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United States Nuclear Regulatory Commission
Protecting People and the Environment

Performance Evaluation and Potential Uncertainties in Spent Fuel Integrity and Source Term

T. Ahn [1], H. Jung [2], and J. Guttmann [1]

[1] Office of Nuclear Material Safety and Safeguards
U.S. Nuclear Regulatory Commission (NRC)
Washington, DC, USA
[2] Center for Nuclear Waste Regulatory Analyses (CNWRA)
Southwest Research Institute, San Antonio, Texas, USA

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Disclaimer:

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Outline

- Purpose
- Performance Evaluation for Various Waste Management Strategies
- Potential Uncertainties Associated with Performance Evaluation in Extended Storage
- Simulated Spent Fuel (SIMFUEL) Corrosion Test
- References

Purpose

- Review performance evaluation for various waste management strategies
- Present potentially important uncertainties with spent fuel (SF) integrity in on-site operation, extended storage, transportation, and long-term waste disposal. An emphasis is on extended storage
- Present corrosion test results of Simulated Spent Fuel (SIMFUEL)

Performance Evaluation for Various Waste Management Strategies

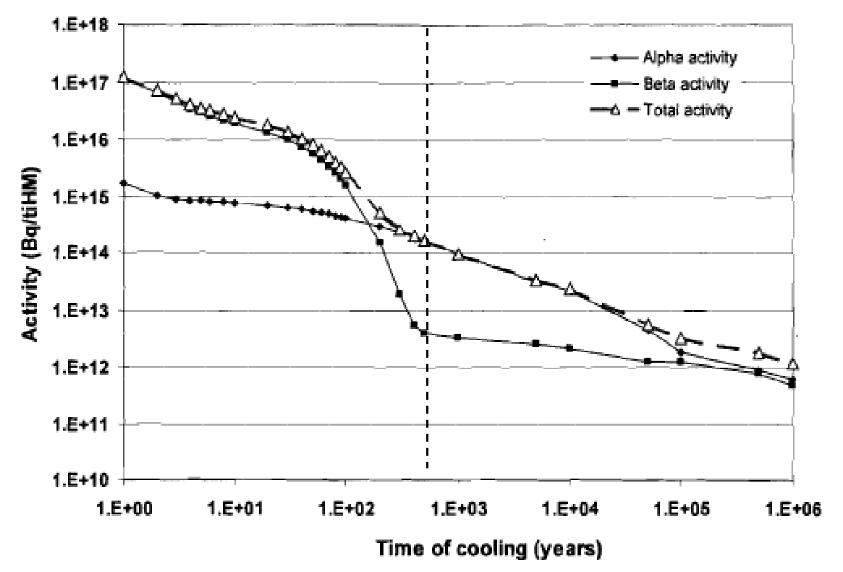
Options for Evaluating Performance

- Deterministic modeling based on short-term experimental results in assessing Spent Fuel conditions:
  - Regulatory and technical gap analysis
  - Mitigation process
  - Iterated with risk assessment
- Probabilistic system approach in assessing risk associated with Spent Fuel integrity:
  - Identification of events and failure mechanisms
  - Probability and probabilistic cut-off of failure mechanisms
  - Consequence analysis by system performance evaluation, including uncertainty and sensitivity analyses
  - Examples of system performance evaluation codes for on-site operation and extended storage, including SAPHIRE (Idaho National Engineering Laboratory, 1998), MELCOR (Gauntt et al., 2000), MACCS2 (NRC, 1998), RSAC (Wenzel, 1994), and PCSA (Dasgupta et al., 2002)
  - Risk assessment at one time and time-dependent system performance assessment

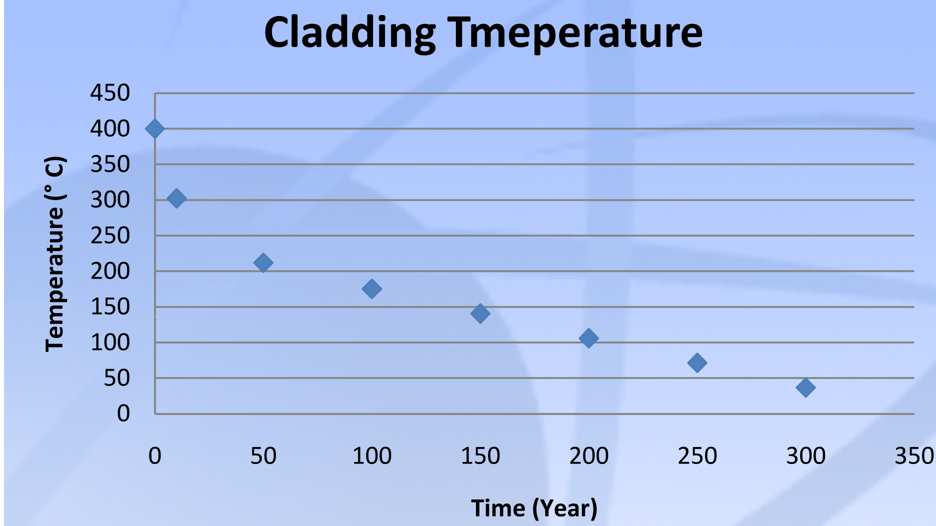
Parameters & Components Related to Radionuclide Source Terms

- **Radionuclide Inventory** (figure on next slide)
- **Heat Generation:** mechanisms, aging, burnup
- **Gas generation** (figure on next slide)
- **SF UO_2 matrix:** powdering by oxidation, powdering by drop/collision/impact, fire, high-burnup rim effects, radionuclide migration and diffusion, dissolution
- **Cladding:** embrittlement, mechanical failure, corrosion
- **Canister:** mechanical failure by drop/collision/impact, pitting corrosion, stress corrosion cracking, aging (phase transformation and segregation)
- **Neutron absorber:** degradation
- **Other structure:** canister (or cask) internal components, concrete materials, building structure (HEPA filter)

Parameters & Components Related to Radionuclide Source Terms (continued)

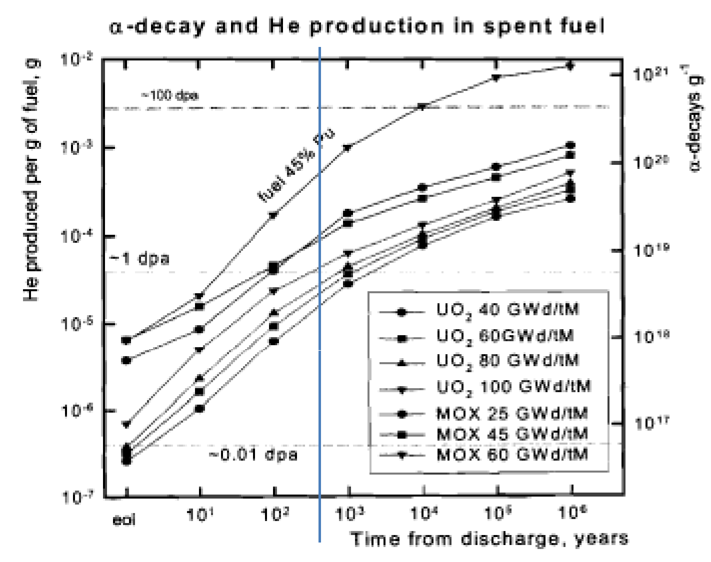


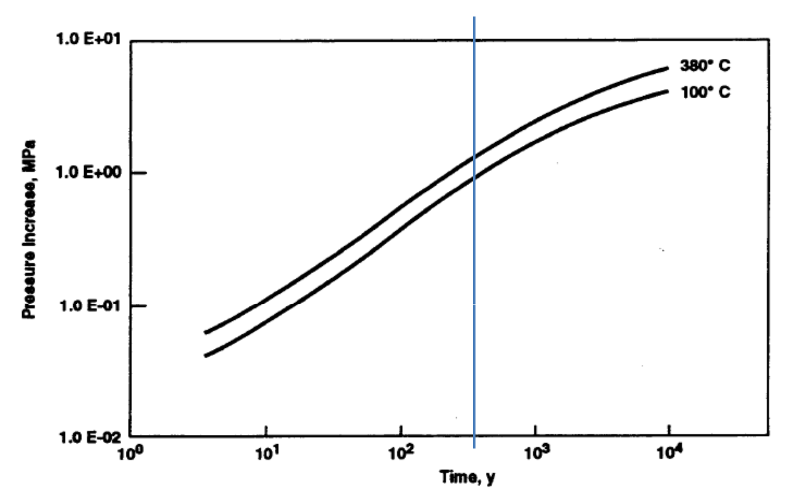
(a) Specific alpha, beta, and total activities in Bq per ton of initial heavy metal (iHM) in UOX fuel of 60 GWd/t versus time (Ferry et al., 2005) [1 Bq = 2.70x10-11 Ci; 1 Mton = 1.1 tons]



(b) Cladding temperature history for high-burnup spent fuel in dry storage casks (100 and 380 ºC = 212 and 716 °F respectively, after EPRI, 2007)

Parameters & Components Related to Radionuclide Source Terms (continued)





(a) Helium production from alpha-damage accumulation during storage of spent fuel at various Burnup (1 g is 0.035 oz, Rondinella et al., 2010)

(b) Calculated pressure increase caused by helium production from actinide decay, Pressurized-water reactor fuel rod with 30 MWd KgU-1 and an assumed 100 percent He release (Johnson and Gilbert, 1983) [1 watt = 3.42 BTU/hr; 1 Mpa = 0.145 ksi; 100 and 380 ºC = 212 and 716 °F respectively]

Event, Sequence, Scenarios

- **Nominal effects: extended storage and geologic disposal**

- **Accidental effects: aircraft impact, fire, wind (hurricane/tornado), operation, transportation, or seismic impacts**

- ***Probability of materials at risk for potential radionuclide release, criticality or shielding (e.g., drop, collision, long-term degradation, and other accidents)***

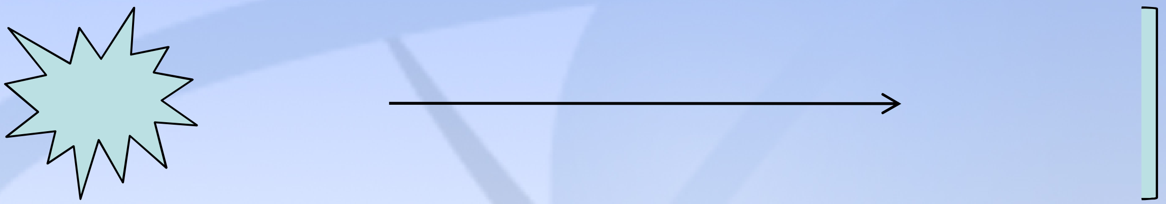
- **Onsite storage Probabilistic Risk Assessment (PRA) for existing reactor facility was completed in 2007 (NRC).**
- **New transportation PRA will be issued in 2011.**

Probability Cut-off and Dose

- **Example Approaches to Evaluate Performance**
  - ***Cut-off probability for exclusion or materials at risk***
  - **Probability weighted dose (e.g., canister drop probability x consequence)**
  - **Dose per one-time effect as consequence from postulated event (e.g., low probability of a severe event)**
  - **Worker dose inside a boundary and public dose outside a boundary**

Release Path

- On-site operation, storage, transportation: operational buildings, casks, controlled boundary, biosphere
- Geologic disposal: waste form, waste package, near-field, far-field, biosphere



Source Term: spent fuel, cladding, other waste forms, canister, building, HEPA, near field

Transport Path: air, groundwater

Boundary: worker dose, public dose

Release Fraction and Release Rate

- **Data Needs for High-Burnup Spent Fuel**
- **Release Fraction at Fixed Time for Operation and Storage:**
  - **Tritium, noble gases, iodine, crud, ruthenium, cesium, strontium, fuel fines**
  - **Release fraction, materials at risk (from initiating event), damage ratio, leak path factors, canister retention factor (e.g., Kamas et al., 2006; Sprung et al., 2000)**
  - **Factors involved: impact energy from collision, seismic or transportation accidents, oxidation from fire or aircraft impact, cladding integrity, and internal structure integrity**

- **Release Rates in Time-Dependent Waste Disposal:**
  - **Fission products and actinides**
  - **Radionuclides bounded in the matrix and gap/grain boundary segregation**

Criticality Control

- Moderator Exclusion – an option only once applied for international transportation
- Configurational Stability of Spent Fuel Assemblies and Internal Structure
- Effectiveness of Neutron Poison
- Burnup Credit
- Low Consequence (mostly at off-site)

Potential Uncertainties Associated with Performance Evaluation in Extended Storage

Uncertainties in Release, and in Criticality and Shielding Control, from Corrosion and Cracking

- **Mechanical Failure of Canister: mechanical puncture upon impact stress, restricted area opening and gas pressure relief from ductile materials**
  - ***Cut-off event probability for exclusion (no breach of confinement) or materials at risk***
  - **Perforation development from mechanical impact failure: V-Notch impact test and weld effects**
  - **Particulate filtration, gas pressure**
  - **Canister drop: review of U.S. Department of Energy standard canister of DOE spent nuclear fuel (Snow et al., 2000), Idaho Spent Fuel Project canister (Snow and Morton, 2003), multi-canister over packs (Snow, 2004), and high-level waste canister (Peterson, 1985)**
  - **Expand NRC storage risk study (NUREG-1864, NRC, 2007) to include high-burnup spent fuel and aged canisters**
- **Long-term Phase Transformation and Corrosion**

Uncertainties in Release, and in Criticality and Shielding Control, from Corrosion and Cracking (continued)

- Stress Corrosion Cracking and Localized Corrosion of Canister: limited area opening and gas pressure relief
  - *Cut-off probability for exclusion (no breach of confinement) or materials at risk:* materials, environments (e.g., chloride concentration, relative humidity and water film thickness, salt content), temperature, and stress
  - Stress corrosion cracking: crack propagation, network of tight cracks (size and density in number), source (impact, residual stress, weld flaw)
  - Pitting and crevice corrosion: initiator for stress corrosion cracking, limited area
  - Coastal and industrial area: susceptibility (Mintz and Dunn, 2009; Tani et al., 2009)
  - Effects of phase transformation

Uncertainties in for Release, and in Criticality and Shielding Control, from Corrosion and Cracking (continued)

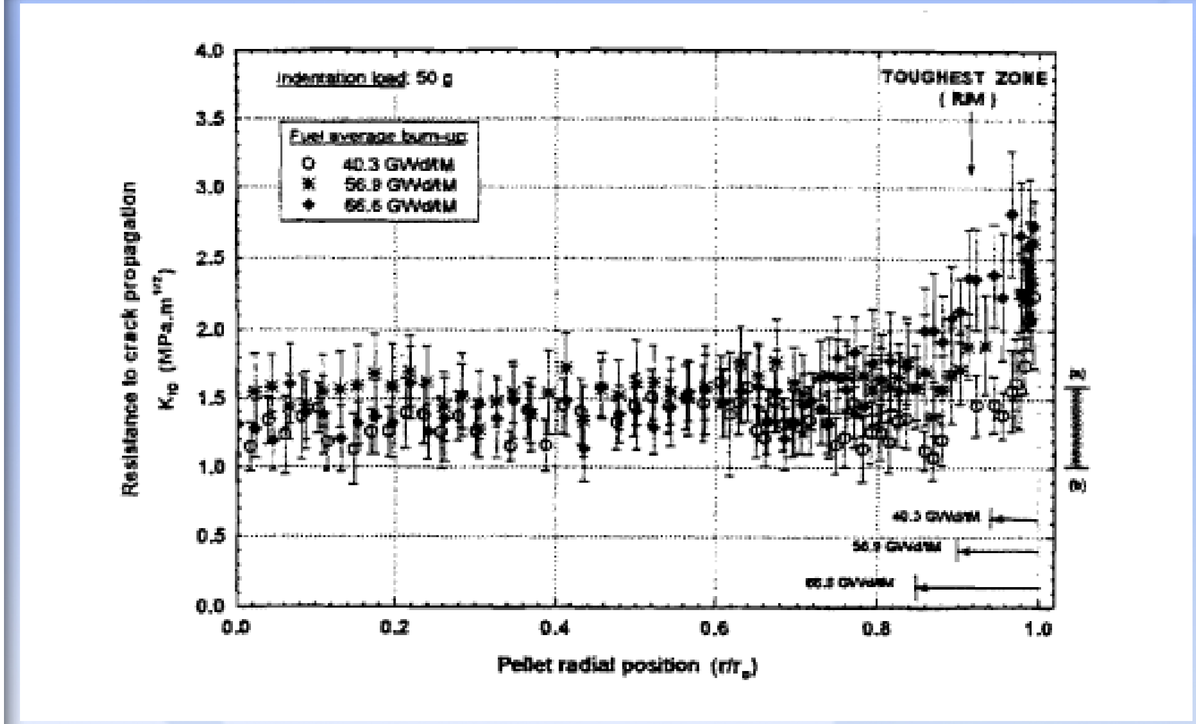
- **Cladding Failure: limited area opening, gas pressure relief, and reconfiguration**
  - *Cut-off probability for exclusion (no opening area) or materials at risk: materials, environments, temperature, and stress*
  - Impact failure
  - Low temperature creep and embrittlement (e.g., hydride reorientation, delayed-hydride cracking) (e.g., Ahn et al., 2007), and stress corrosion cracking
  - Initiation and propagation of cracks: limited size and density in number
  - High-burnup embrittlement: bed filtering effects with Reynold number (Otani et al., 1989)
  - Effect of configurational changes on criticality

Uncertainties in SF Matrix Degradation

- Spent Fuel Matrix
  - Release fraction from rim structure- increase or decrease (figure on next slide)
  - Oxidation (burnup effects)/hydration (e.g., Ahn, 1996b) and radiation damage
  - Impact from operation, seismicity, transportation, fire and aircraft impact: impact energy and oxidation
  - Effects on dissolution: matrix and particle size effects (e.g., Ahn and Mohanty, 2008; Ahn, 1996a)
  - Release by diffusion and migration

Uncertainties in SF Matrix Degradation

(continued)

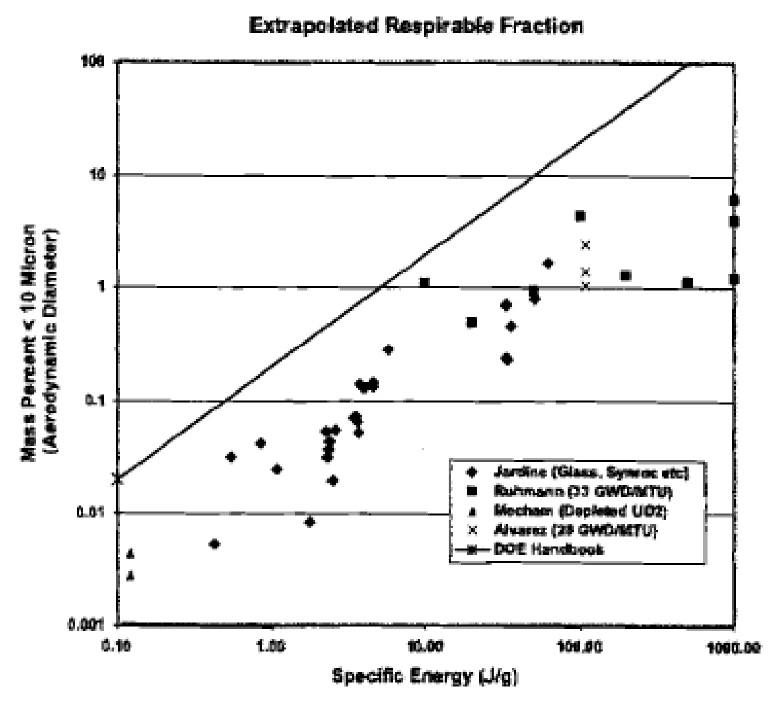


- **Release fraction is sensitive to the number of tears and fraction of rim layer fracture (NRC, 2007) For example, grain/subgrain boundary effects**

Figure: Radial fracture toughness from crack development after microhardness testing at different burnups (Spino et al., 1996). The bar on the right indicates the values under light and heavy loads. [1 Mpa = 0.145 ksi; 1 m = 3.28 feet]. "Resistance to crack propagation" is measured by a combination of the threshold stress and the flaw size necessary for crack propagation, defined here as K_{IC}.

Uncertainties in SF Matrix Degradation

(continued)



- Specific energy (J/g) is the external impact energy (Joule) absorbed in a unit weight (g) of spent fuel.

- Comparison of the DOE Handbook's respirable fraction equation to experimental values of the specific energy input into the brittlement material (NRC, 2007) [1 J = 0.24 cal; 1 kg = 2.2 lb]

- Aging effects

Uncertainties in Neutron Absorber Degradation

- Neutron Absorber
  - Borated Stainless Steel, potentially for long-term storage and disposal:
    - General corrosion (Fix et al., 2004)
    - Pitting and crevice corrosion (Fix et al., 2004)
    - Mechanical failure
    - Phase transformation
  - Others for short-term storage (e.g., BORAL)

Uncertainties in Other Components

- **Concrete Degradation**
  - **Storage overpack and pads**
  - **Freeze-thaw cycles, leaching of calcium hydroxide, radiation embrittlement, thermal expansion, corrosion of embedded reinforcement steel**
  - **Lead to loss of materials, crack formation, and change in concrete properties**
- **Shielding Material (Polymer) Degradation**
  - **Sources: Gamma radiation and high temperature**
  - **Changes in physical configuration (e.g., chain scission, cross-linking, gas evolution)**
  - **Shrinkage**
  - **Boron consumption**
- **Radiation Damaged Elastomer Seals**

Summary

- Potential uncertainties in the risk assessment associated with SF integrity in extended storage and waste disposal
  - Radionuclide inventory
  - Heat generation
  - Gas generation
  - SF UO_2 matrix: powdering by oxidation, powdering by drop / collision / impact, high-burnup rim effects, radionuclide migration and diffusion, dissolution
  - Cladding: embrittlement, mechanical failure, corrosion
  - Canister: mechanical failure by drop/collision/impact, pitting corrosion, stress corrosion cracking, aging (phase transformation and segregation)
  - Neutron absorber
  - Other structures: canister (or cask) internal components, concrete materials, shielding materials, building structure (HEPA filter)

Simulated Spent Fuel (SIMFUEL) Corrosion Test

Background

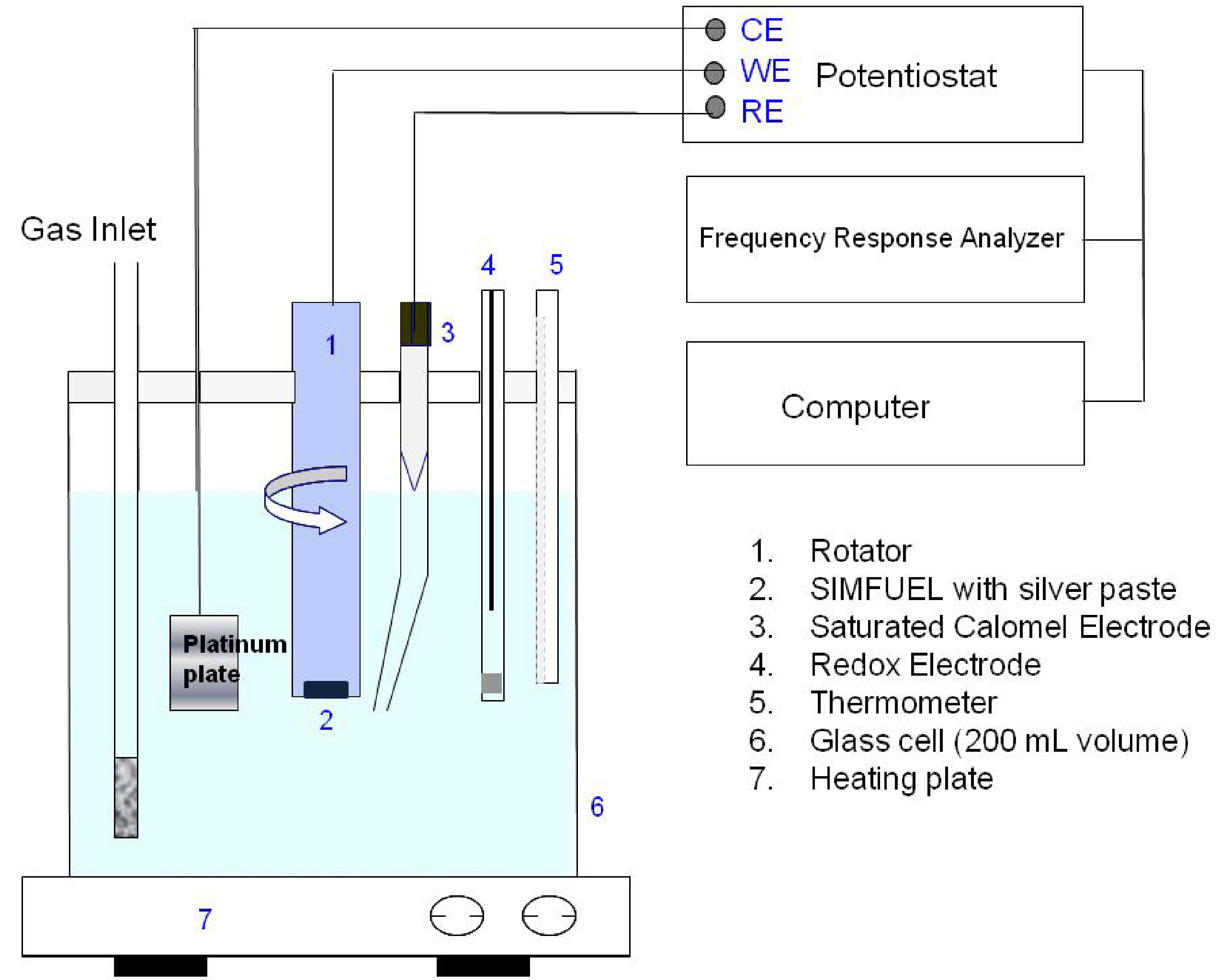
- To understand radionuclide release behaviour from spent nuclear fuel (SNF) to be emplaced in a disposal repository
- To confirm a range of dissolution rates of SNF in a repository relevant environment
- To assess a potential effect of radionuclide (actinides) sorption onto the oxides formed on stainless steel
- SIMFUEL is a synthetic uranium dioxide (UO_2) doped with non-radioactive elements to represent chemical state and phase microstructure of irradiated SNF
- No bubble gases and rim structure in SIMFUEL
- SIMFUEL can represent SNF characteristics in terms of matrix dissolution of SNF
- SIMFUEL can avoid unrealistic radiolysis effects due to relatively strong gamma/beta radiation during an initial repository period after closure (e.g., up to ~1,000 years), because waste package failures could occur much later than such as after ~10,000 years when a level of gamma/beta radiation is expected to be several orders lower and ineffective or negligible on radiolysis of water

Electrochemical Testing

- **Test specimen: (i) rotating disc of SIMFUEL samples with 3 or 6 at% burn-up levels (2 to 3 mm thick, ~12 mm of diameter) and (ii) disc shape of type 316L stainless steel plate for sorption test**
- **Test temperature: room temperature (22 ± 2 ºC) (as a dominant temperature after waste package failure to show significant radionuclide release in consequences)**
- **Test solution (simulated waters for in-package chemistry or ground water, applicable to various disposal sites): Carbonate-based solutions using a mixture of NaCl and $NaHCO_3$ with and without calcium and silica ion addition at pH 7 or 8**

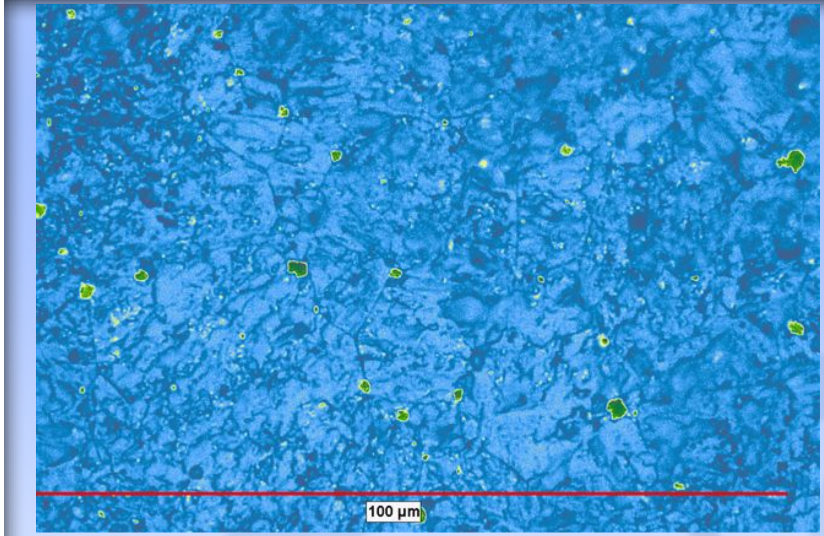
	Solution A (pH = 8, rotation 1000 rpm) Concentration (weight)	Solution B (pH = 7, no rotation) Concentration (weight)
NaCl	0.24 mM (0.014 g)	0.2 mM (0.01117 g)
$NaHCO_3$	0.61 mM (0.0512 g)	0.2 mM (0.0168 g)
$Na_2SiO_4 \cdot 5H_2O$	0.61 mM (0.1391 g)	0.1 mM (0.0228 g)
$CaCl_2$	0.13 mM (0.0144 g)	0.05 mM (0.0056 g)

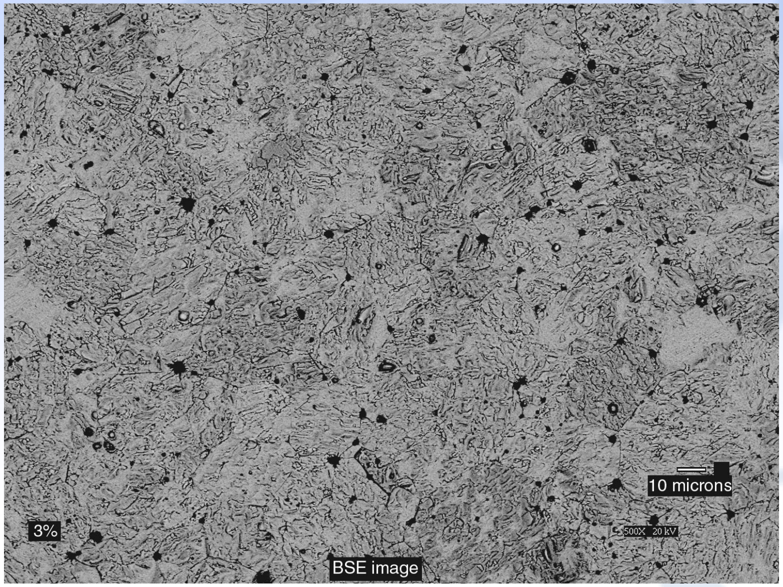
Experimental Set-up

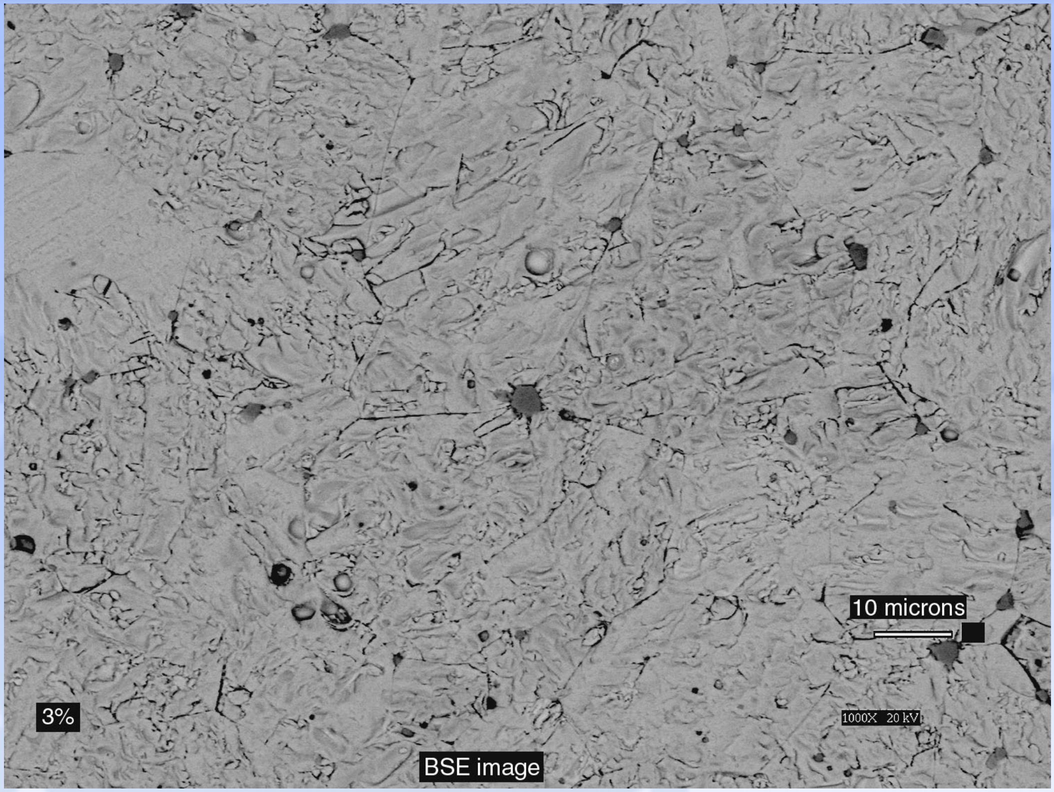


1. Rotator
2. SIMFUEL with silver paste
3. Saturated Calomel Electrode
4. Redox Electrode
5. Thermometer
6. Glass cell (200 mL volume)
7. Heating plate

Microstructure of SIMFUEL (Surface Morphology of 3 at% burn-up)

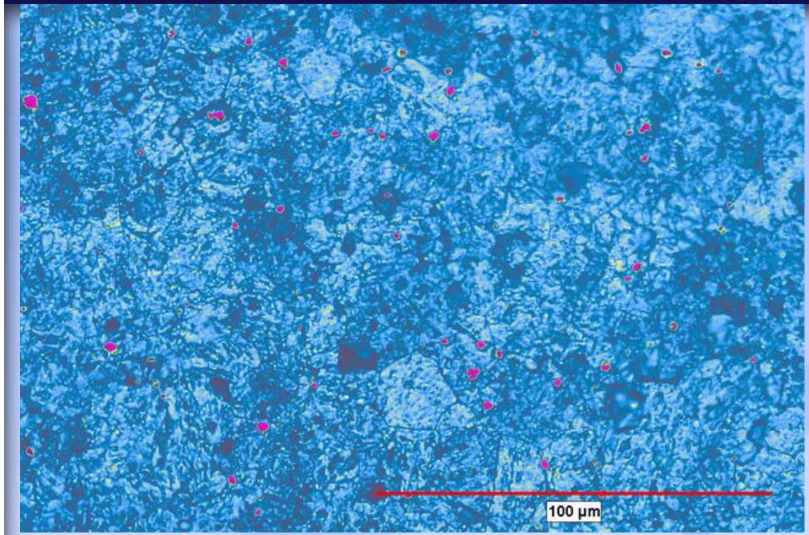


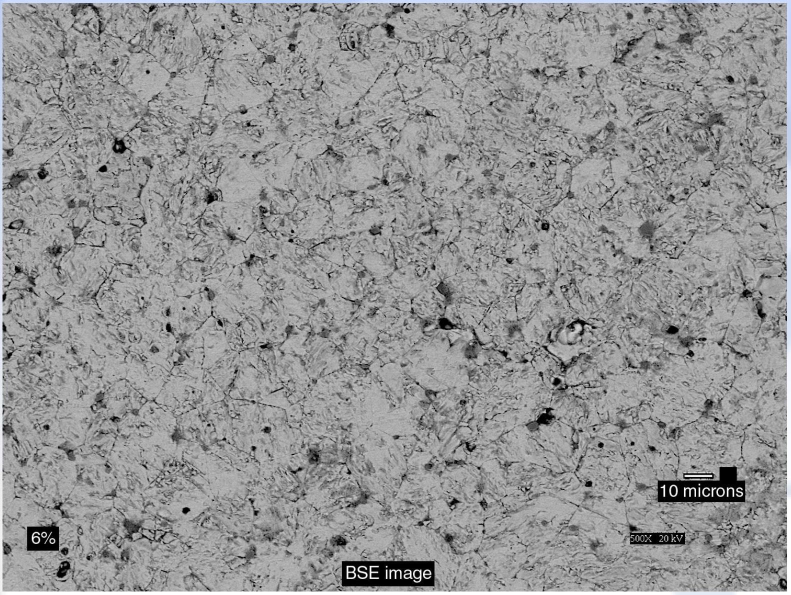


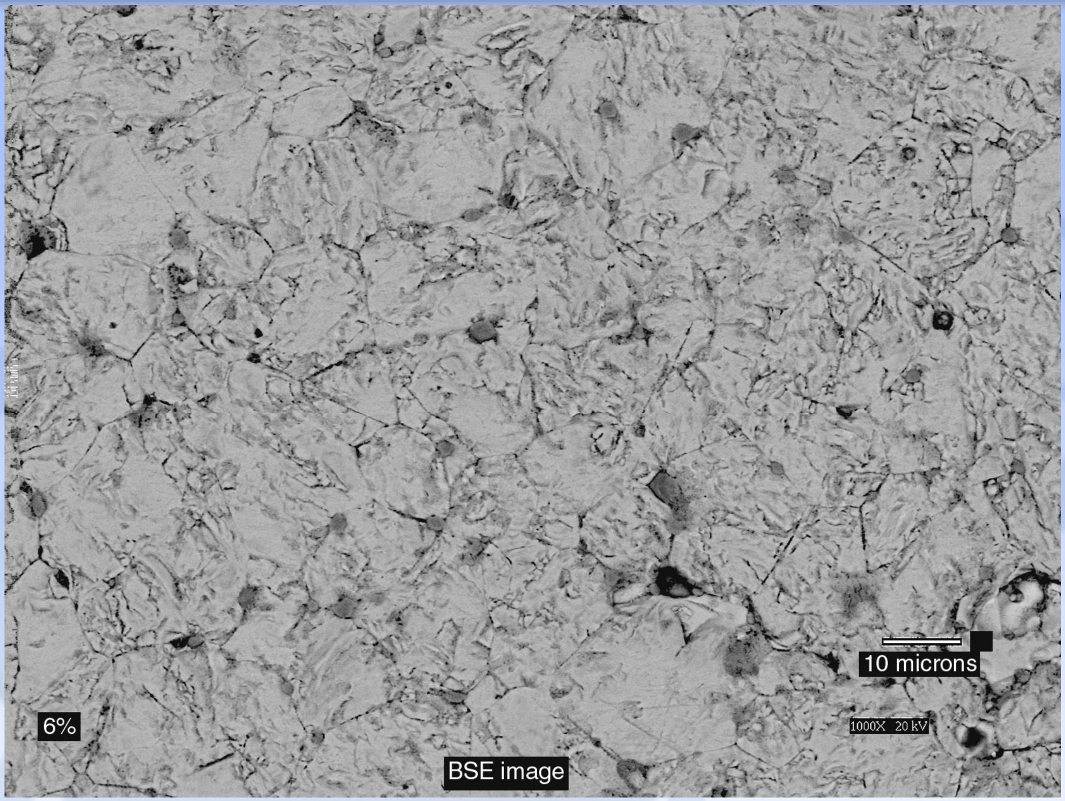


- ❑ Etched in a mixture of sulfuric acid (10 ml), hydrogen peroxide (20 ml), and DI water (70 ml) for 3 minutes at room temperature

Microstructure of SIMFUEL (Surface Morphology of 6 at% burn-up)



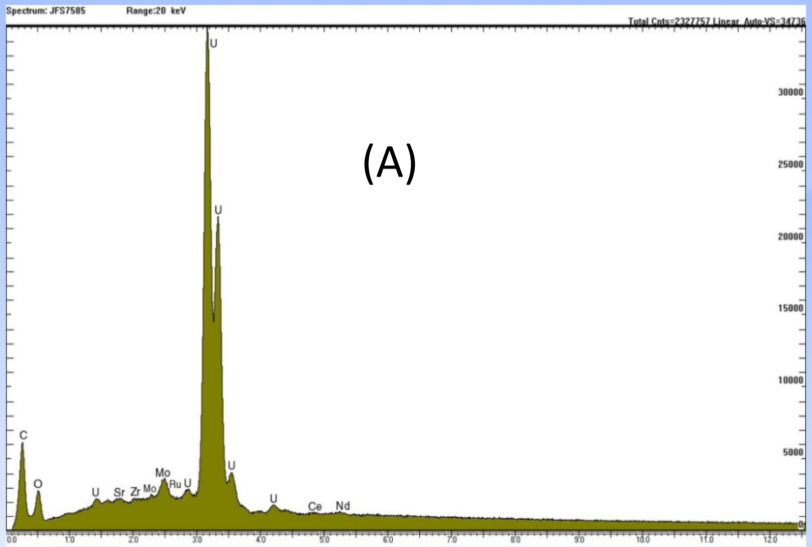


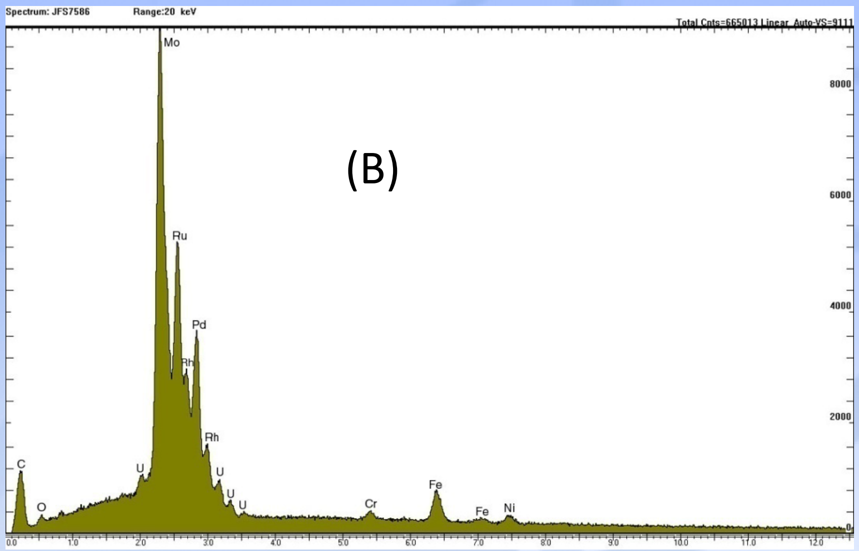


- ❑ Grain size of ~10 µm
- ❑ Precipitates mainly along grain boundary, as well as grain inside

Chemical Composition of SIMFUEL (3 wt% burn-up)

- ❑ Analyzed chemical composition of SIMFUEL by Energy Dispersive Spectroscopy for two different areas including (A) both grain and grain boundary included general surface area (~200 µm×200 µm) and (B) grain boundary precipitates only





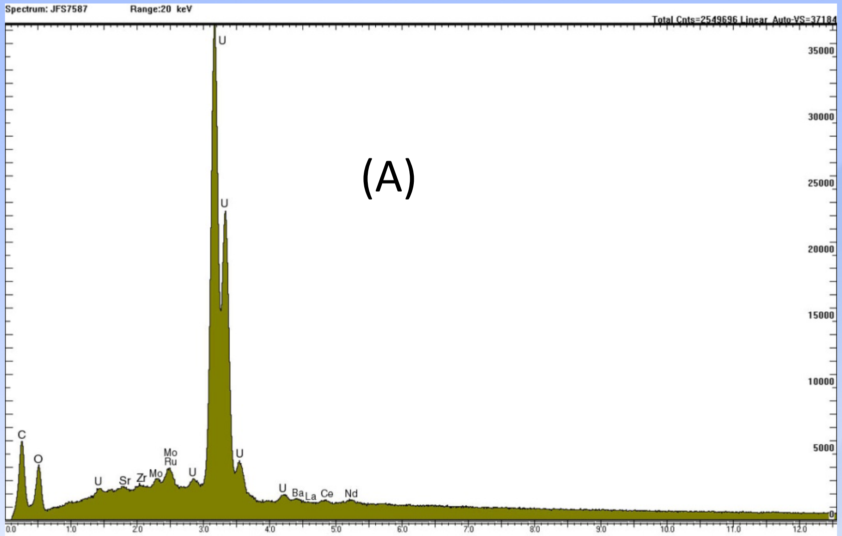
3 at% burn-up	Sr	Y	Zr	Mo	Ru	Rh	Pd	Ba	La	Ce	Nd	U	Cr	Fe	Ni
general surface (wt%)	0.37	<0.01	0.55	0.49	0.51	<0.01	<0.01	<0.01	<0.01	0.37	0.24	97.46	<0.01	<0.01	<0.01
grain boundary precipitate (wt%)				39.26	28.66	3.72	18.09					6.21	0.63	3.42	

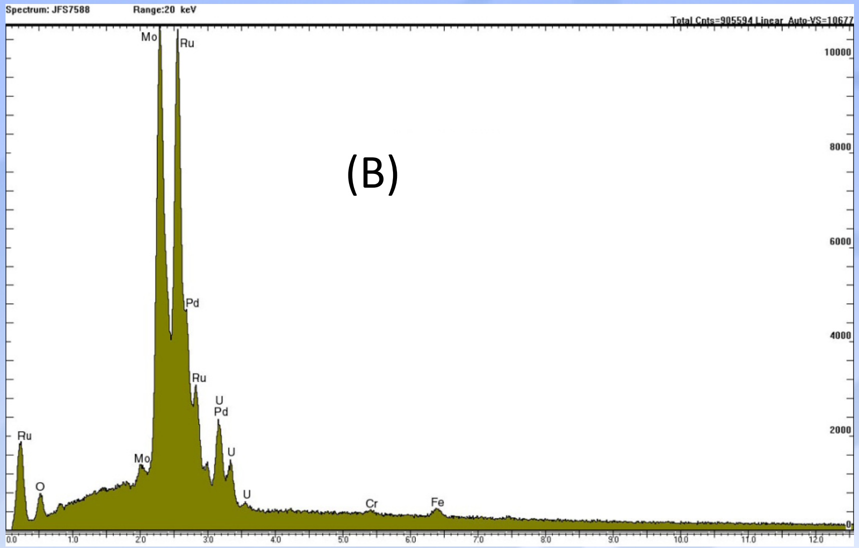
Literature data*	SrO	Y_2O_3	ZrO_2	MoO_3	RuO_2	Rh_2O_3	PdO	$BaCO_3$	La_2O_3	CeO_2	Nd_2O_3	UO_2
3at% burnup (wt%)	0.223	0.04	0.336	0.356	0.36	0.028	0.147	0.15	0.113	0.304	0.494	97.449

* Rondinella and Matzke, J. Nucl. Mat. 238 (1996) 44 - 57

Chemical Composition by EDS (6 at% burn-up)

- ☐ Analyzed chemical composition of SIMFUEL by Energy Dispersive Spectroscopy for two different areas including (A) both grain and grain boundary included general surface area (~200 μm×200 μm) and (B) grin boundary precipitates only





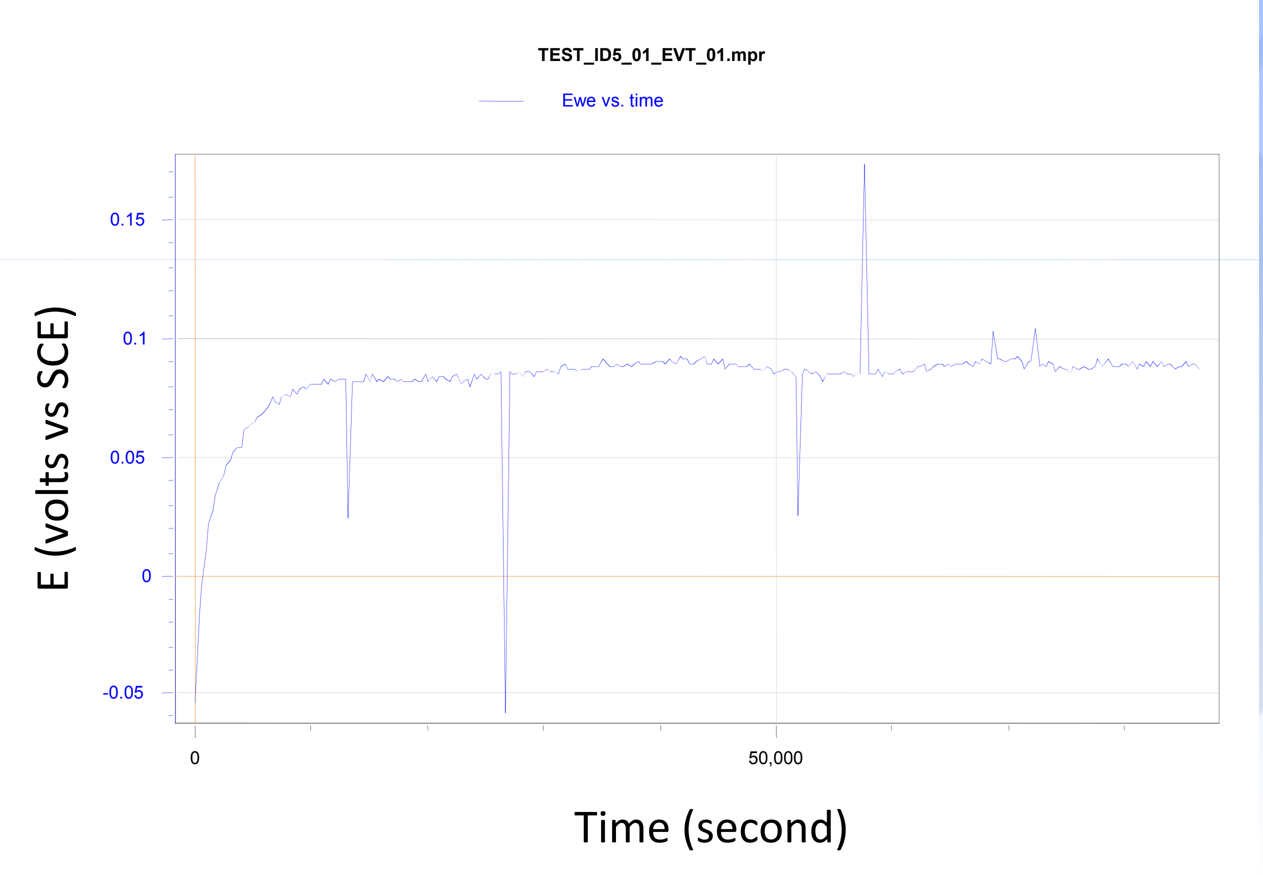
6 at% burn-up	Sr	Y	Zr	Mo	Ru	Rh	Pd	Ba	La	Ce	Nd	U	Cr	Fe	Ni
general surface (wt%)	0.34	<0.01	0.73	1.11	0.52	<0.01	0.21	0.64	0.28	0.66	0.49	95.02	<0.01	<0.01	<0.01
grain boundary precipitate (wt%)				30.47	42.02		10.05					6.21	0.63	3.42	1.31

Literature data *	SrO	Y_2O_3	ZrO_2	MoO_3	RuO_2	Rh_2O_3	PdO	$BaCO_3$	La_2O_3	CeO_2	Nd_2O_3	UO_2
6at% burnup (wt%)	0.406	0.06	0.593	0.72	0.754	0.034	0.434	0.307	0.254	0.545	1.027	94.866

* Rondinella and Matzke, J. Nucl. Mat. 238 (1996) 44 - 57

Open Circuit Potential

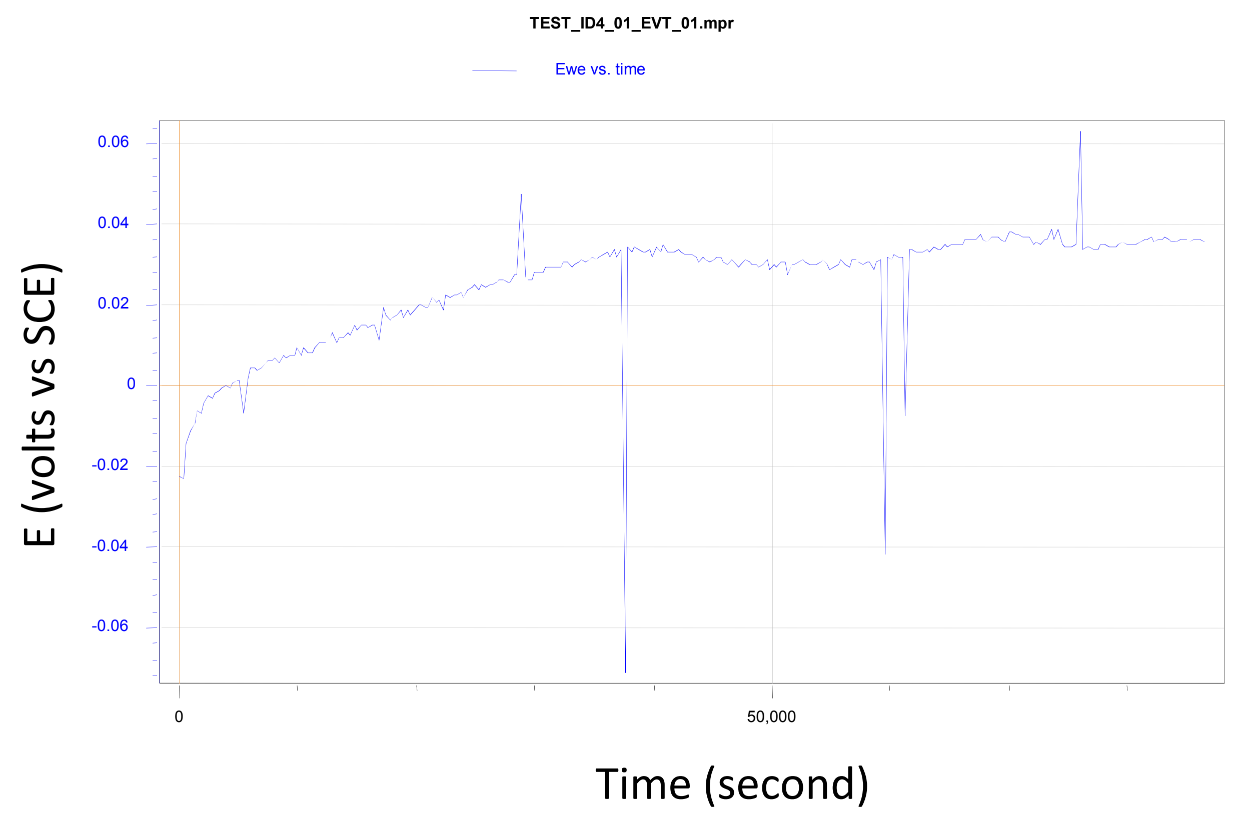
- 6 at% burn-up SIMFUEL tested in the solution A without silica and calcium ions



- Open circuit potential increased with time
- Reached to 0.09 V_{SCE} after 72 hours immersion

Open Circuit Potential

- 6 at% burn-up SIMFUEL tested in the solution A with silica and calcium ions

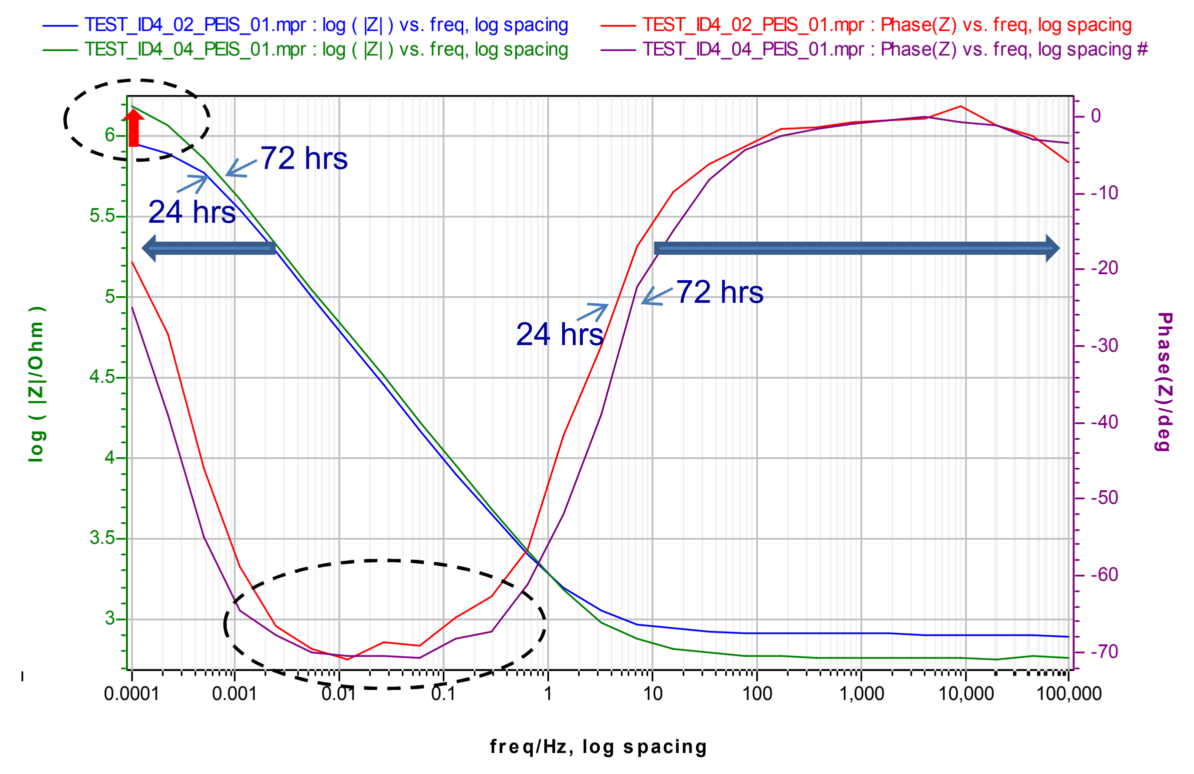


- Slow increase of open circuit potential with time
- Reached to 0.035 V_{SCE} after 72 hours
- Lower open circuit potential than the without silica and calcium addition

Impedance Spectra

(6 at% burn-up SIMFUEL tested in the solution A without silica and calcium ions)

- ❑ Corrosion resistance is generally proportional to the magnitude of impedance (resistance) modulus, |z| (high modulus indicates high corrosion resistance)
- ❑ Reaction mechanisms (e.g., passivation or purely anodic dissolution) can be identified by the shape of phase angle vs. frequency curve (called time constant)

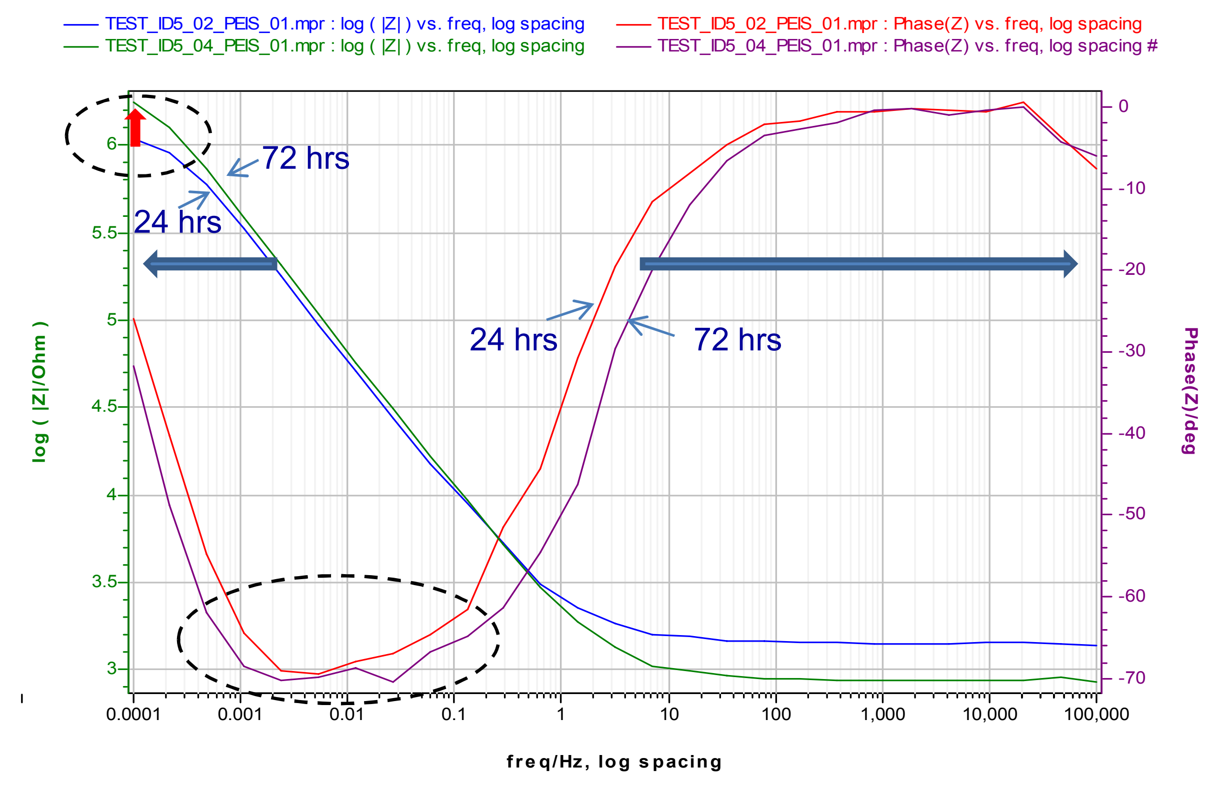


- ❑ The modulus (corrosion resistance) increased with time (e.g. 24 and 72 hrs responses).
- ❑ The shape of phase angle vs. frequency curves for 24 and 72 hrs is very similar, but appears a little broader for 72 hrs curve in low frequency range

Impedance Spectra

(6 at% burn-up SIMFUEL tested in the solution A with silica and calcium ions)

- Corrosion resistance is generally proportional to the magnitude of impedance (resistance) modulus, |z| (high modulus indicates high corrosion resistance)
- Reaction mechanisms (e.g., passivation or purely anodic dissolution) can be identified by the shape of phase angle vs. frequency curve (called time constant)



- The modulus increased with time.
- The shape of phase angle vs. frequency curves for 24 and 72 hrs is very similar, but appears a little broader for 72 hrs curve in low frequency range

Corrosion Rate Estimate

- **Stern-Geary Equation**

  Corrosion current, $i_{corr} = B / R_p$

  $$B = b_a \times b_c / [2.303(b_a + b_c)]$$

  where, R_p, polarization resistance; b_a and b_c, anodic and cathodic tafel slopes (used 0.06 and 0.2 V, respectively)

- **Faraday's law**

  Corrosion rate = $K_2 \times i_{corr} \times EW$

  where, K_2, constant (0.0895 mg cm^2/μA dm^2 day); EW, equivalent weight (for U, 59.5 = 238/4 = W/n)

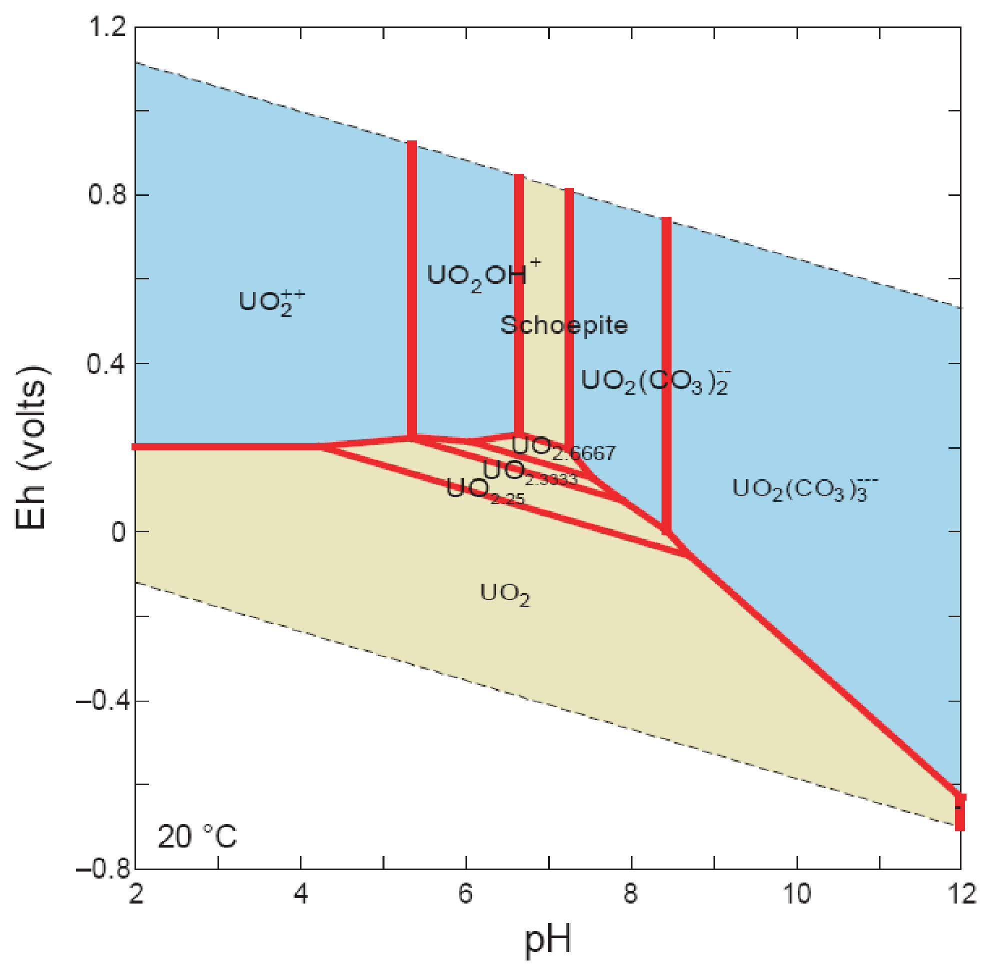
Results Summary

- Exposed surface area of SIMFUEL: 1.13 cm^2
- Electrical circuit for fitting impedance spectra: simple Randle's circuit (R1+Q2/R2)
- Electrical resistivity of SIMFUEL: 2.7 – 2.8 kohms
- Based on the measurement results after 72 hrs immersed

	Solution A (1000 rpm)				Solution B (no rotation)	
Burn-up (at%)	3%		6%		6%	
Silica and Ca addition	Without	With	Without	With	Without	With
Open circuit potential (V_{SCE})	0.12	0.085	0.1	0.06	0.1	0.09
Polarization reistance (Ohms)	5.76E+06	6.66E+06	2.95E+06	2.20E+06	3.48E+06	3.04E+06
Corrosion current (A/cm^2)	3.08E-09	2.66E-09	6.01E-09	8.06E-09	5.10E-09	5.83E-09
Corrosion rate (mg/m^2 d)	**1.6**	**1.4**	**3.2**	**4.3**	**2.7**	**3.1**

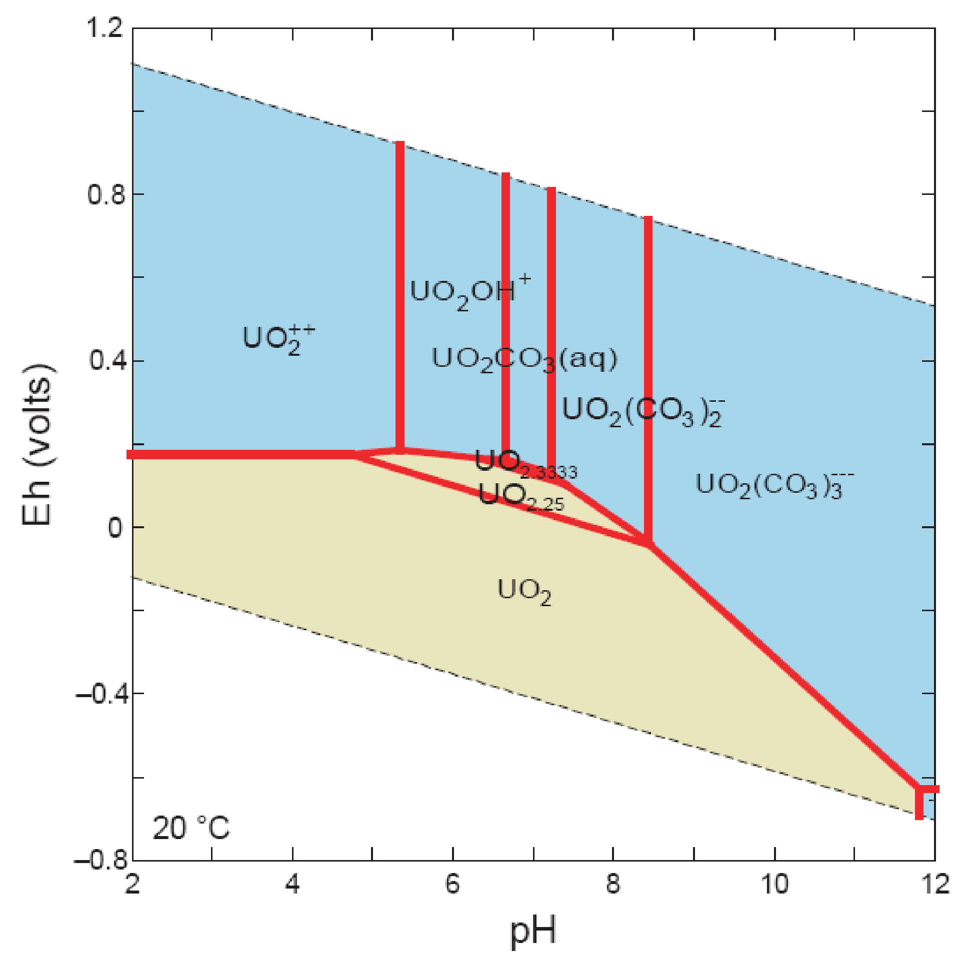
Potential-pH Diagram of U-Simulated Water System at 20 ºC

- Activity of dissolved species (e.g., UO_2^{++}): **10^{-7} M**



Potential-pH Diagram of U-Simulated Water System at 20 ºC

- Activity of dissolved species (e.g., UO_2^{++}): **10^{-8} M**



Preliminary Results – dissolution

- Addition of silica and calcium ions or higher burn-up (6 at%) led to lower open circuit potential after 72 hours compared to without or lower burn-up (3 at%), respectively.

- The polarization resistance of 6 at% burn-up fuel was lower than 3 at%, then higher dissolution rate of 6 at% fuel. However, a difference was minimal.

- Effect of silica (up to 0.61 mM) and calcium ions (up to 0.13 mM) addition was not significant to dissolution rate of SIMFUEL at room temperature.

- A formation of secondary phase such as $UO_3 \cdot 2H_2O$ is thermodynamically possible, which may be a reason of increase in polarization resistance with immersion time under the conditions tested.

Current Status

- ❑ Completed analyses of corrosion test results of 3 and 6 at% burn-up SIMFUEL conducted in a simulated water at room temperature
- ❑ Conducted microstructural characterization of SIMFUEL using optical microscopy, scanning electron microscopy and energy dispersive spectroscopy
- ❑ Conducted sorption test using stainless steel immersed in the post-test solutions
- ❑ Requested solution chemistry analysis using ICP-MS analyses for pre- and post-test solutions to estimate chemical concentrations of dissolved species in the solutions including Ca, Si, Cr, Fe, Ni, Sr, Y, Zr, Mo, Ru, Rh, Pd, Ba, La, Ce, Nd, and U

Future Plan

- ❑ Longer term dissolution tests of SIMFUEL to confirm evolution of polarization resistance with time
- ❑ Conduct dissolution test at different temperatures (e.g., 50 and 70 ºC) to establish an Arrhenius type dissolution rate equation
- ❑ Conduct surface analysis of stainless steel 316L after sorption test to evaluate potential sorption of actinides onto the oxides
- ❑ Analyze the first set of ICP-MS analysis results and compare those results to electrochemical test results in terms of dissolution rate
- ❑ Design and conduct dissolution (degradation) test of SIMFUEL under reducing condition

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Note: All NRC ADAMS documents can be found at <www.nrc.gov/reading-rm/adams.html> using the accession number (MLxxxxxx).

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