2011 Fuel Cycle Technologies Annual Review Meeting

Transactions Report

Argonne National Laboratory

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Fuel Cycle Technologies Program: Introduction

As the largest domestic source of low-carbon energy, nuclear power is making major contributions toward meeting our nation's current and future energy demands. The United States must continue to ensure improvements and access to this technology so we can meet our economic, environmental and energy security goals. We rely on nuclear energy because it provides a consistent, reliable and stable source of base load electricity with an excellent safety record in the United States. To support nuclear energy's continued and expanded role in our energy platform, therefore, the United States must continually improve its knowledge, technology, and policy in order to:

- Support the safe and secure use of nuclear energy and its associated technologies.
- Support transportation, storage and disposal of used nuclear fuel and associated wastes
- Enhance the safety of nuclear energy and stored used nuclear fuel in response to extreme events (e.g., the events at Fukushima-Daiichi have raised calls to develop technologies that improve accident tolerance).
- Improve the long-term sustainability of nuclear energy.

The use of nuclear energy in the U.S. has generated over 60,000 tons of used nuclear fuel and continues to generate approximately 2,000 additional tons per year. The Federal government has the responsibility for management of the nation's commercial used nuclear fuel up to and including permanent disposal.

To address this need, the Department of Energy (DOE) Office of Nuclear Energy (NE) Fuel Cycle Technologies (FCT) program is charged with developing used fuel and waste management strategies and sustainable fuel cycle options that improve resource utilization, responsibly manage wastes, improve safety, and limit proliferation risk. To accomplish this mission, FCT has established short-, intermediate- and long-term strategic goals:

- In the near term, FCT will address the BRC recommendations for used fuel management, increase its focus on nuclear fuels with enhanced accident tolerance, and identify sustainable fuel cycle options for further development.
- In the intermediate term, the program will conduct science-based, engineering-driven research for sustainable fuel cycle options, conduct research to support extended storage of used nuclear fuel, and develop the scientific basis for determining sites and procedures for disposal of used nuclear fuel.

- In the long term, the program will demonstrate specific fuel cycle technologies, establish confidence in extended used fuel storage capabilities, and support the selection and licensing of potential used fuel disposal sites.

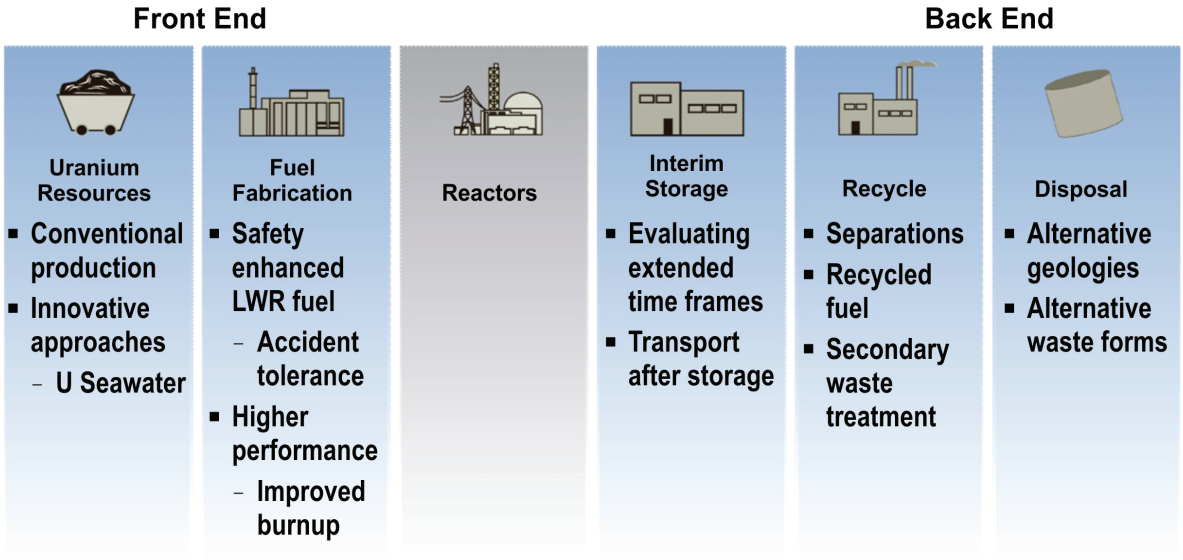

Figure 1: Illustration of the nuclear fuel cycle.

To achieve its mission, FCT has initiated numerous activities in each of the technical campaign areas, of which this report provides a sample. National laboratory and university experts will present these activities in greater detail during the annual review meeting. Discussions include the following technical areas, along with a cross-section of nuclear fuel cycle topics as shown on Figure 1:

- Development of high burnup fuel and cladding materials to withstand irradiation for longer periods of time with improved accident tolerance
- Development of simplified separations, waste management (including storage, transportation, and disposal) and proliferation risk reduction methods
- Development of processes and tools to evaluate sustainable fuel cycle system options and to effectively communicate evaluation results to stakeholders

Used Fuel Disposition Campaign

1.1 Used Fuel Disposition Campaign Overview

Mission

Identify alternatives and conduct scientific research and technology development to enable storage, transportation, and disposal of used nuclear fuel and wastes generated by existing and future nuclear fuel cycles.

Objectives

Near-Term Objectives

- Identify and prioritize gaps in the current technical bases for extended storage of existing used nuclear fuel (UNF).
- Initiate experimentation and modeling activities to address high-priority data gaps associated with extended storage of UNF.
- Identify and prioritize gaps in the current technical bases for disposing of UNF and high-level radioactive waste (HLW) in a range of potential disposal environments.
- Continue model development, validated by experiment, for the evaluation of multiple generic disposal system concepts.
- Support system-level analyses of storage and disposal options in the context of overall fuel cycle performance.

Long-Term Objectives

- Based on both experiment and simulation, develop the technical bases needed to support extended interim storage of all existing and anticipated UNF.
- Demonstrate extended storage strategies through a test and validation complex.
- Develop a robust modeling and experimental basis for the long-term performance of multiple disposal system options.

Challenges

- To provide a sound technical basis for implementation of a new national policy for managing the back end of the nuclear fuel cycle, including identification and evaluation of safe and secure options for storage, transportation, and permanent disposal of radioactive wastes resulting from existing and future fuel cycles.

FY 2011 Funding

Used Fuel Disposition Campaign	
Campaign Management	$1,412K
External and International	$872K
Storage and Transportation	$7,039K
Disposal	$14,562K
Total	**$23,885K**

Major Research and Development Activities

External and International activities have included support for the BRC, participation in international working groups addressing storage and transportation of UNF and HLW, support for bilateral interactions between the United States and the Republic of Korea and the United States and Japan, and planning for U.S. involvement in FY 2012 in disposal research and development (R&D) in European underground research laboratories (URLs).

Storage and Transportation R&D examines three topics: storage, transportation, and security. Storage R&D focuses on closing technical gaps related to UNF storage. Currently available data may be insufficient to support licensing dry cask storage of high-burnup fuels (greater than 45 GWd/MTU) or extended storage of all types of used fuel. For example, uncertainties remain regarding cladding performance following possible hydride reorientation and creep deformation, and also regarding long-term canister integrity. Transportation R&D focuses on ensuring transportability of UNF following extended storage, addressing data gaps regarding fuel integrity, retrievability, and subcriticality. Security R&D focuses on questions related to material attractiveness and self-protection due to surface dose rate, which decreases as UNF ages.

Disposal R&D focuses on identifying multiple viable geologic disposal options, addressing technical challenges for generic disposal concepts in various host media (mined repositories in salt, clay/shale, and granitic rocks, as well as deep borehole disposal in crystalline rock). R&D will transition to site-specific challenges as national policy advances (the Nuclear Waste Policy Act currently precludes site-specific work at locations other than Yucca Mountain). R&D goals at this stage are to reduce generic sources of uncertainty that may impact the viability of disposal concepts, to increase confidence in the robustness of generic disposal concepts, and to develop the science and engineering tools needed to select, characterize, and ultimately license a repository.

Key FY 2011 Outcomes

Key accomplishments in FY 2011 include completion of the following reports:

- *Basis for Identification of Disposal Options for Research and Development for Spent Nuclear Fuel and High-Level Waste,* March 31, 2011, FCRD-USED-2011-000071
- *Disposal Research and Development Roadmap,* March 31, 2011, FCRD-USED-2011-000065 Rev 0
- *U.S. Radioactive Waste Inventory and Characteristics Related to Potential Future Nuclear Energy Systems,* June 2011, FCRD-USED-2011-000068, Rev 2 (prepared for the BRC)
- *Gap Analysis to Support Extended Storage of Used Nuclear Fuel,* June 30, 2011, FCRD-USED-2011-000136
- *Generic Repository Design Concepts and Thermal Analysis (FY11),* August 2, 2011, FCRD-USED-2011-000143, Rev 0
- *Infrastructure Capabilities Needed to Support R&D for Extended Storage,* September 23, 2011 (draft, pending completion of DOE review)

1.2 Abstract: Review of the Used Fuel Disposition Campaign's Storage and Transportation Research and Development Activities

Ken Sorenson, Sandia National Laboratories

Introduction and Objectives

The near-term mission of the storage and transportation R&D being conducted under the Used Fuel Disposition (UFD) Campaign is to develop the technical basis to demonstrate safety and security of UNF storage beyond current regulatory time limits, as well as transportation after storage. Current efforts are focused on legacy commercial used fuel already in storage, as well as all fuel that will be discharged over the life of the U.S. fleet of light water reactors.

Research and Development Overview

NE is supporting R&D to develop the technical basis to demonstrate safety and security of UNF stored in dry cask systems for timeframes well beyond 80 years. FY 2011 efforts have focused on a technical gap analysis, which was done in a deliberate fashion that included development of functional requirements and criteria sets, as well as a thorough review of existing literature and regulatory positions. Throughout development, the identified gaps were shared in a collaborative way with industry (e.g., the Electric Power Research Institute [EPRI], utilities, and cask and fuel vendors) and international organizations in order to obtain the broadest perspectives from the science, engineering, operational, and regulatory communities. This effort culminated in a report that identified and prioritized the technical gaps.

Concomitant with the gap analysis was a related effort that evaluated alternatives for conducting R&D at various facilities within the DOE national laboratory complex. Alternatives for conducting the needed R&D were identified and evaluated using a systematic approach, resulting in a narrowing down of options for more detailed study.

Related work in transportation and security round out the FY 2011 R&D efforts. In transportation, options are being pursued that may result in the need for less materials data. These options include criticality margins (i.e., moderator exclusion and criticality safety analyses) and simulation of the transport environment through over-the-road testing. The security work has focused on issues resulting from UNF storage duration that is longer than initially planned. The primary issue concerns the "self-protection threshold" of the fuel when it is initially placed in storage.

Accomplishments

Key accomplishments in FY 2011 include completion of the following reports:

- *Process and Evaluation Criteria: Testing and Evaluation Alternatives to Demonstrate Very Long Dry Storage of Used Nuclear Fuel*, December 2010, FCRD-USED-2011-000029
- *Functions and Requirements – Used Fuel Storage and Transportation*, December 2010, FCRD-USED-2011-000030
- *Gap Analysis to Support Extended Storage of Used Nuclear Fuel*, June 30, 2011, FCRD-USED-2011-000136 (Level 1 report currently in draft to address industry comments)
- *Infrastructure Capabilities Needed to Support R&D for Extended Storage*, September 23, 2011, FCRD-USED-2011-xxxxxx (Level 2 report currently in draft and in industry review)

1.3 Abstract: Generic Repository Concepts and Thermal Analysis for Advanced Fuel Cycles

Massimiliano Fratoni, Lawrence Livermore National Laboratory

Introduction and Objectives

Geologic disposal of spent nuclear fuel[1] (SNF) and HLW can be implemented using open or closed emplacement modes. For closed emplacement modes, waste packages are in direct contact with encapsulating engineered or natural materials, and it may be desirable to keep temperatures below specified values to limit changes to these materials. This study performed thermal analyses for combinations of projected waste inventories, repository designs, and geologic media, and identified relationships between waste package capacity and the duration of surface decay storage needed to meet those temperature limits.

Research and Development Overview

The Used Fuel Disposition Campaign developed a set of reference geologic disposal concepts that provide context for ongoing research and development (R&D) activities in the areas of SNF long-term storage, transportation, and disposal. A multi-year R&D effort aims to evaluate the interplay between the three major components of these geological disposal concepts: waste inventory, geological settings, and concepts of operation.

Accomplishments

Three waste inventory cases were considered: (1) direct disposal of high-burnup SNF from light water reactors (LWRs), (2) HLW from reprocessing spent LWR fuel and direct disposal of used plutonium–mixed oxide (MOX) fuel, and (3) wastes from reprocessing spent LWR fuel and continuous recycling of metal fuel from fast reactors operating as transuranic burners. Clay/ shale, salt, crystalline rock, and deep borehole settings were selected as disposal concepts. Repository layout and engineered barrier descriptions were selected based on international designs. Based on engineered material and rock temperature constraints, target maximum waste

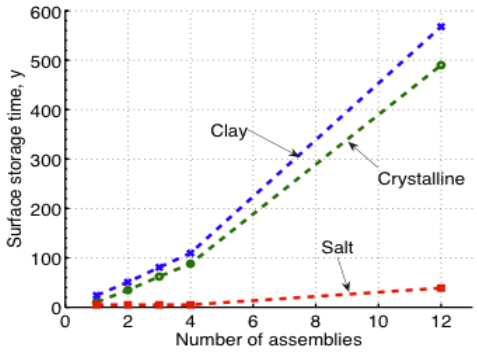

Figure 1. Minimum decay storage duration to limit peak waste package surface temperature to 100°C (for clay buffer or clay/shale media) or 200°C (for salt) as a function of number of UOX assemblies per waste package for crystalline, clay/shale, and salt media.

[1] Spent nuclear fuel is distinguished from UNF in that the latter is presumed to retain value pending subsequent reprocessing to separate out useful isotopes.

package surface temperatures were identified, enabling a sensitivity study to inform the tradeoff between the quantity of waste per disposal package and decay storage duration. In clay and crystalline rock, for surface storage duration of 100 years or less, waste package sizes for direct disposal of SNF are effectively limited to 4-PWR configurations or equivalent size and output, whereas 12-PWR configurations in salt would require less than 50 years of surface storage (Figure 1). Follow-on work will include adding additional reference concepts, verification and uncertainty analysis of thermal models, developing descriptions of surface facilities and other system details, and cost estimation.

1.4 Abstract: Integrated Research Project – Fuel Aging in Storage and Transportation: Accelerated Characterization and Performance Assessment for Nuclear Fuel Storage System

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Introduction and Objectives

This integrated research project involves six university partners with multiple investigators per university as well as supporting advisors from Savannah River National Laboratory and Pacific Northwest National Laboratory. The program will develop methods to characterize and monitor key limiting phenomena related to the performance of UNF storage systems. The overall objective is to create predictive tools in the form of observation methods, phenomenological models, and databases that will enable the design, installation, and licensing of dry UNF storage systems that will be capable of containing UNF for extended periods.

Research and Development Overview

The Fuel Aging in Storage and Transportation (FAST) program is focused on four distinct, yet integrated, technical mission areas (TMAs):

- Low-Temperature Creep
- Hydrogen Behavior and Delayed Hydride Cracking
- UNF Canister Corrosion
- Novel System Monitoring

The TMAs are designed to address challenges relevant to almost every independent spent fuel storage installation (ISFSI) system currently deployed or under development, with a special emphasis on high-burnup fuel.

The first two technical missions deal with potential UNF cladding failure phenomena. Low-temperature creep and delayed hydride cracking may lead to premature fission product releases within the cask that would complicate future transportation, storage, and/or processing operations. The third mission is focused on protecting the integrity of the dry cask to preserve the containment of the fuel even if the cladding fails over time. The fourth mission will develop long-term monitoring methods to equip future decision makers with real-time understanding of the internal state of the UNF being stored.

The identified critical failure mechanisms (low-temperature creep, delayed hydride cracking and canister corrosion) occur over timescales that range from nanoseconds, associated with elementary diffusion events, to the extended periods over which UNF storage systems are expected to function without losing structural integrity. While a rich database exists from experiments that have been conducted over the last several decades, it is not sufficient or prudent to extrapolate the results to several hundreds of years. Thus accelerated tests—in which environmental conditions such as stress, fluence, temperature, and/or chemical concentrations are enhanced—become a viable option to assess the critical failure mechanisms over extended periods.

Fuel Cycle Options Campaign

2.1 Fuel Cycle Options Campaign Overview

Mission

Develop and implement management processes including communications, perform integrated fuel cycle analyses and technical assessments, and provide information that can be used to inform Office of Fuel Cycle Technologies activities objectively and transparently, including R&D, strategy and policy formulation, program decisions and budgets.

Objectives

- Develop and manage the processes that can be used for guiding selection of one or more sustainable alternative fuel cycle options and prioritizing associated R&D.
- Perform analyses and studies of fuel cycle options that are objective and reproducible.
- Provide knowledge management systems that provide documentation to support transparent decision making for R&D investments, ensuring past, present and future program results are traceable and available.
- Develop approaches and materials for communication of FCT program objectives, values, and accomplishments to stakeholders.

Challenges

- Merging Systems Analysis and System Engineering campaigns, developing the new campaign and managing the changing directions and priorities
- Developing the process for evaluating fuel cycle options that will be objective, reproducible, and responsive to evolving national priorities
- Conducting an evaluation and assessment of fuel cycle options that is convincingly comprehensive and conclusive concerning fuel cycle options and their capabilities
- Identification of restrictions on possible options from policy and legislative actions
- Lack of analysis, data, and experience with many technology options, increasing uncertainty of the fuel cycle evaluations
- Availability of sufficient detail from previous analyses and consistency of assumptions

FY 2011 Funding

Systems Analysis Campaign	
Campaign Management	$852K
Systems Analysis, Optimization, Trade-off Studies	$4,983K
Tools and Data Development	$833K
Fuel Cycle Simulator Development	$702K
Total	**$7,370K**
Systems Engineering Campaign	
Systems Engineering (Management and Activities)	$3,565K
Subject Matter Experts Core Group Support	$702K
Options Development	$817K
ANL Support	$250K
Total	**$5,334K**

Major Research and Development Activities

Nuclear Fuel Cycle Evaluation and Screening aims to develop the process and technical basis for conducting the objective and reproducible evaluation of nuclear fuel cycles with respect to criteria reflecting concerns or issues. The evaluations will be used to identify promising fuel cycle options that address these concerns, providing information that can be used to guide R&D directions by defining function and performance goals for each part of the fuel cycle. This entails activities to deliver the following major objectives:

- **Fuel Cycle Option List** – credibly comprehensive
- **Evaluation Criteria and Performance Metrics** – comprehensive and relevant
- **Evaluation and Screening Approach** – objective and flexible
- **The Complete Fuel Cycle** – from mining to disposal

Integrated Fuel Cycle Analysis establishes performance metrics, fuel cycle evaluation must be supported by accurate and consistent technical analysis of fuel cycle performance, including fuel cycles that have not been extensively analyzed and covering a broad range of possibilities, including use of thorium, alternate reactor concepts, and a range of geologic disposal options.

Analysis of Specific Systems Issues includes developing an understanding of the important factors affecting the transition from one fuel cycle to another, including the dynamic interactions between each part of the fuel cycle and the existing and proposed infrastructure; examining the utility of limited recycle approaches and possible new uses for light water reactors; and new approaches for analyzing the economic impact of a nuclear fuel cycle.

Key FY 2011 Outcomes

Key accomplishments in FY 2011 include completion of the following reports:

- *Peer-Reviewed Fuel Cycle Options Screening Report*
- *Dynamic Analysis Scoping Study Report*
- *Report on Impact of Innovative Fuels, Transmuters, and Separations Processes*
- *Report on Assumptions and Technical Basis for Fuel Cycle Systems Options*
- *Report on Modified Open Cycle as a Transition Step*
- *Report on Extended Uses for Light Water Reactors*
- *Energy Return on Investment Report*
- *Complete Report on Contributions for GAINS*
- *Summary Report on Fuel Cycle Research and Development Program*

2.2 Abstract: Economics Analysis Activities

Francesco Ganda, Idaho National Laboratory

Introduction and Objectives

This presentation documents (1) the creation of a new methodological framework for the equilibrium economic analysis of complex fuel cycles and (2) the development and testing of a new code specifically developed for this purpose (NE-COST). This effort has been necessary to enable economic analysis of systems with multiple reactor types. The key concept is to divide each fuel cycle into subsets of facilities (here generally called "islands") which will contain one single reactor or blanket, and a number of fuel cycle facilities. Although the facilities of a given fuel cycle can be assigned arbitrarily to each island without affecting the estimated overall cost of electricity, a careful assignment can (1) simplify interpretation of the cost of each individual island, (2) facilitate communication of results, (3) allow re-utilization of the input for a different system featuring a similar sub-set of facilities, and (4) obtain a substantial logical and computational simplification, supporting robustness and reproducibility.

Research and Development Overview

The island approach and the NE-COST model greatly simplify the cost analyses of complex multi-reactor fuel cycles, an activity of importance in support of the fuel cycle evaluation process. Generally, each reactor type will have a different reloading schedule and operational life, and the timing of the expenditures has economic relevance in an environment in which the cost of capital is non-zero. The system's levelized cost of electricity depends on the time and amplitude of cash flow within each subsystem, and substantial simplification can be achieved by modeling each of these cash streams separately, rather than performing a convolution of the cash flows of each facility in the fuel cycle. The cost of electricity of the overall system will then be approximated by the weighted average of each island's cost of electricity, where the weights are the fractional energy generated by each island. The conditions under which this approximation estimates the overall cost of electricity exactly have been identified and found generally non-stringent for practical systems.

Accomplishments

The main achievements in FY 2011 are as follows:

- Invention and development of the "island approach" for a systematic and simpler economic modeling of multi-reactor fuel cycle systems
- Identification of the minimal logical structure that allows the economic modeling of each island with no code modifications

- Implementation of programming with the desired functionalities in NE-COST
- Full implementation of Monte-Carlo uncertainty analysis capabilities
- Creation of a suite of tools to (1) handle the stochastic combination of uncertainty distributions in the input costs and (2) plot the results, with the intent of facilitating interpretation and communication of the calculated costs and uncertainties
- Successful completion of a benchmark between G4-ECONS and NE-COST for a three-reactor fuel cycle

2.3 Abstract: Energy Return on Investment Analysis

Bill Halsey, Lawrence Livermore National Laboratory

Introduction and Objectives

The objective of this study is to develop a methodology and requisite data to assess the potential Energy Return on Investment (EROI) for nuclear fuel cycle alternatives, and to apply that methodology to calculate EROI for a limited set of initial fuel recycle scenarios. The FCT Systems Analysis Campaign objectives include development of relevant fuel cycle metrics and development of tools and associated data for analysis of fuel cycle systems. This study, conducted by Lawrence Livermore National Laboratory (LLNL) and AREVA Federal Services, represents an initial evaluation of EROI as a metric for fuel cycle facilities, processes and technologies, and includes development of an analysis tool that can be used for future studies. The focus of the LLNL effort is to develop an analysis methodology that is consistent with the broader energy modeling community, and the focus of the AREVA effort is to bring industrial experience and operational data into the study.

Research and Development Overview

A literature survey found no published EROI results for nuclear fuel recycle. A set of recycle scenarios were developed and energy content data established to enable such an evaluation. For nuclear recycle facilities, data from industrial operational experience was applied. An Excel spreadsheet tool was developed, and initial EROI results were obtained.

Accomplishments

This study has successfully developed a methodology, an industrial database, and an analysis tool for EROI analysis of nuclear fuel cycle alternatives. Initial results for several fuel recycle scenarios are shown below. Observations include the following:

- Establishing the “energy boundaries” is a major part of scenario definition.
- EROI for fuel recycle was found to be large compared to many energy alternatives.
- The energy for decommissioning of nuclear facilities may be a significant factor.

Scenario	Total MOX (LWR and SFR) in tHM/y	Total RepU (LWR and SFR) in tHM/y	Number of SFR	EROI (Final Energy)
One-Pass Recycle UNF → Recycle → LWR → Disposal	100	58.5	N/A	71.6
Two-Pass Recycle UNF → Recycle → LWR → Recycle … LWR → Disposal	140.1	52	N/A	81
2nd Pass Recycle to multi-pass SFR burners UNF → Recycle → LWR → Recycle … SFR Burner (Pu) and LWR (RepU) → Disposal	138.9	48.4	5	96
2nd Pass Recycle to single-pass SFR breeders UNF → Recycle → LWR → Recycle … SFR Breeder → Recycle → LWR → Disposal	131.2	71.3	5.6	98.3

2.4 Abstract: Recycle Fast Reactors

Florent Heidet, Argonne National Laboratory

Introduction and Objectives

The current fleet of nuclear reactors is only capable of extracting less than one percent of the thermal energy contained in the mined uranium because of the large amount of depleted uranium generated by the enrichment process and low burnup achieved. The approach commonly proposed to increase fuel utilization is to reprocess the fuel. Because of proliferation concerns and public acceptance, however, existing separations technologies are not generally acceptable.

Conceptually, recycle fast reactors can increase significantly the uranium utilization without requiring actinide separation. Except for the initial fuel loading, only depleted uranium is fed to the recycle fast reactors, and the fuel is reconditioned with a partial separation process. This type of reactor could provide the U.S. electricity capacity for several hundred years using the depleted uranium already accumulated.

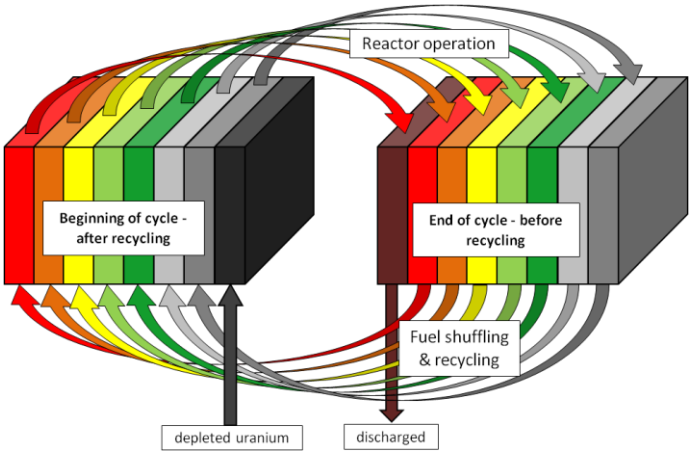


Figure 1. Schematic of the recycle fast reactor mode of operation.

Research and Development Overview

To improve understanding and determine the requirements enabling burnup propagation—the principle on which the recycle fast reactors are based—entails studying the relevant physics. Based on this knowledge, two preliminary recycle fast reactor designs, with power ratings of 3000 MW_{th} and 1600 MW_{th}, respectively, are proposed and their neutronics performance assessed.

Accomplishments

It was found that the maximum uranium utilization achievable with the 3000 MW_{th} and 1600 MW_{th} recycle fast reactors are 55% and 43%, respectively. In addition, when compared to currently operating reactors, the recycle fast reactors enable significantly decreased uranium enrichment requirements; lower waste volume per unit of energy produced; and improved waste characteristics per unit of energy produced. With proper fuel management, the 3000 MW_{th} recycle fast reactor achieves a capacity growth rate of up to 3.9% using only depleted uranium.

During this study, a number of areas requiring additional research have been identified. The most important are the fuel reconditioning, the refabrication of very high-burnup fuel, and the development of advanced cladding materials.

Material Protection, Accounting, and Control Technologies Campaign

3.1 Material Protection, Accounting, and Control Technologies Campaign Overview

Mission

Develop innovative technologies and analysis tools to enable next-generation nuclear materials management for future U.S. nuclear fuel cycles.

Objectives

- Develop and demonstrate advanced material control and accounting technologies that would, if implemented, fill important gaps in existing MPACT capabilities.
- Develop, demonstrate, and apply MPACT analysis tools to assess effectiveness and efficiency of MPACT systems and guide research and development (R&D).
- Develop guidelines for safeguards and security by design, and publish international guidance documents.

Challenges

Key drivers

- Future advanced fuel cycle facilities may be larger, more complex, and more widespread.
- Threats, both insider and outsider, may continue to become increasingly sophisticated and capable.
- Achieving stringent goals for detection timeliness and sensitivity in advanced fuel cycle facilities will be difficult and expensive.
- Satisfying stringent physical protection requirements in advanced fuel cycle facilities will be expensive.
- Addressing stakeholder concerns will require positive assurance that risks of nuclear proliferation and terrorism are minimized.

Technical needs

- Improve the precision and accuracy of key nuclear material measurements.
- Improve the timeliness and cost-effectiveness of measurements and analysis.
- Expand the scope of detection to include more indicators, taking advantage of existing data where possible and new sources of data where appropriate.

- Expand and strengthen detection and assessment algorithms to exploit larger data sets and provide results in near-real time.
- Model and simulate MPACT performance against a wide spectrum of assumed threats, and rigorously demonstrate MPACT effectiveness and efficiency in future U.S. nuclear energy systems.
- Integrate safeguards and security into the design of future nuclear fuel cycle facilities from the earliest stages of the design cycle.

FY 2011 Funding

MPACT Campaign	
Campaign Management and Ad Hoc Technical Support	$637K
Material Control and Accounting Technologies	$2,647K
MPACT Analysis Tools	$1,016K
Safeguards and Security by Design	$500K
Total	**$4,800K**

Major Research and Development Activities

Material Control and Accounting Technologies are being developed with new capabilities that will significantly advance the state of the art in accounting and control. A focused, innovative, science-based R&D program is being conducted to improve precision, accuracy, speed, sampling and monitoring methods, and scope of nuclear material accounting and control. Major technical focus areas include active interrogation methods based on neutron and photon drivers, advanced passive detection methods such as ultra-high-resolution x-ray and gamma-ray spectrometry, advanced sensors, and a range of neutron-based techniques.

MPACT Analysis Techniques are being developed with new capabilities to address the huge quantities of data that can now be extracted from operating processes and utilized for accounting, control, and detection. A fully integrated system in a complex, large processing facility remains in the category of a long-term challenge. MPACT analysis tools will enable more effective monitoring of facility operations and, as a result, better detection timeliness and sensitivity.

Safeguards and Security by Design is a methodology and discipline for integrating nonproliferation and security considerations into the design of nuclear facilities from the earliest stages. The goal is to identify innovative process and facility design features that maximize the effectiveness and efficiency of safeguards and security, and to work with the design team

throughout the design process to introduce such features as appropriate, to minimize costly retrofits.

Key FY 2011 Outcomes

- Demonstration of the capability of a 256-pixel microcalorimeter array to determine plutonium mass and isotopics with better precision and accuracy than traditional high-purity germanium detectors for gamma spectrometry.
- Report on electrochemical actinide sensors, assessing both the physical stability of a new ceramic sensor material under the harsh conditions of electrochemical processes and alternative configurations for a sensor assembly.
- Report on lead slowing-down spectrometer, documenting status of time-spectral analysis methods development, threshold neutron sensor development, and experimental benchmarking of the lead slowing-down spectrometry technique.
- Report on process model validation, documenting experimental validation of the AMUSE process model through targeted validation experiments focused on UREX and COEX processes.
- Report on Safeguards and Security by Design, documenting the initial application of Safeguards and Security by Design methodology to the analysis of new small reactor designs.

3.2 Abstract: Multi-Isotope Process Monitor Abstract

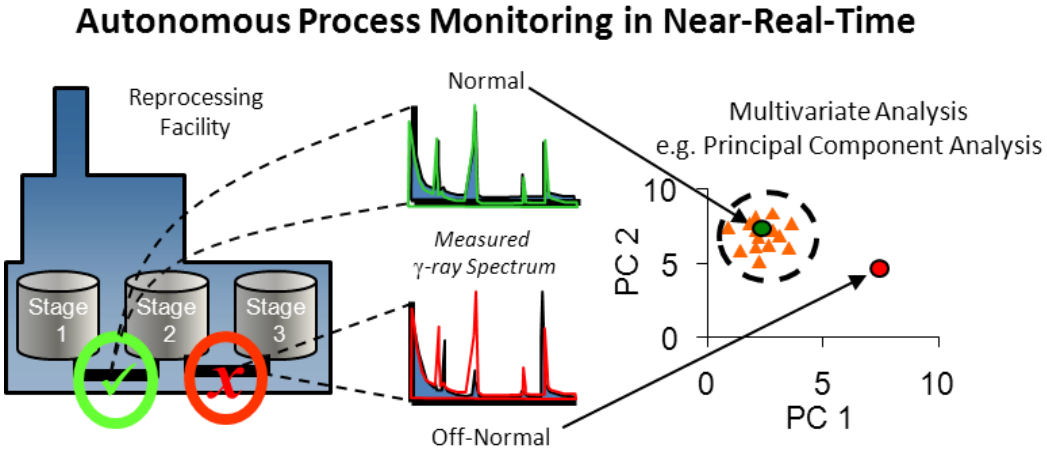
Christopher Orton, Pacific Northwest National Laboratory

Introduction and Objectives

Researchers at the Pacific Northwest National Laboratory, in conjunction with several U.S. universities, are working to develop the Multi-Isotope Process (MIP) Monitor, a system for monitoring used nuclear fuel recycling facilities online, non-destructively, and in near-real time. The goal of MIP project is to improve material accounting and control, process control, safety, efficiency, and overall plant performance to aid both the plant operator and the regulator. Specifically, this technology is primarily intended to support Objective 4 from the *Nuclear Energy Research and Development Roadmap* to "minimize the risks of nuclear proliferation and terrorism." This research also contributes to Objective 3 by potentially improving process control, which can contribute to "sustainable nuclear fuel cycles."

R&D Overview

The MIP Monitor method takes advantage of the patterns of radioactive isotopes that distribute during fuel recycling. By combining information-rich gamma-ray spectroscopy with pattern recognition software, it can identify off-normal conditions in process streams and even quantify some plant and fuel conditions. Because it targets gamma-emitting indicator isotopes, which are inherent to used fuel, the MIP Monitor approach is compatible with several separation processes, including both electrochemical and aqueous methods. The method design supports the use of small, portable, high-resolution gamma detectors that can be deployed throughout the facility at multiple measurement locations. The research has included applying multivariate statistical techniques to simulated and empirical gamma spectra representative of spent fuel separations under various process conditions, demonstrating the basic feasibility and validity of the MIP Monitor methods. Future studies include developing, refining, and optimizing the multivariate approach through simulated and, when possible, experimental data sets.



Accomplishments

During FY 2011, the MIP Monitor project completed several tasks to advance the technology development. These tasks included maintaining and collecting gamma-ray spectra from spent fuel samples using sodium iodide and cadmium-zinc-telluride (CZT) detectors and constructing a flow loop capable of providing dynamic data on spent fuel solutions. Significant studies into advanced multivariate techniques and approaches were conducted. Steps were also taken to secure collaborations with domestic and international partners and acquire large amounts of empirical process data that will aid in the refinement of the data analysis techniques that are central to the MIP Monitor.

3.3 Abstract: Solid State Neutron Detector Abstract

J. Clinton, Y. Danon, J. Huang, R. Dahal, J. Lu and I. Bhat
Rensselaer Polytechnic Institute

Introduction and Objectives

Active and passive neutron interrogation are valuable tools for nuclear safeguards and process monitoring of the nuclear fuel cycle, utilized to verify initial enrichment of low-enriched uranium (LEU) fuel, assay the uranium and plutonium content of spent fuel, and monitor the fissile material content in process and product streams within a reprocessing plant. These tasks are often accomplished with neutron coincidence counters, which typically use moderated thermal neutron detectors. In order to achieve high detection efficiency, ^{3}He-filled thermal neutron detectors are normally utilized. However, due to a shortage in worldwide ^{3}He supply, these detectors have become prohibitively expensive. A better type of detector would be a low-cost solid-state detector that can be mass produced like any other computer chip. The scalability of these devices would allow for compact detection system configurations not feasible with the larger, gas-filled detectors, such as position-sensitive arrays embedded in a moderator.

The goal of this research is to develop a high-efficiency, low-noise, gamma-insensitive, self-powered solid-state neutron detector system with sufficient detector size (up to eight inches in diameter), and integrated with interface electronics (e.g., preamplifier) as a coincidence counter for MPACT applications. In addition to the primary goal, this project will educate and train graduate students in fabrication and testing of solid-state radiation detection systems.

Research and Development Overview

The detectors described above first undergo analytical and Monte Carlo modeling, including several 3-D etched structure configurations, to optimize both the electronic and nuclear device characteristics. Fabrication is then accomplished using standard silicon processing techniques, such as deep reactive ion etching (DRIE). The innovations of this research in material processing are in the fabrication of a continuous p-n junction in the silicon, necessary for full charge carrier collection without increased device capacitance or leakage current, and adaptation of highly developed 3-D integration technology for practical, high-efficiency neutron detector fabrication. A detector test bed, consisting of a calibrated thermal neutron source and dedicated electronics, is then used to accurately characterize device performance.

Accomplishments

Key accomplishments in FY 2011 include the fabrication of a prototype thermal neutron detector, etched in a honeycomb array of hexagonal holes using the Bosch process and filled

with natural boron (19% ^{10}B) via low-pressure chemical vapor deposition (LPCVD), as shown in Figure 1.

(a) **(b)**

Figure 1: Hexagonal microstructures etched into a silicon substrate
(a) before, and (b) after LPCVD boron application.

Testing demonstrated a measured intrinsic efficiency of 4.5%; scaling this result to that of a device filled with 95% ^{10}B gives 21.4%, which is one of the highest efficiencies reported in the literature for a single detector of this type. A second generation of devices has been produced with improved electronic characteristics, resulting in measured leakage currents of -1.7×10^{-8} A/cm^2, which is the best reported value for these detectors.

Future work will include epitaxial silicon deposition to reduce detector series resistance, incorporation of enriched boron, fabrication of multiple wafer devices, and the design and fabrication of on-chip preamplifiers.

Separations, Waste Forms, and Fuel Resources Campaign

4.1 Separations, Waste Forms, and Fuel Resources Overview

Mission

Develop the next generation of fuel cycle separation and waste management technologies that enable a sustainable fuel cycle with reduced processing, waste generation, and potential for material diversion.

Objectives

- Develop a fundamental understanding of methods for the separation of transuranic elements from UNF.
- Develop and demonstrate enabling technologies to separate transuranic elements from UNF.
- Develop and demonstrate enabling technologies to separate and immobilize gaseous fission products from UNF.
- Develop a fundamental understanding of the factors affecting performance of advanced waste forms.
- Demonstrate predictable performance of advanced waste forms with greatly improved durability and waste loadings.
- Investigate alternative separation technologies and waste forms that could lead to transformational breakthroughs.

Challenges

- Separation of americium or americium and curium from lanthanides
- Capture and immobilization of off-gas constituents of used fuel, including iodine, krypton, tritium and potentially carbon
- Development of separation technologies and waste forms, which is inter-related to the types of fuels being processed, the types of fuels being fabricated, and the reactors used to burn recycled fuels
- Proliferation risk assessment of separation technologies, which is very subjective and must be done in the context of the entire fuel cycle (mining to disposal)

FY 2011 Funding

Separations and Waste Form Campaign	
Campaign Management	$929K
Advanced Electrochemical Technology	$3,100K
Advanced Aqueous Technology	$2,942K
Sigma Team for Minor Actinides	$2,775K
Sigma Team for Off-Gas	$2,725K
Fundamental Methods Development	$3,792K
Transformational Separation Technologies	$1,470K
Modified Open Cycle Technologies	$4,984K
Advanced Waste Forms	$2,040K
Waste Forms Characterization	$2,011K
Total	**$26,636K**

Major Research and Development Activities

Minor Actinide Separations Sigma Team is developing simplified approaches to the separation of americium, or americium and curium, to enable future fuel cycles that transmute minor actinides. There is a large international effort—nearly every country involved in fuel cycle research is working on this difficult chemical separation—and the FCR&D program is making significant progress on the development of cost-effective methods of separating the minor actinides americium and curium.

Off-Gas Capture and Immobilization is a critical technology for enabling any new fuel treatment facility to be licensed to meet current regulations. The capture of iodine and krypton could be very costly additions to a new facility, and immobilization of the long-lived iodine will be important to reduce the source term in a geologic repository.

Advanced Waste Forms are necessary for the immobilization of waste streams from the advanced separations processes, including high-halide electrochemical salt wastes, gaseous fission products waste, and separated technetium. The waste forms for streams containing ^{129}I, ^{99}Tc, and transuranics require performance for very long time periods in order to be a sufficient barrier to release. This requires new materials and a better understanding of the alteration and release mechanisms. Waste form and process development is also required to significantly reduce the cost of waste treatment, storage, transportation and disposal.

Alternative Separation Processes are looking at alternative separation processes, beyond aqueous and pyrochemical, that could offer simplification and cost reduction and possibly reduce proliferation risk.

Key FY 2011 Outcomes

- Completed peer review of white papers on modified open cycle separation and waste form concepts and initiated research on four research projects.
- Established Joint Feasibility Study for Pyroprocessing Research with Republic of Korea.
- Completed a workshop on modified open cycle volatilization technologies.
- Demonstrated multi-channel on-line monitoring on a solvent extraction centrifugal contractor flowsheet.
- Completed a peer review of the Sigma Team for Minor Actinide Separations results and activities.
- Completed a peer review of the Off-Gas Sigma Team results and activities.
- Validated model of strontium to zirconium decay in a waste form material.
- Obtained ancient glass samples from Roman terrestrial and seabed locations.
- Made significant progress on development of an international glass corrosion mechanism.
- Completed a peer review of glass corrosion results and activities.
- Completed direct nitration tests on UNF.
- Completed the fabrication and testing of electrochemical salt waste forms from EBR-II process wastes.
- Demonstrated at laboratory scale an aerogel-based iodine capture media with over double the capacity of current technology.
- Developed a new material capable of krypton and xenon separation from air at near-room temperature.

4.2 Abstract: Sigma Team for Minor Actinide Separations

Bruce Moyer, Oak Ridge National Laboratory

Introduction and Objectives

The Sigma Team for Minor Actinide Separation (STMAS) was formed at the beginning of FY 2009 to enable more efficient separation methods for americium (Am) and other minor actinides (MAs) to greatly improve the overall benefit of fuel recycle. Two-fold aims of the STMAS are to provide the scientific basis for 1) an efficient separation of americium and curium (Cm) from fission products and 2) an efficient separation of americium from curium. These two aims are being pursued mainly within the paradigm of aqueous reprocessing of used oxide nuclear fuel, the overarching question being whether major simplification and economy can be achieved. It is anticipated that this will entail new chemistry and likely altogether new separation agents.

Research and Development Overview

Strategies for Am separation are focused on either complexation or exploiting the higher oxidation states of Am. Complexation approaches seek selective binding through use of aqueous-phase complexants or specific extractants or their combinations. Manipulation of the americium oxidation state is a potentially powerful approach for a selective Am separation, but the very high potentials required for oxidation of Am(III) to Am(V) and Am(VI) present a formidable challenge. Efforts are focused on methods for oxidation of Am and for separations involving Am(V) and Am(VI) separation once generated.

Accomplishments

- An Am/europium separation factor of ~50 was demonstrated in extraction from first-cycle raffinate simulant after oxidation of Am to Am(VI) with sodium bismuthate (Figure 1). Selective stripping from co-extracted Ce(IV) was successfully demonstrated.
- A single combined process that does the same job as the tandem TRUEX–TALSPEAK processes is advancing, with minimum actinide/lanthanide separation factors exceeding 20 and fission products dealt with.

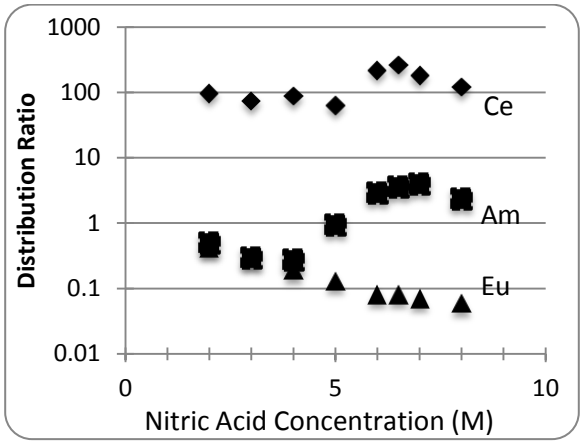


Figure 1. MA extraction from first-cycle raffinate simulant after oxidation with sodium bismuthate.

- A new class of mixed-donor extractants for trivalent actinides has been designed using computational techniques (Figure 2). With both soft- and hard-donor groups, they should be both strong and selective.
- Despite its complexity, the chemistry of the TALSPEAK process is gradually being elucidated, and the understanding has led to a process variation that has much flatter pH dependence.
- A new macrocyclic complexant has been shown to complex Am(III) preferentially over Cm(III), leading to a useful separation factor of 5.9, in principle sufficient for 99% separation in six stages.

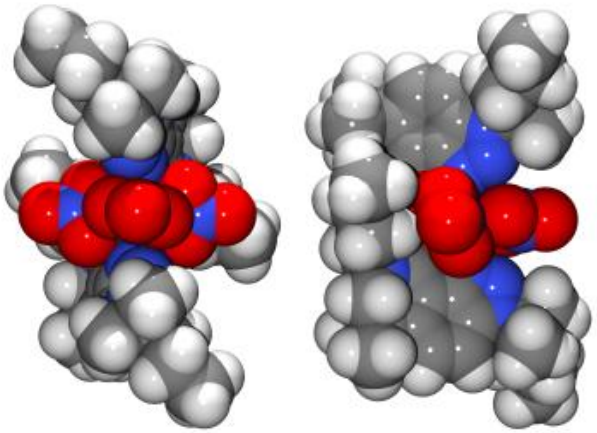


Figure 2. Space-filling models of the structure of $[Am(L)_2(NO_3)_3]$ (L = new mixed-donor ligand).

4.3 Abstract: Combination of Hard and Soft Donor Ligands for Actinide Co-Extraction from UREX Raffinates

Peter Zalupski, Idaho National Laboratory

Introduction and Objectives

The traditional UREX+ concept of actinide (An) recovery from dissolved UNF involves an ensemble of separation steps arranged in tandem according to chemistry selected by the demands of product purity, process handling and proliferation risks. As such, this process manages very difficult separation tasks, while yielding actinide product streams of exceptional quality. Although successfully demonstrated on a laboratory scale, the feasibility of industrial implementation of a separation plant based on such a concept is in question, citing economic reasons. Consequently, the current separations philosophy of the FCR&D program sponsors viable methods of reducing the complexity of separation activities while avoiding the separation of pure plutonium. It is clear that the success of such an enterprise rests on the discovery of efficient group actinide (uranium through curium) extraction from the dissolved fuel streams.

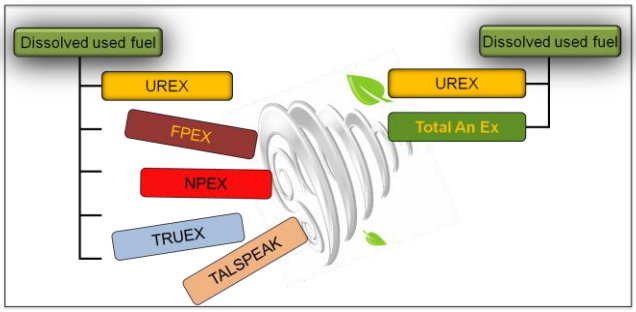


Figure 1. Current multi-step separation process (left) and desired reduced-complexity process using group actinide extraction techniques (right).

This research effort seeks to develop a proof-of-principle demonstration of the feasibility of such co-extraction based on the combination of sulfur-based soft donor reagents with traditional hard-donating ligands. This separation concept relies on finding synergy between the hard donor's affinity for hexavalent and tetravalent actinides and the soft donor's preference to interact with actinides of lower charge density (Np(V), Am(III), Cm(III)). While the extraction of An(VI) and An(IV) is easily accomplished using tributyl phosphate, the lower charge density of An(III) and An(V) impedes efficient electrostatic interaction with a hard donor ligand. Thus the extraction of those tri- and pentavalent ions has to rely on their slightly "softer" nature, where more polarized f-electron orbitals covalently interact with soft-donor ligands. Accordingly, a combination of hard and soft donating ligands may achieve the co-extraction task.

The goals of the project are to evaluate viable ligand combinations, prove or disprove the concept and, if actinide co-extraction is successful, identify other co-extracting soft metal ions.

Research and Development Overview

The identification of a soft donor with sufficient hydrolytic and radiolytic stability that allows separating actinides from lanthanides (Ln) is of the essence to this project. Following this identification will be studies on the characterization of trivalent actinide and lanthanide extraction and the effects of combining hard and soft donor reagents on the extraction. If actinide/lanthanide separation is maintained with the mixed donor solvent, the distribution of all actinides will be investigated. Finally, competitive distribution studies will track group actinide partitioning from surrogate UREX raffinate. This study will identify the composition of the final product, which will be evaluated by the consulting panel of scientists with expertise in process engineering, fuel fabrication and waste forms. The feedback received from the panel will be addressed during the optimization studies.

Accomplishments

Since the inception of the project in June 2011, a methodology towards synthetic preparation of a soft-donating ligand candidate has been completed. The bifunctional sulfur ligand has been synthesized, purified and characterized. The acid degradation stability studies have been completed showing very promising results. Based on ^{31}P nuclear magnetic resonance (NMR) results, the compound shows no signs of hydrolysis after a 60-minute contact with 1 M nitric acid. Elemental analysis is in progress. The upcoming studies will evaluate actinide/lanthanide differentiation properties for the synthesized ligand candidate.

4.4 Abstract: Alpha Radiolysis of Nuclear Solvent Extraction Ligands used for An(III) and Ln(III) Separations

Thomas D. Cullen, Stephen P. Mezyk, California State University, Long Beach
Mikael Nilsson, University of California, Irvine
Bruce J. Mincher, Idaho National Laboratory

Introduction

The development of a quantitative understanding of the fundamental chemistry of separations processes is essential for an optimized approach to allow minimal processing and waste generation. The separation of the minor actinides from dissolved nuclear fuel is one of the more formidable challenges in this regard. The partitioning of americium, and its transmutation in fast reactor fuel, would reduce HLW long-term storage requirements by as much as two orders of magnitude.

Research has been performed worldwide for several aqueous solvent extraction processes for Ln(III) separation from An(III). In the United States, the most developed process for this separation is TALSPEAK based on the competition between HDEHP (bis(2-ethylhexyl) phosphoric acid) in the organic phase and lactate-buffered diethylenetriamine pentaacetic acid (DTPA) in the aqueous phase. In Europe and Japan, more focus has been placed on the use of dithiophosphinic acids, or DMDOHEMA/bis(triazinyl)pyridine-diamide mixtures. However, any large-scale separation process must be robust under high-radiation dose rates and nitric acid hydrolysis conditions. In particular, the radiation effects on solvent extraction formulations may result in decreased ligand concentrations, giving lower metal distribution ratios, as well as reduced separation factors due to the generation of undesired complexing products, and impaired solvent performance due to films, precipitates, and increased viscosity.

Research Overview

In this work, the ligand degradation chemistry is induced by the radiation field in which separations will be quantified. This provides useful information for future process design and possible ways to avoid unwanted degradation products. To date, unfortunately, significant gaps in our knowledge remain. Principal amongst these gaps are the effects of alpha-radiation and the kinetics/degradation products of these solvent (diluent plus extraction ligands) and metal-loaded ligand systems**.**

This project is measuring the effects of gamma and alpha irradiation on these solvent systems using both steady-state irradiations to measure effects on extraction performance changes (distribution and stripping ratios) and to identify decomposition products, and electron pulse radiolysis measurements to determine the kinetic parameters for the important transient species

reactions. The direct comparison of alpha and gamma irradiations allows us to elucidate the mechanisms of ligand decomposition, as the respective yields of radicals produced by the different radiation sources are known. Gamma irradiations can be performed using standard ^{60}Co irradiators; however, the determination of the alpha-radiolysis-induced extraction ligand degradation has been more problematic, with no single reported method being optimal. Therefore, we are conducting studies using three concomitant approaches to identify the best measurement methodology: internal isotope irradiation (^{244}Cm, ^{211}At), ion-beam measurements (alpha particle beams from an accelerator) and nuclear reactor-induced alpha radiolysis. Our initial focus has been on the CMPO ligand in dodecane, where we have now demonstrated that the radiolytic decomposition efficiencies are approximately the same for both gamma and alpha radiolysis, and that the overall decomposition is significantly less when this formulation is irradiated in contact with acidic water. Analogous studies are currently in progress for TBP.

4.5 Abstract: On-Line Monitoring and U/TRU Co-deposition for Pyroprocessing

Brenda Serrano-Rodriguez, Idaho National Laboratory

Introduction and Objectives

These two technologies are being pursued to support development of advanced pyroprocessing methods of used nuclear fuels. Transuranics (e.g. neptunium, plutonium, americium, curium) accumulate in the electrolyte during uranium recovery operations in electrorefining. On-line monitoring provides a means of monitoring this accumulation, and uranium/transuranic (U/TRU) co-deposition provides another means of recovering the transuranics for recycle in fast reactors. The objective of this work is to demonstrate the efficacy of both technologies in an electrochemical used fuel treatment flowsheet.

On-line monitoring can be used to monitor the process and support efficient operations as well as potential use by the International Atomic Energy Agency [IAEA] to correlate changes in electrolyte chemistry to declared pyroprocessing activities. On-line monitoring and U/TRU co-deposition can be used in conjunction to assure that an unsanctioned (high-purity) plutonium product is never recovered from the electrolyte. U/TRU co-deposition has the potential to produce a higher-quality alloy (i.e., less lanthanide carryover) for fuel manufacture than the current liquid metal cathode approach.

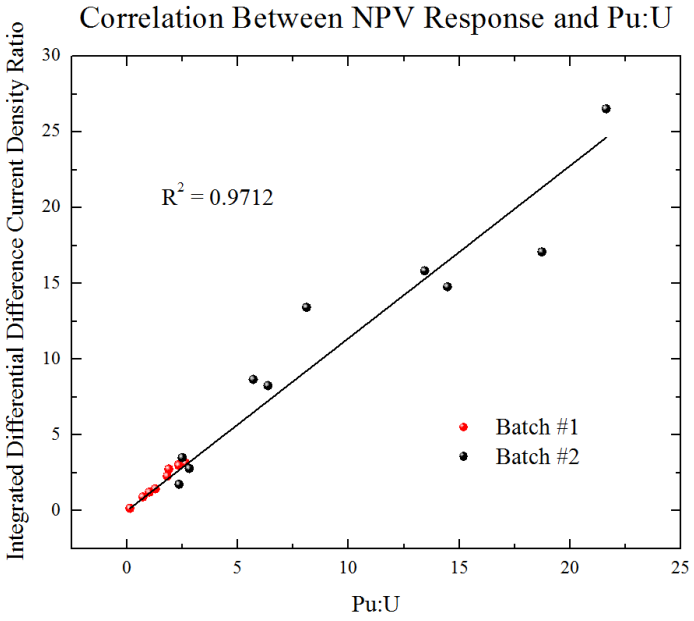


Figure 1. On-line monitoring results.

Research and Development Overview

The fundamental electrochemical aspects of the two technologies are being studied, and electroanalytical techniques are being developed. The molten salt electrolyte systems are becoming more complex with the inclusion of UCl_3, $PuCl_3$, $LaCl_3$, $CeCl_3$ and $GdCl_3$. This

research requires a highly systematic approach to discern and resolve certain electrochemical interactions.

Accomplishments

The basic utilities of the two technologies were demonstrated and verified in FY 2011. Work was performed using UCl_3 and $PuCl_3$ containing electrolytes; on-line monitoring was verified by a series of voltammetry studies, and U/TRU co-deposition was verified by the recovery and analysis of gram-quantities of uranium–plutonium alloy.

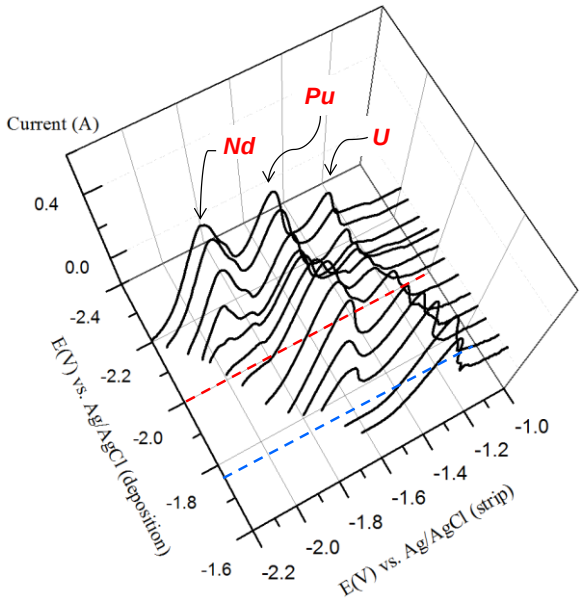


Figure 2. Plot of anodic stripping voltammetry data for uranium—plutonium–neodymium co-deposition tests.

4.6 Abstract: Silver-Functionalized Silica Aerogels for Capture and Immobilization of Gaseous Radioiodine from Reprocessing Off-Gas

J. Matyáš, Pacific Northwest National Laboratory

Introduction and Objectives

To support the future expansion of nuclear energy, an effective method is needed to capture and safely store radiological iodine-129 (^{129}I) released during reprocessing of spent nuclear fuel. Because of its long half-life, 1.57×10^{7} years, any ^{129}I emitted from a reprocessing plant will persist in the environment for tens of millions of years; and although its activity is low, the associated dose would gradually increase the longer these releases are allowed. In addition, being a gas, the iodine is highly mobile in the environment. This project is developing advanced, high-surface-area, silver-functionalized silica aerogels with high I_2-loading and retention capacities that show great promise for removing and immobilizing ^{129}I from an advanced fuel cycle reprocessing plant.

Research and Development Overview

Various materials have been investigated to capture and immobilize iodine. In most cases, however, the materials that are effective for capturing iodine cannot subsequently be sintered/densified to create a stable composite that could be a viable waste form. In contrast, the functionalized silica aerogels can be effective for capturing radioiodine and, once laden with iodine, these materials can be sintered or densified into a durable silica glass. The developed silica aerogel adsorbent and waste will yield an unprecedented control over the management of radioiodine in the stack gases from reprocessing plants, meeting the current and future EPA regulations for concentration of radioiodine in the environment and U.S. Nuclear Regulatory Commission regulations and licensing requirements for long-term storage of radioactive waste.

Accomplishments

Silver-functionalized silica aerogel exhibits decontamination factors in excess of 10,000 in off-gas streams containing 4.2 ppm of iodine and have a maximum iodine sorption capacity of 47.6 mass%, which is more than four times

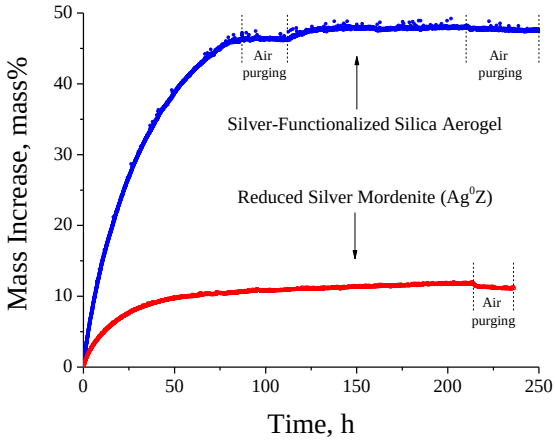


Figure 1. Mass increase over the time for silver-functionalized silica aerogel and silver-reduced mordenite.

higher than that of the silver-reduced mordenite (11.1 mass%), as shown in Figure 1.

Our planned future research activities include further improvement and evaluation of the capture efficiencies of functionalized silica aerogels for I_2, as well as retention of I_2 during the formation of the waste form (densified silica aerogel), investigation of the effect of aging on sorption performance, and evaluation of waste form durability.

Advanced Fuels Campaign

5.1 Advanced Fuels Campaign Overview

Mission

Conduct research, development and demonstration (RD&D) for various fuel forms (including cladding) needed for implementation of the different fuel cycle options defined in the *Sustainable Fuel Cycle Implementation Plan*, ending the campaign mission for any given fuel type when fuel qualification is completed via engineering-scale demonstration of the fabrication processes and irradiation of lead-test assemblies (LTAs) to demonstrate in-pile performance. Develop a state-of-the art R&D infrastructure to support the "goal-oriented science-based" approach.

Objectives

There are two distinct objectives for the campaign with respect to fuel (and cladding) qualification.

- Next generation of LWR fuels with enhanced performance and safety, and reduced waste generation.
- Transmutation fuels with enhanced resource utilization and proliferation resistance.

Challenges

Three- to Five-Year Goals:

- Achieve a state-of-the art R&D infrastructure that supports the use of a "goal-oriented science-based" approach for accelerating the development and qualification of selected fuel concepts.
- Complete conceptual design and document fundamental properties of metallic fast reactor (FR) transmutation fuels for burnup up to 20% burnup. Complete FR cladding R&D towards justification up to 200 dpa.
- Demonstrate improved process towards net shape sintering and microstructural properties tailored for desired performance and fundamental thermal conductivity models (oxide fuels with emphasis on LWR applications).
- Select advanced LWR fuel concepts (including cladding) for further developments towards a lead test rod within the subsequent five to seven years.
- Establish an initial validation database for the multiscale, multiphysics fuel performance code developed by the Nuclear Energy Advanced Modeling and Simulation (NEAMS) program and complete at least two demonstration problems.

FY2011 Funding

Advanced Fuels Campaign		
Activity	**Without Carryover**	**With Carryover**
Campaign Management & Integration	$2,203K	$2,798K
Analytic Support	$2,170K	$2,190K
Innovative Fuel Concepts (3 concepts)	$2,000K	$2,223K
Oxide Fuels	$7,200K	$8,218K
Metallic Fuels	$6,810K	$7,614K
Microencapsulated Fuels	$3,700K	$3,700K*
Core Materials	$3,450K	$4,134K
Characterization/PIE Techniques	$1,600K	$4,951K
Irradiation Testing	$8,050K	$9,527K
Total	**$37,183K**	**$45,355K**

* Microencapsulated fuels had a $5,744K carryover from FY 2010 into FY 2011 under the NGNP program (the Deep Burn program, which was transferred to the Advanced Fuels campaign in the beginning of FY11).

At the end of FY 2011, the Nuclear Data campaign was merged into the Advanced Fuels Campaign. The Nuclear Data activities were funded at the $4,500K level in FY 2011.

Major Research and Development Activities

The following are the major R&D activities as adjusted to the FY 2012 budget and program redirection with respect to next-generation LWR fuels.

International Collaborations include work with France, Japan, Russia, China, South Korea, and the European Union. The emphasis in all collaboration activities is metallic fuel development, joint irradiation testing and data analyses, and characterization/post-irradiation examination (PIE) technique development.

Analytic Support involves reactor impact analyses, fabrication modeling, and sensitivity studies on fuel performance code developed by NEAMS.

Ceramic Fuels tasks comprise advanced sintering process development, thermal performance of oxide fuels, and fission gas gettering concept development (in FY 2012, there will be an emphasis on LWR fuels).

Metallic Fuel tasks include casting technology development, fabrication and characterization of minor actinide- and lanthanide (LN)-bearing fuel samples, fundamental properties measurement and fuel–clad chemical interaction tests (also includes metallic LWR fuel development).

Microencapsulated Fuels tasks support development of LWR fuels with enhanced safety performance.

Core Materials activities support development of FR cladding up to 200 dpa and development of LWR cladding for enhanced performance and accident tolerance.

Characterization/PIE Techniques will be developed, including novel techniques and adaptation of high-resolution equipment to irradiated fuel samples.

Irradiation Testing includes design and execution, PIE and data analyses, and advanced in-pile instrumentation development.

Key FY 2011 Outcomes

The following are the key deliverables/achievements for FY 2011:

- An industry advisory committee was established for the Campaign.
- National and international PIE workshops were conducted.
- AFC-3 metal fuel samples were fabricated and characterization was started. AFC-3A and 3B are ready for insertion into the Advanced Test Reactor (ATR) cycle 150B.
- An assessment was completed for potential agents to immobilize lanthanide fission products in metallic fuels.
- The metallic fuel thermo-physical properties and fission product barriers report was finalized.
- The construction of the multifunctional prototype instrument for laser ultrasound measurements was started.
- A report was issued on microscale laser-based thermal diffusivity measurements on metallic fuels.
- A master sintering curve was established for UO_2 feedstock, and the dynamic oxygen/metal control during sintering was demonstrated.
- Thermal diffusivity evolution during sintering of UO_2-based reference materials was determined.

- An irradiation vehicle was designed for High-Flux Isotope Reactor (HFIR) irradiation of microencapsulated LWR fuels.
- An initial accident testing (two series) of microencapsulated LWR fuels and different cladding materials was completed.
- Charpy, compact tension, tensile and transmission electron microscope testing of ACO-3 samples were performed.
- Coated HT-9 and MA957 samples were fabricated and subjected to iron ion-beam irradiation at 500ºC.
- The PIE report was issued for AFC-1D, 1H, 1G, and 2A. PIE was completed on AFC-2B and AFC-2C.
- Metallography, micro-hardness, and fracture toughness measurements on Fast Flux Test Facility (FFTF) oxide fuels were completed.
- A database for code comparison was generated from X425 experiments in Experimental Breeder Reactor II (EBR II). The database contains the operating conditions for ten metallic fuel pins and the corresponding experimental measurements.

5.2 Abstract: Development of Innovative High Thermal Conductivity UO_2 Ceramic Composite Fuel Pellets with Carbon Nano-Tubes Using Spark Plasma Sintering

Jim Tulenko, University of Florida

Introduction and Objectives

The main disadvantage of UO_2 is its low thermal conductivity. During a reactor's operation, because the thermal conductivity of UO_2 is so low, about 2.8 W/m-K at 1000°C, there is a large temperature gradient in the UO_2 fuel pellet, causing a very high centerline temperature and introducing thermal stresses, which lead to extensive fuel pellet cracking and fission gas release. These cracks will add to the fuel pellet interactions, potentially leading to failure after high burnup. The high fuel operating temperature also increases the rate of fission gas and volatile atom release and fuel pellet swelling caused by fission gases bubbles, which limits the lifetime of UO_2 fuel in reactor.

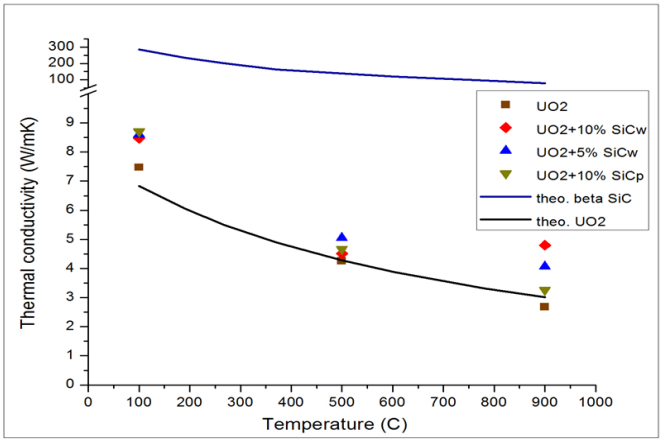


Figure 1. UO_2 pellet thermal conductivity measurements with dopants.

Research and Development Overview

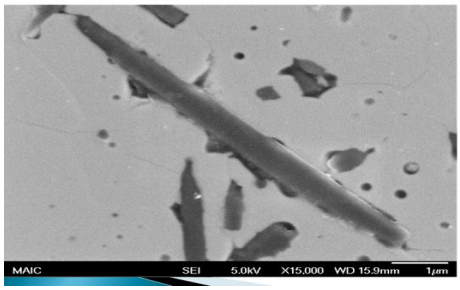


Figure 2. SEM of SPS UO_2 pellet showing SiC whisker.

The University of Florida is looking at silicon carbide (SiC) powder, SiC whiskers, carbon nano-tubes (CNT) and nano-particle diamond additions to UO_2 to determine the positive effect on UO_2 pellet thermal conductivity.

Accomplishments

Initial results show an 8% to 50% improvement with the silicon whiskers and a 10% to 20% improvement with silicon powder. Even greater increases are expected with CNT and diamond particles (see Figure 1).

Figure 3 shows that, with spark plasma sintering (SPS) processing, the project team was able to retain the basic dopants. Figure 2 shows SiC whiskers under a scanning electron microscope (SEM).

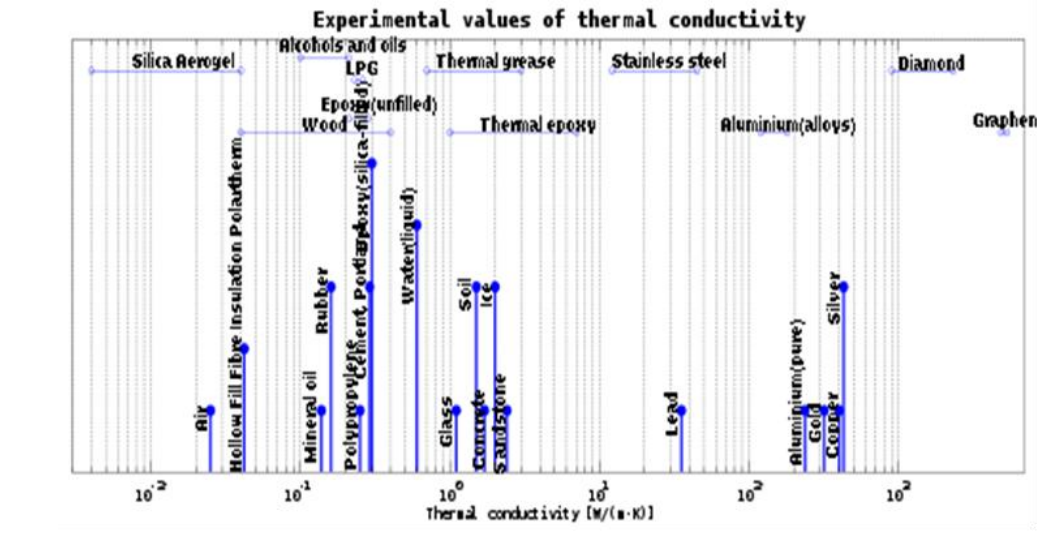


Figure 3. Thermal conductivities of materials of interest for additions to UO_2. Note that natural single-crystal diamond has the highest thermal conductivity of any known material except for CNT.

5.3 Abstract: Metallic Light Water Reactor Fuel: History of Light Water Reactor Fuel Development

Ron Omberg, Pacific Northwest National Laboratory

Introduction and Objectives

The objective of this effort is to develop a more accident-tolerant fuel for LWRs. A metallic fuel was chosen because it has a large thermal conductivity and low specific heat that will provide a greater capacity to remove heat during an accident. The particular alloy chosen was uranium-molybdenum because of the large amount of R&D in the current High Performance Research Reactor Program and because of the extensive earlier research in this area. The desired future state is to demonstrate that this fuel is capable of responding more benignly to major events such as a loss of coolant accident (LOCA). This research supports the overall objectives of the fuel campaign because it pursues a line of research with the potential to change the response of reactor safety systems and the reactor core to an entire suite of major accident events.

Research and Development Overview

The R&D plan has three major components. One is to irradiate a uranium-molybdenum alloy in the ATR to determine its long-term irradiation performance. Specific items to be measured are irradiation swelling and fission gas release as a function of burnup. A second R&D component is to develop a process for diffusing a corrosion-resistant barrier into the exterior of the uranium-molybdenum fuel meat. The third R&D component is to develop an extrusion process that is capable of extruding both the cladding and the fuel meat as a single unit. Each of these areas forms a critical gap that this program is addressing: in-reactor performance, corrosion resistance in the event of a cladding breach, and monolithic extrusion.

Accomplishments

Key accomplishments in FY 2011 include the completion of a preliminary design for an in-reactor experiment for irradiation in the ATR. With this in hand, FY 2012 research will focus on ex-reactor testing—specifically the development of a process to apply an inter-diffusional corrosion barrier and the development of a process to extrude the cladding, inter-diffusional barrier, and metal fuel as a single unit.

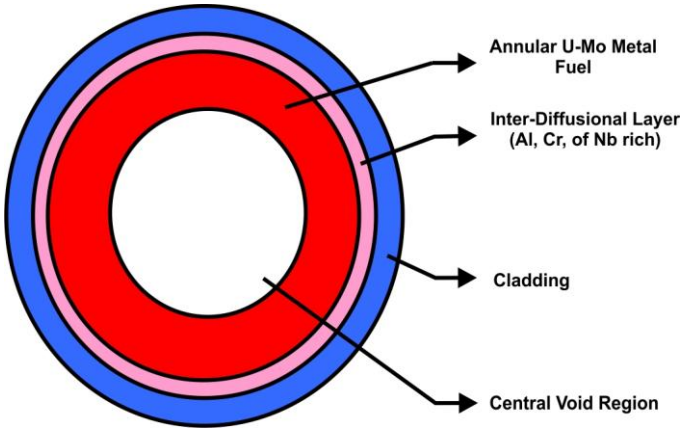


Figure 1. Cross-sectional view of a uranium-molybdenum fuel rod showing key performance features.

5.4 Abstract: Transmutation Fuels Post-Irradiation Examination

Heather Chichester, Idaho National Laboratory

Introduction and Objectives

The Advanced Fuels Campaign is studying fuels that can transmute long-lived transuranic isotopes contained in UNF into shorter-lived fission products. Advanced fuels have been designed, fabricated, and irradiated to test transmutation fuel concepts. PIE of these fuels provides data on fuel behavior and irradiation performance; these data are used to determine feasibility of advanced fuel designs, understand effects of minor actinide additions to fuel, and support modeling and simulation.

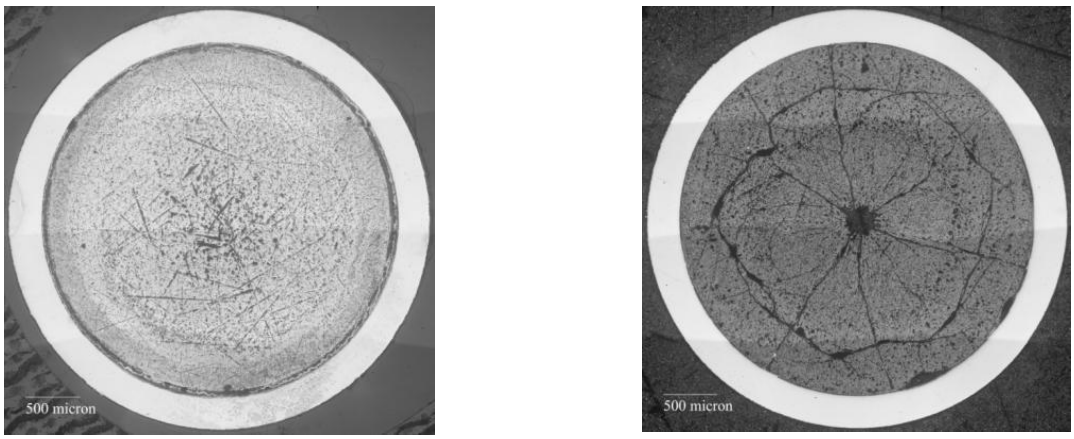
Research and Development Overview

PIE provides data on fuel behavior and irradiation performance. Phenomena studied include dimensional changes, fission gas release, fuel microstructure, cladding and fuel microhardness, and compositional changes. Irradiation testing and PIE are required to determine the feasibility of advanced fuel concepts and designs and may confirm expected performance or provide insight into unexpected phenomena.

Accomplishments

In FY 2011, baseline PIE was completed on three AFC-1 fuel experiments, three AFC-2 fuel experiments, and four legacy FFTF fuel experiments. Baseline PIE includes visual and dimensional inspection, neutron radiography, gamma scanning, fission gas analysis, optical microscopy, microhardness testing, and analytical chemistry. AFC-1 experiments included AFC-1D (metallic), 1G (nitride and metallic), and 1H (metallic). AFC-2 experiments included AFC-2A (metallic), 2B (metallic), and 2C (MOX). FFTF experiments included pins from ACO-3 and FO-2 (MOX), and MFF3 and MFF5 (metallic).

Future work will include PIE using advanced techniques to study higher-resolution fuel microscopy and constituent redistribution, PIE of higher burnup experiments, and comparison of results from ATR irradiation experiments to fast reactor experiments using results from EBR-II, FFTF, and FUTURIX experiments.



(a) (b)

Figure 1. Optical microscopy of AFC fuel rodlets: (a) AFC-1H Rodlet 4 U-29PU-4AM-2Np-30Zr, 3.91E21 f/cm^3, 36.00 at.% HM, 26.68 at.% fissile, and (b) AFC-2C Rodlet 2 $(U_{0.80},Pu_{0.20})O_{1.98}$, 1.25E21 f/cm^3, 7.11 at.% HM, 13.78 at.% fissile.

5.5 Abstract: Microencapsulated Fuels and Support of Accident Tolerant Fuel Development

Lance Snead, Oak Ridge National Laboratory

Introduction and Objectives

The scope of this technical area is the application of coated particle fuel technology for advanced reactor platforms including LWR applications. It is highly integrated with direct modeling, fuel fabrication, irradiation testing, and post-irradiation performance evaluation. FY 2011 focus areas included modeling, TRU tri-isotropic (TRISO) development, LWR microencapsulated fuel development, and the testing of advanced clad and fuel materials being developed for enhanced safety under FCR&D and other programs.

Research and Development Overview

This campaign is broken into functional areas of directed modeling, fuel utilization and safety analysis to guide development efforts, fuel R&D primarily focused on (but not limited to) the new fully ceramic microencapsulated (FCM) fuel, and an irradiation/qualification program. Mid-year the program began a program of assessing the performance of advanced fuels concepts, including those in the FCR&D program, under assumed beyond-design-basis accident conditions, focusing on high-temperature steam reactions.

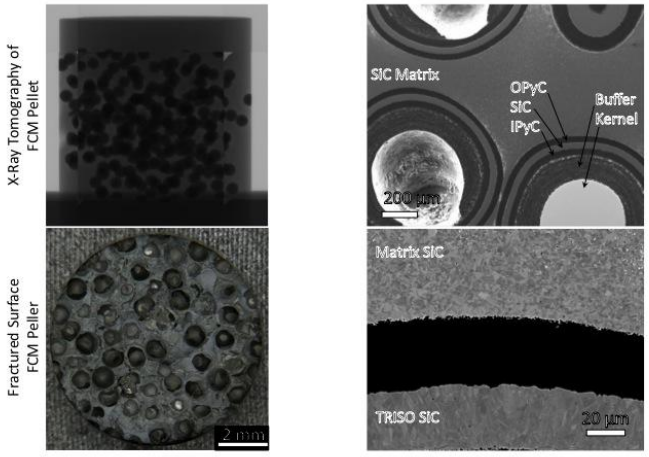


Figure 1. The project fabricated the first uranium-bearing FCM fuel and carried out an intensive program for optimizing the matrix for this fuel system.

Accomplishments

- Fabricated the first uranium-bearing FCM fuel and carried out an intensive program for optimizing the matrix for this fuel system (see figure).
- Designed the irradiation campaign and received the first positive results on FCM fuel.
- Conducted accident testing on FCM fuel and advanced fuel and cladding materials.

- Incorporated SiC and graphite matrix physical properties into the FRAPCON code and performed a preliminary analysis to assess fuel temperatures under LWR operating conditions.
- Performing prolonged high-pressure, high-temperature steam oxidation tests to probe the beyond-design-basis accidents regime.

5.6 Abstract: Ceramic Fuels Technologies

Ken McClellan, Los Alamos National Laboratory

Introduction and Objectives

The Ceramic Fuels Technology area within the Advanced Fuels Campaign addresses fuel fabrication and fuel performance R&D for conventional pellet fuels and targets. Traditionally this technical area has focused on oxide- and nitride-based pellet fuels and targets as well as ceramic/ceramic composite fuels. This area does not include ceramic fuels based upon coated particle technology.

The ceramic fuel development approach has transitioned to a fundamental behavior-based approach and so is substantially spectrum-independent and thus supports LWR and FR fuels. The knowledge and data from this technical area and the coordination with modeling efforts will help enable improved safety and reliability of existing reactors as well as accelerated fuel development and licensing for transmutation and advanced fuels.

Research and Development Overview

The FY 2011 focus of ceramic fuels has been exclusively on oxide-base fuel systems, specifically UO_2, minor actinide-bearing MOX and ThO_2-based fuels. The area substantially completed the transition to a science-based approach structured to employ separate effects testing (SET) to develop increased fundamental understanding of fuel processing and performance and generate data sets necessary to enable development of physics-based predictive models. While the emphasis is on a SET approach, integral effects testing continues to be a critical component of the R&D approach in order to enable periodic assessment of the assumptions made in efforts to isolate or "separate" phenomena and to provide data that can be used to validate codes and to compare to historical irradiation test data.

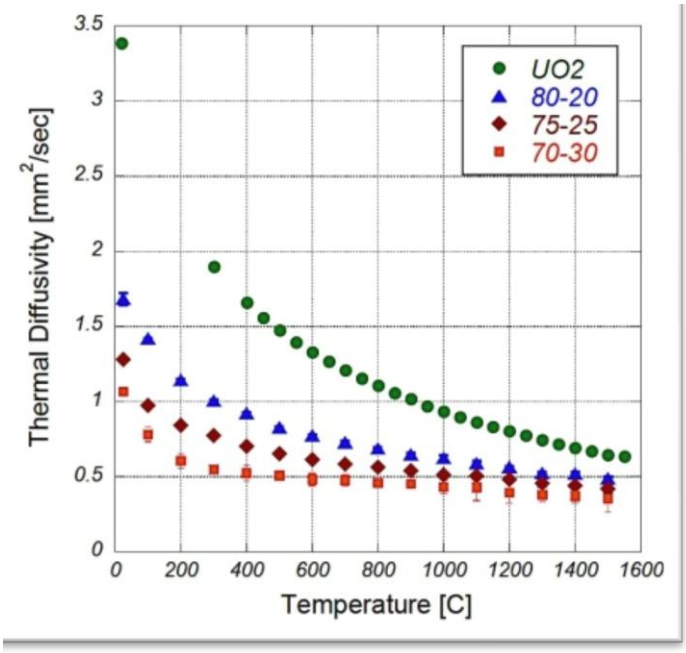


Figure 1. Thermal diffusivity of UO_2 and MOX fuel as a function of temperature and U-Pu ratio.

This technical area uses a conventional materials science

approach to define the relationships between fuel chemistry, feedstock, and fabrication process and the resultant fuel properties and final in-pile performance for the oxide fuel systems of programmatic interest. Critical aspects of this approach include making reference materials through advanced fabrication techniques and subsequent testing of these materials in experiments specifically constructed to increase our fundamental understanding of ceramic fuels and to provide necessary datasets for modeling. The fundamental understanding and SET data are required for predictive model development. Figure 2 shows the relative roles in the process of each collaborating organization.

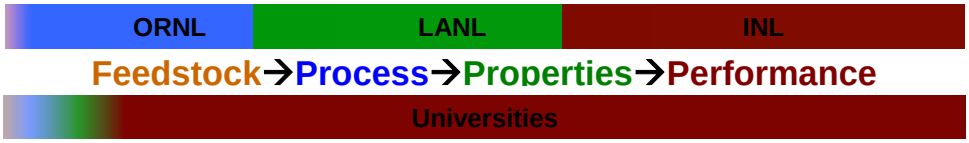


Figure 2. Relative role of project participants.

Accomplishments

- Fuel development and irradiation test plans for UO_2- and ThO_2-based fuels
- Synthesis of reference feedstock and engineered samples for SET experiments
- Development of *in situ* thermal diffusivity determination during sintering and master sintering curves for robust, advanced UO_2 fuel fabrication

5.7 Abstract: Metallic Fuels Technologies

Rory Kennedy, Idaho National Laboratory

Introduction and Objectives

The Metallic Fuels Technology area of the Advanced Fuels Campaign is charged with advancing the science and engineering of metal alloy and related fuel types primarily for application in fast reactors for transmutation of long-lived transuranic actinide isotopes and, recently, for application in LWRs for accident tolerance. In support of these objectives, the technical area develops and/or adapts state-of-art instrumentation and analysis techniques for performing measurements on radioactive and highly radioactive materials. Primary thrust areas include the acquisition and purification of actinide feedstocks, fuel fabrication and fabrication process development, the characterization of irradiation test fuels and materials, the study of the fundamental properties and behavior of fuels and fuel constituents in conjunction with the program's modeling and simulation efforts, the development of advanced characterization techniques, and the development of innovative fuel concepts. An important near-term objective of the technical area is to demonstrate the remote (i.e., hot cell) fabrication of metallic transmutation fuels directly coupled to a remotely operated separations process for irradiation testing.

Accomplishments

Highlights for FY 2011 include the fabrication of the AFC-3A and AFC-3B irradiation test fuels wherein both solid and annular fuel forms were produced (Figure 1). Annular fuel had not previously been produced and required a significant development effort. In addition to the integral irradiation test fuel fabrication, 60 TEM samples intended for short-term irradiations in the HFIR via the recently implemented rabbit system were prepared and characterized (20 were shipped to Oak Ridge National Laboratory).

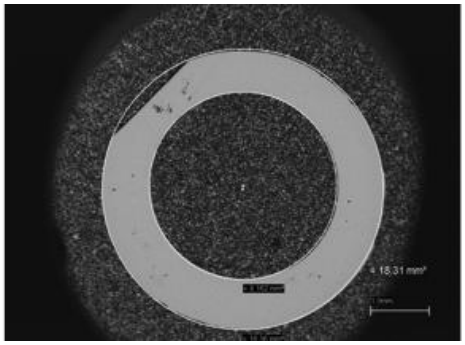


Figure 1. Scanning electron micrograph of a sample of the fabricated AFC-3 annular fuel.

A noteworthy advance in understanding fuel behavior was provided by the successful radial profiling of thermal diffusivity on four irradiated metallic fuel samples at spacing of 25 μm using the newly developed and deployed Scanning Thermal Diffusivity Microscope (STDM). These data are currently under analysis. In addition to the implemented STDM studies, the shielded Electron Probe Micro Analyzer (EPMA) was installed and, along with the microfocus X-ray

Diffractometer (μ-XRD) and the Focused Ion Beam (FIB), operated with radioactive materials. In the case of the FIB, irradiated fuel samples were prepared.

For FY 2012, much of the work will continue within each of the thrust areas. In addition, there is a significant effort under a new Cooperative Research and Development Agreement (CRADA) agreement with the Korea Atomic Energy Research Institute (KAERI) to initiate design of equipment to demonstrate the fabrication of recycle fuel under remote (hot cell) conditions.

5.8 Abstract: Core Materials Technologies

Stuart Maloy, Los Alamos National Laboratory

Introduction and Objectives

The FCR&D program is investigating methods of dealing with transuranics in various fuel cycle options and is supporting the development of next-generation LWR fuels. To achieve this goal, new fuels and cladding materials must be developed and tested to high burnup levels (greater than 20%) and under accident conditions. To achieve such high burnup levels, the fast reactor core materials (cladding and duct) must be able to withstand very high doses (greater than 200 dpa) while in contact with the coolant and the fuel. Thus, these materials must withstand radiation effects that promote low-temperature embrittlement, radiation-induced segregation, high-temperature helium embrittlement, swelling, irradiation creep, corrosion with the coolant, and chemical interaction with the fuel (FCCI).

Research and Development Overview

To develop and qualify materials to a total fluence greater than 200 dpa requires development of advanced alloys and irradiations in fast reactors to test these alloys. The research team is presently testing specimens of ferritic/martensitic alloys (T91/HT-9) previously irradiated in the FFTF reactor up to 210 dpa at a temperature range of 350°C to 700°C. This activity includes analysis of a duct made of HT-9 after irradiation to a total dose of 155 dpa at temperatures from 370°C to 510°C. Advanced radiation-tolerant materials are also being developed to enable the desired extreme fuel burnup levels. Specifically, coatings and liners are being developed to minimize FCCI, and research is under way to fabricate large heats of radiation-tolerant oxide dispersion steels with homogeneous oxide dispersions.

Accomplishments

Significant accomplishments have been achieved in FY 2011 to develop and qualify materials to a total fluence greater than 200 dpa:

- Detailed measurements were made on an ACO-3 duct at Los Alamos National Laboratory, including phase analysis through TEM, small-angle neutron scattering, and neutron diffraction studies; rate jump testing was performed on irradiated tensile specimens to aid development of multi-scale models.
- Thermal annealing studies have been performed at Oak Ridge National Laboratory on specimens from the ACO-3 duct at

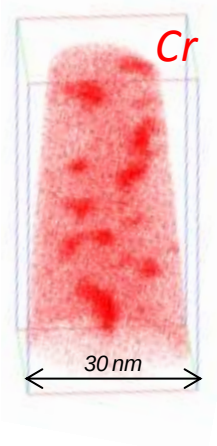


Figure 1. Atom probe analysis on specimen from ACO-3 duct (dose=20 dpa, T_{irr}=380°C).

550°C and 650°C, followed by fracture toughness testing showing significant recovery of properties.

- Mechanical testing has been performed at Pacific Northwest National Laboratory on specimens of MA-957 after irradiation to >100 dpa, showing retention of ductility after testing at room temperature.
- The design of an AFC-3 materials capsule was completed at Idaho National Laboratory, including cladding-fuel-liner diffusion couples for irradiation in the ATR.
- Advanced oxide dispersion-strengthened alloys, 14YWT, have been produced from a >50 kg heat. A uniform oxide dispersion was produced for high creep resistance and improved radiation tolerance.

5.9 Abstract: Nuclear Data

Tony Hill, Idaho National Laboratory

Introduction and Objectives

The poorly understood uncertainties in the existing nuclear data propagate into uncertainties in calculated integral quantities, driving margins and costs in advanced system design, operations and safeguards. This research effort contributes to the resolution of technical, cost, safety, security and proliferation concerns in a multi-pronged, systematic, science-based R&D approach. The objective is identify, prioritize, develop and deliver the required nuclear and covariance data, at the needed precision, to support operation, design and safety performance of future nuclear systems and fuel cycles. Findings will provide fundamental, cross-cutting support for Objectives 2, 3 and 4 from the *Nuclear Energy Research and Development Roadmap* and the FCT campaigns.

Research and Development Overview

Nuclear data plays a fundamental role in many calculations performed throughout the fuel cycle. This research effort identifies and develops small-scale, phenomenon-specific experiments informed by theory and engineering to minimize the reliance and expense of large integral experiments. Also included in this effort is the identification and inclusion of high-quality data from previous large, expensive integral experiments, ensuring past investments are realized and future data requirements are minimized. Research activities are leveraged by effective collaborations between experiment and theory and between DOE programs and offices, national laboratories and universities, both domestic and international.

Accomplishments

New point-wise nuclear data evaluations for the primary fast reactor structural materials were generated, along with energy-dependent covariance data. A Monte Carlo-based nuclear model has been implemented and used to produce the world's first outgoing fission neutron spectrum evaluation, along with full covariance data, as a function of energy with all relevant physical correlations included.

A new Pu-239 capture cross-section measurement was completed at Los Alamos Neutron Science Center (LANSCE) using the newly-developed Parallel Plate Avalanche Counter (PPAC), and the fission Time Projection Chamber (TPC) successfully collected the world's first fully reconstructed fission product tracks emerging from neutron-induced fission of U-238.

University Poster Presentations

6.1 Abstract: Neutron and Gamma-Ray Cross-Correlation Measurements of MOX Fuel Using Liquid Scintillators

Eric C. Miller, Jennifer L. Dolan, Sara A. Pozzi, Marek Flaska,
Shaun D. Clarke, and Paolo Peerani
University of Michigan

The detection of special nuclear material (SNM) is a very difficult problem: typically, the signals are weak, thus difficult to distinguish from the background, and in many practical applications additional shielding can be expected. In order to detect SNM more accurately, new methods to isolate the source signal are needed. Using cross-correlation functions, it may be possible to identify the time-correlated particles from fission events above the background. To examine this feasibility, measurements were performed on 1-kg MOX fuel samples, a ^{252}Cf source, and an americium–beryllium source. Using pulse shape discrimination, the particle types were isolated, and the individual correlation contributions were determined. By comparing the (n,n) peak of the correlation curves, a distinct difference is observed between a spontaneous fission source and an (α,n) source.

6.2 Abstract: Synthesis and Characterization of Low-Valent Binary Technetium Chlorides

Erik V. Johnstone, University of Nevada at Las Vegas

Technetium-99 (^{99}Tc) is a pure beta emitter with a long half-life ($T_{1/2}$ = 2.1x10^5 years) produced in kilogram quantities from the fission of uranium-235 fuel in nuclear reactors. Its complex chemistry and ability to form oxides, halides, nitrides, sulfides, carbides, etc, and range of oxidations states (-1 to +7) make it a very interesting radionuclide to study. In order to better understand how technetium behaves in the nuclear fuel cycle and other areas of applied science, it is necessary to develop the fundamental chemistry of technetium.

One interesting facet of fundamental technetium chemistry is the binary technetium halide systems. Until recently, three binary technetium halides were known: TcF_6, TcF_5, and $TcCl_4$. Research at the University of Nevada at Las Vegas in the past few years has contributed two more binary technetium halides with the additions of $TcBr_4$ and $TcBr_3$. In this work, low-valent binary technetium chlorides were synthesized in the solid state and analyzed using various physicochemical characterization methods, including single-crystal and powder x-ray diffraction (XRD), ultraviolet-visible spectrophotometry (UV-Vis), x-ray absorption fine structure (XAFS), and elemental analysis. Technetium dichloride was synthesized from the stoichiometric reaction of technetium metal and elemental chlorine at elevated temperatures as a novel compound with new structure-type with Tc_2Cl_8 units containing a Tc-Tc triple bond. α-$TcCl_3$ was prepared from the reaction of $Tc_2(O_2CCH_3)_4Cl_2$ with passing HCl (g) at elevated temperatures and characterized as a novel compound isostructural to $ReCl_3$ with a triangular Tc_3^{3+} core structure containing metal-metal double bonds generating the Tc_3Cl_9 motif. A second phase of $TcCl_3$ has also been identified from the reaction of stoichiometric amounts of elemental chlorine with technetium metal. The β-phase is isomorphous with α-$MoCl_3$ and α-$RuCl_3$ with a Tc-Tc metal-metal bond, and has been shown to decompose to the more stable α-phase of the trichloride.

The synthesis and structure of technetium tetrachloride have been previously determined, but reinvestigation of the compound has yielded a new synthetic method employing sealed Pyrex tubes as well as a study of its thermal properties. The low-valent technetium chlorides synthesized exhibit interesting chemical structures and unique physicochemical properties, which suggest these species to be optimal for potential waste forms or use in separations for the nuclear fuel cycle.

6.3 Abstract: Quantum Size Effects in the Electronic Structure Properties of γ –U (100) Nanolayers

Dayla R. Morrison and Asok K. Ray

University of Texas at Arlington

The generalized gradient approximation (GGA) to density functional theory (DFT) and hybrid density functional theory (HDFT) have been used to compute the layer-by-layer properties of γ-uranium (γ-U) in the (100) symmetry. As a precursor to the surface computations, bulk calculations were performed to determine the ground state electronic structure properties. The total energy was optimized at six different levels of theory: non-magnetic (NM), ferromagnetic (FM), and anti-ferromagnetic (AFM) with and without spin-orbit coupling (SOC), all at DFT and HDFT levels. The bulk modulus, B (GPa), was calculated from the Murnaghan equation of state.

For the DFT calculations without SOC, the AFM configuration resulted in the lowest energy, but with SOC included, NM was obtained as the ground state. With HYB-DFT, the NM state was also obtained when spin-orbit coupling was added but, without SOC, FM was found to be the ground state. The research team noted that while the lattice constant indicated relatively little preference to the level of theory, the bulk moduli oscillated significantly. The HYB-DFT showed a marked difference from experimental value, which indicated that while it *might* be a useful tool to describe *some* heavily correlated systems, it *appears* to be unreliable in predicting reasonable values for the bulk modulus for uranium.

The ground state bulk lattice constant and bulk modulus are found to be 3.46 Å and 113.75 GPa, respectively, at the non-magnetic with spin-orbit coupling level of theory, to be compared with the experimental values of be 3.47 Å and 113.3 GPa, respectively. As a consequence, the layer-by-layer surface calculations were performed at the non-magnetic level including spin-orbit coupling. The monolayer displayed a significant shrinking of the "effective" lattice constant of about 23.55% from bulk theoretical values. Further analysis of the change in energy per added "bulk" indicates that after 5 layers the energy stabilized and did not change by more than 10 mRy. The surface energy and the work function of the γ-U (100) surface are predicted to be 1.56 J/m^2 and 3.24 eV respectively. Electronic density of states plots of atoms located at the surface, subsurface and center of a hexa-layer slab indicated *some* localization of the *5f* electrons at or near the Fermi level with a gradual trend toward delocalization with increased depth within the slab.

6.4 Abstract: A Game Theoretic Approach to Safeguards Selection and Optimization

Rebecca M. Ward and Erich A. Schneider

The University of Texas at Austin

The anticipated global expansion of nuclear power promises to place unprecedented demands on the nonproliferation regime, making the optimization of safeguarding resources critical to international security. To that end, a novel safeguards modeling approach is being developed to determine inspector resource allocation strategies that optimize detection probability. The model uses a game theoretic component to compute the resource allocation strategy that optimizes detection probability. This component is coupled to a discrete event simulator, which generates detection probabilities for individual diversion–interdiction strategy pairs and feeds them into the game theory model. The inspector safeguarding strategy is subject to budget constraints, and a background detection probability is introduced to serve as a surrogate for all safeguarding activities not explicitly modeled. To verify the feasibility of this model, a proof of concept has been developed using a simple simulation model that calculates detection probabilities for an insider threat scenario and couples the results to a two-person zero-sum game. A sensitivity analysis was conducted to assess the effect of changes in budget and background detection probability on defender and attacker strategies, as well as overall detection probability.

6.5 Abstract: Using a Modified CINDER90 Routine in MCNPX 2.6.0 for the Prediction of Helium Production in Minor Actinide Targets

Carey M. Read, Jr., Travis W. Knight, and Kenneth S. Allen
University of South Carolina

Fast reactors containing heterogeneous minor actinide target rods are now being modeled. Studies of transmutation in these rods must consider helium production from α-decay since helium is produced in substantial quantities. This research utilized an innovative method to calculate gas production by modifying the CINDER90 depletion code used by MCNPX 2.6.0 to include helium production from α-decay. The modified CINDER90 code was verified using the ORIGEN-ARP module of SCALE6. The code was tested using the Sodium-cooled Heterogeneous Innovative Burner Reactor model created at the University of South Carolina. The modified version of the *cinder.dat* file includes helium production otherwise not available from the current version; therefore, the modified version should be distributed in subsequent MCNPX 2.6.0 releases for use in fast reactor calculations using heterogeneous minor actinide target rods.

6.6 Abstract: Characterization of Uranyl(VI) Nitrate Complexes in a Room-Temperature Ionic Liquid Using Attenuated Total Reflection-Fourier Transform Infrared Spectrometry

Donna L. Quach, Chien M. Wai, and Sofie P. Pasilis
University of Idaho

Room-temperature ionic liquids form potentially important solvents in novel nuclear waste reprocessing methods. Of current interest are the solvation, speciation, and complexation behaviors of actinides and lanthanides in room-temperature ionic liquids. In this study, the research team used attenuated total reflection-Fourier transform infrared spectrometry to characterize the coordination environment of uranyl(VI) in solutions of the room-temperature ionic liquid 1-butyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide containing either tetrabutylammonium nitrate or nitric acid. Both $UO_2(NO_3)_2$ and $UO_2(NO_3)_3^-$ species were detected in solutions containing tetrabutylammonium nitrate. $v_{as}(UO_2)$ for these two species were found to lie at 951 and 944 cm^{-1}, respectively, while $v_{as}(UO_2)$ arising from uranyl(VI) coordinated by bis(trifluoromethylsulfonyl)imide anions in 1-butyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide was found to lie at 968 cm^{-1}. In solutions containing nitric acid, only $UO_2(NO_3)_2$ was detected, due to the high water content. The $UO_2(NO_3)^+$ species was not detected under the conditions used in this study. From the results shown here, the research team concludes that infrared spectroscopy forms a valuable addition to the suite of tools currently used to study the chemical behavior of uranyl(VI) in room-temperature ionic liquids.

6.7 Abstract: Removal of ^{14}C from Irradiated Graphite for Waste Volume Reduction and Bulk Graphite Recycle: Thermal Treatment

Tara E. Smith and Mary Lou Dunzik-Gougar
Idaho State University

Public concerns regarding availability of energy and environmental health are driving the growth of nuclear energy. The United States and other countries are developing advanced "Generation IV" nuclear systems, as established by the Generation IV International Forum.

DOE commissioned Idaho National Laboratory to develop the Next Generation Nuclear Plant (NGNP), a high-temperature gas-cooled reactor (HTGR) which uses coated fuel particles embedded in graphite, a graphite reflector and core structural components, and helium coolant. As graphite is bombarded with neutrons in a reactor, carbon-14 (^{14}C) is produced through three distinct reactions with ^{13}C, ^{14}N, and ^{17}O. In existing irradiated reactor graphite, the majority of ^{14}C has been found on the graphite component surface. This location is indicative of the neutron activation of ^{14}N, because ^{14}N is present on the surface and in surface pores as adsorbed N_2 from air. Carbon-14 is relatively long-lived (half life = 5730 years) and would have significant mobility in groundwater and atmospheric systems as CO_2 evolving over time from irradiated graphite. Disposal of large irradiated graphite components from an HTGR would likely be costly, and the value of such pure nuclear-grade graphite is expected to increase, thus recycle may be an option.

A means of optimally removing the majority surface ^{14}C from the bulk graphite ^{12}C is being investigated. Pyrolysis and oxidation in a steam atmosphere have been suggested as ^{14}C decontamination methods. Fachinger et al. (2006) demonstrated the concept of thermal treatment of irradiated nuclear graphite in the presence of steam or oxygen. During thermal treatment, graphite samples are heated to a temperature in the range of 800°C –1500°C in the presence of inert argon gas, which carries any gaseous products released during treatment. While oxygen is naturally adsorbed onto the graphite, for some experiments argon is mixed with small quantities of oxygen or carbon dioxide gas to increase the oxidation rate of surface species. Oxygen combines with the surface to form carbon–oxygen bonds that are expected mostly in the form of carbon monoxide (CO), which gasify upon heating up the graphite. Graphite oxidation kinetics are highly temperature-dependent. Below 700°C, the oxidation is limited by the rate at which graphite chemically reacts with oxygen. By approximately 800°C, oxidation is limited by the diffusion of oxygen into, and product gases out of, the graphite pore structure. Above ~900°C, the rate limiting phenomenon is oxygen diffusion through the surface-boundary layer of gases. Experimental parameters, including treatment temperature, gas composition, and gas flow rate will be optimized to offer viable thermal treatment parameters to efficiently remove ^{14}C from bulk nuclear graphite.

6.8 Abstract: Extraction of Trivalent Lanthanides and Americium by Tri-n-octylphosphine Oxide from Ammonium Thiocyanate Media

Derek Brigham, Coralie Badajoz, Gerard Cote, and Kenneth L. Nash
Washington State University

An investigation of the solvent extraction of trivalent lanthanides and Am^{3+} from ammonium-thiocyanate media by tri(*n*-octyl)phosphine oxide (TOPO) in toluene has been completed. This system is of interest both for its potential as a means of separating transplutonium actinides from fission-product lanthanides and for use in thiocyanate-based solvent extraction systems. Partitioning was monitored using radiotracer techniques where appropriate, and for others, inductively coupled plasma optical emission spectrometry (ICP-OES) or inductively coupled plasma mass spectrometry (ICP-MS). The research team has investigated extraction behavior of all members of the lanthanide series (except for promethium) plus yttrium. Conditional enthalpies (all exothermic) were determined (for selected systems) from the temperature dependence of the extraction reaction. A comparison with nitrate media shows higher extractive power of TOPO in contact with thiocyanate media, arising at least in part from the lower heat of the phase transfer of thiocyanate (relative to nitrate). The moderate tendency of thiocyanic acid to partition into the extractant phase has been profiled. Slope analysis indicates that TOPO solvation decreases from four ($M(SCN)_3TOPO_4$) for the light members of the series to three (or less) for the heavy lanthanide ions; Am^{3+} is extracted with four TOPO molecules. Despite the decrease in Ln:TOPO stoichiometry across the series, extraction is generally flat for the light lanthanides and increases from Gd^{3+} to Lu^{3+}. The extraction of Am^{3+} from mildly acidic ammonium-thiocyanate media was found to be at least 10 times stronger than that of the lanthanides between La^{3+} and Gd^{3+}.

6.9 Abstract: Characterization of Electrodeposited Technetium on Gold Foil

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The reduction and electrodeposition of TcO_4^- on a smooth gold foil electrode with an exposed area of 0.25 cm^2 was performed in 1 M H_2SO_4 supporting electrolyte using bulk electrolysis with a constant current density of 1.0 A/cm^2 at a potential of -2.0 V. Significant hydrogen evolution accompanied the formation of technetium (Tc) deposits. Technetium concentrations of 0.01 M and $2x10^{-3}$ M were electrodeposited over various times. Deposited fractions of Tc were characterized by powder x-ray diffraction, x-ray absorption fine structure spectroscopy, and scanning electron microscopy with the capability to measure semiquantitative elemental compositions by energy-dispersive x-ray emission spectroscopy. Results indicate the presence of Tc metal on all samples as the primary electrodeposited constituent for all deposition times and Tc concentrations. Thin films of Tc have been observed, followed by the formation of beads that are removable by scratching. After 2000 s, the quantity of Tc removed from solution and deposited was 0.64 mg Tc per cm^2. Following electrodeposition, the solution showed characteristic absorbances near 500 nm, corresponding to hydrolyzed Tc(IV) produced during deposition of Tc metal. No detectable Tc(IV) was deposited to the cathode.

6.10 Abstract: INFUPOD2: The Creation and Use of an International Nuclear Fuel Cycle Modeling Policy Development Tool

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With international models setting standards for possible future nuclear fuel cycles, a common problem when communicating these options is the complexity of the tool in which the model was created. For this reason, a program known as the International Nuclear Fuel Cycle Modeling Policy Development Tool (INFUPOD) was developed. It is based on a reactor and fuel chain with simplistic visual and numerical input and output. The main objectives and constraints of INFUPOD were that it must be simple to use, with the primary task of serving as a tool to educate the user about fuel and dynamic reactor systems. The tool offers options for such variables as initial settings, growth rate, and capacity ratios to allow for model variation. The majority of the options provided to the user will be located in an attached spreadsheet to minimize the need for action in the equations input of the systems dynamics program Vensim. INFUPOD will not include full isotopic tracking but rather will follow only uranium, plutonium, minor actinides, and fission products throughout the entire fuel cycle. These will be input with simple defined "recipes" created from a transmutation library. Options for up to three reactor types and three fuel types will be provided with basic cases available for selection, such as Light Water Reactor–Uranium Oxide Fuel (LWR-UOX), Light Water Reactor–Mixed Oxide Fuel (LWR-MOX), and Fast Reactor–Metallic Fuel (FR). With a wide range of possible output parameters, a development of multiple policy structures is needed to accommodate a global scale of nuclear reactor fuel cycles.

With a wide range of users, the input and output graphics are of great importance to allow for evaluation of model dynamics based on a wide range of options. As noted above, the objective of INFUPOD is to be a dynamic educational tool rather than a completely accurate simulation of the complex fuel and reactor system currently operating; therefore, the tool has a greater flexibility in application to users in preparation for a possible expanded use of dynamic system modeling and simulation. With an objective of an appropriate number of options for model variation, upon interaction with INFUPOD the user should feel comfortable with basic system dynamic of nuclear reactor and fuel systems. The goal of the tool is to assist those who influence policy to become more confident in making informed decisions concerning the nuclear fuel cycle.

6.11 Abstract: Bringing Resolution to the Nuclear Fuel Cycle: The Complexities of Spent Nuclear Fuel

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The use of nuclear power has generated large amounts of waste in the form of spent nuclear fuel. This spent fuel has presented many challenges to the energy industry and the governments responsible for the management of this waste material. Any solution to this problem must create workable fixes to the various dynamics involved in the security, economic, and environmental considerations that accompany high-level nuclear waste disposition. Looking at the most likely management scenarios—temporary dry-cask storage, geological repository, reprocessing—a cost–benefit analysis can be made. The most workable solution to the waste management problem is the expansion of temporary dry-cask storage with eventual geological storage or reprocessing as a long-term resolution.

6.12 Abstract: A Drop-in Concept for Deep Borehole Canister Emplacement

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Disposal of high-level nuclear waste in deep boreholes drilled into crystalline bedrock (i.e., granite) is an interesting repository alternative of long standing. Work at the Massachusetts Institute of Technology over the past two decades, and more recently in collaboration with the Sandia National Laboratories, has examined a broad spectrum of design aspects associated with this approach.

For emplacement, past reports suggest using steel cables to lower each canister into the borehole. This process would require many years to complete and precise control to safely lower the canisters thousands of meters. The current study evaluated a simple, rapid, "passive" procedure for emplacement of canisters in a deep borehole: free-fall release into a water-flooded borehole.

The project involves both analytic modeling and $1/5^{th}$-scale experiments on a laboratory mockup. Experiments showed good agreement and validated the model. Depending on the inputs used for the mass and dimensions of the full-scale canister and the viscosity of water, the model predicted terminal velocities of 2.4–2.6 m/s (4.5-5.8 mph). Further experiments showed that this could be reduced by 50% by making the surface hydraulically rough. Based on these predictions and a structural analysis, there seems to be little risk of damage when a canister bottoms out on a stack of previously loaded canisters. For reference, dropping the canister in air from a height of only 0.3 m (about one foot) would result in an impact velocity of 2.44 m/s.

Cost estimates for the conventional drill string-based method were developed, and the research team concluded that the drop-in method would reduce emplacement costs and time by at least 70%, down to $700,000 per borehole. A simple drop-in procedure deserves serious consideration for adoption as a standard procedure for borehole loading.

Appendix A. Acronyms

μm	Micron(s)
μ-XRD	Microfocus X-Ray Diffractometer
3-D	Three-Dimensional
Å	Angstrom
AFC	Advanced Fuel Cycle
AFM	Anti-Ferromagnetic
Am	Americium
AMUSE	Argonne Model for Universal Solvent Extraction
An	Actinide
At	Astatine
ATR	Advanced Test Reactor
B&B	Breed and Burn
BRC	Blue Ribbon Commission
C	Carbon
Ce	Cerium
Cf	Californium
Cl	Chlorine
cm	Centimeter(s)
Cm	Curium
CMPO	Octyl*(Phenyl)*-N,N-Diisobutylcarbonoylmethyl-Phosphine Oxide
CNT	Carbon Nano-Tubes
Co	Cobalt
COEX	Co-Extraction (of actinides)
CRADA	Cooperative Research and Development Agreement
CZT	Cadmium-Zinc-Telluride
DFT	Density Functional Theory
DMDOHEMA	Dimethyl-Dioctyl-Hexyl-Ethoxy-Malonamide
DOE	Department of Energy
dpa	Displacements Per Atom
DRIE	Deep Reactive Ion Etching
DTPA	Diethylenetriamine Pentaacetic Acid
EBR	Experimental Breeder Reactor
EPA	Environmental Protection Agency
EPMA	Electron Probe Micro Analyzer
EPRI	Electric Power Research Institute
EROI	Energy Return on Investment
eV	Electron-volt
FAST	Fuel Aging in Storage and Transportation
FCM	Fully Ceramic Microencapsulated
FCR&D	Fuel Cycle Research and Development (program)
FCT	Fuel Cycle Technologies (program)
FFTF	Fast Flux Test Facility
FIB	Focused Ion Beam
FM	Ferromagnetic
FR	Fast Reactor
FY	Fiscal Year
G4-ECONS	Generation IV Excel Calculation of Nuclear Systems (an economic evaluation tool for Gen IV reactors)
Gd	Gadolinium
GGA	Generalized Gradient Approximation
GPa	Gigapascal

GWd/MTU	Gigawatt Days per Metric Ton Uranium
HDEHP	Bis-(2-Ethylhexyl) Phosphoric Acid
HDFT	Hybrid Density Functional Theory
He	Helium
HLW	High-Level (Radioactive) Waste
HFIR	High-Flux Isotope Reactor
HTGR	High-Temperature Gas-Cooled Reactor
I	Iodine
IAEA	International Atomic Energy Agency
INFUPOD	International Nuclear Fuel Cycle Modeling Policy Development Tool
ISFSI	Independent Spent Fuel Storage Installation
J	Joule(s)
KAERI	Korea Atomic Energy Research Institute
kg	Kilogram(s)
La	Lanthanum
LANSCE	Los Alamos Neutron Science Center
LEU	Low-Enriched Uranium
LLNL	Lawrence Livermore National Laboratory
Ln	Lanthanide
LOCA	Loss of Coolant Accident
LPCVD	Low-Pressure Chemical Vapor Deposition
LTA	Lead-Test Assembly
Lu	Lutetium
LWR	Light Water Reactor
m	Meter(s)
M	Molar
MA	Minor Actinide
MIP	Multi-Isotope Process
MOX	Mixed Oxide
MPACT	Material Protection, Accounting, and Control Technologies
mph	Miles Per Hour
mRy	milli-Rydbergs
MW_{th}	Megawatts Thermal
N	Nitrogen
NE	Office of Nuclear Energy
NEAMS	Nuclear Energy Advanced Modeling and Simulation
NE-COST	(code for equilibrium economic analysis of complex fuel cycles)
NGNP	Next Generation Nuclear Plant
nm	Nanometer(s)
NM	Non-Magnetic
NMR	Nuclear Magnetic Resonance
Np	Neptunium
O	Oxygen
P	Phosphorus
PIE	Post-Irradiation Examination
PPAC	Parallel Plate Avalanche Counter
ppm	Parts Per Million
Pu	Plutonium
PWR	Pressurized Water Reactor
R&D	Research and Development
RD&D	Research, Development and Demonstration
RepU	Reprocessed Uranium
s	Second(s)
SEM	Scanning Electron Microscope
SET	Separate Effects Testing

SFR	Sodium Fast Reactor
SiC	Silicon Carbide
SNF	Spent Nuclear Fuel
SNM	Special Nuclear Material
SPS	Spark Plasma Sintering
SOC	Spin Orbit Coupling
STDM	Scanning Thermal Diffusivity Microscope
STMAS	The Sigma Team for Minor Actinide Separation
TALSPEAK	Trivalent Actinide–Lanthanide Separation by Phosphorus Reagent Extraction from Aqueous Complexes
TBP	Tributylphosphate
Tc	Technetium
Th	Thorium
tHM/y	Tonnes Heavy Metal Per Year
TMA	Technical Mission Area
TOPO	Tri(*n*-octyl)phosphine Oxide
TPC	Time Projection Chamber
TRISO	Tri-Isotropic
TRU	Transuranic(s)
TRUEX	Transuranium Extraction
UFD	Used Fuel Disposition Campaign
UNF	Used Nuclear Fuel
UOX	Uranium Oxide
UREX	Uranium Recovery by Extraction
URL	Underground Research Laboratory
UV-Vis	Ultraviolet-Visible Spectrophotometry
V	Volt(s)
W/m-K	Watts per Meter-Kelvin
XAFS	X-Ray Absorption Fine Structure
XRD	X-Ray Diffraction