Fuel Cycle Technologies Annual Review Meeting Transactions Report





Idaho National Laboratory November, 2015

About the Cover

Clockwise from upper left corner

- High density (95% theoretical) enriched test pellets were fabricated at LANL for the upcoming test irradiation at ATR.
- Quartz tube coil filled with a two phase mixture of aqueous solution and solvent for the innovative SANEX process in the gamma irradiator at MFC.
- U₃Si₂ pellet samples were fabricated using differing feedstock particle size distribution and sintering temperature; then characterized to examine the as fabricated microstructure. This is a three-year research program at the University of South Carolina includes fabrication, testing, and modeling of a high uranium density, advanced nuclear fuel (U₃Si₂).
- Optical microscopy of AFC-3A Rodlet 5 possibly exhibiting two zone Zr redistribution behavior typically observed in historic irradiations of U-10Zr fuel.
- Optical micrograph of UN/U₃Si₂ composite showing optimized microstructure where the U₃Si₂ phase (light) surrounds the UN (dark) to protect the UN from washout during normal operation.
- Background: An initial melt of a Synroc-based ceramic waste form using the Cold Crucible Induction Melter.



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Fuel Cycle Technologies Annual Review Meeting Transactions Report

November 3-5, 2015

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Introduction



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FUEL CYCLE TECHNOLOGIES PROGRAM

1. INTRODUCTION

The Fuel Cycle Technologies (FCT) program supports the Department of Energy's (DOE's) mission to "Enhance U.S. security and economic growth through transformative science, technology innovation, and market solutions to meet our energy, nuclear security, and environmental challenges." Goal 1 of DOE's Strategic Plan is to develop innovative energy technologies that enhance U.S. economic growth and job creation, energy security, and environmental quality. FCT does this by investing in advanced technologies that could transform the nuclear fuel cycle in the decades to come. Goal 2 of DOE's Strategic Plan is to strengthen national security by strengthening key science, technology, and engineering capabilities. FCT does this by working closely with the National Nuclear Security Administration and the U.S Department of State to develop advanced technologies that support the Nation's nuclear nonproliferation goals.

DOE published its second Quadrennial Technology Review (QTR) in September 2015. The QTR "examines the status of the science and technology that are the foundation of our energy system, together with the research, development, demonstration, and deployment (RDD&D) opportunities to advance them." It is intended to "assist decision makers as they set priorities, within budget constraints, to develop more secure, affordable, and sustainable energy services." The QTR notes that future developments in the electricity production sector will likely include a mix of fossil-based generation with carbon capture and storage, nuclear energy, and renewables, such as solar and wind. FCT is pursuing the RDD&D that the QTR notes is crucial to develop advanced reactor designs that will enable sustainable nuclear fuel cycles of the future. Additionally, FCT is addressing the four key challenges noted in the QTR that would otherwise limit the ability of





September 2015

nuclear energy to widely expand: safety, economics, waste management, and proliferation of nuclear weapons.

The mission of FCT is to (1) conduct generic R&D and other generic activities related to used nuclear fuel (UNF), nuclear waste management, and disposal issues and (2) conduct R&D related to advanced sustainable fuel cycle technologies that have the potential to improve resource utilization and energy generation, reduce waste generation, enhance safety, and limit proliferation risk. In addition, the program is laying the ground work for implementing the Administration's strategy on the management of spent nuclear fuel (SNF) and high-level waste (HLW). The program employs a long-term, science-based approach to foster innovative, transformational technology solutions to achieve this mission. Advancements in fuel cycle technologies and solutions support the enhanced availability, affordability, safety, and security of nuclear-generated electricity in the U.S.

The nuclear fuel cycle consists of acquiring the uranium resources and fabricating fuel on the "front end," using that fuel to produce heat and electricity in reactors, and then storing and disposing of the used fuel on the "back end." This is the "once through" fuel cycle currently being developed in the U.S. The back end could also include recycling used fuel in various ways to produce new fuel. FCT is conducting R&D on advanced technologies covering many aspects of the front end and the back end of the nuclear fuel cycle. FCT also works closely with other programs in DOE's Office of Nuclear Energy (NE) that focus on advanced reactor technology development. FCT employs an integrated systems approach in fuel cycle technology development because technologies employed in one aspect of the fuel cycle will impact the performance throughout other aspects of the fuel cycle. This is accomplished through systems analyses, engineering, and integration.



Figure 1. The front and back end of the nuclear fuel cycle.

FCT R&D consists of eight technical campaigns that correspond to the elements of the nuclear fuel cycle in Figure 1.

Front End

- 1. Fuel Resources works to assure that economic nuclear fuel resources remain available. The campaign is currently investigating the viability of recovering uranium from seawater. FCT also supports DOE's uranium management policies.
- 2. Advanced Fuels conducts near term R&D of advanced light-water reactor (LWR) fuel and longterm R&D of transmutation fuel. The campaign also develops advanced capabilities to support a science-based approach to fuel development.

Back End

- **3.** Nuclear Fuel Storage and Transportation lays the groundwork and develops options for decision makers on the design of an integrated waste management system with special emphasis on a consolidated storage facility and associated transportation activities.
- 4. Material Recovery and Waste Form Development researches advanced fuel cycle material recovery and waste management technologies that improves current fuel cycle performance and enables a sustainable fuel cycle with minimal processing, waste generation, and potential for human diversion.
- 5. Joint Fuel Cycle Studies collaborates with the Republic of Korea to assess the technical and economic feasibility and nonproliferation acceptability of electrochemical recycling for managing used fuel.
- 6. Used Fuel Disposition R&D identifies alternatives and conducts scientific research and technology development to enable storage, transportation, and disposal of used fuel and wastes generated by existing and future fuel cycles.

Integrating Campaigns

- 7. Materials Protection, Accounting and Control Technologies develop the technologies and analysis tools to support the next generation of nuclear materials management and safeguards for future fuel cycles.
- 8. Fuel Cycle Options analyzes complex fuel cycle system options, assesses their overall performance under various scenarios, and improves the understanding of the interdependencies between subsystems and their associated technologies.

The table below provides the funding, including carryover, for each campaign in FY-15. It excludes funding for Nuclear Energy University Programs, Small Business Innovative Research/Small Business Technology Transfer, program management and support at Idaho National Laboratory, and special projects and support services managed at DOE-NE.

Fiscal Year-End 2015 Funding (Includes Carryover)

Fuel Cycle Technologies				
Major Activities	FY-15 Funding			
Fuel Resources Program	\$6,629,866			
Advanced Fuels Campaign	\$61,015,187			
Nuclear Fuel Storage and Transportation Planning Project	\$36,561,477			
Material Recovery and Waste Form Development Campaign	\$22,327,407			
Joint Fuel Cycle Studies Campaign	\$7,546,199			
Used Fuel Disposition R&D Campaign	\$32,473,239			
Material Protection, Accounting, & Control Technologies Campaign	\$6,151,020			
Fuel Cycle Options Campaign	\$10,548,225			
Total	\$183,252,620.00			

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Fuel Resources Program



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2. FUEL RESOURCES PROGRAM

2.1 Overview

S. Kung, Department of Energy

Mission

The Fuel Resources Program seeks to identify and implement actions to ensure that economic nuclear fuel resources remain available. The program will evaluate nuclear fuel resources and develop recovery technologies to enable a sustainable fuel cycle through increasing the fuel resources. Priority attention in the near term will focus on developing the technology for extraction of uranium from seawater.

Objectives

Develop advanced adsorbent materials that can simultaneously enhance uranium sorption capacity, selectivity, kinetics, and materials durability to reduce the technology cost and uncertainties.

- Develop a molecular-level understanding of the coordination modes, sorption mechanisms, and kinetics of uranium extraction.
- Design and synthesize functional ligands for extraction of uranium from seawater with higher capacity, selectivity, and stability.
- Investigate advanced high-surface area hybrid supports for separation of uranium from seawater that could lead to a major breakthrough.
- Develop marine testing methods to evaluate the extraction efficiency of uranium from seawater from polymeric and high-surface area hybrid supports.
- Develop sorbents with enhanced durability and recyclability.
- Assess the cost and energy return on investment for extraction of uranium from seawater with polymeric adsorbents.

Challenges

- Low concentration of uranium in seawater environment (3.3 ppb)
- Graft the chelating agents onto polymers or high-surface-area materials and retaining their selectivity, capacity, recyclability, and kinetics for uranium extraction
- Understand the primary variables that impact the technology cost.

R&D Strategies

The R&D strategy is to take advantage of recent developments in high-performance computing, advanced characterization instruments, and nanoscience and nano-manufacturing technology to enable technical breakthroughs. The technology-driven, science-based R&D efforts are focused on:

• Simulating and predicting structural and functional relationships using modern computational tools

- Applying advanced characterization tools to understand dynamic chemical processes at the atomic and molecular levels
- Synthesizing novel nanoscale adsorbent materials with architectures and functionality tailored for specific chemical performance.

Fiscal Year-End 2015 Funding (Includes Carryover)

Fuel Resources Program			
Major Activities	FY-15 Funding		
Fuel Resources Support Efforts	\$6,629,866		
Total	\$6,629,866		

Major Accomplishments

The Fuel Resources program is based on a multidisciplinary team with members from national laboratories, universities, and research institutes (Error! Not a valid bookmark self-reference.).



Figure 2. The DOE-NE Fuel Resources Program.

Program Goal: Participants seek to take advantage of recent developments in (1) high-performance computing, (2) advanced characterization instruments, and (3) nanoscience and nano-manufacturing technology to enable technical breakthroughs related to uranium recovery from seawater. The goal is to develop advanced adsorbents to reduce the seawater uranium recovery technology cost and uncertainties.

Under this program, Oak Ridge National Laboratory (ORNL) has developed new amidoxime-based adsorbents of high surface area, which tripled the uranium capacity of leading adsorbents synthesized by the Japan Atomic Energy Agency (JAEA). Adsorbent synthesis at ORNL is mainly based on radiation-

induced graft polymerization (RIGP), atom-transfer radical polymerization, and nano-synthesis. The focus is to develop adsorbents that will expand the uranium sorption capacity, improve uranium selectivity, accelerate adsorption kinetics, and enhance durability and reusability. Currently, ~70% of technology costs are attributed to expenditures associated with adsorbent materials. Increasing uranium adsorption capacity proportionally decreases the technology costs. The majority of adsorption binding sites are currently not occupied by uranium. A better understanding of the uranium coordination chemistry and mechanisms enables the development of tailored sites for uranium retention. Increasing reaction kinetics, on the other hand, reduces time required for adsorbent soaking in seawater. Furthermore, reducing materials degradation allows adsorbent reuse for several cycles, thus improving the technology cost.

ORNL adsorbents, as well as adsorbents developed at other laboratories, including universities participating in the Nuclear Energy University Program (NEUP), are tested with natural seawater at the Marine Sciences Laboratory (MSL) of the Pacific Northwest National Laboratory (PNNL). In addition to marine testing, the uranium from seawater program at PNNL/MSL performs adsorbent characterization and material durability, biofouling and toxicity testing, and marine deployment assessment. Three of the ORNL adsorbents (38H, AF1, and AI8) have undergone significant marine testing so far, and among these three adsorbent formulations, the AF1 adsorbent was proven to have the highest uranium adsorption capacity (3.86 ± 0.18 g U/kg adsorbent) at an exposure temperature of 20°C, during a 56-day seawater exposure, with a predicted saturation capacity of 5.4 g U/kg adsorbent. The uranium adsorption capacity of the AI8 was found to increase at a greater rate than AF1 and 38H with increasing temperature. New adsorbent materials have been tested with higher adsorption capacities (6.2 ± 1.5 g/kg for 56-day exposure in seawater). Work is under way to further confirm these results.

Overall, this program produced, by far, the highest-capacity adsorbents ever developed for uranium recovery from seawater. Additional adsorbent characterization is performed mainly by ORNL and PNNL in collaboration with other institutions. This characterization includes (1) physical characterization, e.g., surface morphology via neutron reflectivity and ellipsometry, tensile strength, and visual observation through scanning electron microscopy, and (2) chemical characterization, e.g., Fourier Transform Infrared (FTIR) spectroscopy, Nuclear Magnetic Resonance spectroscopy, X-ray Photoelectron Spectroscopy, and X-ray Absorption Fine Structure (XAFS) Spectroscopy. The objective is to better understand the chemical and physical characteristics of the adsorbents developed in order to further improve their performance and durability in such environment as seawater.

Thermodynamic and structural analyses of the seawater chemistry and the interaction of various seawater species with the adsorbent ligands, performed at Lawrence Berkeley National Laboratory (LBNL) in parallel with ORNL conducted computer-aided ligand modeling, provide the basic knowledge for the design of better ligands to improve uranium capacity and selectivity. Thermodynamic and kinetic adsorption modeling conducted at ORNL and PNNL in collaboration with NEUP participants provides information on rate-limiting adsorption mechanisms and effects of thermodynamic parameters, such as temperature, pH, and concentrations of various species, which can be used to improve the capacity and kinetics of uranium adsorption from seawater.

2.2 Engineering Uranyl Selectivity through Polymer Design and Advanced Spectroscopy

C. W. Abney, Oak Ridge National Laboratory

Introduction

Rapid economic growth of emerging countries in conjunction with development of new technologies drives an accelerating demand for a wide range of industrial metals and minerals. Reserves of such nonrenewable raw materials are often limited to a few countries, where social and political factors can affect supply security and result in severe global economic repercussions. Uranium is one such "critical metal," with nuclear fuel applications making its supply certainty a matter of global energy security. While studies have estimated terrestrial uranium ores can sustain 100 years of power generation at the current consumption rates, more than 1000× this quantity is dissolved in the world's oceans. Development of advanced sorbent materials could afford a near-limitless supply of uranium for the nuclear fuel cycle, or at the least achieve a financial backstop and ensure market stability. Attaining these ambitious tasks demands the design of adsorbents with high uranium affinity and selectivity.

Under the Fuel Resources program, ORNL has developed new high-surface-area amidoxime-functionalized polymeric adsorbents which have tripled the capacity of the previous state-of-the-art technology. However, enabling a financially viable means of harvesting this unconventional uranium reserve requires further advances in adsorbent recyclability, durability, and most of all selectivity, as exchanging the binding of nickel, iron, or vanadium for uranium would increase the uptake by 2, 4, and 10×, respectively.

To these ends, efforts at ORNL have focused on inverting the ratio of U:V as a means to improve adsorbent capacity. Extensive studies have been performed to identify how amidoximation time, temperature, and solvent influence selectivity, while ongoing research seeks to identify optimal comonomers to enhance adsorbent kinetics and promote selective uranium binding. Expanding upon the traditional RIGP techniques developed previously, the application of atom-transfer radical polymerization has afforded an entirely new avenue for surface functionalization of



Figure 3. Qualitative comparison of uranium XAFS spectra reveal distinct structural differences between seawatercontacted adsorbents and small molecule structural analogs proposed in the literature.

polymer adsorbents. Importantly, this approach facilitates a controlled polymerization mechanism, permitting tuning of the polymer morphology to promote stretching of polymer chains away from the adsorbent trunk. This relatively simple phenomenon imparts greater accessibility to the uranium-binding groups upon immersion in water.

Finally, ORNL researchers have leveraged advanced spectroscopic approaches to identify how amidoxime-functionalized polymers bind uranium and other transition metals prevalent in seawater. Relying upon DOE Basic Energy Sciences User Facilities, XAFS spectroscopy has been performed on

polymer adsorbents deployed for 56 days in environmental seawater (Figure 3). This synchrotron radiation-based technique is capable of providing local structural information, such as coordination number and bond lengths, for materials devoid of the long-range structural ordering necessary to accommodate crystallographic investigation. Importantly, as an element-specific technique, the binding environments for uranium, vanadium, and most other transition metals can be readily discriminated. These efforts revealed the coordination environment for uranium was not the same as predicted by previous small molecule-based investigations, challenging many long-held assumptions.

By mastering control of polymer morphology and positively identifying how uranium, vanadium, and transition metals are bound under environmental conditions, ORNL researchers intend to enable the rational engineering of advanced adsorbent materials for the selective extraction of uranium from seawater.

2.3 Computational Ligand Screening and Design for Extraction of Uranium from Seawater

V. S. Bryantsev, Oak Ridge National Laboratory

Extraction of uranium from seawater has received worldwide attention due to very high total amount of uranium dissolved in seawater, but low capacity and degradation of polymer fibers after several cycles of adsorption and stripping have plagued efforts to make this technology competitive compared to traditional uranium mining methods. At present, experimental research programs aimed at improving the performance of separation processes for recovery of uranium from seawater are mostly empirical in nature. Computer-aided molecular design and accurate screening of new host architectures can play a critical role in recognizing and selectively extracting uranium, which can eventually lead to improvements in sorption capacity of amidoxime-based and future generation adsorbents. In this talk, I will present the progress towards developing a computational framework for predictive modeling of complexation behavior of uranium $(UO_2^{2^+})$ and vanadium (VO_2^+, VO^{2^+}) oxometal ions and computational ligand design. We employ density functional and coupled-cluster theory to investigate potential binding motifs and complexation energies of uranyl and oxovanadium metal ions with amidoxime ligands. This information is utilized in the structure-based design of multidentate host architectures that are structurally organized to complex the uranyl ion. Computational modeling of materials and processes for extraction of uranium from seawater provides the essential foundation for the development of next-generation of adsorbent materials with high uranium binding affinity and selectivity.

Advanced Fuels Campaign



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3. ADVANCED FUELS CAMPAIGN

3.1 Overview

J. Carmack, Idaho National Laboratory

Mission

The mission of the Advanced Fuels Campaign (AFC) is to perform research, development, and demonstration (RD&D) activities for advanced fuel forms (including cladding) to enhance the performance and safety of the nation's current and future reactors; enhance proliferation resistance of nuclear fuel; effectively utilize nuclear energy resources; and address the longer-term waste management challenges. This includes development of a state-of-the art RD&D infrastructure to support the development of advanced fuel systems using a "goal-oriented science-based approach."

Objectives

AFC has been given the responsibility to develop advanced nuclear fuel technologies for the Fuel Cycle R&D (FCRD) program. The current focus is on the following (see Figure 4):

- Advanced Light Water Reactor (LWR) Fuels with enhanced accident tolerance, improved performance, and increased utilization.
- Advanced Reactor Fuels with emphasis on actinide transmutation with enhanced proliferation resistance, resource utilization, and waste minimization in future fuel cycles.
- **Capability Development** in tools and techniques, such as advanced in-pile instrumentation, characterization, postirradiation examination (PIE) and separate effects testing to generate data for advanced modeling and simulation activities.
- Advanced Fuel Performance Modeling and Simulation is essential to the development of advanced fuel systems. AFC interfaces with the Nuclear Energy Advanced Modeling and Simulation (NEAMS) program to develop multi-scale, multi-physics fuel performance codes.



Advanced Fuels Campaign





Challenges

- Major Increase in Fuel Burnup, Performance, and Utilization over the Current Technologies. An increase in fuel burnup is desired for all the fuel cycle options. However, the quantitative goals for burnup depend on the reactor type and, more importantly, selected fuel cycle options. In some cases, there are practical and economic limitations to burnup beyond the fuel cycle efficiency and technology limitations. Burnup in once-through cycles is limited by the initial enrichment constraints and cladding material properties. Burnup for fuels under full recycle may be limited by reactor physics, storage, and/or disposal constraints after the discharge of spent fuel. Another important consideration in increasing the burnup is to ensure zero-failure, a standard for which industry strives at its current, moderate burnups. Quantitative limits for the burnup grand-challenge under various fuel cycle scenarios will be developed as the program progresses and fuel cycle scenarios are defined.
- Low-loss Fuel Fabrication Processes. The challenge for fabrication is to substantially lower the irretrievable losses from the current levels, which are typically on the order of 1%. This requires the development of cleaner and more efficient fabrication processes without imposing an economic penalty on fuel fabrication. The objective is to generate less waste during the fabrication process and increase resource utilization. Such improved fabrication processes may also contribute to increased safety of plants and enhance safeguards and materials accounting.
- **LWR Fuels with Increased Performance and Enhanced Accident Tolerance.** Improvements will be measured by increased margin to fuel failure, increased response time during an accident to prevent severe damage to the core, and reduced hydrogen generation when the core is uncovered and the fuels and cladding are in contact with steam.

Major R&D Activities

The following are the major RD&D activities for next-generation LWR fuels, adjusted to the FY-15 budget and program direction.

- International Coordination and Collaboration. Bilateral agreements are supported with France, Japan, the European Union, the Republic of Korea, and China. The emphasis is on advanced LWR fuels with enhanced accident performance; metallic fuel development; irradiation testing and data analyses; and development of characterization and PIE techniques. Two joint irradiation projects have been developed with the Halden Reactor Project (Norway) in advanced LWR fuels: (1) an instrumentation qualification test in the Advanced Test Reactor (ATR) in advance of the Accident-Tolerant Fuel (ATF)-2 loop test and a (2) creep test of FeCrAl and SiC samples in the Halden reactor. Activities are supported under four multinational agreements and arrangements, the Gen IV Sodium Fast Reactor project arrangement, the Organization for Economic Cooperation and Development-Nuclear Energy Agency (OECD-NEA), EURATOM, and coordinated research projects under the International Atomic Energy Agency (IAEA). These multinational agreements allow the review and coordination of fuel development activities worldwide.
- Advanced LWR Fuels with Enhanced Accident Tolerance. Fundamental RD&D activities continued on ATF concepts; screening attributes and metrics were established; and research activities were coordinated between DOE laboratories, industry Funding Opportunity Announcement (FOA) teams, university Integrated Research Project (IRP) teams and Nuclear

Energy University Programs (NEUP). Critical testing capability is required for ATF development. This includes high-temperature steam oxidation testing (recently developed specifically for ATF), material property measurements, and irradiation testing.

- Advanced Reactor Fuels. Primary RD&D areas included advanced fabrication technology development; fabrication and characterization of minor actinide- and lanthanide-bearing fuels; fundamental property measurements and Fuel Cladding Chemical Interaction (FCCI) testing; and irradiation performance testing.
- **Capability Development.** Primary RD&D areas included advanced modeling and simulation (M&S) of fuel performance and fabrication processes; characterization technique development; and unique in-pile and out-of-pile material property measurements. Experimental transient testing capabilities were established for the Transient Reactor Test (TREAT) facility. Three principal test modes are currently under development; a static capsule test capability, a water test loop, and a sodium test loop.

Fiscal Year-End 2015 Funding (Includes Carryover)

Advanced Fuels Campaign			
Major Activities	FY-15 Funding		
AFC Management & International Collaboration	\$ 2,055,000		
Advanced LWR Fuels	\$ 27,038,431		
Industry ATF Project Authorization	\$ 8,000,000		
Advanced Reactor Fuels	\$ 10,902,105		
Capability Development	\$ 13,019,651		
Total	\$ 61,015,187		

Major Accomplishments

Accident-Tolerant LWR Fuel

Accident-tolerant LWR fuel research made significant progress in FY-15 through insertion of fuel experiments from each industry-led FOA project (Westinghouse, AREVA, and General Electric [GE]) in the ATF-1 irradiation experiment in ATR. These projects are integrated into DOE's laboratory funded research, development, and infrastructure activities in AFC with the university NEUPs and university-led IRPs. In addition, ATF fabrication support resources were established and demonstrated at ORNL, INL, and Los Alamos National Laboratory (LANL). Research quantities of ATF compositions were fabricated, demonstrating the capability to support industry FOA fuel fabrication needs as well as the capability to fabricate novel ATF fuel compositions. Design of the initial irradiation of ATF concepts, designated as the ATF-1 test series, was completed. The initial ATF-1 experiments were inserted in ATR in FY-15. The FOA fuel concepts will continue irradiation in ATR over the next two years.

Advanced Reactor Fuels

The Advanced Reactor Fuels area achieved major objectives in FY-15 in the areas of fabrication development, characterization of actinide-bearing fuels, irradiation testing, and postirradiation examination of metallic fuel experiments. Last year, the FUTURIX-FTA experiment was returned from the Phenix fast reactor in France, and in FY-15 postirradiation examination of these fuels was initiated.

The FUTURIX-FTA fuel experiment contained four pins of transuranic-bearing metallic and nitride fuels. The nondestructive postirradiation examination of the FUTURIX-FTA fuel pins was completed, and the destructive evaluation will be completed in FY-16. The campaign has pursued the investigation of fuel alloy additions to immobilize the lanthanide fission products, sodium-free annular fuel concepts, and cladding coatings and liners for mitigation of fuel-cladding chemical interaction. This year several compositions of annular fuel having a variety of additives continued irradiation in ATR. Also, direct casting of metallic fuel into a zirconium liner was demonstrated. A new fresh fuel characterization glovebox began operations at INL with the capability of measuring the thermal properties of a variety of nuclear fuel systems, including fuels containing significant quantities of transuranic materials. In the area of cladding development, a major goal is to develop fast reactor claddings capable of withstanding high irradiation doses (>250 dpa). In collaboration with industry and international partners the program has successfully fabricated candidate ferritic-martensitic materials with improved material properties and has irradiated selected cladding samples to high doses. A first set of material samples irradiated in the BOR-60 reactor have been returned to the United States for materials property characterization.

Capability Development

Activities pursued under the capabilities development area include new techniques and equipment to study irradiated nuclear fuels, as well as new and innovative capabilities for nondestructive evaluation and in-pile experiment measurements. In FY-15, the program was a major contributor to the acquisition of a Talos Transmission Electron Microscope (TEM), a state of the art capability now available at ORNL to study advanced nuclear fuels and materials. A team of LANL and Brookhaven National Laboratory (BNL) scientists performed the first in-situ, time resolved experiment at the new Office of Science National Spallation Light Source-II at BNL, conducting a study on the fundamental behavior of the field assisted sintering process on uranium dioxide. Finally, a new activity has been initiated in to develop, design, and build transient experiment capabilities in TREAT. The conceptual and preliminary designs were completed for a static capsule, a flowing water loop, and a flowing sodium loop to support future transient testing needs of AFC.

3.2 Design of FeCrAl alloys with Enhanced Accident Tolerance through Targeted Irradiation Programs

K. G. Field, S. A. Briggs, P. Edmondson, R. Howard, Y. Yamamoto, K. A. Terrani, Oak Ridge National Laboratory

FeCrAl alloys with minor Y additions are an attractive alloy class for near-term deployment as a cladding material with enhanced accident tolerance for LWR applications. FeCrAl alloys with minor Y additions have shown excellent high-temperature steam oxidation up to 1450°C.¹ This performance results in the potential for increased safety margins by limiting the heat and hydrogen production during high-temperature steam exposures during LWR accident scenarios. Current R&D efforts have focused on optimizing the FeCrAl alloy class through microstructural and chemical control to develop a cladding material that retains the known high-temperature steam oxidation resistance while still having properties conducive to normal operation in LWRs. Such properties include high-temperature mechanical performance, corrosion resistance, formability, thermal stability, and irradiation performance, to name a few.

Currently, a systematic effort is underway to determine the radiation tolerance of these optimized, advanced FeCrAl alloys by using targeted irradiation programs. Several different materials research reactors, including ATR, the High Flux Isotope Reactor (HFIR), and the Halden Reactor, and ion beam facilities including the use of the in-situ irradiation capabilities at the Intermediate Voltage Electron Microscopy (IVEM)-Tandem facility are being used to assess a range of phenomena during irradiation including radiation-hardening and embrittlement, irradiation creep, swelling, and fuel-clad chemical interactions (FCCI). Extensive PIE, including advanced characterization techniques is ongoing or planned to access these phenomena in FeCrAl alloys and determine their controlling mechanism(s).

A quick example of the type of information being gained from this extensive program is shown in Figure 5, which is the result of conducting atom probe tomography (APT) to provide atom-by-atom reconstructions of small volumes. The figure shows the formation of small Cr-rich clusters, otherwise known as α' , formed after irradiation in the HFIR to 7 dpa at 320°C in four different model FeCrAl alloys. The image shows a composition dependence on the size and number density of the Cr-rich α' phase in these alloys and not shown is the change in average cluster composition with bulk-alloy composition. Analysis derived from results such as Figure 5 can be linked to the mechanical properties and material performance using known structure-property relationships allowing for further alloy optimization efforts. For example, the information in Figure 1 has provided insight on the mechanisms for radiation-hardening where Cr-rich α' is linked to significant increases (>50 MPa) in the yield strength using the dispersed barrier hardening model.² This relationship between precipitation, alloy composition, and hardening resulted in an effort to reduce the bulk Cr content of nuclear grade FeCrAl alloys at or below 13 wt.% Cr.

These targeted irradiation programs, with output such as the one briefly described here, are proving critical towards the assessment of the FeCrAl alloy class as an accident-tolerant fuel (ATF) cladding. The foundational knowledge gained from these efforts will assist in providing a robust application for a licensing amendment request (LAR) to FeCrAl alloys in a lead test rod (LTR) insertion into a commercial LWR.



Figure 5. 3D reconstruction using atom probe tomography (APT) of different FeCrAl alloys showing the population of α ' precipitates (magenta clusters) using a concentration iso-surface (30 at.% Cr) irradiated to 7 dpa at 320°C in the HFIR. Black dots represent 2%.

References

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3.3 High-Density Ceramic Fuels for LWR Accident-Tolerant Applications

A. T. Nelson, J. T. White, D. D. Byler, J. T. Dunwoody, K. J. McClellan, Los Alamos National Laboratory

A wide range of accident-tolerant fuel (ATF) concepts are under investigation by the Advanced Fuels Campaign. The fuel cladding is the logical focus of many of these efforts, as improvements in oxidation resistance provide the most direct means of extending coping time during a loss of cooling accident (LOCA). However, modification or replacement of zirconium cladding is unlikely to provide any economic incentive to the fuel vendors and utilities whose sustained investment will determine the success of any proposed ATF. The economics of plant operation as dictated by the fuel-clad system are directly linked to U-235 content and neutronics. If enrichment is capped at existing licensed values, the only means to maintain or exceed current reactor heat output and cycle lengths is to consider fuel materials possessing uranium contents superior to uranium dioxide (UO_2).

Several fuel materials of superior uranium density exist, but all contain deficiencies when evaluated for service in LWR environments. For example, uranium mononitride (UN) exhibits generally favorable performance as a nuclear fuel, but rapidly pulverizes if exposed to water. It must be protected from this degradation if incorporated as a constituent of an ATF concept. The conventional solution is encapsulation in an inert matrix (e.g. SiC). However, this approach returns the fuel to a position where it will be challenged to meet existing reactor performance benchmarks. The novel solution currently under investigation by AFC researchers is development of fissile ceramic composite fuels. Instead of an inert matrix, a uranium-bearing material is chosen to recover some of the lost fissile content and at the same time provide ATF performance benefits to the fuel system. Such an approach offers an economic incentive to the nuclear industry and satisfies primary ATF metrics through improved fuel properties.

The first generation of fissile ceramic composite fuel architectures are centered on use of UN to provide high uranium density and favorable fuel properties augmented by uranium silicide compounds incorporated as a secondary phase at volume fractions ranging from 10 to 35%. Uranium silicide compounds, specifically U₃Si₂ and U₃Si₅, offer significant thermal conductivity improvements over UO₂ when incorporated into a UN composite. This will reduce fuel centerline temperatures thereby limiting fuel restructuring, thermal stresses, and fission product mobility. These benefits combine to provide a monolithic fuel pellet hypothesized to greatly restrict fission product transport to any free surface, effectively retarding fission product release during cladding failure.

Initial experimental assessment of the critical properties of uranium-silicide compounds as well as UN composites has been completed to provide fundamental thermophysical properties to the modeling and simulation communities. Figure 6 highlights the properties of four uranium silicide compounds in comparison to both UN and UO₂. Despite promising results obtained to date, the in-pile performance of these systems must be determined. A series of ATF irradiations are ongoing at INL's ATR. Once the composite microstructure has been optimized, a series of enriched test pellet fabrications have been completed for both UN-15 vol% U₃Si₅ and UN-30 vol% U₃Si₂ compositions (Figure 7). Examination following insertion of these pellets into upcoming ATR cycles will provide initial indications of the viability of these fissile ceramic composite concepts.



Figure 6. Thermal conductivity of uranium-silicide compounds in comparison to uranium mononitride and uranium dioxide. Uranium silicide data presented in this figure was collected through a series of experimental studies under the AFC program.



Figure 7. Optical micrograph *(left)* of UN/U₃Si₂ composite showing optimized microstructure where the U₃Si₂ phase (light) surrounds the UN (dark) to protect the UN from washout during normal operation. This fabrication pathway has been used to produce high-density (95% theoretical) enriched test pellets *(right)* for upcoming test irradiation at ATR.

3.4 U₃Si₂ Fabrication and Testing for Implementation into the BISON Fuel Performance Code

K. Metzger, T. W. Knight, X. Huang, University of South Carolina; J. Harp, M. Meyer, Idaho National Laboratory

This three-year research program includes the fabrication, testing, and modeling of a high uranium density, advanced nuclear fuel (U_3Si_2) . This work is designed to deliver key research data on creep that is complementary to earlier and ongoing programs focused on advanced fuels for greater safety and economics (i.e. power uprates) and supports DOE programs in LWR Sustainability. The project has made significant progress in the following areas:

- The procedure for fabricating single-phase U₃Si₂ specimens for creep testing has been developed and the production of a series of creep test specimens in ongoing.
- Impulse excitation shakedown testing has been performed on a surrogate material and tested over a range of temperatures.
- U₃Si₂ pellet samples were fabricated using differing feedstock particle size distribution and sintering temperature. These pellets were fully characterized to examine the as fabricated microstructure (Figure 8).
- Near term creep data will be obtained from compression creep testing; the test rig design is complete and fabrication of the testing equipment will be complete by the end of 2015.
- Thermal and swelling material models for U₃Si₂ have been implemented in the BISON fuel performance code.
- A series of simulations with Zircaloy and silicon carbide claddings have been run in BISON using the current material models implemented for U₃Si₂.
- Compression creep testing will provide the strain rate data required to develop a thermal creep model for U₃Si₂. This creep model will then be implemented to BISON, which will provide a more robust model for the behavior of U₃Si₂ in a LWR environment.





Figure 8. U₃Si₂ pellet samples were fabricated using differing feedstock particle size distribution and sintering temperature. These pellets were fully characterized to examine the as fabricated microstructure.

3.5 Recent AFC Experimental Postirradiation Examination Results

J. Harp, Idaho National Laboratory

Baseline PIE was recently completed on several tests from the AFC transmutation fuels series of experiments. Transmutation fuels for fast spectrum reactors seek to transmute a wide array of toxic radioactive species (particularly actinides such as americium). Successful demonstration of transmutation fuels could reduce fuel cycle disposal costs by reducing repository long-term radiotoxicity and heat load. These experiments included fuels that were mixed oxides with and without minor actinides (AFC-2D), ternary U-Pu-Zr metallics with and without minor actinides (AFC-2E), and metallic fuels of various forms and compositions to explore high-burnup concepts (AFC-3A and AFC-3B). Baseline PIE includes visual examination, dimensional inspection, neutron radiography, gamma spectrometry, fission gas release analysis, optical microscopy, microhardness testing, and chemical burnup analysis. The observation of historical behavior in AFC tests is important for linking true fast reactor tests to the quasifast reactor conditions achieved in current Advanced Test Reactor (ATR) testing.

Additionally it is important to discover if the addition of minor actinides to create transmutation fuels changes fuel performance compared to historically tested U and U-Pu based fuels. AFC-2D focused on the fuel performance of different mixed oxides (MOX) and minor actinide MOX fuels along with variations in their oxygen-to-metal (O/M) ratios. FCCI in oxide fuel with steel cladding has been well studied and is driven by the oxygen potential and volatile fission product content at the fuel-cladding gap. Optical microscopy examination of AFC-2D confirmed this behavior where there was significantly more FCCI in the higher O/M ratio Rodlet 3 than was present in the lower O/M ratio Rodlet 5. The onset of FCCI is an important factor that limits the irradiation performance and functional lifetime of transmutation fuels. AFC-2E was designed to confirm that fuel behavior observed in EBR-II (a historic sodium cooled fast reactor) testing of U-Pu-Zr ternary fuel was also observed in ATR testing of the same compositions. Some rodlets appear to have been irradiated at conditions very similar to margin tests performed in EBR-II (assembly X-447A). The fuel microstructure observed in these tests is almost identical to the microstructure observed in AFC-2E Rodlet 1.

Overall AFC-3A and AFC-3B performed as expected and comparisons to similar historic fuel compositions can be made. The optical microscopy cross-section from AFC-3A Rodlet 5 (Figure 9) is a good example of possible Zr constituent redistribution historically seen in U-10 wt.% Zr fuels. This fuel also contained 1 wt. % Pd to act as a stabilizing agent for rare earth fission products to help mitigate FCCI, and no FCCI was observed in this sample. Future work on samples from all these experiments will include electron microscopy examination to confirm suspected constituent redistribution, confirm observed precipitate composition, and investigate the composition of different FCCI layers and their interaction with the cladding.

Baseline nondestructive PIE was completed on the four FUTURIX-FTA pins that were irradiated at the Phénix fast reactor in France. All exams indicate good fuel performance



Figure 9. Optical microscopy of AFC-3A Rodlet 5 possibly exhibiting two zone Zr redistribution behavior typically observed in historic irradiations of U-10Zr fuel.

from the four pins. Most notably in the neutron radiography, the metallic fuel pins indicate intact Na plugs and no indications of excessive FCCI, and the nitride pins indicate pellets that are largely intact after irradiation and shipment. PIE results from FUTURIX-FTA pins will be compared to PIE results from matching pins irradiated in ATR.

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Nuclear Fuels Storage and Transportation Project



Idaho National Laboratory November 3-5, 2015 This page left intentionally blank

4. NUCLEAR FUELS STORAGE AND TRANSPORTATION PLANNING PROJECT

4.1 Overview

M. Nutt, Argonne National Laboratory

Mission

DOE-NE is to lay the groundwork for implementing interim storage, including associated transportation, per the Administration's *Strategy for the Management and Disposal of Used*¹ *Nuclear Fuel and High-Level Radioactive Waste,* and to develop a foundation for a new nuclear waste management organization. The organization within NE that is responsible for this planning is the Office of Nuclear Fuels Storage and Transportation (NFST). The technical work to support this planning effort is being conducted by DOE's national laboratories.

Goals and Objectives

DOE's overarching goal is to develop options for decision-makers on the design of an integrated waste management system. The goal is to develop an integrated plan to (1) implement interim storage; (2) improve the overall integration of storage as a planned part of the waste management system; (3) prepare for the large-scale transportation of SNF and high-level waste (HLW), with an initial focus on removing SNF from the shutdown reactor sites; and (4) develop the foundational information, resources, and capabilities needed to support future implementation decisions and actions.

Near-term objectives are to (1) develop and maintain an integrated plan to accomplish the Administration's Strategy goals, (2) improve integration of storage as a planned part of the waste management system, (including evaluating standardization of dry cask storage systems), (3) develop and evaluate design options for an integrated waste management system, (4) develop and apply systems analyses to provide quantitative estimates of system impacts of utility actions' impacts on the system and inform future decisions, (5) prepare for the large-scale transportation of SNF and HLW, (with an initial focus on removing SNF from the shutdown reactor sites), and (6) establish and maintain a unified, integrated SNF database and analysis system to characterize the input to the waste management system.

Major Activities

NFST activities are divided into four major topical areas (1) Consent-Based Siting, (2) Storage, (3) Transportation, and (4) Strategic Crosscuts. Major activities in each area are described below.

Consent-Based Siting activities continued to lay the groundwork for the development of consent based siting process for nuclear waste management facilities that reflects input received from interested parties at the community, state, and tribal level. This included analysis of an annual nationwide survey of public preferences related to national energy sources, awareness of the nuclear fuel cycle, and support for building interim storage facilities. To ensure that future siting efforts are informed by past experience, the Siting Experience Database (SED) continued to be populated, providing an archive of documented experience on nuclear waste facility siting efforts (accessible at www.curie.ornl.gov).

¹ Note "used" and "spent" are used interchangeably by various organizations to refer to nuclear fuel that has been irradiated in a nuclear reactor.

Storage activities included identifying and evaluating a suite of generic modular design alternatives for the receipt and storage of SNF in canisters at an interim storage facility (ISF), with input from industry contractors. Preparations are underway to develop a generic (not site-specific) pilot ISF design and an associated Topical Safety Analysis Report through an industry contract. An effort to address interim storage system aging management considerations was initiated, leveraging on prior investigations completed by the UFD R&D Campaign.

Transportation activities included the awarding of a contract to design/analyze and subsequently fabricate prototype American Association of Railroads (AAR) standard S-2043 compliant cask and buffer railcars for future testing. Collaboration with state regional groups and tribal representatives continued in the development of a transportation planning framework and in the conduct of a Nuclear Waste Policy Act (NWPA) Section 180(c) policy implementation exercise to support potential revision to the policy. The Stakeholder Tool for Assessing Radioactive Transportation (START) was enhanced. Information related to site inventory, site conditions, and near-site transportation infrastructure and experience continued to be collected through a visit to the San Onofre nuclear generating station. Initial site-specific de-inventory analyses were begun for three shutdown sites to understand the tasks and interfaces necessary for the complete de-inventory of the reactor sites. Through an industry contract, concepts for reusable transportation casks, including casks capable of being loaded with all assemblies enclosed in damaged fuel cans, were designed and assessed.

Strategic Crosscut activities included project management, coordination, and planning relative to the NFST mission and objectives. An integrated waste management systems analysis was conducted to inform future decisions and provide quantitative estimates of the impacts of utility and federal actions. The development of the Next Generation System Analysis Model that will be more readily capable, sustainable, and maintainable in the future continued. The unified, comprehensive SNF database and integrated analysis system, referred to as the Used Nuclear Fuel Storage, Transportation & Disposal Analysis Resource and Data System (UNF-ST&DARDS) was enhanced to characterize the input to the waste management system and provide a credible, controlled data source for key information. A collaborative SNF document and data access system, the Centralized Used Fuel Resource for Information Exchange (CURIE; curie.ornl.gov) was maintained and enhanced. Opportunities were evaluated for standardization in the waste management system, including conducting a quantitative assessment and comparison of relevant options, and using input provided from an industry contractor relative to generic design and operational studies for small standardized transportation, aging, and disposal systems.

Nuclear Fuels Storage and Transportation Planning Project				
Major Activities	FY-15 Funding			
Strategic Crosscuts	\$16,658,832			
Storage	\$6,659,708			
Transportation	\$12,335,418			
Consent-Based Siting	\$907,489			
Total	\$36,561,477			

Fiscal Year-End 2015 Funding (Includes Carryover)

Major Accomplishments

A contract for Cask and Buffer Railcar Prototype Development was awarded to a team led by AREVA. The contract included design, analysis, and fabrication of the cask and buffer railcars for testing to meet AAR Standard S-2043, "Performance Specification for Trains Used to Carry High-Level Radioactive Material."

An initial Transportation Planning Framework was completed to remove commercial spent nuclear fuel from shutdown reactors. The framework reviewed lessons learned from prior shipping campaigns; described current transportation planning activities and summarized the current situation at shutdown reactor sites; outlined roles and responsibilities of the shipper, states, tribes, local officials, and industry; incorporated best practices from transportation plans; documented shipping campaigns; and discussed technical and institutional activities that need to be completed in order to have a fully operational transportation system.

The cost implications of an ISF in the Waste Management System were evaluated. This

provided an initial evaluation of the cost implications of incorporating an ISF into the waste management system. Specifically, the impacts of the timing of opening an ISF relative to opening a geologic repository were analyzed to understand the potential effects on the total system cost.

Version 2.0 of the Unified SNF Database and Analysis System was deployed. UNF-ST&DARDS is foundational resource for tracking SNF from reactor power production through ultimate disposition. It provides a comprehensive, controlled source of technical data, referred to as the "Unified Database," for waste management system analysis and evaluation tools, as well as fuel cycle system analyses and safeguards and security studies. UNF-ST&DARDS provides access to key technical data and analysis capabilities to characterize the SNF inventory and includes enhanced capabilities to assess safety, risks, and uncertainties throughout the waste management system.

Capabilities of the Execution Strategy Analysis (ESA) Model were enhanced. The ESA Model is being used to evaluate alternative strategies for deploying an ISF and the associated transportation infrastructure per the Administration's *Strategy*. It allows for ongoing performance assessment of the evolving strategy and project plan that takes into account significant assumptions, risks, and uncertainties throughout the project lifecycle.

The Next-Generation Waste Management Systems Analysis Model (NGSAM) was enhanced to represent all commercial nuclear reactor sites. NGSAM is more capable, usable, and maintainable than the current integrated waste management system analysis tools. The agent-based simulation tool allows for the evaluation of a broad range of potential future waste management system architectures that include both interim storage and ultimate disposition to provide DOE and other stakeholders with information regarding various potential integrated waste management system alternatives.

The Intermediate Standardization Assessment was completed. It documents further evaluation of integrating standardized canisters into the nuclear waste management system, an intermediate step in quantitative assessment of standardization. The report broadens the potential alternatives evaluated, highlights preliminary observations, identifies needed information, and guides future evaluation work.

Version 1.2 of Stakeholder Tool for Assessing Radioactive Transportation (START) was

released. START is a transportation decision-support tool enabling users to represent and evaluate a wide range of transportation scenarios to assist with stakeholder communications and information sharing. It provides route identification process and preliminary routing analysis; NWPA section 180(c) policy development and needs assessments; data collection near shutdown sites; and waste management systems integration.

Industry contracts related to Storage, Transportation, and Standardization were completed.

The following three significant task orders were completed by advisory and assistance services contractor teams and were executed with coordination by the federal staff and laboratory support.

- "Generic Design Alternatives for Dry Storage of Used Nuclear Fuel" by Chicago Bridge and Iron (CB&I) more fully developed and documented generic design alternatives for receipt and storage of SNF and greater-than-Class-C (GTCC) waste contained in dual-purpose canisters.
- "Generic Design for Small Size Standard Transportation, Aging, and Disposal Canister System" by EnergySolutions developed a generic design of a small capacity (4PWR/9BWR) standardized transportation, aging, and disposal (STAD) canister.
- "Operational Requirements for Standardized Dry Fuel Canister Systems," by EnergySolutions developed solutions to address operational impacts of using a standardized canister at utility sites with a smaller capacity than conventional, dual-purpose canisters to move the required number of SNF assemblies from the SNF pool to onsite dry storage in a designated time frame.

4.2 NFST Modeling Tools

J. Jarrell, Oak Ridge National Laboratory

Introduction and Objectives

NFST is developing foundational capabilities and tools to support and inform the future deployment of a consolidated interim storage and large-scale transportation of SNF. These tools include Used Nuclear Fuel Storage, Transportation, & Disposal Analysis Resource and Data System (UNF-ST&DARDS), Stakeholder Tool for Assessing Radioactive Transportation (START), Transportation and Storage Logistics (TSL), Next-Generation System Analysis Model (NGSAM), Execution Strategy Analysis (ESA), Multi-Objective Evaluation Framework (MOEF), and the Centralized Used Fuel Resource for Information Exchange (CURIE).

Overview

These tools work in an integrated fashion to provide a flexible, adaptable evaluation tool. Additionally, each tool has unique capabilities and can be used individually to answer specific questions related to spent fuel management.

The UNF-ST&DARDS tool provides a consistent, traceable source of data related to SNF and the ability to characterize the SNF and related system (e.g., casks, canisters, etc.) at any time after discharge. The UNF-ST&DARDS' Unified Database provides a consolidated archive of SNF and SNF systems. Along with the SNF data, UNF-ST&DARDS can perform automated SNF depletion calculations and canister/cask shielding, thermal (Figure 10), criticality and containment calculations for assessing issues and uncertainties related to the extended storage and transportability of loaded canisters.



Figure 10. Example UNF-ST&DARDS analysis result showing temperatures inside a SNF canister.

The START tool is being developed to evaluate transportation routing options and identify training needs to support emergency preparedness. The tool is a web-based application that uses geographic information systems (GIS) technology to represent transportation network operations as well as proximate features. START is being used for a variety of transportation planning activities, including: (1) route identification and analysis, (2) identifying emergency preparedness training needs, (3) site infrastructure assessment, and (4) waste management systems analysis.

The systems architecture analysis tools provide system-wide logistics and cost information (Figure 11). The legacy simulation tool, TSL, provides a framework for evaluating the entire system for managing commercial SNF, DOE-owned SNF, and DOE-owned high-level nuclear waste. It models the discharge of SNF from reactors, pool inventory changes, fuel handling operations (e.g., receipt of SNF at the ISF, repackaging of SNF into canisters or bare fuel casks), decay heat of SNF assemblies, and shipment requirements of the waste management system. A new tool, NGSAM, is being developed for NFST analysts to perform integrated waste management system analysis that better represent waste handling processes than existing legacy tools. This flexible, agent-based tool will simulate a broad range of

scenarios to provide quantitative information about a broad range of system alternatives and factors, including repository emplacement capability, thermal constraints, repackaging needs, storage and transportation alternatives, and impacts on utility options.



storage at reactor sites for different SNF acceptance rates

The ESA tool supports ongoing performance assessment of the evolving project plan/strategy that takes into account significant assumptions, risks, and uncertainties throughout the project lifecycle. It has been developed as a dynamic simulation tool that explicitly models and assesses the impacts of uncertainties (activity durations and costs), constraints (policy, legislation, regulatory), risks (technical, nontechnical), and opportunities (Figure 12).



Figure 12. ESA analysis of the probability of not exceeding Pilot ISF costs for different implementation strategies.

MOEF is a set of capabilities, methods, processes, and tools that provide a means to evaluate alternative scenarios and system architectures where there are multiple conflicting objectives and differing stakeholder perspectives that must be taken into account. It is suited for integrating diverse types of information, including the results from system analysis models, cost and benefit studies, empirical data, and expert judgment, into clear and well-defined measures of merit for the alternatives being considered.

The CURIE website is a national resource accessible to industry, vendor, federal, laboratory partners, and other stakeholders. CURIE includes a calendar, the SNF image gallery, NFST featured documents, and external links to databases and websites.

Accomplishments

Over the past fiscal year, each tool has made significant progress and produced a number of high-quality reports and analysis. A select few of these accomplishments are described below.

Version 2 of the UNF-ST&DARDS tool was distributed on September 30, 2015. In this version, a number of improvements were introduced including (1) an updated Unified Database with a prereleased version of the GC-859 fuel inventory and corresponding decay heat calculations, (2) enhanced capabilities and frameworks, including as-loaded containment analysis, several new templates for criticality, shielding, and thermal analyses, and self-protecting calculations of discharged assemblies, and (3) a new automated report generation capability.

The FY-15 Waste Management System Architecture Analysis Report was submitted on

September 30, 2015. In the report, system architecture analysis was performed on the waste management system alternatives. In particular, this report focused on (1) alternative allocation strategies, (2) thermal management, (3) alternative acceptance rates and dates, and (4) alternative storage and receipt options.

The Intermediate Standardized Canister Evaluation was submitted on September 30, 2015.

In this report, system architecture tools were used with a focus on shipment of bare fuel to an ISF and loading standardized canisters for storage and subsequent transportation and disposal from that point forward. Scenarios were developed and analyzed that focused on (1) bare SNF transported from reactors to an ISF in reusable bare SNF casks with deployment of a standardized canister system deferred to the ISF, (2) alternative acceptance priority rankings based on conclusions from previous systems architecture studies, and (3) the impact of having to accommodate spent fuel in canisters of different sizes based on different repository geologies.

The Multi-Objective Evaluation of the FY-15 Canister Standardization Scenarios was submitted on September 30, 2015. In this evaluation, MOEF was used to better understand and quantify the relative value of incorporating SNF canister standardization into the waste management system from different stakeholder perspectives.

4.3 NFST Integrated Waste Management System Architecture Analysis

C. Trail, Argonne National Laboratory

Introduction and Objectives

NFST is developing foundational capabilities to support the application of system engineering and decision analysis principles to inform future decisions regarding the deployment of an integrated waste management system. To that end, NFST's Systems Architecture Analysis effort conducts systems analyses to evaluate an integrated approach to transportation, storage, and disposal of SNF in the waste management system. This effort evaluates the implications of the current strategy for at-reactor storage of SNF in large dry storage systems as they relate to the subsequent potential disposal. Additionally, the analysis evaluates alternative strategies and approaches for managing the SNF to identify potential benefits to the waste management system.

Overview

The FY-15 Systems Architecture Analysis effort within NFST built upon the system architecture work from previous years. Industry engagement and data collection efforts further inform the analysis, and include improvements in model accuracy regarding cost and operational logistics.

The TSL simulation tool is used to simulate all scenarios in this report including the reference scenario. The TSL simulation tool is a framework for evaluating the entire system for managing commercial SNF, DOE-owned SNF, and DOE-owned high-level nuclear waste.

Accomplishments

Several key insights were gained as a result of the FY-15 systems architecture analysis. A subset of the results is summarized below:

A range of strategies for the allocation² of SNF for shipment from commercial nuclear reactor sites were evaluated. These alternatives consider a range of acceptance rates (1,500 Metric Ton Uranium per year [MTU/yr] to 6000 MTU/yr), and allocation strategy alternatives to the oldest fuel first (OFF) allocation method defined by the standard contract. Additionally alternative methods for shipping SNF directly from the spent fuel pools at reactor sites are considered dual-purpose canisters (DPCs), or reusable bare fuel transportation casks. Results indicate that larger acceptance rates, alternative allocation strategies, and combinations of the two can clear reactor sites sooner compared to a 3,000 MTU/yr acceptance rate with an OFF allocation strategy. Using a combination of accelerated acceptance and modified allocations is potentially the most efficient approach for clearing SNF from reactor sites, reducing overall at-reactor costs of SNF management.

^{1.} This is a technical report that does not take into account the contractual limitations under the Standard Contract. Under the provisions of the Standard Contract, DOE does not consider spent fuel in canisters to be an acceptable waste form, absent a mutually agreed-to contract modification. To ensure the ability to transfer spent fuel to the government under the Standard Contract, individual spent fuel assemblies must be retrievable for packaging into DOE-supplied transportation casks.

The number of site-years with SNF remaining on site after the last reactor has shut down, shown in Figure 13, is used as a metric to evaluate approaches for clearing SNF from reactor sites. This metric illustrates the reductions in post-shutdown site-years that could potentially be achieved by for different SNF acceptance strategies.



Notes: OFF – Oldest Fuel First SNF Allocation prioritization; DS-SD – site-specific allocation strategy that aims to minimize SNF transfer to dry storage and remove SNF from shutdown sites; Canisters Only – all SNF is transported from the reactor sites in DPCs; Canisters with Bare Fuel – individual fuel assemblies from spent fuel pools transported in reusable transportation casks.



The impacts of alternative SNF acceptance approaches on operations and costs at the interim storage facility (ISF) were also evaluated. Different SNF acceptance strategies impact the configuration and operation of an ISF. Accelerated acceptance rates would increase the size of an ISF because additional storage capacity and fuel processing facilities would be needed. If individual SNF assemblies are received at the ISF in reusable bare fuel transportation casks, a large number of individual assemblies would have to be processed for storage. Those assemblies could either be placed into DPCs for storage or loaded into a large bare fuel assembly storage facility (such as a spent fuel pool). Notably, pool storage of SNF at the ISF was not found to be cost prohibitive in contrast to findings from previous studies. These new findings are the result of updated information provided by industry engagement, but design concepts and detailed cost information is still required for all aspects of ISF storage configurations.

The rate of SNF acceptance from reactor sites and when a repository becomes operational, significantly impacts the ISF costs and operational requirements. Results demonstrate that delays of 10 years in the repository start date increase the peak ISF inventory by 30% to 40% depending on the SNF acceptance rate from reactor sites to the ISF. Imposing the repository emplacement thermal limits on ISF-to-MGR (monitored geological repository) shipments of SNF impacts the overall ISF inventory. These constraints can add decades to the amount of time that the ISF must remain open; 108 years of ISF operation for the most restrictive repository media evaluated (clay/granite) compared with 72 years of ISF operation for the least restrictive repository media evaluated (open media).

Path Forward

NFST's Systems Architecture Analysis continues to support the application of system engineering and decision analysis principles to inform future decisions regarding the deployment of an integrated waste management system. The focus of this effort in FY-16 is the partial integration of the next generation of system analysis tools still in development. This integration involves feature requests from system analysts to software developers, benchmarking the new software against legacy tools, as well as incorporating already completed functionality into systems analysis studies scheduled to be conducted later in the year.

4.4 Transportation Hardware/Railcar

P. Schwab, Department of Energy

Introduction and Objectives

NFST was established to lay the groundwork for implementing interim storage, including the associated transportation of SNF and HLW. The transportation of SNF and HLW will be primarily performed by rail and will require the design of specialized cask, buffer, and escort railcars to be approved by the Association of American Railroads (AAR) under AAR Standard S-2043. AAR Standard S-2043 defines the performance specification for trains used to carry SNF and HLW (collectively referred to by the AAR as high-level radioactive material (HLRM). The planned approach to design new cask and buffer railcars that meet AAR Standard S-2043 is described.

Overview

The first steps in implementing the planned approach were to prepare a request for proposal (RFP), negotiate a contract, and award a contract for the design and prototype fabrication of cask and buffer railcars to transport HLRM.

A significant effort was expended by NFST over 18 months (March 2014 to August 2015) to document the history and requirements of the AAR standard, develop a railcar systems requirements document, and conduct procurement activities from planning through contract award. As a result of these efforts, DOE awarded a contract on August 21, 2015 for the design and associated analysis and prototype fabrication of cask and buffer railcars. The railcar design contract was awarded to AREVA Federal Services (AFS). The end result of this effort will be development of the final cask and buffer railcar designs, including associated analysis, and fabricated railcar prototypes ready for testing. The new cask railcars have been named *ATLAS*.

The ATLAS Railcar Design Project has three phases that are being performed under a single multi-year firm-fixed-price (FFP) contract. Phase 1 is Mobilization and Conceptual Design, Phase 2 is Preliminary Design, and Phase 3 is Prototype Fabrication and Delivery. The summary-level project schedule is shown in Figure 14.

The schedule does not include any hold points between phases, so the contractor does not need approval from DOE to start work on any of the phases. The scheduled completion dates for Phases 1, 2, and 3 are August 2016, March 2017, and March 2019, respectively. Note that Phase 2 is scheduled to start in April 2016, which is four months before Phase 1 is scheduled to be finished.

Tasks	2014	2015	2016	2017	2018	2019	2020	2021	2022
Issue RFI & Sources Sought Notice	4/28/14								
RFI/SSN Responses Due to DOE	7/2/14	1							
Issue Presolicitation Notice	-	11/21/14							
Issue RFP		12/8/14							
Preproposal Teleconference		12/22/14							
RFP Responses Due to DOE	•	2/23/15							
Contract Award		8/2	1/15						
Phase 1: Mobilization and Conceptual Design									
Phase 2: Preliminary Design									
Phase 3: Prototype Fabrication and Delivery									
Acquisition of Escort Railcars									
Testing and Approval of Railcars									

Figure 14. Schedule for ATLAS Railcar Design Project.

The major accomplishment in FY-15 was awarding the contract in August for the design, associated analysis, and prototype fabrication of cask and buffer railcars. The railcar design contract was awarded to AFS. The principal subcontractor to AFS is KASGRO Rail, a company with extensive experience in the design and fabrication of heavy-duty railcars. DOE, AFS, and KASGRO Rail held a kickoff meeting on September 10, 2015 to discuss how specific tasks in this contract would be performed.

DOE-ID initiated the 18-month procurement effort by conducting various activities including market research and combined sources sought notice and request for information (RFI). The information obtained from these activities was reviewed and any resulting changes were implemented into the requirements documents. For example, DOE's original statement of work (SOW) required the railcar design to fit into AAR clearance plate B, but based on feedback received from industry, DOE changed the requirement to AAR clearance plate C.

A much larger change was made to the SOW between release of the RFI (April 2014) and the RFP (December 2014). During this time, DOE decided not to include railcar testing and approval in the SOW, so the current contract will not include testing of the railcars or approval by the AAR. Therefore, a follow-on contract will be required to perform this technical scope.

The RFP was posted to the web portal on December 8, 2014, as solicitation number DE-SOL-0006863 for the design, associated analysis, and prototype fabrication of railcars. A preproposal teleconference was conducted on December 22, 2014, that provided an overview of the RFP, a description of the SOW, instructions on responding to the RFP, and answers to questions received by DOE. DOE received the proposals on February 23, 2015 and immediately began the proposal review and evaluation process.

Following the evaluation process, AFS was selected, contract discussions were conducted, and the contract was awarded for design and prototype fabrication. The resulting contract includes a 12-month schedule reduction (compared to AFS's original proposal) and a single, negotiated FFP contract for all three project phases thus avoiding the riskier cost-plus-fixed-fee (CPFF) contract that was anticipated for Phases 2 and 3. Both cost and schedule were reduced in the final FFP contract along with reduced project risk to DOE. The successful negotiations resulted in DOE-ID awarding a FFP contract on August 21, 2015 to AFS.

This acquisition continues to lay the groundwork to implement an integrated nuclear waste management disposition system by preparing for future large-scale transport of HLRM. This acquisition is for railcar designs which will be owned by DOE, as well as prototype fabrication of one cask railcar and two buffer railcars. The end result of this effort will be final cask and buffer railcar designs, associated analysis, and fabricated railcar prototypes ready for testing.

Path Forward

Future activities are envisioned to perform the required testing and to obtain final approval of the cask and buffer railcars from AAR. The effort to design and develop railcar prototypes, conduct the necessary testing, and secure approval of S-2043–compliant railcars is estimated to take 7 years to complete. AAR approval will require extensive full-scale testing of the individual railcars and the complete railcar consist (e.g. cask, buffer and escort cars coupled together as they would be during actual HLRM transport).

In order to have a fully compliant rail consist, AAR approval of an escort railcar will also be necessary. Currently, DOE is evaluating alternatives for development of an escort railcar to meet AAR S-2043 requirements. One alternative would be to use an S-2043 compliant escort railcar that the US Navy is already engaged in developing. DOE is currently working with the US Navy to see if this option will work for both parties.

DOE will have several options to choose from (lease, purchase, etc.) to establish and maintain the necessary railcar fleet for HLRM shipments. Once the design and prototype testing is under way, DOE will decide whether to procure or lease the necessary rolling stock. DOE does not plan to develop any new locomotives but will expect private railroad companies to provide locomotives compatible with the S-2043 compliant railcars.

While a complete transportation system cannot be fully developed until a destination site is known, long lead-time activities necessary for transportation system development such as railcar design and prototype fabrication can be addressed now. NFST is proactively laying the groundwork so that a transportation system capability will be available to ensure safe, secure, efficient movement of SNF from commercial nuclear power reactor sites in a timely manner when a receiving site becomes available.

4.5 Task Orders Completed in FY-15

R. Howard, Oak Ridge National Laboratory

Introduction and Objectives

The Administration released its *Strategy for the Management and Disposal of Used Nuclear Fuel and High-Level Radioactive Waste* in January 2013³. The Strategy includes a phased, adaptive, and consentbased approach to siting, and implementing a comprehensive management and disposal system. These plans include activities to establish one or more ISFs using consent-based siting and to prepare for largescale transport of SNF. NFST completed the following three Task Orders, using industry Advisory and Assistance Contracts related interim storage and transportation. These design concepts provide important information to the waste management systems analysis regarding throughput capacities, operational considerations, and cost estimates.

- *Generic Design Alternatives for Dry Storage of Used Nuclear Fuel-* Chicago Bridge and Iron (CB&I) developed and documented generic design alternatives for receipt and storage of SNF and GTCC waste contained in dual-purpose canisters.
- *Generic Design for Small Size Standard Transportation, Aging, and Disposal Canister System*-Energy*Solutions* developed a generic design of a small capacity (4PWR/9BWR) STAD canister.
- Operational Requirements for Standardized Dry Fuel Canister Systems- EnergySolutions developed solutions to address operational impacts of using a standardized canister at utility sites with a smaller capacity than conventional, dual-purpose canisters.

Task Order 16: Generic Design Alternatives for Dry Storage of Used Nuclear Fuel

All alternatives evaluated were capable of receiving SNF in DPCs, without the capability to open the dry storage canisters or handle bare fuel assemblies. The studies included engineering evaluations, staffing analyses, time and motion studies, and cost/schedule analyses, all based on uniform assumptions that were made and applied consistently to each alternative. Key output variables include total cost, manning requirements, total dose to workers, throughput achieved (i.e., canisters placed in storage per week), and schedule estimates.

Design Study #1 investigated alternative storage systems for commercial DPCs. The following alternatives were evaluated for storage of DPCs at the Pilot ISF and the Expanded ISF:

- Commercial DPCs using above ground storage currently deployed and licensed (see Figure 15)
- Commercial DPCs using standardized overpacks
- Commercial DPCs using licensed and deployed underground storage
- Commercial DPCs using above /below-grade vault.

³ http://energy.gov/downloads/strategy-management-and-disposal-used-nuclear-fuel-and-high-level-radioactivewaste



Artist's View of a Pilot ISF Using Existing Storage Systems

Figure 15. Schematic of Pilot ISF using currently deployed and licensed storage systems.

Design Study #2 evaluated various cask handling methods and configurations, given a range of DPC receipt rates. The objective was to determine which alternative methods improve time and motion for each process step in handling DPCs and reduce worker radiation doses. The following alternative cask handling configurations and methods were evaluated:

- Current canister transfer methods used at most nuclear power plants
- Automated canister transfer using a fixed-movement standard transfer cask and other features that remove labor and dose intensive steps
- Remote canister transfer without a transfer cask which requires a radiation shielded facility (hot cell)
- Simplified Cask Handling Operations that would not require a CHB.

Design Study #3 responds to the renewed emphasis on standardization of storage and transportation systems the same as for Study #1.

Task Order 18: "Generic Design for Small Standardized Transportation, Aging and Disposal Canister Systems" and Task Order 21: "Operational Requirements for Standardized Dry Fuel Canister Systems"

Task Orders 18 and 21 are related to the design and operations/ implementation aspects of Standardized Transportation, Aging and Disposal Canister systems and were performed by Energy*Solutions* and its team partners NAC International, Exelon Nuclear Partners, Talisman International, and Petersen

Task Order 18 entailed the development of a generic design of a small-capacity (4 PWR or BWR assemblies) STAD canister system.

Based on the design information developed under Task Order 18, Task Order 21 entailed the development of operational approaches and assessment of the associated impacts for moving predefined SNF throughput quantities (including limitations based on the number of reactors per site and cycle length) in a standardized canister to an onsite dry storage facility. Identification of innovative operational approaches was emphasized in order to minimize impacts on utility operations. The following standardized canister capacities were considered:

- 4-, 12-, and 21-PWR assembly capacity canisters
- 9-, 32-, and 44-BWR assembly capacity canisters.

For the medium and large STAD canisters, it was determined that they would be loaded individually utilizing a loading process similar to that used to load sixty-one 37-PWR DPCs in less than 52 weeks at the Zion Nuclear Power Plant.

For the small STAD canisters (4-PWR and 9-BWR), two loading processes were identified and evaluated; each of which involves loading, welding, drying and transferring small STAD canisters in groups of four:

- "STAD-in-Can" Four small STAD canisters in an overpack can with a welded lid. A shield plug (with a lifting ring) is installed and welded to each small STAD canister.
- "STAD-in-Carrier" This reflects the design concept from Task Order 18. Four small canisters, each with their inner (shield plug) and outer (top plate with lifting ring) lids installed and welded closed, and are jointly held within an open-sided carrier (see Figure 16).

The throughput parametric time and motion studies determined that each of the eight STAD system variants (4 PWR STAD systems and 4 BWR STAD systems) has the potential to meet the throughput requirements for each of the predefined nine plant cases investigated, assuming that dual transfer casks and process technology improvements are used (i.e. the "Optimized" loading processes). The medium PWR and BWR STAD canisters have the lowest margin of all analyzed cases.



Produced by EnergySolutions and team partners: NAC International, Talisman International, Petersen Incorporated and Exelon Nuclear Partners for the U.S. Department of Energy's Nuclear Fuels Storage and Transportation Planning Project

Figure 16. EnergySolutions STAD-in-Carrier design concept.

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Material Recovery and Waste Form Development Campaign



Idaho National Laboratory November 3-5, 2015 This page left intentionally blank

5. MATERIAL RECOVERY AND WASTE FORM DEVELOPMENT CAMPAIGN

5.1 Overview

T. Todd, INL, J. Vienna, PNNL

Mission

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

Material Recovery and Waste Form Development (MRWFD) applies expertise and technical capabilities to a wider array of applications than just separations. The Campaign also leverages its expertise by working with others in areas such as environmental remediation, national security missions, critical materials, as well as civilian nuclear applications.

Objectives

- Develop technologies that support the current once-through fuel cycle and have near-term potential application.
- Develop a fundamental and practical understanding of methods for the separation of uranium and transuranic elements from used fuel.
- Develop a fundamental and practical understanding of the factors affecting performance of advanced waste forms.
- Develop and demonstrate enabling technologies to separate and immobilize gaseous fission products from used nuclear fuel.
- Develop and demonstrate advanced waste forms and processes with greatly improved cost and performance.

Challenges

- Separation of trivalent actinides from lanthanides.
- Capture and immobilization of off-gas constituents of used fuel, including iodine, krypton, tritium and potentially carbon in a cost-effective manner meeting current US regulations.
- Development of separation technologies and waste forms is interrelated to the types of fuels being processed, the types of fuels being fabricated, and the reactors used to burn recycled fuels.
- Measuring waste form lifetimes in a laboratory is impossible considering they are on the order of hundreds of thousands to millions of years.
- Proliferation risk assessment of separation technologies is very subjective and must be done in the context of the entire fuel cycle (mining to disposal).

Major R&D Activities

Reference Technologies and Alternatives is developing and evaluating mass balances to enable objective comparison of technology performance for new separation technologies. Additionally, this activity supports development of co-extraction processes for uranium and plutonium, online monitoring tools, evaluation of solvent degradation mechanisms, and development of tritium removal technologies (for open and closed fuel cycle applications).

Sigma Team for Advanced Actinide Recycle is developing more robust and simplified approaches to the separation of actinides, to enable future fuel cycles that transmute transuranics for improved resource and waste management. There is a large international effort in nearly every fuel cycle country working on this difficult chemical separation and the FCT program is making significant progress on the development of cost effective methods of separating the transuranics from used fuel.

Sigma Team for Off-Gas Capture and Immobilization is focused on this critical capability required to enable any new fuel treatment facility to be licensed under current regulations. The capture of iodine with the required efficiency and the capture of 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.

Fundamental Separation Data/Methods is developing the research tools and methods to enable a science-based, engineering-driven research approach focused on understanding the fundamental scientific properties of separation processes, to move away from empiricism.

Advanced Waste Forms and Processes are necessary for the immobilization of waste streams from the advanced separations processes, including high-halide electrochemical salt wastes, gaseous fission products waste, cladding hulls, aqueous high-level waste, and separated technetium. The waste forms for streams containing ¹²⁹I, ⁹⁹Tc, and TRU 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 forms and process development is also required to significantly reduce the cost of waste treatment, storage, transportation and disposal.

Advanced Waste Form Characterization is used to develop sufficient understanding of waste form degradation behavior to predict radionuclide release over geologic timescales. This fundamental understanding is required to optimize waste form chemistry and support disposal system performance assessment.

Domestic Electrochemical Processing is developing technologies to enhance performance in the treatment of fast reactor fuels. This technology is ideally suited to treatment of metallic fuels for recycle of transuranics.

Fiscal Year-End 2015 Funding (Includes Carryover)

Material Recovery and Waste Form Development Campaign				
Major Activities	FY-15 Funding			
Campaign Management and Integration	\$ 1,403,554			
Reference Technologies and Alternative	\$ 3,943,854			
Sigma Team for Minor Actinides	\$ 3,977,152			
Sigma Team for Off-Gas	\$ 3,937,970			
Fundamental S&M / M&S	\$ 2,392,578			
Waste Form Development and Performance	\$ 5,516,302			
Domestic Electrochemical Processing	\$ 1,155,997			
Total	\$22,327,407			

Major Accomplishments

- Issued the FY-14 Annual Technical Accomplishments report. This report highlights the research accomplishments from the previous fiscal year and places the research activities in context of the overall program objectives.
- Demonstrated electrochemical oxidation of americium (from III to VI) for the first time in a noncomplexing aqueous acidic medium. The electrolysis makes use of a high-surface-area, fluoride-doped tin oxide electrode coated with nanoparticles of a tin-doped indium oxide (nITO) with an americium-selective terpyridine ligand attached to the nITO surface.
- Demonstrated co-crystallization of americium (VI) with uranyl nitrate for the first time. This activity is exploring the co-precipitation of actinyl ions (U, Np, Pu, Am) from aqueous media as a group actinide separation concept.
- Completed a number of solvent irradiation tests in the INL radiolysis/hydrolysis test loop. These tests not only supported program needs, but also international collaborations with the European Union SACSESS (Safety of Actinide Separation Processes) Program. The INL test loop is a one-of-a-kind research capability in the world and has drawn significant attention from the European research community. Solvents are being tested for Advanced TALSPEAK and ALSEP processes, and for the EU program (innovative SANEX process). A picture of a two phase mixture of aqueous solution and solvent for the innovative SANEX process in the gamma irradiator at the Materials and Fuels Complex (MFC) is shown in Figure 17.



Figure 17. Quartz tube coil filled with innovative SANEX solvent and strip solution in the gamma irradiator at MFC.

- Hosted an international workshop on radiation chemistry (Radical Behavior 2015) on July 20-22, 2015 at INL. The workshop was organized and chaired by Bruce Mincher, an INL Laboratory Fellow and internationally recognized authority on radiation chemistry. The three day workshop involved DOE national laboratory participants, universities, and participants from Germany, UK, France, and the Czech Republic. A tour of the INL's separation and radiation chemistry research laboratories was also part of the workshop.
- Synthesized and evaluated molecular sieve zeolite membranes to separate and concentrate deuterated water from ordinary water. Deuterated water was used as the nonradioactive simulant for tritiated water. Several silicoaluminophosphate (SAPO-34) molecular sieve zeolite membranes were synthesized and characterized with gas and vapor permeation measurements. The pervaporation process performance was evaluated for the separation and concentration of deuterated water, and a high separation factor (>15) was achieved.
- Completed the "Closed Fuel Cycle Waste Treatment Strategy Report," (PNNL-24114). This report summarizes the current status of development of waste forms for a closed nuclear fuel cycle and summarizes the masses and volumes of waste forms for both primary and secondary waste streams expected from both aqueous and electrochemical reprocessing plants. The report reflects the changes in process flowsheets and waste management strategies that have occurred since a similar study was conducted in 2007 for the Global Nuclear Energy Program (GNEP).
- Designed and fabricated a sorbent-based capture system designed to integrate into the existing dissolver off-gas treatment system at the ATALANTE facility. The system has undergone shakeout testing at ORNL. A photograph of the simplified system is shown in Figure 18.



Figure 18. Photograph of ATALANTE iodine capture system.

• Monitored a glass corrosion experiment for corrosion extent and pH in real time. This was the first time for this type of experiment. Although the experimental set-up needs to be improved, the results for the U.S. reference environmental assessment glass performed well with prior experiments and confirmed the utility of the technique with measurements real-time. Additional tests have been initiated to obtain immediate knowledge of "Stage III" corrosion in detail (Figure 19).



Figure 19. Real time Raman probe measurement of EA glass corrosion progress.

 Completed an initial melt test of a Synrocbased ceramic waste form using the Cold Crucible Induction Melter (CCIM). This is a significant accomplishment and represents only the third time that a ceramic waste form was melted. On this occasion, unlike the previously reported attempts, the melter was started using a prototypic production starter ring and after the melt was formed (at above 1500°C) continuous feeding of the melter was demonstrated. Figure 20 shows the Synroc ceramic waste form melt in INL's CCIM.



Figure 20. Photograph of Synroc ceramic waste form melt in INL cold-crucible melter.

• Completed a study of ZrCl4 purification requirements to determine the effectiveness of chlorination in obtaining a sufficiently pure Zr source for reused in nuclear applications or disposal as low-level waste. These results indicate that single pass chlorination would result in a ZrCl4 condensate that would be appropriate for low-level waste (LLW) disposal, but, further purification would be required for reuse. Four purification options were investigated and show promise (Figure 21).



Figure 21. Photograph of Zircaloy cladding chlorination test at ORNL.

• Fabricated an advanced ceramic waste form (ACWF) at INL with roughly 40% higher waste loading than the baseline using a frit formulated at PNNL (Figure 22). This proof-of-concept study led to the conclusions that the approach is successful and further development is warranted.



Figure 22. Photograph of ACWF samples fabricated at INL with 40% higher waste loading.

5.2 Electrochemical Oxidation of Am(III)

C. J. Dares, T. J. Meyer, University of North Carolina at Chapel Hill B. J. Mincher, Idaho National Laboratory

Introduction and Objectives

The separation of Am from the lanthanides and other minor actinides is a crucial step in the implementation of useful fuel recycling schemes. Its presence in the nuclear waste stream greatly limits the storage capacity of geologic repositories due to heat production. ²⁴¹Am is also a major contributor to the long-term radiotoxicity of HLW. There is therefore a need to develop closed nuclear fuel recycling schemes which not only improve uranium efficiency, but also minimize the volume of HLW generated. In these schemes, Am must be separated from the lanthanides prior to transmutation as their high neutron cross-section would otherwise disrupt the fission efficiency of the recycled fuel.

Separating Am from the waste stream is difficult. It has a comparable charge density to the lanthanides and minor actinides which exist in a common +3 oxidation state, and have similar ionic radii. In aqueous solutions, its coordination chemistry is therefore quite similar leaving few options for selective separation. One approach to the separation of Am is to exploit the slightly more diffuse 5f-orbitals of Am compared to the 4f-orbitals of the lanthanides. This provides a stronger, more covalent interaction between actinides and N-donor ligands. While notable progress has been made in Am(III) complexation based strategies, significant challenges have been encountered when attempting to adapt their narrow pH range requirements for process scale-up, stimulating efforts to find alternatives.

With the exception of the Ce(IV), the lanthanides are not oxidized beyond the +3 oxidation state in acidic media. Americium can be oxidized in acidic solutions, and form thermodynamically stable *trans*-dioxo cations in both the +5 (AmO_2^+), and +6 (AmO_2^{2+}) oxidation states. These species have significantly different properties than the trivalent lanthanide and actinide species including altered coordination chemistries. These differences are being investigated to facilitate superior selectivity during separation. AmO_2^+ has a low charge density and forms very weak coordination complexes requiring separation schemes in which Am is left in solution, and the lanthanides are removed. Initial separation schemes that selectively remove Am(VI) are therefore favored because of the potential improvement in selectivity.

Am(III) oxidation in acidic media is difficult as a result of the high oneelectron oxidation potential (2.6 V in 1 M acid). It has been accomplished with a small number of strong oxidants including persulfate, bismuthate, and ozone (when Ag(I) is added as a redox mediator). Use of chemical oxidants is not ideal as their inclusion into the waste stream may complicate and necessitate additional steps, often with additional complexity.

R&D Overview

We approach the separation of Am from waste solutions by electrochemical oxidation of Am(III) followed by the selective removal of Am(VI). Electrochemical oxidation of Am(III) has been attempted previously with limited success. We have developed new electrodes



Figure 23. Structure of the ptpy derivatized *n*ITO electrode for Am oxidation.

Am(III)

Am(V)

Am(VI)

comprised of a thin layer of conductive metal oxide nanoparticles (tin-doped indium oxide, nITO) which were subsequently derivatized with a covalently bound ligand known to have an affinity for Am(III) (Figure 23). We have previously demonstrated that surface modification by covalent attachment of molecules can dramatically alter electrode behavior by imparting the reactivity and interfacial properties of the molecules to the surface. Our initial ligand of study involved a phosphonic acid derivatized Ndonor terpyridine ligand (p-tpy). It is capable of binding Am(III) strongly enough to alter redox potentials, and effect electron transfer events.

Accomplishments

Am speciation during electrochemical experiments was determined by visible spectroscopy monitoring the characteristic f-f transitions of the various Am oxidation states. We have demonstrated for the first time, the oxidation of Am(III) to Am(V) and Am(VI) at potentials as low as 1.8 V vs. the saturated calomel electrode (SCE) in pH 1 nitric acid (Figure 24). Control experiments using nITO electrodes not derivatized with the p-tpy ligand showed no evidence of Am(III) oxidation with applied potentials as high as 2.7 V – but instead resulted in rapid electrode decomposition. Decomposition of the underivatized electrodes is the result of oxidation of the electrode surface, and deterioration by oxygen bubble formation during water oxidation processes. It is clear that the surface-bound p-tpy has a beneficial effect on the oxidation of Am(III), and, imparts stability to the electrode by increasing the over potential for, and decreasing the rate of water oxidation. Increasing the applied potential using the p-tpy|n|TO electrodes results in a greater proportion of Am(VI) formed (Figure 25). To understand the factors limiting quantitative production of Am(VI), we investigated the dynamics and mechanism of auto-reduction. Am(VI) is not infinitely stable in acidic media because of radiolysis, which produces reducing agents such as hydrogen peroxide. We identified and measured rate constants for both the one-electron reduction of Am(VI) to Am(V), as well as the direct two-electron reduction of Am(VI) to Am(IV) which immediately disproportionate to yield Am(V) and Am(III).

60

 $Am(III) + nITO|p-tpy \Rightarrow nITO|p-tpy-Am(III)$ nITO|p-tpy-Am(III) $\xrightarrow{-e^{-}}$ nITO|p-tpy-Am(IV) nITO|p-tpy-Am(IV) $\xrightarrow{-e^{-}}$ nITO|p-tpy-Am(V) nITO|p-tpy-Am(V) $\xrightarrow{+2H_2O-4H^+}$ nITO|p-tpy+ [Am^VO₂]⁺ $[Am^{V}O_{2}]^{+} \xrightarrow{-e^{-}} [Am^{V}O_{2}]^{2+}$



Figure 24. Proposed oxidation mechanism using a p-tpy|nITO electrode.

Figure 25. Am species composition at the conclusion of electrolysis at different applied potentials.

These results have produced a publication which has been accepted for publication to *Science*.¹ In addition, a provisional patent describing this technology has been submitted to the USPTO.²

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5.3 Room Temperature Separation of Krypton and Xenon using Metal Organic Frameworks

P. K. Thallapally, D. Banerjee, Jian Liu, R. K. Motkuri, J. E. Cabe and M. D. Bearden; Pacific Northwest National Laboratory

Introduction and Objectives

During reprocessing, fission-product noble gases are released when the fuel cladding is breached and any fuel processing is performed. Two of these gases are xenon (Xe) and krypton (Kr). Although Xe is generated as fission product, by the time the fuel is reprocessed, all the radioactive isotopes have decayed to very low concentrations. Nevertheless, nonradioactive Xe is at a higher concentration than Kr. ⁸⁵Kr is the isotope of regulatory concern and cannot be freely released into the atmosphere. The objective is to develop and demonstrate technologies to enable the effective capture of gaseous radionuclides (particularly ⁸⁵Kr) from used nuclear fuel at near room temperature.

R&D Overview

Metal organic frameworks (MOFs) are being evaluated to remove Xe and Kr from nuclear reprocessing applications at near room temperature. MOFs have improved capacities (~60 mass% at 1 bar and RT) and selectivities at room temperature. Among the materials tested at PNNL, nickel dioxobenzenedicarboxylic acid (NiDOBDC) and a partially fluorinated MOF with copper (FMOFCu) have shown improved Xe and Kr capacities. With these MOFs, series of tests were

Process	Decontamination	Total Project Capital	Proposed Annual Consumables ^A		
	Factor	(\$10 ⁶)	(\$)		
Cryogenic Distillation	67	8.77	$267~000^{\mathrm{A}}$		
MOF Adsorption	100 ^B	8.42	78 000		

performed with more complex simulated off-gas streams. These tests show MOFs can remove parts per million levels of Xe and Kr at room temperature. The selectivity of FMOFCu was found to change from Xe > Kr (6.0 mass% at RT) to Xe < Kr (14.7 mass% at -40°C) simply by changing the temperature. Results from single column experiments suggest that Xe at parts per million levels in process off-gas stream could be removed. Similarly, two-column experiments suggest that removing the Xe first on a MOF column and then the Kr on a second bed, both with the same MOF could separate both gases from air and from each other. Given the success in separating Xe and Kr at near room temperature, an initial economic evaluation was performed to determine the viability of using a MOF-based adsorption system relative to a cryogenic distillation system. A summary of the results from the economic study are shown in Table 1. These results indicate that there is an economic advantage to the use of MOF-based processes for the removal of Kr from reprocessing off-gas streams. These results indicated that a MOF-based process was cost competitive despite the concern over the projected high materials costs. Our economic analysis concluded that further research on MOFs and related porous materials is required to improve the adsorption capacity and selectivity of Kr from process off-gases. Obtaining better capacities and selectivities would further reduce the cost per kg to remove Kr.

Accomplishments

A new metal organic framework composed of cheap starting materials with high thermal stability

(~500°C) was found to adsorb high Xe uptake (0.075 mmol/g at 3 kPa); considerably higher than the current benchmark materials NiDOBDC and CC3 under similar experimental conditions. On the contrary, the Kr adsorption isotherm exhibits a smaller uptake and does not saturate even at 100 kPa, indicative of a much weaker framework-Kr interaction. This hints that MOF is highly

Figure 26. Ca based Metal Organic Framework and cycle data at room temperature.

discriminatory for Xe over Kr. At 100 kPa, the MOF adsorbs 1.3 mmol/g of Xenon at room temperature, under the same condition Kr is adsorbed, 0.8 mmol/g but the Kr loading can be significantly improved by lowering the temperature (1.3 mmol/g; 5°C). Secondary evidence of strong Xe-selective nature of CaSDB lies in its fast adsorption kinetics, with maximum Xe uptake reached within 10 minutes. Similarly up to 20 cycles were performed on CaSDB (at 1 bar and 400 ppm) without any loss of Xe loading capacity at room temperature (Figure 26).

Single column breakthrough experiments (Figure 27) with gas mixture (400 ppm Xe, 40 ppm Kr, 78% N₂, 21% O₂, 0.9% Ar, 0.03% CO₂) at room temperature indicate a very high Xe adsorption capacity (14 mmol/kg), higher than the NiDOBDC (7 mmol/kg) and CC3 (11 mmol/kg). The Xe/Kr selectivity was found to be 14; twice the value we observed for NiDOBDC. Remarkably, the CaSDB retains its Xe uptake capacity even in the presence of water vapor (40% relative

target gas on 0.4 \cap ပိုပ 0.2 0.0 1.6 1.2 ပိုပ 0.8 0.4 1.2 0.0 ပ္ိ 0.8 \mathcal{O} 0.4 0.0 6x10⁻⁴ CO 4x10⁻⁴ ပိုပ 2x10⁻⁴ 0 6x10⁻⁵ 4x10⁻⁵ ပိုပ 2x10⁻⁵ 6x10⁻⁴ 0 ပိုင် 4x10⁻⁴ 2x10⁻⁴ 0 30 40 50 10 20 60 70 80 0 Time (min)

Figure 27. Single column breakthrough measurements using CaSDB at room temperature.

humidity), whereas water competes for the same adsorption sites as Xe/Kr in NiDOBDC under similar conditions. The outstanding Xe capacity under simulated conditions with and without water vapor makes this new MOF a leading candidate for separating the Xe/Kr from nuclear reprocessing applications.

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5.4 Tritium Separation from Dilute Aqueous Solutions

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Objectives

The release of tritium [as tritiated water (HTO)] from nuclear facilities poses a serious hazard to the environment. As a result, there is a need to control tritium release to the environment. A novel option of utilizing molecular sieve membranes to preferentially separate tritium from high-volume dilute HTO-bearing aqueous streams was evaluated to address this issue. The use of membranes for radioactive decontamination has been reported in the literature.¹⁻³ However, the proposed approach of utilizing molecular sieve membranes on robust high-surface area, high-integrity supports has not been reported.¹⁻³ Traditional tritium separation technologies have evolved around the highly energy intensive cryogenic distillation and use of adsorbents such as zeolite 3A, zeolite 4A, and Ca-mordenite .⁴⁻⁷ A low-energy alternative utilizing highly selective zeolite molecular sieve membranes was evaluated, which may demonstrate significant improvement over the available alternatives for tritium separation.

The proof of principle research was performed for the separation and concentration of deuterated water (HDO). Molecular sieve zeolite membranes were synthesized and evaluated to separate and concentrate HDO from ordinary water (${}^{1}\text{H}_{2}\text{O}$, more commonly written H₂O). HDO was used as the nonradioactive simulant for HTO. The goal in this proof of concept study is to determine the feasibility of separating tritium by taking advantage of the differences in adsorption and diffusion rates through the zeolite molecular sieve membranes. It was recently reported that silico alumino phosphate (SAPO-34) layered on porous alumina supports can perform selective separations by taking advantage of the differences in mass and kinetic diameter of the molecules being separated.⁸

R&D Overview

The SAPO-34 seed crystals and membrane were characterized by X-ray diffraction, and the synthesized membranes were tested for single-gas permeation in which the driving force is a pressure gradient. The X-ray diffraction patterns were analyzed to confirm the previously known and established phase identification of the synthesized seed crystals (Figure 28).

The analysis of HDO concentration in the feed and permeate from the pervaporation system was performed using Fourier Transform Infrared Spectroscopy (FTIR). The feed D_2O concentration was varied from about 10 to over 1,000 ppm. In nuclear waste processing, when both acid and water streams are recycled, HTO concentration is estimated to be up to 400 Ci/m³. The anticipated tritium concentration in the tritiated water is in the range of a few ppm, which is at the low end of deuterated water concentration evaluated in this study.

Several process and membrane parameters were evaluated. These include concentration of D_2O (presumed to rapidly equilibrate with natural water and form HDO), operating temperature and membrane thickness. Pervaporation experiments were performed at feed temperatures ranging from 25 to 60°C to determine the effect of vapor pressure driving force on permeance and selectivity. In order to verify that the SAPO-34 zeolite membrane layer is responsible for the observed separation of deuterated water, experiments were performed with a bare alumina support without the presence of the membrane layer. These results confirm that no selectivity was observed for HDO over H₂O in the absence of the SAPO-34 membrane layer.



Figure 28. Schematic of the membrane separation system.

Accomplishments

The results of membrane separation performance tests show the preferential permeation of HDO resulting in a substantially higher concentration of HDO in permeate as compared to the feed concentration. Separation factors calculated from the measured deuterium concentrations ranged from about 0.3 to 18.8, depending on membrane properties and operating conditions. An invention disclosure was filed at ORNL (IDSA 3549) in June 2015 which describes the selective separation and concentration of deuterated and tritiated water.

The separation factor of HDO over H₂O is defined as:

$$\alpha_{HDO/H_2O} = \frac{C_{HDO}^P / C_{H_2O}^P}{C_{HDO}^F / C_{H_2O}^F}$$

where, C_{HDO}^{P} and $C_{H_2O}^{P}$ are concentrations of HDO and H₂O in the permeate, respectively, and C_{HDO}^{F} and $C_{H_2O}^{F}$ are concentrations in the feed solution.

The effect of temperature on HDO adsorption and transport was evaluated at feed temperatures of 25, 50, and 60°C. A higher selectivity was obtained at lower operating temperature, indicating the adsorption of HDO is stronger at lower temperatures. Preparations for experiments with tritiated water are currently in progress to demonstrate the suitability of zeolite membranes for the separation and concentration of tritium from tritiated water.

Preliminary capital and operating cost estimates compared to other tritium separation methods such as combined electrolysis and catalytic exchange (CECE) will be performed in FY-16.
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5.5 Development of Titanate-Based Ceramic Waste Forms for HLW Immobilization

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Introduction and Objectives

Durable ceramic waste forms tailored to incorporate all the elements present in high-level waste (HLW), including the fission products and actinides, have the potential to broaden the available disposal options and thus minimize the storage and disposal costs associated with used nuclear fuel reprocessing. These multiphase ceramics are based on naturally occurring and geochemically stable minerals that have immobilized uranium, thorium, and other natural radioactive isotopes in the environment for millions of years, substantiating their long-term durability.

The primary advantage of ceramic waste forms is the waste ions are incorporated into thermodynamically stable crystalline phases that are substantially less soluble in water compared to glassy waste forms. A simplified melt and solidification process, has been demonstrated using Cold Crucible Induction Melter (CCIM) technology, makes ceramic waste forms an attractive solution for HLW immobilization. A primary objective of this project is to demonstrate a scaled melt-solidification process for ceramic waste forms and to characterize the ceramic waste forms produced.

R&D Overview

Two ceramic compositions were developed and tested considering the CCIM capabilities at INL. Unlike the glasses and glass-ceramics that have been processed in CCIM, ceramic materials generally melt at higher temperatures and electrically couple to the induction field at a different frequency. Therefore, viscosity behavior, electrical conductivity, and crystallization kinetics were investigated to down-select a single composition for processing in the CCIM.

X-ray diffraction, electron microscopy, inductively coupled plasma-atomic emission spectroscopy (and inductively coupled plasma-mass spectroscopy for Cs), and product consistency tests were used to characterize the ceramic materials processed in the CCIM (Figure 29). Samples were analyzed from various radial and vertical areas of the solidified ceramic to provide an indication of the homogeneity throughout the CCIM with respect to phase assemblage, chemical composition, and chemical durability.

Accomplishments

The first proof of principle CCIM test with a nonradioactive ceramic waste form for immobilization of HLW generated from an envisioned used nuclear fuel cycle was conducted in October of 2014. Characterization analyses confirmed that a crystalline ceramic with desirable phase assemblage was produced from a melt using a CCIM (Figure 30). Major hollandite, zirconolite, and perovskite phases were identified in addition to highly substituted pyrochlore phases. Expected minor phases rich in Fe, Al, or Cs were also identified but, the impact of such phases on the properties of the CCIM material appeared significantly less compared to laboratory results. Only minor differences were observed vertically or radially in the CCIM material with respect to chemical composition, phase assemblage, and durability. This recent CCIM test and the resulting characterization in conjunction with demonstrated compositional improvements support continuation of melt solidification processing of ceramics for immobilization of HLW.



Figure 29. Transmission electron microscopy (TEM) image showing crystallinity and interface between hollandite and zirconolite particles. Selected Area Electron Diffraction (SAED) pattern inset corresponds to the zirconolite phase.



Figure 30. Summary of primary phase abundances in core samples compared to estimated abundances based on feed composition and target phase assemblage.

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Joint Fuel Cycle Studies



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6. JOINT FUEL CYCLE STUDIES

6.1 Overview

M. Goff, Idaho National Laboratory

Mission

The mission of the Joint Fuel Cycle Studies Campaign is to provide technical information to enable assessment of the technical and economic feasibility and nonproliferation acceptability of electrochemical recycling and other options for managing used nuclear fuel for relevant stakeholder scenarios.

A joint study is being performed to explore options for managing used nuclear fuel, including the technical and economic feasibility and nonproliferation acceptability of the electrochemical recycling process. This 10-year collaboration is described as the Joint Fuel Cycle Studies (JFCS). The collaboration is guided by a Steering Committee consisting of representatives from multiple government agencies. To execute the JFCS, three topical working groups have been formed: the Electrochemical Recycling Working Group (ERWG); the Safeguards and Security Working Group (SSWG); and the Fuel Cycle Alternatives Working Group (FCAWG).

The JFCS is currently within the second of three phases:

- Phase I: Evaluation of laboratory-scale feasibility (2 year duration)
- Phase II: Evaluation of integrated kilogram-scale process operation (5 year duration)
- Phase III: Evaluation of recycled fuel fabrication, irradiation performance, and postirradiation inspection (3 year duration).

The Joint Fuel Cycle Studies Campaign provides broad support to DOE for the JFCS, and leads the U.S. activities of the ERWG. Major milestones and planned major activities for the ERWG during Phase I and Phase II of the collaboration are presented in Figure 31.

Objectives

- Develop and install process technologies (processing, fuels, and safeguards) to test an integrated electrochemical process at kilogram-scale.
- Complete an integrated test of LWR fuel recycles via electrochemical recycling, including fuel fabrication from recycled materials.
- Perform irradiation testing and postirradiation examination of fuel fabricated from recycled LWR constituents.
- Support the development and testing of technology for safeguarding an electrochemical recycling process.
- Support the assessment of the range of options for used fuel management relevant to JFCS stakeholders.
- Generate data to support assessment of the technical and economic feasibility and nonproliferation acceptability of electrochemical recycling.

Challenges

- Administrative challenges of an international, multi-agency collaboration
- Integration of fuel fabrication and separations recovery requirements
- Adaptation of safeguards technologies and methodologies into the integrated recycling process
- Scaling of the suite of electrochemical recycling technology to the kilogram-scale and generation of data regarding the technical and economic feasibility of further scale-up
- Receipt of modern used commercial LWR fuel to utilize as testing feedstock.



Figure 31. Major Phase I and Phase II activities of the JFCS.

Major R&D Activities

The Campaign Guidance & Technical Support activity coordinates the technical activities of the study through the ERWG of the JFCS. These activities include coordination of stakeholder interests such as the governing collaboration documents, selection of baseline and advanced process flowsheets, development of a predictive process model, oversight of integrated testing and Critical Gap R&D activities, and integration of safeguards considerations into process development.

The Integrated Recycling Test (IRT) is a kilogram-scale demonstration of electrochemical recycling with irradiated materials. This activity will allow assessment of electrochemical recycling flowsheets and provide material balance information for process modeling. This work includes the fabrication, irradiation, and postirradiation examination of metal fuel rodlets produced from recycled LWR fuel. To

the extent possible, potential safeguards technologies will be deployed and evaluated during integrated testing activities.

Critical Gap R&D activities are a key component of the JFCS to acquire applied and fundamental knowledge necessary to close process modeling gaps, resolve critical technical issues important to success of the Integrated Recycling Test, and develop and demonstrate improved technologies that are critical to commercial-scale process feasibility. Highlights of experimental activities which are currently underway include experiments toward improved long-life anodes for oxide reduction processes, exploration of methods to reduce lanthanide content in recovered U/TRU products, and investigation of waste forms with simplified production processes and high tolerance for chloride constituents.

Fuel Fabrication and Performance activities include the preparation of a remote sampling and fuel casting process, the utilization of an established test capsule preparation process for irradiation experiments in the Advanced Test Reactor, and established postirradiation capabilities. Activities also include the research of key fuel fabrication or performance issues, such as rodlet inspection technology, reduction of fuel-cladding chemical interaction through cladding barrier layers, and development of fuel fabrication technology for the recycling of LWR fuel with rare earth fission products.

Fiscal Year-End 2015 Funding (Includes Carryover)

Joint Fuel Cycle Studies Campaign	
Major Activities	FY-15 Funding
Non-Cost-Shared Activities	\$ 914,212
Electrochemical Recycling Activities	\$ 5,634,877
Fuels	\$ 997,110
Total	\$ 7,546,199

Major Accomplishments

- The Phase IIb CRADA was signed by program stakeholders.
- Facility preparations necessary to receive commercial fuel pins for use in the IRT were completed. Fuel rods were loaded into a cask liner by the fuel vendor in preparation for delivery.
- The suite of process equipment moved through the fabrication and qualification process. Notably, the molten salt furnace for the oxide reduction process step was installed into a workstation inside the Hot Fuel Examination Facility.
- Slugs of simulated recycled fuel were cast using prototypic materials.
- A study on the behavior of carbon anodes under typical oxide reduction conditions was completed.

6.2 Processes for an Integrated Study of Electrochemical Recycling

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Introduction and Objectives

The IRT is a kilogram-scale test designed to evaluate a baseline flowsheet for electrochemical recycling of LWR fuel and to provide material balance information for an integrated process model. The IRT is a critical requirement for the overall goal of the Joint Fuel Cycle Study to explore the technical and economic feasibility as well as nonproliferation acceptability of the electrochemical recycling process. The IRT will be performed at the Hot Fuel Examination Facility (HFEF) at Idaho National Laboratory. The presentation will discuss the processes to be utilized for the IRT.

Several unit operations are required for kilogram-scale processing of LWR fuel. The feedstock fuel material will be recovered primarily using mechanical decladding. After separation of the fuel and cladding the oxidized fuel will be reduced electrolytically to metal in a molten salt solution. The reduced fuel with residual salt will then be subjected to a salt removal distillation process in order to avoid salt cross-contamination in subsequent processes. After salt removal, the reduced fuel is electrorefined for group separation of low-enriched uranium (U) and uranium/transuranic/rare earth (U/TRU/RE) products. The electrorefined products contain residual salt or cadmium that is also removed through vacuum distillation. Final U/TRU/RE products will be cast into fuel slugs via a remote casting process. These steps are shown graphically in Figure 32.



Figure 32. Overview of material flows and the major processes planned for the IRT.

6.3 Interfacial Kinetic Studies of Actinides in Electrochemical Systems

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Introduction and Objectives

Electrorefining is the main actinide separation process in electrochemical reprocessing. During this process, actinides are separated from the bulk of the fission product elements by electrotransport onto a solid or liquid cathode. Passing current between the anode and cathode of the cell simultaneously dissolves used fuel from the anode, and electrodeposits metallic uranium at one cathode and/or uranium – transuranic elements at a different cathode. The goal of electrochemical processing is to maximize the separation and recovery of actinides from chlorinated fission products so that minimal amounts of actinides are lost to the process waste stream. Availability of accurate electrochemical kinetic data for actinides is essential to maximizing the processing rate of the electrorefining step and enabling effective technology development.

Very few studies have been conducted to determine interfacial electron transfer kinetic information for actinides and lanthanides in molten salts (i.e., standard rate constant (k^o), exchange current density (i_o), and transfer coefficient (α)). Most of the studies have been done to measure the basic thermodynamic and electrochemical properties (deposition potentials, diffusion coefficients, etc.) of actinides, which do not involve kinetic considerations and do not provide any information about the processing rate of the electrorefining step. The specific objective of the present work is to determine the interfacial electron transfer kinetics for actinides in molten salts using electroanalytical methods.

R&D Overview

A typical electrodeposition process involves at least two consecutive steps: mass transfer of the reactant from the bulk solution to the electrode interface and charge transfer at the electrode surface to form a product. A simple schematic of the electrode processes occurring at the electrode/electrolyte interface during the electrodeposition of U^{3+} to U^0 is shown in Figure 33. The main requirement to measure the heterogeneous electrode kinetics using electroanalytical techniques is that the heterogeneous electron transfer rate of the overall electrode process must be manifested in the electrochemical experiment. Therefore, the electrode reaction rate must be controlled either partially (quasi-reversible system) or totally (irreversible system) by the charge-transfer reaction that occurs between the electrode and an ion or molecule, and involves the transfer of electrons across the electrode/electrolyte interface rather than the process being controlled purely by diffusion. In theory by changing the time scale of the experiment, one can render even a completely reversible electrode reaction quasi-reversible or even totally irreversible in order to determine its kinetic parameters. Practically, however, this approach is limited by the resistive and capacitive effects, which cause degradation of the electrochemical response and impair the measurements. Depending on the technique used, there are different ways to achieve varying degrees of kinetic irreversibility, and thus different restrictions apply for achieving effective measurements.

In addition to experimentally achieving and verifying kinetic irreversibility, the theory and mathematical relationships describing the irreversible experimental dependences measured by these methods (e.g., peaks, plateaus) must be available in order to analyze the system. For the majority of electrochemical techniques involving soluble-soluble reactions, well-established theories and fundamental equations are available. However for reactions involving the formation of an insoluble product (e.g., electrodeposition),

no mathematical relations have been established describing the current-potential characteristics of the irreversible curves generated by any of the available electrochemical techniques. Because redox reactions of interest for actinide ions in molten salts usually involve the formation of an insoluble product, additional effort is required to derive the desired relationships and to thoroughly verify their accuracy.



Figure 33. A schematic of the electrode processes occurring during uranium electrodeposition.

Accomplishments

Available direct-current (DC) electrochemical techniques capable of measuring the interfacial kinetic parameters have been explored and experimental conditions essential for successful determination of electrode kinetics have been reviewed. Not surprisingly, it was found that the interfacial electron transfer kinetics of actinides at elevated temperatures are quite facile and difficult to measure with conventional methods that impose potential sweeps, potential steps, or current steps because of the complications arising from nonfaradaic charging current and ohmic effects in the system. Fortunately, a different method has been identified that is based on analyzing the faradaic impedance and observing the response of the system to low-amplitude sinusoidal signal perturbations as a function of frequency. This method, AC voltammetry, allows for discrimination against the background current and allows accurate evaluation of interfacial kinetic parameters even for systems with very fast kinetics. In addition to the ability to measure fast electron transfer kinetics, the fact that AC voltammetry provides dual time domain measurements plus additional phase angle measurements, permits direct evaluation of three independent electrochemical parameters (i.e., exchange current density (i_o), transfer coefficient (α), and diffusion coefficient (D_o)).

Using the available information in the literature on faradaic impedance interpretation and the fundamentals of AC voltammetry, the theory describing the current-potential characteristics for the deposition of an insoluble substance has been derived. The mathematical relationships describing the AC current-potential characteristics for two time domains (reversible and irreversible) as well as phase angle were obtained. Analysis of these equations and their dependence on experimental parameters revealed that measurements using AC voltammetry enable direct evaluation of three independent electrochemical parameters (D_o , i_o , and α), which is the great advantage of this technique compared to the commonly used DC techniques. The presentation will cover the results of both preliminary experiments using the AC technique and the experimental parameters necessary to acquire the desired information, as well as the procedures required to obtain accurate and reproducible data.

Used Fuel Disposition R&D Campaign



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7. USED FUEL DISPOSITION RESEARCH AND DEVELOPMENT CAMPAIGN

7.1 Overview

P. Swift, Sandia National Laboratories

Mission

The Used Fuel Disposition R&D (UFD) Campaign identifies alternatives and conducts scientific research and technology development to enable storage, transportation, and disposal of used nuclear fuel (UNF) and wastes generated by existing and future nuclear fuel cycles.

Objectives

Near-Term Objectives (2016–2020)

- Support the DOE-Industry high burn-up fuel full-scale storage demonstration project
- Develop understanding of how temperature and pressure affect cladding integrity in high-burnup UNF through experimentation and predictive modeling
- Develop understanding of how corrosion and stress corrosion cracking affect performance of stainless steel dry storage canisters through collection of material and environmental data and predictive modeling
- Characterize external loadings on UNF during normal conditions of transport
- Field a deep borehole test, with drilling beginning in 2016 and testing complete in 2019
- Complete evaluation of the direct disposal of dual-purpose canisters
- Develop the experimental and modeling basis for understanding long-term performance of disposal systems in clay/shale rock.

Long-Term Objectives (2020–2025)

- Support the implementation of a full-scale, NRC-licensed confirmatory storage demonstration facility via significant collaboration with industry.
- Develop the technical basis necessary to support eventual transportation of UNF.
- Develop the technical basis of the Deep Borehole Disposal concept with Deep Borehole Field Test data.
- Support implementation of integrated storage, transportation and disposal concepts.

Challenges

Provide a sound technical basis for supporting the DOE strategy for managing the back end of the nuclear fuel cycle, including the identification and evaluation of safe and secure options for storage, transportation, and permanent disposal of radioactive wastes resulting from existing and future fuel cycles.

Major R&D Activities

Storage and Transportation R&D examines three topics: Storage, Transportation, and Security. Storage R&D focuses on closing technical gaps related to extended storage of UNF, including uncertainties with high-burnup used nuclear fuel cladding performance and long-term canister integrity. Transportation R&D focuses on ensuring transportability of UNF following extended storage, addressing data gaps regarding nuclear fuel integrity, retrievability, and understanding stresses and strains during normal conditions of transport. Security R&D focuses on questions related to material attractiveness and self-protection due to surface dose rate, which decreases as UNF ages. The UFD campaign participates in the international Extended Storage Collaboration Project, led by the Electric Power Research Institute (EPRI) with input from the DOE, the U.S. Nuclear Regulatory Commission (NRC), and programs in multiple other nations.

Disposal R&D focuses on identifying multiple viable geologic disposal options, addressing technical challenges for generic disposal concepts in various host media (e.g., mined repositories in salt, clay/shale, and granitic rocks, and deep borehole disposal in crystalline rock). 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. International collaborations include: DECOVALEX (Development of Coupled Models and their Validation against Experiments, with participation from multiple nations in Europe and Asia); the Mont Terri underground research laboratory (Switzerland); Colloid Formation and Migration (Switzerland); SKB Task Force (Sweden), Salt R&D (Germany); and crystalline disposal R&D with Korean Atomic Energy Research Institute (KAERI) Underground Research Tunnel. The deep borehole disposal concept has not been demonstrated anywhere in the world, and the campaign has therefore identified support for a DOE-managed field test using nonradioactive surrogate waste as one of its highest priorities for the coming years. Drilling for the deep borehole field test is planned to begin in the fall of 2016.

DOE-managed High Level Waste and Spent Nuclear Fuel Research. This disposal research activity is intended to establish a path for the permanent disposal of high-level radioactive waste and SNF derived from national defense nuclear activities and R&D activities of the Department of Energy (DOE). In FY-16, specific attention will be given to the activities needed to implement a plan for disposal of the subject wastes within the DOE's existing authority under the Atomic Energy Act of 1954, and fully consistent with the requirements of the Nuclear Waste Policy Act of 1982, as amended.

DOE-managed Industry Storage Cask Demonstration. The storage cask demonstration project, led by EPRI under contract to DOE, is conducting a multiyear test to collect data from a UNF dry storage system (located at the North Anna Nuclear Power Plant in Virginia) containing high burnup fuel. The primary goals of the test are to provide confirmatory data for model validation and potential improvement, provide input to future dry storage cask design, support license renewals and new licenses for Independent Spent Fuel Storage Installations, and support transportation licensing for high burnup used fuel.

Fiscal Year-End 2015 Funding (Includes Carryover)

Used Fuel Disposition R&D Campaign		
Major Activities	FY-15 Funding	
Campaign Management and Integration	\$ 1,089,762	
Storage and Transportation Research	\$10,777,536	
Disposal Research	\$14,605,941	
DOE-managed Industry Storage Cask Demonstration	\$ 6,000,000	
Total	\$ 32,473,239	

Major Accomplishments

- Supported the industry storage cask demonstration by actively participating in:
 - Support for finalization of the EPRI Test Plan.
 - R&D in support of nondestructive evaluations of the demonstration cask.
 - Selection and eventual placement of assemblies to obtain the greatest diversity in cladding and heat exposures during the duration of the test.
 - Planning for cask lid design, licensing issues, and approval of final design.
- Completed cladding bend test on irradiated Zircaloy-4 and M5 cladding. Results showed that the irradiated fuels can withstand millions of bending cycles at potentially higher than representative loading levels before breaking.
- Tested a surrogate PWR fuel assembly to simulated rail and truck normal transport conditions on shaker tables and an actual over-the-road truck test to better understand the integrity of high burnup fuel during transport. Results provide further confirmation that stresses and strains applied to the fuel rods and assembly are well below those predicted to damage cladding or the assembly.
- Updated analyses of dual-purpose canister (DPC) disposal alternatives indicate that DPC direct disposal could be technically feasible, at least for some DPCs in some disposal concepts. Operational engineering challenges can be met, thermal load management objectives through combinations of aging, ventilation, and spacing, and postclosure criticality concerns can be addressed for individual DPCs as needed. DPC disposal is probably not an all-or-nothing option: SNF in some DPCs may require repackaging regardless of the selected disposal concept.
- Supported the DOE in issuing a Request for Proposals for the deep borehole field test (for a site, site management team, and drilling capabilities for the characterization borehole), published on July 9, 2015.

Deliverables

Storage & Transportation:

- Test Plan for Sister Rod Characterization and Testing, M2FT-15IN08020111, March 2015
- Evaluate the Frequency for Gas Sampling for the High Burn-up Storage Demonstration Project, M2FT-15IN0802013, April 2015

- Conduct ring compression tests on HBU PWR cladding alloys at 350°C, M2FT-15AN0805011, September 2015
- Conduct cyclic bend tests of used nuclear fuel, M2FT-15OR0805031, September 2015
- Stress Corrosion Cracking Investigation on a Full Scale Stainless Steel Canister Mock-up, M2FT-15SN0805051, August 2015
- Thermal profile analyses of in-situ industry storage systems identified for inspection, M2FT-15PN0810049, September 2015
- Development of Uncertainty Quantification Methodology as Applied to Storage and Transportation R&D, M2FT-15SN0810051, July 2015.

Disposal Research

- Evaluation of Used Nuclear Fuel Disposition in Clay-bearing Rocks, M2FT-15SN0806071, September 2015
- Evaluation of Used Nuclear Fuel Disposition in Crystalline Rocks, M2FT-15SN0807071, September 2015
- Application of Generic Disposal System Models, M2FT-15SN0808011, September 2015
- International Collaboration Activities in Different Geologic Disposal Environments, M2FT-15LB0811012, September 2015
- Investigations on Technical Feasibility of Direct Disposal of Dual-Purpose Canisters, M2FT-15SN0816021, May 2015
- Site Evaluation for Deep Borehole Field Test, M2FT-15SN0817061, June 2015
- Conceptual Design and Requirements for Characterization and Field Test Boreholes, M2FT-15SN0817081, September 2015
- Deep Borehole Field Test Specifications, M2FT-15SN0817091, September 2015
- Draft Test Plan for Phased Large-Scale Thermal Testing, M2FT-15LA0819016, April 2015.

7.2 Deep Borehole Field Test Overview

G. Freeze and D. Sassani, Sandia National Laboratories

Using a reference design that is expected to be achievable in crystalline rocks with currently available commercial drilling technology, the implementation of the deep borehole disposal concept may be technically feasible, cost effective, and have sufficient capacity for some waste forms. In the reference design (Figure 34), waste packages are emplaced in the lower 2,000 m of a 5,000-m-deep borehole, with the upper 3,000 m of the borehole appropriately sealed with a combination of bentonite, cement plugs, and cement/crushed rock backfill (Arnold et al 2011).

DOE's Assessment of Disposal Options for DOE-Managed High-Level Radioactive Waste and Spent Nuclear Fuel (DOE 2014a) recommended "that DOE retain the flexibility to consider options for disposal of smaller DOE-managed waste forms in deep boreholes …, and that DOE conduct the deep borehole field test needed to confirm the safety and feasibility of the concept." Accordingly, the DOE UFD Campaign is proceeding with a Deep Borehole Field Test (DBFT) to further evaluate the deep borehole disposal concept and technical capabilities for implementation, *without use of any radioactive waste*.

Deep Borehole Field Test

The DBFT Project Plan (SNL 2014) includes drilling two boreholes to a depth of about 5,000 m (16,400 feet) into crystalline basement rock in a geologically stable continental location. An initial smallerdiameter (~21.6 cm [8.5"] diameter at total depth) Characterization Borehole (CB) will be drilled and completed to facilitate downhole scientific testing and analysis. If site conditions are found to be suitable, DOE may decide to drill a larger-diameter (43.2 cm [17"] diameter at total depth) Field Test Borehole (FTB) to facilitate proof-of-concept of emplacement activities using surrogate waste packages.

Two boreholes provide a robust approach to achieve the overall goals of the DBFT. First, downhole characterization can be achieved with standard logging technology and methodology in the CB, whereas characterization in the larger-diameter FTB would present additional technical challenges. Second, the diameter of the CB will be large enough to accommodate testing the system for emplacing test packages representative of smaller waste forms such as Cs/Sr capsules. Third, by conducting characterization activities in the smaller CB, the costs associated with those activities could be reduced significantly compared to the costs for characterization activities in the FTB. Finally, two holes will provide the unique opportunity for cross-hole testing at depth, if such testing is deemed warranted. Both geophysical and geohydrological cross-hole testing could be used in the DBFT to assess the viability of performing future characterization at a deep borehole disposal site from only a single borehole.

As a first step, DOE issued a Request for Information (RFI) (DOE 2014b) to "seek interest in, and input from industry, States, local communities, individuals, private groups, academia, or any other stakeholders willing to host a Deep Borehole Field Test." Subsequent to the RFI, the DOE issued Draft (DOE 2015a; April 7, 2015) and Final (DOE 2015b; July 9, 2015) Requests for Proposal (RFP) to seek competitive bids from contractor teams capable of providing (1) a site suitable for the DBFT (accommodating both boreholes), and (2) site-management, science and technology support, and drilling services for the CB. FTB drilling and construction and the emplacement demonstration will be performed under a second RFP and contracting process.

The DBFT activities address several important areas to confirm the viability of the deep borehole disposal concept:

- Evaluation and verification of geological, geochemical, geomechanical, and geohydrological conditions at a representative location;
- Demonstration of drilling technology and borehole construction to 5 km depth in crystalline basement with sufficient diameter for cost-effective waste disposal;
- Evaluation of package, waste, and seals materials at representative temperature, pressure, salinity, and geochemical conditions in the laboratory;
- Development and testing of engineering methods for test package loading, shielded surface operations, test package emplacement, and borehole seals deployment; and
- Demonstration of preclosure and postclosure safety.



Figure 34. Generalized concept for deep borehole disposal of radioactive waste.

References

- 1. Arnold, B.W., P. V. Brady, S. J. Bauer, C. Herrick, S. Pye and J. Finger, 2011, *Reference Design and Operations for Deep Borehole Disposal of High-Level Radioactive Waste*, SAND2011–6749. Sandia National Laboratories.
- 2. DOE, 2014a, Assessment of Disposal Options for DOE-Managed High-Level Radioactive Waste and Spent Nuclear Fuel, U.S. Department of Energy, Washington DC, October 2014.
- 3. DOE, 2014b, Request for Information (RFI) Deep Borehole Field Test, Solicitation Number DE-SOL-0007705, U.S. Department of Energy Idaho Operations Office.
- 4. DOE, 2015a, Draft Request for Proposals (RFP) Deep Borehole Field Test (April 7, 2015), Solicitation Number DE-SOL-0008071, U.S. Department of Energy Idaho Operations Office.
- 5. DOE, 2015b, Request for Proposals (RFP) Deep Borehole Field Test (July 9, 2015), Solicitation Number DE-SOL-0008071, U.S. Department of Energy Idaho Operations Office, Idaho Falls, ID.
- 6. SNL, 2014, *Project Plan: Deep Borehole Field Test*, FCRD-UFD-2014-000592, Rev. 0; SAND2014-18559R, US Department of Energy Used Fuel Disposition Campaign.

7.3 International Collaboration Activities in Disposal Research

J. Birkholzer, Lawrence Berkeley National Laboratory

In 2012, the Used Fuel Disposition Campaign (UFD) initiated an International Disposal R&D Program that significantly advanced active collaboration with several international geologic disposal programs in Europe and Asia. Such collaboration has allowed the campaign to benefit from a deep knowledge base with regards to alternative repository environments developed over decades, and to utilize international investments in research facilities (such as underground research laboratories), saving millions of R&D dollars that have been and are being provided by other countries. Guiding principles for selection of collaboration opportunities are as follows:

- Focus on activities that complement ongoing disposal R&D within UFD (e.g., the science and engineering tools developed in UFD are tested in comparison with international experiments).
- Select collaborative R&D activities based on technical merit, relevance to safety case, and cost/benefit, and strive for balance in terms of host rock focus and repository design.
- Emphasize collaboration that provides access to and/or allows participation in field experiments conducted in operating underground research laboratories not currently available in the U.S. (i.e., clay, crystalline).
- Focus on collaboration opportunities for active R&D participation (i.e., U.S. researchers work closely together with international scientists on specific R&D projects relevant to both sides).

To date, the International Disposal R&D Program has established formal collaboration agreements with five international initiatives and several international partners, and national lab scientists are participating in a balanced portfolio of international R&D activities, addressing relevant R&D challenges in fields like near-field perturbation, engineered barrier integrity, RN transport, and integrated system behavior. These form a considerable portion of UFD disposal research, in particular in the Crystalline and Argillite work packages, and significant advances have been made over the past few years in developing the technical basis for disposal in a range of potential host rock environments. Comparison with experimental data provided by international partners has contributed to testing and validating predictive computational models for evaluation of disposal system performance in a variety of generic disposal system concepts. Comparison of model results with other international modeling groups, using their own simulation tools and conceptual understanding, have enhanced our confidence in the robustness of predictive models used for performance assessment. The possibility of linking model differences to particular choices in conceptual model setup provides guidance into "best" modeling choices and understanding the effect of conceptual model variability. Promising opportunities exist for further expansion of the international program.

The following gives a brief overview of the five multinational cooperation initiatives DOE has joined as a formal partner:

DECOVALEX Project

The DECOVALEX Project is an international research collaboration and model comparison activity for coupled processes simulations in geologic repository systems (currently 10 project partners). The project develops modeling test cases that involve experimental data sets from international underground research facilities. Typically, these experimental test cases are proposed by one of the project partners, and are

then collectively studied and modeled by all DECOVALEX participants. Currently, the project involves test cases from four international underground research laboratories (URLs) in France (Tournemire), Japan (Horonobe), Switzerland (Mont Terri), and the Czech Republic (Bedrichov Tunnel). Modeling cases with UFD involvement include, for example, an engineered-barrier heater test and the use of environmental tracers for estimating fracture properties.

Mont Terri Project

The Mont Terri Project is an international research partnership for the characterization and performance assessment of a clay/shale formation (currently 15 partners). The partnership essentially provides open access the existing Mont Terri URL in Switzerland. Partner organizations can conduct experiments in the URL, can participate in experiments conducted by others, and have access to all project results from past and ongoing efforts. In the current phase, the Mont Terri Project comprises about 40 separate experiments that are relevant to all relevant phases in the lifetime of a repository. UFD researchers have engaged in several projects ranging from large-scale heater tests (Figure 35) to damage zone and diffusion experiments.





Colloid Formation and Migration (CFM) Project

The CFM Project is an international research project for the investigation of colloid formation, bentonite erosion, colloid migration, and colloid-associated radionuclide transport. This collaborative project (currently nine partners) is one of several experimental R&D projects associated with the Grimsel Test Site (GTS) in the Swiss Alps, a URL situated in sparsely fractured crystalline host rock and one of few facilities underground that permits radionuclide studies. The CFM project conducts radionuclide migration experiments in a fracture shear zone complemented by laboratory and modeling studies. UFD researchers have interpreted field measurements conducted as GTS using semi-analytical and numerical

methods, and have supported the field interpretation with laboratory experiments on colloidal transport and sorption.

FEBEX Dismantling Project

The Full-scale Engineered Barriers EXperiment (FEBEX) experiment at GTS consists of an *in situ* fullscale heater test conducted in a crystalline host rock with bentonite backfill (currently 10 partners). The heating phase of the experiment, which ended in Spring 2015 after 18 years of operation, was followed by a new project, the FEBEX Dismantling Project (FEBEX-DP), aimed at dismantling the test site and conducting postmortem analysis of engineered and natural barrier components to better understand the long-term performance of barrier components after heating and natural resaturation. UFD researchers have been participating in the planning and predictive modeling of the experiment, and will soon conduct sample analysis and interpretation of long-term engineered barrier behavior.

SKB (Swedish Nuclear Fuel and Waste Management) Task Forces

The SKB Task Forces are a forum for international collaboration in the area of conceptual and numerical modeling of performance-relevant processes in natural and engineered systems (currently 12 partners). One task force focuses on flow and radionuclide migration processes in naturally fractured crystalline rock (GWFTS Task Force); another task force tackles remaining challenges in predicting the coupled behavior of the engineered barrier system (EBS Task Force). The task force topics center on experimental work conducted at the Äspö Hard Rock Laboratory (HRL) situated in crystalline rock. DOE joined both task forces in January 2014. UFD researchers are actively engaged in the interpretation and modeling of a bentonite-rock interaction experiment currently under way at HRL.

UFD has also explored bilateral collaboration opportunities for active collaboration, and has selected additional R&D activities with potential for substantial technical advances. One example for fruitful bilateral collaboration is the joint work with the Korea Atomic Energy Research Institute (KAERI) regarding the KURT underground research laboratory hosted by a shallow tunnel in a granite host rock, located in a mountainous area near Daejeon, Republic of Korea. UFD researchers are developing improved techniques for *in situ* borehole characterization and are also testing methods for measuring streaming potential (SP) to characterize groundwater flow in a fractured formation. The approach will soon be tested in the field in KURT following an ongoing expansion of the underground facility.

7.4 Modeling Used Fuel under Transportation and Storage Loads

N. A. Klymyshyn, Pacific Northwest National Laboratory

Introduction and Objectives

Finite element modeling of a detailed used fuel assembly is being used to help understand and quantify the loading that used nuclear fuel assemblies may experience during different stages of transportation and storage. The level of detail in the fuel assembly includes realistic rod to rod contact, nonlinear spacer grid leaf spring behavior, and spacer grid geometry that can realistically crush under sufficient loading. This finite element model has been validated against experimental fuel assembly surrogate dynamic testing performed by SNL, and interpretation of the results is aided by the experimental rod bend test data collected by ORNL. The modeling and experimentation together are forming the technical basis needed to reach conclusions about the safety of transporting and storing used nuclear fuel in an extended dry storage environment.

Transportation Modeling

Transportation modeling supports the ongoing fuel assembly test campaign led by SNL, which uses a real fuel assembly with surrogate fuel rods to study the fuel's response to simulated normal condition of transport dynamic loads. Pretest modeling was used to guide the placement of strain gages and accelerometers for testing. Posttest modeling provided validation of the detailed LS-DYNA fuel assembly model. Figure 36 shows the shaker table model configuration with the detailed fuel assembly model. After validation, the model was used to estimate the difference in response between the surrogate fuel used in testing and realistic used fuel. Modeling was also used to address questions and technical concerns about the test configuration, including the influence of surrogate fuel configuration on the cladding strains, the influence of the conveyance structure on transmitted loads, and whether a one foot drop of a truck package is potentially more limiting than the normal conditions of transport (NCT) shock and vibration environment. The transportation modeling work is continuing to support development of a rail test plan, and includes a 1-foot drop model of the ENSA package in early FY-16. Some of the key transportation modeling conclusions made to date:

- The LS-DYNA model of the fuel assembly shows good agreement with recorded test data.
- The NCT shock and vibration environment for truck transport is relatively low in magnitude.
- The 1-foot drop of a hypothetical truck transportation package is potentially more limiting than the NCT shock and vibration environment.
- Recent modeling results suggest that cladding strains are not strongly sensitive to surrogate fuel composition or conveyance design in the low-magnitude truck NCT shock and vibration environment. Similar sensitivity studies will be performed for the rail NCT shock and vibration environment to support the development of a rail test plan.

Storage Modeling

The long-term storage modelling considers loading scenarios associated with extended dry storage: seismic events, canister handling drops, and dry storage cask tip-overs. The goal is to determine the loads associated with these three events and then consider how much material degradation may occur before the loads become a safety problem. The intent of this analytical study is to help prioritize the material testing

programs. Models of a vertical dry storage cask system were developed and were used to evaluate the canister response. Detailed fuel assembly models were added to the cask system models to evaluate the fuel assembly response, particularly the cladding strains. Figure 37 shows the tip-over finite element model and contours of bending moment in the cladding beams. The potential for stress corrosion cracking in the canister welds has started, and will continue into FY-16. Some of the key storage modeling conclusions made to date:

- The tip-over load case causes the highest loads of the three scenarios considered. The canister handling drop and seismic load cases are both relatively benign compared to tip-over.
- None of the three load cases predict MPC wall failure from direct loading in the as-built condition. Significant material degradation or wall thinning would be necessary to challenge the MPC integrity.
- The potential for stress corrosion cracking in the MPC welds was analyzed in the tip-over case. It was found that ductile failure of the wall section is more plausible than failure due to crack propagation.
- Peak cladding strains are relatively high in the cask tip-over case. Further model evaluation is needed to determine if peak cladding strain is near the failure threshold or over it.



Shaker Table Model Configuration

Figure 36. Transportation model, shaker table configuration.



Figure 37. Storage model, cask tip-over scenario.

7.5 Multi-Sensor Inspection and Robotic Systems for Dry Storage Casks

C. J. Lissenden, Professor of Engineering Science and Mechanics, Pennsylvania State University

Inspection of dry storage casks for used nuclear fuel is a significant challenge due to environmental conditions and geometric access constraints. The primary modes of degradation are anticipated to be stress corrosion cracking in the heat affected zone of welds in stainless steel containment canisters and general deterioration of the protective concrete overpack. This presentation will highlight progress from the first year of an integrated research project awarded by the Nuclear Energy University Program. The project includes development of: laser induced breakdown spectroscopy (LIBS) to characterize surface composition (especially the presence of chlorides), sensors and ultrasonic guided wave inspection methodology for detection of cracking, as well as ultrasonic systems for characterization of concrete degradation. A key aspect of the project is that the sensor systems used for these inspections must be deployed by robotic delivery systems that operate in a harsh constrained environment. A novel sensor delivery method is being developed for the geometry of vertical axis casks. Furthermore, data management techniques are under development to ensure that the acquired data are traceable to specific locations inside the cask.

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Material Protection, Accounting, & Control Technologies



Idaho National Laboratory November 3-5, 2015 This page left intentionally blank

8. MATERIAL PROTECTION, ACCOUNTING, AND CONTROL TECHNOLOGIES

8.1 Overview

M. Miller, Los Alamos National Laboratory

Mission

Develop innovative technologies and analysis tools to enable next-generation nuclear materials management for existing and future U.S. nuclear fuel cycles, to manage and minimize proliferation and terrorism risk.

Objectives

- Develop tools, technologies, and approaches in support of used fuel safeguards and security for extended storage, electrochemical processing, and other advanced nuclear energy systems.
- Develop and demonstrate advanced material control and accounting technologies that would, if implemented, fill important gaps in existing Materials Protection, Accounting and Control Technology (MPACT) capabilities.
- Develop, demonstrate and apply MPACT analysis tools to assess effectiveness and efficiency of MPACT systems, guide R&D and support advanced integration capabilities.
- Perform technical assessments in support of advanced fuel cycle concepts and approaches.
- Develop guidelines for safeguards and security by design and apply to new facility concepts.

Challenges - Key Drivers

- It is likely that used fuel will be stored for an extended time until an ultimate disposition pathway is available.
- 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.
- Lab-scale demonstration of an advanced safeguards and security system in the early 2020 time frame.

Technical needs

- Improve the precision and accuracy nuclear material accountancy measurements, while at the same time improving their timeliness and cost-effectiveness.
- 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 in an integrated manner that quantitatively takes into account uncertainties and correlations.
- 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.

Major R&D Activities

Safeguards and Security by Design – Electrochemical: Safeguards and Security by Design is a methodology and discipline for integrating next generation MPACT considerations into the design of nuclear facilities from the very 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, minimizing the need for costly retrofits. Electrochemical processing is being used as the test case for application to advanced fuel cycle technologies, in coordination with the Material Recovery and Waste Forms Campaign and the Joint Fuel Cycle Study. Advanced concepts and approaches, analysis tools, and instrumentation are being developed and applied in an integrated manner to optimize the overall system effectiveness.

Used Fuel Extended Storage: Concepts and approaches are being developed for integrated safeguards and security for used fuel extended storage. This includes the risk-informed security analysis (vulnerability and consequence), assessment of and addressing technology gaps, and providing leadership in the area of international best practices for security of dry storage. This effort is coordinated with the Used Fuel Disposition Campaign and the Nuclear Fuel Storage and Transportation Planning Project.

Exploratory Research/Field Tests: Advanced instruments are being developed with new capabilities that will significantly advance the state of the art in accounting and control. A focused, innovative, engineering-driven 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. As the technical readiness level of these technologies increases, we are planning and executing field tests in fuel cycle facilities to obtain operational experience and demonstrate their effectiveness.

Advanced integration and analysis of data streams at the process, facility, and fuel cycle levels requires new methods to understand and propagate uncertainties, including correlations, for significantly enhanced system performance.

Fiscal Year-End 2015 Funding (Includes Carryover)

Material Protection, Accounting, & Control Technologies Campaign		
Major Activities	FY-15 Funding	
Campaign Management, Integration, Technical Support	\$837,216	
Safeguards and Security by Design - Electrochemical	\$2,334,608	
Used Fuel Extended Storage	\$1,864,881	
Exploratory Research/Field Tests	\$1,114,315	
Total	\$6,151,020	

Major Accomplishments

Electrochemical Process Monitoring for Enhanced Safeguards advanced process monitoring instruments (level/density, voltammetry, and actinide) are being developed for electrochemical processing as part of the safeguards and security by design effort. Actinide sensor development went beyond surrogate testing this year, with initial ion exchange runs being completed with plutonium. We also began qualification of the level/density and voltammetry sensors with field tests planned in concert with the Joint Fuel Cycle Study. In addition, a micro-analytic sampling system is being developed using a droplet generator and spectroscopic characterization was completed using strontium-doped molten salt. Finally, a hybrid statistical method has been developed that incorporates process monitoring data in combination with traditional nuclear material accountancy data for diversion detection.

Modeling and Simulation for Electrochemical Processing Safeguards advanced radiation transport calculations (using MCNP coupled to application-specific algorithms) have been performed for the PEER and Mark-IV electrorefiners to identify signatures for advanced monitoring instrumentation development. A parametric study was conducted with variable initial enrichments, burnups, and cooling times. Results of the study indicate that neutron and delayed gamma signatures may be exploitable to monitor process parameters. Additional fidelity in radiation transport simulations can be enabled by mass flow models under development using chemical process models that include dynamics. The Dynamic Electrorefiner (DyER) code is currently being developed for this purpose. At the facility level, we continued to develop the Safeguards and Security Performance Model (SSPM), incorporating results from the other modeling activities, including the hybrid statistical model (details follow in the next section).

Security Evaluations of Used Nuclear Fuel in Extended Storage focused on a preliminary evaluation of a generic pilot storage facility design from NFST, continued numerical investigation of the spent fuel ratio, consequence analysis for dry cask sabotage, and dynamic self-protection assessment capability. The evaluation of the generic pilot storage facility included sensor and security perimeter design, simulated force-on-force assessments, and consequence analyses from successful sabotage events and is documented in a classified report. Results of the numerical evaluation of the spent fuel ratio support a ratio closer to 1 rather than the currently adopted 3 and were presented at the International High Level Radioactive Waste Management conference. We continue to hold regular technical exchange meetings with the NRC on this topic as well.

Advanced Neutron and Gamma-Ray Instrumentation are being developed to support the broad range of instrumentation needed to support next generation nuclear materials management for the back end of the fuel cycle as well as for new advanced fuel cycle facilities.

- A dry cask monitoring system is being developed based on neutron signatures as developed by MCNP and by the UNF ST&DARDS database (which documents actual spent fuel cask inventories in the U.S.) This system shows promise for application to shipper-receiver checks during transport, cask integrity monitoring at the storage site, and recovery of Continuity of Knowledge based on a ratio method that eliminates the effect of source decay. Laboratory benchmark testing is expected to begin in FY-16.
- Development of the super-high-resolution gamma spectrometer based on microcalorimetry continued in FY-15, where the systematics of total measurement uncertainty are beginning to be uncovered. This investigation showed that the key advantage of increased resolution can in principle lead to an improvement in determination of plutonium isotopics by a factor of 10-60, thereby showing promise to break the current 1% uncertainty barrier. In addition, the team has begun to investigate the application of a microwave multiplexing scheme that would enable much higher count rates in practical application of the instrument.
- A neutron counter that can withstand very high gamma dose is under development using a ¹⁰Blined parallel plate technology originally developed for replacement of the IAEA standard High Level Neutron Coincidence Counter. In addition to adaptation to high-dose applications, this technology allows for the extraction of average neutron energy, important for complex sample matrix applications such as assay of the product ingot from electrochemical processing.
- The Multi-Isotope Process (MIP) Monitor has begun a series of field tests at the Savannah River H-Canyon in FY-15, with encouraging initial results. Of particular interest was the observation of MOX source tank mixing and the ability of the MIP Monitor to distinguish between the original solutions and the final mix, using gamma-ray based spectral principle components analysis (details follow in the next section).

8.2 The Multi-Isotope Process (MIP) Monitor Deployment Project at H-Canyon

D. Meier, A. Casella, D. Abrecht, Pacific Northwest National Laboratory L. Sexton, Savannah River National Laboratory J. Coble, N. Shoman, University of Tennessee-Knoxville

Introduction and Objectives

The Multi-Isotope Process (MIP) Monitor was developed by a collaboration of researchers at PNNL, The Ohio State University, UT-Austin, and Penn State. The MIP Monitor measures spectral distributions of certain radionuclides present within feed, product and waste streams of a nuclear recycling or separation facility. These radionuclides are monitored online by gamma spectrometry and compared, in near-real-time (NRT), to spectral patterns representing "normal" process conditions using multivariate analysis. In addition to identifying "off-normal" conditions, multivariate regression techniques can also be used with the MIP Monitor to quantify major process conditions simultaneously in NRT. By focusing on gamma-emitting radionuclides, the MIP Monitor approach is compatible with the use of small, portable, relatively high-resolution gamma detectors that may be deployed throughout an existing nuclear facility. The objective for the current work will be to deploy a MIP Monitor detection system in H-Canyon nuclear separation facility near Savannah River National Laboratory (SRNL) during their processing campaigns. The spectra obtained will be processed using principal component analysis and other regression techniques.

R&D Overview

The integration of online, NRT process monitoring has the potential to enhance the efficiency and accuracy by which MC&A and process controls are currently applied at nuclear facilities. Current accountancy techniques rely heavily on Thermal Ionization Mass Spectrometry (TIMS) for analyses of special nuclear materials. While highly accurate and precise, time intensive TIMS measurements require extensive preprocessing by skilled staff. In the time it takes to generate these analytical results, significant quantities of SNM could be diverted within a large recycling facility. In addition, errors associated with these results, when used for material balance purposes, scale with the size of the batches. Without augmenting current DA-based MC&A approaches, the propagation of such errors might ultimately limit the size of a material balance area or even the overall size of a recycling facility. However, online, NRT, process monitoring technologies, when applied in concert with traditional DA techniques, may have the ability to significantly reduce required resources and improve the timeliness of MC&A at recycling and separation facilities. Also, since the MIP Monitor has the ability to provide enhanced process information in NRT, this technique could improve the manner in which a process is controlled, resulting in enhanced system efficiency and effectiveness for the operator.

Accomplishments

This year's primary accomplishment was developing a feasibility test plan for the deployment of the MIP Monitor detection system in the H-Canyon reprocessing plant at SRNL while gathering preliminary data and developing methods to process the raw data though preprocessing and chemometric algorithms. This test plan provides details for many aspects of the deployment effort including, deployment locations, detector specifications, environmental considerations, as well as facility limitations and opportunities. It also identifies the need for deployable detection systems that are rugged, stable, reliable and can operate

with a minimum of maintenance. The primary detector that is being used are NaI(Tl) (nominally 6-7% keV FWHM at 661 keV) detectors. Lanthanum bromide detectors-LaBr3 (nominally 2.8% keV FWHM at 661 keV) are also being used for benchmarking and calibration purposes. The data is gathered using the Maestro software package in list data acquisition mode. Also, an adjustable test rig is being developed in parallel that will not only hold and geometrically adjust the detection systems but also allow for the incorporation of either collimating or shielding materials as needed.

Another important accomplishment has been the engagement of the Savannah River Site (SRS) staff and the development of a deployment team that consists of Savannah River National Lab (SRNL), SRS and PNNL staff. Consequently of this engagement, PNNL staff are now allowed to perform hands-on work in H-Canyon facility with radiological and facility oversight from SRNL and the SRS team. The project has also integrated H-canyon operations management with the deployment planning discussions. This relationship has proven invaluable as it provides a behind the scenes view of the operational schedule of the H-Canyon facility and also has provided the hidden benefit of identifying other targets of opportunity that may not be normally available.

The chemometric analysis team at the University of Tennessee-Knoxville conducts both the post processing of the data and the chemometric analysis of the acquired spectra. Figure 38 is an example of raw list mode data captured (intensity vs channel as a function of time). Figure 39 is an example of the PCA analysis of two separate MOX source tanks blending into a third product tank.



Figure 38. Raw list mode data of a plutonium tank.

Figure 39. PCA analysis of the blending of two different MOX tanks.

This effort is well positioned to support the MPACT 2020 milestone that is developing laboratory-scale demonstrations of advanced safeguards and security systems. The flexibility of the MIP monitor combined with the support of H-Canyon is a unique capability that is well positioned to help achieve MPACT goals.
8.3 Modeling and Simulation for Analysis of Safeguards Performance

B. Cipiti, Sandia National Laboratories

Introduction and Objectives

The purpose of the modeling and simulation for safeguards performance work is to help guide the safeguards design of future electrochemical reprocessing facilities. Electrochemical plants require new approaches for safeguards due to the unique material form and processing environment as compared to aqueous plants. Process monitoring technologies were examined as part of the safeguards and security design, and a diversion scenario analysis was completed.

R&D Overview

An analysis of the unit operations was performed to examine nonsampling process monitoring measurements that may be useful for safeguards. The technologies that were found to be research priorities include salt/level measurements of the electrorefiner, voltammetry or actinide sensor technologies for oxide reduction and electrorefiner monitoring, current and voltage monitoring in the electrorefiner, and bulk mass measurements throughout the plant.

SSPM was used to develop and test the preliminary safeguards design (Figure 40). This was a culmination of past work on the design of the accountancy system along with the integration of the process monitoring measurements. The design is based around four key measurement points (KMPs) including the input spent fuel, electrorefiner salt inventory, U product, and U/TRU product measurements. Additional measurements will also be required for confirmatory measurements or for areas processing very low actinide quantities, but these measurements do not drive the overall measurement error.

A parametric diversion scenario analysis was completed based on the safeguards system design. Both IAEA and NRC regulations were used as a basis for the results. A variety of abrupt and protracted diversions were examined to help define the measurement uncertainty goals for the four KMPs. These results are being used to help guide the design and placement of the technologies being examined for electrochemical safeguards.

The integration of materials accountancy and process monitoring data with physical protection was also examined in order to help protect against insider diversion scenarios. Both the SSPM and STAGE software were used to evaluate this integration. Multiple scenarios were examined that included removal of fractions of the U/TRU product and subsequent removal from the facility, with a goal of removing 8 kg of Pu in total. Results showed significant improvements in the ability to stop theft scenarios by integrating the accountancy and process monitoring data.

Finally, a subtask to evaluate an alternative input accountancy measurement was finalized. Previous work had identified the need for alternative input accountancy options for electrochemical plants. Modeling was used to evaluate active neutron interrogation and delayed gamma assay of shredded fuel in baskets going into the process. The work this year found that the counting statistics would be too poor assuming a 10^{10} n/s D-D source, and increasing the source strength to the levels required would require a device that is not practical for use in this application.

The modeling and simulation work will fit into the MPACT 2020 milestone to provide a distributed test bed for safeguards systems and technologies. The SSPM along with other modeling capabilities will be used to help tie together the measurement technologies and test beds which are applicable to the back end of the fuel cycle.



Figure 40. Electrochemical safeguards system design.

Fuel Cycle Options Campaign



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9. FUEL CYCLE OPTIONS CAMPAIGN

9.1 Overview

R. Wigeland, Idaho National Laboratory

Mission

The Fuel Cycle Options (FCO) Campaign mission is to develop and implement analysis processes and tools and perform integrated fuel cycle evaluations to provide information that can be used to objectively and transparently inform DOE-NE as decisions are made about overall R&D directions and to integrate Office of Fuel Cycle Technologies activities through R&D efforts on common fuel cycle goals. An illustration of how FCO informs the DOE-NE decision-making process is shown in Figure 41 for the Nuclear Fuel Cycle Evaluation and Screening (E&S) Study.



Figure 41. Nuclear Fuel Cycle Evaluation and Screening Study provides input to the DOE decision-making process.

Objectives

- Develop understanding of the challenges and benefits associated with current and alternative nuclear fuel cycles, including quantifying the potential for improving fuel cycle performance, identifying and studying the issues associated with transition and deployment, and exploring options for managing fuel cycle issues by performing steady-state and dynamic analyses of complete integrated fuel cycles (mining through disposal), including consideration of specific existing and developing technologies, accident-tolerant fuels, specific fuel cycle concepts, and various deployment strategies including the use of modular facilities.
- Develop understanding of the potential role of nuclear power in the domestic and global energy market considering alternative nuclear fuel cycles and the effects of economics and other external issues such as CO₂ management and identifying what aspects of fuel cycles can influence the domestic and global use of nuclear power by analyzing possible future nuclear power deployment including using market-driven scenarios.

- Develop improved fuel cycle economic analysis capability, providing results of greater credibility mainly by analyzing historical economic data to inform on projected costs of reactors and other fuel cycle facilities, increasing modeling capabilities, and continuing improvement of supporting economic data.
- Contribute to a broader understanding of the required characteristics of the promising fuel cycles capable of providing substantial improvements with respect to the current U.S. fuel cycle by communicating the results of the Nuclear Fuel Cycle Evaluation and Screening Study both within the FCT program and outside the program, and developing and improving communication tools such as the online Fuel Cycle Catalog to foster understanding of nuclear fuel cycles, their capabilities, and their limitations.
- Develop and manage processes and data for identifying sustainable alternative fuel cycle options for informing R&D prioritization, including documentation that supports transparent decision making for R&D investments.

Challenges

- Development of the range of deployment and transition options that can affect the attractiveness of alternative nuclear fuel cycles and identification of the relevant performance parameters.
- Development of approaches to address issues that arose during the Evaluation and Screening study, including methods and data required to improve estimates of fuel cycle economics and financial risks, and studies of the overall viability of nuclear power both domestically and globally.
- Development of effective means to communicate the information on the promising fuel cycle options that promotes understanding of the reasons why only certain options are promising and what are the potential benefits, and communicating all aspects of nuclear fuel cycle capabilities and limitations to a variety of stakeholders.
- Identification of areas of analysis and other activities where the Fuel Cycle Options campaign can inform DOE/NE and other stakeholders on alternative fuel cycles and technology options.

Major R&D Activities

In FY-15, the campaign performed analyses in three general areas, mainly based on the results from the Nuclear Fuel Cycle E&S study that identified promising fuel cycle options which could substantially improve nuclear fuel cycle performance as compared to the current U.S. fuel cycle.

Integrated Fuel Cycle Analysis. In this area, perform analyses of promising fuel cycles from the E&S study to improve the understanding of how specific sets of technology options function as a system including working with other campaigns to consider the effects of a range of specific implementing technologies for all parts of the fuel cycle to identify any interactions (favorable or otherwise), e.g., separations, waste forms, storage and disposal technologies. Specific activities included:

- Issue the final report of the E&S study and the associated computer-based decision-analysis software.
- Complete analyses of the promising Evaluation Groups to narrow the range of fuel cycle options that would be considered promising.

- Refine and enhance the approach and the information assessment of economics and financial risk for future fully-developed fuel cycles.
- Analyze the most promising options from the E&S study to identify and quantify the benefits of minor actinide recycle and the associated challenges.
- Analyze the promising thorium-based fuel cycle options from the E&S study to further quantify benefits and challenges of using thorium.
- Analyze the effects of using modular facilities such as SMRs, distributed fuel fabrication, used fuel processing, and disposal to explore the effects of fuel cycle facility modularization.
- Analyze the effects of using extended storage prior to irradiation of recycle fuel, prior to used fuel processing, prior to fuel fabrication, and prior to disposal to explore the effects of extended storage within the fuel cycle
- Analyze the economics of using modular fuel cycle facilities.

Development and Deployment Issues. In this area, perform analyses of transition from the current U.S. fuel cycle to an alternative fuel cycle, considering deployment and implementation options, economics, and impact of modular facilities to inform on choices, decision timing, and costs.

- Continue investigation into the numerous issues concerning the ability to perform economics analysis, including supporting data, interdependence of cost estimates between facilities, uncertainties (for an estimate, among multiple estimates, experience vs. variation, etc.), and identification of what is needed to support credible fuel cycle cost estimates and evaluation of financial risk.
- Analyses of transition to the most promising fuel cycles, including estimates of yearly costs utilizing the best-informed cost and financial data from previous year's activities in the campaign.
- Analyses of U.S. regional reactor retirement and electricity demand to explore the interaction of regional reactor retirement schedule, projected energy demand, and the potential deployment of LWRs or a transition to an alternative fuel cycle in the context of other energy sources.
- Analyze transition from a uranium-based fuel cycle to a thorium-based fuel cycle, recognizing that thorium needs a separate supply of fissile material to start up.
- Analyses of transition to the most promising fuel cycles including the effects of using modular facilities such as SMRs, distributed fuel fabrication and used fuel processing, and disposal to explore the effects of choices, decision timing, and costs when using modular fuel cycle facilities.
- Analyze the global impacts of nuclear energy on reducing greenhouse gas emissions.

Fuel Cycle Catalog and Emerging Issues. Further development of the public online Fuel Cycle Catalog for fuel cycle information, which was made operational in FY-14, but maintenance, improvement, and expansion are needed.

• Maintain publicly-available fuel cycle catalog to disseminate fuel cycle information, including further development of the software supporting the Catalog such as additional interactive capabilities.

• Increase the range of fuel cycles and supporting technology information in the catalog, including the results of ongoing analyses, and filling gaps in information.

Fiscal Year-End 2015 Funding (Includes Carryover)

Fuel Cycle Options Campaign		
Major Activities	FY-15 Funding	
Campaign Management	\$ 858,365	
Integrated Fuel Cycle Analysis	\$4,841,464	
Development and Deployment Issues	\$4,382,395	
Fuel Cycle Catalog and Emerging Fuel Cycle Issues	\$466,001	
Total	\$10,548,225	

Major Accomplishments

Issued the final report, "Nuclear Fuel Cycle Evaluation and Screening Report," and the supporting decision-analysis software, "Screening and Evaluation Tool (SET)," available on the INL website as follows:

https://inlportal.inl.gov/portal/server.pt/community/nuclear_science_and_technology/337/nuclear_fuel_cycle_evaluation_and_screening_final_report

- Completed the investigation of the fuel cycle groups contained in the promising Evaluation Groups (collections from the Nuclear Fuel Cycle Evaluation and Screening study to reduce the number of potentially promising fuel cycle groups for R&D, and issued the report "Analysis of the Promising Evaluation Groups."
- Completed a review of LWR and reprocessing facility costs, including construction, operation, and maintenance, to provide a basis for estimating facility costs in general, developed new data for sodium-cooled fast reactors, and summarized in the report "Economics Evaluation of the Promising Options."
- Completed a study on the relative challenges and benefits for the most promising fuel cycle options from the Nuclear Fuel Cycle Evaluation and Screening, U/Pu and U/TRU recycle in fast reactors, considering effects on implementation and deployment, leading to the conclusion that R&D on both approaches should be supported at this time.
- Completed a study on the impact of ATF on the performance of once-through LWRs ("basis of comparison" for the E&S study), supporting to the planned prioritization of ATF concepts, with the analyses and results documented in the report "Fuel Cycle Impacts of Accident Tolerant Fuel."
- Issued the report, "Transition to an Alternative Uranium-Based Fuel Cycle Choices, Timing, and Costs," that documents the analyses of transition to a fast reactor recycle fuel cycle, exploring the general effects of deployment choices and timing and identifying some potentially viable transition approaches, but is likely to occur over many decades due to the effects of facility lifetime and timing of technology availability.

- Performed market analyses of nuclear power in the U.S. for scenarios with and without a carbon tax, with the difference in the nuclear share of the electricity market between the once-through and recycle scenarios driven by the higher cost of fuel for the SFRs. For the LWR-to-SFR transition, the system was constrained by fuel availability, with results documented in the report "Regional Reactor Retirement and Electricity Demand Transition and Overall Energy Supply."
- Implemented market analyses using regional-scale representation of the U.S. to better characterize the overall U.S. energy use and electricity generation in particular, enabling simulation of nuclear energy deployment with respect to other energy sources and the assessment of nuclear fuel cycle issues at the state and regional scale, with the analyses and result reported in "Simulations of Regional US Electricity Generation and Nuclear Power Deployment."

9.2 Fuel Cycle Transition Analysis: Evaluating the Transition from Current U.S. Nuclear Energy System to Fast Reactor Continuous Recycle System

E. Hoffman, Argonne National Laboratory

Introduction and Objectives

The Nuclear Fuel Cycle E&S Report¹ provides the results of the E&S study. Four of the most promising options were identified as well as other potentially promising options. The E&S study considered performance at steady-state when evaluating the potential benefits and identified a set of challenges (Development and Deployment Risk, Institutional Issues, and Financial Risk and Economics) to achieving that eventual end state.

Following the completion of the E&S study, the FCO Campaign initiated analyses of the transition to alternative fuel cycles, particularly those identified as most promising. The objective of the transition analysis is to help inform decisions about choices that can be made that affect transition, how long transition might take, and what the performance of the overall nuclear enterprise would be during the transition from one fuel cycle to another.

This paper summarizes work completed in support of the transition analysis, including (1) developing the approach and methodology for extending the evaluation of the fuel cycle options over the period of transition from the current state of the U.S. fuel cycle to new eventual steady state system; (2) applying that method to a potential transition scenario involving one of the most promising options identified by the E&S study; (3) identifying issues and challenges associated with such transition; and (4) verifying that at the end of transition the system is performing as expected based on the E&S study. Other related efforts included evaluating the impact on transition of using modular fuel cycle technologies and developing approaches and methods for evaluating the economics during transition.

R&D Overview

Fuel Cycle Modeling and Analysis. Much of the effort has focused on improving modeling capability, with significant developments in expanding our ability to evaluate a particular transition scenario in areas of potential interest to decision makers (e.g., LLW source and quantity generated by year, annual expenditures by source). This analysis capability will allow decision-makers to optimize transition performance based on their value judgments. This ability to look at the dynamic behavior of an expanded set of metrics will increase the understanding of transition to an alternative fuel cycle option.

The transition analysis extended the basic approach developed in the E&S study to include metrics appropriate for transition, including dependencies on time, fleet size, and reactor/fuel cycle technology to quantitatively analyze the projected behavior of the U.S. nuclear fuel cycle system. The E&S study was chartered with a set of nine Evaluation Criteria, six related to benefits and three related to challenges, and within each criterion, developed a set of metrics for evaluating the technology-neutral (physics driven) performance and challenges towards getting to first-of-a-kind deployment. All metrics considered in the E&S study were considered for inclusion in the expanded transition analysis and they were included if there was any time-dependent behavior of potential interest (even if there is no transition to an alternative fuel cycle option), if there was a significant change in performance between the initial system or final

system, or any reason that the time-dependent behavior may be of interest. This was the primary focus of the work completed in FY-15. Utilizing this work to inform on the transition to alternative fuel cycle options is the focus of the work in FY-16.

Example Transition. An *Example Scenario* was defined assuming U.S. nuclear energy production growing at an annual rate of 1%. The transition analysis modeling starts at 2015 and extends to 2200. This growth rate is thought to be representative of nuclear energy roughly maintaining its current market share of electricity based on projection of current electricity demand growth rates. The *Example Scenario* assumes that existing inventories of UNF generated prior to 2015 will be disposed as SNF (SNF), but all UNF generated in 2015 and after will be available for recycle. The *Example Scenario* also includes defining when each reactor in the current fleet is expected to shut down. The assumed lifetime of the existing fleet was that in terms of energy generation, half would operate for 60 years and half would have their lifetime extended to 80 years.

The set of assumptions that defines the specific transition pathway for the *Example Scenario* is referred to as the *Base Case*. The *Base Case* assumes that transition will be to a fuel cycle in Evaluation Group EG23 (Continuous recycle of U/Pu with new natural-U fuel in fast critical reactors), one of the most promising fuel cycle groups identified by the E&S study. The key assumptions are that the end-state technologies are sodium-cooled fast reactors (SFRs), offsite aqueous reprocessing of UNF (no separated Pu), and glove-box fabrication of the recycled metal fuel. The SFRs are assumed to be available for commercial deployment beginning in 2050. Prior to that, all demand for new or replacement generation to maintain the 1% growth rate is met by constructing new light-water reactors (LWRs) with lifetimes of 80 years. All LWR UNF produced beginning in 2015 is recycled and used for deployment of the SFR fleet.

A *Basis of Comparison* was defined to understand the behavior of the *Example Scenario* without a transition to a different fuel cycle and to quantify the impacts of transition. The *Basis of Comparison* assumed that the growing energy demand continues to be met by new LWRs operating with the same assumptions as the *Base Case*.

Selected Results. A credible transition path was developed for the *Base Case* to illustrate transition from the current U.S. fuel cycle to a fuel cycle using sodium-cooled fast reactors with continuous recycle of U/Pu, meaning that sufficient time would be available for all technologies to be developed and deployed on the timescales needed and that the material necessary to operate the system was available. Figure 42 shows the annual electricity generation by reactor type for the *Base Case*. For the *Basis of Comparison*, the electricity generation would be the same except that all of the electricity produced by SFRs in the *Base Case* would be produced by new replacement LWRs instead. Two types of SFRs were used to model the transition: the first type deployed (SFR-A) has a higher breeding ratio to enable and expedite transition. The second (SFR-B) has a breeding ratio that is set to sustain operations at the 1% growth rate without producing excess materials.



Figure 42. Annual electricity generation by source for the base case.

The E&S study found a significant reduction in the Natural Uranium Required per Energy Generated when comparing EG23 and the current U.S. fuel cycle. The current transition analysis found that not only does the normalized (per Energy Generated) value matter, but the cumulative demand is also likely to be an important measure of performance. Figure 43 shows the cumulative natural uranium required. By 2200, the *Basis of Comparison* in the U.S. will require in excess of the 2014 Red Book ² world identified resources, which suggests potential future supply issues. In contrast, the *Base Case* will require approximately 40% of the world's uranium resources identified today, which is a very large improvement, but does not clearly resolve the potential for a future supply issue even with the benefits of transition to a fuel cycle that makes much better use of fuel resources. Significant reduction in the cumulative need for uranium resources for the *Base Case* appears unlikely because it is driven by:(1) the need to build LWRs until the SFRs can be deployed independently and (2) the continued operation of these LWRs until the end of their lifetimes.



Figure 43. Comparison of cumulative natural uranium required.

The analysis also indicates that the system would take well into the next century before the system is performing near equilibrium conditions for this metric due to the persistence of a large LWR fleet.

The Volume of LLW per Energy Generated was another metric considered by the E&S study under the Nuclear Waste Management Criteria. While no significant difference was identified between the current

U.S. fuel cycle and EG23 by the E&S study, due to the large contribution of decontamination and decommissioning wastes to the overall LLW production for each facility's life cycle, fuel cycles in EG23 were evaluated as having somewhat higher LLW generation, and the transition analysis shows that there is dynamic behavior that could have implications for system design and public policy. Even for the *Basis of Comparison*, there will be an increase of about a factor of 3 in the annual rate of LLW generated when the existing LWRs reach the point where a significant fraction are being decommissioned and decontaminated (D&D), at around 2045. The *Base Case* results in a generally higher annual rate of all sources of LLW and a significant variation over time in the types and volumes of LLW generated (Figure 44).



Figure 44. Total LLW generation rate in basis of comparison and base case.

These types of time-dependent analyses were applied to all of the metrics covering a wide range of areas of potential interest. The cause of these results can be traced back to the source to understand what the driving function is, whether an assumption or an aspect of the fuel cycle transition.

Accomplishments

The evaluation of fuel cycle transition identified and considered potentially important parameters and evaluated them to inform on the performance of the overall fuel cycle during transition. This will allow informing on the many questions related to how long it takes to deploy the new system, how long it takes to achieve the anticipated benefits, how to minimize costs, how to maximize benefits, and an identify a wide variety of potential issues and address more complex questions related to understanding the deployment of a particular fuel cycle option.

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9.3 The Potential of Nuclear Energy for Addressing Climate Change

S. H. Kim, Pacific Northwest National Laboratory

Introduction and Objectives

Addressing climate change to "avoid dangerous human interference with the climate" implies changes to the global energy system of unprecedented scale. Limiting global warming to 2° C, for instance, requires the stabilization of atmospheric greenhouse gases (GHG) to approximately 450 ppm CO₂-equvalent and the near complete reduction of CO₂ emissions by the end of the century. The current US and global energy system is dominated by fossil fuel use with associated GHG emissions.

There is greater interest in the expanded use of nuclear energy for addressing climate change since nuclear energy already contributes to the global energy system without directly contributing to CO_2 or other GHG emissions. Increasing nuclear energy use in response to climate change, however, implies an increase in the number and distribution of nuclear reactors worldwide. Furthermore, minimizing the burden of addressing climate change implies that nuclear energy must be competitive with other carbon-free energy forms. The future of expanded nuclear energy use imposes additional demands on all aspects of the nuclear energy system and challenges for the sustainability of the nuclear fuel cycle.

The potential role of nuclear power in the US and globe is explored within the context of the broader energy system, and alternative promising nuclear fuel cycles and advanced reactor technologies under investigation by the FCO Campaign. The PNNL's Global Change Assessment Model (GCAM) is utilized to simulate alternative long-term futures of domestic and global economic growth, energy use, and CO₂ emissions, and to assess the nuclear energy contribution to domestic and global energy market and for mitigating climate change.

R&D Overview

PNNL's GCAM model is an "integrated assessment model" that has been widely utilized over the past several decades to understand the relationships between human activities and the Earth system in the context of integrated agriculture, energy, water, and climate change analysis. GCAM includes representations of the economic, energy, agricultural, land use, and climate systems for 32 global regions, including the US, and provides simulations from 1990 to 2100 in 5-year time steps. GCAM has detailed technological representation of the overall energy system, and the nuclear energy system in particular. The representation of the nuclear fuel cycle and reactor technologies for power generation has been developed in GCAM to simulate alternative promising fuel cycles and advanced reactor technologies under investigation by the FCO Campaign within the overall energy and carbon emissions context.

For FY-15, a version of GCAM that further disaggregates the US into 50 states, including the fuel cycle and reactor technology representation at the state-level, has been developed and employed for assessing the regional potential of nuclear energy. Data developed by the FCO activities are inputs to the GCAM model. GCAM scenarios utilize information and data on nuclear fuel cycle and reactor characteristics and costs that are consistent with the FCO Campaign.

Alternative carbon mitigation scenarios and sensitivity studies of nuclear energy technologies were generated and the potential of nuclear energy with and without climate mitigation policies were explored. Scenario results provided potential range of nuclear energy deployment and associated demands for natural uranium and generation of nuclear wastes. CO₂ emissions differences with and without nuclear

power deployment allowed the assessment of the potential value of nuclear energy in addressing climate change.

Summary of Achievements

Simulations of US and global electricity generation show that nuclear energy remains an important component of the expanding global energy system. As shown in Figure 45, nuclear energy contribution in the US maintains its current share in the long-term. Near-term contribution of nuclear energy in the US decreases as a result of retirement of legacy reactors and the competitiveness of power generation from natural gas and wind. Global demand for electricity outpaces the growth in the US and the global demand for nuclear energy increases significantly by the end of the 21st century.

Regional simulations of the US energy system indicate the potential contribution of nuclear energy by state, as displayed in Figure 46. Population and economic growth and the demand for electricity by state, and the availability and cost of competing power options drive the nuclear energy deployment by state.



Figure 45. Electricity Generation by Fuel for the US and Globe (EJ/yr).



Figure 46. US nuclear electricity generation by state (EJ/yr).

\$50 PPM

550 POM

650 PPM

The contribution of global nuclear energy use to CO_2 emissions reduction under global carbon mitigation policies shows that nuclear energy has tremendous value as a carbon mitigation option. Changes to the global nuclear power capacity are shown in Figure 47. The global value of nuclear energy in addressing climate change is measured in trillions of US dollars. The value of nuclear energy is significantly greater when fewer carbon-free technology options, such as fossil Carbon Capture & Storage (CCS), are available and climate mitigation targets become increasingly stringent (Figure 48).

12

10

8

6

4

2

0

without CCS





Figure 48. Value of nuclear energy under alternative climate mitigation policies.

withCCS

Value of Nuclear

Appendix A: Acronyms



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10. APPENDIX A: ACRONYMS

AAR	Association of American Railroads
ACWF	advanced ceramic waste form
AFC	Advanced Fuels Campaign
AFS	AREVA Federal Services
APT	atom probe tomography
ATF	accident-tolerant fuel
ATR	Advanced Test Reactor
BNL	Brookhaven National Laboratory
BWR	boiling-water reactor
CCIM	Cold Crucible Induction Melter
CEC	cation exchange capacity
CECE	Combined Electrolysis and Catalytic Exchange
CURIE	Centralized Used Fuel Resource for Information Exchange
DFC	damaged fuel can
DOE	Department of Energy
DOE-NE	DOE Office of Nuclear Energy
DPC	dual-purpose canister
DSC	dry storage cask
E&S	evaluation and screening [Study]
EPRI	Electric Power Research Institute
ERWG	Electrochemical Recycling Working Group [JFCS]
ESA	Execution Strategy Analysis
EURATOM	European Atomic Energy Community
FCAWG	Fuel Cycle Alternatives Working Group
FCCI	fuel-cladding chemical interaction
FCO	fuel cycle options
FCRD	Fuel Cycle Research and Development
FCT	Fuel Cycle Technologies
FFP	firm fixed price [Contract]
FOA	Funding Opportunity Announcement [Industry-Led Teams]
FTIR	Fourier Transform Infrared [Spectroscopy]
GNEP	Global Nuclear Energy Program
GTCC	Greater Than Class C
GTS	Grimsel Test Site
HDO	deuterated water

HFEF	Hot Fuel Examination Facility
HFIR	High Flux Isotope Reactor
HLW	high-level waste
HLRM	high-level radioactive material
HRL	Äspö Hard Rock Laboratory
НТО	Tritiated Water
IAEA	International Atomic Energy Agency
INL	Idaho National Laboratory
IRT	Integrated Recycling Test
ISF	Interim Storage Facility
IRP	Integrated Research Project [University-Led Teams]
IVEM	Intermediate Voltage Electron Microscopy [Tandem Facility]
JAEA	Japan Atomic Energy Agency
JFCS	Joint Fuel Cycle Studies
KAERI	Korean Atomic Energy Research Institute
KMP	Key Measurement Point
LANL	Los Alamos National Laboratory
LAR	licensing amendment request
LBNL	Lawrence Berkeley National Laboratory
LIBS	laser induced breakdown spectroscopy
LLW	low-level waste
LOCA	loss of coolant accident
LTR	lead test rod
LWR	light-water reactor
MFC	Materials and Fuels Complex [INL]
MOEF	multi-objective evaluation framework
MOF	metal-organic-framework
M&S	modeling and simulation
MGR	monitored geological repository
MPACT	Materials Protection, Accounting and Control Technology
MPC	multi-purpose canister
MRWFD	Material Recovery and Waste Form Development [Campaign]
MSL	Marine Sciences Laboratory
NCT	Normal Conditions of Transport [Shock]
NEAMS	Nuclear Energy Advanced Modeling and Simulation
NEUP	Nuclear Energy University Program
NFST	Nuclear Fuels Storage and Transportation (Project)
NGSAM	Next-Generation Waste Management Systems Analysis Model
NWPA	Nuclear Waste Policy Act

NRC	Nuclear Regulatory Commission
O/M	Oxygen-To-Metal Ratio
OECD/NEA	Organization for Economic Cooperation and Development /Nuclear Energy Agency
OFF	oldest fuel first
ORNL	Oak Ridge National Laboratory
PIE	Postirradiation Examination
PNNL	Pacific Northwest National Laboratory
PWR	pressurized-water reactor
QTR	Quadrennial Technology Review
R&D	Research & Development
RD&D	Research, Development, and Demonstration
RDD&D	Research, Development, Demonstration, and Deployment
RFI	Request for Information
RFP	Request for Proposal
RIGP	Radiation Induced Graft Polymerization
SACSESS	Safety of Actinide Separation Processes
SED	Siting Experience Database
SEM	Scanning Electron Microscopy
SET	Screening and Evaluation Tool
SNF	spent nuclear fuel
SFR	Sodium-Cooled Fast Reactor
SOW	scope of work
SRNL	Savannah River National Laboratory
SRS	Savannah River Site
SSPM	Safeguards and Security Performance
SSWG	Safeguards and Security Working Group
STAD	Standardized Transportation, Aging, and Disposal [Canister]
START	Stakeholder Tool for Assessing Radioactive Transportation
TREAT	Transient Reactor Test [Facility]
TRU	transuranic
TSL	Transportation and Storage Logistics
UFD	used fuel disposition
UNF	used nuclear fuel
UNF-ST&DARDS	Used Nuclear Fuel Storage, Transportation & Disposal Analysis Resource And Data System
URL	Underground Research Laboratory
XAFS	X-Ray Absorption Fine Structure [Spectroscopy]