The Nuclear Fuel Cycle: Safe Management of Spent Nuclear Fuel

R.L. Sindelar

Laboratory Fellow, Savannah River National Laboratory, Aiken, SC

The aim for storage of spent nuclear fuel (SNF) either in wet or in dry storage systems is to ensure general safety objectives are met throughout a desired storage period. Staff at the Savannah River National Laboratory (SRNL), in collaborations with partners at other national laboratories, industry research organizations, and the University of South Carolina (UofSC), have performed materials aging testing and analyses, and have established nuclear materials aging management programs to support extended periods of safe storage of research reactor (RR) SNF and of commercial power reactor (PR) SNF pending ultimate disposal. Several example challenges include susceptibility of aluminum SNF from research reactors to corrosion in poor quality water (wet storage). In dry storage, aluminum SNF can release hydrogen via radiolysis of the hydrated oxides on the aluminum cladding. Austenitic stainless steel canisters used for dry storage are susceptible to chloride-induced stress corrosion cracking (outside-in attack) that threaten the confinement boundary provided by the canister. This paper further describes these challenges, among others, and the formulated solutions to support extended safe storage of SNF.

Introduction

The nuclear fuel cycle for fission reactors spans the set of functions and processes from the initial mining of uranium through to the permanent disposal of the spent fuel itself, or, if reprocessed to recover useful species, a waste form for disposal in a repository (see Figure 1). An important stage in the back end of this cycle is the storage of the spent nuclear fuel (SNF) which can be in a wet storage system (e.g., a pool or water basin) or in a dry storage system (e.g., stainless steel canisters with radiation shielding overpacks on a concrete pad outside).

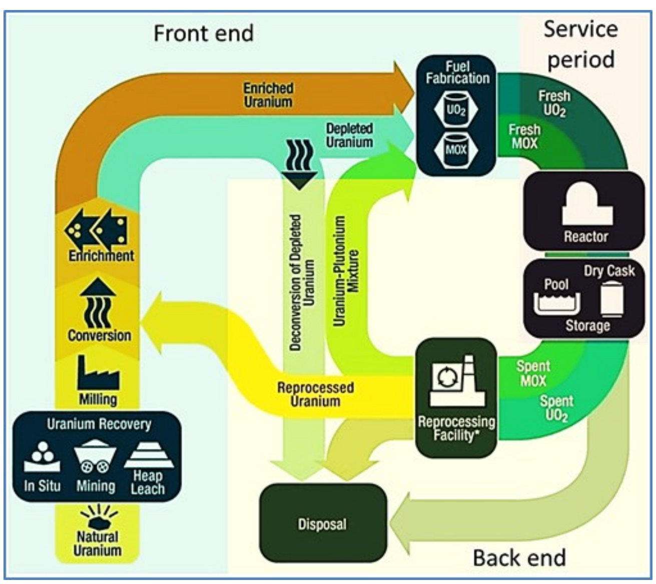

Figure 1. Stages in a Generic Nuclear Fuel Cycle for a PR with Reprocessing [courtesy of U.S. Nuclear Regulatory Commission]

Figure 2 shows examples of commercial power reactor (PR) fuel and research reactor (RR) fuel and their wet and dry storage systems. Following reactor service, the SNF is discharged to a pool for cooling, for both power reactors (PR) and research reactors (RR). The SNF can remain in pool storage for additional cooling and shielding of the attendant decay radiation. In the U.S., PR fuel was originally intended to be stored in the water pool several years prior to reprocessing. However, the U.S. has followed a "once-through" (i.e., no reprocessing) nuclear fuel cycle strategy since the late 1970's, and this has led to interim storage in pools at full capacity or soon-to-be full pools at most of the U.S. reactor sites, and in dry storage systems owned by utilities and licensed by the U.S. Nuclear Regulatory Commission (NRC). At present, over 80,000 metric tonnes of heavy metal (MTHM) of SNF have been discharged from PR in the U.S., of which nearly half is stored in dual-purpose canisters[1] (DPC) made of stainless-steel numbering over 3000 DPCs [1].

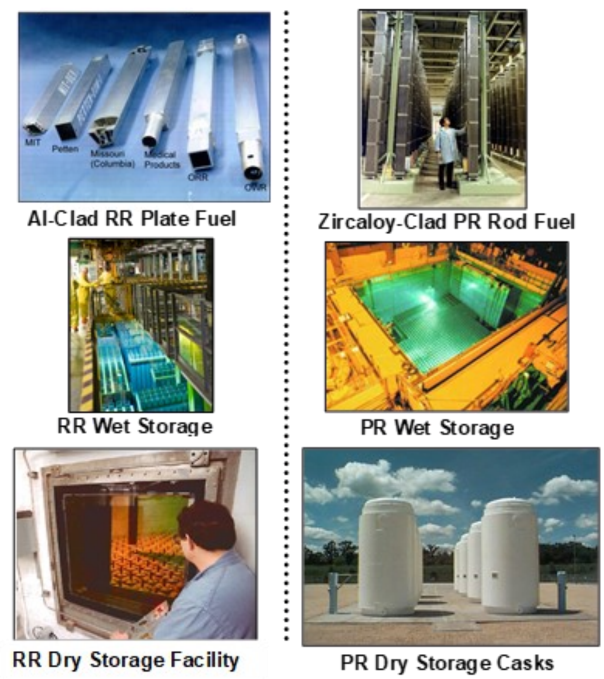

Figure 2. Research Reactor (RR) and Power Reactor (PR) Fuel in Storage Systems

The U.S. Department of Energy - Environmental Management (DOE-EM) office is responsible for the receipt and storage pending disposition of spent nuclear fuel that was used in research and test reactors worldwide that contains U.S. origin and certain non-U.S. origin enriched uranium. This foreign and domestic RR fuel is primarily aluminum-based, aluminum-clad fuel that is being stored in L Basin, a water basin at the Savannah River Site (SRS) in South Carolina. Additional aluminum fuel, the majority of which originated from the Advanced Test Reactor, is in dry storage in the Irradiated Fuel Storage Facility (IFSF) at the Idaho Nuclear Technology and Engineering Center (INTEC) at the Idaho National Laboratory (INL).

The lack of a federal geologic repository as the ultimate end state for the SNF from PR and RR has led to de facto extended storage with no certain end date.

[1] The DPC would be multi-purpose for storage, transportation, and disposal

Common to both these reactor categories of fuel for both types (wet and dry) of storage systems is the aim to maintain general safety objectives throughout the storage period. The safety objectives are met through the systems, structures, and components (SSCs) in the storage systems, whose designs are predicated on the performance of the fuel in the storage environment.

The general safety objectives listed below have been derived from those prescribed by the International Atomic Energy Agency (IAEA) [2, 3] and the U.S. NRC requirements for dry storage systems as listed in its Standard Review Plan for Dry Storage Systems [4]:

- Maintain criticality safety
- Maintain cooling of the fuel
- Maintain radiation shielding
- Maintain confinement
- Maintain retrievability (on canister or fuel basis)

Specifications and limits (e.g., for fuel peak cladding temperature [4]) and aging management programs (e.g. in-service inspections) for the fuel and SSCs of the storage systems can be prescribed to ensure these safety objectives are met. The U.S. NRC regulates the wet and dry storage of PR SNF and grants operating licenses for the storage facilities.

This paper looks at selected aging materials challenges and solutions supporting the extended storage safe storage of RR and PR in wet and dry storage systems. The work of the Savannah River National Laboratory (SRNL), including with its partnerships, is emphasized.

Aluminum-Clad RR SNF – Extended Wet Storage

Aluminum is a common material for RR fuel cladding used worldwide, for RR fuel storage racks in basins, and in material systems for neutron absorbers in PR SNF storage pools. In poor quality water, aluminum is particularly susceptible to corrosion [5]. Control against corrosion attack can be achieved by demineralization and filtration systems to maintain low solids, low conductivity water, and within a range of pH to minimize aluminum solubility. Using experiences of corrosion in reactor systems, including the vast experience at the Savannah River Site (SRS), guidelines were prepared by the International Atomic Energy Agency [6] to prescribe limits for water quality parameters important to corrosion control in water systems for research reactors.

A broader interrogation of the set of systems, structures, and components used to meet the general safety objectives for extended fuel storage in the L Basin at SRS was performed in 2011 [7]. The overall conclusion was that the fuel can be stored in L Basin, meeting the general objectives for safe fuel storage for an additional 50 years and beyond, contingent upon continuation of existing fuel management activities and several augmented program activities. This work was cited by the Nuclear Waste Technical Review Board in its 2017 topical report on the Management and Disposal of U.S. Department of Energy (owned) Spend Nuclear Fuel [8].

The augmented program activities by SRNL and the Spent Fuel Project Engineering organization at SRS have been the development and deployment of non-destructive examination tools to perform enhanced remote visual inspection of the stored aluminum SNF [9] (see Figure 3), and of the containers storing the non-aluminum SNF in L Basin [10]. A primary conclusion from the inspection report of 10 assemblies of aluminum SNF verified that the fuel storage and management practices avoid corrosion and furthermore mitigate existing corrosion of the fuel [9]. Additional inspection of the containers for the non-aluminum SNF in L Basin is in progress [10].

A separate investigation on aluminum-based materials in reactor systems was the investigation on the corrosion performance of a Neutron Absorber Material (NAM) by SRNL for the U.S. Nuclear Regulatory Commission [11]. The NAM is attached to stainless steel racks and used for criticality control in PR spent fuel (storage) pools. The specific NAM is a composite sheet containing a cermet core of boron carbide (B_4C) particles from 35 to 65 wt.% in aluminum (Al 1100) powder, sandwiched between aluminum (Al100) sheets as shown in Figure 4.

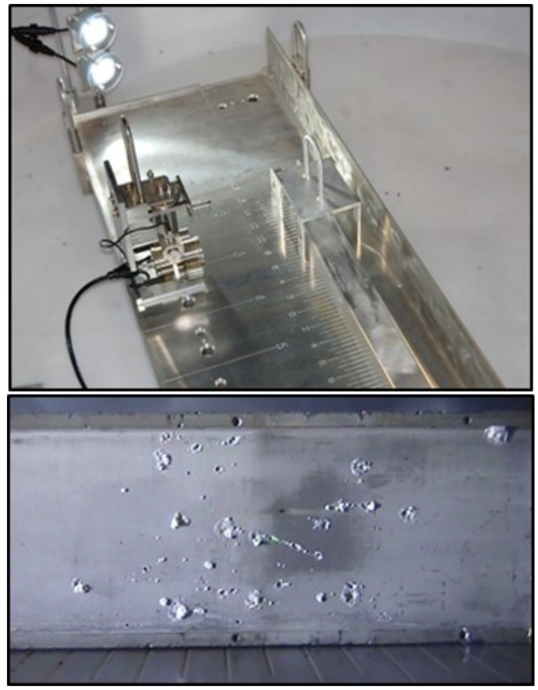

Figure 3. Fuel Inspection Table (top photograph) and an image of a plate-design aluminum SNF assembly following 25-years' storage in L Basin (bottom). The pitting corrosion damage occurred prior to fuel receipt at L Basin; no change to the fuel condition has occurred while in L Basin [9]

Figure 4. Laser Confocal Microscope montage image of a cross-section specimen from a Neutron Absorber Material (NAM) panel from the Zion Nuclear Power Plant Spent Fuel Pool following 22 years' of service [11]

Using standard electrochemical test methods, and immersion test methods with special specimen preparation techniques, the corrosion rates of the aluminum panel, and of the aluminum-based core materials were determined at temperature and water chemistry conditions. Arrhenius relationships were established to allow predictive modeling of corrosion performance at long-term nominal (typical water quality for spent fuel pool service) and at long-term base (spent fuel pool water quality limits) water chemistries' exposure, and at hypothetical transient (off-normal) water chemistry and temperature exposure conditions.

Maintaining neutron absorber efficacy of the NAM is required throughout service in the spent fuel pool. The results from the electrochemical corrosion testing showed that the controlled water chemistry conditions of typical Pressurized Water Reactor and Boiling Water Reactor Spent Fuel Pools would allow very long service periods for the NAM. In addition, the immersion testing showed a key result that the core material (if exposed by loss of aluminum sheet clad) would not corrode significantly to cause loss of the absorber material (B_4C particles) even under severe off-normal water chemistry and temperature conditions for up to well over a week of exposure [11].

Aluminum-Clad RR SNF – Extended Dry Storage

An extensive body of work had been done at the Idaho National Laboratory and the Savannah River National Laboratory in the late 1990s to develop the storage system and evaluate materials interactions for DOE SNF in a standard canister destined for imminent (at that time) repository disposal. A subset of this fuel, the highly-enriched aluminum SNF, posed particular challenges to extended dry storage (> 50 years), as highlighted in a 2017 report [12] by the DOE Spent Nuclear Fuel Working Group (SNFWG).

A primary challenge, unique to the aluminum-clad SNF vis-à-vis zircaloy or stainless-steel-clad SNF, is the presence of hydrated oxides of aluminum formed on the SNF cladding by virtue of irradiation and wet storage service. These hydrated oxides can release hydrogen gas under gamma radiation [12, 13]. Experiments and theory on hydrogen release from boehmite and bayerite, two hydrated aluminum oxides, was published by SRNL in 2014, demonstrating the phenomenon. Figure 5 displays the results of the computational chemistry theory modeling that indicates hydrogen production is possible with energy input (e.g., via gamma radiation) to the hydrated oxides.

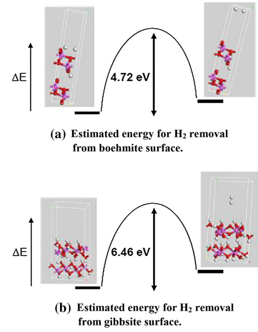

Figure 5. Energy barrier diagrams illustrating removal of a hydrogen molecule (H_2) from the hydrated oxides boehmite (a) and bayerite (b). [figures reproduced from reference 13]

A comprehensive set of activities to address the challenges from drying and dry storage of aluminum SNF and establish the technical bases for extended dry storage was sponsored by the DOE-EM, Office of Technology Development beginning in late 2017. The work, led by INL, and its partners of SRNL, UofSC and industry has been conducted over the last several years. The work included additional first-time radiolysis testing of hydrated oxides on aluminum substrates to build a library of G-values for hydrogen generation; modeling of SNF-in-canister system including the head-space gas evolution; and drying tests for the technologies of vacuum drying and of forced-gas dehydration. An example of the results of laboratory-scale testing work shows the profound reduction of radiolytic hydrogen by drying of specimens with hydrated oxides to promote removal of physisorbed waters and thermal decomposition of the hydrated oxides (see Figure 6).

The lab-scale work and the technical bases for the drying and dry storage of aluminum SNF have been largely completed - a status report summarizes the key findings of the program to date [14]. A primary conclusion is that sealed dry storage of aluminum SNF would not cause flammable gas mixtures to evolve, and that the pressure rating of the DOE Standard Canister for DOE RR SNF would not be exceeded under extended dry storage. Additional recipes for drying with subsequent radiolysis testing is in progress in FY22.

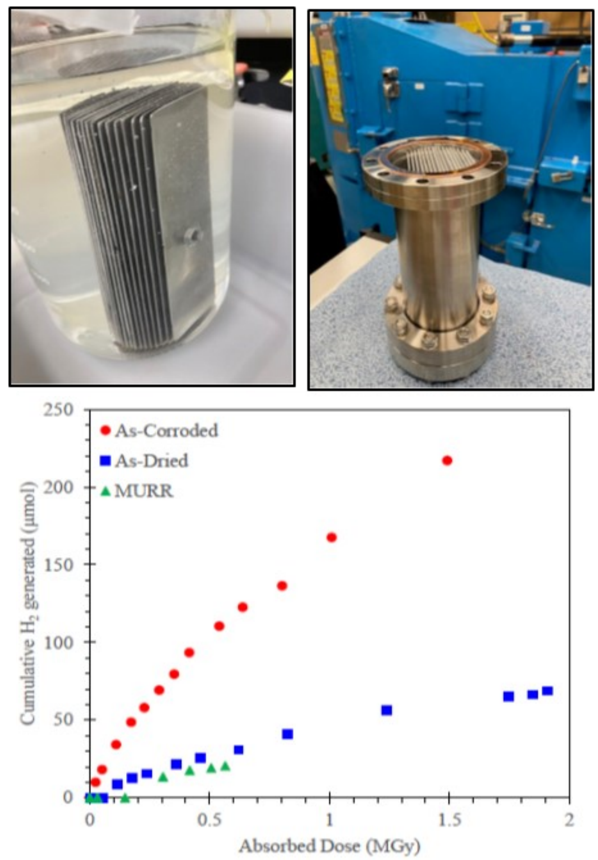

Figure 6. Aluminum plate specimens with hydrated oxides loaded in a "mini-canister" (top). Data of radiolytic hydrogen from the mini-canister specimens with gamma dose for conditions of vacuum drying ("As-Corroded") and heated air (220°C) ("As-Dried"), as well as results for a much smaller sample with no drying ("MURR") (bottom) [figures reproduced from reference 15].

Zircaloy-clad PR SNF – Extended Wet Storage

Stainless steel and zirconium alloys are more resistant to corrosion in water than aluminum. Zirconium cladding alloys are subject to general corrosion with an adherent passive oxide film of ZrO2 that grows with time in water at power reactor operating temperatures (e.g., 288°C bulk core water temperature for Pressurization Water Reactors). After discharge from the reactor and in pool storage, zirconium alloy and stainless-steel claddings are highly resistant to corrosion due to their robust passive oxide layers. There are typically no technical specifications or requirements for water chemistry to mitigate corrosion of zirconium alloys in pool storage due to the relatively innocuous environment of low temperature (nominal 40°C) water exposure vis-à-vis reactor operation temperature exposure. Water clean-up is required in pool storage to limit water radioactivity levels [6]; also keeping the conductivity and water temperature low in pool storage will minimize the intensity of electrochemical reactions [16].

New-build pools for PR SNF storage are not feasible given construction and operating & maintenance costs. Dry storage of PR SNF is the only cost-effective pathway for its extended storage pending repository disposal.

Zircaloy-clad PR SNF – Extended Dry Storage

Circa 2009, with the 20-year initial licenses of PR fuel storage facilities reaching expiration, and with the license application to the U.S. NRC for the federal repository (Yucca Mountain Project) being vacated by the U.S. government, the pathway to imminent permanent disposition of SNF (and also High Level Waste from the U.S. defense production mission) by repository disposal in the U.S. became uncertain, and the technical bases for extended safe storage and re-licensing of storage facilities was needed. This need was recognized by the broad nuclear power industry with stakeholders of the U.S. NRC, electric power utilities, Electric Power Research Institute (EPRI), SNF storage system vendors, DOE – Office of Nuclear Energy (NE), and nuclear system experts at the U.S. national laboratories.

A gross bounding estimate for the period of extended dry storage of PR SNF storage systems of "up to 300 years" was suggested by R.E. Einziger of the U.S. NRC at that time, and various organizations worked to identify "technical information gaps" in predicting aging materials performance over such very long storage periods. The U.S. NRC contracted SRNL to perform such a gap analysis, and the results were published in an NRC contractor report (CR) [17].

Much work to provide the bases for extended dry storage (for at least up to 40-year re-license being granted by the U.S. NRC) has been completed. The remaining ongoing investigations including materials testing and analyses, and development and deployment of tooling and systems for inservice inspection are under work sponsored by EPRI, the NRC, and DOE-NE in addition to work by the storage system vendors. A common bi-annual meeting forum, the Extended Storage Collaboration Program is hosted by EPRI to share the information being generated.

One materials' aging challenge with key input provided by the SRNL staff involves the phenomenon of Chloride-Induced Stress Corrosion Cracking (CISCC). The CISCC phenomenon has a potential to occur at SNF canister weldments as this location has weld residual stresses. Deposits of chloride-containing marine salts and industry dust may build up during outside storage, and with cooling of the canister surface with time, the salts can deliquesce and form a brine to provide the requisite conditions for CISCC. The growth of cracks could penetrate the nominal 5/8" thick steel shell of the canister to cause a loss in the confinement boundary of the canister.

Two aspects of this challenge include flaw mechanical stability against loadings to avoid a large rupture of the canister, and the crack growth rate (CGR) of CISCC cracks, should they form. The mechanical stability of flaws in a canister subject to in-service loads and weld residual stresses was rigorously analyzed by SRNL [18]. Various locations and sizes of flaw postulates including throughwall flaws (assumed a breach of the canister) were considered. As shown in Figure 7, the size of the most limiting location flaw (perpendicular to a circumferential weldment) and under the accident loading conditions and with weld residual stress, was over several inches.

The second aspect, the CGR prediction, is an important input for flaw disposition, should flaws be detected in a canister (none have to date of this paper). A team of staff from Sandia National Laboratories, Dominion Engineering, Inc., and SRNL, defined a temperature-based CGR that may be used to project the future growth of a flaw [19]. This work was an important part of the technical bases for an ASME code case for inservice inspection of the canisters [20, 21]. The code case is now available for utilities to adopt into their specific aging management programs for their fuel storage installations to allow their re-licensing and continued operation.

There has been additional important work by SRNL staff for safe dry storage of PR SNF. In 2014, the staff organized a workshop of an expert team to evaluate the impact of hydriding on the zircaloy-clad SNF for storage and transportation [22]. An international consensus code for drying SNF [23], shepherded by SRNL, was recently published by ASTM International to provide guidance for drying the SNF in the first place!

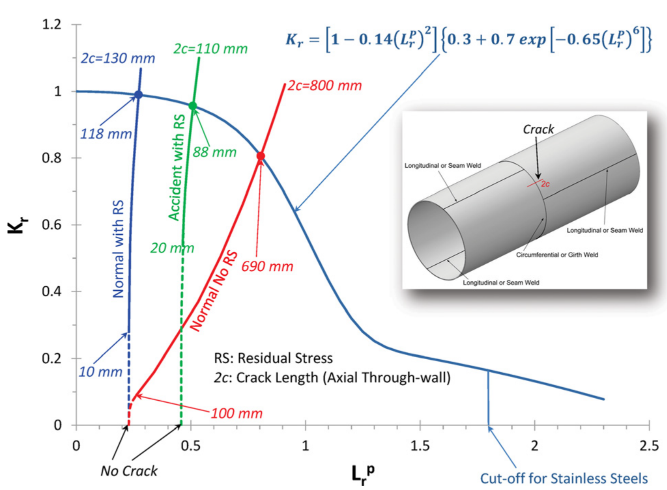

Figure 7 - Failure Assessment Diagram evaluation for flaw stability in a SNF canister showing long flaws remain stable even under bound accident conditions including weld residual stress (RS) [reproduced from reference 18]

Conclusions

The approach to enable safe extended storage of SNF in wet or in dry storage systems involves materials' aging testing and analyses and establishment of aging management programs (e.g. inservice inspection programs) to provide the technical underpinnings and support the demonstration that general safety objectives for storage are met throughout the desired storage period in consideration of materials aging. Several challenges for extended storage of RR and PR SNF were described as examples of work performed by materials scientists and engineers to provide solutions to meet these challenges. The staff at the Savannah River National Laboratory have had prominent roles in this work sponsored by the U.S. Department of Energy and U.S. Nuclear Regulatory Commission.

The practical experience and success in providing solutions for nuclear systems positions SRNL well to solve future challenges.

Acknowledgements

This work was produced by Battelle Savannah River Alliance, LLC under Contract No. 89303321CEM000080 with the U.S. Department of Energy. Publisher acknowledges the U.S. Government license to provide public access under the DOE Public Access Plan (http://energy.gov/downloads/doe-public-access-plan).

References

1. Peters, S., Vinson, D., and Carter, J.T., "Spent Nuclear Fuel and Reprocessing Waste Inventory," FCRD-NFST-2013-000263. Rev. 8, U.S. Department of Energy, Office of Nuclear Energy, Spent Fuel and Waste Disposition, November 2021.
2. IAEA Safety Standard Series, Safety Guide No. NS-G-4.3, "Core Management and Fuel Handling for Research Reactors."
3. IAEA Safety Standards, Specific Safety Guide No. SSG-63, "Design of Fuel Handling and Storage Systems for Nuclear Power Plants."
4. NUREG 2215, Standard Review Plan for Spent Fuel Dry Storage Systems and Facilities – Final Report, U.S. Nuclear Regulatory Commission, ADAMS accession number ML20121A190.
5. Sindelar, R.L., Chandler, G.T., and Mickalonis, J.I., "Water Quality and Corrosion: Considerations for Nuclear Reactor Systems," Journal of the South Carolina Academy of Science, Vol. 9, No.1, ISSN 1553-5975, Spring 2011.
6. Sindelar, R.L. (USA-SRNL), R. Ramanathan (Brazil-IPEN), R. Haddad (Argentina-CNEN), F. Bakker (Netherlands - PETTEN), "Good Practices for Water Quality Management in Research Reactors and Spent Fuel Storage Facilities," IAEA Nuclear Energy Series, No. NP-T-5.2, 2011.
7. Sindelar, R.L. and Deible, R.W., "Demonstration of Long-Term Storage Capability for Spent Nuclear Fuel in L Basin," SRNL-STI-2011-00190, April 2011.
8. "Management and Disposal of U.S. Department of Energy Spent Nuclear Fuel," a Report to the United States Congress and the Secretary of Energy, U.S. Nuclear Waste Technical Review Board, December 2017.
9. Hromyak, M., Sindelar, R., Verst, C., Boerstler, J., and Colebeck, A., "Augmented Monitoring and Condition Assessment Program (AMCAP) Material Test Reactor (MTR) Fuel Inspection Program Report," SRNL-STI-2020-00152, May 2020.
10. Ward, L.N., Trimm, M., Verst, C., and Boland, S., "Visual Examination of Aluminum Containers for Extended Wet Storage of Non-Aluminum-Clad Spent Nuclear Fuel – FY21," SRNL-STI-2021-00311, June 2021.
11. Fuentes, Roderick E., Kesterson, Ronald L., Verst, Christopher G., Sindelar, Robert L., and Focht, Eric. "Electrochemical Testing of Aluminum-Boron Carbide Cermet Panels from the Decommissioned Zion Nuclear Power Plant Spent Fuel Pool," CORROSION 2021, paper no. NACE-2021-16548, April 2021.
12. Lacroix, L.L., et al., "Aluminum-Clad Spent Nuclear Fuel: Technical Considerations and Challenges for Extended (>50 Years) Dry Storage," DOE-ID/RPT-1575, June 2017.
13. Westbrook, M.L, Sindelar, R.L. and Fisher, D.L., "Radiolytic hydrogen generation from aluminum oxyhydroxide solids: theory and experiment," Journal of Radioanalytical and Nuclear Chemistry, 2014, DOI 10.1007/s10967-014-3563-6.
14. Eidelpes, E. and Sindelar, R., "Technology Development for Aluminum-clad Spent Nuclear Fuel for Extended Dry Storage – Status Report FY21," INL/EXT-21-01417, September 2021.
15. d'Entremont, Anna, Verst, Christopher G., Randall, Benton, C., McNamara, Joy D., Duncan, Andrew J., and Sindelar, Robert L., "Radiolysis Testing of

SNF Materials and Surrogates in a Mini-Canister," Paper 22162 WM2022 Conference, March 2022 (to be published).

16. U.S. NUCLEAR REGULATORY COMMISSION, Regulatory Guide 1.13, "Spent Fuel Storage Facility Design Basis," Revision 2, March 2007.
17. Sindelar, R.L., Duncan, A.J., Dupont, M.E., Lam, P.-S., Louthan, Jr., M.R., and Skidmore, T.E., NUREG/CR-7116, SRNL-STI-2011-00005 "Materials Aging Issues and Aging Management for Extended Storage and Transportation of Spent Nuclear Fuel," U.S. Nuclear Regulatory Commission, November 2011
18. Lam, P.-S. and Sindelar, R.L., "Flaw Stability Considering Residual Stress for Aging Management of Spent Nuclear Fuel Multiple-Purpose Canisters," ASME Journal of Pressure Vessel Technology, August 2016, Vol. 138.
19. Broussard, J., Bryan, C., Sindelar, R., and Lam, P.-S. "Crack Growth Rate Model for CISCC of Stainless Steel Canisters," Paper Number PVP2019-94055, Proceedings of the ASME Pressure Vessels & Piping Conference, San Antonio, Texas, USA, July 2019.
20. Section XI ASME Boiler and Pressure Vessel Code Case N-860, 2019 ASME Code Cases: Nuclear Components, Supplement 6, Approved July 6, 2020
21. Broussard, J., "Technical Basis Summary for Code Case N-860: Inspection Requirements and Evaluation Standards for Spent Nuclear Fuel Storage and Transportation Containment Systems," Paper Number PVP2021-64176, Proceedings of the ASME Pressure Vessels & Piping Conference, Virtual Online, July 2021.
22. Sindelar, R.L., Louthan, Jr., M.R., and B.D. Hanson, "White Paper Summary of 2nd ASTM International Workshop on Hydrides in Zirconium Alloy Cladding," May 29, 2015, DOE-NE Fuel Cycle Research & Development, FCRD-UFD-2015-000533, SRNL-STI-2015-00256.
23. ASTM C1553-21, "Standard Guide for Drying of Spent Nuclear Fuel," ASTM International, August 2021.