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Spent Nuclear Fuel Technology Development Activities at the Idaho National Laboratory

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Overview—Presentation Content

- **Spent nuclear fuel (SNF) technology development (TD) at the Idaho National Laboratory (INL):**
	- − Goals and objectives
	- − Scope

 • **Research on extended dry storage of aluminum-clad spent nuclear fuel (ASNF):**

- − Fiscal Year 2022 (FY-22) Technical Basis for Extended Dry Storage of ASNF
- − Status and ongoing ASNF dry storage TD activities
- − Impact on U.S. Department of Energy's (DOE's) ability to store ASNF
- **Addressing Nuclear Waste Technical Review Board (NWTRB) 2017 recommendations**
	- Advanced neutron absorber

Program Goals and Objectives

- **Program goals: Perform TD for the National Spent** Nuclear Fuel Program to **address issues related to storing, transporting, processing, and disposing of DOE-owned and managed SNF**
- **Program objectives:**
	- − Defined each FY under consideration of input from DOE, DOE Spent Nuclear Fuel Working Group (SNFWG), NWTRB, Independent Technical Reviews (ITRs), and other subject matter expert input
	- − **Provide support to DOE**, INL, Idaho Site, Savannah River National Laboratory (SRNL), Savannah River Site (SRS), and other national laboratories and sites in their SNF management responsibilities
- **Led by INL** and executed in close collaboration with SRNL

Various DOE-managed SNF types [1].

Key Input to Program and Previous Interactions

• **DOE:**

- − 2017 SNFWG report [2]: Five technical gaps on extended dry storage of ASNF
- − SNFWG meetings
- − DOE Office of Environmental Management (EM) Technology Operations (TO) and Nuclear Materials (NM) planning and coordination meetings

• **NWTRB**

- − Applicable recommendations of 2017 NWTRB report [1], e.g.:
	- Measuring and monitoring conditions in existing and new storage systems
		- DOE standard canister lid and remote canister monitoring system (RCMS)
	- Impact on residual water on SNF storability, specifically ASNF
	- Research and development for the DOE standard canister and its contents, such as remote welding and weld inspection TD, treatment of sodium-bonded SNF, and compliance with DOE standard canister design specifications
- **Participation in meetings**

Program Scope

Extended Dry Storage of ASNF—Technical Challenges

DOE-EM ASNF inventory:

- − Nationwide: ~13 metric tons of heavy metal (MTHM)
- − Idaho Site: ~3 MTHM (dry)
- − SRS L Area: ~6.3 MTHM (wet)
- − Hanford Site: ~3.4 MTHM (dry)
- − Low mass but 15 vol%
- **Packaging and extended dry storage:**
	- − Idaho Site Chemical Processing Plant (CPP)-603 and SRS L Area were built in 1950s
	- − Agreements with states, such as Idaho

Need for the development of a technical basis for extended dry storage of ASNF

Technical Basis for Extended Dry Storage of ASNF— Strategy for Addressing Identified Technical Challenges

PIE: Post irradiation examination

Technical Basis for Extended Dry Storage of ASNF—Key Activities and Findings

Task 1—(Oxy)hydroxide Behavior Evaluations

• Activities:

- − Aluminum alloy microstructure changes at elevated temperatures
- Water vapor corrosion experiments

• Findings:

- − High-temperature drying could lead to alloy phase changes
- No hydrogen is expected to be produced due to corrosion under representative dry storage conditions Test chamber water vapor corrosion of aluminum samples [9].

Task 2—Radiolytic Gas Generation Experiments

- Activities:
	- Radiolytic gas generation experiments (hydrogen H_2 , oxygen O_2)
	- Effects of different backfill gas types (air, argon, helium)
	- Supported by SRNL mini-canister testing

• Findings:

- Definition of *G*-values, describing the quantity of H₂ generation formed per unit of deposited energy
- Insignificant quantities of $O₂$ is generated
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Technical Basis for Extended Dry Storage of ASNF—Key Activities and Findings

Task 3—ASNF Dry Storage M&S

Activities:

- Thermal simulations of ASNF dry storage configurations
- Chemical simulations for storage periods of 50 years and more

• Findings:

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- Insignificant oxygen generation (<1 ppm) prevents sealed canister flammability (H₂ < 17%)
- Oxygen inhibits hydrogen generation in vented systems
- − Canister pressurization of no concern to integrity (<2.6 atm)
- 1nsignificant generation of nitric acid (<5,000 ppm) M&S of H₂ generated during 50 years of

Task 4—ASNF Dry Storage Performance Evaluations

- Activities:
	- − Characterization of actual ATR ASNF
	- − Comparison of ATR materials stored wet and dry for up to 22 years
	- − Analysis of chemical composition, surface morphology, and chemisorbed water quantity
- Findings:
	- No significant changes during multiyear wet or dry storage
	- No evidence for significant post-operational oxide growth on ASNF
	- − Occasionally found oxide plumes caused by abrasion, but remained stable during storage Scanning electron microscopy of an ATR
Cocasionally found oxide plumes caused by abrasion, but remained stable during storage (exv)

Advanced Test Reactor (ATR) dry storage [12].

⁽oxy)hydroxide layer cross section [13].

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Technical Basis for Extended Dry Storage of ASNF—Key Activities and Findings

Task 5—ASNF Drying Experiments

- Activities:
	- − Vacuum drying and forced helium dehydration
	- − ATR elements in mock canister
	- Thermogravimetric analyses to evaluate procedure effectiveness
- Findings:

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- Highly effective processes for removal of free water
- − Chemisorbed water removal during forced helium dehydration >220°C
- $\frac{1}{2}$ Significant reduction of H₂ generation due to drying Drying Drying experiment setup [4].

Task 6—Preparation of ASNF Surrogates

- Activities:
	- Development of corrosion protocols to reproduce (oxy)hydroxides on aluminum alloys
	- − Characterization of morphologies, thicknesses, and chemical compositions
	- − Comparison with actual dry- and wet-stored ASNF (SRS L Area)
- Findings:
	- Surrogate can effectively simulate the types of phases and morphologies of actual ASNF
	- − (Oxy)hydroxides appear in mixed monohydrate and trihydrate phases
	- − Passivation and protection by existing oxide layers is expected Scanning electron microscopy: Missouri University
Passivation and protection by existing oxide layers is expected Reaction Busing electron Reactor ASNF [14

Relative mass loss of dried samples [4].

Research Reactor ASNF [14].

A Technical Basis for Extended Dry Storage of ASNF

Task 2 FY-24 Continuation—Radiolytic Gas Generation and Corrosion Layer Behavior

• **Problem statement:**

- − Irradiation of corroded ASNF and backfill atmosphere promotes the formation of radiolysis products such as molecular hydrogen gas (H₂) and nitric acid (HNO₃)
- − Development of a quantitative understanding of the various parameters that impact the radiolytic formation and consumption of H_2 and $HNO₃$
- **Near-term objectives:**
	- − Evaluate the impact of breakaway corrosion phenomena on radiolytic gas generation, specifically H_2 and $HNO₃$
	- Determine the extent of radiation-induced $HNO₃$ production under dry storage conditions
	- − V&V of predictive models
	- − Address preliminary ITR recommendations
- **Expected outcomes:**
	- − Continue to deepen the understanding of radiation-induced phenomena
	- − Essential for V&V of predictive models and to strengthen the technical basis

Aluminum alloy 1100 coupon corrosion (A and B) and a gamma-irradiated flame-sealed 6061 coupon in the presence of water and an air atmosphere (C).

Task 2 FY-24 Continuation—Radiolytic Gas Generation and Corrosion Layer Behavior: Preliminary Results

- **Hypothesis:** Previous program work established that the radiation-induced **yield of chemical species, such as HNO3, H2, and O2, do not raise any canister corrosion, overpressurization, or flammability concerns.** However, **HNO₃** claims were model predictions only.
- **Preliminary findings:**
	- − The radiolytic yield of HNO₃ increases with absorbed gamma radiation dose in sealed-air systems.
	- The radiolytic yield of $HNO₃$ also increases with increasing relative humidity (relative humidity (RH) = -0% , -50% , and >95%) in sealed-air systems.
	- − The formation of HNO₃ is strongly affected by the amount of available water. This is indicated by the detection and trends of other NOx species when water depletes.
	- In the presence of surrogate aluminum-cladding coupons the radiolytic yield of $HNO₃$ in the gas phase is lower.

Yields of $NO₃$ and $NO₂$ generated from the cobalt-60 gamma irradiation of sealed-air under 0% RH in the presence and absence of a corroded aluminum alloy 6061 coupon corrosion. Data acquired by ion chromatography. *Inset:* Fourier transform infrared spectroscopy results highlighting speciation and absorbed dose trends in detectable **NOx** species in irradiated air sealed at ~95% RH.

Task 3 FY-24 Continuation—M&S of ASNF Dry Storage Canisters

• **Problem statement:**

- − The gas space composition, temperature, and pressure evolution within ASNF dry storage canisters need to be well understood
- **Near-term objectives:**
	- − Refine three-dimensional multiphysics computational fluid dynamics (CFD) simulations coupled with a chemical model to predict chemical species and pressure buildup
	- − V&V of computational models through comparison with experimental data collected from the ASNF Dry Storage Pilot
	- Enhancing the safety basis of the ASNF Dry Storage Pilot and ASNF dry storage applications in general
- **Expected outcomes:**
	- − Well-supported modeling approach for ASNF-in-canister behavior, including pressure, temperature, and gas space composition evolution predictions
	- − Implementation of verified chemical model for gas-phase and surfacemediated radiolysis reactions

Development of $H₂$ concentration in DOE standard canister during 200 years of sealed dry storage [17].

Task 5 and Instrumented Lids TD FY-24 Continuation— Mini-Canister Testing

• **Problem statement:**

- − There is a need for additional radiolysis data for higher accumulated doses to support predictions of ASNF dry storage behavior
- − ITR recommendations need to be addressed, such as the development of an improved understanding of ASNF corrosion processes and the impacts of conditioning and radiation
- **Near-term objectives:**
	- − Measure H2 yields via periodic gas sampling for doses up to 15 Megagray (Mgy)
	- − Testing of impacts of various canister conditioning processes
	- − Comprehensive reevaluation of available data and models
- **Expected outcomes:**
	- − Support model on H₂ yields from surrogates and actual ASNF material
	- − Address questions on corrosion risks during dry storage and define hydrogen yield measures (i.e., *G*-value) specific to ASNF dry storage

 $H₂$ vs. absorbed gamma dose [18].

Instrumented Lids FY-24 Continuation—CPP-603 RCMS

- **Problem statement:**
	- − The feasibility of extended ASNF dry storage in existing configurations needs to be confirmed
	- − For repackaging purposes, an in-depth understanding of in-canister conditions in existing ASNF dry storage configurations is needed
- **Near-term objectives:**
	- − Perform wireless transmission testing in CPP-603 fuel-handling cave
	- − Complete preliminary and final design of RCMS
	- − Collect temperature, relative humidity, pressure, radiation, and hydrogen concentration data
- **Expected outcomes:**
	- − Strengthen the technical basis for extended dry storage of ASNF
	- − Demonstrate remote-sensing capability adaptable to other applications and DOE-EMmanaged SNF types

Canister lid system in 3D-printed mock-up enclosure [19].

In-canister sensor mechanical assembly [19].

ASNF Dry Storage Pilot FY-24 Continuation

- **Problem statement:**
	- − V&V of M&S capabilities is required
- **Objectives:**
	- − Loading and monitoring of High-Flux Isotope Reactor (HFIR) ASNF dry storage utilizing best practice conditioning
- **Expected outcome:**
- For Strengthening the technical basis for extended dry storage of ASNF through confirmation of predicted behavior

Impact of Current Activities on the Technical Basis for Extended Dry Storage of ASNF

Support the defensibility of the technical basis for extended dry storage of ASNF:

- The continuation of ASNF experimental and M&S work will enable the INL/SRNL project team to **address ITR feedback**.
- The continuation of **subject matter expertise development** in this space, including a deep understanding of **one-of-akind chemistry models** of a broad range of ASNF dry storage configurations. This expertise is needed for evaluations of various ASNF dry storage strategies currently under consideration.
- The development of the instrumented lids will enable **V&V of the technical basis for extended dry storage of ASNF** and enable DOE-EM to collect nonexistent data from ASNF dry storage canisters that are in DOE-EM's custody and located in aging facilities.
- The ASNF Dry Storage Pilot will complete **one-of-a-kind sealed ASNF dry storage data collection**
- The ASNF Dry Storage Pilot will enable **risk mitigation** through the exploration of alternative ASNF disposition technologies and approaches. V&V of the technical basis will further the **development of actionable alternatives to the dissolution of ASNF** after the expected upcoming shutdown of SRS H Canyon.
- The ASNF Dry Storage Pilot will be used to evaluate placing **HFIR ASNF into dry storage** without utilizing SRS L Basin. This will support DOE in completing **planning of maintenance and decommissioning of aging facilities** such as L Basin.

Summary of Activities Focused on Enabling Extended Dry Storage of ASNF

• **DOE-EM-managed ASNF:**

- − Continues to manage ~13 MTHM of ASNF
- − Sources for additional ASNF generation:
	- ATR at the INL—Study radiation effects on reactor materials and fuels
	- HFIR at the Oak Ridge National Laboratory —Used to study neutron effects on materials, and to produce isotopes for medical, industrial, and research purposes
	- Domestic and foreign research reactors at universities and other research entities
- A technical basis for extended dry storage of ASNF is needed to enable DOE to package and store ASNF for **extended periods of time:**
	- − Will provide DOE with an alternative to dissolution of ASNF
	- − 1995 Idaho Settlement Agreement (ISA) includes the requirement to remove ASNF from the Idaho Site by 2035
	- − Reduces risk of not being able to package SNF in the short term or meeting the ISA 2035 deadline, which would expose DOE to financial fines
- **Current activities focus on V&V and strengthening of the technical basis**

Neutron Absorber Evaluation—Purpose

• **Problem statement:**

− Evaluations conducted for the Yucca Mountain Project license application required neutron absorbers for criticality control for some configurations of DOE-managed SNF

• **Approach:**

- − Reduce the probability of criticality for the relevant period of performance per 10 CFR Part 63
- − A new absorber material was proposed for use in DOE standard canisters: Advanced neutron absorber (ANA) made of Ni, Cr, Mo, and Gd
	- Initial corrosion tests showed poor corrosion tests for borated stainless steel
	- Gd was determined to be less soluble than B [21]
	- The American Society for Testing Materials (ASTM) accepted the ANA as the ASTM standard B 932-04 (UNS N06464 \rightarrow pictured right) [22]
	- In 2005 the ANA was approved by the American Society of Mechanical Engineers (ASME) for ASME Section III, Division 3 applications as Code Case N-728 [23]

Neutron Absorber Evaluation—Continued

• **Superiority of ANA challenged:**

− Other fabrication techniques for borated stainless steel (304B4, 304B5) showed better corrosion performance (in specific environments)

• **Supporting analyses and decisions:**

- − It was proposed that borated stainless steel be used for the Transportation, Aging, and Disposal (TAD) canister for commercial SNF
- − A study conducted in 2011 determined 304B4 borated stainless steel was less corrosive than ANA in limited testing environments [24]
- − An analysis concluded the criticality control ability of ANA did not exceed that of 304B4 borated stainless steel [25]

Intact (left) and degraded (right) ATR fuel in an intact DOE standard canister surrounded by intact high-level waste canisters [25].

Recent Neutron Absorber Work

- **Borated stainless steel (304B4) and boron carbide (B4C) were used in criticality evaluations [26]:**
	- − Some ATR elements are stored in ATR4 buckets at INL in dry storage (CPP-603)
	- − Throughput could be reduced with alternative packaging options
	- − With additional neutron absorber inserts all scenarios had a k-effective less than 0.93
- **Additional analyses were conducted evaluating the susceptibility of ANA to seawater corrosion [27]:**
	- − Results showed that the Ni5Gd second phase, distributed along face-centered cubic grain boundaries in Ni-Cr-Mo-Gd, has lower corrosion potential than the substrate
	- − Isolating the Ni5Gd from the corrosion medium through advanced manufacturing is a viable way to improve the ANA corrosion resistance

ATR4 bucket configurations in DOE standard canister [26].

- The DOE-EM SNF TD Program continues to **perform TD for the National Spent Nuclear Fuel Program to address issues related to storing, transporting, processing, and disposing of DOE-owned and managed SNF**
- A **technical basis** for extended dry storage of ASNF was developed
	- − This basis is needed to enable DOE to package and store ASNF for extended periods of time
	- − Currently efforts focus on strengthening the technical basis
	- − An ASNF Dry Storage Pilot will be part of the V&V efforts
- DOE and the DOE-EM SNF TD Program continue to work toward **addressing NWTRB recommendations**, such as research on ANA

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