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ABSTRACT

An assessment of the possible material interactions arising from the movement of previously wet stored spent nuclear fuel (SNF) into long-term dry interim storage has been conducted for selected fuels in the Idaho Chemical Processing Plant (ICPP). Three main classes of fuels are addressed: aluminum (Al) clad, stainless steel (SS) clad, and unclad Uranium-Zirconium Hydride ($UZrH_x$) fuel types. Degradation issues for the cladding, fuel matrix material, and storage canister in both wet and dry storage environments are assessed. Possible conditioning techniques to stabilize the fuel and optimum dry environment conditions during storage are also addressed.

I. INTRODUCTION

What are the issues and options for acceptable dry storage of Al-clad, SS-clad, and unclad $UZrH_x$ SNF currently in wet storage at the ICPP in the CPP-603 and CPP-666 wet storage basins? This is the central question being addressed by Pacific Northwest Laboratory (PNL) for the Westinghouse Idaho Nuclear Company, Inc. (WINCO). This assessment will assist in evaluating failure mechanisms for these SNFs to support the identification of vulnerabilities during their movement and eventual placement in an interim dry storage environment. These spent fuel types were selected for evaluation because of known corrosion in the CPP-603 for aluminum alloys (fuel elements and storage components) and potential degradation issues for SS-clad and $UZrH_x$ fuels. It is assumed that the present condition of the fuels will play a significant role in the eventual performance of these fuels in future long-term storage environments. Distinct issues for each spent fuel type are anticipated for either continued wet storage or future long-term dry storage.

II. BACKGROUND

This evaluation has focused on obtaining information from documentation of past fuel storage histories and conditions to assess the performance behavior of SNF in a dry storage environment. The SNF evaluated includes four types of fuel material: uranium-aluminum (UAl_x), uranium oxide (UO_2), uranium metal, and $UZrH_x$. The UAl_x fuel is generally clad in aluminum; the UO_2 fuel and U metal are clad in SS; the $UZrH_x$ fuel is clad in aluminum, SS, or has had the cladding removed. The three major contributors to ICPP's spent fuel inventory, based on the number of units stored, are from the Experimental Breeder Reactor II (EBR-II, ~55%), the Advanced Test Reactor (ATR, ~13%), and TRIGA spent fuel with aluminum cladding (TRIGA Al, ~9%). A list of all of the fuels evaluated in this assessment is provided in Table 1.

These spent fuels were placed in wet storage in anticipation of eventual reprocessing. With Idaho National Engineering Laboratory's (INEL) focus away from reprocessing SNF, these fuels have been left in storage far longer than was originally designed. Additionally, those fuels stored in the CPP-603 basin were subjected to excessive microbiological, chloride, and other contaminant levels during the 40-year history of the facility. With some of the fuel observed to be degrading (aluminum alloys), and the other fuels likely to be suffering from unobserved attack as well, an assessment of the possible degradation mechanisms most likely to occur in the wet environment was initiated. Very little as-stored data are available for the fuels and what is available is primarily qualitative. The results, conclusions, and recommendations presented below constitute a comprehensive assessment of the current known information on these fuels.

TABLE 1 Types of SNF Being Evaluated

Fuel	Abbreviation	Fuel Matrix/Cladding
Atomics International (a SNAP fuel)	AI	UZrH/declad
Army Package Power Reactor	APPR	UO_2 SS/SS
Advanced Reactivity Measurement Facility	ARMF	UAl/Al
Advanced Test Reactor	ATR	UAl/Al
Battelle Memorial Institute, Battelle Thermal Reactor	BMI	UO_2 SS/SS
Boiling Reactor Experiment V	BORAX V	UO_2 SS/SS
Experimental Breeder Reactor II	EBR II	U-5% Fissium/SS
EBR II Scrap	EBR II, ANL-6	U-5% Fissium/SS
Enrico Fermi Atomic Power Plant (SS clad only)	Fermi I Blanket	UMo/SS
High Flux Beam Reactor	HFBR	UAl Al cermet/Al U_3O_8 Al cermet/Al
University of Missouri Research Reactor	MURR	UAl/Al
Oak Ridge Reactor	ORR	UAl/Al
Pathfinder	Pathfinder	UO_2 SS cermet/SS
SM-1A Army Reactor	SM-1A	UO_2 SS/SS
Systems for Nuclear Auxiliary Power	SNAP	UZrH/declad
Organic Moderated Reactor Experiment	SPEC (OMRE)	UMo/SS
Special Power Excursion Reactor	SPSS (SPERT)	UO_2 SS/SS
Kansas State, Cornell, Texas, Hanford, MSU, and GA TRIGA SNF with Al cladding	TRIGA Al	UZrH/Al
Berliner Experimenter Reactor II	TRIGA Ber II	UZrH/SS
Neutron Radiography Reactor	TRIGA FLIP	UErZrH/SS
MSU, GA, and Berkeley TRIGA SNF w/SS cladding	TRIGA SST	UZrH/SS
University of Washington Argonaut Reactor	UW	UAl/Al
Vallecitos Boiling Water Reactor	VBWR (GENEVA)	UO_2 SS/SS

III RESULTS

Degradation mechanisms that appeared to be significant for the assessed fuel types were evaluated and ranked as shown in Table 2. For simplicity, the unclad $UZrH_x$ fuels are not shown in Table 2 because they were determined to be susceptible to only one degradation mechanism, crumbling. Because of the limited information on the as-stored condition for many of these fuels, the validity of these rankings and their absolute levels of importance will require verification by future work. The results of this initial assessment are summarized below for each of the major types of SNF evaluated.

A. Aluminum-Clad SNF

Three major issues exist for the Al-clad SNFs. The first issue is that the cladding is degrading (extensive pitting has been observed) in the CPP-603 environment and possibly in the CPP-666 pool,[a] based on visual examinations and open literature information.[1,2] Extensive pitting can lead to fuel matrix corrosion, fuel washout, and eventual weakening of the cladding material from excessive material loss. It has been shown from post-irradiation examinations of ATR plate-type fuel elements that the fuel can be physically removed from between the exterior plates (cladding) during reactor operation.[3] In addition, excessive pitting can eventually compromise the physical integrity of the fuel element structure, causing the fuel to lose configuration.

The formation/presence of UH_3 is thermodynamically possible for some of the fuel/environment combinations. This is of concern because of its low-temperature ignition and combustion that could be an issue in drying operations or subsequent dry storage, if sufficient quantities have formed. Previous studies have indicated that hydride formations as small as 5g can form "match heads" capable of igniting the surrounding uranium in the fuel matrix.[4] This reaction would not take place under normal conditions, but the hydrides provide enough thermal energy to start the reaction. Handling, conditioning, and dry storage strategies can be devised if UH_3 is detected or suspected.

Moisture contained in hydrated corrosion products could contribute to continued degradation and hydrogen buildup during storage if sufficient release occurs; however, the release cannot be presently quantified. The release would occur either by thermal or radiological decomposition of the corrosion products while the fuel is in a dry environment. One of the main concerns with this possible reaction is the formation and buildup of hydrogen gas resulting from decomposition of the corrosion product. Hydrogen gas may accumulate inside the storage container, making retrieval of the fuel difficult. However, because the amount of liberated moisture is expected to be low at storage temperatures of <150°C, this problem should be minimal.

B. Stainless Steel-Clad SNF

The SS-clad spent fuels appear to have three primary degradation mechanisms: intergranular stress corrosion cracking, degradation due to sodium interactions, and potential pyrophoricity from UH_3 that may be in the fuel. These issues are discussed below; additional degradation mechanisms are shown in Table 2.

Intergranular stress corrosion cracking of the cladding is suspected in these spent fuels because the irradiation temperatures were in the range for sensitization and embrittlement of the SS cladding. This is a significant concern even for dispersion-type fuels. Due to the inhomogeneous mixing of the fuel compound, powdered metal matrix, and burnable poison, large agglomerations and stringers may form inside the fuel matrix, producing localized "hot spots" on the cladding.[5] Thus, localized areas on the cladding may have been subjected to sensitization temperatures without the entire fuel element experiencing this condition. In addition, the plate-type fuels are also susceptible to swelling of the fuel at these stringer areas, potentially compounding the problem by providing stress to the sensitized area.

Since EBR-II fuels constitute a significant portion of the fuel inventory, specific problems with this particular fuel type become noteworthy. Staff from Argonne National Laboratory-West indicate that sodium has been present on some

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[a]Unirradiated Al-alloy materials have been subject to pitting attack in CPP-666; some evidence suggests that the cause is microbiological. Whether irradiated materials are susceptible remains to be determined.

TABLE 2 Potential Degradation Mechanisms for Selected Al- and SS-Clad SNF at INEL

Cladding Group	Fuel	Cladding Degradation Mechanisms											Fuel Degradation Mechanisms			
		Corrosion	Stress Rupture	Incipient Defects	Embrittlement	Sensitization*	LTS	IGSCC	IASCC	FAE	Hydrate Release	Crud Spallation	Corrosion	Pyrophoric	Fission Product Release	Crumbling
Aluminum	ARMF	Y1*	N		>150°C	N	N	N	N	N	>150°C	Y2	Y1	Y2	Y1	
	ATR	Y1	N	Y1	>150°C	N	N	N	N	N	>150°C	Y2	Y1	Y2	Y1	
	MURR	Y1	N		>150°C	N	N	N	N	N	>150°C	Y2	Y1	Y2	Y1	
	ORR	Y1	N		>150°C	N	N	N	N	N	>150°C	Y2	Y1	Y2	Y1	
	UW	Y1	N		>150°C	N	N	N	N	N	>150°C	Y2	Y1	Y2	Y1	
	HFBR	Y1	N		>150°C	N	N	N	N	N	>150°C	Y2	Y1	Y2	Y1	
	TRIGA Al	Y1	N		>150°C	N	N	N	N	N	>150°C	Y2	N	N	N	Y1
Stainless Steel	EBR-II	N	>400°C	Y7	Y1	Y1	>200°C	Y1	Y6	Y5	N	N	N	Y1	Y2	Y2
	ANL-6	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	Y2	Y1	Y2	Y3
	SPEC	N		Y2	Y1	Y1	>200°C	Y1	Y3	Y1	N	Y1	N	Y1	Y1	
	Fermi Cores 1 & 2	N	NA	Y2	N	Y2	N	Y2	Y2	Y2	N		Y1	Y1	Y1	Y1
	APPR	N	N		Y1	Y1	>200°C				N		N	N	N	N
	BMI	N	N		Y1	Y1	>200°C				N		N	N	N	N
	BORAX V	N	N		Y1	Y1	>200°C				N		N	N	N	N
	YBWR	N	N	Y2	Y1	Y1	>200°C				N		N	N	N	N
	Pathfinder	N	N	N	N	Y1	>200°C		N	N	N		N	N	N	
	SM-1A	N	N	N	N	Y1	>200°C	Y1	N	N	N		N	N	Y1	
	SPSS	N	N	N	N	Y1			N	N	N		N	N	N	
	TRIGA FLIP	N	N		Y1	N	>200°C	N	N	N	N		N	N	N	Y1
	TRIGA SST	N	N		Y1	N	>200°C	N	N	N	N		N	N	N	Y1
	TRIGA Ber II	N	N		Y1		>200°C				N		N	N	N	Y1

FAE = Fuel Agency Effect;   IASCC = Irradiation assisted stress corrosion cracking.
N indicates mechanism is of low concern; Y indicates mechanism is of concern; lower numbers indicate higher order of importance. NA = Not applicable. * Y indicates that the cladding may have been sensitized during irradiation.

of the EBR-II fuel assemblies before shipment. Sodium has been shown to attack the SS cladding in reactor by preferentially reacting with the protective chromium and nickel, potentially exacerbating the problems of embrittlement and cracking.[6] This detrimental reaction may occur (albeit at a decreased rate) if sodium is in contact with the cladding over an extended period of time. Additionally, the free sodium (both outside the fuel pins and as a bonding agent inside) has been identified as a vulnerability issue for potential reactions with the water environment, causing possible hydrogen explosions.

Formation of UH_3 may be an issue in the EBR-II, ANL-6, and SPEC fuels, although the rates of formation should be slower than for pure uranium. If the cladding of a uranium-metal-bearing fuel element should fail, water can react with the sodium present to release both heat and hydrogen. A moist hydrogen environment is optimal for uranium hydride formation and unlike the dispersion-type fuels with small fuel particles, the solid uranium alloys are undiluted and can potentially yield an uncontrolled pyrophoric reaction if exposed to an oxygen atmosphere (such as during drying). This sequence of events would require breached fuel elements with the storage can possessing a moist environment. The ANL-6 and SPEC fuels are breached and the nominal EBR-II fuels exhibit cracking and irradiation creep failures as the dominant failure mechanism during in reactor service.[7] The storage cans are SS but the actual environment under which the spent fuel has been exposed, inert or air, moist or dry, varies with the age of the fuel and when it was canned. This perceived issue could affect over half of the spent fuel inventory, depending on its prevalence. To date, no examinations of the fuel have been conducted to determine the condition of these fuels.

C. $UZrH_x$ Spent Fuel

The $UZrH_x$ fuels are considered very stable, and as a result very few degradation issues could be determined. No quantitative data were found for the performance of $UZrH_x$ in these environments to confirm the conclusion on inertness. If the $UZrH_x$ is inert, then failure of aluminum or SS cladding containing this material will be less important to the overall performance of this fuel type in a dry storage environment.

While the unclad $UZrH_x$ should be relatively inert in the current and potential storage environments, but it is brittle. The inertness is primarily due to the thin, tenacious oxide layer (ZrO_2) formed on the exposed surfaces of the ZrH_x matrix material.[8] This oxide "skin" has been shown to provide an impervious structure that readily resists attack by corrosive agents. However, the ZrH_x matrix material is also extremely brittle and can potentially crack or break if mishandled. Such breakage could lead to problems of confinement if the containers storing the fuel fail catastrophically, resulting in difficulty in retrieving the fuel from the dry storage container.

IV STORAGE AND CONDITIONING REQUIREMENTS

Solutions to the above issues appear possible based on information in the literature on degradation mechanisms, experience with similar fuels or materials at other facilities, and extensive experience with wet and dry storage of commercial spent fuel. A first estimate of the acceptable conditioning methods and dry storage conditions for the aluminum-clad, SS-clad, and unclad $UZrH_x$ fuels is summarized below.

- A careful drying scheme will need to be developed to ensure that water-logged fuel types do not further degrade during drying operations. Any remaining water inside the plate-type structures can produce excessive stresses during the liquid-gaseous-phase change and must be avoided to minimize further degradation.

- For drying operations involving all the spent fuels evaluated, initial limits appear to be temperatures below 300°C (except SS-clad U alloy fuel) and evacuation to <5 torr static pressure prior to filling with a cover gas. Limits for the SS-clad U alloy fuel appear to be <200°C. Limiting concerns include residual moisture for all of the fuels (which is the main reason for the 300°C limit) plus concern over U oxidation and pyrophoric reactions involving UH_3 for the EBR-II fuel for storage temperatures >200°C.

- For dry storage of the spent fuels evaluated (except the SS-clad $UZrH_x$ and the unclad $UZrH_x$), initial dry storage limits of <150°C and 99.995% He, Ar, or N_2 are suggested. If UH_3 is proven to not be an issue, then it might be possible to justify air storage for aluminum-clad UAl_x and U_3O_8 fuels.

- Dry storage limits for the SS-clad $UZrH_x$ and the unclad $UZrH_x$ are initially proposed to be temperatures <400°C with 99.995% He, Ar, or N_2 cover gas. Limiting concerns include corrosion, crumbling, and dehydration, depending on the fuel type.

- For a given fuel, if UH_3 is detected in significant amounts, conditioning treatments may be justified; drying and canning operations must be conducted to avoid ignitions.

Based on the evaluation thus far on the spent fuel conditions and known or potential failure mechanisms, several recommendations can be made to determine whether the initial limits proposed for drying operations and dry storage are valid. These recommendations are outlined below.

A. Aluminum-Clad Spent Fuels

- Determine whether UH_3 is present and in sufficient quantity to warrant conditioning to remove the hydride from the fuel matrix. If the hydride is present, it will need to be determined whether a potential pyrophoric reaction is self-sustaining in the dilute dispersion type fuels, including those with SS as well as aluminum fuel matrices. If the reaction is not self-sustaining, then the hydride formation issue becomes less significant.

- Determine how much moisture is generated from actual spent fuel and any remaining corrosion products present to clarify whether moisture will build up during drying or dry storage. A determination of the hydrogen gas generation would be readily available from this information as well. If UH_3 is not prevalent or is not an issue, determine whether air storage for the aluminum-clad fuels is feasible.

B. Stainless Steel-Clad Spent Fuels

- Determine what was and is the actual storage environment for the EBR-II spent fuel, including the amount of moisture at the time the fuel was canned. It is important to know whether the double containment of these fuels has been breached and allowed water to come in contact with the fuel elements and generated hydrogen gas.

- Determine the extent of cladding damage resulting from prolonged exposure to sodium. If it appears that the EBR-II fuel canisters and cladding are failed, verify the extent of UH_3 formation in the U-fissium fuel and the probability of a self-sustaining pyrophoric reaction taking place either during drying or dry storage. Both the extent of formation and the pyrophoricity of this alloy is expected to be less than for unalloyed uranium metal.

C. Unclad $UZrH_x$ Spent Fuel

- Determine whether the fuel is stable in water and in long-term dry storage. No definitive data on diffusion, leach, or corrosion rates in ambient water storage were found to verify such conclusions.

- Verify the brittleness of the $UZrH_x$ fuel to determine whether unclad or clad fuel of this type will present a handling/retrieval issue due to fuel element breakage.

V. CONCLUSIONS

Evaluation of dry storage for the spent fuels in the CPP-603 and CPP-666 basins is a challenging task because of the wide variety of fuel and cladding materials that have been exposed to many different irradiation and storage histories. If evaluation of the as-stored condition and failure mechanisms of the fuels can be conducted to satisfy the issues, then dry storage requirements may be less restrictive than those proposed.

The fuels could be segregated into failed and unfailed categories, possibly reducing the extent of any conditioning required based on experience noted in the literature on moving some unfailed Al-clad fuels from dry storage with minimal processing. However, selection of failed and unfailed fuel would require an assessment of the available techniques for making such a selection, and might be a significant task in itself. On one hand, it appears that

additional information will be required to verify these initial storage limits or to determine how to condition the fuel, particularly for fuel known to have failed cladding. On the other hand, dry storage is now a mature and widely applied technology. While no failed uranium-bearing fuel that has been subject to aqueous corrosion has yet been moved from wet to long-term dry storage, methods have been demonstrated to safely condition fuel that may include uranium hydrides. In any case, the stakeholders of this facility may be required to make hard choices between proposed schedules, costs, and adequately understanding the needed conditioning and storage environments.

ACKNOWLEDGMENTS

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