DOE-EM Sponsored Research on Long-Term Dry Storage of Aluminum-Clad SNF

Michael Connolly, Ph.D.

Nuclear Waste Technical Review Board
Fall 2019 Board Meeting
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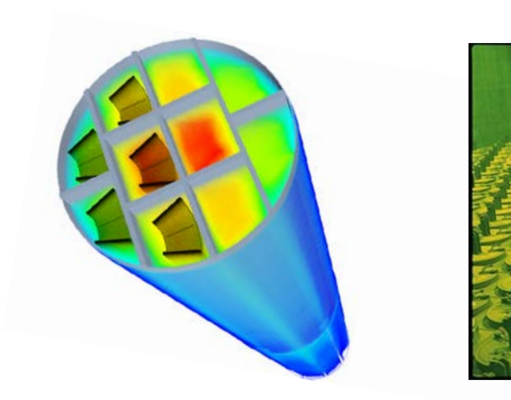
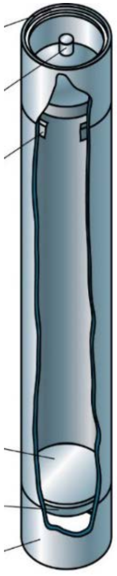




Presentation Overview

- Description of DOE Research Test Reactor Al clad SNF
- Identification of knowledge and technical data gaps associated with long-term dry storage of ASNF
- Description of DOE EM funded activities to address identified knowledge and technical data needs
- Summary and future activities

Program Contributors

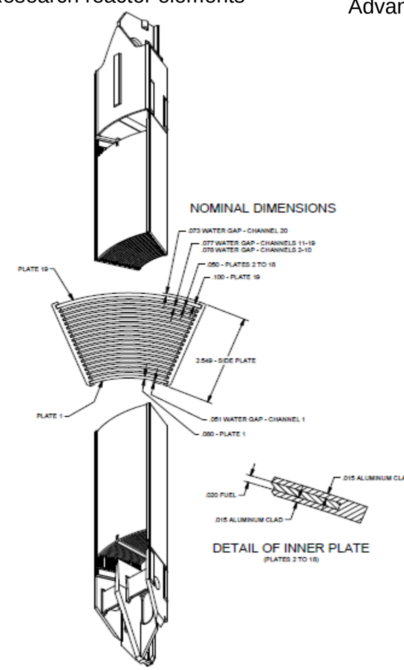
- National Laboratory, University, and Industry collaboration
  - Idaho National Laboratory
  - Savannah River National Laboratory
  - University of South Carolina
  - Holtec International
  - Fluor Idaho
- DOE Environmental Management and Nuclear Energy
- More than 30 individual contributors

DOE Aluminum Clad SNF

- Mostly HEU fuel
  - ✓ U-235 enrichments to 93%
- Surface hydroxides/oxyhydroxides formed as a result of in-reactor and post-discharge conditions
  - ✓ Free, physisorbed, and chemisorbed water
  - ✓ Radiolytic gas generation
- Aluminum physical characteristics (vis-à-vis Zr, SS)
  - ✓ Lower neutron capture cross-section
  - ✓ Susceptible to corrosion
  - ✓ Lower melting temperature
- Research test reactor fuel elements
  - ✓ Higher surface area
  - ✓ Thinner clad (<0.5 mm)
  - ✓ Experience extreme and variable reactor conditions

Research reactor elements

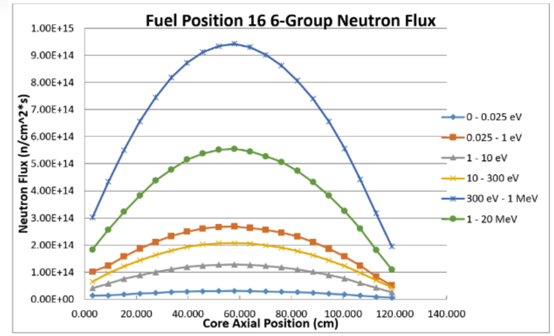

Advanced Test Reactor element



Advanced Test Reactor element

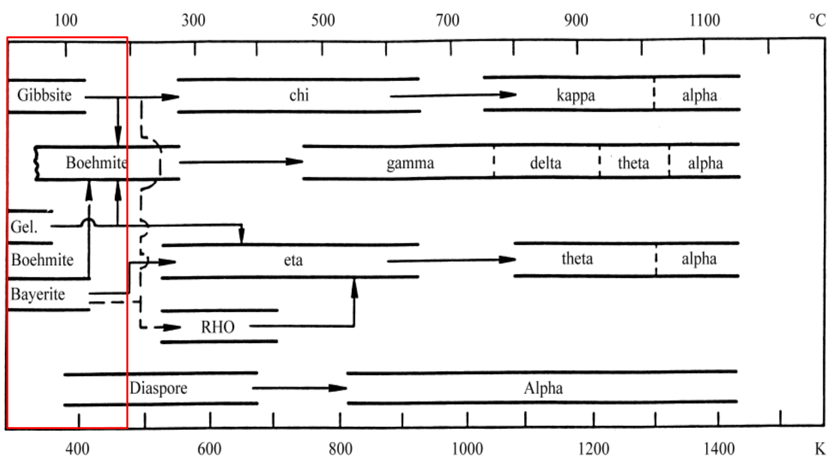
Potential Technical Issues

- In-reactor effects on cladding
- Water storage chemistry
- Resulting hydrated oxide (e.g. boehmite, gibbsite, bayerite) corrosion layer
- Radiolytic production of gases (e.g. H_2)
- Chemical/Thermal behavior of corrosion layer
- Resulting challenge to storage systems
- $2Al + 6H_20 \rightarrow 2Al(OH)_3 + 3H_2$ (< 77°C)

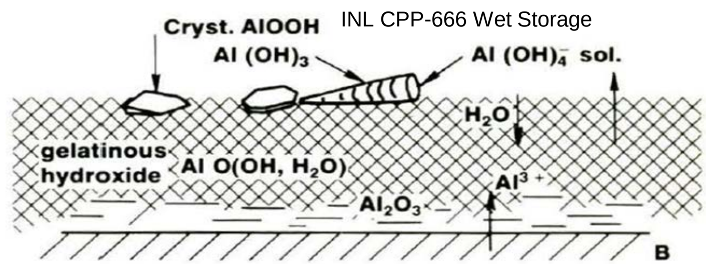


M.Glazoff, T.Lister, R.Smith, INL/Ext-18-51694, Nov 2019

INL CPP-666 Wet Storage



K Wefers and C. Mistra, Oxides and Hydroxides of Aluminum, Alcoa Technical Paper No. 19, Alcoa Laboratories, 1987



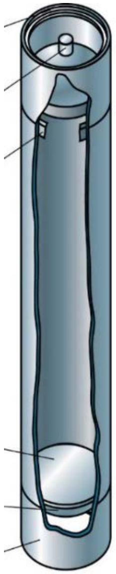
A: Al Immersed in Water, 1 day at 335K
B: Film Texture, Schematic

Multilayered Hydroxide Film on Aluminum

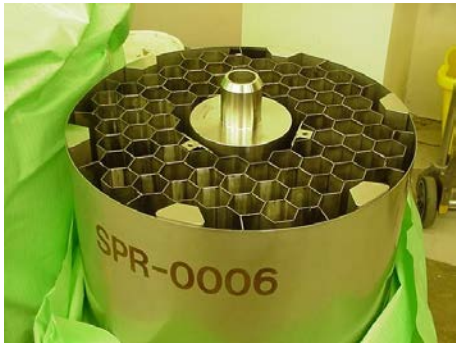
Evaluation of Al SNF Dry Storage Sytems

- Evaluated existing and planned storage systems
- Evaluated technical and engineering studies performed since the late 1950s
- DOE issued report documenting study results - *Aluminum Clad Spent Nuclear Fuel: Technical Considerations and Challenges for Extended (>50 Years) Dry Storage, DOE-ID/RPT-1575, June 2017*
  - ✓ Chemistry/behavior of hydrated oxide layers in dry storage systems
  - ✓ Radiolytic gas generation data for hydrated oxide layers
  - ✓ Combined effect of episodic breathing and radiolytically generated corrosive gases
  - ✓ Performance of ASNF in existing dry storage
  - ✓ Effects of high (>100^{o}C) temperature drying on oxyhydroxide layers

INL CPP-603 dry storage facility



DOE Standard Canister

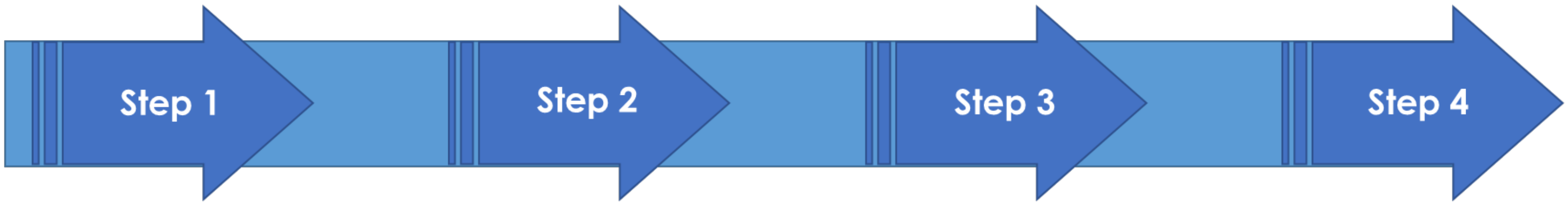


Hanford MCO Single Pass Reactor Fuel Basket

ASNF Extended Dry Storage Action Plan – Technical and Engineering Activities, INL/EXT-17-43908, Nov 2017

- Developed in response to DOE-ID/RPT-1575
  - ✓ Recommends multi-year *laboratory* based studies coupled with modeling and simulation
  - ✓ Action plan identifies six tasks to address identified knowledge gaps and technical data needs
- Six integrated technical tasks
  - ✓ Hydrated oxide layer behavior and chemistry
  - ✓ Hydrated oxide layer radiolytic gas generation characterization
  - ✓ Sealed and vented dry system modeling
  - ✓ Performance of ASNF in dry storage
  - ✓ Hydrated oxide layer response to drying
  - ✓ Surrogate sample preparation
- Funded by EM Office of Technology Development
  - ✓ Initiated January, 2018

Enabling Road Ready Dry Storage of Aluminum Spent Nuclear Fuel – Technology Readiness



Identify Problem

- DOE H-Canyon options analysis
- Al clad-SNF WG Gaps Report
- 5 technology gaps identified

Laboratory Studies

- Surrogate studies
- PIE to validate surrogates
- Modeling and simulation
- Drying studies

Develops preliminary technical basis for drying and dry storage of ASNF

Validation/Verification

- Drying + Instrumented canister demo (Validation)
- PIE post-canister demo (verification)
- Improved modeling and simulation
- Draft final technical basis

Improves the technical basis for drying and dry storage of ASNF

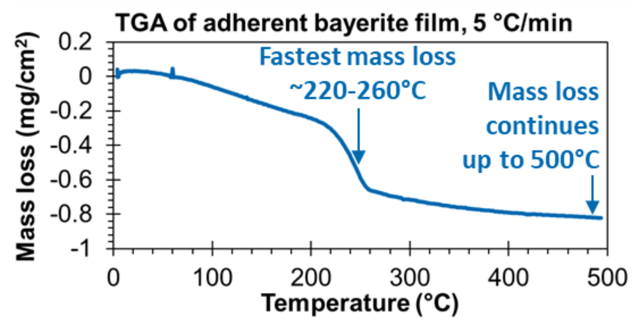
Deployment

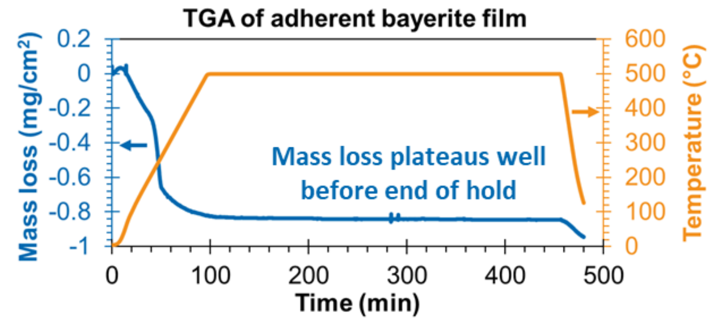
- Technical basis enables licensing of a road-ready ASNF dry storage system

Multi-year DOE EM funded program addressing steps 2-3

Chemistry and Reactivity of Hydrated Oxide Films

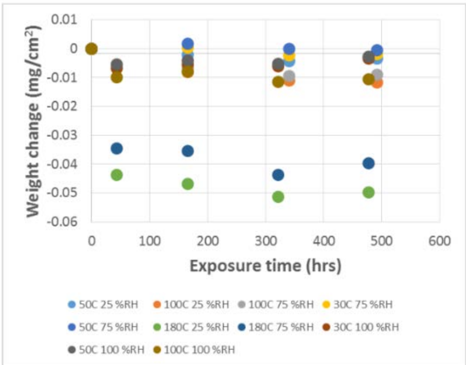
- Lab studies on prefilmed substrates across expected humidity and temperature ranges
- Higher temperature studies to support elevated temperature fuel drying studies
- Preliminary results show
  - ✓ Elevated (~200^{o}C) temperature required for decompose hydrated oxides
  - ✓ Weight loss due to dehydration
- Advanced thermodynamic and kinetics modeling support results



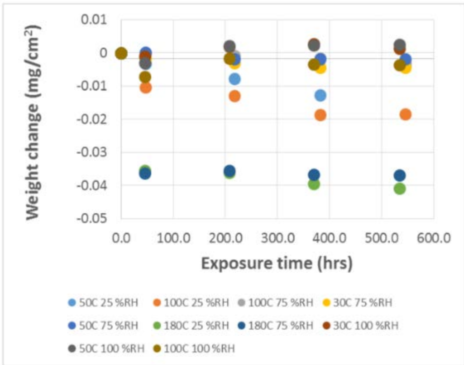


Savannah River National Laboratory FY19 LDRD Results

Drying temperature (°C)	200	220	260	500
Hold time (h)	24	24	4	6
Mass loss (mg/cm^2)	0.78	0.87	0.90	1.02



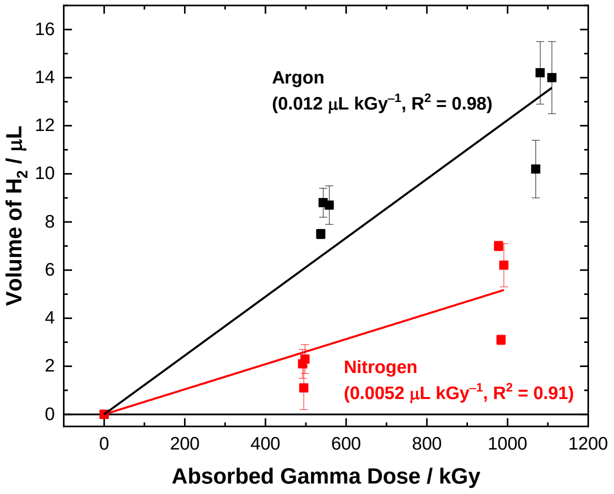
Weight change vs exposure for AA1100



Weight change vs exposure for AA6061

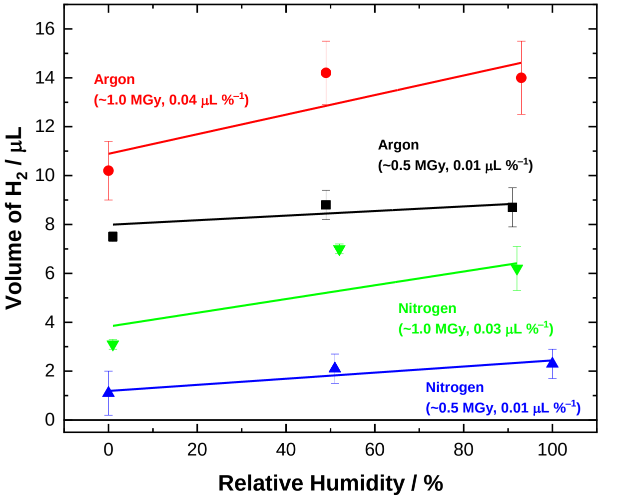
Radiolytic Gas Generation Characterization

- Lab studies on prefilmed substrates and across humidity, temp, and gas environments
- Gamma irradiation using ^{60}Co source (46 Gy/min)
- Initial results show
  - ✓ Films promote H_2 formation
  - ✓ H_2 G-values appear dependent on gas composition
- H_2 G-values based on energy partitioning between corrosion layer and substrate

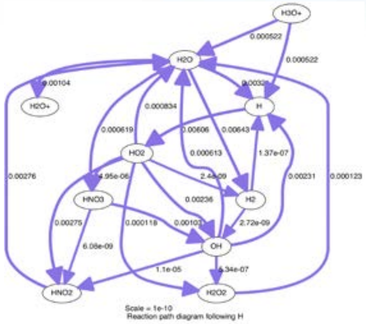


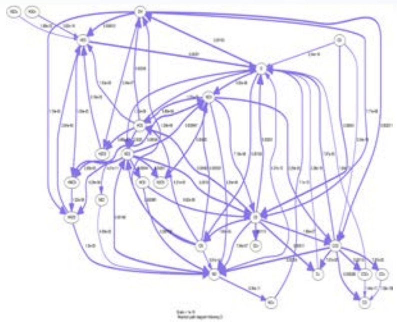
gas phase	relative humidity (%)	absorbed gamma dose (kGy)	H_2 production (μL)	*G*-values for boehmite (molecule/100 eV)	*G*-values for bayerite (molecule/100 eV)
Ar (max)	49	543	8.8 ± 0.6	1.9313 ± 0.0998	1.8831 ± 0.0973
Ar (min)	0	1070	10.2 ± 1.2	1.1441 ± 0.1047	1.1155 ± 0.1519
N_2 (max)	52	978	7.0 ± 0.2	0.8201 ± 0.0161	0.7996 ± 0.0157
N_2 (min)	0	495	1.1 ± 0.9	0.2639 ± 0.1388	0.2574 ± 0.1354
Ar (max)	0 (100 °C)	500	7.6 ±0.5	1.7449 ± 0.0671	1.7013 ± 0.1308
Ar (min)	0 (200 °C)	1000	11.3	1.3554	1.3215

Calculated H_2 G-value assuming boehmite or bayerite

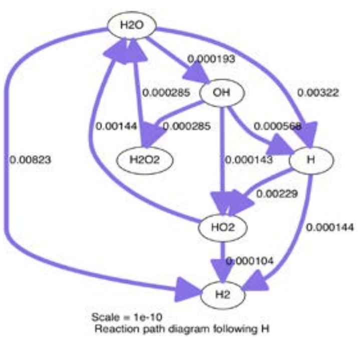


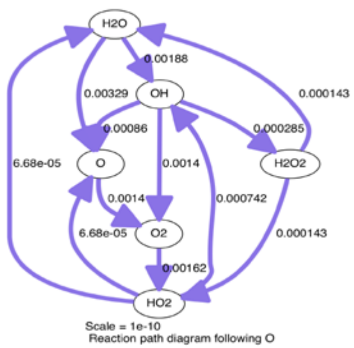
Modeling and Simulation of ASNF Dry Storage System



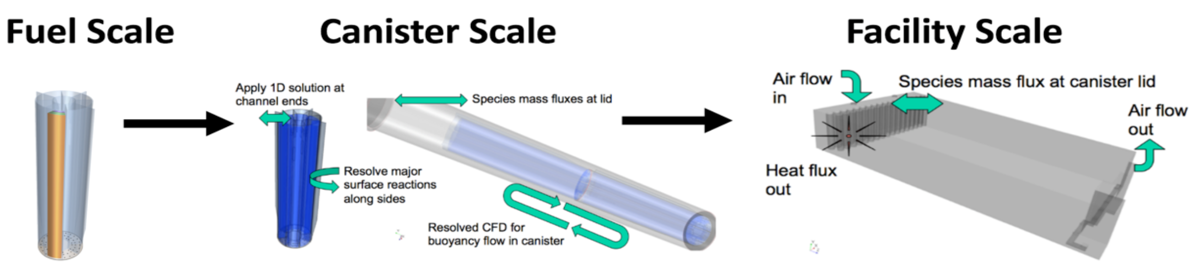


Example full reaction pathways, above, for H (left) and O (right) based on Wittman and Hanson's (2015) radiolysis model for 99% Helium and 1% residual H_2O in fill gas. 40 species and 115 total reactions. Reduce reaction pathways, below, for H (left) and O (right). 8 species and 22 total reaction.



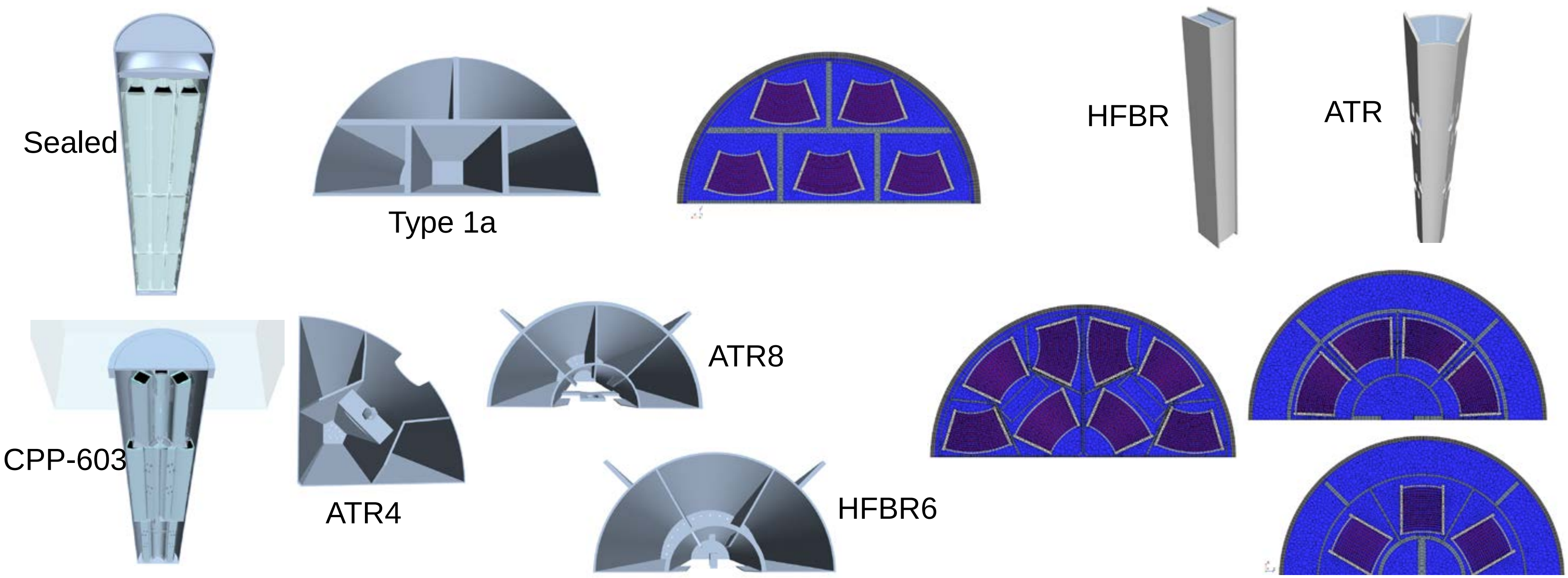


- Coupling all relevant processes
- Multiphysics CFD models
  - ✓ Thermal convection and mass transport inside canister
  - ✓ Gas production kinetics of Al oxyhydroxide layer
  - ✓ Bulk gas phase radiolysis reactions
  - ✓ Mass and heat exchanges with ambient air of transient humidity/temperature changes
- Reliable predictions of long-term evolutions of the temperature and gas phase concentrations for
  - ✓ Sealed canister (e.g., DOE standard canister)
  - ✓ Vented canister (CPP-603)
- Close integration with experimental tasks
  - ✓ Model validations
  - ✓ Guiding experimental designs



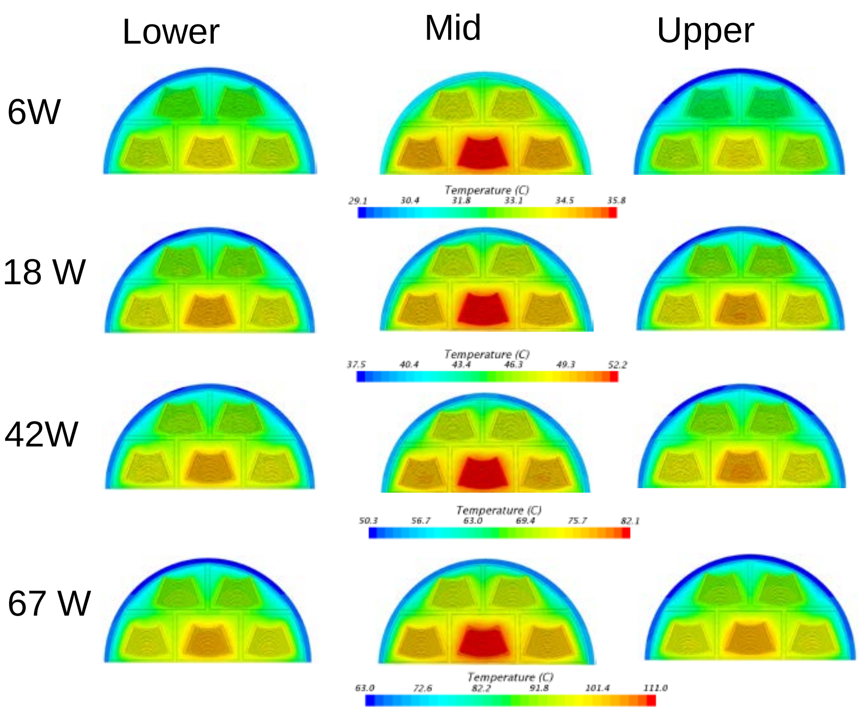
Dry Storage System Canister Models

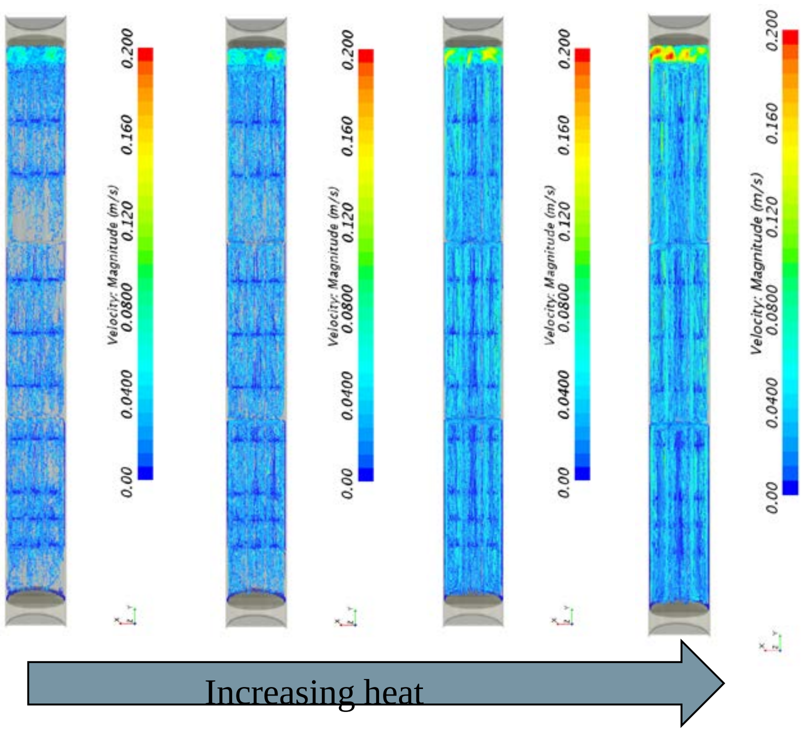
- 18” Diameter canister, 15-foot height with 3 type-1a buckets loaded with 10 ATR elements each
- 18” Diameter, ~11 foot with 1 ATR-8 bucket, 4 ATR-4 buckets, 1 HFBR bucket (24 ATR, 6 HFBR elements)



Sealed Canister Velocities/Temperatures

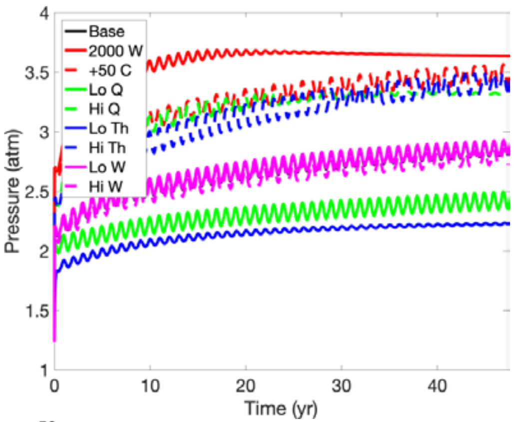
- Use CFD temperature to initialize Cantera with 5 temperature zones, and use recirculation flow as mass exchange between zones

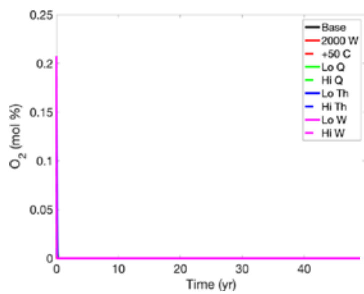


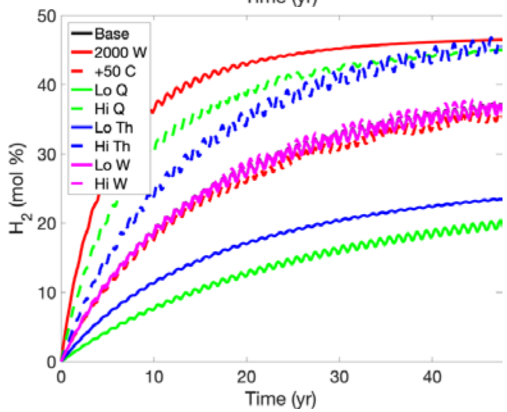


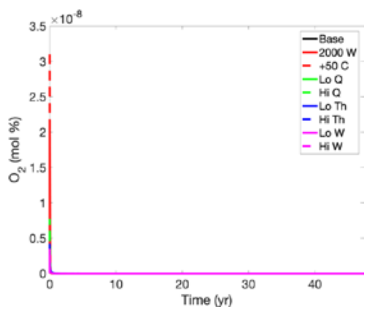
Preliminary Sealed Results

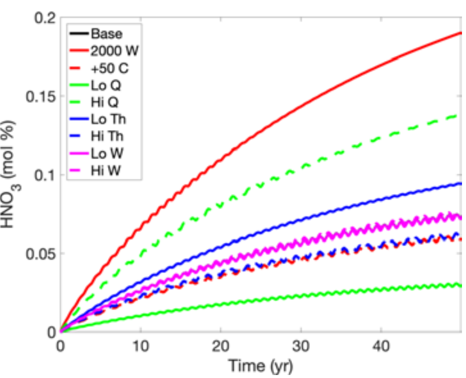
- All cases – even low heat decay reach well above 4% H2, but all show negligible quantities of O_2 present, with small amounts 100-1000 ppm HNO_3 across cases
- The pressure increases sharply at first from thermal dehydration of pseudo boehmite and slowly from H_2 release from radiolysis of boehmite layer
- Main parameters show 2-4 atm variation, large thickness variation shows up to 5.5 atm with 65% H_2

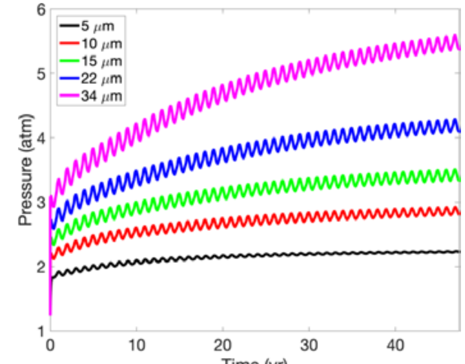


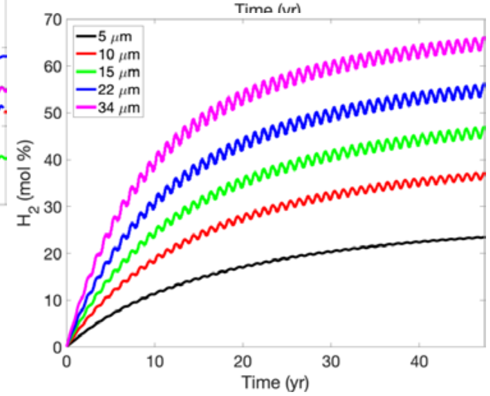










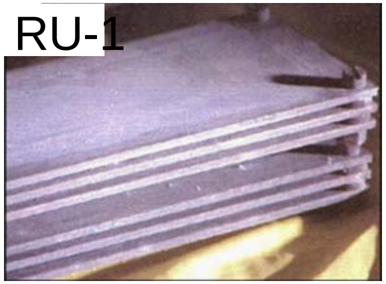


Characterization of Stored ASNF

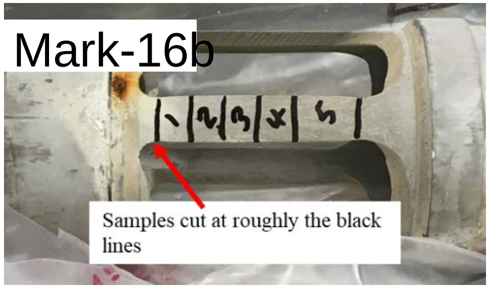
- Investigate performance of ASNF stored in wet and dry storage
- ASNF and ASNF components from
  - ATR operating canal
  - INL CPP-603 dry storage
  - L Basin wet and dry
- Characterize attendant surface film composition and morphology (e.g. bayerite, gibbsite, boehmite)

ATR end box and end box piece - wet storage

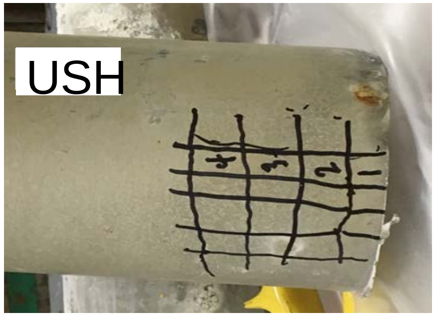

ATR element with end box removed



Uruguay RU-1 reactor element – dry storage



SRS Mark-16B from production reactor – wet storage



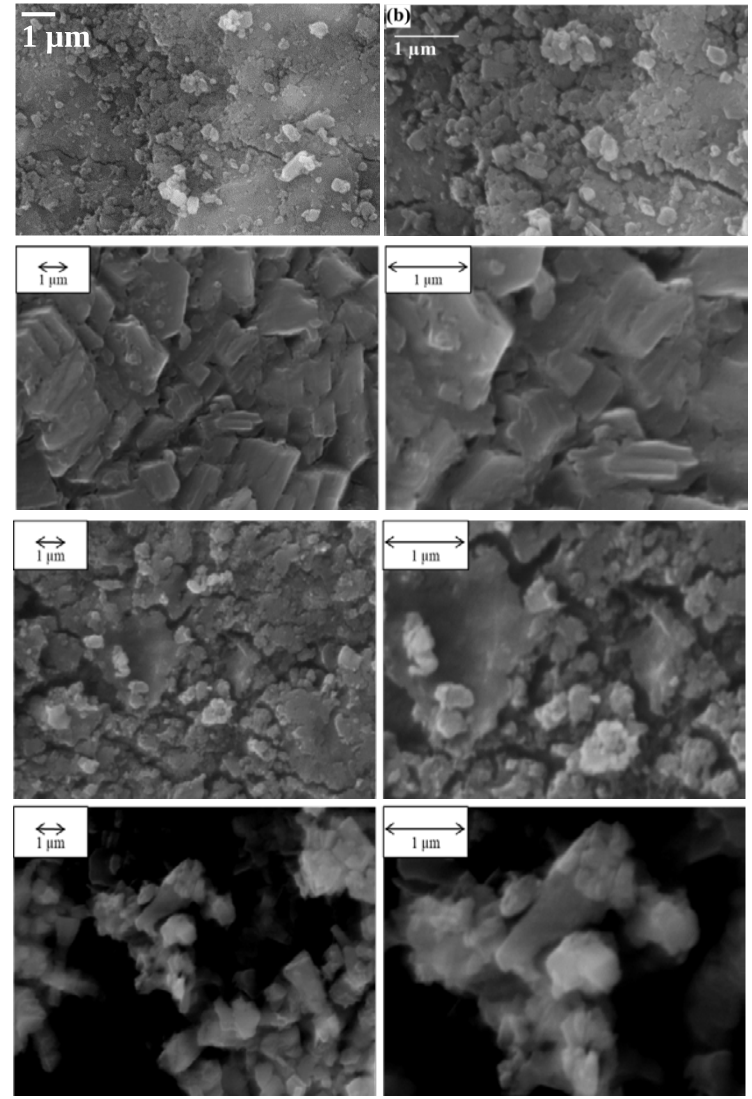
SRS universal sleeve housing from production reactor – wet storage

Missouri University Research Reactor element – wet storage

Characterization of Stored ASNF (cont)

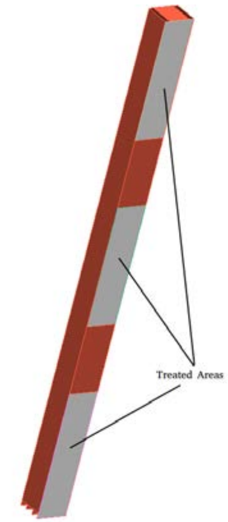
- RU-1 (Al-1100)
  - ✓ In-reactor ~70° C for 8 y; ~30 y dry storage
  - ✓ XRD: gibbsite (P), possible boehmite (S)
  - ✓ Film appears dense, 200 nm–25 μm thick
  - ✓ Irregularly shaped blocks or flakes
- MURR (Al-6061)
  - ✓ In-reactor ~113 d at ≥60° C; <18 y wet storage (~22° C)
  - ✓ XRD: bayerite (P), boehmite (S)
  - ✓ Film appears dense, 5-10 μm thick
  - ✓ Hexagonal blocks or scales
- USH (Al-6063)
  - ✓ In-reactor ~1800 d at ≤93° C; ~40 y wet storage (~22° C)
  - ✓ XRD: boehmite (P) and bayerite (S)
  - ✓ Film appears dense, thickness undetectable in SEM
  - ✓ Irregularly shaped blocks or flakes
- Mk-16b (Al-6061 or Al-6063)
  - ✓ In-reactor ~220 d at ≥34 ° C; ~40 y wet storage (~22° C)
  - ✓ XRD: bayerite (P), boehmite (S), and gibbsite (T)
  - ✓ Film appears porous, 5-15 μm thick
  - ✓ Irregularly shaped blocks or flakes

(P): primary, (S) secondary, (T) tertiary

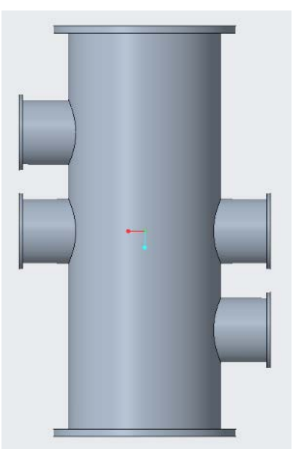


ASNF Hydrated Oxide Layer Response to Drying

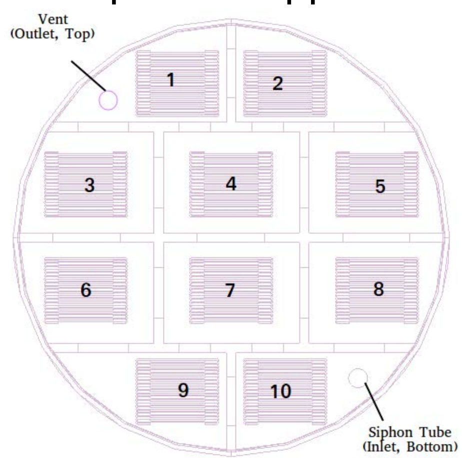
- Objective to assess effectiveness of drying processes at elevated temperatures (>100^0 C)
  - ✓ Vacuum drying
  - ✓ Forced gas dehydration
- Engineering scale test
  - ✓ 1/3 height DOE Standard Canister
  - ✓ Mock ATR elements
  - ✓ 10 elements/test
  - ✓ 3 oxide treated plates/element & 4 treated elements/test
  - ✓ Heated elements
  - ✓ CFD based model developed to support



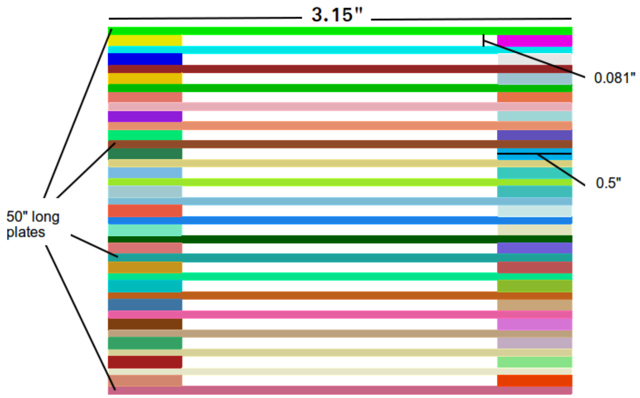
Mock treated ATR element plate



Main test chamber w/view ports



Chamber with Type 1A basket



Mock ATR element

Summary and Path Forward

- ASNF long-term dry storage knowledge and technical gaps have been identified
- A multi-year program has been implemented to address identified gaps
  - ✓ Needed preliminary information and data has been generated
  - ✓ An initial multi-physics model has been developed to predict dry storage system performance
- Follow-on studies are planned
  - ✓ Hydrated oxide radiolysis experiments
  - ✓ Hydrated oxide chemistry and reactivity experiments
  - ✓ Complete characterization of dry stored ATR SNF
  - ✓ Complete engineering scale ASNF drying studies
  - ✓ Update and refine dry storage system model and dry storage system performance simulations

Questions?

Idaho National Laboratory

Backup Slides

Program Reports

Aluminum Clad Spent Nuclear Fuel Long Term Dry Storage Technical Issues Action Plan – Technical and Engineering Activities, INL-EXT-17-43908, November 2017, M. Connolly.

Aluminum Clad Spent Nuclear Fuel Long Term Dry Storage Technical Issues Project Execution Plan, PLN-5596, May 2018, M. Connolly and J. Jarrell.

Transient Coupled Chemical-Thermal-Fluid Field Simulation for Sealed Aluminum-clad Spent Nuclear Fuel Storage Canister, INL/EXT-18-51683, Rev. 0, June 2018, A. Abboud and H. Huang.

Aluminum Clad Spent Nuclear Fuel Task 1: Oxyhydroxide Layer Behavior and Chemistry Experimental Test Plan, INL/EXT-18-45857, July 2018, T. Lister.

Aluminum Clad Spent Nuclear Fuel Task 2: Oxide Layer Radiolytic Gas Generation Resolution Experiment Test Plan, INL/EXT-18-45858, July 2018, P. Zalupski.

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Aluminum Clad Spent Nuclear Fuel Task 4: Performance of Aluminum SNF in Dry Storage Experiment Test Plan, INL/EXT-18-45861, July 2018, P. Winston.

Aluminum Clad Spent Nuclear Fuel Task 6: Surrogate Sample Preparation and Validation Experiment Test Plan, INL/EXT-18-45864, July 2018, M. Connolly.

Preparation of Aluminum Oxide Films Under Water Exposure – Preliminary Report on 1100 Series Alloys, SRNL-STI-2018-00427, Rev. 0, August 2018, K. Metzger, R. Fuentes, A. d'Entremont, L. Olson, R. Sindelar.

Preparation of Aluminum Oxide Films Under Water Exposure – Preliminary Report on 6061 Series Alloys, SRNL-STI-2018-00449, Rev. 0, August 2018, A. d'Entremont, R. Fuentes, L. Olson, R. Sindelar.

Program Reports (cont)

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Development of Transient Coupled Chemical-Thermal-Fluid Multiphysics Simulation for Unsealed, Vented Aluminum-clad Spent Nuclear Fuel Storage Canister, INL/EXT-18-51681, Rev. 0, September 2018, A. Abboud and H. Huang.

Characterization of Oxyhydroxides on a Dry-Stored Fuel Plate From L-Basin, SRNL-STI-2018-00428, Rev. 0, October 2018, L. Olson, R. Fuentes, A. d'Entremont, R. Sindelar.

Preparation of Aluminum Oxide Films Under Water Exposure – Preliminary Report on 5052 Series Alloys, SRNL-STI-2018-00646, Rev. 0, November 2018, A. d'Entremont, R. Fuentes, L. Olson, R. Sindelar.

Modeling activities concerning aluminum spent nuclear fuel cladding integrity, INL/EXT-18-51694, December 2018, M. Glazoff and T. Lister

Vapor Phase Corrosion Testing of Pretreated AA1100, INL/EXT-18-52249, December 2018, T. Lister.

Sensitivity Study of Coupled Chemical-CFD Simulations for Sealed and Unsealed Aluminum-clad Spent Nuclear Fuel Storage Canisters, INL/EXT-19-52650, January 2019, A. Abboud and H. Huang.

Radiation-Induced Changes in Corrosion of AA1100, INL/EXT-19-52738, Rev. 1., February 2019, E. Parker-Quaife, G. Horne, C. Heathman, P. Zalupski.

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