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Materials Recovery and Waste Form Development Campaign

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U.S. Department of Energy

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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**

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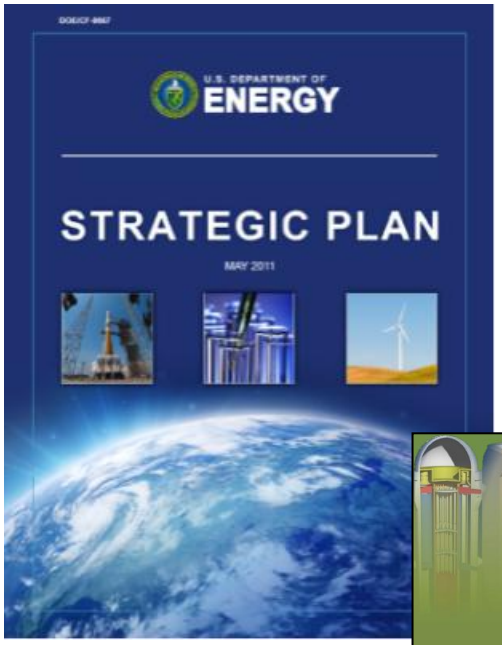
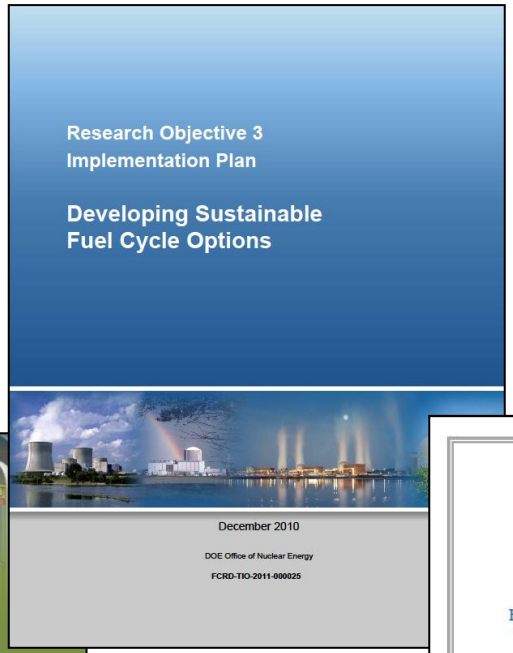
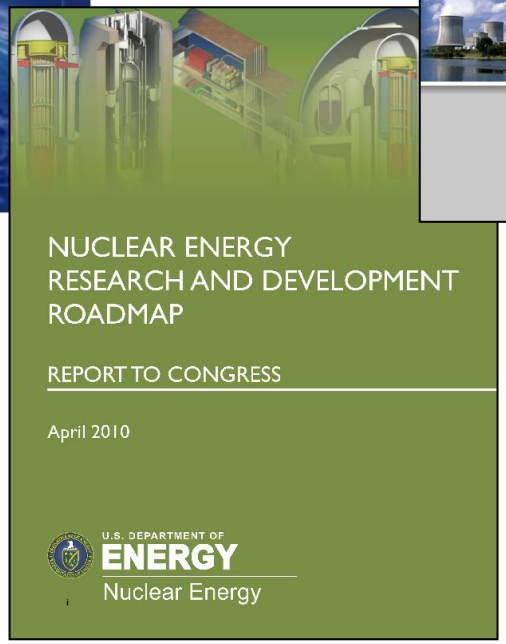
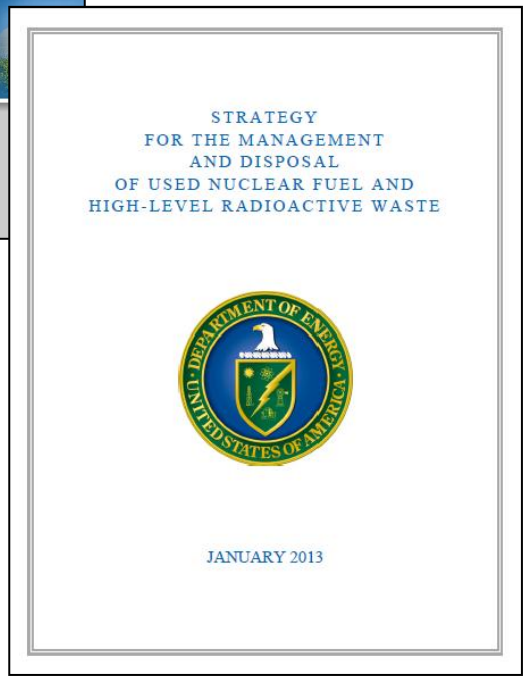
Program Drivers









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FCRD Mission and Program Objectives

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.
DOE

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.
NE

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

Program Objectives

Near Term

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

Medium Term

- **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.**

Long Term

- **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.**

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Office of Fuel Cycle Technologies: an Integrated Approach

Front End

Uranium Resources

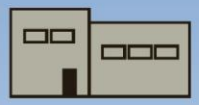
- Conventional production
- Innovative approaches
  - U Seawater

Fuel Fabrication

- Safety enhanced LWR fuel
  - Accident tolerance
- Higher performance
  - Improved burnup

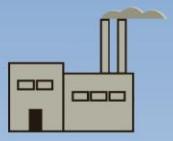
Reactors

Back End



Interim Storage

- Evaluating extended time frames
- Transport after storage



Recycle

- Separations
- Recycled fuel
- Secondary waste treatment

Disposal

- Alternative geologies
- Alternative waste forms

←----------Safeguards and Security By Design----------→

Optimize through systems analysis, engineering, and Integration

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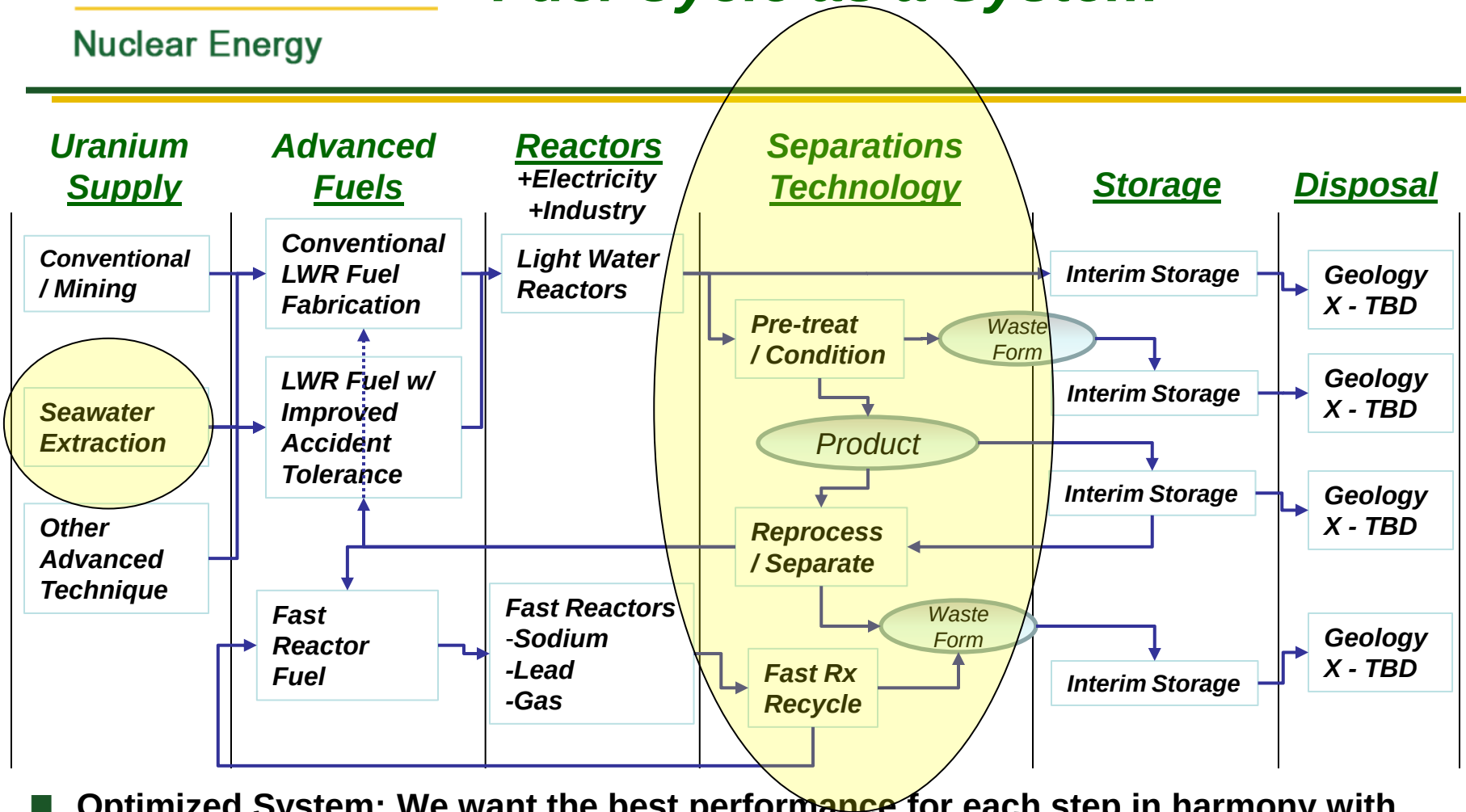
Fuel Cycle as a System



- 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

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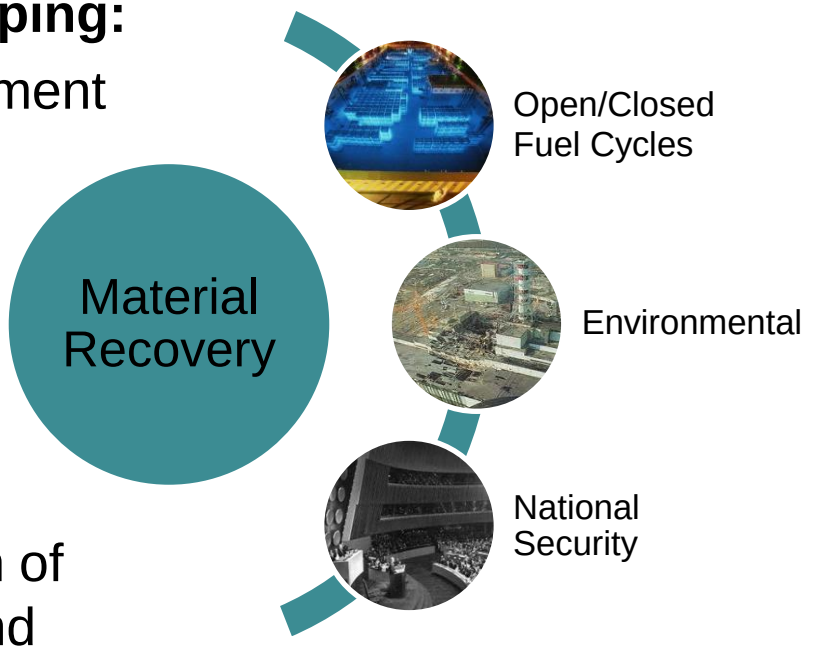
Campaign Objectives

- **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



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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***

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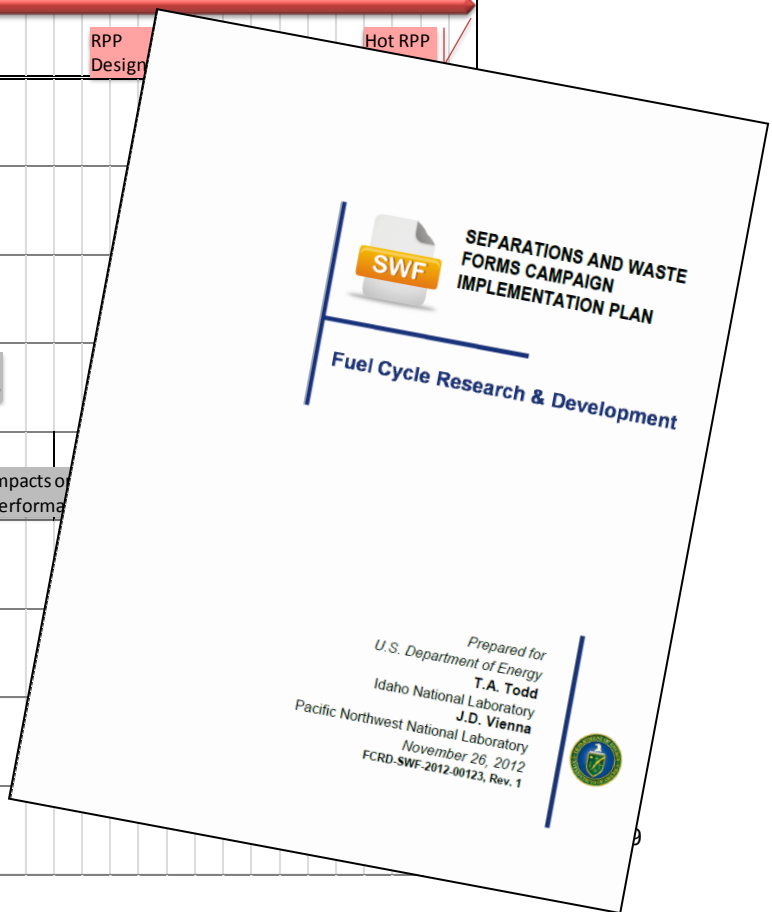
Campaign Implementation Plan

Fiscal Year	12	13	14	15	16	17	18	19	20	21	22	23	24	25	26	27	28	29	30	31	32	33	34	35	36	37	38	39	40	41	42	43	44	45	46	47	48	49	50	51	52	53	54	55	56	57	58	59	60
Case study technologies			Preliminary FS Case study						Integrated Lab-Scale Test Setup								Complete Integrated Lab-Scale Testing																																		
LWR fuel reprocessing plant									ES Simulant Testing						Hot ES Startup																											Hot RPP Startup									
Fast reactor fuel reprocessing plant															Lab-scale UNF tests										Hot ES Startup										RPP Design								Hot RPP				
Glass ceramics			Proof of Principle Testing		Reference Formulation				Conceptual Design		Preliminary Design				Pilot Testing																																		
Develop high durability Tc waste form			Select Waste Form		Performance Model				Process Development						Impacts on Performance				Pilot Testing		Waste Form Compliance																												
Epsilon metal for Tc and noble metals			Evaluate Processes		Optimize Process				Performance Model			Pilot Testing			Impacts on Performance						Waste Form Compliance																												
Fast reactor electrochemical salt waste forms			Salt Waste Managment Approach				Select Waste Form										Develop Salt Process				Performance Model		Pilot Testing			Impacts on Performance																						
Fast reactor metal waste forms							Characterize Waste					Reference Formulation									Alloy Process			Performance Model				Pilot Testing																					
Zr recycle			Select Process	Initial Test			Optimize Process					Process Design			Pilot Testing		Process Model																															
Fuel fabrication transuranic waste forms							Waste Estimates					Select Waste Form			Select Waste Process		Performance Model				Pilot Testing			Impacts on Performance				Waste Form Compliance																				
Electrochemical separations			Demonstrate U/TRU Seps w/ Simulants				Demonstrate U/ TRU Seps w/ UNF								Develop Reference Echem Process Ops						Develop High Value Process Alternatives																														
Glass performance			Characterize Ancient Analog		Consensus Rate Law in DIW				General Consensus Rate Law																																							



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Thrust Area – Fuel Cycle Overlay

Near-term & Open Fuel Cycle

Long-term & Closed Fuel Cycle

Capability Development/Maintenance

- ***Uranium from seawater***
- ***Tritium Separations / Treatment***
- ***Off-gas Sigma Team***
- ***Waste Form Performance***
- ***Waste Form and Process Development***
- ***Aqueous Recycle Technology***
- ***Electrochemical Recycle Technology***

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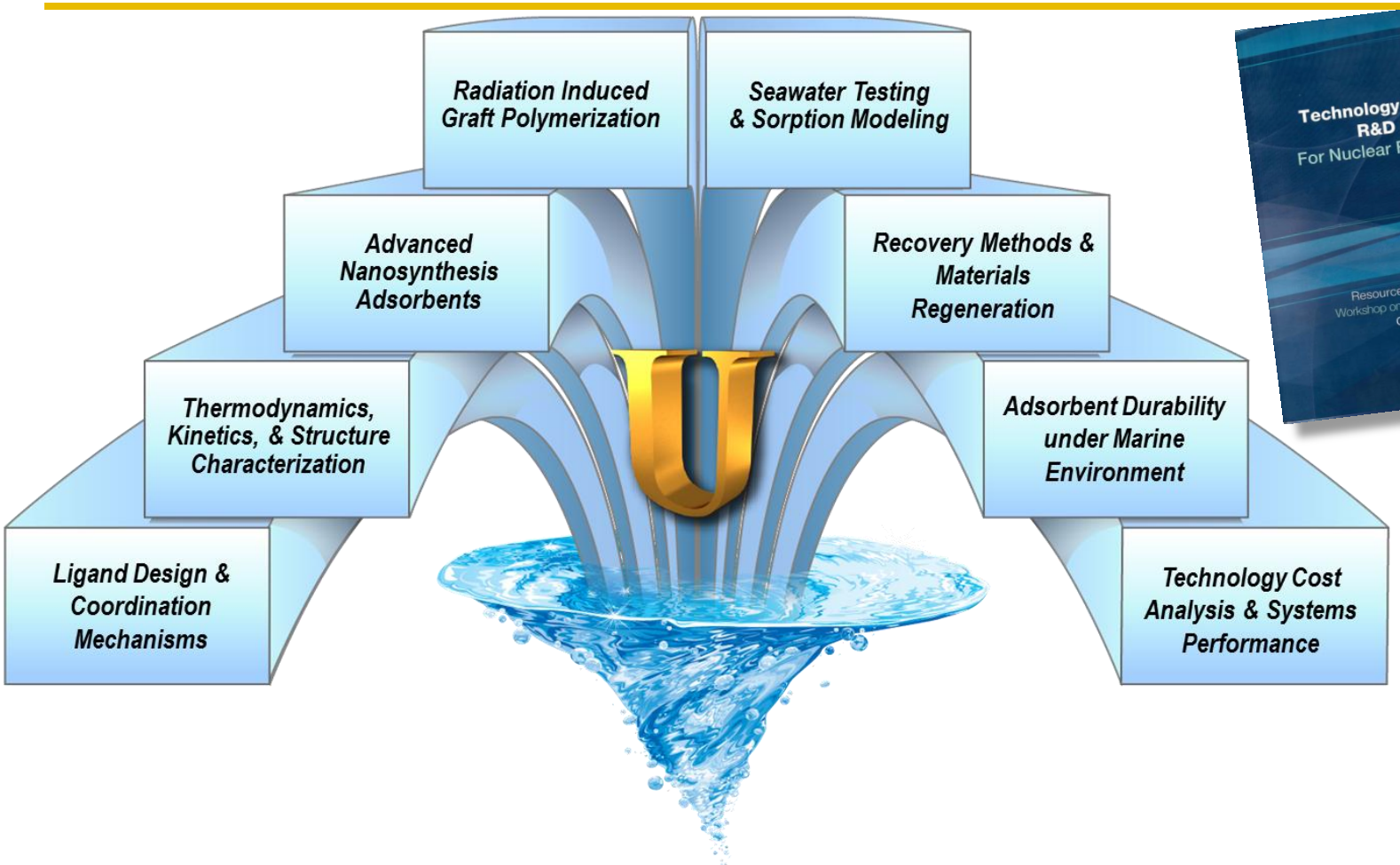
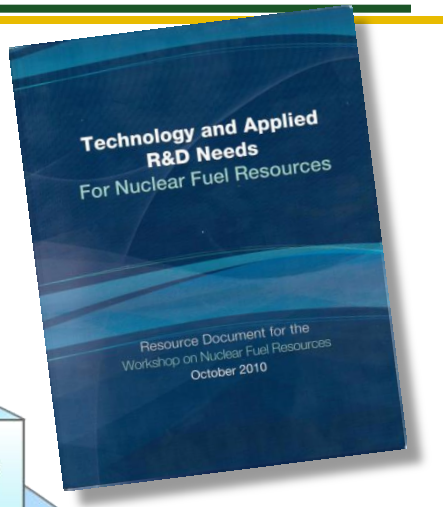
Uranium from Seawater





"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."

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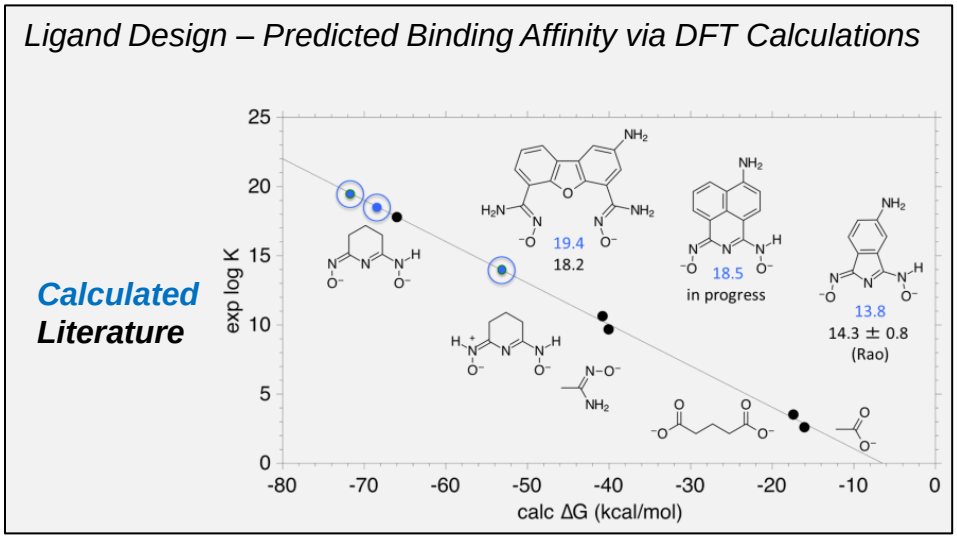
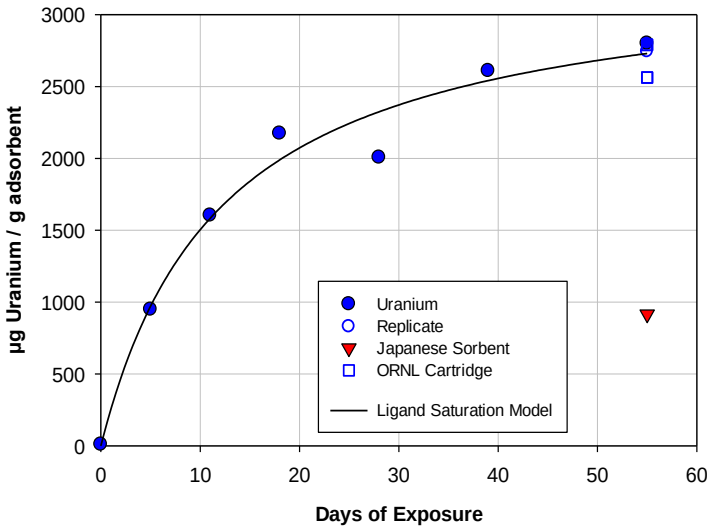
Uranium from Seawater Accomplishments

- 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





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Tritium from High Volume Water

- **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

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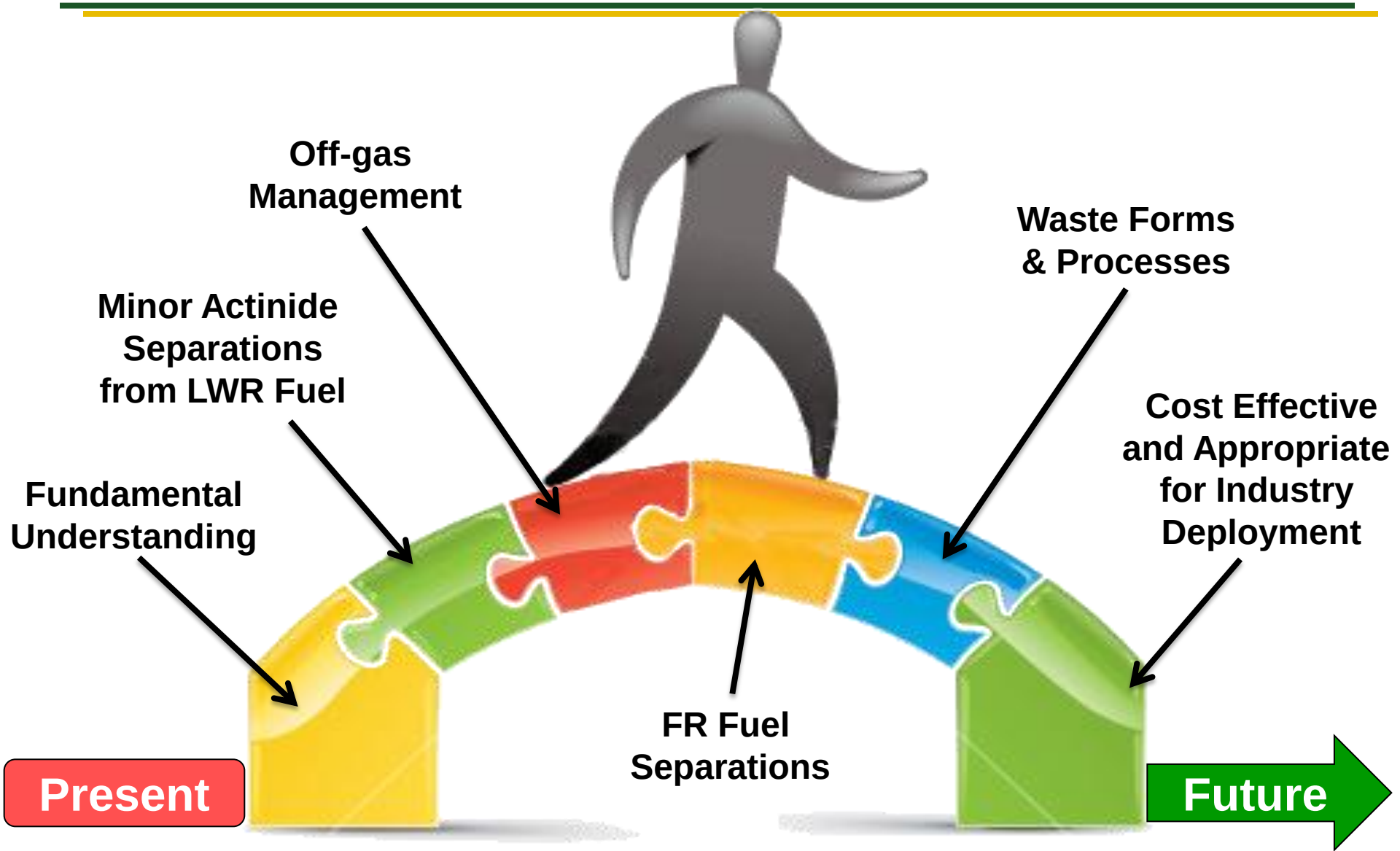
Closed Fuel Cycle Technology Gaps



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Sigma Teams

- **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

Los Alamos NATIONAL LABORATORY EST. 1943
OAK RIDGE National Laboratory
Pacific Northwest NATIONAL LABORATORY
INL Idaho National Laboratory
Actinide Science at WSU
Argonne NATIONAL LABORATORY
SRNL
Sandia National Laboratories

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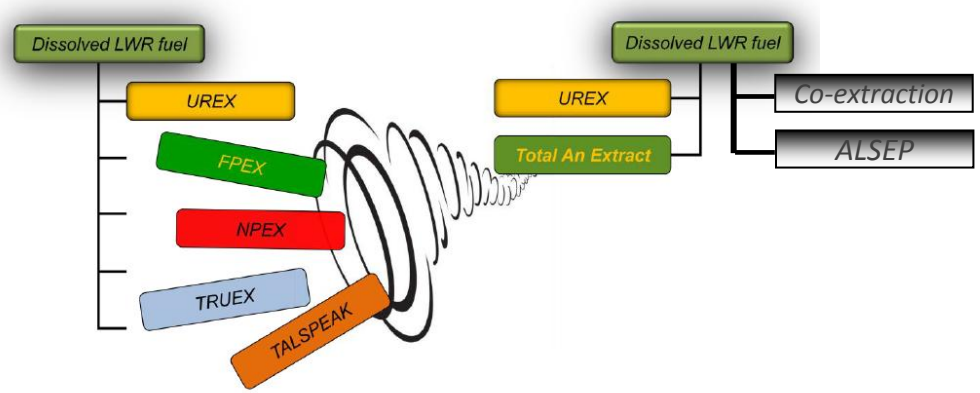
Sigma Team for Minor Actinide Separations

- 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

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Sigma Team for Minor Actinide Separations

- **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*

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Off-Gas Sigma Team

- **U.S. regulations require management of off-gases to a degree not industrially demonstrated (^{3}H, ^{129}I, ^{85}Kr, and potentially ^{14}C)**
- **HTO pretreatment can remove ^{3}H 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**

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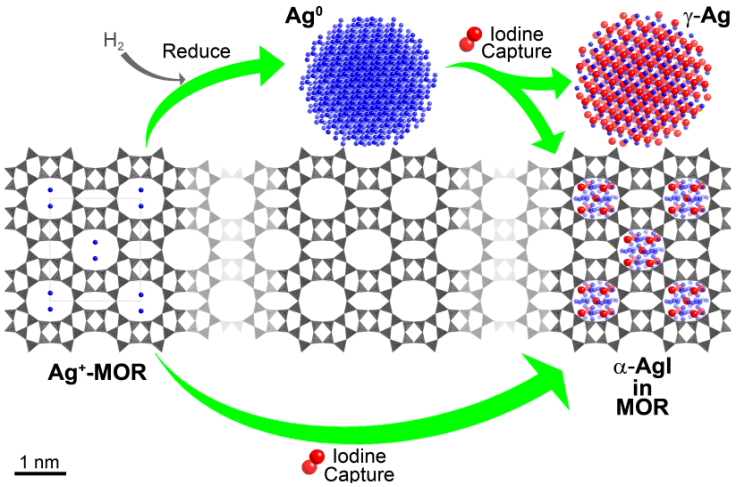
Off-gas Sigma Team: Iodine Management

- **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)**

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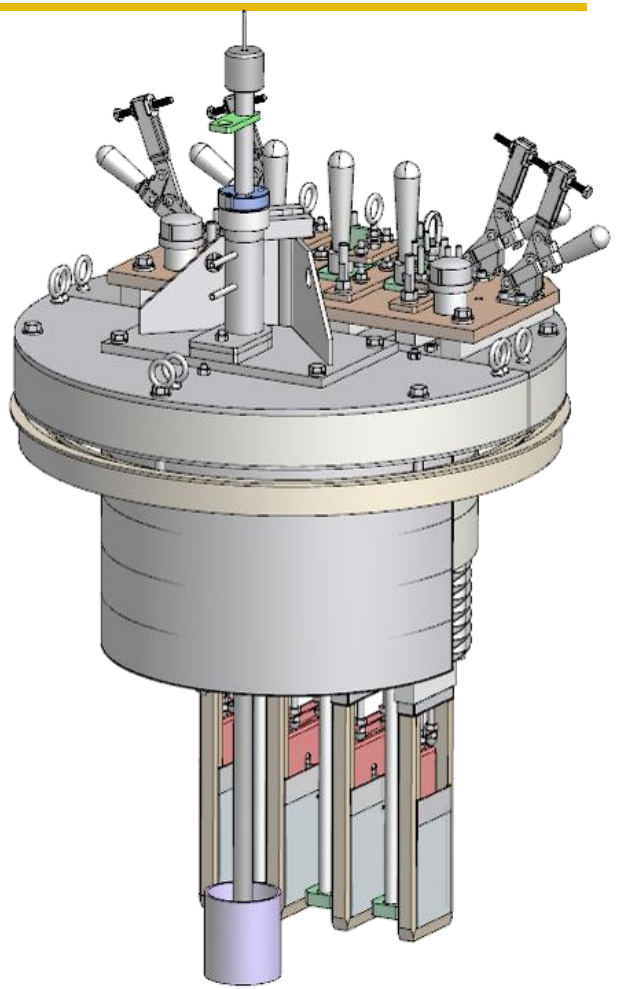
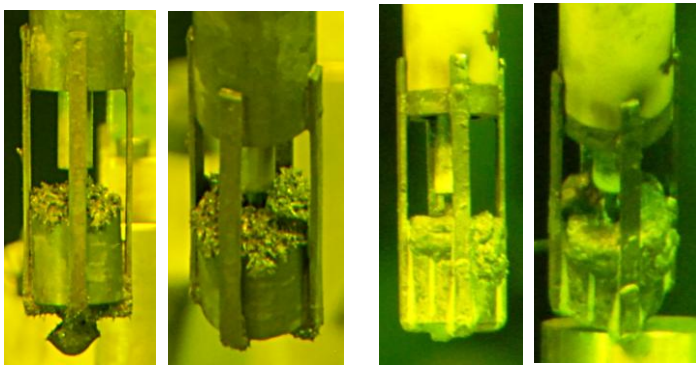
Electrochemical Recycle of FR Fuel (Domestic Program)

- **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**





U deposition

U – Pu co-deposition

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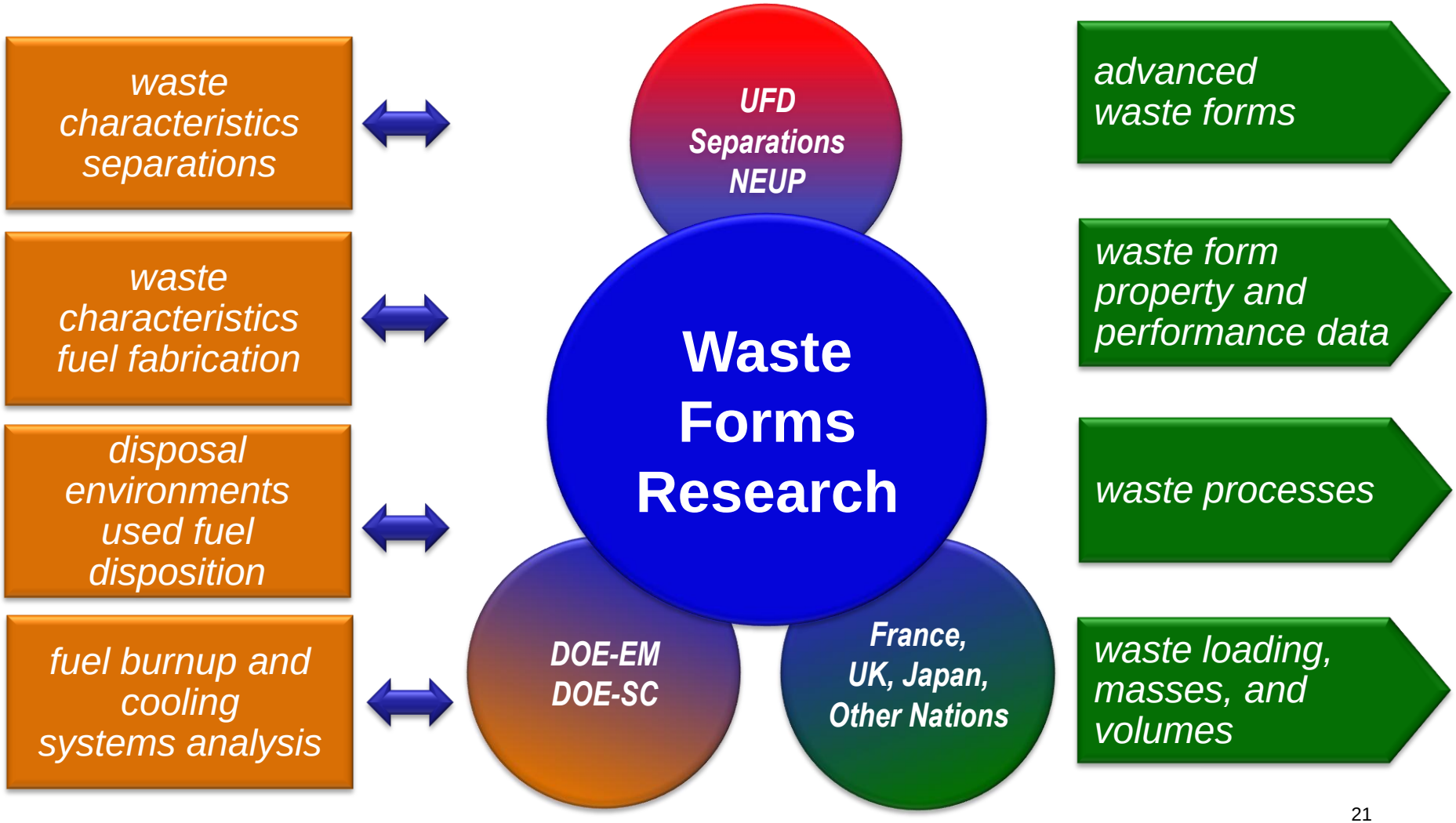
Waste Forms Research



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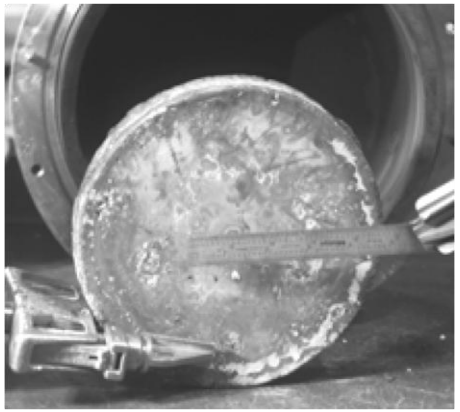
Waste Forms and Processes

- **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.



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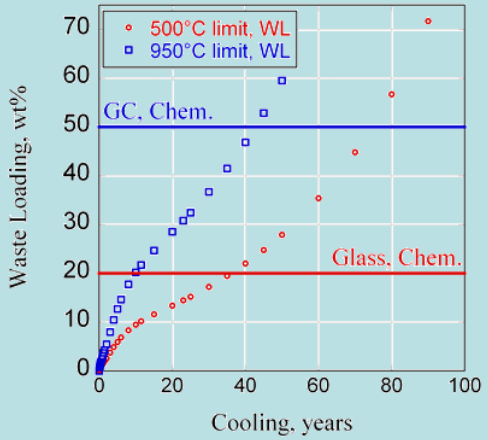
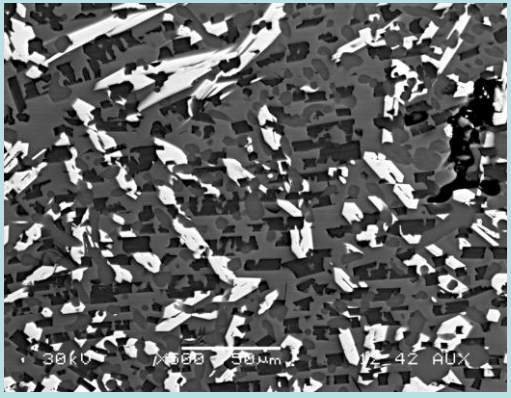
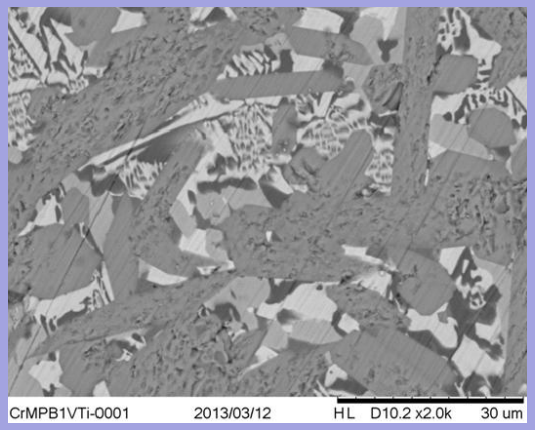
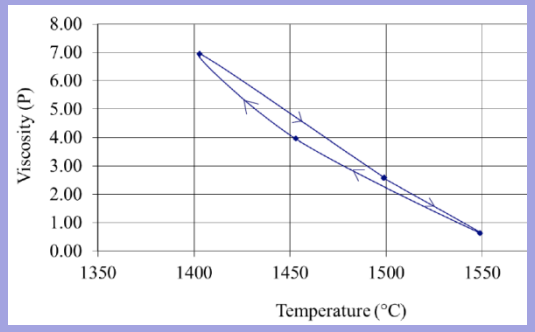
Waste Forms Recent Accomplishments

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

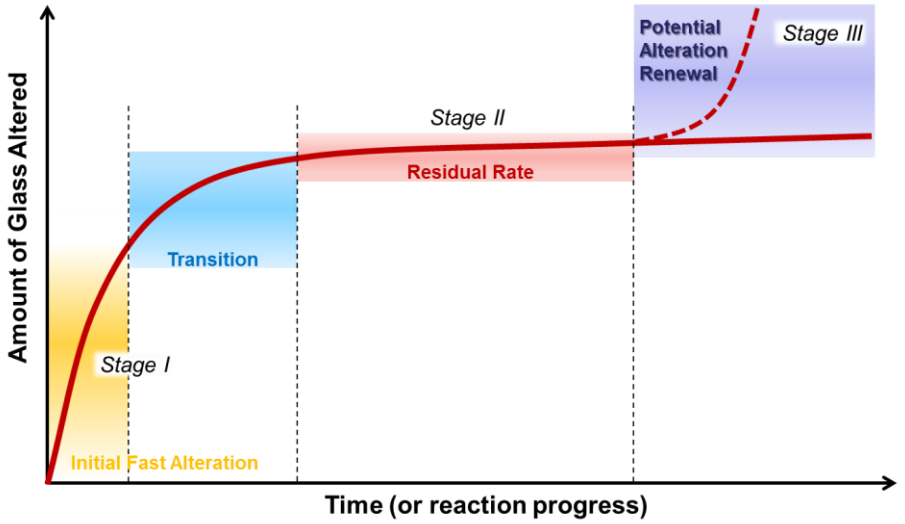
Fuel components (U, Pu, MAs, FPs) DFs of 10^3 to 10^6 were obtained

Reaction:				
Temperature		356-371°C	400°C	
Time		3.50 hours	10.5 hours[1]	
Unconverted clad		10.2 g	~0	
Converted Zr		5.5 g (35%)	~12 (~100%)	
%/hour		10%/hour	9.5%/hour	
		Bq/g Zr in Cladding		
Gamma Spectroscopy	Fuel and Clad	$ZrCl_4$ Product	$ZrCl_4$ Product	Final DF
^{241}Am	1.8E+08	1.9E+03	9.4E+04	1.9E+03
^{243}Am	3.8E+06	<2.2E+01	<5.1E+01	>7.5E+04
^{137}Cs	2.7E+09	3.7E+02	3.6E+03	7.5E+05
^{125}Sb			4.7E+02	
^{60}Co	< 4E+05	1.9E+01	<5.5E+01	
^{154}Eu	1.2E+07	<2.9E+01	<7.4E+01	1.7E+05
^{155}Eu		<3.1+01	6.3E+01	

[1]Argon purge every 2 hours for ~ 30-45 min.

Waste Form Performance

- **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



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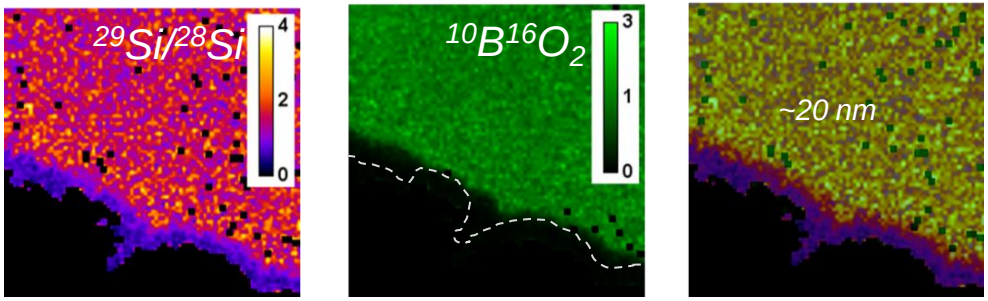
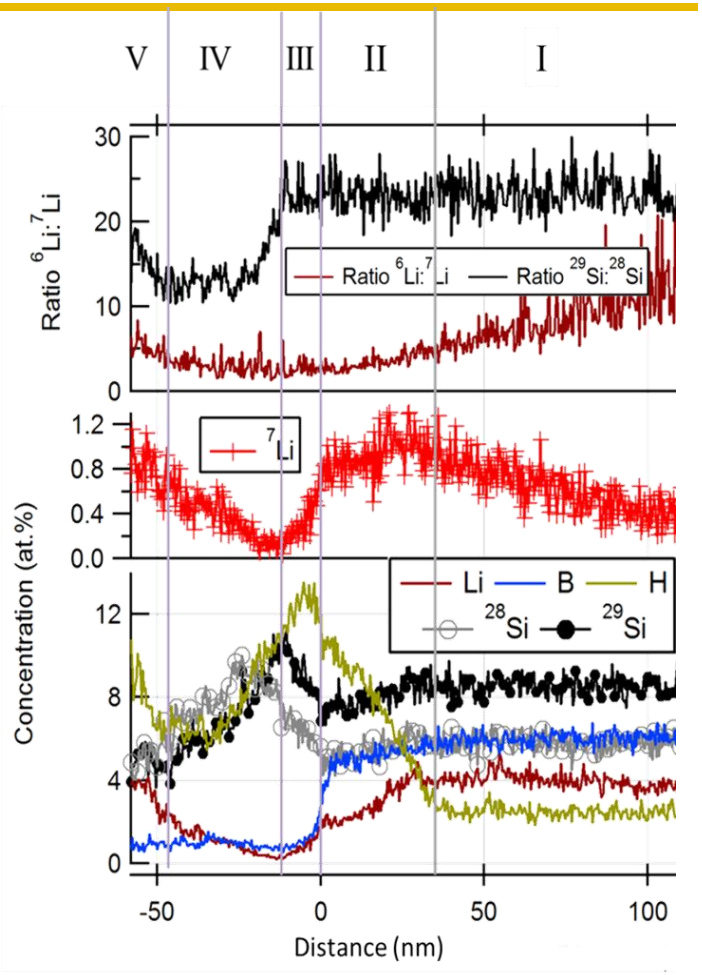
Glass Corrosion Recent Accomplishments

- 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}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

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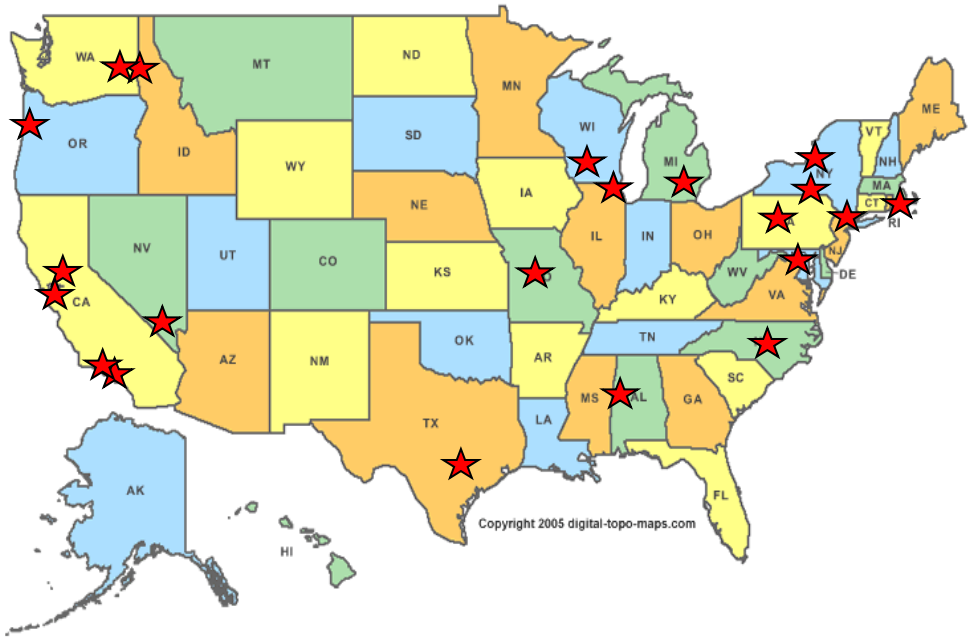
Campaign University Support

- 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



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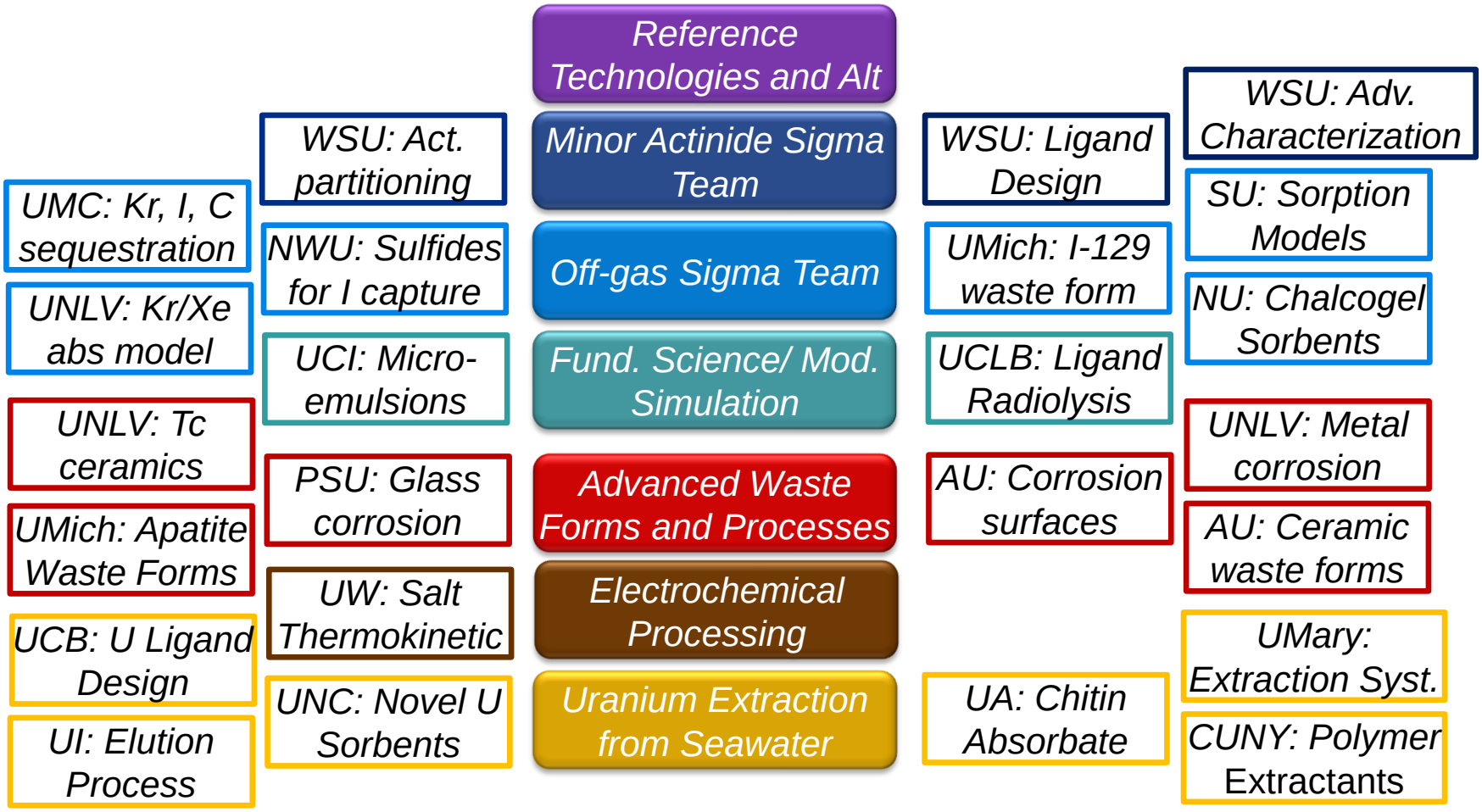
Ongoing NEUP Collaborations (FY-09 through FY-12 project starts)



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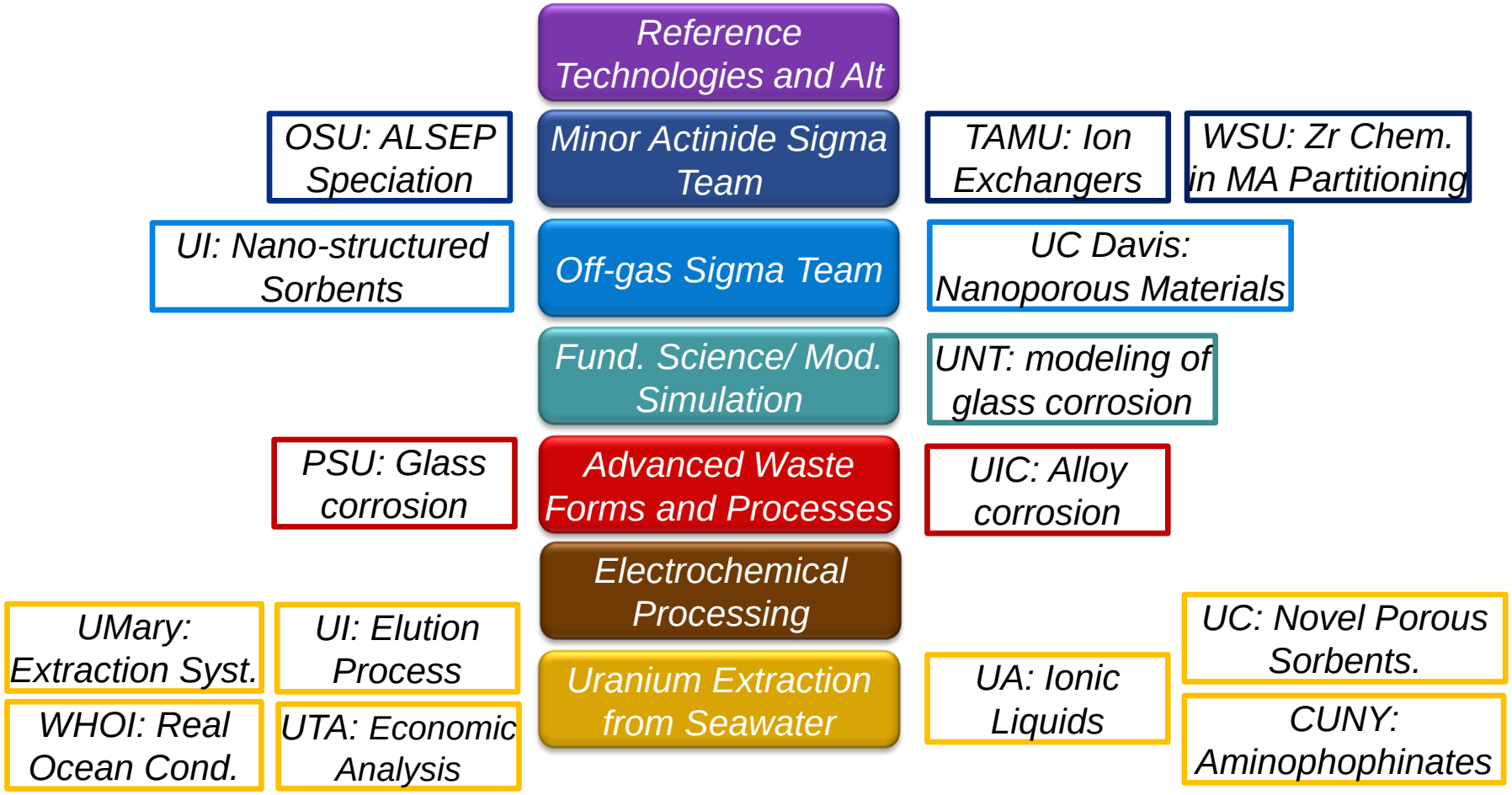
FY-13 New Start NEUP Collaborations



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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*

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Summary and Conclusions

- **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**