Materials Recovery and Waste Form Development Campaign

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U.S. Department of Energy

Nuclear Waste Technical Review Board Meeting
November 20, 2013
Outline

Introduction – Program Drivers
Campaign Structure
Uranium from Seawater
Tritium Treatment
Fuel Cycle Gaps
  – Sigma Teams
  – Domestic Electrochemical Process
  – Advanced Waste Forms
Partnerships
Summary and Conclusion
Program Drivers

Nuclear Energy

Research Objective 3
Implementation Plan
Developing Sustainable Fuel Cycle Options

NUCLEAR ENERGY RESEARCH AND DEVELOPMENT ROADMAP
REPORT TO CONGRESS
April 2010

STRATEGY FOR THE MANAGEMENT AND DISPOSAL OF USED NUCLEAR FUEL AND HIGH-LEVEL RADIOACTIVE WASTE
JANUARY 2013
**Mission**

- Ensure America’s security and prosperity by addressing its energy, environmental, and nuclear challenges through transformative science and technology solutions.
  
  **Goal 3: Secure Our Nation,**
  
  *Enhance nuclear security through defense, nonproliferation, and environmental efforts.*

- Advance nuclear power as a resource capable of making major contributions in meeting the Nation’s energy supply, environmental, and energy security needs by resolving technical, cost, safety, security and regulatory issues through research, development, and demonstration.

- Develop used fuel waste management strategies and sustainable fuel cycles that improve resource utilization, minimize waste generation, improve safety and limit proliferation risk.

**Program Objectives**

- Address BRC recommendations for Used Fuel Disposition.
- Increase focus on accident tolerant fuels.
- Down select fuel cycle options for further development.

- Conduct science-based, engineering-driven R&D for selected fuel cycle options.
- Complete plans for developing a dry storage demonstration project for extended storage of used nuclear fuel.
- Evaluate benefits of various geologic media for disposal.

- Demonstrate the selected fuel cycle options at engineering scale.
- Operate a dry storage demonstration project for extended storage of used fuel.
- Conduct engineering analysis of disposal site(s) for selected geologic media.
### Office of Fuel Cycle Technologies: an Integrated Approach

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**Optimize through systems analysis, engineering, and Integration**

**Safeguards and Security By Design**
Fuel Cycle as a System

**Uranium Supply**
- Conventional / Mining
- Seawater Extraction
- Other Advanced Technique

**Advanced Fuels**
- Conventional LWR Fuel Fabrication
- Light Water Reactors
- LWR Fuel w/ Improved Accident Tolerance
- Fast Reactor Fuel
- Fast Reactors - Sodium - Lead - Gas

**Reactors**
- +Electricity +Industry

**Separations Technology**
- Reprocess / Separate
- Pre-treat / Condition
- Waste Form
- Fast Rx Recycle

**Storage**
- Interim Storage
- Geology X - TBD

**Disposal**
- Interim Storage
- Geology X - TBD

- **Optimized System:** We want the best performance for each step in harmony with other parts of the system
- **Near-Term/Long-Term Balance:** Seek near-term applications while maintaining the long-term objective of a sustainable fuel cycle
Campaign Objectives

- Develop advanced fuel cycle material recovery and waste management technologies that improve current fuel cycle performance and enable a sustainable fuel cycle, with minimal processing, waste generation, and potential for material diversion to provide options for future fuel cycle policy decisions.

- Campaign strategy is based on developing:
  - **Technologies** for economical deployment
    - *Concept through engineering-scale demonstration*
  - **Capabilities** for long-term science-based, engineering driven R&D, technology development and demonstration
  - **People** to provide the next generation of researchers, instructors, regulators and operators
MrWFD Campaign Leadership

- Terry Todd, INL, National Technical Director
- John Vienna, PNNL, Deputy National Technical Director
- Jim Bresee, DOE-NE, Federal Program Director
- Stephen Kung, DOE-NE, Federal Program Director
- Kimberly Gray, DOE-NE, Federal Program Director
Campaign Implementation Plan

<table>
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<tr>
<th>Fiscal Year</th>
<th>Case study technologies</th>
<th>LWR fuel reprocessing plant</th>
<th>Fast reactor fuel reprocessing plant</th>
<th>Glass ceramics</th>
<th>Develop high durability Tc waste form</th>
<th>Epsilon metal for Tc and noble metals</th>
<th>Fast reactor electrochemical salt waste forms</th>
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<th>Fuel fabrication transuranic waste forms</th>
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Thrust Area – Fuel Cycle Overlay

Near-term & Open Fuel Cycle

Uranium from seawater

Tritium Separations / Treatment

Long-term & Closed Fuel Cycle

Off-gas Sigma Team

Waste Form Performance

Waste Form and Process Development

Aqueous Recycle Technology

Electrochemical Recycle Technology
"The research effort for extracting uranium from seawater began in FY-11 with the goal of doubling the baseline sorption capacity previously established by Japanese researchers."
Uranium from Seawater
Accomplishments

- Advanced understanding of U absorption on amidoximes
- Developed improved absorbents based on improved understanding
- Demonstrated roughly 3× capacity of new material in laboratory testing
- Developed preliminary cost study to identify key drivers and uncertainties
- Began to evaluate biofouling, competitive absorption, elution process, material lifetime

**Ligand Design – Predicted Binding Affinity via DFT Calculations**

*Calculated Literature*

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**Graphical Data**

- **Ligand Saturation Model**
- **ORNL Cartridge**
- **Japanese Sorbent**
- **Replicate**

*Uranium*:
- 0 µg Uranium/g adsorbent
- 500 µg Uranium/g adsorbent
- 1000 µg Uranium/g adsorbent
- 1500 µg Uranium/g adsorbent
- 2000 µg Uranium/g adsorbent
- 2500 µg Uranium/g adsorbent
- 3000 µg Uranium/g adsorbent

*Days of Exposure*:
- 0 days
- 10 days
- 20 days
- 30 days
- 40 days
- 50 days
- 60 days

*Uranium Absorption*:
- 0 µg Uranium/g adsorbent
- 50 µg Uranium/g adsorbent
- 100 µg Uranium/g adsorbent
- 150 µg Uranium/g adsorbent
- 200 µg Uranium/g adsorbent
- 250 µg Uranium/g adsorbent
- 300 µg Uranium/g adsorbent

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**Graphical Representation**

- Plot of ligand saturation model over days of exposure.
- Data points indicating uranium uptake over time.
- Comparison with other sorbents and cartridges.
The campaign has planned a study to evaluate and develop materials/processes for tritium separations from high volume water for application to:

- reactor operation, nuclear accident scenarios, fuel processing, (may also benefit legacy waste site cleanup)

PWR operation generates $^3$H. Discharge of which is a public perception, and potentially a regulatory, challenge.

Nuclear accidents (e.g., TMI and Fukushima) generate high-water volumes with sufficient $^3$H to preclude free release.

Recycle operations may produce high volume of tritiated water that must be immobilized and disposed.
Closed Fuel Cycle Technology Gaps

- Minor Actinide Separations from LWR Fuel
- Off-gas Management
- Waste Forms & Processes
- Cost Effective and Appropriate for Industry Deployment
- FR Fuel Separations
- Fundamental Understanding

Present: Fundamentals
Future: Deployment
To address some of the most difficult challenges in the separations and waste forms area, we formed two “Sigma Teams”

- Multidisciplinary, multi-laboratory teams of experts focused on solving a single challenge
- Greatly enhanced collaboration and synergy

**Am (Cm) separation from lanthanides**

- Led by Dr. Bruce Moyer of ORNL
- Participants: ORNL, ANL, INL, SRNL, PNNL, LANL, WSU, other universities
- Began in FY-09

**Off-gas capture and immobilization**

- Led by Dr. Robert Jubin of ORNL
- Participants: ORNL, INL, SNL, PNNL, universities
- Began in FY-10
Several approaches to separation of Am alone, or Am and Cm together, are being investigated.

The goal of the research is to develop a simplified, robust method for minor actinide separation.

However, the program is working toward possible deployment of a future fuel cycle around mid-century, so the emphasis of the program, *until now*, has been on scientific understanding and discovery of approaches.

We are just beginning to transition into development of conceptual separation flowsheets.

- Fewer separation processes
- More robust separation processes
- Simplified/improved material accountability
- Fewer waste form processes
- Fewer waste form handling/storage facilities
- Fewer waste forms to qualify
Why separate actinides?
- Pu and Am dominate repository heat load in 300-3000 year time frame
  - *Heat load will be a primary driver for any repository design*
  - *Recovery/transmutation of actinides provides additional energy and keeps long-term heat generating isotopes out of the repository*
- Minimize volume/mass of material going to a geologic repository
  - *Could preclude the need for more than one repository (but we will always need at least one repository)*

Major accomplishments
- Demonstrated extraction of Am(VI) from curium and lanthanides
  - *Major issue is the oxidation of Americium*
- Developed single process to combine former TRUEX and TALSPEAK processes
  - *More robust operating conditions (less dependence on pH)*
  - *More predictable behavior than TALSPEAK process*
U.S. regulations require management of off-gases to a degree not industrially demonstrated (³H, ¹²⁹I, ⁸⁵Kr, and potentially ¹⁴C)

HTO pretreatment can remove ³H before fuel dissolution and thereby capture in a low volume concentrated stream (issues of co-absorption of iodine being addressed)

Unprecedented levels of iodine removal and immobilization technologies being developed and demonstrated

- Iodine capture also being considered for reactor accident scenarios (capture media on containment vessels)
- Iodine waste form performance directly influences repository dose estimates under most scenarios

Cryogenic distillation of Kr is expensive and challenging, near room-temp separations methods being developed
Iodine management options:

1. Capture on Ag-Mordenite(Z), encapsulate in glass to form glass composite material
2. Capture on Ag-Aerogel, sinter to fused silica matrix with nano-AgI
3. Capture on Chalcogel, sinter to chalcogenide glass
4. Separate from capture media, immobilize in apatite or SiC

Demonstrated immobilization of AgZ in GCM and began waste form performance studies

Developed and demonstrated effectiveness of Ag-Aerogel for iodine capture with high DF (10 000+) and capacity (48 wt%)

Demonstrated the fabrication of fused silica waste form from I loaded Ag-Aerogel with minimal I loss

Performed initial test of I solidification in SiC

Began development of apatite for iodine immobilization (NEUP)
Metallic fast reactor fuel is effectively recycled using Echem process in molten chloride salt.

Echem is being used to treat a fraction of DOE Na-bond fuel at INL but without TRU recycle and using inefficient processes.

Further development in waste management, U/TRU codeposition, equipment design, materials accountancy, Am/Ln separation.

Recent development of a solid cathode process for more effective U/TRU deposition.
Waste Forms Research

- waste characteristics
- separations
- waste characteristics
- fuel fabrication
- disposal environments
- used fuel disposition
- fuel burnup and cooling systems analysis

UFD
Separations
NEUP

- advanced waste forms
- waste form property and performance data
- waste processes
- waste loading, masses, and volumes

DOE-EM
DOE-SC
France,
UK, Japan,
Other Nations

DOE
- EM
- SC

U.S. DEPARTMENT OF
ENERGY
Nuclear Energy
Waste form/process development is aimed at making a U.S. fuel cycle more effective by the development of the next generation of waste management technologies with focus on:

- Technologies to enable the reuse of UNF components that would otherwise require waste treatment, storage, transportation, and disposal (e.g., cladding, noble metals, noble gases).
- Waste forms for dose impacting radionuclides that have orders of magnitude improvements in durability (compared to HLW glass) to reduce the reliance on engineered and natural barrier systems and thereby open new disposal options and lower cost disposal.
- Waste forms and processes that will facilitate lower cost management to include less expensive and complex processing, lower storage and disposal costs, and more flexible to wider ranges of disposal environments and fuel cycles.
Waste Forms Recent Accomplishments

Developed and demonstrated silicate-based glass ceramics with high loading and improved properties.

Developed Synroc formulation capable of forming desired phases by melt-quench process (bound alkali in Hollandite, not previously demonstrated).

Demonstrated Zr purification from irradiated hulls.

Fuel components (U, Pu, MAs, FPs) DFs of $10^3$ to $10^6$ were obtained.

### Decladding Hot Tests

<table>
<thead>
<tr>
<th>Reaction</th>
<th>Temperature (°C)</th>
<th>Time (hours)</th>
<th>Unconverted clad</th>
<th>Converted Zr</th>
<th>%/hour</th>
<th>Beq/g Zr in Cladding</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>356-371</td>
<td>3.50 hours</td>
<td>10.2 g</td>
<td>5.5 g (33%)</td>
<td>10%/hour</td>
<td>~12 (~100%)</td>
</tr>
<tr>
<td></td>
<td>400</td>
<td>10.5 hours</td>
<td>~0</td>
<td>~12 (~100%)</td>
<td>9.5%/hour</td>
<td>~12 (~100%)</td>
</tr>
</tbody>
</table>

**Gamma Spectroscopy**

<table>
<thead>
<tr>
<th>Zr-Cl Product</th>
<th>356-371 °C</th>
<th>400 °C</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{241}$Am</td>
<td>$1.8\times10^8$</td>
<td>$1.9\times10^3$</td>
</tr>
<tr>
<td>$^{243}$Am</td>
<td>$3.8\times10^6$</td>
<td>&lt;2.2 \times10^1</td>
</tr>
<tr>
<td>$^{137}$Cs</td>
<td>$2.7\times10^9$</td>
<td>3.7 \times10^2</td>
</tr>
<tr>
<td>$^{154}$Eu</td>
<td>&lt;4E+05</td>
<td>&lt;2.9 \times10^1</td>
</tr>
<tr>
<td>$^{155}$Eu</td>
<td>&lt;3.1+01</td>
<td>6.3 \times10^1</td>
</tr>
<tr>
<td>$^{125}$Sb</td>
<td>1.2E+07</td>
<td>&lt;2.9E+01</td>
</tr>
<tr>
<td>$^{60}$Co</td>
<td>&lt;4E+05</td>
<td>&lt;5.5E+01</td>
</tr>
<tr>
<td>$^{154}$Eu</td>
<td>&lt;3.1+01</td>
<td>6.3E+01</td>
</tr>
</tbody>
</table>

*Argon purge every 2 hours for ~30-45 min.*
Objectives are to develop sufficient understanding of waste form performance in disposal environment to provide defensible source-term model for PA while minimizing conservatism:

- Initial focus is glass and Fe-based alloy corrosion later to be expanded to other waste forms for immobilization of dose contributing radionuclides (UO$_2$ fuel performance is studied under UFD).
- Glass corrosion studies seek to develop and international consensus rate law that accounts for impacts of glass composition and environmental parameters.
- Alloy corrosion studies seek to develop an initial rate law suitable for incorporation into a PA.
Glass Corrosion Recent Accomplishments

- Organized international program to develop consensus glass corrosion rate law
- Applied advanced characterization techniques to identify the elusive transport barrier associated with residual rate reduction and quantified the relative impacts to transport control and surface reaction control under certain controlled conditions
- Successfully modeled experimental results from a 26-year corrosion experiment using micro-continuum reactive transport model
- Developed theory for coupled kinetics of secondary phase precipitation and glass dissolution to account for accelerated corrosion

Nano-SIMS of cross-section of corroded SON68 glass showing (left) surface enrichment of $^{28}\text{Si}$, (center) total depletion of B at surface, and (right) inner layer theorized to provide transport barrier

Atom probe tomography of reacting glass surface after isotope exchange test
Approximately 20% of the Fuel Cycle Technologies budget is awarded to universities to perform research in support of the program via NEUP projects (with an additional ~5% direct university support).

It is essential to leverage this funding to make technical progress in the campaign.

It is also very important to educate the next generation of nuclear science and technology researchers.

The NEUP program is highly integrated with the MRWFD campaign.

FCR&D review of all MRWRD NEUP projects in FY-13 to assess efficacy of projects.
Ongoing NEUP Collaborations (FY-09 through FY-12 project starts)

- Reference Technologies and Alt Fund. Science/ Mod. Simulation
  - WSU: Act. partitioning
  - NWU: Sulfides for I capture
  - UCI: Micro-emulsions

- Minor Actinide Sigma Team
  - PSU: Glass Thermokinetic
  - UW: Salt Thermokinetic
  - UNC: Novel U Sorbents

- Advanced Waste Forms and Processes
  - AU: Corrosion surfaces
  - AU: Ceramic waste forms
  - UCB: U Ligand Design
  - UNLV: Metal corrosion

- Electrochemical Processing
  - UCLB: Ligand Radiolysis
  - SU: Sorption Models
  - UNLV: Metal corrosion

- Uranium Extraction from Seawater
  - UA: Chitin Absorbate
  - CUNY: Polymer Extractants

- UMC: Kr, I, C sequestration
  - UNLV: Kr/Xe abs model
  - UNLV: Tc ceramics

- NWU: Sulfides for I capture
  - UMich: Apatite Waste Forms
  - UNLV: Metal corrosion

- PSU: Glass Thermokinetic
  - UW: Salt Thermokinetic
  - UNLV: Tc ceramics

- UW: Salt Thermokinetic
  - UNLV: Metal corrosion
  - UNC: Novel U Sorbents

- UNLV: Metal corrosion
  - UNLV: Tc ceramics
  - UNC: Novel U Sorbents

- UNLV: Tc ceramics
  - UMich: I-129 waste form
  - UCLB: Ligand Radiolysis

- UCLB: Ligand Radiolysis
  - SU: Sorption Models
  - UNLV: Metal corrosion

- SU: Sorption Models
  - UNLV: Metal corrosion
  - UNLV: Tc ceramics
FY-13 New Start NEUP Collaborations

Reference Technologies and Alt

Minor Actinide Sigma Team

Off-gas Sigma Team

Fund. Science/ Mod. Simulation

Advanced Waste Forms and Processes

Electrochemical Processing

Uranium Extraction from Seawater

OSU: ALSEP Speciation

UI: Nano-structured Sorbents

PSU: Glass corrosion

UMary: Extraction Syst.

WHOI: Real Ocean Cond.

UTA: Economic Analysis

TAMU: Ion Exchangers

UC: Novel Porous Sorbents.

WSU: Zr Chem. in MA Partitioning

UC Davis: Nanoporous Materials

UNT: modeling of glass corrosion

UIC: Alloy corrosion

UC: Ionic Liquids

CUNY: Aminophophinates
Key International Collaborations

**France: (CEA)**
- Off-gas capture and immobilization
- On-line process monitoring
- Radiation chemistry
- Fundamental understanding of minor actinide separation chemistry
- Glass corrosion rate-law

**Japan:**
- Uranium extraction from seawater
- Minor actinide separations
- Off-gas capture

**Russian Federation:**
- Tritium removal
- On-line process monitoring
- Glass corrosion
- Process modeling

**China:**
- Uranium extraction from seawater
- Pyroprocessing
- Off-gas capture

**European Union:**
- Participation in EU SACSESS program (Safety of Actinide Separations)

**Others:**
- Active participation in OECD/NEA, IAEA workshops and studies
- Initial meeting with Czech Republic
- Possible future UK collaborations
The **Materials Recovery and Waste Forms Development Campaign** is performing research to support a range of nuclear fuel cycle options by:

- Developing technologies to improve fuel current fuel cycle performance and enable future fuel cycles
- Improving the fundamental understanding of processes important to materials recovery, separations, waste form development, and waste form performance
- Developing and maintaining expertise needed to solve current and future challenges in nuclear materials management

The **Campaign includes leaders in their respective fields from eight national laboratories (ANL, INL, LANL, LBNL, ORNL, PNNL, SNL, and SRNL) and 30 universities**

**Active collaborations are ongoing with a number of other federal and international organizations**

**Significant technical achievements have kept the campaign at the forefront of technology development in a number research areas**