researchers at Argonne National Laboratory and Oak Ridge National Laboratory, the research team will also examine the extension of these techniques to fiber-optic deployment, including evaluation of the PRM under engineering-scale demonstration conditions. Through these experiments, along with continued laboratory efforts, the potential extension of these optical spectroscopy techniques to the higher actinides (plutonium in particular) will be examined.



Impact of process conditions on uranium absorbance spectrum. Blue lines indicate peaks used for determining nitrate for the Peak Ratio Methodology.

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Task 30

Combined Radiation Detection Methods for Assay of Higher Actinides in Separation Processes

D. Beller, C. Sanders, and W. Kernan

BACKGROUND

Monitoring of higher actinides (HA; includes neptunium, plutonium, americium, and curium) during the separation of used nuclear fuel has been identified as a critical research area in the U.S. Advanced Fuel Cycle Initiative. Recycling of used fuel by chemically separating it into uranium, fission products, and HA would be the first step in this new fuel cycle. Material Protection, Accounting, and Control (MPAC) is necessary for materials accounting, criticality monitoring, and assurance of proliferation resistance. The objective of this MPAC project is to develop technology to detect and accurately measure quantities of higher actinides in used fuel assemblies and processing systems without taking frequent samples. Process systems may include separations batches, pipelines, storage tanks, and fuel fabrication equipment. A variety of measurements may be combined to calculate flow rates of actinide elements with a to-be-determined precision.

RESEARCH OBJECTIVES AND METHODS

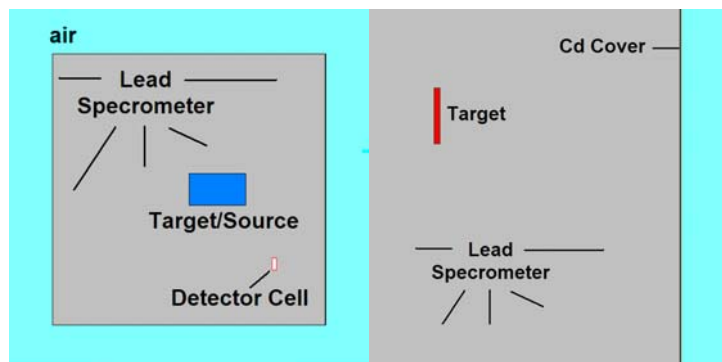
In the MPAC project, faculty and students are investigating the potential to use combined neutron and gamma-ray detector systems to measure quantities and isotopic constituents contained during separations and intermediate storage. This will require knowledge of the nuclear and decay characteristics of materials during processing, the development of conceptual designs of monitoring systems, radiation transport studies to develop an understanding of operational regimes, and experiments to confirm performance. In addition, both passive and active concepts will be investigated, including collaborations with the Idaho Accelerator Center at Idaho State University (ISU) to use electron linear accelerators for producing photoneutrons in situ, for photon activation of HA, or for stimulating emissions processes (e.g., X-ray fluorescence).

Radiation transport and scoping studies will be conducted to investigate combined gamma-ray, neutron, and active and passive detection techniques to measure quantities and isotopic constituents contained during separations and intermediate storage. Scoping and design studies will first be performed using validated data sets (decay properties and reaction cross sections) and the radiation transport code MCNPX. Basic measurements will then be performed and compared to predictions. Planned experiments have not been conducted.

RESEARCH ACCOMPLISHMENTS

Lead and Carbon Slowing Down Spectrometers

Lead Slowing Down Spectrometer (SDS) and Carbon SDS (CSDS) were modeled. This modeling was expanded to include the UNLV ^3He Neutron Multiplicity Detector System (NMDS), which was developed as part of Task 6) with multiple neutron detectors in up to four layers surrounding used nuclear fuel rods or assemblies. Modeling also included simulation of decay gamma



Cross sections of the UNLV benchmark of the LANL lead SDS taken from MXNPX models. (left) The overall geometry, (right) A close-up of a cross section of the thin target. The response to neutrons of various energies (time constants) is influenced by the position and isotopic composition of the detector.

rays for coincidence counting investigations.

In preparation for future experiments, MCNPX was used to model neutron transport characteristics in LSDS and CSDS. However, the detector system must be able to resolve the time dependence of the neutron signal. Because ^3He detectors, such as those contained in the NMDS, have a slow response, SDS configurations must be studied before conducting experiments. A Los Alamos National Laboratory (LANL) LSDS was modeled to benchmark computational methods for determining energy-time correlation constants. The benchmark exercise was completed and published. Multiplicity measurements for spontaneous fission and spontaneous-fission induced fission for isotopes of U, Pu, and Cf were verified within Monte Carlo statistical uncertainty limits. Simulations were conducted for other isotopes (Am, Np, and others) for multiplicity measurement experiments using a subset of the NMDS detectors.

In future work, the MCNPX code will be used to design an experiment using the ISU CSDS and electron linac, followed by experiments conducted at ISU. During this period, plans will be developed to conduct experiments at ISU with the CSDS to develop technology for assaying fuel rods and/or assemblies.

Neutron Multiplicity Detector System

Collaboration with the V.G. Khlopin Radium Institute (KRI) was developed for an upgrade to and maintenance on the Neutron Multiplicity Detector System. KRI completed design studies for a coincidence/anti-coincidence using a multi-plate plastic scintillator system to reduce background and provide greater neutron-muon discrimination. KRI procured components for the upgrade, installed them and coupled outputs to their NMDS channels, and began measurements in St. Petersburg, Russia.

In preparation for the KRI visit, the NMDS was reconnected to the original Russian data acquisition system and several neutron multiplicity counts were conducted to confirm its performance. The

data files were transmitted to KRI where they were evaluated. All detectors appear to be operating as they were when the system was initially delivered and set up at UNLV several years ago. Two specialists from KRI visited UNLV in November to service the NMDS; they cleaned and secured connections, replaced some parts, and adjusted bias voltages on some detectors.

A commercial control program was installed to automate the collection and storage of data on the 60-channel NMDS. It will automatically take data and has operated reliably since this upgrade.

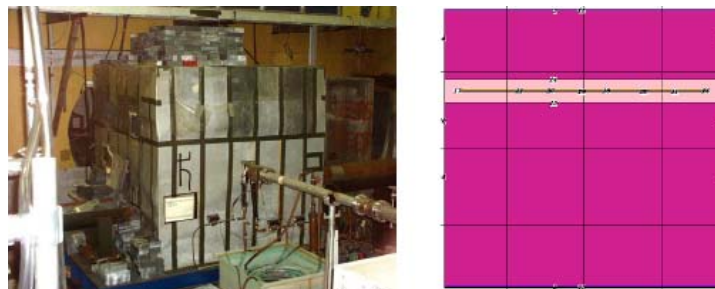
Rensselaer Polytechnic Institute collaboration

Rensselaer Polytechnic Institute (RPI) collaborators investigated neutron slowing down spectroscopy for quantitative analysis of ^{239}Pu , ^{235}U and possibly ^{241}Pu in used fuel assemblies or rods. Several fuels, configurations, and parametric studies were modeled.

RPI completed various parametric studies to compare computed response to measurements in the LSDS. First order calculations were conducted with one fuel pin based on data from an AP1000 reactor. A detector was placed in the assay channel of the lead and the tally was convoluted with the ^{235}U fission cross section. This data was compared to experimental fission data for ^{235}U collected in the LSDS in 2007. Differences between calculated and experimental data are due to the broadening of the neutron energy resolution at lower neutron energies. Also, the reaction rate decreases as the slowing-down time increases, so that background becomes a larger factor as the neutron energy decreases.

Other RPI results:

- Simulation of ^{235}U fission chamber response was compared to measured data indicating that hydrogen content and impurities in the lead must be included to accurately simulate the measured response.



RPI's LSDS (left), and a cross section at $x = -37.5\text{cm}$ showing the assay channel and fuel pin (the x axis is positive along the beam line).

ACADEMIC YEAR HIGHLIGHTS

- ♦ A.A. Rimsky-Korsakov and N.A. Kudryashev, "General Upgrade Design, Instrumentation Upgrade Of Neutron Multiplicity Detector System," Progress Report 1, V.G. Khlopin Radium Institute, St. Petersburg, Russia, September 2007.
- ♦ A.A. Rimsky-Korsakov and N.A. Kudryashev, "Detailed Equipment Specifications and List Of Materials and Components to be Purchased, Instrumentation Upgrade Of Neutron Multiplicity Detector System," Progress Report 2, V.G. Khlopin Radium Institute, St. Petersburg, Russia, September 2007.
- ♦ Timothy Beller graduated with an M.S. in Materials and Nuclear Engineering, December 2007.
- ♦ Ryan LeCounte graduated with an M.S. in Materials and Nuclear Engineering, December 2007.
- ♦ Lawrence Lakeotes graduated with an M.S in Materials and Nuclear Engineering, May 2008.
- ♦ Sandra De La Cruz graduated with a B.S.M.E. (nuclear option), May 2008.
- ♦ Luis Durani graduated with a B.S.M.E. (nuclear option), May 2008.

- Simulation of a single SPERT fuel pin and a ^{238}U assay detector were performed.
- Analysis of optimization and biasing methods for LSDS calculations demonstrated that, due to the nature of the LSDS, all areas of the lead as well as all neutron energies are critical to the problem and that even biasing the source will affect the resulting spectrum in the ^{238}U detectors.
- A model of an AP1000 fuel assembly was completed to study effects of self shielding within the assembly. These studies demonstrated that MCNP can produce results that agree with measurements of time-dependent fission rates in the LSDS; however, great effort must continue to be made to optimize the calculations. Particular effort must be made to increase efficiency to allow for smaller statistical error with a minimum of calculation time.

FUTURE WORK

UNLV is discussing follow-on research with Pacific Northwest National Laboratory, Idaho National Laboratory, LANL, ISU, and RPI. Current work includes continuation of studies at RPI as well as an undergraduate research internship at LANL during the summer 2008 to model fuel assemblies.

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 James Laidler, Argonne National Laboratory
 Thomas Ward, UNLV Science Adviser for Russian Collaborations, TechSource, Inc.
 Yaron Danon, Professor of Nuclear Engineering, and Catherine Romano, Graduate Student, Aerospace and Nuclear Engineering, Rensselaer Polytechnic Institute

Task 31

Decoupling and Disturbance Rejection Control for Target Circulation

J. Ma, J. Lee, and W. Yim

BACKGROUND

The Target Complex loop TC-1 was originally conceived as part of an accelerator-driven system (ADS) pilot plant that was designed and developed by the Institute of Physics and Power Engineering (IPPE) and Experimental and Development Organization (EDO) “Gidropress” in Obninsk, Russia, under the International Science and Technology Center Project #559 in 1998. It was to be used as the target in a 1 MW_{th} ADS experiment run off of the LANSCE proton accelerator at Los Alamos National Laboratory (LANL). When the U.S. transmutation program changed priorities from accelerator-driven systems towards nuclear fission reactors, the TC-1 loop was brought to UNLV to be developed as an academic research tool.

Liquid lead-bismuth eutectic (LBE) is employed as a spallation target, as well as a heat transfer fluid or coolant, in the TC-1 loop. The TC-1 loop can play a role as a testing facility in the U.S. to support research in heavy liquid metal coolant for the nuclear industry. During a thermal and engineering test of the TC-1 loop in 2005 at UNLV, it was observed that the existing control algorithm led to a very slow convergence to the target temperature setting, and also showed unstable oscillatory behavior. The existing algorithm was not robust enough to handle the complicated heating system of the TC-1 loop, where nine heating zones or elements are compacted in one tight container. This interaction and coupling between each heating zone, as well as a heat disturbance from a low efficiency electromagnetic (EM) pump, caused the overall temperature control system to be complex and nonlinear.

RESEARCH OBJECTIVES AND METHODS

The primary objective of this task was to study and modify the coupling effect between heating zones and the existing control algorithm to achieve precise temperature control in the TC-1 loop system. Safety concerns, the alarm system, and a user-friendly design became the secondary objective.

The TC-1 loop system has more than a single input and a single output, and it exhibits a nonlinear interactive property between the heater inputs and temperature outputs. For effective temperature control in multiple locations of the TC-1 loop, these nonlinear interaction terms must be eliminated (decoupled) in the control loop. Eliminating these interaction terms requires the identifica-

tion of these interacting (coupling) terms. After successful elimination of the coupling terms, a closed loop control algorithm can be designed which can achieve the precise tracking of the temperature on multiple locations of the TC-1 loop under external temperature disturbance from an electromagnetic pump. One example of such an algorithm is a Proportional-Integral-Derivative (PID) control law. This law can easily be implemented within the existing LabView codes of the Monitoring, Controlling, and Scram Protection System (MCSPS).

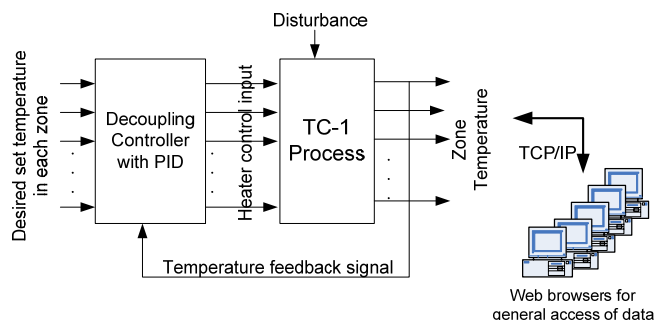
In addition, the electromagnetic pump, used for molten lead circulation, becomes a large heat source. This, in particular, is due to its low efficiency. A “disturbance observer”-based control method was used to compensate modeling uncertainties as well as external disturbance. The disturbance observer regards the difference between the actual output and the output of the nominal model as an equivalent disturbance applied to the nominal model. The disturbance observer based control algorithm achieves a precise tracking of set temperatures despite the highly coupled thermal disturbance existing in the loop. Finally, the alarm system and a 24-hour monitoring and dial-out system was designed.

RESEARCH ACCOMPLISHMENTS

The interacting terms between heater inputs and target temperature outputs in each zone were identified experimentally. These identified terms were then expressed in a discrete transfer function matrix. The system identification was carried out by heating up one zone from room temperature to 50 °C, while keeping others off. 50 °C was selected to avoid large temperature differences between zones. The transfer functions, which are used to describe dynamic response between individual inputs and outputs, were identified. One non-interacting (decoupling) control algorithm, based on the identified model, was developed to reduce the influences from each zone.

Significant improvement in the controller performance was achieved by upgrading the existing controlling algorithm. The heaters of all heating zones were well controlled to maintain the temperature of all zones within the desired range. The difference between maximum and minimum was only about 5 °C. In the previous algorithm, the temperatures of different zones were not regulated well at the desired levels. It is desired that the temperature of all zones should be controlled within a range of 190 to 200 °C. However, the temperature difference between all heating zones was about 80 °C. This exceeds the safety range and has a high risk to cause thermal inhomogeneity.

Wired to the main program, the 24-hour monitor device can automatically dial out when a TC-1 loop temperature is too high or there is abnormal current passing through EM pump. In addition, if the communication between computer and watch-dog is not connected, the alert signal will be sent out. The current transformers were assembled to detect actual heater on/off status. Signals from these current transformers were regulated and recorded by the data acquisition system for investigation.



Overall Control Block Diagram of the TC-1 Loop System.

An external cooling system was developed and installed on the TC-1 loop. The monitoring system for cooling fluid flow rate, temperature, and pressure was coded in the main program to make this main system sophisticated. The improved program will help to find the optimized parameter for the pump to continuously operate the TC-1 loop for extended periods of time.

TASK 31 PROFILE

Start Date: September 2006

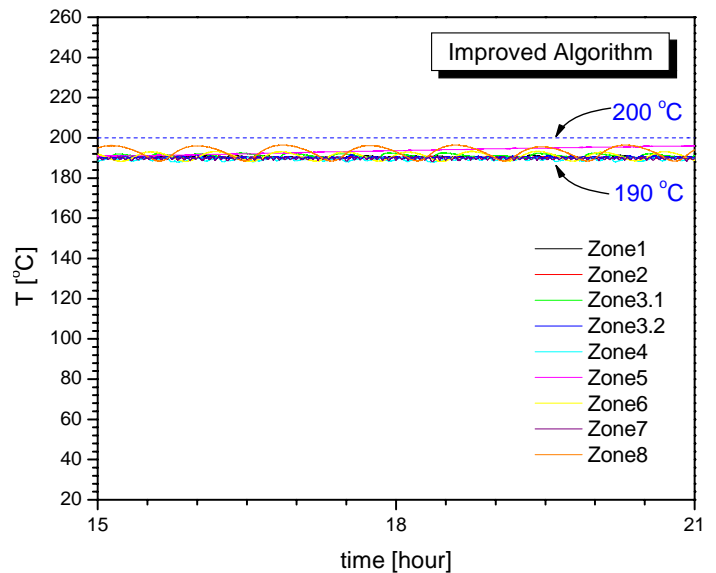
Completion Date: December 2007

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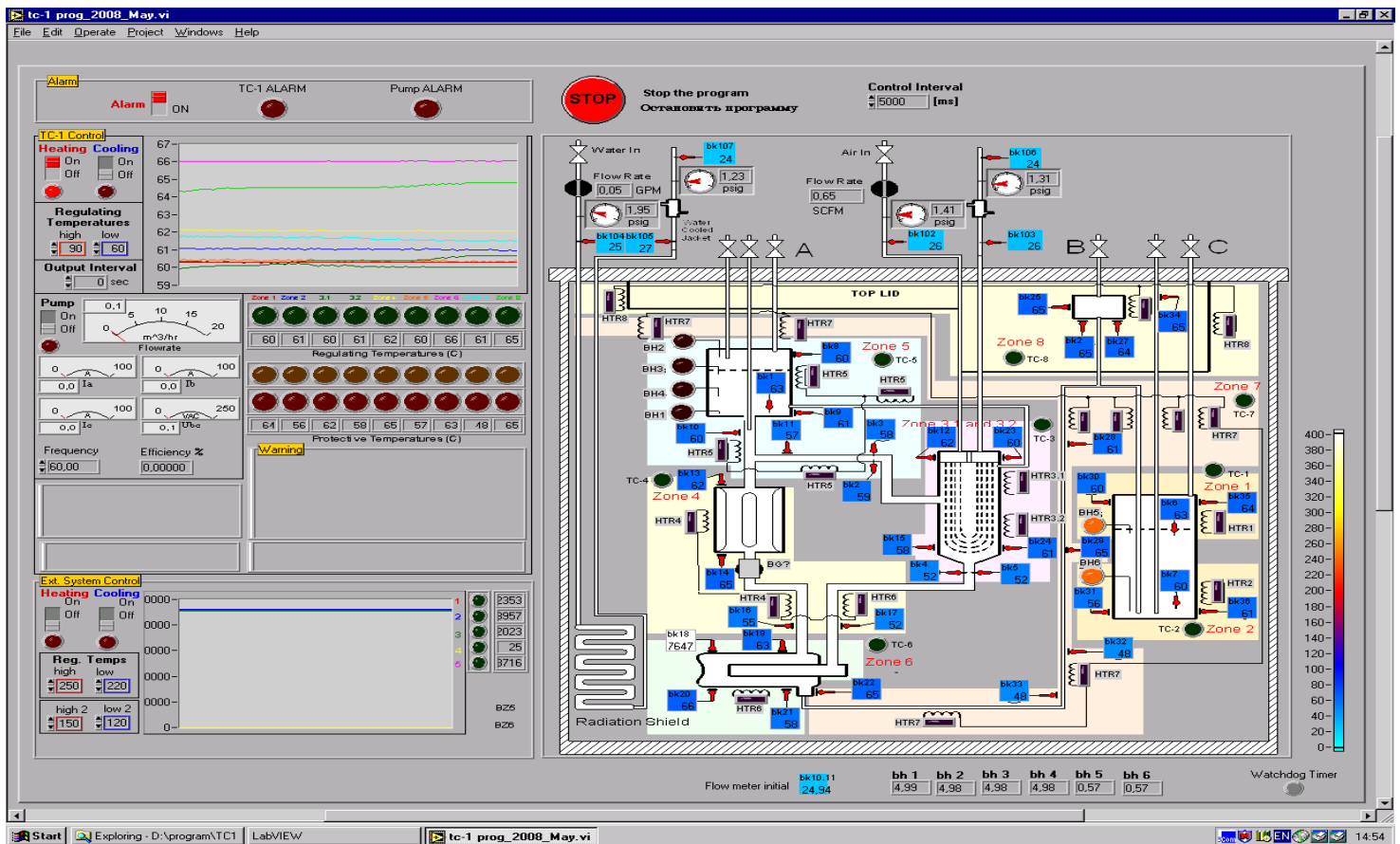
Xiuju (Julia) Tan, M.S. thesis, Mechanical Engineering Department, "Thermal Control for the Liquid metal Coolant Circulation Loop," December 2007.

Conference Proceedings:

X. Tan, J.S. Lee, J. Ma, and W. Yim, "Disturbance Observer based Thermal Hydraulic Control of the Lead Bismuth Eutectic Loop," Eighth International Topical Meeting on Nuclear Application and Utilization of Accelerators (AccApp 07), Pocatello, ID, July 30-August 2, 2007.



Temperature response of nine heating zones. As obtained by the new algorithm.



Interface of Control System for TC-1.

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Joon Soo Lee, Assistant Research Professor, Mechanical Engineering Department
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Task 32

Modeling and Design Algorithms for Electromagnetic Pumps

D.P. Cook, Y. Chen, and J. Ma

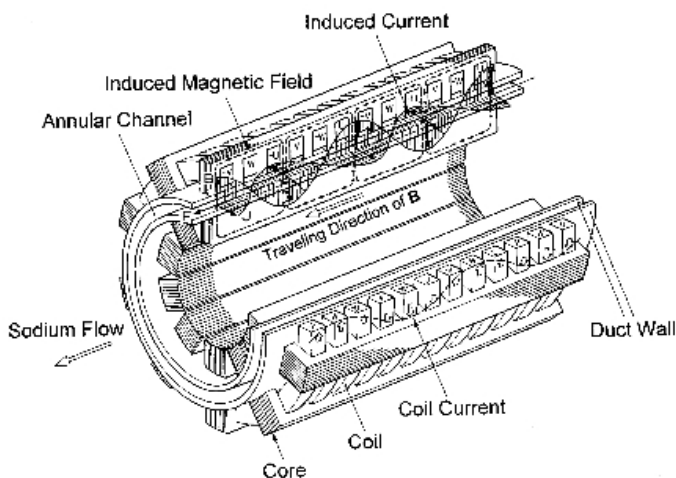
BACKGROUND

Electromagnetic (EM) induction pumps are used extensively in current and proposed nuclear power systems and industrial molten metal transfer operations. Although the Magnetohydrodynamic (MHD) theory that underlies the operation of these types of pumps has been studied extensively in the past few decades, the design of specific EM pumping systems for specific flow cases requires computational tools and expertise, which is lacking in the U.S. In recent years, the majority of research done on these systems has been done in Japan, Korea, France, and the former Soviet-bloc countries. However, for the past two years, researchers at UNLV have been utilizing the TC-1 liquid metal loop system at UNLV and an Annular Linear Induction Pump (ALIP) to drive the liquid metal and to develop such computational tools that will allow the accurate and efficient optimization of EM pump systems for nuclear applications.

A cut-away picture of an ALIP such as has been used in prototype Sodium Fast Reactors and the TC-1 loop at UNLV is illustrated below. These ALIPs consist of three main parts:

- An inner cylindrical core fabricated from a ferromagnetic material
- An annular channel through which the liquid sodium flows
- An outer ferromagnetic core in which a set of inductor coils are embedded

During operation, a 3-phase, alternating current travels through the inductor coils. This current produces a magnetic field which, in turn induces a current in the liquid sodium in the pump annulus and inner core. Pumping forces develop in the liquid sodium due to the interaction of the magnetic field and the induced current, causing the liquid sodium to flow down the length of the annulus. The magnitude of these pumping forces, and the operational efficiency of the pump, is dependent on a large number of design pa-



Cut-away picture of an annular, linear induction pump (ALIP).



TC-1 Loop laboratory located at UNLV.

rameters including coil current and position, material selection for the inner and outer cores, and size of the annular gap.

RESEARCH OBJECTIVES AND METHODS

The research objectives of this task are:

- A literature review of topics pertinent to EM pump design. These topics include the equations governing the physical phenomena occurring in EM pumps and mathematical algorithms used in modeling these physical phenomena, different EM pump configurations, and the effects of materials properties on pump performance.
- Development of computational models of the TC-1 loop at UNLV.
- Evaluation of the computational models through comparison with experimental data taken on the TC-1 loop.
- A parametric study of the TC-1 loop investigating the pumping efficiency as a function of operating conditions, materials properties, and geometric parameters.

RESEARCH ACCOMPLISHMENTS

On-line EM pump literature database

The on-line literature database has been available for over a year and now contains over 140 entries. New entries are added on a regular basis by the various researchers working on the project. Currently, this database can be found at:

<http://nstg.nevada.edu/mmrg/research/LitSurvey/EMP-Literature.html>.

Calculations of EM phenomena and pump efficiency

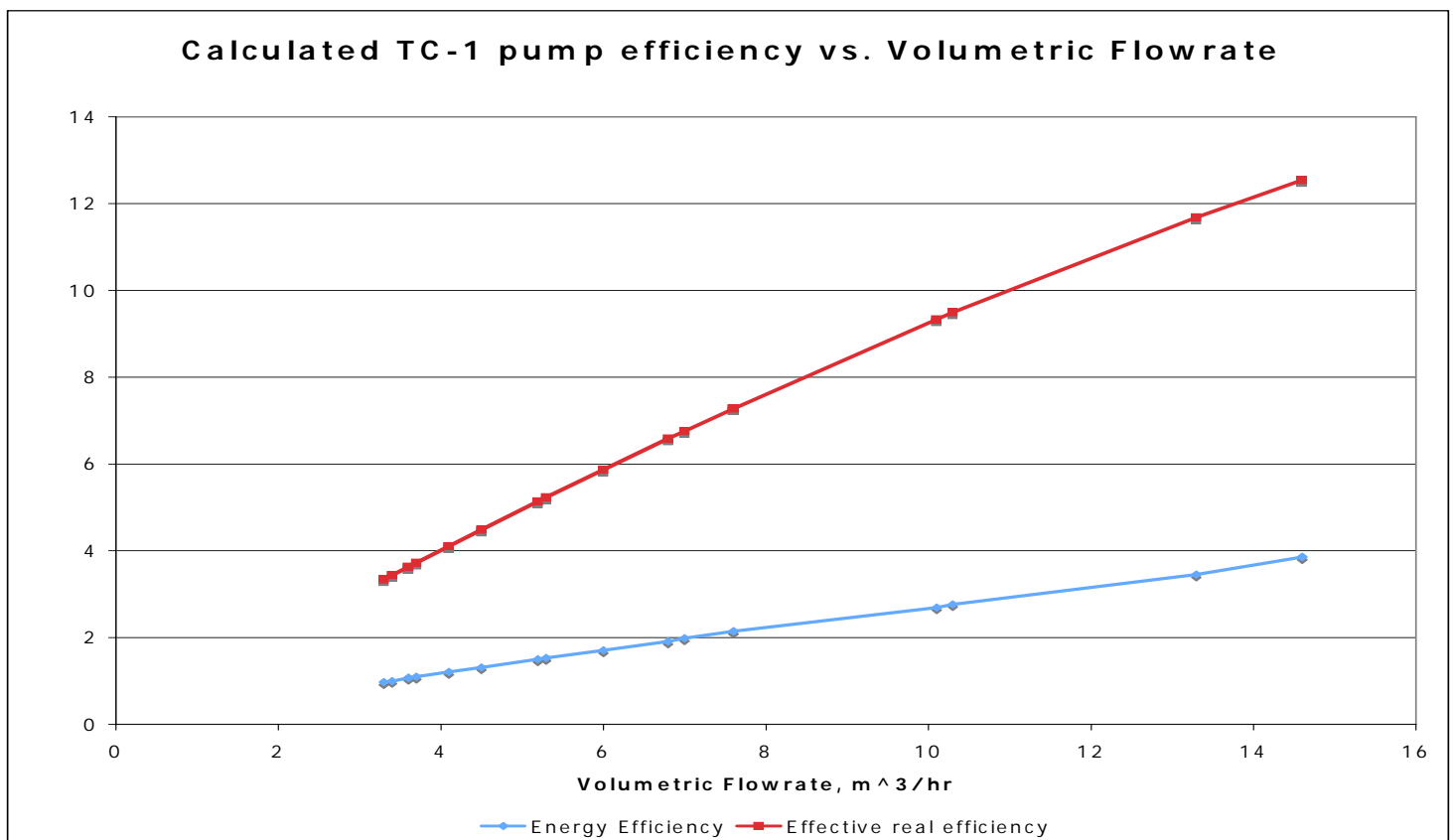
An analytic model of the EM phenomena in a generic ALIP has been developed using the method of separation of variables and Fourier transforms applied to the magnetic vector potential form of the induction equation. The model is written in Fortran and consists of a library of functions that calculate the various EM phenomena (magnetic vector potential, magnetic field components, Lorentz forces, induced current density distribution, etc.).

While working through these results, some of the earlier results from this project were revisited, in particular, the calculated values for the TC-1 pump efficiency. There are several ways to calculate the efficiency for an EM pump. One way, often referred to as the energy efficiency of the pump, is to divide the energy of the flowing fluid, i.e., pressure times flow-rate, by the energy it takes to run the pump, i.e., voltage times current. A second way to calculate the efficiency of an EM pump, often termed the effective real

efficiency, basically relates the speed of the fluid to the speed of the traveling magnetic field. Both of these types of efficiency are illustrated below as a function of mass flow rate through the TC-1 loop. It should be noted that previously calculated efficiencies were always less than 1%, whereas the new calculations are all significantly above 1%. This discrepancy needs to be explored further and clarified.

FUTURE WORK

The next phase of the project involves development of a fully-coupled MHD solver for calculating both the EM and fluid flow phenomena in the EM pump. For this model, the EM field phenomena will be calculated using an integral formulation of the Maxwell's equations, and the fluid flow phenomena will be calculated using a finite volume formulation of the Navier-Stokes equations.



Calculated efficiencies for TC-1 loop pump.

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Collaborator

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Task 33

Separation of Technetium from Uranium and Waste Form Synthesis

K. Czerwinski

BACKGROUND

In the Advanced Fuel Cycle Research & Development (AFC R&D) activities, the uranium extraction (UREX+1) process is proposed as one of the most promising technique to separate transuranic elements (TRU) from light water reactor spent nuclear fuel in the years to come. The isotope ^{99}Tc will be separated together with U within the first process steps. After the separation of U, Tc must be immobilized by their incorporation in a suitable storage and waste form.

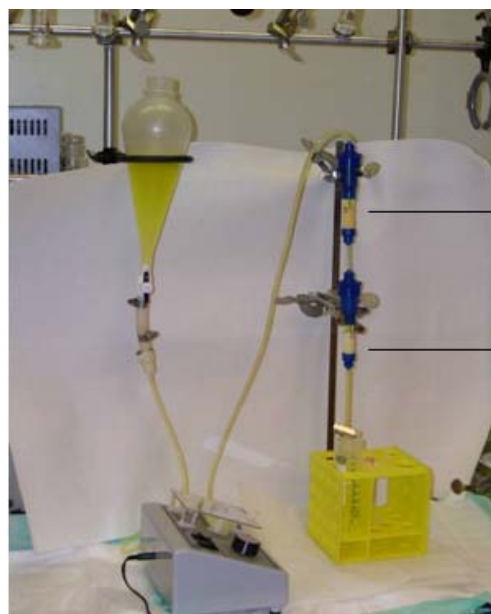
A candidate process to immobilize ^{99}Tc is to alloy metallic Tc with excess metallic zirconium. This material has potential advantages in terms of the future reuse of ^{99}Tc and its potential transmutation. Providing a Tc storage/waste form strongly promotes the AFC R&D and the separation of TRU elements using the UREX+1 process. However, little thermodynamic data in the binary technetium–zirconium metal system exist, and only few data are available on the synthesis of Tc-Zr alloys and on their potential performance under temporary or geological storage conditions.

In this project, systematic investigations on the Tc-Zr binary metal system will be evaluated for the first time. The synthesis of metallic Tc as well as its alloys with Zr will be evaluated. In order to provide valuable data to AFC R&D, the thermodynamic equilibrium phases, as well as their performance under repository conditions, will be examined.

RESEARCH OBJECTIVES AND METHODS

The research objectives of this project are as follows:

- Evaluate anion exchange methods for achieving the separation of Tc from U.
- Synthesize metallic Tc from the separated product.
- Synthesize and characterize Tc alloys.



Separation process with two columns.

- Investigate Tc-corrosion and Tc-leaching of binary Tc-Zr phases under a range of conditions.

The following experimental techniques are used in the evaluation of the solutions and solids from the experiments: ultraviolet-visible spectroscopy, time-resolved laser fluorescence spectroscopy, X-ray Absorption Fine-Structure Spectroscopy (XAFS), and microscopy.

RESEARCH ACCOMPLISHMENTS

Separations

Static and dynamic experiments were performed to investigate the interaction of technetium with selected anion exchange resins. The kinetic static experiments showed Reillex resin had superior Tc sorption kinetics and was selected for further study. Reillex treated by nitric acid (Resin 6) was also used in the studies.

Dynamics experiments were performed on simulated UREX solution in order to determine the separation factor for uranium and technetium and to study the possibility of a Tc elution from the resin. The one column experimental set-up consisted of a reservoir of 250 mL, linked to a peristaltic pump, with a constant flux which can be between 1.2 to 8.4 mL/min. It is connected to a small column (length = 5 cm and diameter = 1 cm) and contained 1 g of resin (Reillex HP or Resin 6).

The process is divided into 3 steps: absorption, washing, and elution. For absorption, 150 mL of the UREX solution were placed in the reservoir and pumped with a peristaltic pump through the column at a constant flow rate (2, 4 or 8 mL/min). Samples were collected into centrifugation tubes (45 mL \pm 0.5 mL for the first two and 10 mL \pm 0.2 mL for the others). For each sample, 10 μL of solution were removed and mixed with 10 mL of liquid scintillation fluid in a plastic vial, and ^{99}Tc was counted by liquid scintillation. The samples were eluted with a 50 mL solution of NH_4OH (1M or 2 M) and were introduced in the reservoir and poured through the column with the same constant flow rate.

Samples were collected into centrifugation tubes of 10 mL \pm 0.2 mL with 10 μL of samples were removed and mixed with 10 mL of liquid scintillation fluid for ^{99}Tc analysis. For samples containing a high amount of technetium, a dilution (1/100) was necessary for the liquid scintillation analysis in order to be able to use the calibration curve. To optimize U/Tc separation, treated and untreated resin were combined. The set-up is composed as before by a reservoir, a pump and two columns.

The absorption and washing steps were carried out with exactly the same conditions, as presented above.

Concerning the elution step, the columns are disconnected and only the first one containing the treated resin is eluted, whereas the second one is kept for a re-use and for an eventual Tc reprocessing by pyrolysis. A suitably pure Tc and U stream resulted from the experiment.



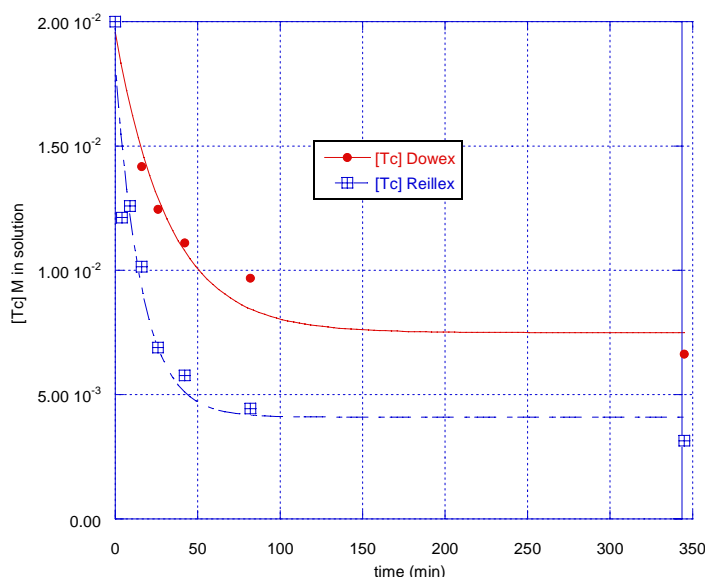
Tc/Zr sample before and after arc melting.

Solid phase synthesis

Steam reforming of pertechnetate in the presence of carbon yielded Tc metal at 700°C under Ar or N₂. Alloy samples were prepared by grinding Tc and Zr metal in different ratios, from 25 to 75 % Tc. Cylindrical pellets were obtained by pressing the powder and arc melting to produce the alloys. After arc melting, samples were analyzed by X-ray diffraction and optical microscopy.

Other progress

- Sm₂Tc₂O₇ oxide was prepared. This was the first of a series for lanthanide-Tc oxide waste form.
- 2 % Tc electrode studies were completed and evaluated.
- Studies were conducted on Tc/Nd /Sm ternary oxide formations. The compounds targeted were (RE)₂Tc₂O₇ (RE = Nd, Sm). Mainly unreacted TcO₂ and Sm₂O₃ was observed.



Kinetics of TcO₄⁻ removal from solution with initial [TcO₄⁻]=0.02 M by Dowex Marathon and Reillex resins.

ACADEMIC YEAR HIGHLIGHTS

- ♦ E.E. Rodriguez, F. Poineau, A. Llobet, K. Czerwinski, R. Seshadri, and A.K. Cheetham, "Preparation and Crystal Structures of Bismuth Technetates: A New Metal Oxide System," *Inorg. Chem.* 47(14), 6281-6288, 2008.
- ♦ F. Poineau, A.P. Sattelberger, and K.R. Czerwinski, "XAFS spectroscopic study of Tc₂(O₂CCH₃)₄X₂ (X = Cl, Br)," *J. Coord. Chem.*, 61(15), 2356-2370, 2008.
- ♦ F. Poineau, A.P. Sattelberger, S.D. Conradson, and K.R. Czerwinski, "Octachloro- and Octabromoditechnetate(III) and Their Rhenium(III) Congeners," *Inorganic Chemistry*, 47(6), 1991-1999, 2008.
- ♦ P.F. Weck, F. Poineau, T. Hartmann, E. Kim, G. Jarvinen, and K.R. Czerwinski, "Combined experimental and theoretical study of Tc-Zr alloys as possible waste form," European Research Materials Society, Strasbourg, France, May 2008.
- ♦ F. Poineau, E. Mausolf, and K.R. Czerwinski, "Electrochemical Behavior of Metallic Technetium in Aqueous Solution," Atlanta 2008, Nuclear Fuel Cycles for a Sustainable Future, Montpellier, France, May 2008.
- ♦ E. Mausolf, C.S. Gong, J. du Bouchaud du Mazabrun, F. Poineau, D.K. Ford, G. Jarvinen, K.R. Czerwinski, "Elution of technetium from anion exchange resins," 235th ACS National Meeting, New Orleans, LA, April 2008.
- ♦ E.E. Rodriguez, F. Poineau, A. Llobet, K. Czerwinski, and A.K. Cheetham, "Synthesis of new ternary technetium oxides," 235th ACS National Meeting, New Orleans, LA, April 2008.
- ♦ F. Poineau, E.E. Rodriguez, A.P. Sattelberger, and K. Czerwinski, "Synthetic chemistry of high valence technetium oxides," 235th ACS National Meeting, New Orleans, LA, April 2008.
- ♦ F. Poineau, A.P. Sattelberger, and K. Czerwinski, "Technetium Chemistry: Fundamental Studies and Applications to the Fuel Cycle," Cadarache, France, September 2007.
- ♦ F. Poineau, G. Jarvinen, and K. Czerwinski, "Synthesis and Behavior of Metallic Technetium Waste Forms," MIGRATION 07, Munich, Germany, August 2007.
- ♦ F. Poineau, G. Jarvinen, and K. Czerwinski, "Synthesis and Characterization of Technetium Waste Forms," Idaho National Laboratory, June 2007.
- ♦ J. Du Mazabrun, F. Poineau, K. Czerwinski, G. Jarvinen, and D. Ford, "Separation uranium/technetium for the UREX process," 62nd Northwest Regional Meeting of the American Chemical Society, Boise, ID, June 2007.
- ♦ G. Jarvinen, D. Ford, K. Long, K. Czerwinski, F. Poineau, and J. Du Mazabrun, "Technetium Recovery and Conversion from the UREX Process Stream using Anion Exchange," Actinide Separations Conference, Las Vegas, NV, June 2007.
- ♦ J. Du Mazabrun, F. Poineau, K. Czerwinski, G. Jarvinen, D. Ford, and K. Long, "Separation and Recovery of Technetium in the UREX process," Actinide Separations Conference, Las Vegas, NV, June 2007.

Research Staff

Ken Czerwinski, Principal Investigator, Associate Professor, Department of Chemistry
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 Philippe Weck, Computational Chemist, Department of Chemistry
 Cynthia Gong and Frederic Poineau, Radiochemistry Post-Doctoral Researchers, Harry Reid Center for Environmental Studies

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Collaborators

Gordon Jarvinen and Doris Ford, Los Alamos National Laboratory
 Al Sattelberger, Argonne National Laboratory

Task 34

Solution-Based Synthesis of Nitride Fuels

K.R. Czerwinski and T.A. Sullens

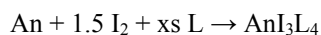
BACKGROUND

Nitride fuels have appropriate properties for advanced fuels, including high thermal conductivity, thermal stability, solid-state solubility of actinides, fissile metal density, and suitable neutronic properties. The presence of oxide and carbide impurities that occur during the current nitride fuel synthesis limits their utility in advanced fuel cycles. The presence of these impurities can lead to the formation of phases with different properties from the bulk material, including the formation of secondary phases. Even a small amount of oxygen in the nitride fuel can reduce thermal conductivity by 10%. This limits the ability to fully develop nitride fuels for advanced reactors. To avoid the entrainment of carbides and oxides in the final actinide nitride product, a non-aqueous, carbon free solution phase route is proposed and investigated in this project.

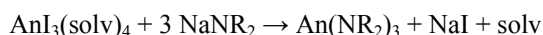
A drawback of nitride fuels involves their synthesis. For nitride fuels, the current synthetic route is carbothermic reduction from the oxide to the nitride, which is based on solid phase reactions involving a stepwise process from the metal oxide, to the carbide, and finally the nitride. This high temperature, solid-phase approach is plagued by impurities in the final nitride product and difficulties in the synthesis and fabrication steps. A direct solution-based synthesis would eliminate, or at least minimize, the impurities and other synthetic problems. The proposed solution route to nitride would also have the added benefit of providing several adjustable parameters that would allow control of the properties of the final solid product.

RESEARCH OBJECTIVES AND METHODS

The objective of this project is to develop solution phase synthetic routes for actinide nitrides for use in nuclear fuels. In the 1970s, a synthesis for actinide nitrides was proposed based on the reaction of plutonium triiodide (PuI_3) and uranium tetraiodide (UI_4) with sodium metal in liquid ammonia giving PuN and UN , respectively, as the reaction products. The synthetic chemistry of trivalent actinide complexes was advanced greatly with the discovery at Los Alamos National Laboratory (LANL) of efficient routes to soluble iodide starting materials of the type AnI_3L_4 , where An is uranium, neptunium, or plutonium, and L is tetrahydrofuran (thf), pyridine, dimethylsulfoxide, etc.

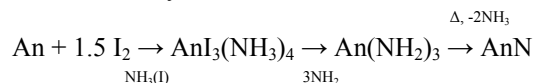


PuI_3 is soluble in ammonia, presumably as the ammonia adduct $\text{PuI}_3(\text{NH}_3)_4$. More recent efforts by ANL and LANL collaborators have investigated amido reactions with actinides to produce $\text{An}(\text{NR}_2)_3$ complexes, where An = U, Np, Pu:



These results suggest that the amido complexes could provide a non-aqueous solution method for the synthesis of nitrides. The

resulting sodium iodide is readily soluble in liquid ammonia, making separation of these products possible. From these results, a plausible route for the synthesis of nitride fuels is:



where An is uranium, neptunium, plutonium or americium. To achieve the solution synthesis of actinide nitrides the fundamental chemical coordination and speciation involved in the above reaction will be examined. The non-aqueous synthetic route, based on amido chemistry, potentially provides property control over the nitride product. The resulting nitride product should be free of the impurities inherent in the carbothermic reduction technique.

The experiments are being performed by a joint research team from UNLV, LANL, and Argonne National Laboratory (ANL), with actinide experiments performed at each laboratory. This project maintains special emphasis on student participation in the project research with all team members collaborating in the educational efforts. The young researchers from UNLV have the opportunity to perform research at ANL and LANL, helping train the next generation of radiochemists and actinide scientists with experience in DOE laboratories. The synthesis of nitride fuels through this method is complementary to the ongoing carbothermic reduction syntheses being performed within the UNLV Radiochemistry program. The ANL and LANL participants are experts in synthetic actinide chemistry, including non-aqueous chemistry and materials science.

The experimental methods for the synthesis of the amido actinide species relies upon the use of inert atmosphere and elevated pressure. These conditions can be reached with the use of Schlenk lines, pressure vessels, and glove boxes. A combination of these techniques is being utilized in the project. Characterization of the resulting amido species will primarily utilize X-ray diffraction,



A depiction of the Schlenk line experimental set-up for the generation and storage of liquid ammonia, solvent transfer for the dissolution of iodine, and the cannula transfer of solution for reaction with uranium.

employing methods and equipment already developed by the UNLV, ANL, and LANL partnership.

Single crystal X-ray diffraction will be used for examination of final and intermediate products at the atomic level for the determination of molecular structure, atomic interaction and spacing, crystal structure, speciation, and elemental oxidation state. X-ray Powder Diffraction, high resolution Transmission Electron Microscopy in combination with Energy Dispersive X-ray Analysis, Parallel-collection Electron Energy-Loss Spectroscopy, Electron-beam Microprobe Analysis and Scanning Electron Microscopy are currently in use at UNLV and will also be used to evaluate the actinide nitrides. For complete product characterization, X-ray Absorption Fine-structure Spectroscopy will also be used for analysis. Thermal analyses (Thermogravimetric Analyzer, Differential Thermal Analyzer, and Differential Scanning Calorimetry) of the final products will also be performed at UNLV.

RESEARCH ACCOMPLISHMENTS

One of the original synthetic routes devised for the synthesis of U(III)N involved the entire reaction taking place in liquid ammonia. Several experimental reactions were conducted in an attempt to synthesize the $UI_3(NH_3)_x$ and $U(NH_2)_3(NH_3)_x$ precursors of U(III)N. Each attempt involved cleaning of the uranium metal to remove the oxide coating of the metal reagent with 3 washes of concentrated nitric acid, each followed by a rinse with liquid ammonia. Success of this cleaning procedure was varied, with a majority of cleaned metal oxidizing rapidly once in contact with the liquid ammonia, despite the precautions taken to eliminate oxygen contamination in the reaction flasks. Due to the continual presence of oxide coating of the uranium metal, it was decided to alter the proposed synthetic route to utilize the synthesis of the $UI_3(THF)_4$ precursor, as described in *Inorganic Chemistry*. This involves the purification of THF through distillation, cleaning of the metal uranium, and slow reaction of U and I_2 . Equipment to perform this synthesis was not available for immediate use, but all necessary glassware was purchased.

It was discovered that the uranium metal turnings being used for the synthesis had a possibility of containing niobium as well. Waste solutions were analyzed by ICP-AES, and no niobium was identified. Other reaction waste solutions were also analyzed in order to determine the reactivity of uranium in the liquid ammonia solution. These analyses determined that only a minimal amount of uranium became dissolved, indicating that either there was little reactivity in the liquid ammonia or the reaction was only taking place on the surface of the metal. Neither of these possibilities is desirable, therefore the synthetic route involving THF was selected to avoid these outcomes.

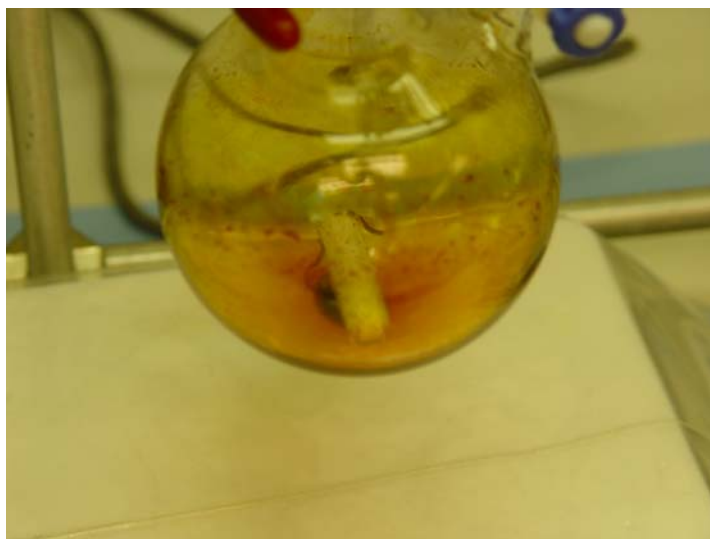
Single crystal and powder diffractometers are necessary instruments for the determination of the intermediate and final products.

ACADEMIC YEAR HIGHLIGHTS

- ◆ D.B. Rego and K.R. Czerwinski, "Unusual Chemistry From F-Element Starting Materials: New Reactivity From F-Element Metallocenes And Halides," 25th Rare Earth Research Conference, University of Alabama, Tuscaloosa, AL, June 2008.
- ◆ D.B. Rego and K.R. Czerwinski, "Unexpected Reactivity Of Simple F-Element Metallocenes and Halides," 25th Rare Earth Research Conference, University of Alabama, Tuscaloosa, AL, June 2008.

Other progress

- Reaction of $UCl_4 + 4 NaNH_3$ in liquid ammonia was performed. $UI_3(THF)_4 + 3 NaNH_2$ in liquid ammonia was performed. The samples were treated and processed.
- Samples were prepared for EA Modeling reactions and crystals were obtained. Modeling reactions were extended to scorpionates. This represents a new synthetic route actinide nitride. Initial characterization indicated other products are present.
- UN was obtained from UI_3 and Li_3N direct reaction and was characterized.
- Metallocenes were synthesized from starting materials. Reaction and monitoring of these metallocenes with amide salts was conducted.
- Liquid ammonia reaction between uranium (III) iodide and sodium amide yielded a new product with indications of being a UN pre-cursor. The product was characterized by XAFS.



A depiction of dissolved iodine reacting with uranium metal after the liquid ammonia solvent has evaporated.

Research Staff

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Tyler Sullens, Co-Principal Investigator, Assistant Research Professor, Harry Reid Center for Environmental Studies
Daniel Rego, Radiochemistry Post-doctoral Researcher, Harry Reid Center for Environmental Studies

Collaborators

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Gordon Jarvinen, Los Alamos National Laboratory

Task 35

Criticality Studies for UREX Processes

D. Beller

BACKGROUND

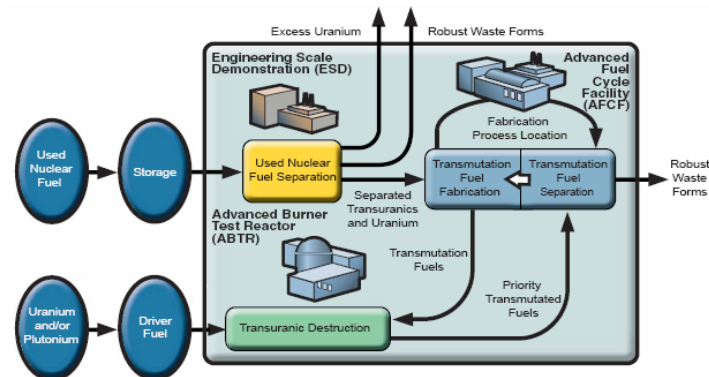
The completion of criticality experiments for mixtures of transuranic actinides (TRU; includes neptunium, plutonium, americium, and curium) that will be created during the separation of used nuclear fuel may be a requirement in order to construct prototype plants for the Global Nuclear Energy Partnership (GNEP). In this program and the Advanced Fuel Cycle Research and Development (AFC R&D) program that supports it, economic and environmental methods are being developed to reduce the impact of waste from commercial nuclear fuel cycles.

Recycling of used fuel by chemically separating it into U, fission products, and TRU would be the first step in this new fuel cycle. Proposed mixtures and concentrations of TRU covering a wide range of conditions must be examined theoretically and experimentally to demonstrate criticality safety in advance of construction of a processing facility. Theoretical studies may be limited because of insufficient nuclear data for the rarer isotopes of Np, Pu, Am, and Cm. These data limitations include reaction cross sections in some energy regimes, thermal feedback coefficients, and delayed neutron fractions.

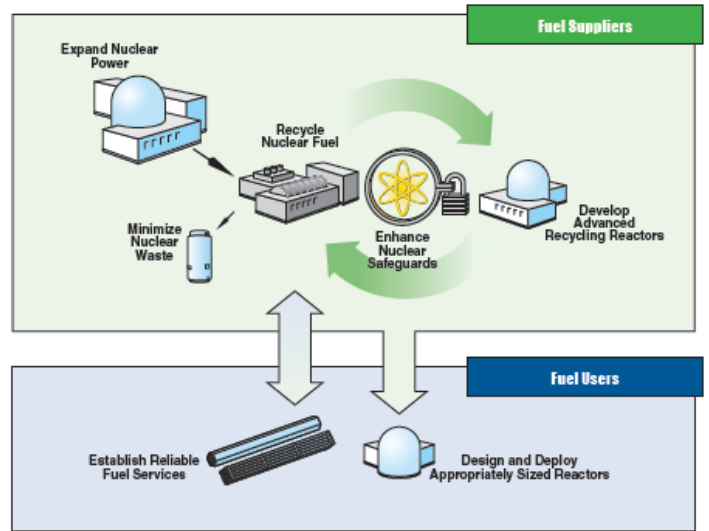
RESEARCH OBJECTIVES AND METHODS

The completion of criticality experiments for mixtures of transuranic actinides that will be created during the separation of used nuclear fuel may be a requirement to construct prototype plants such as the Engineering-Scale Demonstration (ESD) and the Advanced Fuel Cycle Facility (AFCF) for the Global Nuclear Energy Partnership (see illustration below).

GNEP is a program to develop a worldwide consensus to enable the expanded use of economical, environmentally-friendly nuclear energy to meet growing electricity demand. In this program and the Advanced Fuel Cycle R&D program that supports it, participants are developing economic and environmental methods to reduce the impact of waste from commercial nuclear fuel cycles. Recycling of used fuel by chemically separating it into uranium,



The Global Nuclear Energy Partnership research facilities.



The Global Nuclear Energy Partnership (GNEP) structure.

fission products, and transuranic actinides would be the first step in this new fuel cycle. Proposed mixtures and concentrations of TRU covering a wide range of conditions must be examined theoretically and experimentally to demonstrate criticality safety in advance of construction of a processing facility. Theoretical studies may be limited because of insufficient nuclear data for the rarer isotopes of plutonium, americium, curium, and neptunium. These data limitations include reaction cross sections in some energy regimes, thermal feedback coefficients, and delayed neutron fractions (β).

In collaboration between UNLV, Los Alamos National Laboratory (LANL), Argonne National Laboratory (ANL), and Oak Ridge National Laboratory (ORNL), criticality studies will be conducted to support the development of future fuel cycle facilities.

The first step in determining requirements for criticality studies is an examination of past experiments and criticality and sensitivity studies, as well as available databases. Further sensitivity studies will determine what kinds of experiments should be performed to ensure criticality safety in advanced processes. This information can then be used to formulate an optimum set of experiments that can be analyzed in advance using state-of-the-art radiation transport codes. As these facilities and experiments will include complex geometries, a Monte Carlo N-Particle (MCNP) transport code will be used in these sensitivity, scoping, and design studies.

The work may also require generation of new cross section libraries and thermal scattering coefficient databases. Future criticality studies may include cross section sensitivity studies and design of critical experiments including dilute mixtures of Pu, mixed higher actinides in solution, and fuels.

RESEARCH ACCOMPLISHMENTS

Progress implementing NJOY for cross section processing continued with the assistance of LANL and ORNL specialists. NJOY was successfully implemented and is now used to produce temperature-broadened point-wise cross section libraries from eight isotopes of higher actinides for sensitivity studies. Benchmarking of libraries and criticality studies is underway; a benchmark analysis of the Jezebel critical assembly with two temperature-broadened libraries was completed to validate variational and criticality methods.

Processing cross-sections in NJOY99.259 was completed and a review of criticality benchmark experiments was initiated for establishing the range of applicability of physical and spectral parameters. Modeling of criticality sensitivity testing with minor actinides in the UNM AGN-201 reactor continued. A review of criticality safety benchmark experiments was completed for the established range of applicability of physical and spectral parameters and reactivity sensitivity studies began on higher actinides. Fast spectrum reactivity sensitivity studies were completed on eight higher actinide isotopes at six different temperatures. Void and replacement reactivity changes were calculated with three fast benchmark experiments.

The SHEBA liquid-core critical assembly was modeled in preparation for sensitivity studies. Reactivity sensitivity studies of the SHEBA liquid-core critical assembly was completed and modeling detectors for determining ability to measure reactivity changes was initiated. A californium source was added opposite a neutron detector to model neutron transport through the liquid-core SHEBA for reactivity sensitivity studies.

Other Accomplishments

Dr. Charlotta Sanders joined the UNLV research faculty and this project as a co-investigator. Her expertise is in radiation transport, criticality, and shielding analyses, with substantial experience with investigations related to used nuclear fuel. She is advising students and developing new research projects for continuation of MPAC research.

Funding was approved for a contract with Professor Robert Busch of the University of New Mexico (UNM) to investigate the use of UNM's AGN-201 reactor for criticality sensitivity experiments with higher actinides. A MCNP model of AGN-201 reactor was

ACADEMIC YEAR HIGHLIGHTS

- ◆ Lawrence Lakeotes, M.S. thesis, Department of Mechanical Engineering, "Neutron Cross-Section Sensitivity Studies in Higher Actinides for Criticality Safety in Reprocessing," University of Nevada, Las Vegas, NV, April 2008.
- ◆ L. Lakeotes, "Neutron Cross-Section Sensitivity Studies in Higher Actinides for Criticality Safety in Reprocessing," oral presentation, 2008 American Nuclear Society Student Conference, College Station, TX, March 1, 2008.
- ◆ Timothy Beller graduated with an M.S. in Materials and Nuclear Engineering, December 2007.
- ◆ Ryan LeCounte graduated with an M.S. in Materials and Nuclear Engineering, December 2007.
- ◆ Lawrence Lakeotes graduated with an M.S. in Materials and Nuclear Engineering, May 2008.
- Sandra De La Cruz graduated with a B.S.M.E. (nuclear option), May 2008.
- UNLV submitted a closely related, multi-year proposal to the Nuclear Energy Research Initiative for Consortia (NERI-C) with Georgia Tech and UNM as consortium members for computational and experimental examination of actinide cross sections and criticality experiments.
- ◆ Dr. Charlotta Sanders joined the UNLV research faculty and this project as a co-investigator.

obtained from UNM, some programming errors were corrected, and successful criticality runs were conducted in preparation for sensitivity studies.

These studies have begun by modeling small quantities of higher actinides in the reactor hole to determine reactivity sensitivity for potential experiments. Using this funding to prepare for future work, UNLV and UNM conducted a Reactor Experimentation Laboratory Course at UNM using the AGN 201M reactor. In addition, a research proposal was submitted to DOE/NE for collaboration with UNM and Georgia Tech for criticality studies using the UNM AGN reactor.

FUTURE WORK

During the following year the SCALE and MCNP/MCNPX will be used to investigate cross section sensitivity of UREX process and conceptual integral experiments.

Research Staff

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Charlotta Sanders, Research Professor, Mechanical Engineering Department

Students

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Tanya Sloma and Sandra De La Cruz, Undergraduate Students, Mechanical Engineering Department

Collaborators

Robert Busch, Professor, Nuclear Engineering Department, University of New Mexico
Richard McKnight, Argonne National Laboratory
Michael Dunn, Nuclear Data Division, Oak Ridge National Laboratory
David Hayes, Los Alamos National Laboratory
Thomas Ward, UNLV Russian Collaboration Science Adviser, TechSource, Inc.

Task 36

Evaluation of Cs/Sr Waste Form for Long Term Storage and Disposal

G. Cerefice and L. Ma

BACKGROUND

To maximize the utilization of a proposed repository facility, the short-term decay heat generated by high-level radioactive waste must be removed from the waste stream. One proposed advanced fuel cycle strategy calls for the separation of cesium and strontium from used nuclear fuel in order to minimize the short-term heat loading in a repository facility. The separated waste stream will be converted to an aluminosilicate waste form, stored for decay (approximately 300 years), then managed as low-level radioactive waste. The goal is to examine two potential concerns regarding the long-term performance of this proposed cesium/strontium waste form.

To facilitate long-term storage, the disposal containers will need to be able to survive for the entire storage interval. The first aspect of the project will explore the potential interaction of the aluminosilicate waste form with the storage canister materials to determine if there is any corrosion or chemical interaction concerns for the storage of the materials. At the end of the storage interval, most of the cesium (^{137}Cs) in the waste form will have decayed to its daughter, barium (^{137}Ba). While this decay provides a significant reduction in the decay heat generated by the waste form, it poses a new concern. Barium is hazardous, and is identified by the U.S. Environmental Protection Agency as a hazardous constituent under the Resource Conservation and Recovery Act (RCRA). To dispose of any material containing a RCRA-identified constituent, the material must be demonstrated to be durable enough to prevent the release of the hazardous component or must be treated as hazardous waste. For the Cs/Sr waste stream, failure to contain the barium within the waste form would require disposal as a mixed waste stream, greatly increasing the disposal costs. Understanding the potential impacts of radioactive damage, high storage temperatures, and the crystallographic im-

pacts of the decay transmutation itself on the performance of the waste form 300 years from now poses a significant challenge.

RESEARCH OBJECTIVES AND METHODS

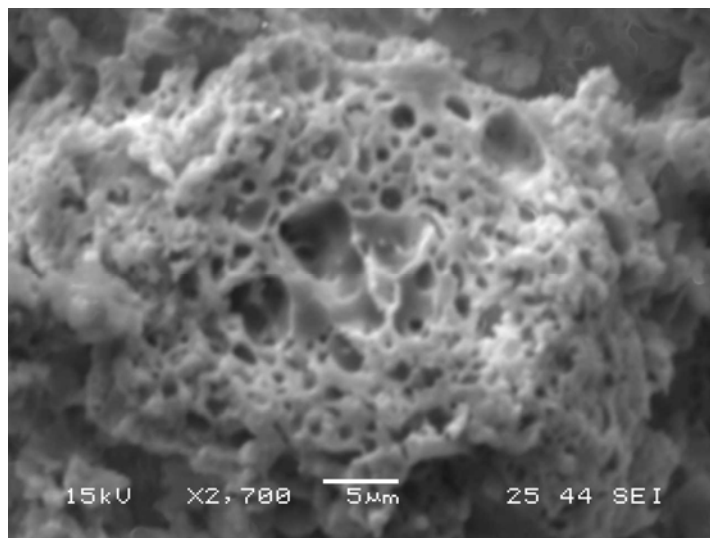
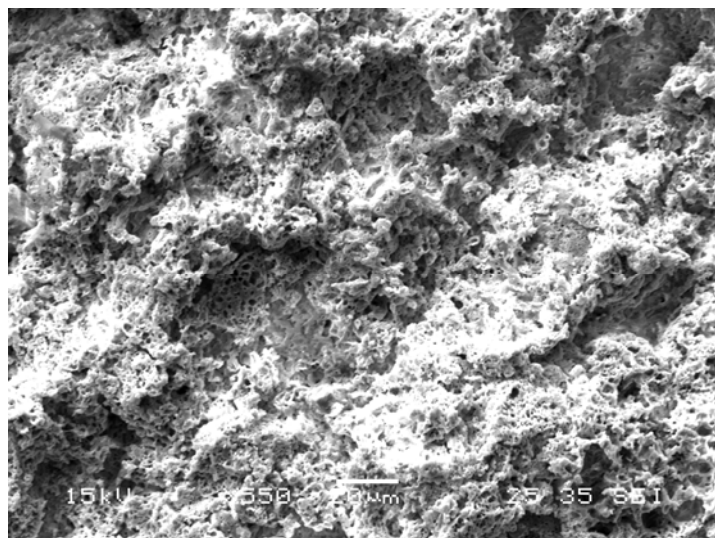
The goal of this task is to develop, characterize, and optimize the proposed aluminosilicate waste form for a separated cesium and strontium waste stream. The research effort at UNLV will be divided into two subtasks:

- Materials compatibility, and
- Waste form optimization and performance.

The materials compatibility subtask will examine the potential for chemical interactions between the waste form material and proposed structural materials for the disposal container (carbon steel, stainless steel, etc.). The waste form performance task will examine the leach resistance of the waste form, with particular attention to barium retention.

The research objectives of this project are:

- To characterize the Cs/Sr-loaded aluminosilicate waste form ceramic,
- To examine the impact of fabrication process parameters on the product waste form,
- To evaluate the potential for chemical interactions between the waste form and container material, and
- To examine the degradation and alteration behavior of the waste form.

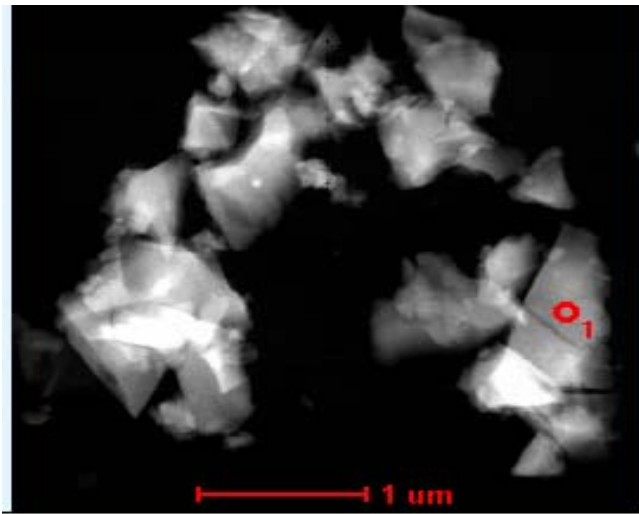


Cs/Sr/Ba-loaded waste form matrix at 550X (a) and 2700X (b) magnification.

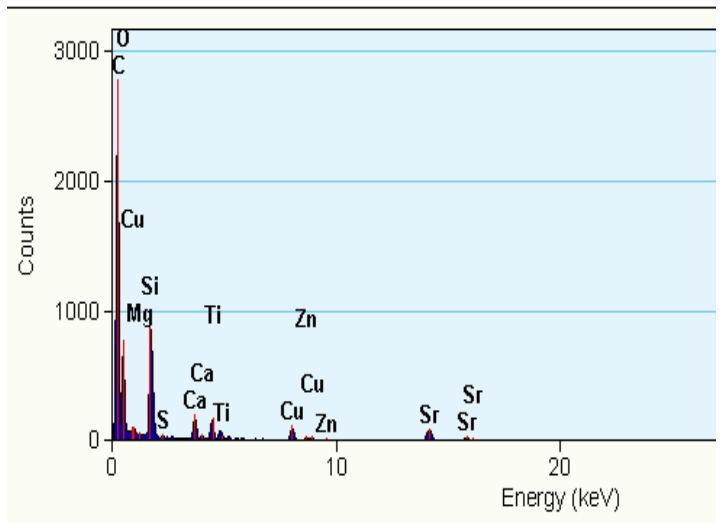
RESEARCH ACCOMPLISHMENTS

To optimize the Cs/Sr waste form, the impact of fabrication process parameters on the final product had to be evaluated. Initial experiments examined the impact of sintering temperatures on the waste form. Thermogravimetric analysis (TGA) identified the temperatures where sorbed and interstitial water is driven from the waste former matrix, as well as the temperatures where the bentonite waste former material undergoes a phase transition to a glass, establishing an upper temperature limit for the fabrication of the waste form. The impact of sintering time and temperature on the waste form and on the starting materials was examined by X-ray Diffraction (XRD) analysis. Significant differences in the crystallinity as well as phase composition of the Cs/Sr-loaded waste form were observed from increasing the sintering temperature from 800 °C to 1000 °C, with additional changes observed when the sintering time at temperature was increased to over 20 hours. Characterization of the product waste forms, including identification of all the composite phases, will continue through the end of the project.

Characterization of the product morphology by scanning electron microscopy showed that the current batch processing method (in which the bentonite clay is loaded with Cs and Sr in solution, dried, then sintered) yields a highly porous material with a sponge-like morphology. Coupled with the results of the TGA analysis, this is likely due to the release of interstitial or matrix water during the early stages of the sintering step. Electron microprobe analysis (EMPA) showed a uniform distribution of Cs, Sr, and Ba throughout the sample matrix, at least at the micron scale.



(a)



(b)

TEM image of Cs/Sr/Ba-loaded Waste Form Matrix (a) with elemental analysis of indicated spot (b).

ACADEMIC YEAR HIGHLIGHTS

- ♦ Identified glass transition temperature for bentonite waste former materials to establish waste form fabrication process limits.
- ♦ Identified and characterized barium- and strontium-bearing phases in sintered waste form matrix.

Further analysis by transmission electron microscopy (TEM) indicated, however, that the waste stream components are actually segregated into discrete phases, and that these discrete, sub-micron particles are distributed throughout the matrix. Initial characterization has identified separate Sr- and Ba-containing aluminosilicate phases in the host matrix. The Cs-bearing phase (or phases) in the matrix have not been identified and confirmed at this time.

FUTURE WORK

With the change in national program direction towards a combined fission product waste stream, work on the waste forms for separated cesium and strontium has been reduced to a lower priority. The work at UNLV will focus primarily on wrapping up the characterization of the waste matrices so that they could be pursued in the future if program directions change, focusing on the identification of cesium-bearing phases within the waste form matrix.

Research Staff

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Longzhou Ma, Assistant Research Professor, Harry Reid Center for Environmental Studies

Student

K.M. Nazmul Ahsan, Graduate Student, Department of Mechanical Engineering

Collaborator

Michael Kaminski, Argonne National Laboratory

Task 37

Thermal Transient Flow Rate Sensor for High Temperature Liquid Metal Cooled Nuclear Reactor

Y. Jiang and J. Ma

BACKGROUND

In nuclear power plants and accelerator driven systems (ADS) for nuclear waste treatment, it is important to monitor the coolant flow rate in the reactor core and pipe-line. In such a strong irradiation, high pressure, and temperature environment, the existing flow measurement techniques (such as Electromagnetic flow meters, Ultrasonic flow meters, Turbine flow meters, etc.) are not accurate and reliable.

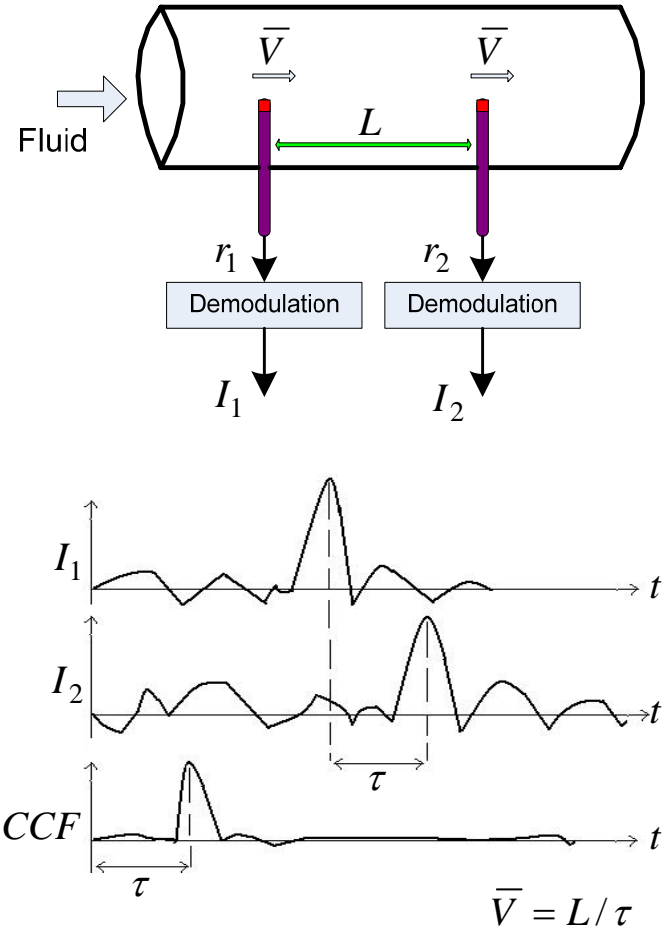
The measurement of flow rates (mass flow rates or volume flow rate) plays a notable role in monitoring and controlling the experimental conditions. The bulk flow rates can be obtained through direct methods, which measure the amount of discharged fluids over a period of time. Alternatively, flow rates can also be obtained using indirect methods. For example, they can be derived through the measurement of fluid velocities. So far, the velocities have been found in strong correlation with signals of pressure, temperature, optical wave, ultrasonic wave, etc. based on diverse physical principles. Note that with some exceptions, the flow rate measurement systems require calibration or empirical corrections, especially after long term operation. In the application of liquid metal coolant flow rate measurement, the high temperature, pressure, and corrosion environment limit most flow meter devices from being used in long term and maintenance-free operation.

As the temperature measurement technique is well developed for high temperature applications, one flow rate measurement technique is proposed based on the correlated thermal signals. This way, the measurement errors due to long term corrosion will be easily counteracted using this proposed method. Correlated thermal signals are measured to deduce the flow velocity.

RESEARCH OBJECTIVES AND METHODS

An alternative flow rate measurement technique has been proposed for this task based on correlated thermal signals obtained from a pair of temperature sensors placed a certain distance apart along the flow. A widely used cross-correlation algorithm, however, suffers from the ambiguity in reading of measurements. To alleviate this problem, a new algorithm is introduced to further improve the accuracy in the transit time estimation using an adaptive inverse system model at a higher cost of computation. When real-time computation is a concern, a second algorithm is proposed based on an adaptive filtering approach which makes a sound trade-off between accuracy and computation cost. This algorithm incurs less processing time than the first proposed algorithm with higher accuracy than the two aforementioned conventional algorithms. These algorithms were evaluated with experiments in a water-based testing loop.

This idea is based on the transit time estimation, using a pair of thermocouples along the flow to provide the temperature readings at two locations along the flow. It is safe to assume that there is a negligibly small change in the characteristics of flow structures,



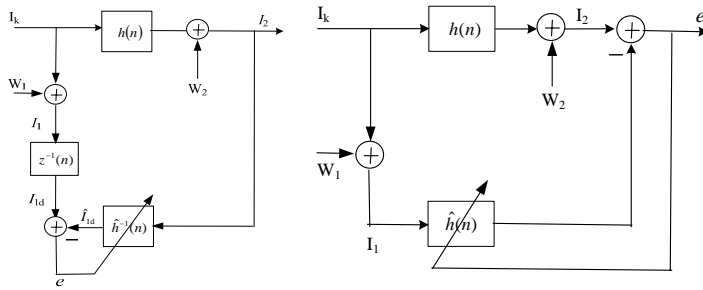
Time Transit Configuration.

provided the sensors are within certain distances. In the illustrated case (above), the upstream thermocouple records a flow signature L/V seconds earlier than the downstream one, where L is the distance between the two sensors and V is the flow velocity. By comparing the signals from the two thermocouples, the time delay, t , can be determined and thus the velocity can be given as follows:

$$V = L/t$$

Methods

The transition time can be obtained using two techniques: cross-correlation-function- or transfer-function-based methods. The first technique, which uses the maximum value of the cross correlation function (CCF) of a measured signal, has two main problems: (i) the obtained peak is too wide, having a negative impact on the result accuracy and (ii) besides the main peak, there can be other undesirable peaks. To alleviate this problem, the transfer function estimation approach was recently proposed. This approach tends to give a narrower peak to get the transit time. A new algorithm was introduced to further improve the accuracy in the transit time estimation using an adaptive inverse system model at a higher cost



New Algorithm I

New Algorithm II

of computation. When real-time computation and measurement is a concern, a second algorithm is proposed based on an adaptive filtering approach. This algorithm incurs less processing time than the first algorithm, with higher accuracy than the two aforementioned conventional algorithms.

Objectives

This project focuses on experimental investigation of a correlation velocity measurement technique by analyzing the temperature fluctuations naturally existing in turbulent flows. Thermocouple temperature sensors are employed in the experiments to obtain local temperature fluctuations. The objectives of the proposed research are as follows:

- To design and construct a correlation velocity measurement device that utilizes the thermocouple temperature sensors to obtain temperature information;
- To develop a data processing scheme and to implement the scheme to build a LabVIEW-based data acquisition system;
- To test the correlation velocity measurement technique in a thermal-hydraulic experimental test facility (water-based), and to compare the results with those obtained from other commercial flow meters;
- To test the measurement device in the UNLV TC-1 loop test section;
- To suggest any improvements for the measurement technique based on the experimental results; and,
- To develop circuit boards for signal conditioning, signal processing, and system integration.

RESEARCH ACCOMPLISHMENTS

The flow velocity of liquid metal coolant, e.g., lead bismuth eutectic (LBE) can be determined through the measurements of temperature fluctuation recorded by a pair of temperature sensors placed a certain distance apart along the flow. Traditionally, this was done using a cross-correlation algorithm to estimate the transit time of the coolant, and thus its velocity. This widely used cross-correlation algorithm, however, suffers from the ambiguity in

ACADEMIC YEAR HIGHLIGHTS

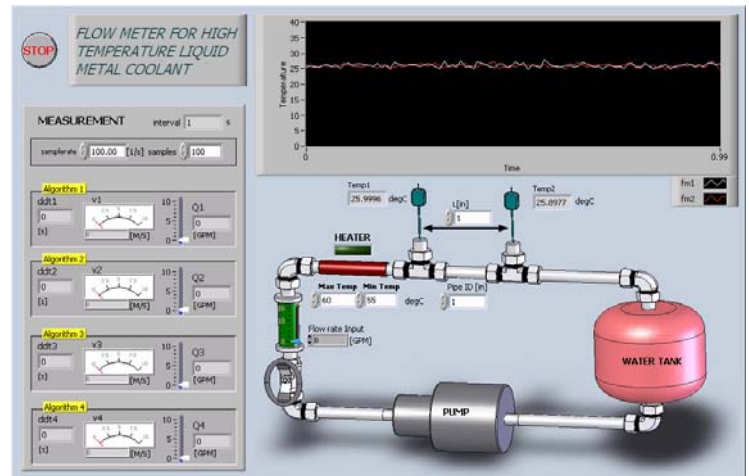
- ♦ T. Moazzeni, Y. Jiang, J. Ma, and N. Li, "Algorithms for the measurement of liquid metal coolant flow velocity with correlated thermal signals," International conference on Communications, Circuits and Systems, Xiamen, Fujian Province, China, May 25-27, 2008.

reading of measurements. To alleviate this problem, the transfer function estimation approach was recently proposed and tends to give more accurate results. Both algorithms have been coded and compared with two algorithms developed by the principal investigators and the students.

A new algorithm has been introduced which can further improve the accuracy in the transit time estimation using an adaptive inverse system model at a little higher cost of computation. When real-time computation is a concern, a second algorithm is proposed based on an adaptive filtering approach which makes a sound trade-off between accuracy and computation cost. This algorithm incurs less processing time than the first proposed algorithm with higher accuracy than the two aforementioned conventional algorithms. A test rig has been built to test the proposed algorithms.

FUTURE WORK

In order to increase the thermal signal amplitude, one customer designed heater will be designed and installed in the water testing rig in the near future to evaluate the signal processing algorithms. The optimized specifications of flow meter (such as distances, heating cycles, etc.) will be obtained in the water based testing rig. Based on the experience on water, this flow meter will be installed in the testing section in TC-1 loop that uses LBE.



The console of the data acquisition system program in LabView.

Research Staff

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Task 38

f-Element Electrochemistry in RTIL Solutions: Electrochemical Separation of Lanthanides and Actinides

D.W. Hatchett and K. Czerwinski

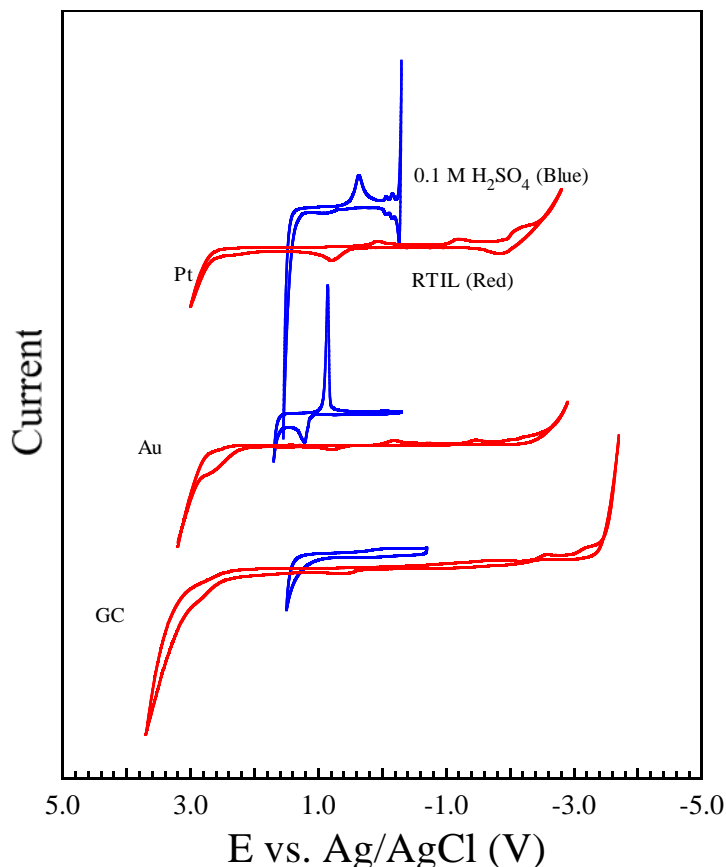
BACKGROUND

Electrochemical methods can be used to effectively separate actinide and lanthanide species from complex mixtures. This is based on the unique electrochemical properties of each specific target species. In studies it has been found that, with the exception of Ce, aqueous solutions provide unsuitable electrochemical windows to effectively evaluate the thermodynamic properties that are useful for chemical separations. Therefore, a more novel approach was examined which eliminated the aqueous solution with a room temperature ionic liquid (RTIL) solution. RTIL solutions do not suffer from the side reactions that are prominent in aqueous environments. In addition, the potential window is much larger for the RTIL solutions. They are a new starting point for the electrochemical separation of individual species from a mixture.

The ultimate goal is to fully characterize the oxidation/reduction of f-elements in RTILs to establish the baseline thermodynamic and kinetic data for these systems. The data will be used to critically evaluate the ability to use electrochemical methods for controlled, potential mediated, separation of f-elements by electroplating on electrode surfaces. Factors that will influence the ability to measure the redox processes in f-elements in RTIL solutions and electroplating on electrode surfaces include the structure, solubility, and stability of the target species in these solutions.

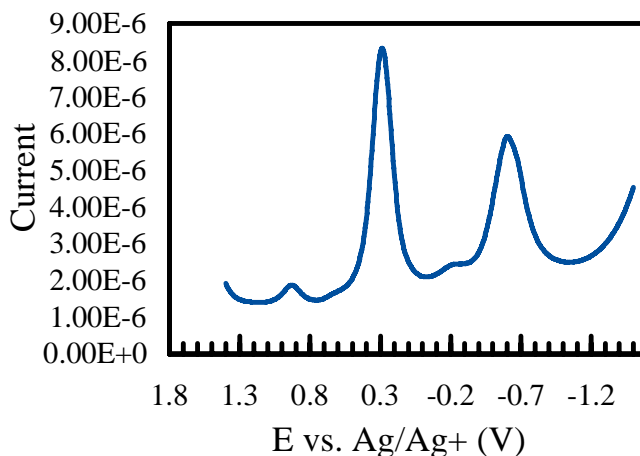
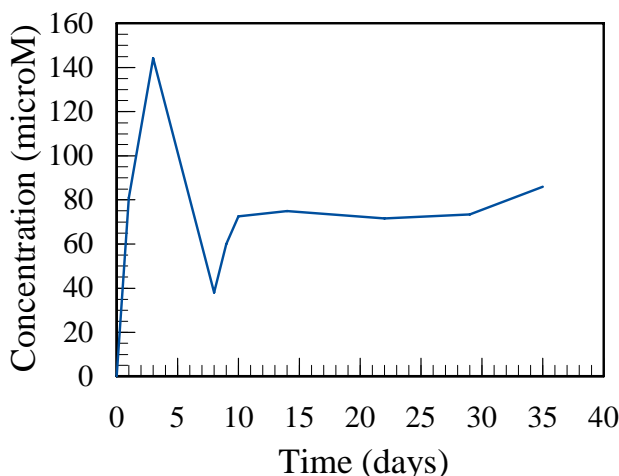
RESEARCH OBJECTIVES AND METHODS

The objective of this project is to use electrochemical techniques to develop a thermodynamic understanding of actinide and lanthanide species in RTIL solutions, and to use this data to effectively separate species with very similar chemical properties. In consultation with a U.S. Department of Energy national program collaborator, electrochemical methods and materials will be evaluated and used to exploit the thermodynamic differences between similar chemical species enhancing the ability to selectively target and



Electrochemistry of glassy carbon (GC), Au, and Pt electrodes in RTIL ([MeBu₃N][NTf₂]) (red) and 0.1 M H₂SO₄ (blue).

sequester individual species from mixtures. This project is in its first year and has successfully completed Phase 1. This research allows for expansion into Phases 2 and 3 for the next year.



(left) Solubility of UO₂CO₃ in a RTIL solution. (right) Square wave voltammetry of UO₂CO₃ in RTIL after 24 hours.

Phase 1 objectives:

- To prepare and characterize RTIL solutions.
- To examine the electrochemical window and to evaluate the electrochemical properties of lanthanide and actinide species in the non-aqueous ionic environment.

Phase 2 objective:

- To examine the solubility and complex formation of Lanthanide and Actinide species.

Phase 3 objective:

- To examine extraction properties of NTf_2^- ion complexes into RTIL solutions with common ions.

Phase 4 objective:

- To electrodeposit lanthanide and actinide species from RTIL.

RESEARCH ACCOMPLISHMENTS

The advantage of using RTIL solutions and eliminating aqueous side reactions and expanding the potential window is that previously inaccessible electrochemical reactions may be observed. These systems provide a huge potential window for the observation of oxidation and reduction of lanthanides and actinides.

In addition, the potential mediated deposition of f-elements is possible, indicating that controlled separation of chemical species is thermodynamically feasible. For example, the reduction of Pu^{3+} , Am^{2+} , Am^{3+} , Cm^{3+} , and U^{3+} all occur before $E = -1.90 \text{ V}$ vs. Ag/AgCl , well within the negative potential limit for the RTIL at platinum (-2.6 V vs. Ag/AgCl), gold (-2.2 V vs. Ag/AgCl), and glassy carbon (-3.4 V vs. Ag/AgCl) electrodes. The figure on the opposite page (top) provides an example of the potential windows associated with GC, Au, and Pt electrodes in aqueous and RTIL solutions.

ACADEMIC YEAR HIGHLIGHTS

- ♦ Synthesis of bulk quantities of RTIL.
- ♦ Electrochemical characterization of the UO_2^{2+} system.

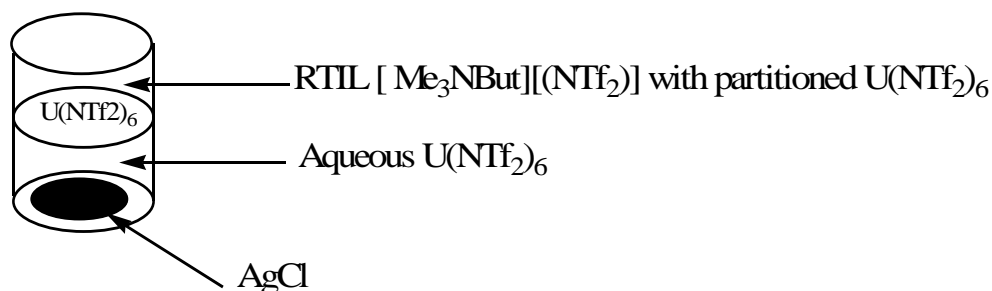
A second advantage is the increased solubility of lanthanide and actinide species in the RTIL. RTIL solutions have been utilized in the waste cycle for extraction of species. These solutions could be useful in the potential dependent separations of lanthanide and actinide species. The figure on the opposite page (bottom) provides the preliminary evaluation of UO_2CO_3 solubility in RTIL. The electrochemical response if UO_2^{2+} is provided also.

Preliminary studies suggest that the extraction and electrode position of lanthanide and actinide species into RTIL solutions is possible. Work will continue with increasing the solubility of target species in RTIL.

FUTURE WORK

- Precipitation/complexation using $\text{Ag}(\text{NTf}_2)$ with chloride derivative of lanthanides and actinides.
- Extraction of the complex using RTIL solutions and common ions.

Future work will focus on the complexation of simple species like CeCl_3 , CeCl_4 . This will be performed through the aqueous precipitation of AgCl and the formation of triflate complexes of Ce. The complexes will then be extracted without the use of additional complexing species such as tributylphosphate (TBP). The goal is to provide a common complex anion for the metal that will allow neutral species to be extracted more efficiently than the TBP/n-Dodecanese system. Once the species are extracted into the RTIL, the electrochemistry will be examined using this solution. The ultimate goal is to electrodeposit and separate a given species using potential dependent methods.



Schematic of experimental set-up for precipitation/complexation using $\text{Ag}(\text{NTf}_2)$.

Research Staff

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Student

Wendy Pemberton, Graduate Student, Department of Chemistry

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Task 39

Knowledge-Based Information Resource Management System for Materials of Sodium-Cooled Fast Reactor

S. Hsieh

BACKGROUND

In the development of advanced fast reactors, materials and coolant/material interactions pose a critical barrier for higher temperature and longer core life designs. For sodium-cooled fast reactors (SFRs) such as the Experimental Breeder Reactors in Idaho and the Fast Flux Test Facility in Hanford, experience has shown that qualified structural materials and fuel cladding severely limits the economic performance.

Liquid sodium has been selected as the primary coolant candidate for the Advanced Burner Reactor (ABR) of the Global Nuclear Partnership (GNEP). Materials improvement has been identified as a major thrust to improve fast reactor economics. Researchers from universities, national laboratories, and related industrial participants have been continuously generating data and knowledge about materials and their interactions with coolants for the past few decades. Considering cost and time, the paradigm of designing and implementing a successful advanced nuclear system can be shifted and updated via the integration of information and internet technologies. Such efforts can be better visualized by implementing collective (centralized or distributed) data storages to serve the community with organized material data sets. Material property data provided by MatWeb.com and the ongoing development of a web-based material handbook for the Generation IV (GEN IV) advanced reactors are a few examples.

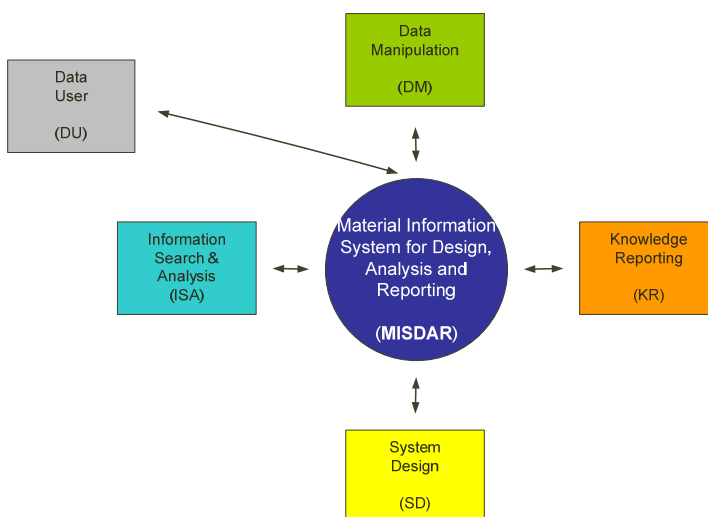
From a system design perspective, SFRs proposed in the GEN IV system have been significantly developed. Major activities defined in the current SFR research and development (R&D) plan can be summarized as the following:

- Ensuring that the needs and goals of the program are followed by the GEN IV International Forum (GIF) countries,
- Documenting and sharing the R&D progress and accomplishments, and
- Integrating relevant activities from GIF SFR R&D with GNEP.

All of these activities follow the path of data generation, analysis, knowledge discovery and, finally, decision making and implementation.

RESEARCH OBJECTIVES AND METHODS

This project proposes to create a modularized web-based information system with models to systematically catalog and analyze existing data, and guide the new development and testing to acquire new data. Technically speaking, information retrieval and knowledge discovery tools will be implemented for researchers with both information lookup options from material databases and technology/development gap analysis from intelligent agent and reporting components. The goal of the system is not only to provide another database, but to also create a distributable and expandable, platform-free, location-free online system for research institutes and industrial partners. Such knowledge discovery and data mining processes generally include data integration, prepara-



Main data flow diagram for the MISDAR.

tion and transformation, data mining and evaluation, and data visualization. Parallel to the development of these front-end analysis tools, web-based data updating and portal administration interfaces will also be designed and developed. Data collection will start during the early stage of the project due to its time consuming nature. The research objectives have been further divided into seven parts, described below:

1. To effectively identify research gaps. Past research on liquid metals need to be initially collected and documented.
2. To conduct requirement analysis on stakeholder identification, data-structure definition and analysis tool definition.
3. To design the multi-tier application architecture based on the requirement analysis. Two data sources, configuration settings and application contents, are stored in an XML (eXtensible Markup Language) file and Microsoft (MS) SQL Server database, respectively.
4. To develop the online system prototype to include database development, portal functionality development and portal presentation development.
5. To implement the web-based resource management system that integrates web portal programming and web server hardware configuration. A Windows-based resource management system will be designated as a web server while database servers can be located onsite with the web server, or at various locations depending on the accessibility of data sources.
6. To conduct system testing, debugging and refinement after the system is initially implemented.
7. To instruct graduate students and publish results related to information retrieval, and material search algorithm development.

RESEARCH ACCOMPLISHMENTS

The final project scope of the “Material Information System for Design, Analysis and Reporting” (MISDAR) was developed in collaborators from LANL. Identified functionalities of the MIS-

DAR are:

- Data manipulation (DM) with data uploading/updating/validation capabilities,
- Information search and analysis (ISA) with advanced search engine and organization algorithms
- System design (SD) with flexible data search engine and analysis tools
- Knowledge reporting (KR) with on-demand reporting tools.

Although the MISDAR is designed to document sodium-related data and information, the “Handbook on Lead-bismuth Eutectic Alloy and Lead Properties, Materials Compatibility, Thermal-hydraulics and Technologies (2007 Edition),” by the OECD/NEA Nuclear Science Committee, was initially used as a baseline reference and guideline on data classification, analysis, and presentation. The system under development will initially include thermo-physical and electrical properties, materials and testing issues, thermal-hydraulics and system technologies, existing test facilities, safety guidelines and open issues and perspectives. Unlike the linear chapter arrangement seen in the paper material handbooks, the material properties and design objects of interest will be effectively presented through a sequence of search results, summary

ACADEMIC YEAR HIGHLIGHTS

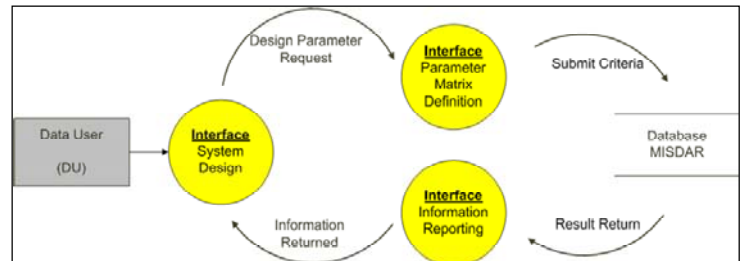
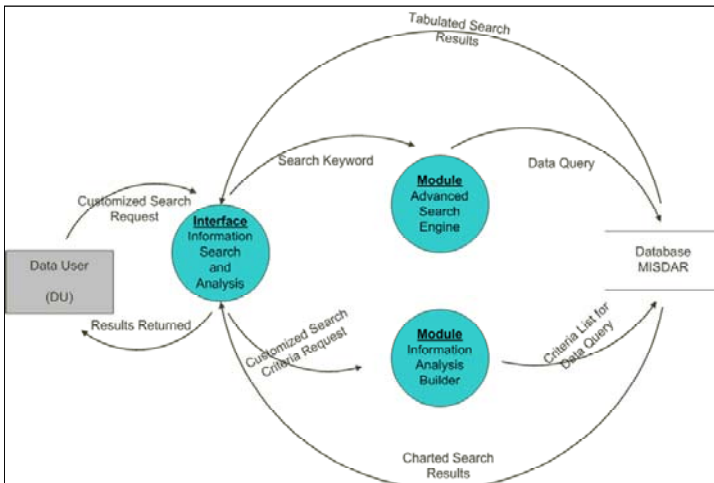
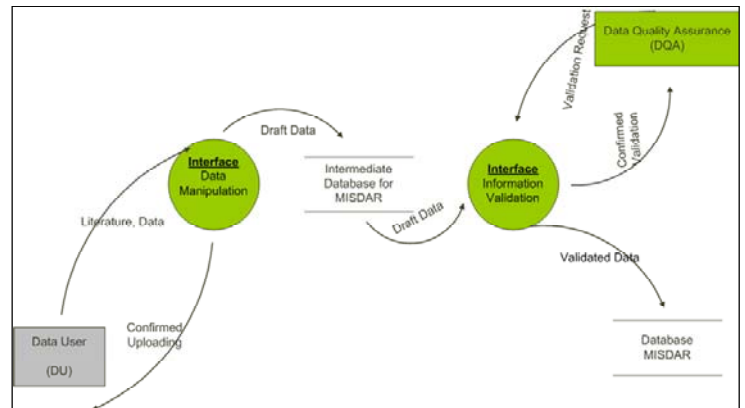
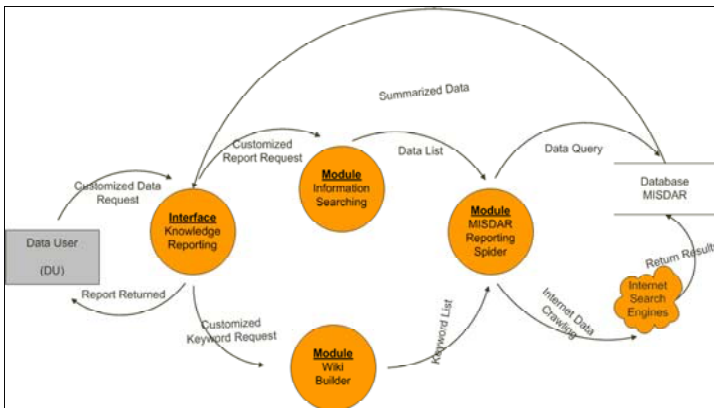
- ♦ A graduate student from the Department of Informatics, was identified to work on information reporting and algorithm development subtasks.
- ♦ More than 70 sodium related references have been collected.

tables, and charts with reference hyperlinks.

The functional structures with the associated priority for the MISDAR implementation are: Portal Management (high), Portal Presentation (high), Data Manipulation (high), Knowledge Reporting (high), Information Search and Analysis (medium), and System Design (low).

FUTURE WORK

Future work will include implementing and testing online system interfaces, and providing a feedback interface for further improvement.



Detailed data flow diagrams for modules related to data manipulation (DM), knowledge reporting (KR), information search and analysis (ISA) and system design (SD).

Research Staff

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Ning Li and Jinsuo Zhang, Los Alamo National Laboratory

Task 40

Evaluation of Fundamental Radionuclide Extraction Data for UREX

K. Czerwinski

BACKGROUND

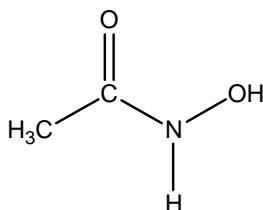
The speciation of technetium and actinides in advanced solvent extraction systems is the basis for their manipulation in separations. The ability to understand and predict radionuclide speciation is paramount to successful modeling of proposed separation systems. This project will examine the speciation of radionuclides in different stages of the uranium extraction (UREX) separation scheme, providing data useful to modeling. The areas to be examined include the speciation of uranium and plutonium with tributylphosphate and the kinetics and thermodynamics of lanthanides and actinides in the TALSPEAK (Trivalent actinide lanthanide separation by phosphorous reagent extraction from aqueous complexes) system. The complexation constants of uranium and plutonium with tributylphosphate (TBP) will be evaluated. In the TALSPEAK system, studies will elucidate the difference in complexation kinetics for the lanthanides and actinides. Computational studies based on density functional theory will be performed for both systems.

RESEARCH OBJECTIVES AND METHODS

Tributylphosphate-Based Extractions

The extraction of tetravalent Pu in nitric acid solution by TBP is well known and has been the basis of Pu purification for a number of decades. However, the data needed for detailed modeling of Pu extraction is not available for all conditions of concern to the Advanced Fuel Cycle Initiative.

Understanding the role of nitrate in plutonium speciation is important for determining the necessary data for plutonium extraction modeling. Data indicates the dinitrate complex of plutonium is strong and may account for difficulties in modeling plutonium extraction under some nitrate conditions. The formation of hydrolysis products may also form an extractable species that will need to be included in modeling. In the UREX process, acetohydroxamic acid (AHA, see figure below) is expected to be used. For the separation of Pu in a solvent extraction system using acetohydroxamic acid, it is necessary to determine the complexation kinetics and thermodynamics. In addition, since plutonium is a redox sensitive element, the change in speciation due to oxidation-reduction reactions upon complexation needs to be evaluated. This project will experimentally evaluate the fundamental speciation of plutonium and uranium in the TBP-dodecane-nitric acid system, with the main emphasis on nitrate speciation and subse-



Acetohydroxamic acid.

Examples of ligands for actinide and lanthanide separations.

| | |
|--|---|
| | Bis(2-ethylhexyl)phosphoric acid (HDEHP) for TALSPEAK |
| | Diphenyl-N,N'-dibutylcarbamoyl phosphine oxide (CMPO) for TRUEX |
| | N,N'-dimethyl-N,N'-dibutyl-2-tetradecyl-malonamide (DMDBDMA) for DIAMEX |
| | Quaternary ammonium salts (Aliquat 336) |
| | α -hydroxyisobutyrate |

quent third phase formation at high nitric and metal ion conditions. Experiments on the AHA systems will be conducted in collaboration with Argonne National Laboratory (ANL) with input from the U.S. Department of Energy Separations Working Group. For this project the focus will be on the evaluation on U and Pu complexation constants with TPB.

Lanthanide-Actinide Separations

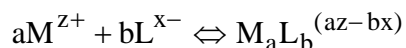
The separation of trivalent lanthanides from trivalent actinides, and americium from curium, has been identified as a topic for investigations by the Global Nuclear Energy Partnership (GNEP). These separations are based on slight differences in ionic radii, small variations in Lewis acidity between the lanthanides and actinides, and redox state speciation.

Numerous ligands have been examined for actinide and lanthanide separations, some of which are the basis of separation schemes. In this work efforts will focus on the TALSPEAK process through the determination of thermodynamic and kinetic data necessary for separation modeling. The TALSPEAK conditions include an aqueous phase with 1 M lactic acid and 0.1 M diethylenetriamine-pentaacetic acid (DTPA). In the original work, the lanthanides are extracted from the aqueous phase at pH 3 with an organic phase of 0.3 M Bis(2-ethylhexyl)phosphoric acid (HDEHP). The actinides americium and curium are then extracted into 0.3 M HDEHP at pH 1.5. The actinides can be removed from the organic phase with dilute mineral acid. Recent GNEP related results have focused on evaluating kinetic and thermodynamic parameters for the extraction of actinides and lanthanides in the TALSPEAK process. While complexation data is available for the interaction of americium and curium with some ligands used in the TALSPEAK process, further efforts are necessary to complete the appropriate dataset for modeling separations in AMUSE (Argonne Model for Uni-

versal Solvent Extraction). Acquiring the necessary kinetic and thermodynamic data for AMUSE modeling of lanthanide and actinide extractions will be an important element of this project.

Evaluation of Chemical Thermodynamic and Kinetic Data

The goal of our plutonium and uranium speciation studies will be to obtain data for modeling the behavior of the actinides under a range of extraction conditions, including acid concentration, metal ion concentration, and temperature. Speciation calculations can be performed if the stability and solubility constants for the chemical species formed under the examined conditions are known. If conditions under which precipitation of the actinide occurs, solubility studies of the precipitated species will be included. A general chemical reaction is described as:



for complexation where M is the metal ion and L is a ligand. In the case of the extraction system under investigation, two ligands may be considered, namely nitrate and TBP. The kinetics of the reaction can be measured to establish conditions needed for equilibrium. For the extraction system under study the kinetics are expected to be rapid. However, kinetic evaluation of third phase formation may be needed.

Enthalpy and entropy will be investigated by evaluating the stability constant as a function of temperature. For a system with different species, if the stability constants are known, then all the species can be calculated at any given pH. The thermodynamic data can also easily be incorporated into existing codes such as AMUSE or the geochemical code CHESS.

Computational studies

Density functional theory (DFT) calculations will be performed to



TBP-HNO₃-Np⁴⁺ third phase appearance..

ACADEMIC YEAR HIGHLIGHTS

- ◆ C.S. Gong, F. Poineau, W.W. Lukens, and K.R. Czerwinski, "Something New From Something Old: Acetohydroxamic Acid for Complete and Efficient Separation of Technetium from Uranium Using Liquid-Liquid Extraction," Atalante 2008, Nuclear Fuel Cycles for a Sustainable Future, Montpellier, France, May 2008.

evaluate the electronic structure of the radionuclide species in the extraction system. Initial DFT calculations on the actinides have been performed using an all electron relativistic calculation rather than a relativistic effective core potential. The DFT calculations will provide a means to compare experimental spectroscopy data that investigates structure and electron transitions, providing a basis for examining and comparing the role of 5f electron mechanisms involved in the crucial species in separation systems.

RESEARCH ACCOMPLISHMENTS

- Extractions were performed at constant ionic strength. Aqueous solutions contained HNO₃, UO₂(NO₃)₂, LiNO₃, and the ionic strength was adjusted using NaClO₄. The organic phase consisted of pre-equilibrated 30% TBP/dodecane. It was mixed for 2 minutes, centrifuged, and separated for analysis. Diluted samples were analyzed by IC and ICP-AES.
- Four sets of extractions were performed for determining Eu-DTPA complexation constant for TALSPEAK separations studies. In the aqueous phase, DTPA varied from 1x10⁻⁷ to 5x10⁻⁷ M, ionic strength was 0.1M, pH of 3.60, and trace amount of ¹⁵²Eu. In the organic phase, 2x10⁻⁴ M HDEHP in dodecane pre-equilibrated three times. Analysis was performed on organic and aqueous phases by Liquid Scintillation Counting. The pH was measured in the aqueous phase after equilibration. The electrode was calibrated and adjustments were made in order to determine H⁺ concentration.
- A slug flow problem was identified that affected UV-Vis spectra. The physical positions of the instrumentation was changed to alleviate issue.
- Steady-state conditions were successfully demonstrated in the contactors with a non-radioactive feed solution (after repairing leaks and checking flow rates via mass balances and flow-meters). The radioactive feed solution used after this steady state was achieved, and the data acquired shows hot feed growing in as cold feed is pushed out.
- DFT calculations were performed of the possible equilibrium structures of the UO₂²⁺/NO₃⁻/H₂O system for various concentrations of NO₃⁻ with determination of the corresponding molecular orbital energy levels and population analysis.

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Task 41

Implementation of Uncertainty Propagation in TRITON/KENO

C. Sanders and D. Beller

BACKGROUND

Monte Carlo methods are beginning to be used for three-dimensional fuel depletion analyses to compute various quantities of interest, including isotopic compositions of used nuclear fuel. The TRITON control module, available in the SCALE 5.1 code system, can perform three-dimensional (3-D) depletion calculations using either the KENO V.a or KENO-VI Monte Carlo transport codes, as well as the two-dimensional (2-D) NEWT discrete ordinates code. To overcome problems such as spatially non-uniform neutron flux and non-uniform statistical uncertainties in computed reaction rates and to improve the fidelity of calculations using Monte Carlo methods, uncertainty propagation is needed for depletion calculations.

RESEARCH OBJECTIVES AND METHODS

To enhance and expand the proper/informed use of Monte Carlo methods for 3-D depletion analyses, statistical uncertainty propagation will be developed and implemented in the TRITON/KENO sequence of SCALE. In particular, work will focus on development and implementation of an approach to determine the uncertainty in isotopic predictions based on the compound effects of multiple calculations (depletion time steps) with stochastic uncertainties in the spatial fluxes in each time step. Subsequently, an evaluation of the statistical uncertainties for an actual commercial used fuel sample will be performed to verify the implementation and develop a better understanding of the importance of statistical uncertainties in the prediction of isotopic compositions.

RESEARCH ACCOMPLISHMENTS

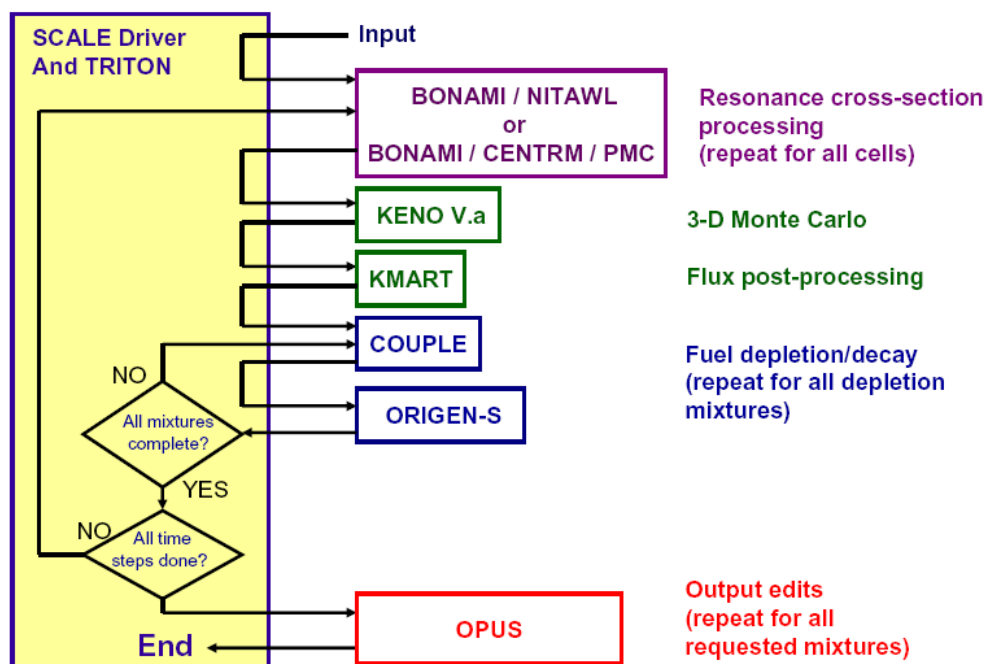
This project was initiated in collaboration with Oak Ridge National Laboratory. The

ACADEMIC YEAR HIGHLIGHTS

- ♦ T. Sloma and C. Sanders, "Evaluation of PWR Rim Effect Utilizing TRITON," oral presentation, 2008 Student Conference of the American Nuclear Society, College Station, TX, February 29, 2008.

first step was initial acquisition of the TRITON/KENO code system from ORNL and identification of a test problem for evaluating the code. An initial test problem was identified and modeled in TRITON and KENO.

Studies of the Pressurized Water Reactor Rim Effect were used to verify the TRITON code implementation. Examination of uncertainty propagation will follow verification and benchmarking studies.



Flowchart of TRITON-K-5 Sequence (photo from the Nuclear Science and Technology Division, Oak Ridge National Laboratory).

Research Staff

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Denis Beller, Co-Principal Investigator, Research Professor, Mechanical Engineering Department

Students

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Quinten Newell, Graduate Student, Ph.D. Candidate, Mechanical Engineering Department (Nuclear Concentration)

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Michael Dunn, Nuclear Data Group Leader, Oak Ridge National Laboratory

Task 42

Monaco/MAVRIC Evaluation for Facility Shielding and Dose Rate Analysis

C. Sanders and D. Beller

BACKGROUND

The dimensions and the large amount of shielding required for Global Nuclear Energy Partnership (GNEP) facilities, advanced radiation shielding, and dose computation techniques are beyond today's capabilities and will certainly be required. With the Generation IV Nuclear Energy System Initiative, it will become increasingly important to be able to accurately model advanced Boiling Water Reactor and Pressurized Water Reactor facilities, and to calculate dose rates at all locations within a containment (e.g., resulting from radiations from the reactor as well as the from the primary coolant loop) and adjoining structures (e.g., from the spent fuel pool).

In complex geometry problems, Monte Carlo methods are often used to compute fluxes or dose rates over large areas using mesh tallies. For problems that demand that the uncertainty in each mesh cell be less than some set maximum, computation time is controlled by the cell with the largest uncertainty. This issue becomes quite troublesome in deep-penetration problems, and advanced variance reduction techniques are required to obtain reasonable uncertainties over large areas. To overcome this issue, Oak Ridge National Laboratory (ORNL) has developed a new sequence, MAVRIC, which will be available with the release of SCALE 6. In this sequence, a methodology called Consistent Adjoint-Driven Importance Sampling (CADIS) has been incorporated for effective variance reduction. This was developed to quickly and automatically determine the biased source distribution and weight windows over a rectangular mesh and a given energy group structure. The method first determines the approximate adjoint particle flux, usually using a discrete ordinates code. The source for this calculation is the detector energy-group response for the process of interest (e.g., dose rate) at the location(s) of interest. The resulting adjoint flux at each location and energy is equated to the importance of particles and is combined with the source distribution to generate the biased source and weight window values that control particle populations at all locations. Very recently, a variation of the CADIS methodology, referred to as the Forward-Weighted CADIS (FW-CADIS) method has been developed, implemented in MAVRIC, and demonstrated for optimization of dose maps.

RESEARCH OBJECTIVES AND METHODS

The MAVRIC sequence is being evaluated along with the Monte Carlo engine MONACO to investigate its effectiveness and use-

ACADEMIC YEAR HIGHLIGHTS

- ♦ Luis Durani, B.S.M.E. (nuclear option) graduated May 2008.

fulness in facility shielding and dose rate analyses. A previously MCNP-evaluated cask array from the Yucca Mountain Project's proposed aging pad and/or buffer area design will be utilized for evaluation and benchmarking purposes. In addition, dose mapping will be performed inside the surface facilities utilizing a transportation cask to evaluate the effectiveness of the code systems. The ability to calculate doses in deep-penetration problems will also be evaluated.

RESEARCH ACCOMPLISHMENTS

The project was initiated in collaboration with ORNL. The first step was initial acquisition of the MONACO/MAVRIC code system from ORNL and identification of a cask array configuration for evaluating the code. 3-D importance/tally mesh was optimized for neutron-photon transport simulation of a single used fuel aging cask. This configuration was modeled in MAVRIC and the model was transmitted to ORNL for review. In addition, inputs were established for a variety of 4x4 cask arrays to match cask storage configurations anticipated at the Yucca Mountain Project. Analyses for the 4x4 aging cask arrays were optimized with respect to dose analysis in between and far from the casks. In addition, the practicality of analyzing a 12 x (4x4) aging cask array is being evaluated with respect to memory limitations.

Other progress:

- Previously encountered memory issues with the SCALE/MONACO/MAVRIC code were resolved by ORNL.
- Dose assessment of a single aging cask was completed. The data indicates excellent agreement to previous data obtained analyzing the same geometry with the MCNP code.
- Complex cask geometry incorporating air vents has been evaluated. As anticipated, the dose rate profile around the cask is slightly different when compared to a simple geometry, and higher dose rates are observed close to the air vents just outside the cask.
- Neutron-neutron and neutron-photon source-response analysis is being examined to determine if it significantly alters the results for much quicker photon-photon analysis.

Research Staff

Charlotta Sanders, Principal Investigator, Research Professor, Mechanical Engineering Department
Denis Beller, Co-Principal Investigator, Research Professor, Mechanical Engineering Department

Students

Steven Simner, Graduate Student, Nuclear Engineering, Mechanical Engineering Department
Luis Durani, Undergraduate Student, Mechanical Engineering Department (Nuclear Engineering Option)

Collaborator

Douglas E. Peplow, Project Technical Advisor, Radiation Transport and Criticality Group, Oak Ridge National Laboratory

UNLV TRANSMUTATION RESEARCH PROGRAM EVENTS

June 18-20, 2007

UNLV hosted the Global Nuclear Energy Partnership Safeguards Working Group meeting.

July 9, 2007

Five researchers and journalists visited UNLV to discuss waste management and nuclear fuel recycling for a project titled “Civil Society and the Nuclear Industry” under the International Visitor Leadership Program.

July 30-August 2, 2007

UNLV co-sponsored the Eighth International Topical Meeting on Nuclear Applications and Utilization of Accelerators (AccApp’07), a joint American Nuclear Society-International Atomic Energy Agency meeting, held at Idaho State University in Pocatello, ID.

August 9-11, 2007

About a dozen students and staff joined others from the local section of the American Nuclear Society for a tour of the San Onofre Nuclear Generating Station, General Atomics DIII-D Fusion Facility, and the USS Topeka Nuclear Submarine.

August 24, 2007

The UNLV Radiochemistry Ph.D. Program conducted a New Graduate Student Orientation that included presentations by the various technical subgroups.

September 10-14, 2007

Thomas Ward, DOE liaison and science adviser, visited UNLV to review Academic Year 2007-2008 student project proposals.

October 1, 2007

Dr. Charlotta Sanders joined the UNLV research faculty. Her expertise is in radiation transport, criticality, and shielding analyses, with substantial experience with investigations related to used nuclear fuel.

October 5-7, 2007

Twelve graduate students from the UNLV Radiochemistry Ph.D. Program and five UNLV faculty members staffed an informational booth at the Town of Pahrump Fall Festival. This is the program’s major community outreach to discuss nuclear issues with the citizens of Nye County, NV.

November 1-15, 2007

Two specialists from the V.G. Khlopin Radium Institute visited UNLV to perform maintenance on the Neutron Multiplicity Detector System and to examine it in preparation for modifications to add a capability for coincidence/anti-coincidence detection of charged particles.

November 3, 2007

Graduate students from the UNLV Radiochemistry Ph.D. Program host and participate in the Nuclear Science Merit Badge Day at UNLV and the Atomic Testing Museum.

January 7-11, 2008

Twenty-four people from across the country attended the MCNPX training held at UNLV.

February 14, 2008

UNLV TRP Director Anthony Hechanova presented the UNLV



UNLV Students toured the control room at the General Atomics DIII-D Fusion Facility in La Jolla, CA on August 10, 2007.

Health Physics Seminar entitled “Transmutation Research Program: Recycling Waste Management Strategy.”

February 22, 2008

UNLV hosted the Second Integrated Symposium between UNLV and NSTec on “Nuclear Forensics.”

February 28-29, 2008

The UNLV Radiochemistry Ph.D. program hosted six prospective students during a 2-day recruitment event.

March 5, 2008

Several UNLV faculty members attended the GNEP Safeguards Working Group Meeting at the Pacific Northwest National Laboratory in Richland, WA.

May 1-2, 2008

Faculty visited the Waste Isolation Pilot Plant in Carlsbad, NM, and discussed common issues.

May 14, 2008

UNLV TRP Director gave an invited presentation titled “Used Fuel Recycling and Advanced Fuel Cycles” to the Nevada Legislature Committee on High Level Nuclear Waste, Las Vegas, NV.

May 14-16, 2008

Rob Price, DOE Advanced Fuel Cycle Research and Development Program Manager, and Craig Martin (Savannah River National Laboratory) visited UNLV to tour facilities, and meet with faculty and students.

May 15-16, 2008

UNLV hosted the Global Nuclear Energy Partnership/Advanced Fuel Cycle Research and Development Campaign Manager’s meeting.

May 2008

UNLV and University of New Mexico conducted a Reactor Experimentation Laboratory Course at University of New Mexico in Albuquerque, NM, using their AGN 201M reactor.

Transmutation Research Program Financial Statement

| Program Administration | FY01 | FY02 | FY03 | FY04 | FY05 | FY06 | FY07 | TOTAL |
|--------------------------------------|--------------------|--------------------|--------------------|--------------------|--------------------|--------------------|--------------------|---------------------|
| Program Support | | | | | | | | |
| Labor | \$187,305 | \$348,324 | \$564,755 | \$452,274 | \$575,395 | \$628,000 | \$680,000 | \$3,436,053 |
| QA Program | \$0 | \$0 | \$0 | \$0 | \$0 | \$0 | \$115,000 | \$115,000 |
| Other Costs | \$121,779 | \$72,336 | \$115,047 | \$108,039 | \$136,638 | \$59,703 | \$120,000 | \$733,542 |
| LANL Subcontract | \$150,000 | \$162,500 | \$0 | \$0 | \$0 | \$0 | \$0 | \$312,500 |
| SRNL Subcontract | \$0 | \$0 | \$0 | \$0 | \$70,250 | \$0 | \$0 | \$70,250 |
| Infrastructure Augmentation | | | | | | | | |
| New Hires | \$112,692 | \$240,648 | \$433,832 | \$391,028 | \$691,846 | \$428,000 | \$428,177 | \$2,726,223 |
| Facilities | \$1,492,446 | \$756,709 | \$838,661 | \$604,933 | \$394,910 | \$515,792 | \$580,160 | \$5,183,611 |
| Academic Programs | \$0 | \$0 | \$0 | \$179,122 | \$154,527 | \$82,642 | \$120,000 | \$536,291 |
| International Collaboration | \$144,973 | \$377,175 | \$155,200 | \$121,700 | \$13,565 | \$70,000 | \$112,000 | \$994,613 |
| Subtotal | \$2,209,195 | \$1,957,692 | \$2,107,495 | \$1,857,096 | \$2,037,131 | \$1,784,137 | \$2,155,337 | \$14,108,083 |
| Student Research Tasks Totals | \$790,805 | \$2,542,308 | \$1,752,505 | \$1,642,904 | \$1,930,869 | \$2,175,863 | \$1,934,663 | \$12,769,917 |
| Totals | \$3,000,000 | \$4,500,000 | \$3,860,000 | \$3,500,000 | \$3,968,000 | \$3,960,000 | \$4,090,000 | \$26,878,000 |

Student Research Task by Task

| | FY01-06 | | FY04-06 | FY07 | TOTAL | | FY05 | FY06 | FY07 | TOTAL |
|---------|----------------|---------|----------------|-------------|--------------|---------|-------------|-------------|-------------|--------------|
| Task 1 | \$424,291 | Task 17 | \$470,944 | \$0 | \$470,944 | Task 28 | \$119,694 | \$77,097 | \$83,323 | \$280,114 |
| Task 2 | \$505,568 | Task 18 | \$557,853 | \$79,818 | \$637,671 | Task 29 | \$151,217 | \$100,596 | \$48,660 | \$300,473 |
| Task 3 | \$576,563 | Task 19 | \$362,098 | \$26,101 | \$388,199 | Task 30 | \$144,201 | \$126,420 | \$187,188 | \$457,809 |
| Task 4 | \$432,679 | Task 20 | \$412,656 | \$27,290 | \$439,946 | Task 31 | \$0 | \$90,973 | \$8,458 | \$99,431 |
| Task 5 | \$274,994 | Task 21 | \$352,800 | \$71,863 | \$424,663 | Task 32 | \$0 | \$92,511 | \$0 | \$92,511 |
| Task 6 | \$369,218 | Task 22 | \$169,742 | \$1,647 | \$171,389 | Task 33 | \$0 | \$178,666 | \$333,513 | \$512,179 |
| Task 7 | \$453,388 | Task 23 | \$294,908 | \$1,257 | \$296,165 | Task 34 | \$0 | \$176,199 | \$116,405 | \$292,604 |
| Task 8 | \$350,090 | Task 24 | \$270,940 | \$81,206 | \$352,146 | Task 35 | \$0 | \$134,785 | \$189,291 | \$324,076 |
| Task 9 | \$221,002 | Task 25 | \$283,870 | \$3,192 | \$287,062 | Task 36 | \$0 | \$15,680 | \$74,483 | \$90,163 |
| Task 10 | \$243,371 | Task 26 | \$375,341 | \$0 | \$375,341 | Task 37 | \$0 | \$0 | \$120,776 | \$120,776 |
| Task 11 | \$181,019 | Task 27 | \$305,152 | \$11,237 | \$316,389 | Task 38 | \$0 | \$0 | \$118,980 | \$118,980 |
| Task 12 | \$281,975 | | | | | Task 39 | \$0 | \$0 | \$61,724 | \$61,724 |
| Task 13 | \$424,247 | | | | | Task 40 | \$0 | \$0 | \$201,043 | \$201,043 |
| Task 14 | \$313,998 | | | | | Task 41 | \$0 | \$0 | \$43,826 | \$43,826 |
| Task 15 | \$268,484 | | | | | Task 42 | \$0 | \$0 | \$43,382 | \$43,382 |
| Task 16 | \$250,024 | | | | | | | | | |

Infrastructure Facilities Augmentation

| | FY01-02 | FY03 | FY04 | FY 05 | FY06 | FY07 | Total |
|--|--------------------|------------------|------------------|------------------|------------------|------------------|--------------------|
| Electron Microanalysis and Imaging Laboratory | \$105,313 | \$63,000 | \$63,000 | \$63,000 | \$62,160 | \$62,160 | \$418,633 |
| Materials Performance Laboratory | \$227,493 | \$0 | \$0 | \$0 | \$0 | \$0 | \$227,493 |
| Transmission Electron Microscope | \$1,459,369 | \$321,582 | \$150,000 | \$166,914 | \$135,000 | \$150,000 | \$2,382,865 |
| ICP – Atomic Emission Spectrometer | \$100,163 | \$0 | \$0 | \$0 | \$0 | \$0 | \$100,163 |
| Lead-Bismuth and Other Molten Materials Facilities | \$234,198 | \$121,579 | \$217,227 | \$66,843 | \$56,027 | \$65,000 | \$760,874 |
| Machining System and Equipment | \$72,619 | \$22,500 | \$934 | \$0 | \$0 | \$0 | \$96,053 |
| Prof. B.J. Das Startup Package Equipment | \$50,000 | \$0 | \$49,972 | \$0 | \$0 | \$0 | \$99,972 |
| Radiochemistry Laboratories | \$0 | \$260,000 | \$123,800 | \$98,153 | \$262,605 | \$303,000 | \$1,047,558 |
| Oxygen Control System | \$0 | \$50,000 | \$0 | \$0 | \$0 | \$0 | \$50,000 |
| Totals | \$2,249,155 | \$838,661 | \$604,933 | \$394,910 | \$515,792 | \$580,160 | \$5,183,611 |

International Collaboration

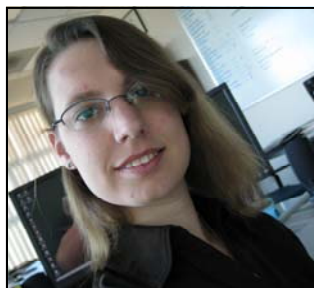
| | FY01-02 | FY03 | FY04 | FY 05 | FY06 | FY07 | Total |
|---|------------------|------------------|------------------|-----------------|-----------------|------------------|------------------|
| Khlopin Radium Institute Contracts | \$342,550 | \$23,950 | \$0 | \$0 | \$0 | \$102,000 | \$468,500 |
| International Science and Technology Center | \$87,500 | \$98,250 | \$51,700 | \$800 | \$0 | \$0 | \$238,250 |
| International Collaboration Program | \$92,098 | \$33,000 | \$70,000 | \$12,765 | \$70,000 | \$10,000 | \$287,863 |
| Totals | \$522,148 | \$155,200 | \$121,700 | \$13,565 | \$70,000 | \$112,000 | \$994,613 |

Photo Gallery

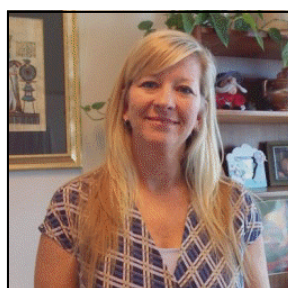
Transmutation Research Program Administration



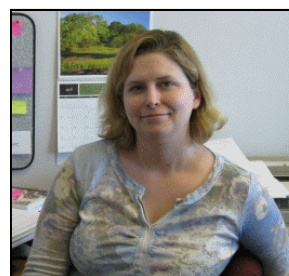
**TRP Director
Anthony E. Hechanova**



**Administrative Assistant
Rebecca Paulson**



**TRP Finance Director
Leisa Rodriguez**



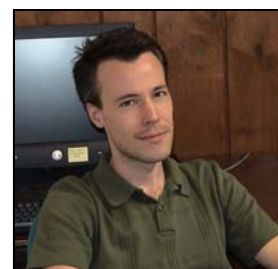
**Finance Assistant
Sandra Boydston**



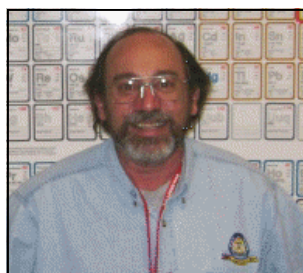
**Finance Assistance
Hallie Lyons**



**HRC Travel Coordinator
Avrina Smith**



**HRC Network Admin
Kris Davidson**



**Laboratory Director
Thomas O'Dou**

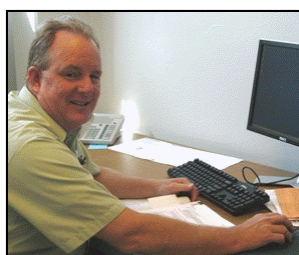
Not pictured:
Joseph Zimmerman
John Knoten



**Laboratory Manager
Trevor Low**



**Project Assistance
Kathy Lauckner**



**Program Developer
Steven Curtis**

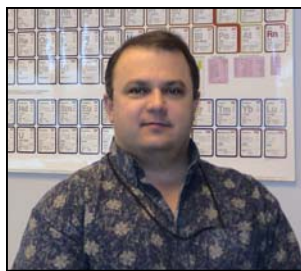


**International Programs
Adviser
Thomas Ward**

Staff, Faculty Researchers and Students



**Group Leader
Radiochemistry
Ken Czerwinski**



**Asst. Research Prof.
Radiochemistry
Gary Cerefice**



**Asst. Research Prof.
Radiochemistry
Tyler Sullens**



**Visiting Professor
Radiochemistry
Al Sattelberger**



**Group Coordinator
Materials
Ning Li (LANL)**



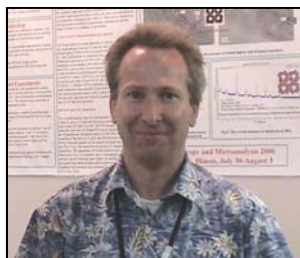
**Group Leader
Materials & Chemistry
Allen Johnson**



**Asst. Research Prof.
Molten Materials
Jian Ma**



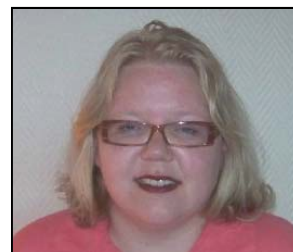
**Asst. Research Prof.
Materials
Longzhou Ma**



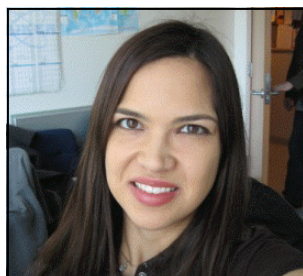
**Asst. Research Prof.
Materials
Thomas Hartmann**



**Group Leader
Nuclear Engineering
Denis Beller**



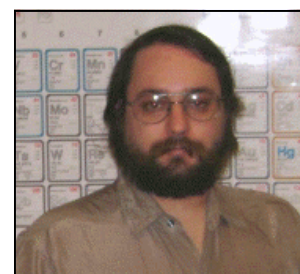
**Asst. Research Prof.
Nuclear Engineering
Charlotta Sanders**



**PhD Researcher
Cynthia Gong**



**PhD Researcher
Frederic Poineau**



**PhD Researcher
Daniel Rego**

Not pictured:
Leslie Jardine, Russian Program Coordinator

Task 1:



Yitung Chen
Associate Professor
Associate Director, NCACM
Mechanical Engineering Department



Darrell W. Pepper
Professor
Mechanical Engineering Department
Director, NCACM



Taide Tan



Randy Clarksean
Adjunct Professor
Mechanical Engineering Department

Yulien Chen
Paul Lawson

Not pictured



Xiaolong Wu

Task 2:



Robert A. Shilll, Jr.
Associate Professor
Electrical and Computer
Engineering Department



William Culbreth
Associate Dean for Research
College of Engineering

Mohamed B. Trabia
Chair
Mechanical Engineering
Department



Anoop George



Satishkumar
Subramanian



Qin Xue



Myong Holl



Greg Loll

Task 3:



John Farley
Professor
Department of Physics



Dale Perry
Adjunct Professor
Lawrence Berkeley National Lab

Allen Johnson
Assistant Professor
Department of Chemistry



Umar Younas



Brian Hosterman



Dan Koury



Thao Trung Ho



Julie Manzerova



Denise Parsons



Lindsay Wylie



Chris Harland



Stacy Sidle

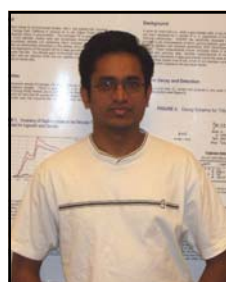
Task 4:



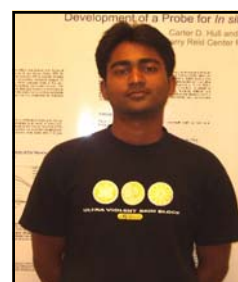
Ajit Roy
Professor
Department of Mechanical Engineering



Mohammad Hossain



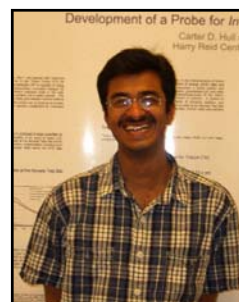
Sudheer Sama



Venkataramakrishnan
Selvaraj



Ramprashad Prabhakaran



Phandi Gudipati

Nikata Agarwal
Aaron Tippetts

Not pictured

Task 5:



Samir Moujaes
Associate Professor
Department of Mechanical Engineering

Yitung Chen
Associate Professor
Associate Director, NCACM
Department of Mechanical Engineering



Kanthi Dasika



Chao Wu

Narain Armbya

Not pictured



Guanjun Li

Task 6:



Denis Beller
Research Professor
Department of Mechanical Engineering



Timothy Beller



Brice Howard



Dean Curtis



Steve Curtis



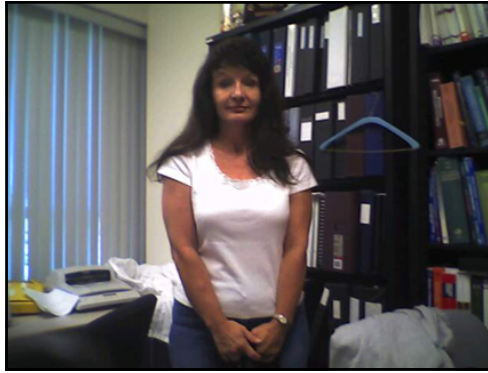
Shruti Patil

Task 7:



Phillip Patton
Associate Professor
Department of Health Physics

Mark Rudin
Associate Professor
Department of Health Physics



Deanna Tuttle

Tserenpagma Chaoui
Ashley Gann
Sung Yop Kim
Jeffrey Wyler
John Shanahan
Yayun Song

Not pictured

Task 8:



Yitung Chen
Associate Professor
Associate Director, NCACM
Mechanical Engineering Department

Sean Hisieh
Research Associate Professor
Mechanical Engineering Department



Sushma Gujjula
Sridhar Munaga

Not pictured



Darrell W. Pepper
Professor
Mechanical Engineering Department
Director, NCACM



Jianhong Li



Haritha Royyuru

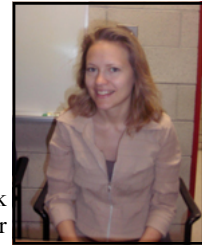


Lijian Sun

Task 9:



Georg Mauer
Professor
Department of Mechanical Engineering



Caroline Wiejak
Visiting Scholar



Richard Silva



Jae-Kyu Lee



Jamil Renno

Task 10:



Ajit Roy
Professor
Department of Mechanical
Engineering

Brendan O'Toole
Associate Professor
Department of Mechanical Engineering



Venkata Potluri



Srinivas Kukatla

Prad Koppula
Raymond Kozak

Not pictured



Mark Jones



John Motaka



Martin Lewis



Bhagath Yarlagadda

Tasks 11 & 12:



William Culbreth
Associate Dean for Research
College of Engineering

Denis Beller
Research Professor
Mechanical Engineering Department



Robin Jenkins
Ashraf Kaboud

Not pictured



Robert O'Brien



Trevor Wilcox



Suresh Sadineni

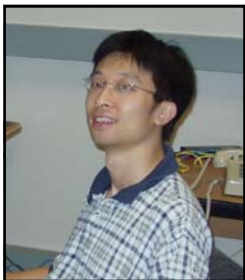


Daniel Lowe



Elizabeth Bakker

Task 13:



Yingtao Jiang
Assistant Professor
Department of Electrical and Computer Engineering

Bingmei Fu
Assistant Professor
Department of Mechanical Engineering



Yi Lu
Shahidur Rahaman

Not pictured



Jian Ma
Assistant Research Professor
Department of Mechanical Engineering



Xiaolong Wu

Task 14:



Ajit Roy
Professor
Department of Mechanical Engineering



Vikram Marthandam



Satish Dronavalli

Raghunandan Karamcheti
Bhagath Yarlagadda

Not pictured



Silpa Budugur



Anand Venkatesh



Srinivas Chanda



Subhra Bandyopadhyay

Task 15:

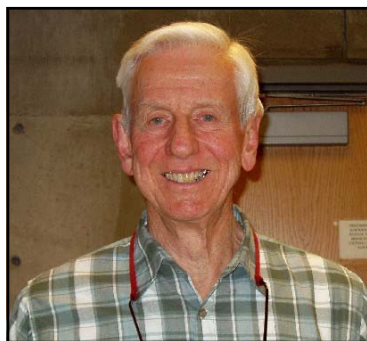


Spencer Steinberg
Professor
Department of Chemistry

Gary Cerefice
Research Scientist
Harry Reid Center



Ginger Kimble



David Emerson
Emeritus Professor
Department of Chemistry

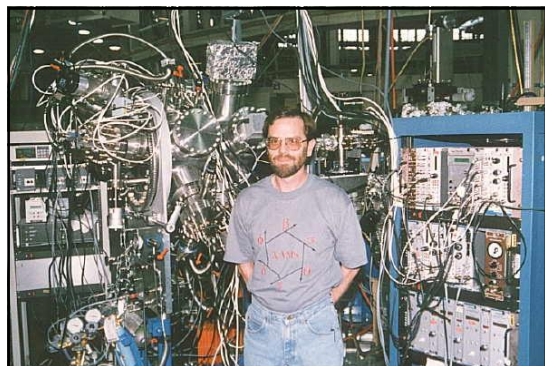


James Dorman



Nancy Birkner

Task 16:



Dennis Lindle
Professor
Department of Chemistry



Dale Perry
Adjunct Professor
Department of Physics



Oliver Hemmers
Assistant Research Professor
Department of Chemistry

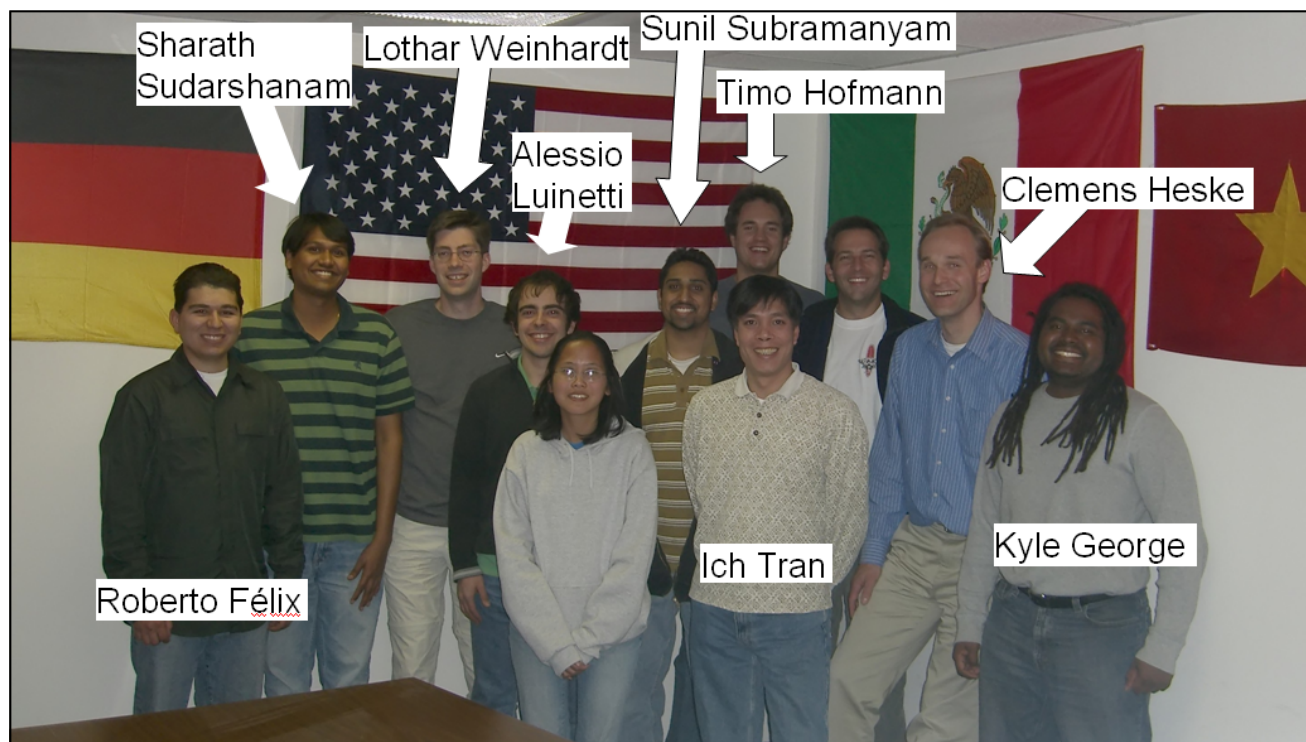


Chinthaka Silva



Chirantha Rodrigo

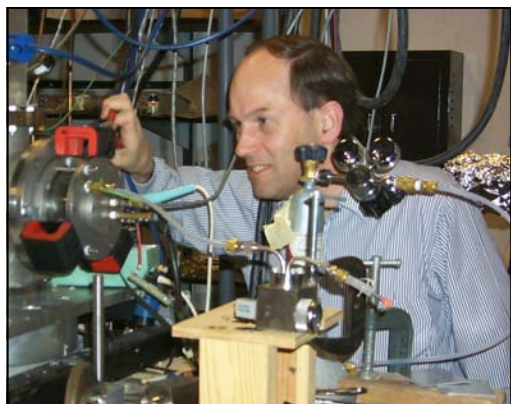
Task 17:



Lothar Weinhardt
Post-Doctoral Fellow
Department of Chemistry

Clemens Heske
Associate Professor
Department of Chemistry

Task 18:



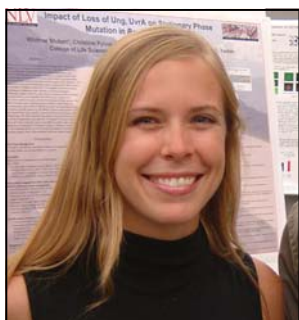
John Farley
Professor
Department of Physics



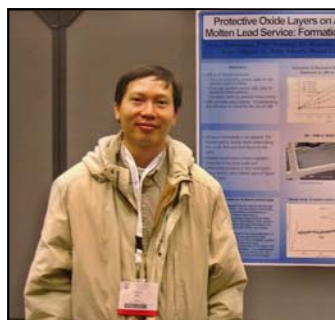
Allen Johnson
Assistant Professor
Department of Chemistry



Dan Koury



Jenny Welch



Thao Trung Ho



Brian Hostermann

Task 19:



Ken Czerwinski
Associate Professor
Department of Chemistry



Frederic Poineau



Kiel Holliday

Task 20:



Ajit Roy
Professor
Department of Mechanical Engineering

Vinay Virupaksha

Not pictured



Harish Krishnamurthy



Pankaj Kumar



Sreenivas Kohir



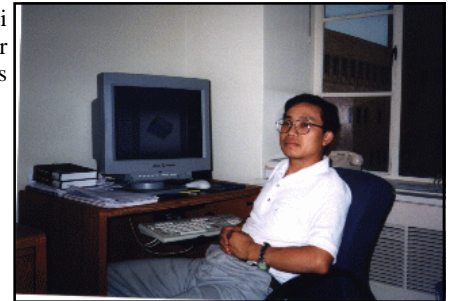
Debajyoti Maitra

Task 21:



Yitung Chen
Associate Professor
Department of Mechanical Engineering

Jichun Li
Assistant Professor
Department of Mathematics



Chaigyod Soontrapa



Taide Tan

Task 22:



Georg Mauer
Professor
Department of Mechanical Engineering

Kamalaku Aluri
Kyle Gohres
Tobias Kotthaeuser
Prathima Sajja

Not pictured



Koushik Damera



Douglas Wettekin



Jamil Renno



Andreas Roderburg

Task 23:



Biswajit Das
Associate Professor
Department of Electrical and Computer
Engineering

Karina Diestra
Nilanjan Halder
Rani Krishna
S. Mookerjea
Lillian Ratliff
Kishire Sreenivasan
Michael Sadowitz
Ich Con Tran
Varadarajan

Not pictured



Jonathan Ross



Pavan Singaraju



Tan Wu



Arghya Banerjee

Task 24:



Yitung Chen
Associate Professor
Department of Mechanical Engineering

Sean Hisieh
Research Associate Professor
Mechanical Engineering Department



Matthew Hodges

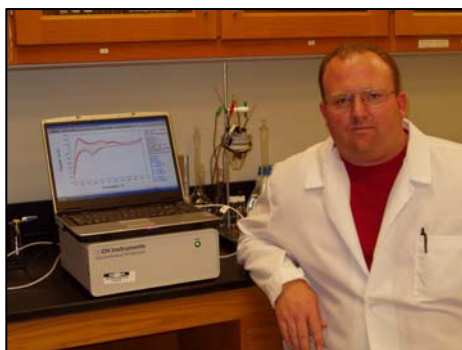


Ming Chang



Jen-Yuan Huang

Task 25:



David Hatchett
Assistant Professor
Department of Chemistry

Ken Czerwinski
Associate Professor
Department of Chemistry



Patrick Houlihan
Moustapha Eid Moustapha

Not pictured



Sandra Elkouz



Sujanie Gamage



Wendy Pemberton

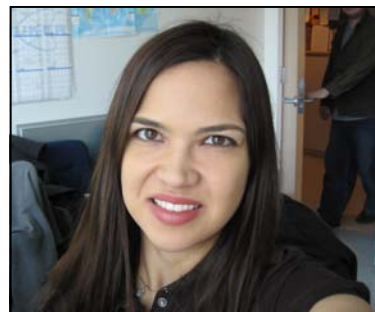
Task 26:



Ken Czerwinski
Associate Professor
Department of Chemistry



Frederic Poineau



Cynthia Gong



Wendy Pemberton

Inna Bashta
Mari Bonaud
Michel Draye
Jeremy Maute
Jade Morgan
Trent Ohman
Ira Racoma
Lena Wright

Not pictured



Amber Wright



Amber Wright

Task 27:



Denis Beller
Research Professor
Department of Mechanical Engineering



Evgeny Stankovskiy



Brice Howard



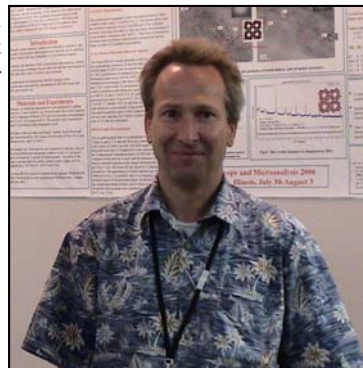
Timothy Beller & Ryan LeCounte set up the High-Power RACE Target experiment at the Idaho Accelerator Center.

Task 28:



Ken Czerwinski
Associate Professor
Department of Chemistry

Thomas Hartmann
Staff Scientist
Harry Reid Center



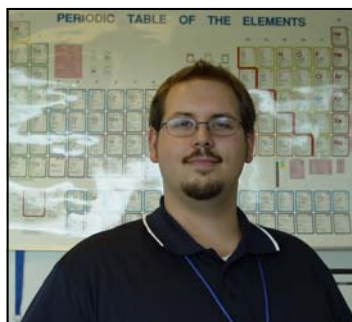
Chinthaka Silva

Task 29:

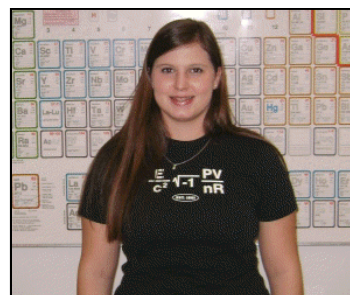


Gary Cerefice
Research Scientist
Harry Reid Center

Ken Czerwinski
Associate Professor
Department of Chemistry



Nicholas Smith



Jamie Warburton

Task 30:



Denis Beller
Research Professor
Department of Mechanical Engineering



Charlotta Sanders
Research Assistant Professor
Department of Mechanical Engineering



Timothy Beller

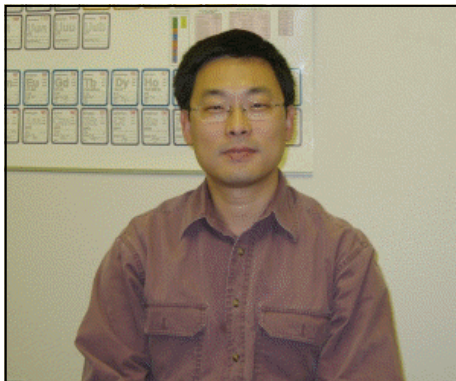
Sandra De La Cruz
Luis Durani
Lawrence Lakeotes
Catherine Romano

Not pictured



Quinten Newell

Task 31:



Jian Ma
Assistant Research Professor
Department of Mechanical Engineering



Woosoon Yim
Professor
Department of Mechanical Engineering

Joon Soo Lee
Assistant Research Professor
Department of Mechanical Engineering



Xiuju (Julia) Tan

Task 32:



Daniel Cook
Assistant Professor
Department of Mechanical
Engineering

Lucas Bang
Lillian Ratliff
Siul Ruiz
Karthek Yalamanchili

Not pictured



Huajun Chen
Postdoctoral Researcher
Department of Mechanical
Engineering



Yitung Chen
Associate Professor
Department of Mechanical Engineering

Jian Ma
Assistant Research Professor
Department of Mechanical Engineering



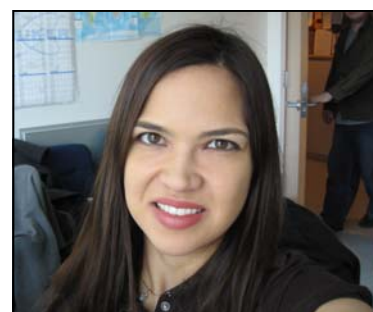
Task 33:



Ken Czerwinski
Associate Professor
Department of Chemistry



Frederic Poineau



Cynthia Gong



Ed Mausolf



Maryline Ferrier

Task 34:



Ken Czerwinski
Associate Professor
Department of Chemistry



Dan Rego

Task 35:



Denis Beller
Research Professor
Department of Mechanical Engineering



Charlotta Sanders
Research Assistant Professor
Department of Mechanical Engineering

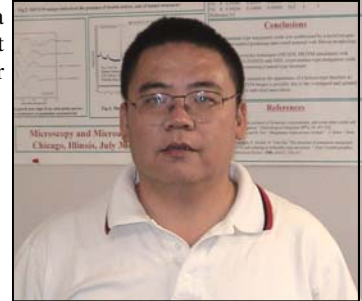
Sandra De La Cruz
Lawrence Lakeotes
Ryan LeCounte
Tanya Sloma

Not pictured

Task 36:



Gary Cerefice
Research Scientist
Harry Reid Center

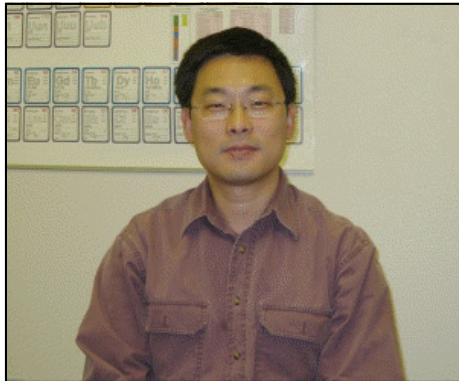


Longzhuo Ma
Research Scientist
Harry Reid Center

KM Nazmul Ahsan

Not pictured

Task 37:



Jian Ma
Assistant Research Professor
Department of Mechanical Engineering

Yingtao Jiang
Assistant Professor
Department of Electrical & Computer Engineering

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Ye Ai
Taleb Moazzeni
Rohit Reddy Saddi

Not pictured

Task 38:



David Hatchett
Assistant Professor
Department of Chemistry

Ken Czerwinski
Associate Professor
Department of Chemistry



Wendy Pemberton

Task 39:



Sean Hisieh
Research Associate Professor
Mechanical Engineering Department



Chang Yeol Lee

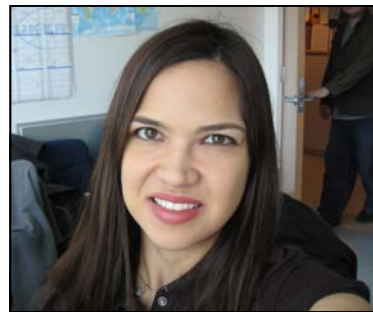
Task 40:



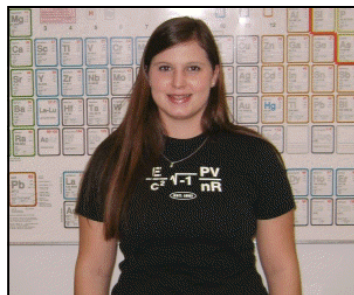
Ken Czerwinski
Associate Professor
Department of Chemistry



Frederic Poineau



Cynthia Gong



Jamie Warburton

Solenne Michaud

Not pictured



Amber Wright

Task 41:



Denis Beller
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Department of Mechanical Engineering



Charlotta Sanders
Research Assistant Professor
Department of Mechanical Engineering



Quinten Newell

Tanya Sloma
Matthew Voegelle

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Task 42:



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Department of Mechanical Engineering



Charlotta Sanders
Research Assistant Professor
Department of Mechanical Engineering

Luis Durani
Steven Simner

Not pictured

Acronyms

| | | | |
|-------|--|-----------|---|
| ABR | Advanced Burner Reactor | HAZ | Heat Affected Zone |
| ACS | American Chemical Society | HDEHP | Bis(2-ethylhexyl)phosphoric acid |
| ADO | ActiveX Data Objects | HE | Hydrogen embrittlement |
| ADSS | Accelerator-driven subcritical systems | HOPG | Highly Ordered Pyrolytic Carbon |
| AFC | Advanced Fuel Cycle | HR | High resolution |
| AFCI | Advanced Fuel Cycle Initiative | HTGR | High Temperature Gas-Cooled Reactor |
| AFM | Atomic Force Microscopy | IAC | Idaho Accelerator Center, Idaho State University |
| AHA | Acetohydroxamic acid | | |
| ALI | Annual limit on intake | ICONE | International Conference on Nuclear Engineering |
| ALIP | Annular, Linear Induction Pump | ICRP | International Commission on Radiological Protection |
| AMAD | Activity median aerodynamic diameter | IEEE | Institute of Electrical and Electronics Engineers, Inc. |
| AMPT | Advance in Materials and Processing Technologies | INL | Idaho National Laboratory |
| AMUSE | Argonne Model for Universal Solvent Extraction | IPPE | Institute for Physics and Power Engineering |
| ANL | Argonne National Laboratory | ISA | Information Search and Analysis |
| ANS | American Nuclear Society | ISOPro | Integrated System Optimization Program |
| ASM | American Society of Metals | ISRS | International Symposium of Research Scholars |
| ASME | American Society of Mechanical Engineers | ISU | Idaho State University |
| ASTM | American Society for Testing and Materials | KR | Knowledge Reporting |
| CA | Cellular Automata | KRI | Khlopin Radium Institute, St. Petersburg, Russia |
| CAD | Computer aided design | KRI-KRISI | Khlopin Radium Institute Research Industrial Enterprise, St. Petersburg, Russia |
| CADIS | Consistent Adjoint-Driven Importance Sampling | | |
| CEA | French Atomic Energy Commission | LANL | Los Alamos National Laboratory |
| CFD | Computational fluid dynamics | LANSCE | Los Alamos Neutron Science Center |
| CL | Constant load | LBE | Lead-bismuth eutectic |
| CSDS | Carbon SDS | LBL | Lawrence Berkeley National Laboratory |
| CTD | Chemical Technology Division | LFS | Laser Fluorescence Spectroscopy |
| DAC | Derived air concentration | LWR | Light Water Reactor |
| DC | Dose coefficient | MCNP | Monte Carlo n-particles code |
| DCAL | Dose and risk calculation software | MCNPX | Monte Carlo n-particles code, extremely high-energy version |
| DCD | Design Class Diagrams | MCSPS | Monitoring, Controlling, and Scram Protection System |
| DFT | Density Functional Theory | MD | Molecular Dynamics |
| DM | Data Manipulation | MHD | Magnetohydrodynamic |
| DOE | U.S. Department of Energy | MISDAR | Materials Information System for Design, Analysis, and Reporting |
| DSA | Dynamic Strain Ageing | MPAC | Materials Protection, Accounting, and Control |
| DSC | Differential scanning calorimetry | MPI | Message passing interface |
| DTPA | diethylenetriaminepentaacetic | MS&T | Materials Science and Technology |
| ECS | Electrochemical Society | MS SQL | Microsoft Standard Query Language |
| EDO | Experimental and Developmental Organization | MS VB | Microsoft Visual Basic |
| EDX | Energy Dispersive X-Ray Analysis | MSZ | Magnesia Stabilized Zirconia |
| %El | Percent elongation | NACE | National Association of Corrosion Engineers |
| EM | Electromagnetic | NETL | Nuclear Engineering Teaching Laboratory |
| ENSDF | Evaluated nuclear structure data file | NMDS | Neutron multiplicity detector system |
| ESD | Engineering-scale Demonstration | NMR | Nuclear Magnetic Resonance |
| FCC | Fullerene containing carbon | NOM | Natural organic matter |
| FEM | Finite Element Method | NSC | Nuclear Science Center |
| FGR | Federal Guidance Report | | |
| FL | Fusion-Line | | |
| FLEX | Fluoride extraction process | | |
| GIF | GEN IV International Forum | | |
| GNEP | Global Nuclear Energy Partnership | | |
| GRG | Generalized Reduced Gradient | | |
| GUI | Graphical user interface | | |
| HA | Higher Actinides | | |

| | | | |
|-----------|--|--------|--|
| NTS | Nevada Test Site | UREX | Uranium extraction process |
| NUBASE | A nuclear properties database | UREX+ | Uranium extraction process plus |
| OCS | Oxygen Control System | UT | University of Texas |
| ORNL | Oak Ridge National Laboratory | UV | Ultra-violet |
| OTA | Oxygen Thermodynamic Activity | UV-Vis | UV-visible |
| PAS | Positron annihilation spectroscopy | WDX | Wavelength Dispersed X-Ray |
| PID | Proportional-Integral-Derivative | XAFS | X-ray absorption fine-structure spectroscopy |
| PNNL | Pacific Northwest National Laboratories | XEDS | X-Ray Energy Dispersive Spectrometry |
| PRM | Peak Ratio Method | XML | Extensible markup language |
| PUREX | Plutonium extraction process | XPS | X-ray photoelectron spectroscopy |
| PVM | Parallel virtual machine | XRD | X-ray diffraction |
| %RA | Percent reduction in area | XRF | X-Ray Fluorescence |
| RACE | Reactor-accelerator coupled experiment | YS | Yield strength |
| RCRA | Resource Conservation and Recovery Act | YSZ | Ytria-stabilized zirconia |
| R&D | Research and Development | | |
| RELAP | Plant Safety Modeling Code | | |
| RGA | Residual gas analyzer | | |
| RPI | Rensselaer Polytechnic Institute | | |
| RSL | Remote Sensing Laboratory | | |
| RT | Room temperature | | |
| RTIL | Room Temperature Ionic Liquid | | |
| SAD | Selected Area Diffraction | | |
| SAMPE | Society for the Advancement of Materials and Process Engineering | | |
| SCC | Stress corrosion cracking | | |
| SD | System Design | | |
| SDP | Sputter Depth Profiling | | |
| SDS | Slowing Down Spectrometer | | |
| SEE | Secondary electron emission | | |
| SEM | Scanning electron microscope | | |
| SFR | Sodium-Cooled Fast Reactor | | |
| SFTF | Spent Fuel Treatment Facility | | |
| SRNL | Savannah River National Laboratory | | |
| SS | Stainless steel | | |
| SSR | Slow-strain-rate | | |
| STAR-CD | A commercial computational fluid dynamics software package | | |
| STFT | Spent Fuel Treatment Facility | | |
| SXF | Soft X-ray Fluorescence | | |
| TALSPEAK | Trivalent Actinide Lanthanide Separation by Phosphorous Regent Extraction by Aqueous Complexes | | |
| TBP | Tributylphosphate | | |
| TC-1 | Target Complex 1 | | |
| TEM | Transmission electron microscope | | |
| TG/DSC | Thermogravimetry and Differential Scanning Calorimetry | | |
| TRIGA | Training, Research, Isotopes, General Atomics | | |
| TRISO | A silicon carbide and pyrocarbon composite coating | | |
| TRP | UNLV Transmutation Research Program | | |
| TRPSEMPro | TRP System Engineering Model Program | | |
| TRU | Transuranic actinides | | |
| TTF | Time-to-failure | | |
| UHV | Ultra High Vacuum | | |
| UML | Unified Modeling Language | | |
| UNLV | University of Nevada, Las Vegas | | |
| UNM | University of New Mexico | | |
| UPS | UV-Photoemission | | |

References

UNLV Transmutation Research Program references, current activities and other miscellaneous information can be found on the TRP website at:
<http://aaa.nevada.edu>

Credits

The University of Nevada, Las Vegas Transmutation Research Program is administered by the Harry Reid Center for Environmental Studies under the auspices of the U.S. Department of Energy (Grant No. DE-FG07-01AL67358 & Grant No. DE-FC07-06ID14781).

This report was produced by Anthony Hechanova and Rebecca Paulson.

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