2012 Fuel Cycle Technologies Annual Review Meeting

Transactions Report

Argonne National Laboratory

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Fuel Cycle Technologies Program: Introduction

As a large domestic source of clean, nearly greenhouse gas-free energy, nuclear power is making major contributions toward meeting our nation's current and future energy demands. The United States must continue to ensure improvements and access to this technology so we can meet our economic, environmental and energy security goals. We rely on nuclear energy because it provides a consistent, reliable and stable source of base load electricity with an excellent safety record in the United States. In order to continue or expand the role for nuclear power in our long-term energy platform, the United States must:

- Continually improve the safety and security of nuclear energy and its associated technologies worldwide.
- Develop solutions for the transportation, storage, and long-term disposal of used nuclear fuel and associated wastes.
- Enhance the resilience of nuclear plants and used nuclear fuel in storage to extreme events and beyond-design-basis accidents such as Fukushima–Daiichi.
- Improve the long-term sustainability of nuclear energy.

To address these high-level goals, the Fuel Cycle Technologies (FCT) program of the Department of Energy (DOE) Office of Nuclear Energy (NE) is charged with identifying promising sustainable fuel cycles and developing strategies for effective disposition of used fuel and high-level nuclear waste, enabling policymakers to make informed decisions about these critical issues. Sustainable fuel cycles will improve uranium resource utilization, maximize energy generation while minimizing waste, improve safety, and limit proliferation risk.

To effectively accomplish its mission, FCT invested nearly $112 million in FY 2012 for parallel and complementary research and development (R&D) in five technical campaign areas that span the entire nuclear fuel cycle:

- **The Fuel Cycle Options Campaign** is developing management processes and tools and performing integrated fuel cycle technical assessments to provide information that can be used to objectively and transparently inform and integrate Office of Fuel Cycle Technologies activities, guiding the selection of sustainable options.
- **The Advanced Fuels Campaign** is developing proliferation-resistant, next-generation metallic fuels for recycling of transuranics, along with advanced accident-tolerant fuel for current light water reactors.
- **The Separations, Waste Forms, and Fuel Resources Campaign** contributes to both a sustainable fuel cycle and improved waste management by effectively separating transuranic

elements from used nuclear fuel and seeking transformational breakthroughs in waste forms with greatly improved performance.

- **The Used Fuel Disposition Campaign** is enabling the technology for storage, transportation, and disposal of used nuclear fuel (UNF) and wastes generated by existing and future nuclear fuel cycles.
- **The Materials Protection, Accounting, and Control Technologies Campaign** is developing the technologies, monitoring tools, and analysis techniques for next-generation nuclear safeguards and security, minimizing risks of proliferation.

Table 1: Relationship of FCT Campaign Areas to High-Level Goals

Campaign	Safety & Security	Transportation & Storage	Accident Tolerance	Sustainability
Fuel Cycle Options	◆	◆	◆	◆
Advanced Fuels	◆		◆	◆
Separations & Waste Forms	◆	◆		◆
Used Fuel Disposition	◆	◆		
MPACT	◆	◆		◆

The program has established short-, intermediate- and long-term strategic goals to implement the respective campaign objectives. Near-term goals include addressing the Blue Ribbon Commission's technical recommendations for used fuel management, increasing the focus on nuclear fuels with enhanced accident tolerance, identifying sustainable fuel cycle options for further development, introducing "safeguards by design" concepts into the international market, and enabling a fundamental scientific understanding of separations and waste behavior.

In the intermediate term, the program will conduct science-based, engineering-driven research for sustainable fuel cycle options, conduct research to support extended storage of used nuclear fuel, develop the scientific basis for determining sites and procedures for disposal of used nuclear fuel, develop advanced material control instrumentation and analytical techniques, and fabricate prototype waste forms and systems for the capture and immobilization of off-gas.

In the long term, the program will demonstrate specific fuel cycle technologies. FCT will work with industry to license a retrievable fuel storage facility, support the selection and licensing of potential used fuel disposal sites, implement a fully integrated MPACT system in a large-scale processing facility, and construct facilities to demonstrate off-gas capture and waste form technologies on an engineering scale.

Figure 1 illustrates how these activities span the entire nuclear fuel cycle.

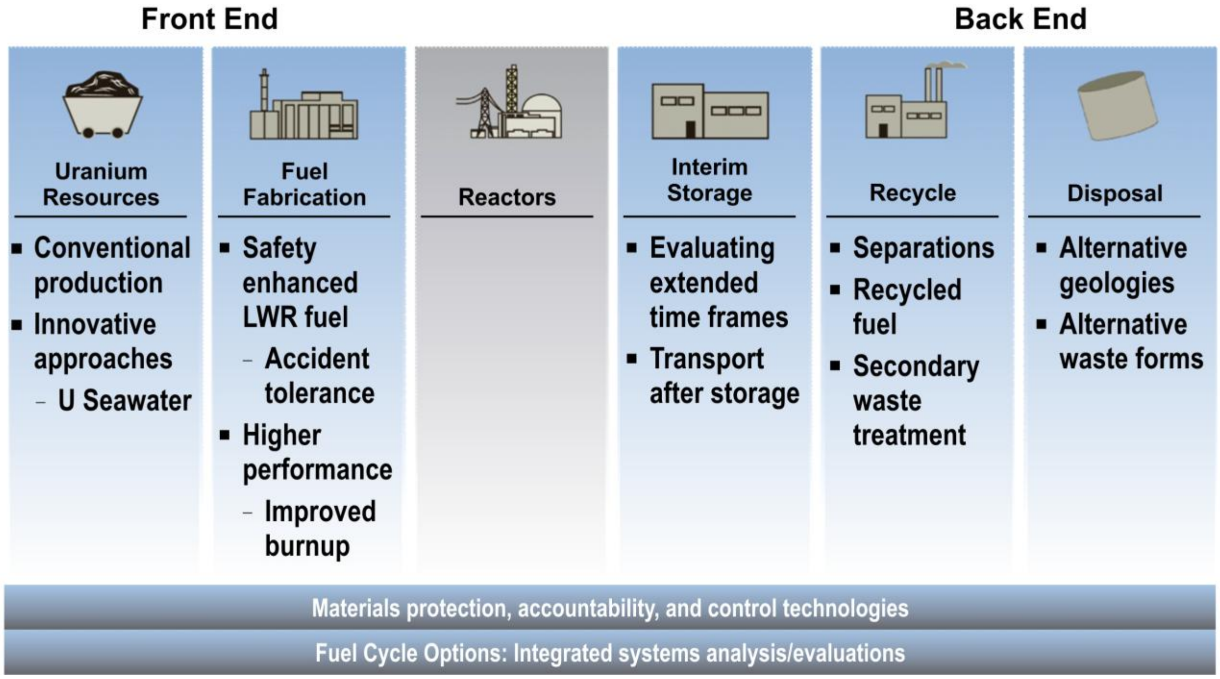


Figure 1: FCT is involved across the entire nuclear fuel cycle.

To achieve its mission, FCT has initiated numerous activities in each of the technical campaign areas, of which this report provides a sample. Highlights of these projects and some of their accomplishments in FY 2012 follow:

Fuel Cycle Options

- Defined 38 evaluation groups with 40 representative options as the basis for screening in FY 2013.
- Found evidence to support the promise of a breed and burn (B&B) reactor design that converts depleted uranium to plutonium and burns it without chemical separation.
- Investigated a core design, including safety attributes, for a high-conversion water reactor (HCWR) for potential use in fuel recycling with LWR technology .

Advanced Fuels

- Undertook analysis of advanced fuel systems (nitride and carbo-nitride), conducted extensive post-irradiation examination (PIE) of high-burnup mixed oxide fuel.
- Employed novel characterization methods to better understand material performance in nuclear environments.

Separations, Waste Forms, and Fuel Resources

- Won a prestigious R&D100 award for demonstrating new sorbent materials for extraction of uranium from seawater with seven-fold higher capacity for uranium over previously developed adsorbents.
- Continued efforts to develop technology to recover uranium from vast seawater reserves; capture and sequester volatile radionuclides from off-gas streams; improve understanding of solvent extraction processes through computational modeling tools and novel microfluidic experimental techniques; and explore glass behavior in corrosive environments.

Used Fuel Disposition

- Developed a Used Nuclear Fuel Storage and Transportation Research, Development and Demonstration Plan to support the extended storage of UNF and initiated key experimental activities to evaluate the performance of high-burnup cladding under conditions relevant to dry storage.
- Entered into agreements with international consortiums to perform collaborative R&D with established programs in other countries and initiated collaborative activities.
- Completed an integrated system-level evaluation of the back-end of the current once-through fuel cycle, considering a range of options for centralized storage and re-packaging of used nuclear fuel into disposal canisters.
- Conducted R&D on high-priority research gaps identified in the Used Fuel Disposition Campaign *Disposal Research and Development Roadmap* in the areas of generic engineered and natural system performance.

Materials Protection, Accounting, and Control Technologies

- Identified key issues and best practices for security of used fuel in storage.
- Developed a new harsh-environment alpha detector that can recognize actinides in a pyrochemical fuel processing stream.

The remainder of this document provides an overview of each campaign, followed by brief synopses of the key R&D project activities highlighted above. During the annual review meeting, national laboratory and university experts will provide more detailed presentations of each FCT campaign area along with the representative sampling of R&D projects being undertaken.

Fuel Cycle Options Campaign

1.1 Fuel Cycle Options Campaign Overview

Campaign Mission

The Fuel Cycle Options Campaign mission is to develop and implement analysis processes and tools and to perform integrated fuel cycle technical assessments to provide information that can be used to objectively and transparently inform and integrate Office of Fuel Cycle Technologies activities.

Campaign Objectives

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

Campaign Challenges

- Completing development of the process for evaluating fuel cycle options that will be objective, reproducible, and responsive to evolving national priorities.
- Conducting an evaluation and assessment of fuel cycle options that is convincingly comprehensive and conclusive concerning fuel cycle options and their capabilities.
- Screening of fuel cycle options to provide a basis for identifying R&D that would be needed to support development of such fuel cycles.
- Communicating all aspects of the fuel cycle options evaluation and screening process to a variety of stakeholders.

Fiscal Year 2012 Funding

Fuel Cycle Options Campaign	
Major Activities	**FY 2012 Funding**
Campaign Management	$750,000
Fuel Cycle Evaluation and Screening	$3,140,000
Integrated Fuel Cycle Analysis	$7,113,000
Fuel Cycle Information Management	$1,328,000
Program Communications	$397,000
Technical Program Integration	$200,000
Total	**$12,928,000**

Major Research and Development Activities

Nuclear Fuel Cycle Evaluation and Screening aims to develop the process and technical basis for an objective and reproducible evaluation of nuclear fuel cycles with respect to nine critical criteria, as well as conducting the evaluation. The evaluations will be used to identify a small number of promising fuel cycle options, providing information that can be used to guide R&D directions such as defining function and performance goals for each part of the fuel cycle. This entails activities to deliver the following major objectives:

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

Integrated Fuel Cycle Analysis ensures that fuel cycle evaluation is supported by accurate and consistent technical analysis of fuel cycle performance for the performance metrics, including fuel cycles that have not been extensively analyzed. Analyses cover a broad range of possibilities, including use of thorium and alternate reactor concepts , as well as to provide data for a broad range of performance metrics. A range of geologic disposal options will be included as concepts are further developed.

Analysis of Specific Systems Issues includes evaluation of potential binning of U.S. spent fuel available for disposal, research and future processing, MOX fuel utilization for a Report to Congress, nuclear lifecycle analysis and its impact on climate change, developing effective communication approaches for FCT program activities, and supporting knowledge management activities to collect, manage and preserve valuable program data and documents.

Key Fiscal Year 2012 Outcomes

Key accomplishments in FY 2012 include completion of the following reports:[1]

- *Fuel Cycle Grouping Approaches (FCRD-FCO-2012-000069)*
- *Nuclear Energy System Evaluation and Screening – Approach and the Evaluation Metrics (FCRD-FCO-2012-000322)*
- *DRAFT of the Fuel Cycle Options List to be Used for the 2013 Screening (FCRD-FCO-2012-000164)*
- *Fuel Cycle Evaluation and Screening Charter*
- *FCR& D FY2011Annual Summary Report*
- *Potential External Constraints on the Evaluation of U.S. Fuel Cycle Options for Long Term Research and Development (DRAFT) (FCRD-FCO-2012-000235)*
- *Nuclear Energy System Evaluation and Screening - Evaluation Approach and Metric for Financial Risk and Economics (Draft) (FCRD-FCO-2012-000270)*
- *Environmental Impacts, Health and Safety Impacts, and Financial Costs of the Front-End of the Nuclear Fuel Cycle (DRAFT) (FCRD-FCO-2012-000124)*
- *Fuel Cycle Database Catalog*

[1] The referenced documents may not all be available in the public domain at the time this document was published.

1.2 Development of the Comprehensive Options List for 2013 Evaluation and Screening

Michael Todosow – Brookhaven National Laboratory

Introduction and Objectives

The Charter for the Evaluation and Screening of Fuel Cycle Options authorizes a study to investigate the potential for alternative nuclear energy systems to provide significant performance advantages in comparison to the current once-through system based on light water reactor (LWR) usage. The purpose of this study is to inform decisions for an R&D program by identifying fuel cycles that have significant benefits in a variety of policy objectives.

Research and Development Overview

The evaluation and screening is based on several major elements:

1) A set of high-level criteria for evaluating performance improvement in terms of broadly defined economic, environmental, safety, nonproliferation, security, and sustainability goals

2) Development of evaluation metrics to describe and characterize the performance of options relative to the defined set of high-level criteria

3) Development of a comprehensive list of "**fuel cycle options**" (sometimes referred to as "**nuclear energy systems**") for the fuel cycle evaluation and screening, in terms of potential performance with respect to the high-level criteria

4) Development of an approach to **group** options with similar high-level features expected to affect performance such that:

(a) There is likely to be a meaningful discrimination of expected performance from one group to the next with respect to the evaluation metrics.

(b) The options comprising an individual group are sufficiently similar that the group's collective performance can be represented by a single option (the "**representative option**") with accuracy sufficient to allow the evaluation and screening of the most promising option group(s).

(c) The total number of groups is such that the evaluation and screening process is tractable.

Accomplishments

The study addressed the third and fourth elements described above, and achieved the following:

- Developed an approach for the grouping of fuel cycle options based on the fundamental characteristics that affect performance (e.g., reactivity, spectra, need for enrichment, incoming fuel material, etc.).
- Developed an approach for reducing the number of evaluation groups (and associated representative options) based on criteria that allowed combining groups based on similar fuel cycle performance with respect to selected key high-level criteria.
- Defined thirty-eight (38) evaluation groups with forty (40) representative options for once-through, and single and multi-stage recycle scenarios (both limited and continuous) that will serve as the basis for the 2013 evaluation and screening process.

1.3 Fuel Cycle Options Offered by Liquid Metal-Cooled Breed and Burn Reactors

***Christian Di Sanzo, Florent Heidet,* Staffan Qvist and Ehud Greenspan** –*
University of California, Berkeley
(* now at ANL)

Introduction and Objectives

A Breed-and-Burn (B&B) reactor is a breeder reactor that converts a significant fraction of a fertile feed fuel into plutonium and then fissions a significant fraction of the bred plutonium without having to reprocess the fuel or with limited processing of the used nuclear fuel (UNF), termed reconditioning. In order to initiate the chain reaction, the B&B core has to be started with an adequate amount of fissile fuel such as enriched uranium, plutonium or transuranium elements (TRU) extracted from light water reactor (LWR) UNF.

Research and Development Overview

Since 2008, with primary support from DOE Nuclear Energy University Programs (NEUP), a group of scientists at University of California (UC), Berkeley, have been studying feasibility of designing B&B cores, attainable core performance, and implications of successful reactor development. The work performed and the findings are described in detail in five journal papers, ten conference proceedings papers, two white papers, twenty-two presentations, and one invention disclosure.

Accomplishments

Following is a brief summary of the findings of the UC Berkeley team:

- It is neutronically feasible to establish a self-sustaining B&B mode of operation in large fast reactor cores provided the fuel and clad can operate safely up to an average burnup of approximately 20%. The uranium utilization of such B&B reactors is about 40 times that of LWRs.
- After reconditioning, fuel discharged from a B&B core at 20% burnup can be used for the "starter" fuel of a new B&B reactor core, thus enabling expansion of the B&B reactor fleet without need for additional fissile LWR fuel beyond that used for starting the initial core. The primary functions of fuel reconditioning are to remove the gaseous fission products (FPs) and to encase the fuel rods in new clad; no separation of actinides from solid FPs is needed.

- Alternatively, the fuel can be recycled in the B&B, after reconditioning, up to a maximum burnup of approximately 55%, with a corresponding uranium utilization about 100 times that of LWRs.
- When used in B&B fast reactors, stockpiles of depleted uranium (currently a waste material) that will accumulate in the United States by 2050 will be able to generate the total U.S. electricity demand for at least eight centuries—twenty centuries if 50% burnup is attainable. No fuel reprocessing and very little (if any) uranium enrichment will be required.
- It is possible to design even large B&B cores to be passively safe.
- It is possible to design a B&B core to fit within the reactor vessel of Super-PRISM.
- It is possible to start implementing the B&B mode of operation without exceeding the already proven clad limits of 200 displacements per atom (dpa) by driving a sub-critical B&B blanket with neutrons leaking from a critical sodium fast reactor (SFR) driver. The blanket discharge burnup could be gradually increased as new supporting experimental evidence becomes available until reaching an acceptable dpa level while establishing a self-sustaining B&B mode of operation. With a 200-dpa constraint, a depleted uranium B&B blanket can generate at least two-thirds of the total core power. The fuel cycle cost of such a seed-blanket SFR is expected to be significantly lower than that of a conventional SFR.
- The amount of TRU in B&B UNF, particularly fissile inventory, can be significantly smaller per unit of electricity generated than that in LWR UNF.
- If LWR UNF could be economically reconditioned for feed fuel of B&B reactors, it will be possible to generate at least double the electricity already produced by the same fuel in LWRs, while continuing to operate on a once-through fuel cycle. That is, the B&B core could provide a very efficient “interim” solution for the LWR UNF.

The UC Berkeley team concluded that successful development of B&B reactors and associated fuel re-reconditioning technologies offer promising new options for the nuclear fuel cycle that could provide a great measure of energy security and energy cost stability. This prospect justifies addressing the challenging technological issues that need be solved before B&B reactors and fuel reconditioning could become commercial.

1.4 Fuel Cycle Option Offered by Reducing the Void Coefficient of Reactivity in High-Conversion Water Reactors

Bo Feng – Argonne National Laboratory
Francesco Ganda – Idaho National Laboratory
Staffan Qvist – University of California, Berkeley

Introduction and Objectives

The Fuel Cycle Research and Development (FCR&D) Program of the DOE Office of Nuclear Energy will conduct an evaluation and screening of fuel cycle options in 2013. To support this effort, fuel cycle data are being collected and, in cases where existing data are insufficient or unavailable, the program is conducting analyses to provide the pertinent information. Such is the case for a system in which transuranic elements are continuously recycled in high-conversion water reactors (HCWRs); there is insufficient information regarding the feasibility of a HCWR design that can produce more fuel than it consumes while exhibiting sufficient safety performance.

The reference HCWR for this study is the GE-Hitachi Resource-Renewable Boiling Water Reactor (RBWR), an un-finalized design of such a system, which employs axially alternating depleted UO_2 blankets and mixed-oxide (MOX) fuel zones in a short pancake-shaped core, a tight triangular lattice, and high void fractions via increased boiling in order to achieve the low moderator-to-fuel ratio required for breeding. However, a potential safety and regulatory obstacle for the RBWR and similar HCWR designs is the positive void coefficient of reactivity.

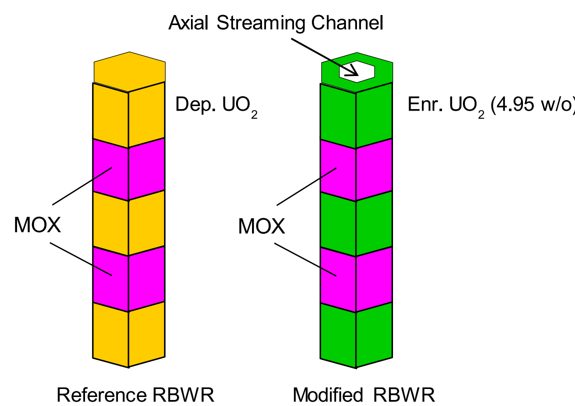


Figure 1: Reference RBWR fuel assembly (L) and modified version (R) showing axial streaming channel and enriched UO_2 blankets

Therefore, the objective of this study is to investigate the feasibility of obtaining negative void coefficients of reactivity in a HCWR design while achieving a fissile inventory ratio (FIR) greater than 1.0. If feasible, such a design could be an attractive option since it could back-fit existing light water reactor cores, use established water-based technology and infrastructure, and potentially achieve a sustainable nuclear fuel cycle.

Research and Development Overview

This study confirmed, through a coupled reactor physics and thermal hydraulics analysis, that GE-Hitachi's RBWR core design showed positive void coefficients at various times of the equilibrium cycle. The pin, assembly, and core models used in this study were created via the stochastic reactor physics codes MCNP and KENO, coupled with lumped channel thermal hydraulics models. In attempts to reduce the void coefficient, the research team investigated various material modifications to the RBWR design, including adding $ZrH_{1.6}$, thorium, erbium, and low-enriched UO_2, and geometric modifications, including employing axial checkerboard fissile/fertile heterogeneity and axial streaming channels. None of these modifications by themselves was able to reduce the void coefficient sufficiently while maintaining the original FIR and fuel discharge burnup. Employing both material and geometric changes simultaneously seemed to be the most promising approach, so a modified RBWR core design was proposed: the reference RBWR core's axial blankets of depleted UO_2 were replaced with 4.95 weight percent (w/o) enriched UO_2, and the four innermost rings of fuel pins in every assembly were replaced with an axial streaming channel.

Accomplishments

The modified RBWR design proposed in this study is able to achieve negative void coefficients of reactivity, lower un-rodded axial power peaking factors, and an increased fissile plutonium inventory ratio (from 1.04 to 1.16), while maintaining the same average discharge burnup of 45 GWd/t. However, the core power is reduced by 14% to maintain the same linear heat rate, and the total FIR (including fissile plutonium *and* uranium-235) is less than 1.0, which means that it consumes more fuel than it produces. It cannot be concluded that a HCWR can achieve negative void coefficients at all times during the equilibrium cycle while maintaining an economical discharge burnup and a total FIR greater than 1.0, which is a requirement to achieve sustainable fuel cycles. However, based on the efforts from this study, it can be concluded that achieving all these performance goals simultaneously, if at all possible, may require substantial changes to conventional HCWR designs.

Advanced Fuels Campaign

2.1 Advanced Fuels Campaign Overview

Campaign Mission

Using a goal-oriented science-based approach, the Advanced Fuels Campaign (AFC) conducts RD&D for various fuel forms (including cladding) needed to implement different fuel cycle options, as defined in the DOE-NE Research and Development Roadmap, Report to Congress, April 2010. The campaign mission for any given fuel type ends when fuel qualification is completed via engineering-scale demonstration of the fabrication processes and irradiation of lead-test assemblies (LTAs) to demonstrate in-pile performance. The mission also includes the development of a state-of-the art R&D infrastructure to support the goal-oriented science-based approach.

Campaign Objectives

AFC has been given the responsibility to develop advanced nuclear fuel technologies for the FCR&D program. The campaign currently focuses on:

- **Advanced Light Water Reactor (LWR) Fuels** with enhanced accident tolerance, improved performance, and reduced waste
- **Metallic Transmutation Fuels** for transmutation with enhanced proliferation resistance
- **Crosscutting Capability Development** such as separate effects testing for modeling and simulation, advanced in-pile instrumentation, characterization and post-irradiation examination (PIE) techniques

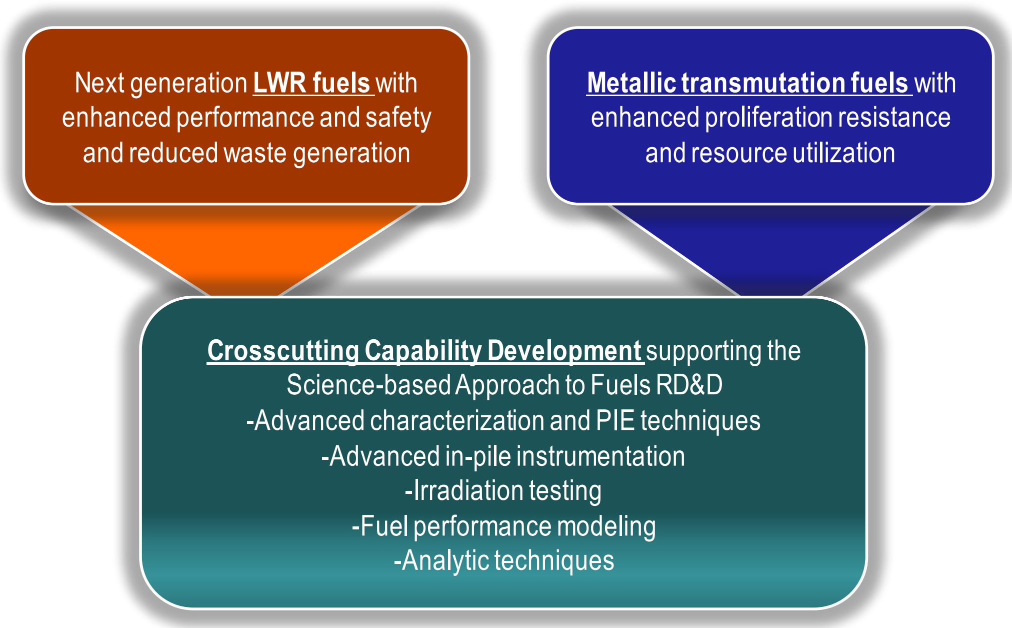


Figure 1: The three AFC pillars.

Campaign Challenges (3 – 5 year goals):

- Complete the conceptual design for baseline transmutation fuel technologies with emphasis on fundamental understanding of fuel fabrication and performance characteristics.
  - Low-loss fabrication capability
    - Hot-cell furnace within three years
  - Comparison of fast spectrum versus filtered thermal spectrum testing
    - Final report in three years (after FUTURIX tests are analyzed)
  - Impact of lanthanides on burnup limits – quantitative assessment complete in five years. Options are:
    - Minimize initial lanthanides – joint studies with pyroprocessing
    - Perform tests with realistic feedstock – lanthanide precipitation limits
    - Immobilize lanthanides – getters used in the fuel
    - Use lanthanide barriers for fuel–cladding chemical interaction (FCCI) – lined/coated cladding
- Identify and demonstrate feasibility of innovative concepts that provide considerable advantage compared to baseline technologies.
- Achieve state-of-the art R&D infrastructure that can be used to transition to a science-based approach to accelerate further development of selected concepts, with emphasis on:
  - PIE capabilities
  - *In situ* instrumentation
- Identify and select advanced LWR fuel concepts for development towards lead-test rod testing within the subsequent five to seven years.
  - Feasibility studies for four years
    - Small-scale irradiation testing/PIE
    - Sample fabrication
    - Out-of-pile testing (including high-temperature steam testing)
    - Assessments of impacts on economic, fuel cycle, operations and design-basis accidents
- Support development of the predictive, multi-scale, multi-physics fuel performance code.

Fiscal Year 2012 Campaign Funding

Advanced Fuels Campaign	
Major Activities	**FY 2012 Funding**
Campaign Management and Integration	$2,519,000
Analytical Support	$1,925,000
Characterization Technique Development	$1,800,000
Irradiation and PIE	$5,175,000
Core Material	$3,020,000
Ceramic Fuel Fabrication and Characterization	$3,200,000
Metal Fuel Fabrication and Characterization	$5,600,000
Microencapsulated Fuels and Support of Accident-Tolerant Fuel Development	$4,100,000
Innovative Fuels	$1,750,000
Nuclear Data	$2,687,000
Total	**$31,776,000**

Major Research and Development Activities

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

International Collaborations: China, the European Union, France, Japan, Russia, and South Korea. The emphases in all collaboration activities are metallic fuel development, joint irradiation testing and data analyses, and characterization/PIE techniques development.

Analytic Support: reactor impact analyses, fabrication modeling, and sensitivity studies on fuel performance code developed by the Nuclear Energy Advanced Modeling and Simulation Program (NEAMS)

Ceramic Fuels: advanced sintering process development, thermal performance of oxide fuels, and fission gas gettering concept development (with a FY 2012 emphasis on LWR fuels)

Metallic Fuel: casting technology development, fabrication and characterization of minor actinide- and lanthanide-bearing fuel samples, fundamental properties measurement and FCCI tests (also includes metallic LWR fuel development)

Microencapsulated Fuels: development of LWR fuels with enhanced safety performance

Core Materials: development of fast reactor cladding up to 200 dpa and development of LWR cladding for enhanced performance and accident tolerance

Characterization/PIE techniques: development of novel techniques and adaptation of high-resolution equipment to irradiated fuel samples

Irradiation Testing: irradiation test design and execution, PIE and data analyses, and advanced in-pile instrumentation development

Presentation Summary

In the near term, the Advanced LWR Fuel effort is focused on enhancing fuel accident tolerance during an extended loss-of-coolant event. Metallic Transmutation Fuels RD&D is focused on the ability to recycle transuranics to achieve partially or fully closed fuel cycles. Fabrication processes, in- and out-of-pile performance, and characterization are elements of the research activities, which also support the joint fuel cycle demonstration studies. Finally, the Crosscutting Capability Development elements of the campaign support the science-based approach focusing on developing a microstructural understanding of nuclear fuels and materials. The science-based approach combines theory, experiments, and multi-scale modeling and simulation to develop a fundamental understanding of the fuel fabrication processes and fuel and clad performance under irradiation. The objective is to use a predictive approach to design fuels and cladding to achieve the desired performance (in contrast to more empirical observation-based approaches traditionally used in fuel development). The experimental activities (separate effects testing) to support the modeling and simulation effort are also included in this program element.

Major FY 2012 Accomplishments

- Researchers at the Idaho National Laboratory have completed Advanced Test Reactor (ATR) irradiation of AFC-2D (High Burnup MA-MOX Experiment) and AFC-2E (Metallic Fuels Fabrication Variables). These experiments will contribute important data toward the development of a connection between historical fast reactor fuel experiment data and testing of advanced fast spectrum fuels in the ATR.
- Researchers have completed the first year of hydraulic rabbit testing (of metallic fuel specimens) in the High Flux Isotope Reactor (HFIR). These tests provide valuable low-dose irradiation performance data to inform and validate the advanced modeling and simulation effort on nuclear fuels.
- Researchers have completed ATR irradiation of AFC-3A (Advanced Metallic Fuel Concept Feasibility Test) on metallic fuels with alloying additions for lanthanide control and annular (sodium-free) slug geometry.

- Researchers have completed baseline PIE campaign on legacy metallic and oxide fuel experiments from the Experimental Breeder Reactor II (EBR-II) and Fast Flux Test Reactor (FFTF) with relevant data for the current transmutation fuel program (including low-smear-density metallic fuels, high-temperature HT9 cladding, and ultra-high-burnup annular MOX fuels).
- The Joint Fuel Cycle Study with the Korea Atomic Energy Research Institute (KAERI) has shown major progress; notably, design packages for the remote hot-cell casting furnace and for the remote sodium settler-bonder are under final review.
- Collaborations with universities have demonstrated that straightforward alloy modifications are capable of stabilizing metallic fuel phases for increased fuel performance toward achieving ultra-high burnup and transuranic burning.
- A ceramic fuel melt point determination apparatus and capability has been established at Los Alamos National Laboratory for materials up to ~3000°C. This capability was demonstrated for five different urania-rare earth oxide compositions in the stoichiometric condition to provide a data set for model development and validation. The capability to map melting point and phase diagrams of materials is critical to understanding and predicting basic material behavior.
- In collaboration with the Japan Atomic Energy Agency (JAEA), researchers have developed a simultaneous thermal analysis system for measuring oxidation of uranium dioxide in controlled atmospheres containing up to 100 percent water vapor. The technique can measure oxidation of LWR fuels in situ during exposure to accident-relevant environments at temperatures up to 1250°C.
- Near-net shape sintering and fabrication of UO_2 pellets have been demonstrated at the Los Alamos National Laboratory to a tolerance of better than ±20 micrometers. Advanced fabrication techniques such as this may be able to significantly lower fabrication process losses and improve fuel performance.
- Researchers have demonstrated a pellet measurement system that provides 3-D mapping of green and sintered pellets, along with a full pellet surface image that can be analyzed to identify defects (e.g., cracks and chips) in sintered pellets. The system can also be used for process monitoring via green pellet monitoring.
- An ion-beam assisted deposition (IBAD) process has been demonstrated for producing thick films of UO_2 with xenon in solution without irradiation. Post-synthesis annealing was shown to yield xenon bubbles that have dimensions and morphologies closely resembling those seen in irradiated fuels.
- Researchers at Idaho National Laboratory, in collaboration with Texas A&M University, produced TiN coated HT-9 tubes (to prevent FCCI) for a future high-burnup ATR irradiation.

- Researchers at Pacific Northwest National Laboratory (PNNL), in collaboration with University of California (UC), Berkeley, tested tensile properties and investigated microstructure using transmission electron microscopy (TEM) and atom probe on MA-957 after irradiation to 100 dpa in FFTF. The MA-957 oxide dispersion-strengthened (ODS) steel alloy is a candidate for advanced nuclear fuel and reactor systems. These data provide a basic understanding of the alloy performance under neutron irradiation environments.
- Researchers at Oak Ridge National Laboratory performed oxidation tests in steam at temperatures up to 1300°C for candidate steel alloys. FeCrAl alloys were the most promising. The FeCrAl alloys are candidates for enhanced accident-tolerant LWR fuel cladding, and steam reaction is a key metric determining the alloy's performance in off-normal reactor conditions.
- Through a cooperative research and development agreement with Terrapower, researchers at Los Alamos National Laboratory (LANL) and PNNL are preparing specimens of HT-9 (which were previously irradiated in FFTF to doses up to 200 dpa) for inclusion in an irradiation in Russian reactor BOR-60. A key goal for the Advanced Fuels Campaign is to develop an understanding of material performance at very high doses, greater than 200 dpa. These samples will be irradiated in BOR-60 to greater than 200 dpa providing some of the first such data available.
- Researchers at LANL and ORNL, in collaboration with UC Berkeley and the Zoz Center, UC Santa Barbara, and ATI, have completed milling of 45 kg (in 15 kg batches) of 14YWT (an advanced radiation-tolerant material). ODS steels, such as 14YWT, need to be fabricated in bulk quantities. Working with universities and industry in this regard ensures that, when ODS alloys are ready for industrial deployment, they can be fabricated at industrial scales.
- Researchers have demonstrated irradiation stability of fully ceramic microencapsulated (FCM) fuel surrogate fuel to a LWR lifetime dose.
- Researchers carried out steam accident testing to identify advanced FeCrAl and SiC-based clad candidates for accident tolerance.
- Researchers designed, constructed and demonstrated a first-of-its-kind 1600°C steam/ hydrogen severe accident test station.
- The team published the first thermochemistry information on the UCN system, applying it to uranium nitride kernel fabrication.

2.2 Understanding Nuclear Fuel Behavior: Thermochemistry of Oxides and Carbo-Nitrides

T. M. Besmann, S. L. Voit, D. Shin, J. W. McMurray – Oak Ridge National Laboratory

The continuing interest in fast reactors and more recent concerns about extending the burn-up of LWR systems and their safety have generated an effort to better understand the behavior of existing fuels and the development of advanced fuel materials. Current LWR and projected fast reactor oxide fuel pins are difficult to model, owing to their high operating temperatures and the significant concentrations of large numbers of fission product elements that result from high burnup. This unprecedented complexity offers an enormous challenge in the thermochemical understanding of these systems and generates opportunities to advance solid solution models to describe these materials.

In the fissioning of uranium, lanthanides and yttrium are generated in significant amounts. These elements typically dissolve in the fuel fluorite oxide phase, having a strong influence on oxygen behavior as a function of oxygen-to-metal ratio and temperature and, thus affecting other thermophysical properties. Thermochemical solid solution models for actinide oxides containing lanthanides and yttrium are thus important for predicting properties and have been the focus of recent work, supported by experimental measurements in urania-lanthanide systems. They have resulted in reasonably accurate representations of behavior using approaches that account relatively simply for the oxygen ion defects in the phase. These and other phase thermochemical values, together with computed fission product concentrations, allow calculation of composition and properties (Figure 1).

Alternative fuel concepts for melt-resistant and accident-tolerant LWR fuels include the tristructural isotropic (TRISO) technology, which consists of an actinide-containing kernel coated with carbon and silicon carbide (SiC) layers that act as pressure vessel and fission product containment. For use in an LWR, these approximately 1–mm-diameter particles will be embedded in either a SiC or zirconium alloy matrix. Because of the limited actinide volume in this fuel form for standard PWR or BWR fuel rod dimensions, it may be necessary to use a higher fuel loading than can be accommodated by UO_2. As a result, uranium nitride (UN) fuel is being considered because of its higher uranium concentration, and efforts to prepare and assess the fuel are being pursued. In addition, interest has been expressed in utilizing pellets of UN fuel, directly replacing UO_2, as this would have a possible advantage in higher thermal conductivity and fission product retention.

Historically, the preparation of UN fuel has been by carbothermic reduction of the oxide with subsequent/simultaneous nitridation, and thus an understanding of the U-C-N system is important. Data on the phase equilibria and thermochemistry of the U-C-N system are, however, limited. Sufficient information does exist to model the U(C,N) phase reliably as a solid solution of UC and UN. With this assumption, researchers can compute system nitrogen pressures and determine phase diagrams.

A thermochemical analysis of the U-C-N system will be described, including the U(C,N) solid solution and prediction of high-temperature behavior.

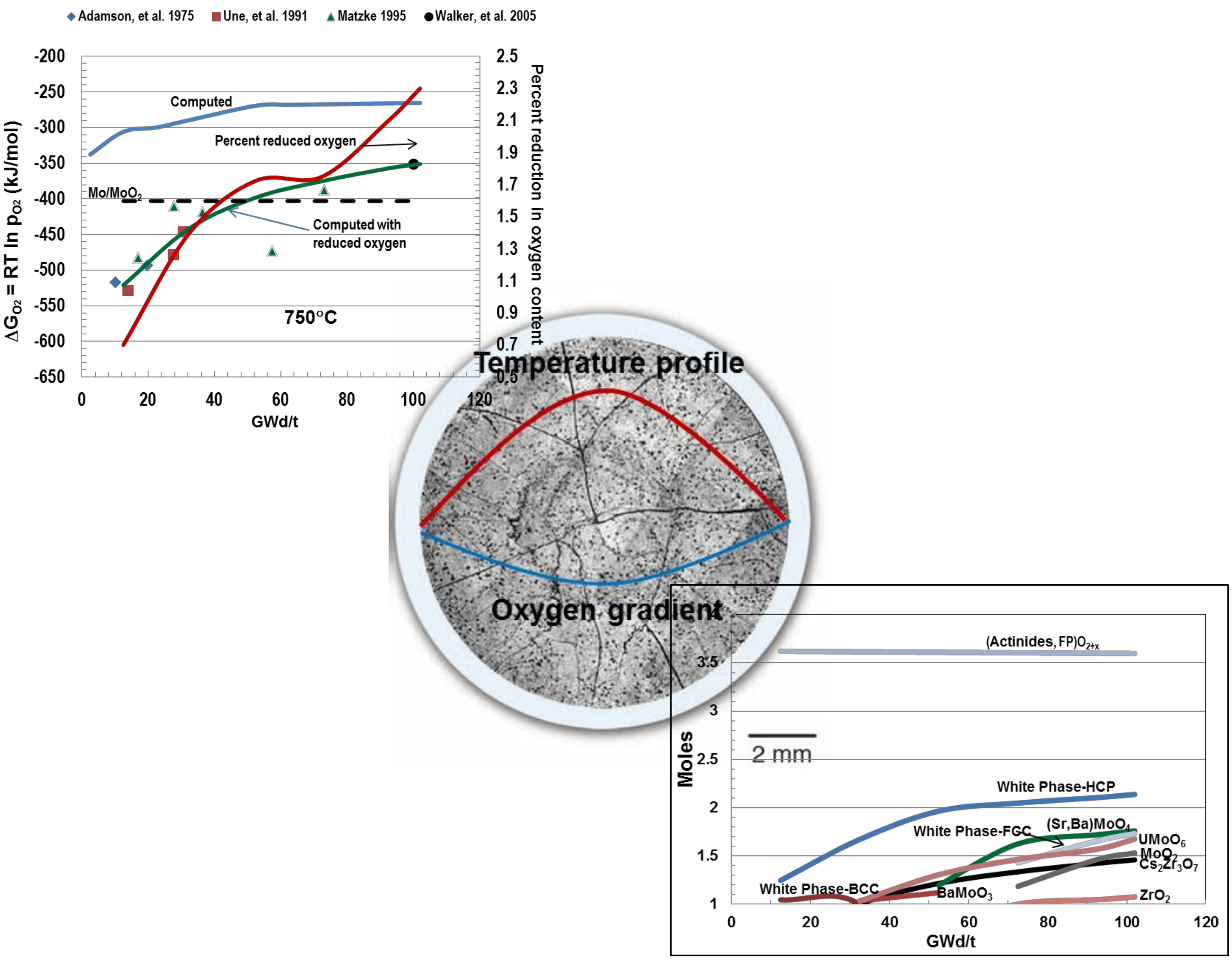


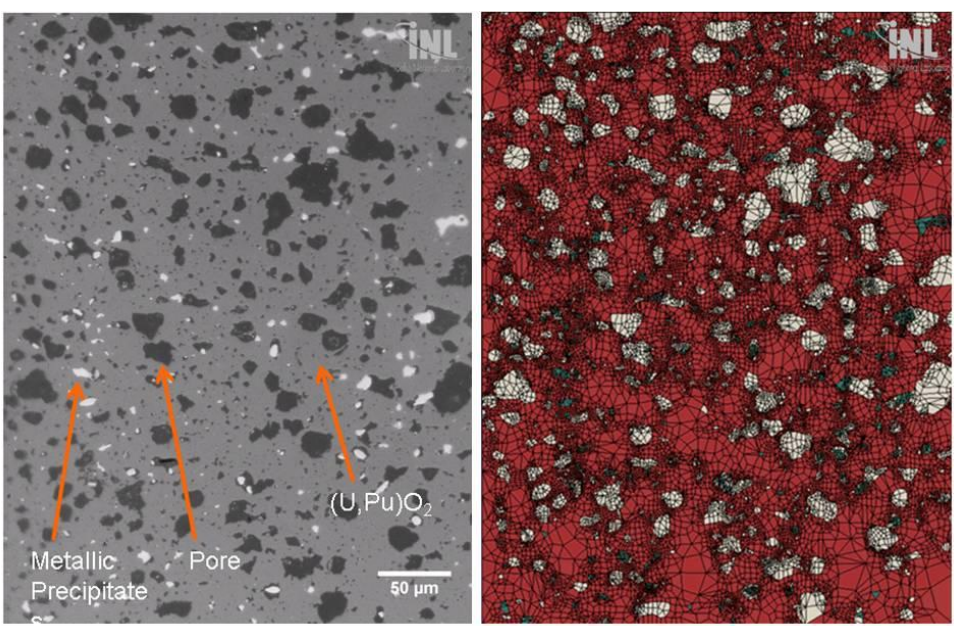
Figure 1: Image of irradiated UO_2 pin cross-section illustrating oxygen and thermal gradients and plots of the variation of oxygen potential in fuel, with and without the effect of clad oxidation, and phase composition during burnup.

2.3 Characterization and Microstructural Modeling of High Burnup Oxide Fuel from the Fast Flux Test Facility

Melissa Teague – Idaho National Laboratory

The increasing demand for cost-effective green energy has led to a renewed interest in nuclear energy, including the commercialization of fast breeder reactors (FBRs). In order for FBRs to become economically competitive with current light water reactors (LWRs), the average burnup of fuel assemblies in an FBR will need to exceed about150 GWd/tHM (~15% fissions per initial metal atom [FIMA]). A secondary reason for interest in FBRs is their potential to “burn,” or transmute, long-lived transuranic isotopes in used nuclear fuel produced by the current fleet of LWRs. Currently, fast reactor performance is largely defined by the limitations of the materials used in reactors, especially the metallic or mixed oxide ($(U, Pu)O_2$) fuel itself. Problems include fission gas generation, changes in thermal conductivity, microstructure changes within the fuel, fuel swelling, and fuel–cladding chemical interaction (FCCI).

The research team conducted optical microscopy, with results showing the evolution of the microstructure from burnups of 3% to 24% FIMA. Data show the formation of a high burnup structure in the rim of the higher burnup samples and re-opening of the pellet fuel gap. Using object-oriented finite element (OOF) software, researchers converted experimentally observed microstructures into finite element analysis (FEA) meshes. An example of a micrograph and its conversion to a FEA mesh is shown in Figure 1. These experimentally derived meshes were then used to model the effective thermal conductivity of the high burnup fuel.



(a) (b)

Figure 1: (a) Optical micrograph of high burn-up oxide fuel;
(b) FEA mesh generated using OOF from the optical micrograph.

The team obtained a burnup of 6.7% FIMA for a mixed oxide (MOX) fuel using a dual-beam focused ion beam (FIB), then conducted post-irradiation examinations (PIE). Analytical tools and techniques applied included three-dimensional microstructure, energy dispersive Xray spectroscopy (EDS), and electron backscatter diffraction (EBSD). The extensive PIE data collected will support key insights into the behavior of high burn-up oxide fuel.

2.4 Benefits and Challenges of Novel Material Characterization Methods for Nuclear Applications

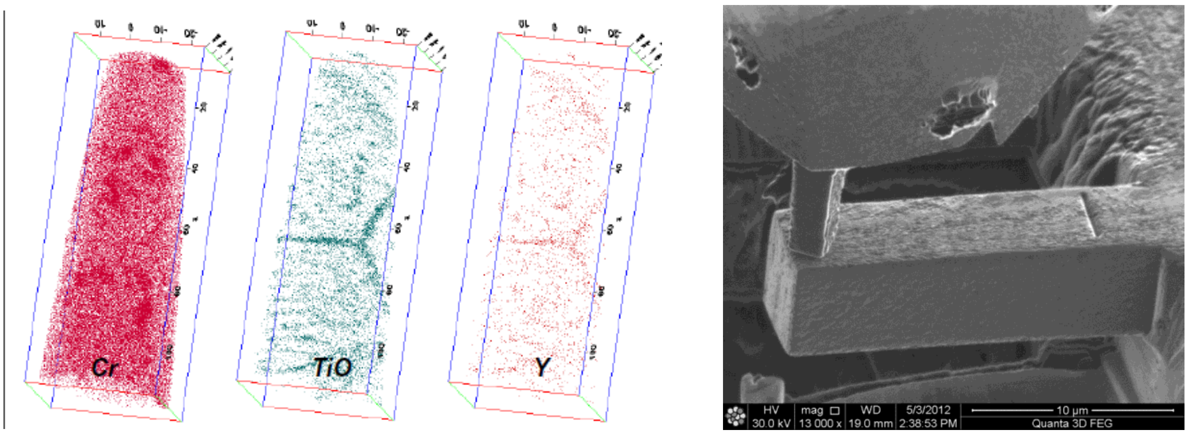
P. Hosemann – University of California, Berkeley

Since the beginning of the nuclear age, scientists and engineers have been concerned about materials degradation in nuclear environments. Therefore, significant efforts have been made to study the related phenomena, such as embrittlement, swelling, and corrosion. Tremendous advances have led to efficient design, structures and materials solutions for challenging problems. Despite these significant advances, however, some basic scientific questions remain unanswered.

Novel materials characterization methods have recently become available, allowing unprecedented insight into old and new problems and leading to new solutions or the confirmation of old data. Enhanced understanding of materials in nuclear environments can improve prediction of materials degradation and the basic understanding of various phenomena. In addition, increased knowledge of materials in challenging nuclear environments can improve the materials development process, thanks to more appropriate and target-oriented accelerated materials testing.

This project is utilizing novel materials characterization methods, conducting small-scale mechanical testing (micro- and nano-compression testing, micro-bend bar testing, nano-indentation) on ion beam and spallation source irradiated materials to obtain mechanical properties. Because of experimental limitations and safety, only micrometer-sized specimens are available for study. These specimens allow an increased statistical sample size, optimizing the limited number of macroscopic samples available and reducing the uncertainty of the data obtained from individual macroscopic mechanical tests, as mechanical properties measured are only as valuable as the microstructural data employed during the measurements.

Therefore, this research team used advanced microstructural characterization tools, such as the double aberration-corrected transmission electron microscope (TEM) and local electrode atom probe, to investigate changes in microstructural features due to exposure in a reactor environment. Researchers studied unique samples previously irradiated in the Fast Flux Test Facility (FFTF) and found that the formation of intermetallic precipitates is clearly an issue that must be considered.



(a) (b)

Figure 1: Local electrode atom probe data obtained from the oxide dispersion-strengthened (ODS) steel MA957 after high-dose irradiation in FFTF: (a) *in situ* micro-bend bar testing of protective oxide layer grown on steel in (b) liquid metal environment.

Separations, Waste Forms, and Fuel Resources Campaign

3.1 Separations, Waste Forms, and Fuel Resources Campaign Overview

Campaign 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.

Campaign Objectives

- Develop a fundamental understanding of methods for the separation of transuranic elements from used fuel.
- Develop a fundamental 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 enabling separation technologies to separate transuranic elements from used light water reactor and fast reactor fuel.
- Demonstrate predictable performance of advanced waste forms with greatly improved durability and waste loadings.
- Investigate alternative separations technologies and waste forms that could lead to transformational breakthroughs.

Campaign Challenges

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

Fiscal Year 2012 Funding

Separations, Waste Forms and Fuel Resources Campaign	
Major Activities	**FY 2012 Funding**
Campaign Management	$815,000
Domestic Echem Technology	$1,188,000
Advanced Aqueous Technology	$1,050,000
Sigma Team for Minor Actinides	$3,135,000
Sigma Team for Off-Gas	$3,715,000
Fundamental Methods Development	$2,580,000
Advanced Separation Alternatives	$2,345,000
Advanced Waste Forms	$2,005,000
Waste Forms Characterization	$1,878,000
Waste Form Processing	$600,000
Modeling and Simulation	$935,000
Fuel Resources	$3,747,000
Total	**$23,993,000**

Major Research and Development Activities

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

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

Advanced Waste Forms are necessary for the immobilization of waste streams from advanced separations processes, including high-halide electrochemical salt wastes, gaseous fission products waste, and separated technetium. The waste forms for streams containing iodine-129, technetium-99, and transuranium elements require performance over very long time periods to be sufficient barriers 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.

Fundamental Methods Development is developing the tools and methods to enable a science-based, engineering-driven research approach focused on understanding the fundamental scientific properties of separation processes and waste form behavior, rather than a purely empirical approach.

Key Fiscal Year 2012 Outcomes

- Completed independent relevancy review of Separations and Waste Forms Campaign.
- Issued revised Separations and Waste Forms Campaign Implementation Plan.
- Demonstrated feasibility of spectrophotometric monitoring of lanthanides in molten salt media. This represents the first time metal concentrations have been monitored by spectrophotometric means (a technique used for aqueous processes) in a molten salt.
- Completed successful demonstration of glass ceramic waste forms production in a cold crucible induction melter. This is the first-ever demonstration of producing a glass-ceramic waste form, which must be made at higher temperatures than can be used in joule-heated melters.
- Designed, built and demonstrated a prototypic micro-fluidic-based, robotic liquid sampling system to automate sampling, reduce waste and reduce personnel exposure.
- Demonstrated proof of concept for the selective extraction of uranium and its recovery using supercritical fluids, as a potential alternative to conventional aqueous processing.
- Demonstrated the ability to efficiently produce epsilon metal waste forms by spark plasma sintering or hot isostatic pressing. Epsilon metal waste forms have been demonstrated to have extreme durability over millennia and could be a potential waste form for undissolved solids.
- Developed and demonstrated a silica aerogel capable of high capacity and high separation factor for iodine. This has the potential of greatly improving iodine recovery from off-gas streams to meet stringent decontamination factors for iodine to meet regulations.
- Demonstrated dissolution and electrochemical recovery of uranium in ionic liquid media. This is an important demonstration, showing potential for future improvements in processing used fuel.

- Developed a simplified one-step An(III)/Ln(III) solvent extraction system with improved operating characteristics. This is a significant step in simplifying the separation of americium and curium from used fuel.
- Developed a conceptual flowsheet for americium (III) extraction using conventional extractants. This flowsheet is substantially more simple than currently developed processes and could be a major step forward in the separation of americium (without curium).
- Demonstrated new sorbent materials for extraction of uranium from seawater with significantly higher capacity for uranium over previously developed and tested materials.
- Completed analyses of a 26-year-old glass corrosion test jointly with CEA (France). This collaborative program is working toward an international consensus on glass corrosion mechanisms.

3.2 Fuel Resources: Cost and System Analysis of Recovery of Uranium from Seawater

Erich Schneider – University of Texas at Austin

Introduction and Objectives

The 4.5 billion tonnes of uranium dissolved in seawater exceed conventional terrestrial resources by a factor of roughly 1,000. Since this uranium is present at extremely low concentrations of 3.3 parts per billion (ppb), the Fuel Resources Campaign is focusing on developing novel materials that surpass the sorption capacity, selectivity and durability of the best existing technology. In parallel with this R&D effort is assessment of the technology's cost and energy return on investment (EROI) - the focus of this task. By identifying the system's highest-impact components, these key viability metrics may guide the technology R&D campaign.

Research and Development Overview

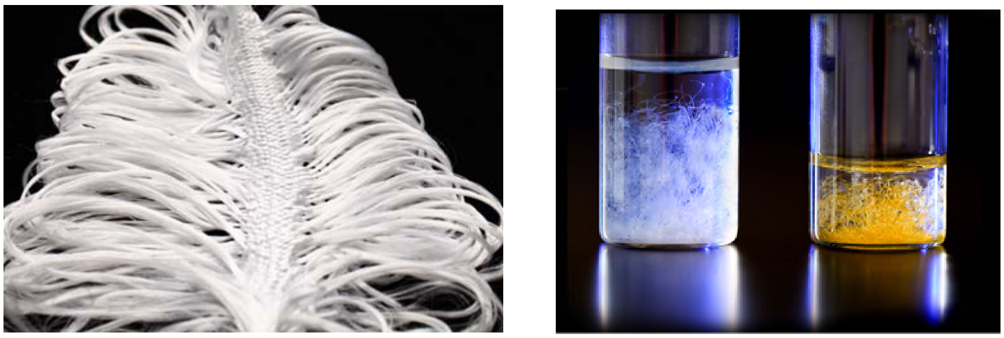
R&D activities focus on adsorbent material development, coordination chemistry and thermodynamics, computer-aided ligand design, marine testing, cost and systems analysis. Six NEUP-funded university-led projects are working on aspects of this topic area. The adsorbent development and ligand design effort focuses on the reference amidoxime-based ligand grafting process, as well as advanced ligands. Novel high-surface-area and nanoporous adsorbents are also being developed. Improvements in ligand selectivity, radiation-induced grafting efficiency, and group density in turn advance the uranium adsorption capacity. Parallel work to improve the adsorbent regeneration process and uranium stripping methodology (using carbonate solutions or supercritical CO_2, for example) is enhancing the materials' durability for reuse in the sea.

A marine testing program carried out at sites in Washington State and Florida is closely integrated with the materials development R&D. The testing program quantifies the performance of candidate materials in seawater using both batch and continuous flow-through apparatus. Interaction of materials with the marine biosphere and mitigation of biofouling are additional focus areas for the marine testing team.

These activities are in turn tightly coupled with the cost and systems analysis effort. New materials must not only offer superior capacity, selectivity and durability, as demonstrated by the marine tests, but also enhance the technology's viability as measured by the cost of producing uranium.

Accomplishments

Advances in high-surface-area trunk materials development have led to a 2012 R&D 100 Award for the Oak Ridge National Laboratory (ORNL) team and partner Hills, Inc., for their HiCap adsorbent (see Figure 1). Marine testing results have shown that new materials are achieving uranium adsorption levels approaching twice that of the best performance reported for the Japanese amidoxime adsorbent. The presentation will document both the uranium production cost estimate based on the Japanese adsorbent—ca. $1,200/kg uranium—and the cost benefits of advances made by the Fuel Resources Campaign.



(a) (b)

Figure 1: (a) R&D 100 award-winning HiCap adsorbent; (b) before and after adsorption of uranium.

3.3 Off-Gas Sigma Team

Bob Jubin – Oak Ridge National Laboratory

Introduction and Objectives

The Off-Gas Sigma team was formed in late FY 2009 to address the broad technical challenges surrounding the capture and sequestration of the volatile (iodine-129, carbon-14, krypton-85, and tritium) and semi-volatile radionuclides associated with virtually every fuel cycle option being considered. Starting with the highest priority elements, materials for the capture of iodine, tritium and krypton are being evaluated for implementation across the entire head-end processing operation as well as in other off-gas streams requiring treatment. Carbon-14 will be addressed for specific cases, if required, to meet regulatory requirements.

Research and Development Overview

For iodine capture, the technical challenges include achieving decontamination factors (DFs) of over 1000 from off-gas streams containing ppm to ppb levels of radioiodine in multiple chemical forms. For krypton capture, high selectivity and the ability to operate near room temperature are significant challenges. For tritium capture, the challenge is primarily recovery from very dilute streams and avoiding co-adsorption of long-half-life radionuclides. The R&D efforts encompass activities that have utilized unique capabilities within the DOE complex to (1) probe the movement of silver atoms to form mobile nanoclusters in iodine capture media, (2) develop new iodine and krypton adsorbents, (3) demonstrate iodine capture DFs > 1000 in small-scale engineering studies using deep-bed column performance, (4) explore the degradation effects of long-term operations on iodine capture media, and (5) evaluate integrated operations.

Accomplishments

- Significant advances have been made in understanding the behavior of the silver in silver-exchanged mordenite with impacts on the sorbent pretreatment, aging, iodine loading, and conversion to waste form.
- Co-absorption of water (tritium)/iodine on silver-exchanged mordenite has been evaluated. Rapid and significant tritium co-adsorption was observed (Figure 1) but with only limited short-term impact on iodine capacity.
- A silver (Ag^0)-functionalized silica aerogel for highly selective and efficient capture of gaseous iodine-129 was developed that, once laden with iodine, can be consolidated into a durable high-iodine-loaded silica-based waste form. Initial aging tests and deep-bed performance tests were completed.

- Removal of xenon and krypton was demonstrated at ppm concentrations from a simulated process off-gas stream at room temperature with Ni/DOBDC, HKUST-1 metal organic frameworks (MOFs), and demonstrated thermal switching of krypton/xenon selectivity with a FMOFCu MOF (Figure 2).
- A position paper that establishes practicable target performance criteria for the capture efficiencies for volatile radionuclides was completed. In this paper, the role of fuel age as well as operational considerations was examined.

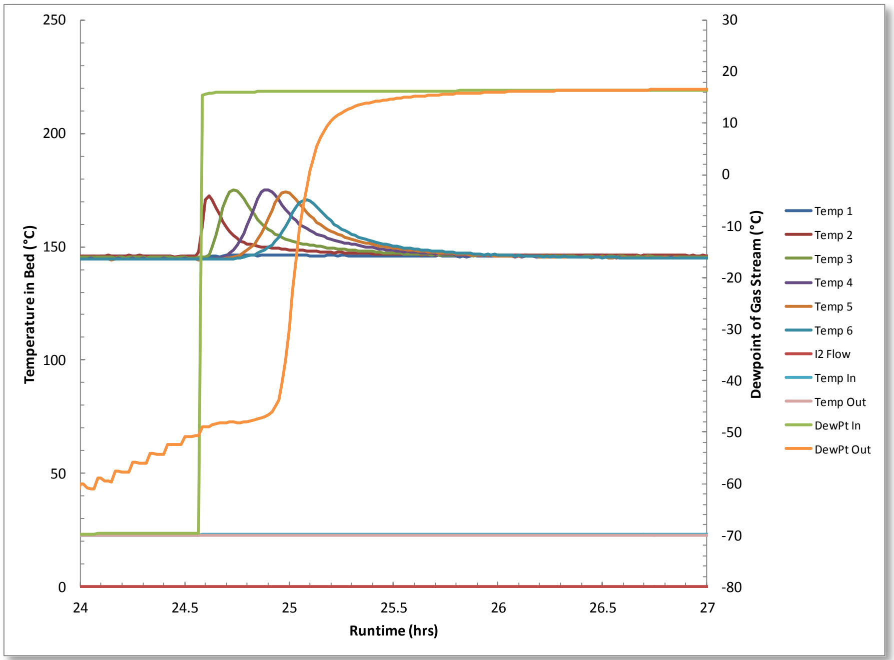


Figure 1: Progress of water (tritium) sorption front indicted by temperature raise and breakthrough curve.

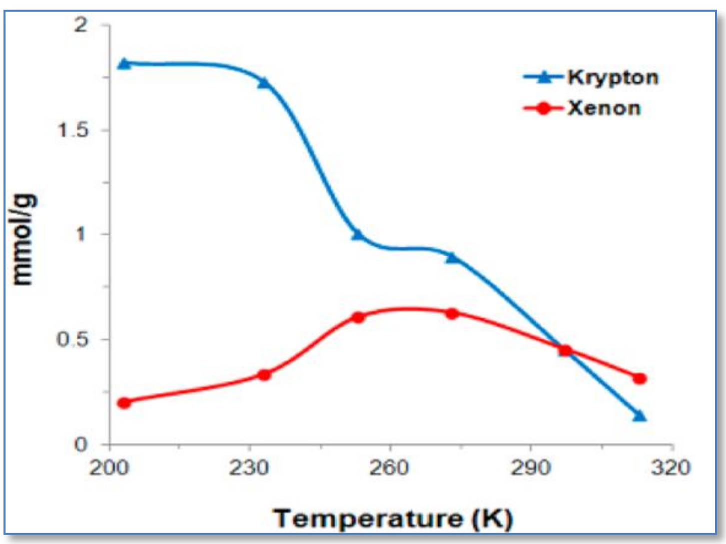


Figure 2: Adsorption of krypton and xenon as function of temperature on FMOFCu MOF.

3.4 Liquid–Liquid Contactor Computational Fluid Dynamics Modeling

Kent E. Wardle – Argonne National Laboratory

Introduction and Objectives

Solvent extraction process simulation tools are critical for advanced process design, evaluation, and optimization and, in turn, provide the quantitative basis for system-level evaluation of process options. Process-level simulations depend on both accurate chemical data and engineering understanding of unit operations performance over the range of process conditions. This effort seeks to deliver computational tools for simulation of solvent extraction contactor unit operations, providing a pathway for predicting key operational performance measures (e.g., stage efficiency and extent of separation) for any conditions using computational fluid dynamics (CFD). To this end, methods are required which can predict liquid–liquid mixing and interfacial area generation, as well as the formation and transport of small droplets.

Of the equipment types used for aqueous-based recycling of used nuclear fuel, annular centrifugal contactors have the largest relative knowledge gap and, at the same time, the greatest opportunity for significant benefits due to their compact size and efficiency. While the tools developed here are generally applicable to other solvent extraction equipment types, the present focus has been on prediction of the complex flow in centrifugal contactors. CFD simulations of these devices can provide insight into improvements in design, enable operational optimization over a wider range of conditions than is currently available, and provide tools to support the confident deployment of this technology in various areas of chemical processing.

Research and Development Overview

As multiphase CFD methods tend to be regime-dependent, the turbulent multiphase flows in centrifugal contactors and other liquid–liquid extraction devices present a unique challenge as they inherently span multiple flow regimes from fully phase-segregated, free-surface flow to fully dispersed multiphase flow. Successful simulation of such flows requires a computational framework capable of combining free-surface capturing methods with multi-fluid dispersed flow modeling. In addition, droplet breakup/coalescence models are needed to capture the evolution of the dispersed phase droplet size distribution in

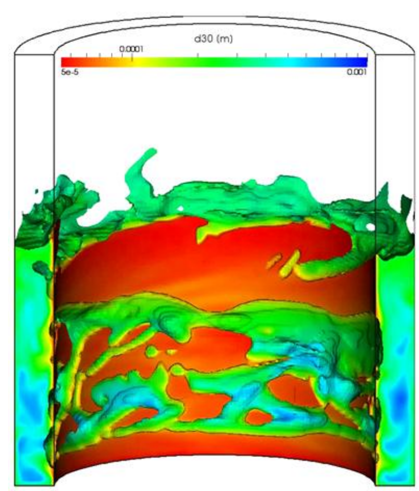


Figure 1. Dispersed phase droplet diameter in an annular mixer (3600 RPM).

contactor flows to predict liquid–liquid interfacial area and enable future implementation of interphase mass transfer for prediction of extraction efficiency. Experimental validation of these advanced models with detailed multiphase data from actual contactors is also essential.

Accomplishments

Using the open-source CFD toolkit OpenFOAM, researchers have developed a hybrid multiphase CFD solver based on the combination of an Eulerian multi-fluid solution framework (per-phase momentum equations) coupled with sharp interface capturing using volume of fluid (VOF) on selected phase pairs. The solver provides unique capability for multiple flow regime multiphase simulation previously non-existent among CFD packages and has been released as part of OpenFOAM v2.1. Relevant to solvent extraction, the solver enables three-phase, liquid–liquid–air simulations in which a sharp interface is maintained between each liquid and air, but dispersed phase modeling is used for the liquid–liquid interactions. The tool has also been extended to include a reduced population balance model to enable calculation of liquid–liquid interfacial area.

3.5 Microfluidic Solvent Extraction Kinetics

Kevin P. Nichols, Candido Pereira, Artem V. Gelis – Argonne National Laboratory

Introduction

One bottleneck in the design of next-generation solvent extraction-based nuclear fuel reprocessing schemes is a lack of interfacial mass transfer rate constants obtained under well-controlled conditions for lanthanide and actinide ligand complexes; such rate constants are a prerequisite for mechanistic understanding of the extraction chemistries involved and are of great assistance in the design of new chemistries. In addition, rate constants obtained under conditions of known interfacial area have immediate, practical utility in models required for the scaling-up of laboratory-scale demonstrations to industrial-scale solutions. Existing experimental techniques for determining these rate constants suffer from one of two fundamental drawbacks: slow mixing or unknown interfacial area. The volume required by traditional methods is an additional practical concern in experiments involving radioactive elements, both from disposal costs and from the reduced actinide concentrations required for experimental safety from process relevant conditions to tracer levels.

Existing solvent extraction kinetics tools consist primarily of either Lewis Cell type systems or highly stirred tanks, each of which has severe practical limitations. A Lewis Cell allows for well-defined interfacial areas in kinetics experiments through baffling and low-speed mixing. The primary disadvantage of a Lewis Cell is the low specific interfacial area typically available ($<50\ m^{-1}$) and the large reagent volumes required, which limit permissible actinide concentrations. A highly stirred tank allows for much greater interfacial areas ($2000\ m^{-1}$), approaching process equipment, but does not possess well-defined interfacial areas.

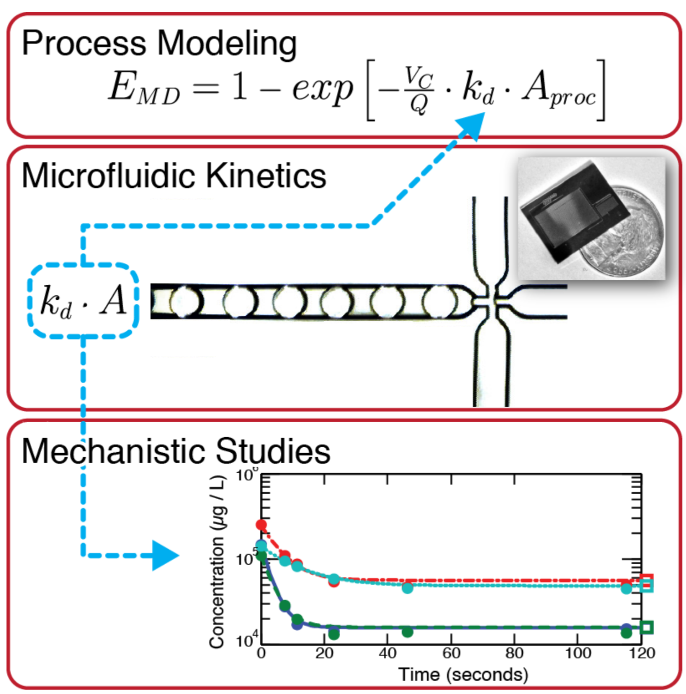


Figure 2: Schematic of a plug-based microfluidic system.

Research and Development Overview

In this work, the research team developed a plug-based microfluidic system that uses flowing plugs (droplets) in microfluidic channels to determine absolute interfacial mass transfer rate constants under conditions of both rapid mixing and controlled interfacial area with extremely high specific interfacial areas (10000 m^{-1}). The small absolute volumes also allow actinide experiments at concentrations approaching industrial loadings. The researchers utilized this system to determine the rate constants for interfacial transfer of all lanthanides, minus promethium, plus yttrium, under TALSPEAK process conditions. In addition, the team presents a path forward for utilizing their system to obtain well-defined interfacial areas for industrial processing equipment (such as centrifugal contactors or pulse columns) that can in turn be used to determine Murphree efficiencies. Knowledge of both rate constants and process interfacial areas is necessary for accurate dynamic modeling of solvent extraction systems.

Accomplishments

A microfluidic kinetics platform has been successfully demonstrated, as has been documented in one peer-reviewed publication and several conference proceedings. Current work involves utilizing the system in basic kinetics research and process modeling.

3.6 Exploration and Modeling of Structural Changes in Waste Glass under Corrosion

Karl Mueller – Pacific Northwest National Laboratory / Portland State University

Introduction and Objectives

Two general models have been proposed to explain the long-term leaching rate of waste glasses, the ***affinity concept*** and the ***protective layer concept.*** There is international agreement that both concepts are valid and must be taken into account when considering the decrease in dissolution rate. This project aims to characterize the molecular structure of the altered layers that form due to leaching and corrosion. The research team is combining aqueous-phase dissolution/reaction experiments with using state-of-the art analytical methods, such as nuclear magnetic resonance (NMR) spectroscopy, to probe the resulting surface layers.

Research and Development Overview

To investigate the rate of dissolution and reprecipitation from *complex* glasses in a mature state of corrosion, a protocol for analysis and interpretation is needed using *simplified* glass compositions. A set of isotopically substituted glasses and those with natural elemental abundances have been synthesized and allowed to corrode in equivalent conditions. Then the corrosion solutions from the enriched and natural isotopic glasses are swapped. Through the use of NMR, researchers can analyze the solid to identify the most active species in the altered layer.

Accomplishments

- Exchange of silicic acid between solution and gel has been confirmed, including changes in local coordination in the gel layer; this indicates a continuous remodeling of the gel layer. How these rearrangements impact transport is still to be determined.
- The presence and identity of ordered/crystalline species can be detected within the gel layer long before they can be observed by diffraction or microscopy.
- The speciation in the gel layers for simplified AFCI and SON68 are comparable even though their dissolution rates differ; thus, linkages between the species and the resulting density are critical characteristics for further evaluation of the competing models.

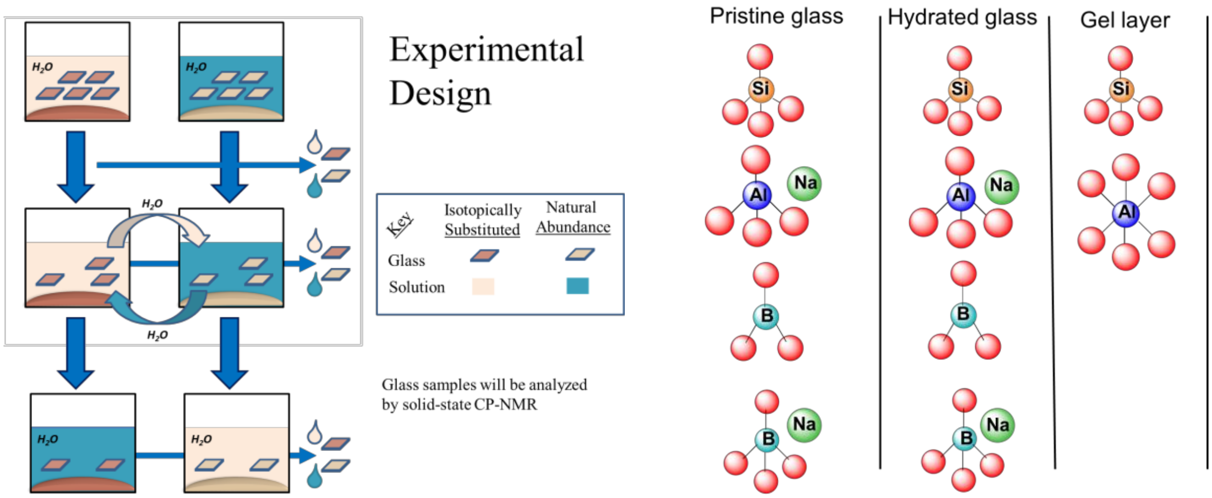


Figure 1: Chemical speciation for different layers of altered glass as determined by NMR analysis. Future work will focus on the determination of exact connections and further differences between the hydrated glass and gel layer.

Used Fuel Disposition Campaign

4.1 Used Fuel Disposition Campaign Overview

Campaign Mission

The Used Fuel Disposition (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.

Campaign Objectives

Near-Term Objectives (2013–2015)

- Conduct testing of high-burnup UNF cladding properties and canister performance.
- Develop a technical basis for licensing transportation systems designed to transport high-burnup UNF.
- Develop models for and conduct analyses of technical and logistical aspects of the interfaces between storage, transportation, and disposal to support decision making.
- Develop the scientific basis for multiple geologic options for permanent disposal of UNF and high-level radioactive waste, including computational models supported by experiments.

Long-Term Objectives (2015–2025)

- Collaborate with industry to field a full-scale NRC-licensed storage facility with monitoring and inspection capabilities to assess long-term performance.
- Collaborate with industry to support the transport of UNF from orphaned independent spent fuel storage installations (ISFSIs) to a consolidated storage facility.
- Develop and implement integrated storage, transportation, and disposal concepts that ensure safe and secure storage and transportation and timely disposal of waste.

Campaign Challenges

- Provide a sound technical basis for implementation of a new national policy 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.

Fiscal Year 2012 Funding

Used Fuel Disposition Campaign	
Major Activities	**FY 2012 Funding**
Crosscut Activities	$4,997,000
Disposal Research	$16,150,000
Storage and Transportation Research	$15,843,000
Total	**$36,990,000**

Major Research and Development Activities

Crosscut activities have included supporting development of the DOE strategy in response to the recommendations from the Blue Ribbon Commission on America's Nuclear Future, implementing the UFD international collaboration, conducting system-level evaluations of the UNF management system (storage, transportation, disposal interface), and supporting the Fuel Cycle Options Campaign's Screening and Evaluation of Fuel Cycle Options effort.

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. For example, uncertainties remain regarding high-burnup nuclear fuel cladding performance following possible hydride reorientation and creep deformation, and also regarding long-term canister integrity. Transportation R&D focuses on ensuring transportability of UNF following extended storage, addressing data gaps regarding nuclear fuel integrity, retrievability, and demonstration of subcriticality. Security R&D focuses on questions related to material attractiveness and self-protection due to surface dose rate, which decreases as UNF ages.

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

Key Fiscal Year 2012 Deliverables

- *Used Nuclear Fuel Storage and Transportation Research, Development, and Demonstration Plan,* March 2012, FCRD-FCT-2012-000053
- *Managing Aging Effects on Dry Cask Storage Systems for Extended Long-Term Storage and Transportation of Used Fuel,* June 30, 2012, FCRD-USED-2012-000119
- *Used Nuclear Fuel Storage and Transportation Gap Prioritization,* April 30, 2012, FCRD-USED-2012-000109
- *Phase I Ring Compression Testing of High-Burnup Cladding,* December 31, 2011, FCRD-USED-2012-000039
- *Evaluation of Generic EBS Design Concepts and Process Models: Implications to EBS Design Optimization,* June 15, 2012, FCRD-USED-2012-000140
- *Radionuclide Interaction and Transport in Representative Geologic Media,* June 20, 2012, FCRD-USED-2012-000154
- *Influences of Nuclear Fuel Cycles on Uncertainty of Long-Term Performance of Geologic Disposal Systems,* July 2012, FCRD-UFD-2012-000088
- *Modeling Coupled Processes in the Near Field of a Clay Repository,* August 15, 2012, FCRD-USED-2012-000223

4.2 Waste Management System Interface Analyses

Mark Nutt – Argonne National Laboratory

Introduction and Objectives

In the 1990s, the U.S. Department of Energy (DOE) completed a number of system analyses investigating consolidated interim storage as part of the waste management solution. These analyses are dated and do not reflect the present situation regarding at-reactor used nuclear fuel (UNF) management, alternatives for away-from-reactor UNF management, and alternatives for ultimate UNF disposal. The Blue Ribbon Commission for America's Nuclear Future and the Nuclear Waste Technical Review Board have pointed out the need for such analyses.

In Fiscal Year (FY) 2012, the Used Fuel Disposition Campaign initiated system-level analyses of the overall interface between at-reactor, consolidated storage, and ultimate disposition, along with development of supporting logistic simulation tools. The objective of this effort, which is continuing in FY 2013, is to provide DOE and other stakeholders information regarding the various alternatives for managing UNF generated by the current fleet of light water reactors operating in the United States.

Research and Development Overview

When developing waste management systems for the ultimate disposal of UNF , one important interface consideration is the need to size fuel assembly waste packages for compatibility with different geologic media. The Used Fuel Disposition Campaign completed thermal analyses, as presented by M. Fratoni at the 2011 Fuel Cycle Technologies Annual Review Meeting. Results indicate that waste package sizes for the geologic media under consideration by the Used Fuel Disposition Campaign are significantly smaller than the canisters being used for on-site dry storage by the nuclear utilities. Therefore, at some point along the UNF disposition pathway, there may be a need to re-package fuel assemblies already loaded into the types of dry storage canisters currently in use.

Figure 1 shows a high-level diagram of the alternative UNF disposition pathways, which involve UNF storage at a consolidated storage facility (CSF) and UNF packaging/re-packaging prior to ultimate disposal.

Accomplishments

The analysis began with the development of a disposition pathway framework, which is a detailed and comprehensive expansion of Figure 1.

While the reactors will continue to transfer UNF to dry storage, there will always be UNF in the used fuel pools, at least until a reactor is shut down and decommissioned. Another important aspect is how the UNF residing in the used fuel pools is managed when fuel acceptance from the reactor sites begins. UNF residing in the pools can be either transported off-site in re-useable transportation casks or placed in dual-purpose canisters suitable for both storage and transportation. This choice affects the design of both a CSF (canistered fuel storage only or canistered and bare fuel storage) and the quantity of UNF that would ultimately have to be re-packaged.

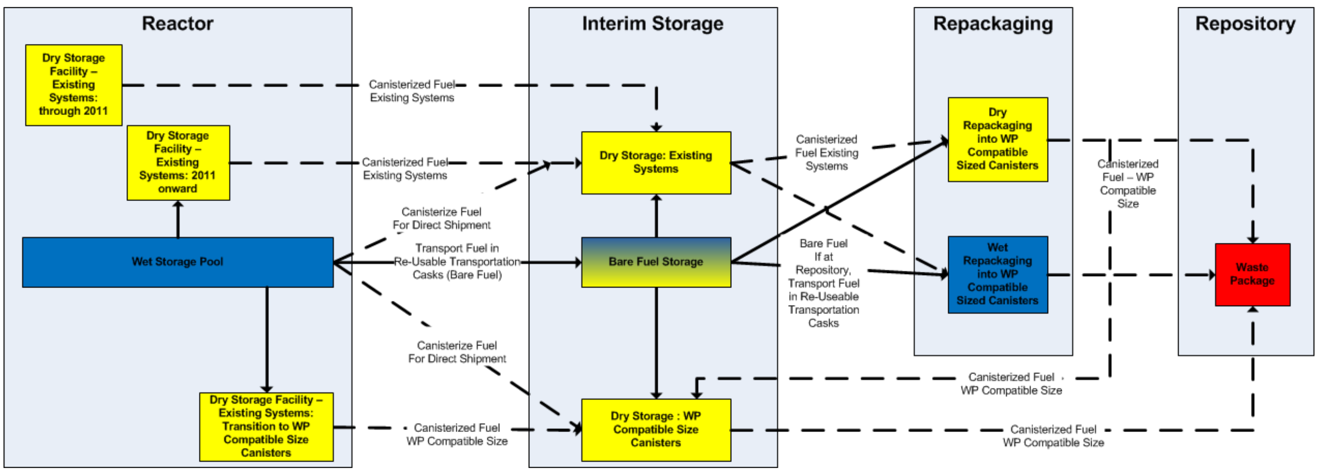


Figure 1: Alternative UNF disposition pathways

These considerations resulted in the identification of nine potential disposition pathways addressing how UNF would be transported from the reactors, where UNF packaging/re-packaging would be performed (repository or CSF), and when UNF packaging/re-packaging would be performed (at CSF receipt or prior to shipment from the CSF to a repository). The analysts then evaluated these nine disposition pathways considering complexity and flexibility, resulting in a down-selection of the disposition pathways for consideration in FY 2012 (re-packaging at CSF receipt was omitted).

A range of input parameters was then determined for evaluating each disposition pathway. Parameters selected include start of CSF operations (2020 or 2035), start of repository operations (2040 or 2055), UNF acceptance rates (1500, 3000, and 6000 MTHM/yr), and waste package size (4, 12, or 21 PWR assemblies and 9, 24, or 44 BWR assemblies). Combining disposition pathways and input parameters, the analysts evaluated a total of 36 individual scenarios.

In parallel, the research group developed the Transportation–Storage Logistics (TSL) simulator. They began with legacy UNF logistics simulators previously developed by DOE, then modified and enhanced them to produce a logistic simulator capable of modeling the range of disposition pathways and input parameters discussed above. The TSL was used to simulate each of the 36 individual scenarios, providing information on a range of logistic parameters including quantities of UNF in at-reactor dry storage, shipping rates for the different types of dry storage canisters and bare-fuel assemblies from the reactors, receipt rates at the CSF and the repository, quantities of UNF and canister types in dry storage, and the number of canisters and UNF assemblies that are packaged and re-packaged.

This quantitative logistic information was then used as input to pre-conceptual design work for consolidated storage facilities, packaging/re-packaging facilities, and repository surface facilities. The CSF design concepts and facility sizes differ depending on the scenario and UNF receipt rates (vertical, horizontal, and bare fuel storage). In addition, the existing legacy and continued use of dual-purpose canisters (and single-purpose storage casks) must be managed, and the inventory and mix of canisters (vertical/horizontal) depends heavily on the scenario and canister receipt rates, directly influencing the CSF and packaging/re-packaging facility design concepts. The strategy for managing UNF in fuel pools once CSF begins operation also affects these design concepts.

4.3 Sensitivity Analyses Supporting a Generic Repository Model for Fuel Cycle Analysis

Kathryn D. Huff – Argonne National Laboratory

Introduction and Objectives

Repository metrics such as necessary repository footprint and peak annual dose could be affected by heat and radionuclide release characteristics specific to variable spent fuel compositions associated with alternative nuclear fuel cycles. For this reason, a generic disposal model designed for integration with a systems analysis framework is necessary for illuminating performance distinctions of potential repository geologies and facility design concepts (including engineering components) in the context of nuclear fuel cycle options.

To calculate geologic repository behavior metrics with respect to candidate fuel cycle options, the Cyder software library under development will be integrated with the Cyclus computational fuel cycle systems analysis platform. By abstraction of more detailed geologic repository performance models (i.e., thermal and radionuclide transport), Cyder aims to capture the dominant physics of important phenomena affecting repository performance in various geologic media and as a function of arbitrary waste composition (i.e., directly disposed used fuel or high-level waste generated from recycling).

To support this abstraction effort in the area of long-term repository performance, sensitivity analyses were performed with respect to various key processes and parameters affecting long-term post-closure performance of geologic repositories in clay media. Based on the detailed computational clay generic disposal system model (GDSM) developed by the Used Fuel Disposition (UFD) Campaign, these results provide an overview of the relative importance of processes that affect the repository performance of a generic clay disposal concept model.

Research and Development Overview

A generic repository model appropriate for nuclear fuel cycle systems analysis must emphasize modularity and speed. The sensitivity analysis conducted using the UFD clay GDSM tool captures the dominant physics of detailed repository performance analysis so that abstracted models can be robustly and flexibly implemented in Cyder without sacrificing simulation speed.

The clay GDSM radionuclide transport toolset, built on the GoldSim simulation framework and contaminant transport model, simulates chemical and physical attenuation processes including radionuclide solubility, dispersion phenomena, and reversible sorption. Input parameters supporting modeling of these transport processes include geometry specifications (e.g., repository depth), geologic material properties (e.g., clay porosity), geochemical data (e.g., elemental solubility limits), and environmental parameters that affect repository performance (e.g., natural system velocity).

Accomplishments

This work utilized an analysis strategy to develop a multi-dimensional overview of the key factors in modeled repository performance. Both individual and dual parametric cases were performed, and repository performance was quantified by means of the peak annual radiation dose to a hypothetical receptor. Individual parameter cases varied a single parameter of interest in detail over a broad range of values. Dual parameter cases were performed for pairs of parameters expected to exhibit some covariance. For each parameter or pair of parameters, forty simulation groups varied the parameter or parameters within the range considered. Each case and its parametric range are detailed in Table 1. For each simulation group, a 100-realization simulation was completed.

Table 1. Parameter Range for Clay GDSM Sensitivity Analyses

Simulation Cases

Case	Parameter	Units	Min. Value	Max. Value
I	D_{eff}	$[m^2 \cdot s^{-1}]$	10^{-8}	10^{-5}
	Inventory	[MTHM]	10^{-4}	10^{1}
II	$V_{adv,y}$	$[m \cdot yr^{-1}]$	6.31×10^{-8}	6.31×10^{-4}
	D_{eff}	$[m^2 \cdot s^{-1}]$	10^{-8}	10^{-5}
III	S_i	$[mol \cdot m^{-3}]$	$(1 \times 10^{-9})\langle S_i \rangle$	$(5 \times 10^{10})\langle S_i \rangle$
IV	$K_{d,i}$	$[m^3 \cdot kg^{-1}]$	$(1 \times 10^{-9})\langle K_{d,i} \rangle$	$(5 \times 10^{10})\langle K_{d,i} \rangle$
V	$R_{WFDeg.}$	$[yr^{-1}]$	10^{-9}	10^{-2}
	Inventory	[MTHM]	10^{-4}	10^{1}
VI	t_{WPFail}	$[yr]$	10^{3}	10^{7}
	D_{eff}	$[m^2 \cdot s^{-1}]$	10^{-8}	10^{-5}

The results of this work illuminated the character of transitions between diffusive and advective transport regimes, highlighted the importance of solubility and partitioning coefficients, and demonstrated the relatively limited importance of waste form degradation rate when the diffusive geologic pathway was the primary barrier to release. These insights are currently are being implemented in combination with analytic models of solute transport to improve the speed and accuracy of nuclide transport models implemented in the Cyder repository analysis library.

4.4 Fuel Aging in Storage and Transportation (FAST): Accelerated Characterization and Performance Assessment of the Used Nuclear Fuel Storage System

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Introduction and Objectives

This integrated research project (IRP) involves six university and two national laboratory partners. The overall objective is to create predictive tools in the form of observation methods, phenomenological models, and databases that will enable the design, installation, and licensing of dry used nuclear fuel (UNF) storage systems that will be capable of containing high burnup UNF for extended periods. The FAST IRP is focused on four distinct yet integrated technical mission areas (TMAs):

- TMA1: Low-Temperature Creep
- TMA2: Hydrogen Behavior and Delayed Hydride Cracking
- TMA3: UNF Canister Corrosion
- TMA4: Novel System Monitoring

The first two technical missions deal with potential cladding failure phenomena that would complicate future transportation, storage, and/or processing operations. The third mission is focused on experimental characterization of UNF stainless steel canister behavior and creation of a predictive model. The fourth mission is to develop long-term monitoring methods to equip future decision makers with real-time information regarding the internal state of the UNF being stored.

Research and Development Overview

TMA-1: Low-Temperature Creep focuses on the low-temperature creep of UNF cladding enabled by decay heat from fission products and stress arising from internal pressure. The major objectives are to 1) obtain creep data using highly oxidized/hydrided tubing under relevant stresses and temperatures, 2) characterize and translate that data to enable input to codes developed to predict UNF behavior in dry storage, and 3) formulate atomistic simulations to understand long-term creep behavior.

TMA-2: Hydrogen Behavior and Delayed Hydride Cracking focuses on characterization and understanding of delayed hydride cracking (DHC) in UNF cladding (i.e., Zircaloy). Samples with low and high hydrogen loadings are being prepared for these studies using proton irradiation, electrochemical methods, and gas phase intrusion. Ion irradiation will also be used to "accelerate" DHC mechanisms to enable long-term behavior modeling.

TMA-3: Corrosion of UNF Canisters addresses currently recognized gaps in understanding mechanisms relevant to UNF canister corrosion. The objectives include 1) electrochemical corrosion testing, 2) stress corrosion cracking (SCC) studies, 3) characterization of the corrosion of welds, bolted joints, and seals under prototypic conditions, and 4) integration of the results of corrosion tests and microstructural analyses to develop predictive models.

TMA-4: Novel System Monitoring will develop *in situ* prognostic (predictive) monitoring methods to verify the condition of the UNF and the internal canister environment without having to open the inner cask. This will include the development of self-powered sensors to provide physical, safety, and security information, both during storage and to the end user that may be transporting UNF for ultimate disposal or reprocessing. The parameters of interest include internal temperatures, internal pressure, and the atmospheric concentration of O_2, H_2O, and krypton in the helium cover gas.

Program Integration Activities: The FAST IRP team is actively integrating with various UNF storage programs, including the Department of Energy Used Fuel Disposition Campaign (primary integration point), the Electric Power Research Institute's (EPRI's) Extended Storage Collaboration Project (ESCP), as well as dialogue with U.S Nuclear Regulatory Commission personnel. Further, dialogue has begun regarding information sharing with the Korean Atomic Energy Research Institute's (KAERI) used fuel storage program.

Accomplishments

The first year of the program has been focused on establishing methods, databases, and codes while forming the implementation and integration strategy for the various mission areas. Highlights from the first six months include the following:

- Multiple experimental systems have been assembled to evaluate the insertion of hydrogen into zirconium alloys. Initial results from electrochemical methods have produced clear rim structures, and vapor phase reactions have produced hydride gradients.
- FRAPCON 3.4 is being used with customized input and time-step strategies to simulate cladding creep performance using the DATING creep model.
- An updated model for low-temperature creep has been created, and validation is under way.
- A suitable interatomic potential has been identified for simulating zirconium behavior in cladding modeling simulations.
- Corrosion testing has been initiated using candidate alloys 304L and 316L in seawater/ coastal conditions using "Salt-On-Sample Hydration" tests and electrochemical methods using prototypic seawater (specified in ASTM D1141).
- A test system has been established to measure stress-induced crack growth in 304L and 316L under various environments.
- An array of sensor and monitoring options has been identified, and initial designs are being tested (patents pending). The first-year products are focused on external methods for canister corrosion, but internal sensors (the harder problem) are also being developed.

Material Protection, Accounting, and Control Technologies Campaign

5.1 Material Protection, Accounting, and Control Technologies Campaign Overview

MPACT Mission

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

MPACT Objectives

- Develop tools, technologies, and approaches in support of used fuel security for extended storage.
- Develop and apply new tools to assess proliferation and terrorism risk.
- Develop and demonstrate advanced material control and accounting technologies that would, if implemented, fill important gaps in existing MPACT capabilities.
- Develop, demonstrate and apply MPACT analysis tools to assess effectiveness and efficiency of MPACT systems and guide R&D.
- Develop guidelines for safeguards and security by design and publish international guidance documents.

MPACT 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.

Technical needs

- Improve the precision and accuracy of key nuclear material measurements.
- Improve the timeliness and cost-effectiveness of measurements and analysis.
- Expand the scope of detection to include more indicators, taking advantage of existing data where possible and new sources of data where appropriate.
- Expand and strengthen detection and assessment algorithms to exploit larger data sets and provide results in near-real time.
- Model and simulate MPACT performance against a wide spectrum of assumed threats and rigorously demonstrate MPACT effectiveness and efficiency in future U.S. nuclear energy systems.
- Integrate safeguards and security into the design of future nuclear fuel cycle facilities from the earliest stages of the design cycle.

Fiscal Year 2012 Funding

MPACT Campaign	
Major Activities	**FY 2012 Funding**
Campaign Management and Ad Hoc Technical Support	$500,000
Material Control and Accounting Technologies	$1,900,000
MPACT Analysis Tools	$1,550,000
Safeguards and Security by Design	$2,550,000*
Total	**$6,500,000**

*Includes $2,000,000 from the Nuclear Reactor Technology program to initiate the Proliferation and Terrorism Risk Assessment project

Major Research and Development Activities

The MPACT campaign continued to re-baseline in FY 2012 to shift focus towards emerging priorities in used fuel security for extended storage, proliferation and terrorism risk assessments, and instrumentation modeling to support MPACT capability enhancement for electrochemical processing.

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

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

Safeguards and Security by Design is a methodology and discipline for integrating nonproliferation and security considerations into the design of nuclear facilities from the 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. The security aspects of used fuel extended storage are being evaluated within the safeguards and security by design framework to support disposition activities and identify R&D needs.

Key Fiscal Year 2012 Deliverables

- *Document Proliferation and Security Evaluation Criteria*, using a cost-based approach driven by NRC licensing requirements, in support of fuel cycle options screening. Determining proliferation and nuclear material security risk for nuclear fuel cycles is challenging. Since NRC licensing must take risk to the public into account, a cost-based approach is one method of comparing different fuel cycles.
- *Test Electrochemical Actinide Sensor Designs*, in lab-scale configuration and process-relevant temperatures, to determine impedance and compatibility with the electrolyte. Electrochemical processing presents challenges for nuclear materials management and accounting. Developing sensors that can directly measure actinides in the electrochemical process environment will greatly advance the ability manage, account and control nuclear materials.
- *Perform Proof-of-Concept Imaging Measurements*, in a configuration simulating process holdup, to quantify special nuclear materials. Process holdup inevitably represents a major source of uncertainty in nuclear materials management, particularly for bulk facilities. Radiation-based imaging can provide improvements in both accounting as well as process operations (e.g., criticality safety, isotopic blends, etc.).

- *Develop Automatic Algorithm for the Multi-Isotope Process Monitor*, capable of isotope identification and assessing spent fuel parameters such as cooling time and burn up. This technology provides earlier detection of possible nuclear material diversion and also has potential to improve process operations by detecting unwanted deviations in process chemistry.
- *Demonstrate Kilohertz Count Rate Capability of Microcalorimeter*, using the high-yield 256-pixel array and a high-burnup plutonium sample, to demonstrate practical utility of the technique. Microcalorimetry improves the resolution of gamma-ray spectroscopy, a standard method in nuclear material accounting, by an order of magnitude. This capability not only improves accuracy but also can reduce the number of samples needed for chemical analysis (which is costly and less timely).
- *Develop a Performance Model for Active Interrogation*, using liquid scintillator detectors and including fast neutron multiplicity analysis, to optimize system parameters. Thermal neutron multiplicity counting is a fission-based method currently in use for nuclear material management and accounting. By extending this technique to fast neutrons, and potentially gamma-rays, accurate measurements of advanced fuel cycle materials will be possible that are currently a challenge to standard techniques.
- *Complete Baseline Electrochemical Process Model*, including capability to perform MPACT sensitivity analyses to aid in overall system optimization and identify capability gaps. Models like this can aid in understanding where the greatest leverage is in terms of R&D investment to improve nuclear materials management, accounting and control.

5.2 Used Fuel Storage Security Analysis, Guidance, and Best Practices

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Scott DeMuth – ***Los Alamos National Laboratory***

Introduction and Objectives

In light of the Blue Ribbon Commission (BRC) report, lessons learned from the accident at Fukushima, and a variety of other factors, increased emphasis is being placed on extended storage of used fuel, including dry storage, potentially for many decades. In addition to domestic security needs, the BRC stressed the importance of "active U.S. leadership in international efforts to address safety, nonproliferation and security concerns." In FY 2012, two efforts were initiated in the MPACT campaign related to the security of extended used fuel storage. These efforts are focused on technical analyses and guidance documents needed to assure that the security risks associated with extended storage are understood and minimized, and that reliable and technically sound information is available to address any stakeholder concerns that may arise. Specifically, Sandia National Laboratories (SNL) and Los Alamos National Laboratory (LANL) are working to develop (1) a prioritized issues list for used fuel storage security and (2) best practices guidance for security of dry-cask used fuel storage facilities.

Research and Development Overview

Efforts to develop a prioritized issues list extend work performed in the Used Fuel Disposition (UFD) Campaign on security issues relevant to extended storage; these efforts included review of the 2006 National Academy of Sciences evaluation of spent fuel safety and security. In addition, the research team considered issues from the BRC report specific to security of used fuel as well as input from other technical experts. The prioritized issues list will provide the basis for R&D objectives for used fuel storage security in the MPACT campaign.

For the best practices guidance, SNL and LANL will work with the World Institute for Nuclear Security (WINS) to research information to develop a best practices workshop focused on the security of dry-cask storage of used fuel. WINS will then host, facilitate, and lead the international workshop and collect lessons learned from personnel with experience directly related to security of used fuel storage. With SNL and LANL participation, WINS will also develop a best practice guide based on workshop results.

Accomplishments

For FY 2012, the following deliverables have been prepared: joint SNL/LANL reports on (1) the prioritized issues list for used fuel storage security and (2) initial recommendations on development of best practices for used fuel storage security (LA-UR-12-22831 included SNL input). In addition, the WINS contract awarded through LANL but not yet issued is awaiting DOE-NE FCR&D FY13 funding decisions.

Plans for FY 2013 include development of MPACT R&D objectives for used fuel security, issuance of the WINS contract, execution of the best practices workshop for security of dry-cask storage of used fuel, preparation of the related best practices guidance document, and continuation of security assessments for extended storage initiated under the UFD Campaign.

5.3 Silicon Carbide Schottky Diode Detectors for Measurement of Actinide Concentrations from Alpha Activities in Molten Salt Electrolyte

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Introduction and Objectives

Silicon carbide (SiC) Schottky diode detectors offer a new means to detect alpha particles in harsh environments, since SiC is radiation hard and chemically inert. Among the potential applications of such detectors is real-time monitoring of alpha emitter inventories in a pyrochemical fuel processing stream. This project encompasses a collaborative effort between researchers from two departments at The Ohio State University: Materials Science and Engineering and Nuclear Engineering. The team is investigating the use of SiC Schottky diode detectors for measuring the concentrations of actinides in the process stream for pyrochemical processing of spent nuclear reactor fuel. The project includes device fabrication, testing, and a modeling-assisted study to demonstrate detector function with device-modeling software. The final step links damage to detector performance, reliability and lifetime, including an experimental and computational study of post-irradiation structural evolution as well as modeling the influence of damage on the detector signal.

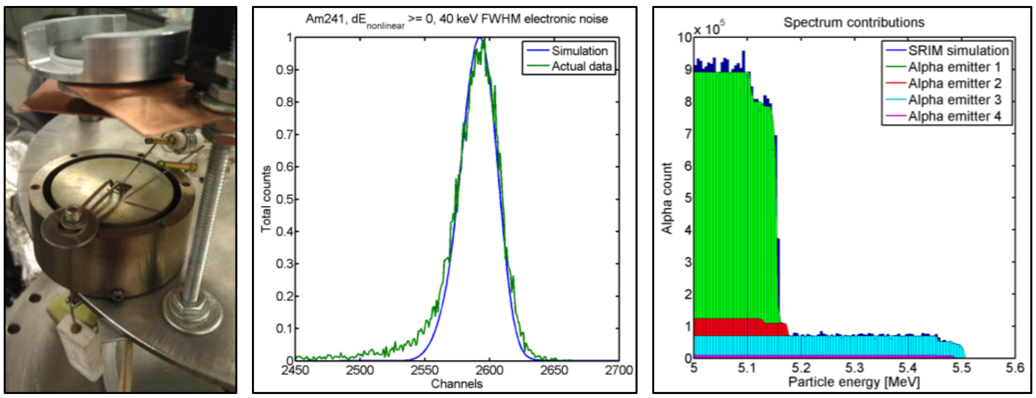
Research and Development Overview

Schottky barrier diode detectors with nickel contacts were fabricated from 4H-SiC wafers, contacted with mechanical micro-positioning electrical clamps, and tested in a vacuum test chamber with an electroplated americium-241 disk source. Spectra for several detectors were taken at room and increasing temperatures until a discernible peak was no longer present. The device fabrication was mirrored by device modeling, which was calibrated to reproduce the measurements for the americium-241 alpha source. The device modeling results were then coupled with SRIM and MATLAB codes in order to predict measurement of characteristic alpha particles that would be present in Idaho National Laboratory’s (INL) Mark V electrorefiner. Finally, efforts are currently under way to use a combination of modeling and experiments at The Ohio State University Research Reactor to study radiation-induced damage.

Accomplishments

During FY 2012, a number of significant tasks were accomplished. Working 4H-SiC Schottky diode detectors were tested up to 500°C, with an increase of full width at half maximum

(FWHM) and shift in centroid, which was successfully replicated through device modeling. The alpha particle spectrum in the INL Mark V electrorefiner was then simulated and shown to be spread out in energy because of salt screening; however, the simulations show that measurements will be able to recover both alpha emitter energy and isotope concentration. This knowledge led to determination of a relationship between detector resolution and detectability of isotopes.



(a) (b) (c)

Figure 1: (a) a 6-mm square detector placed in the test facility on a high-temperature stage, with electrical probes, americium-241 source, and copper shutter; (b) fabricated 4H-SiC detector americium-241 spectra, and simulation result using Sentaurus device and SRIM; (c) ideal 4H-SiC detector room temperature simulated spectrum of the Mark V electrorefiner at INL.

Appendix A. Acronyms

Appendix A. Acronyms

3-D	Three-Dimensional
Ag	Silver
Al	Aluminum
ASTM	American Society for Testing and Materials
ATR	Advanced Test Reactor
B&B	Breed and Burn (reactor)
BRC	Blue Ribbon Commission (on America's Nuclear Future)
BWR	Boiling Water Reactor
C	Carbon
C	Celsius
ca.	Circa
CEA	Commissariat à l'énergie atomique et aux énergies alternatives
CFD	Computational Fluid Dynamics
Cr	Chromium
CSF	Consolidated Storage Facility
Cu	Copper
DF	Decontamination Factor
DHC	Delayed Hydride Cracking
DOBDC	2,5-dioxido-1,4-benzenedicarboxylate
DOE	U.S. Department of Energy
dpa	Displacements per Atom
EBR	Experimental Breeder Reactor
EBSD	Electron Backscatter Diffraction Analysis
EDS	Energy Dispersive Xray Spectroscopy
EPMA	Electron Microprobe Analysis
EPRI	Electric Power Research Institute
EROI	Energy Return on Investment
ESCP	Extended Storage Collaboration Project
FAST	Fuel Aging in Storage and Transportation
FBR	Fast Breeder Reactor
FCCI	Fuel Cladding Chemical Interaction
FCM	Fully Ceramic Microencapsulated
FCRD	Fuel Cycle Research and Development (program)
FCT	Fuel Cycle Technologies (program)
Fe	Iron

FEA	Finite Element Analysis
FFTF	Fast Flux Test Facility
FIB	Focused Ion Beam
FIMA	Fissions Per Initial Metal Atom
FIR	Fissile Inventory Ratio
FP	Fission Product
FWHM	Full Width at Half Maximum
FY	Fiscal Year
GDSM	Generic Disposal System Model
GWd	Gigawatt Day(s)
H	Hydrogen
HCWR	High-Conversion Water Reactor
HFIR	High Flux Isotope Reactor
HKUST-1	$[Cu_3(btc)_2(H_2O)_3]\cdot xH_2O$ (a metal organic framework)
IBAD	Ion-Beam Assisted Deposition
INL	Idaho National Laboratory
IRP	Integrated Research Project
ISFSI	Independent Spent Fuel Storage Installation
JAEA	Japan Atomic Energy Agency
KAERI	Korea Atomic Energy Research Institute
kg	Kilogram
LANL	Los Alamos National Laboratory
LTA	Lead-Test Assembly
LWR	Light Water Reactor
m	Meter(s)
mm	Millimeter
MOF	Metal Organic Framework
MOX	Mixed Oxide
MPACT	Material Protection, Accounting, and Control Technologies
MTHM	Metric Ton of Heavy Metal
MWd	Megawatt-Day(s)
N	Nitrogen
NEAMS	Nuclear Energy Advanced Modeling and Simulation Program
NEUP	Nuclear Energy University Programs
NMR	Nuclear Magnetic Resonance
NRC	U.S. Nuclear Regulatory Commission
O	Oxygen
ODS	Oxide Dispersion-Strengthened

OOF	Object-Oriented Finite Element (software)
ORNL	Oak Ridge National Laboratory
PIE	Post-Irradiation Examination
ppb	Parts Per Billion
ppm	Parts Per Million
PRISM	Power Reactor Innovative Small Module
Pu	Plutonium
PWR	Pressurized Water Reactor
R&D	Research and Development
RD&D	Research, Development and Demonstration
RBWR	(GE-Hitachi) Resource-Renewable Boiling Water Reactor
RPM	Revolutions Per Minute
SEM	Scanning Electron Microscopy
SFR	Sodium Fast Reactor
SiC	Silicon Carbide
SNL	Sandia National Laboratories
SRIM	Stopping and Range of Ions in Matter
TALSPEAK	Trivalent Actinide–Lanthanide Separation by Phosphorous Reagent Extraction from Aqueous Komplexes
TEM	Transmission Electron Microscope
THM	Tons of Heavy Metal
Ti	Titanium
TMA	Technical Mission Area
TRISO	Tristructural Isotropic
TRU	Transuranium (elements)
TSL	Transportation–Storage Logistics (simulator)
U	Uranium
UC	University of California
UFD	Used Fuel Disposition (campaign)
UN	Uranium Nitride
UNF	Used Nuclear Fuel
VOF	Volume of Fluid
w/o	Weight Percent
WINS	World Institute for Nuclear Security
Zr	Zirconium