DISPOSAL OPTIONS AND POTENTIAL CHALLENGES TO WASTE PACKAGES AND WASTE FORMS IN DISPOSAL OF SPENT (IRRADIATED) ADVANCED REACTOR FUEL TYPES

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ABSTRACT

Degradation mechanisms for both solid coated particle fuel and the associated possible waste forms for nuclear metal fuel were examined to identify possible challenges for disposal of spent (irradiated) advanced reactor fuel (ARF) in a geologic repository. ARF types considered in this report include solid coated particle fuel, commonly known as tristructural isotropic (TRISO), and nuclear metal fuel characteristic of compact fast reactors. Nuclear metal fuel consists of uranium alloys such as U-Pu, U-Fs, U-Zr, U-Mo, and U- Pu-Zr, often with Na between the fuel and cladding to enhance heat transport. To date, there has been no operating geologic repository for disposal of spent fuel and associated high-level waste, including irradiated ARF; however, research conducted by the U.S. Department of Energy (DOE) into material performance of spent ARF in disposal environments provides insights into possible challenges related to disposal of spent ARF.

The SiC and pyrolytic carbon (PyC) layers of spent coated particle fuel discharged from advanced reactors, in both pebble-bed and prismatic block designs, perform as barriers against certain degradation mechanisms. These protective layers are expected to better contain radionuclides than light water reactor (LWR) fuel, and enhance TRISO fuel performance in both oxidizing and reducing repository environments when compared to LWR fuel. Whole-block spent coated particle fuel assemblies for direct disposal are often larger per unit mass compared to LWR spent nuclear fuel assemblies, as only approximately 24 percent of the volume is represented by fissile material in these fuel assemblies. Therefore, waste packages used for whole-block disposal are often larger, and for space-limitation reasons, additional possible options for waste conversion have been considered to separate the fission products and allow the graphite to be disposed as Class C low-level waste. Fission product diffusivities and corrosion rates were analyzed to understand the performance and longevity of solid coated particle fuel under hypothetical disposal environments. However, information on diffusivity through SiC under repository conditions is not available in the literature. Although there has been no experience with disposal of TRISO fuel to date, quantitative evaluation of the bulk diffusivities in SiC of the radionuclides of interest at repository temperatures would provide some insight into future disposal performance for spent ARF.

High-level waste forms from treating spent driver metal fuel for disposal—including converted salt waste, ceramic waste, and metallic waste without sodium—and spent blanket metal fuel have been characterized to understand degradation mechanisms in disposal environments. Waste forms converted from spent metal fuel would be packaged into containers, filled with inert gas, and then placed within a sealed over-pack prior to disposal. Salt wastes are chloride-rich and soluble and if they dissolve, chloride is known to induce pitting corrosion and stress corrosion cracking in many metals such as stainless steel and nickel alloys. Conversion of spent metal fuel to ceramic waste immobilizes radionuclides in the ceramic structure and dilutes Pu concentration, but this ceramic waste form is subject to dissolution, as salt-containing ceramic materials are hygroscopic, as well as to radiation embrittlement. Metallic waste could be subject to oxidation, pitting, and galvanic corrosion, and when chloride is present in the disposal environment, localized corrosion would be enhanced. For the spent blanket fuel, the presence of water inside the disposal package can cause uranium to react and form uranium oxides and hydrogen gas, some of which can be absorbed into the spent metal fuel to form uranium hydrides, which are pyrophoric materials that can challenge waste package performance.

Material performance of these fuels as characterized in DOE research offers key insights into possible challenges to disposal of spent ARF in deep geologic repositories; however, additional research into specific waste package performance in relation to postulated repository environments can better characterize the possible disposal performance, given the specific properties of spent ARF that are important to waste package performance, including enrichment levels, and attributes that are unique to processed metallic waste.

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ABBREVIATIONS/ACRONYMS

ARF	advanced reactor fuel
AVR	Arbeitsgemeinschaft Versuchsreaktor
CFR	*Code of Federal Regulations*
CNWRA®	Center for Nuclear Waste Regulatory Analyses
DOE	U.S. Department of Energy
EBR-II	Experimental Breeder Reactor-II
FFTF	Fast Flux Test Facility
FHR	fluoride salt-cooled high-temperature reactor
FSV	Fort St. Vrain
HLW	high-level radioactive waste
HTGR	high-temperature gas-cooled reactor
INL	Idaho National Laboratory
LEU	Low enriched uranium
LWR	light water reactor
LLW	low-level waste
NRC	U.S. Nuclear Regulatory Commission
RH-TRU	remote-handled transuranic
SNF	spent nuclear fuel
THTR	Thorium High Temperature Reactor
TN-FSV	Transnuclear Fort St. Vrain
TRISO	tristructural isotropic
U.S.	United States
WIPP	Waste Isolation Pilot Plant

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QUALITY OF DATA, ANALYSES, AND CODE DEVELOPMENT

DATA: There are no original CNWRA-generated data in this report. Sources of other data should be consulted for determining the level of quality of those data.

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1 INTRODUCTION

1.1 Background

As the U.S. Nuclear Regulatory Commission (NRC) staff prepares for regulatory interactions and potential license applications for non-light water reactor (non-LWR) technologies, a need to develop an understanding of potential challenges associated with regulating the long-term storage, transportation, and disposal of advanced reactor fuel (ARF) types has been identified. For example, revisions may be needed to guidance documents and rules in Title 10 of the *Code of Federal Regulations* (10 CFR) Part 71 and 10 CFR Part 72. Potential ARF types that may be subject to NRC regulation in the future include metal fuels (i.e., uranium alloys such as U-Pu, U-Fs, U-Zr, U-Mo, U- Pu-Zr, often with Na between the fuel and cladding), uranium fuels for high-temperature gas-cooled reactors (HTGR) and molten salt reactors.

The Center for Nuclear Waste Regulatory Analyses (CNWRA®) has been tasked with identifying and assessing the significance of potential technical challenges and issues associated with the storage, transportation, and disposal of ARF types. This report evaluates potential challenges to the performance of waste packages and waste forms during disposal. ARFs and associated waste forms may degrade and interact with other materials in ways that would need to be considered in the design of waste packages and potentially the selection of repository sites. Early identification of the distinctive characteristics and degradation mechanisms of ARFs and associated waste forms and challenges to material performance in the repository setting may aid planning and preparing for future NRC safety reviews.

1.2 Purpose and Scope

This report, based on a literature review, presents available information on the topic of disposal of non-LWR fuel types, which include solid coated particle fuel, commonly known as tristructural isotropic (TRISO), and nuclear metal fuels such as uranium alloys. (UF_4, PuF_3, or ThF_4 used in liquid fueled molten salt reactors are not part of this review because TRISO fuel is more commonly proposed to be used as fuel for molten salt reactors in the U.S.) Based on the unique physical and chemical properties of spent non-LWR fuel and possible alternative waste forms, proposed disposal options were assessed in order to identify possible degradation mechanisms that could impact disposal options and decisions. This report also discusses possible safety and material performance characteristics for the different fuel types in proposed disposal systems.

2 DISPOSAL OPTIONS FOR SPENT ARF TYPES

Geologic disposal is generally adopted worldwide for managing high-level radioactive waste (HLW), including spent ARF types. Many countries have been exploring the feasibility of HLW disposal in mined geologic repositories constructed in salt, crystalline rock, or clay, including Canada, China, Czechia, Finland, Germany, Japan, Sweden, and Switzerland (NWTRB, 2017; King, 2017, 2013). Figure 2-1 illustrates the concept of one mined repository concept proposed by the Swedish Nuclear Fuel and Waste Management Co. (SKB, 2011). Deep underground repositories are typically planned to be in stable rock formations (e.g., granite), which are about 500 m [0.31 mi] to 1,000 m [0.62 mi] deep in the saturated zone below the water table. Because of the relatively high thermal conductivity of saturated backfill and host rock, the temperature at such deep geologic repositories is lower and the supply of oxidant is limited (King, 2017, 2013). The proposed U.S. repository design is in the unsaturated zone above the water table. The repository environment in that design is aerobic and hotter and drier than repositories located below the water table. Deep borehole disposal is another concept that has been considered at various times (Figure 2-1). In one concept discussed in NWTRB (2017), 5-km [3.1-mi]-deep boreholes would be drilled into extremely low permeability crystalline basement rock with a bottom-hole diameter of 0.43 m [17 in]. Waste forms would be emplaced in the lower 2 km [1.2 mi] of the borehole. The upper part would be sealed with concrete, compacted clay, or asphalt.

For most of the proposed repositories, an engineered barrier system is designed to work with the natural barrier to help isolate and contain the HLW after the repository is closed. The main part of the engineered barrier is the waste container. The containers are in general designed as double-walled metallic cylinders (e.g., Figure 2-2), with each cylinder serving a specific function. The inner container directly holding the HLW is usually designed to shield radiation and provide mechanical strength to facilitate safe waste handling and emplacement operations. The outer container is usually a corrosion-resistant barrier, such as the copper canister shown in Figure 2-2 in the waste package proposed by SKB, and the Alloy 22 outer container for the proposed U.S. repository.

2.1 Coated Particle Fuel Disposal Options

Spent coated particle fuel elements discharged from HTGRs or other advanced reactors, in both the pebble-bed and prismatic block designs, are expected to better retain radionuclides than LWR spent nuclear fuel (SNF) in a geologic repository. The multiple coating layers on the fuel particles and the graphite fuel elements are considered engineered barriers that could provide an effective long-term barrier against fission product transport and minimize reliance on performance of the waste package or surrounding geologic media for disposal. To date, there is no disposal experience with spent coated particle fuel. Conceptually, there are three possible options for disposal of spent coated particle fuel in a repository (Shropshire and Herring, 2004; Lotts et al., 1992), including

(i) direct disposal of prismatic block or graphite-matrix coated particle fuel,
(ii) disposal with removal of graphite block, or
(iii) reprocessing of spent fuel to separate the fuel and fission products.

Since commercial reprocessing of spent fuel is currently not practiced in the U.S., only the first two options are discussed in this report.

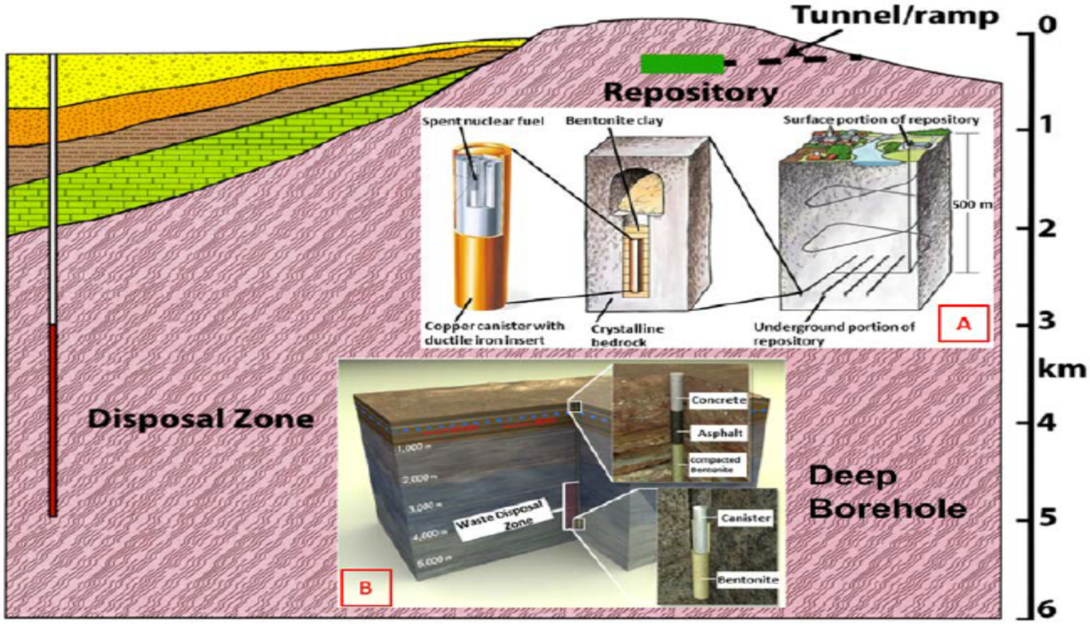


Figure 2-1. Concepts of a mined geologic repository and deep borehole for high-level waste disposal (NWTRB, 2017)

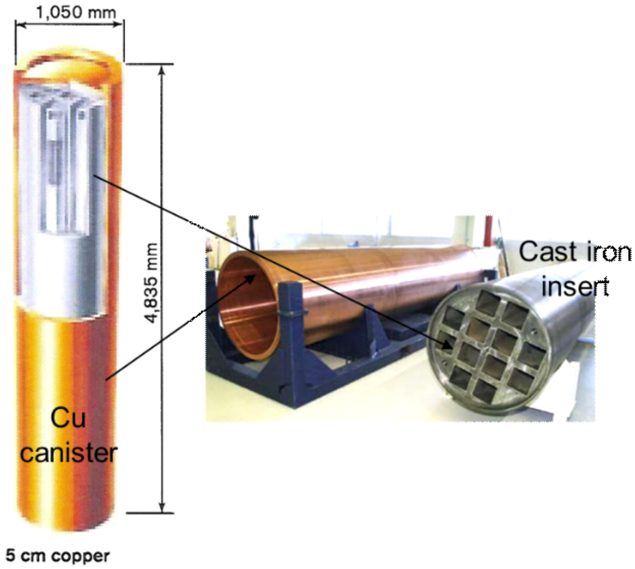


Figure 2-2. Waste container proposed by the Swedish Nuclear Fuel and Waste Management Co. (SKB, 2011)

The first option—direct disposal of the spent coated particle fuel elements in a repository after suitable packaging—is the only option proposed in Germany for final disposal of coated particle fuel pebbles without any treatment or reprocessing (IAEA, 2012; Fachinger et al., 1997). Whole-block disposal of GT-MHR SNF has also been described as a preferred option because of advantages related to ease of implementation, proliferation risks, safeguards requirements, and cost (IAEA, 2012; Richards, 2002; Kiryushin et al., 1997). There have been a number of studies on direct disposal of spent coated particle fuel in different geological media (Gelbard and Sassani, 2018; Van den Akker and Ahn, 2013; Fachinger et al., 2006). The general consensus is that repository performance for both oxidizing and reducing environments would be orders of magnitude better than for LWR SNF because of the chemical characteristics of the spent coated particle fuel (e.g., protective SiC layer). The coated particles alone could potentially prevent water from contacting the fuel kernels for several million years, primarily due to the protective SiC layer (see Section 3.2).

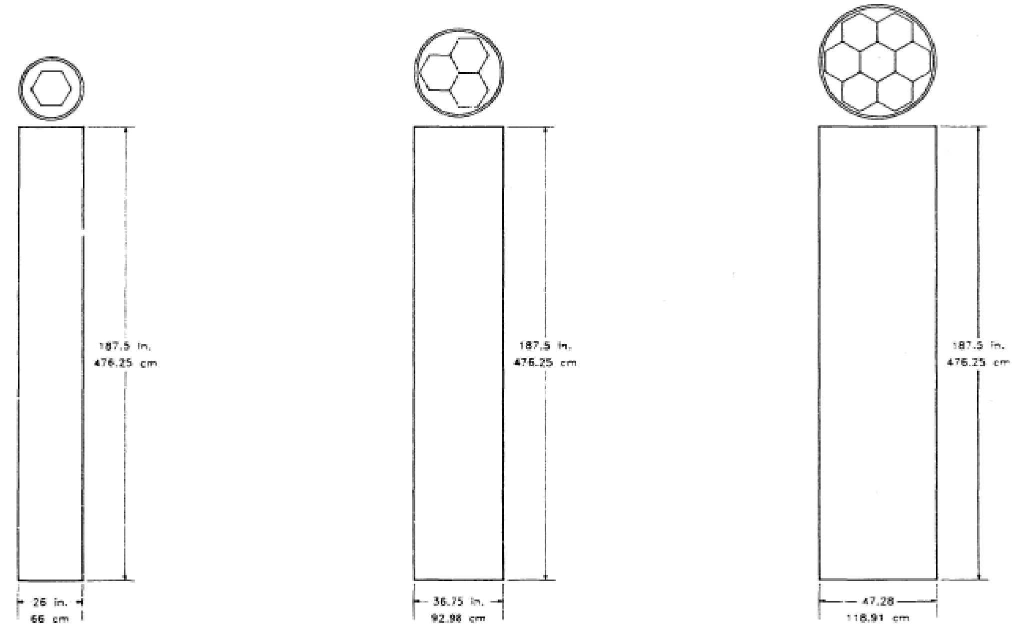
Direct disposal of the spent coated particle fuel elements has the disadvantage of requiring considerably more waste package volume because of the large volume of the fuel elements relative to the typical fuel content in an LWR SNF assembly (Shropshire and Herring, 2004; Lotts et al., 1992). However, spent coated particle fuel has a lower decay heat load by a factor of 5 to 10 compared to LWR SNF. The repository waste package size is limited, among other factors, by spent fuel decay heat load to avoid overheating the waste. As a result, larger diameter or taller waste packages, to maximize storage capacity within thermal design limits, may be an option for direct disposal of spent coated particle fuel. Spent coated particle fuel can be placed in the same waste package designs proposed for LWR SNF. For example, the Fort St. Vrain (FSV) reactor generated a total of 2,208 spent fuel elements in the form of TRISO-coated particles in graphite prismatic blocks. In the context of the LWR SNF waste package in the U.S. design, Figure 2-3 shows a conceptual waste package of similar dimensions and two larger waste package sizes for spent coated particle fuel, which contain 6, 18, and 42 FSV spent fuel elements, respectively (Shropshire and Herring, 2004; Lotts et al., 1992).

The second option is to separate the spent prismatic block fuel elements into the spent fuel matrix material and the carbon from the graphite block (Lotts et al., 1992). There are two separation processes involving a physical or mechanical separation of the fuel from the graphite block and a chemical process such as burning, where the graphite is oxidized to carbon dioxide (CO_2). Separation of the fuel particles from the graphite matrix has been demonstrated to be feasible by separating by physical methods, such as simply pushing the fuel rods out of the graphite block or by mechanical means such as crushing and screening (Lotts et al., 1992). Feasibility of separation by burning either whole fuel elements or crushed fuel elements has also been demonstrated with effective processing of the gaseous products. Fission product contamination resulting from physical separation of the graphite block remains a concern because physical removal of graphite from fuel rods will not likely be a clean separation with existing fuel elements. For chemical separation of graphite block, burning would produce a large quantity of CO_2 that must be processed because of the presence of ^{14}C in the irradiated graphite. Conversion of the CO_2 into a solid form, such as calcium carbonate ($CaCO_3$), would significantly increase the total waste volume over the volume of the original fuel elements.

If the bulk graphite could be cleanly separated from the spent prismatic block fuel, this option would allow the graphite to be disposed as Class C low-level radioactive waste (LLW) and the spent fuel to be emplaced at lower volume in the geologic repository. For example, the fuel rods of the FSV spent fuel elements take up only approximately 24 percent of the overall fuel assembly volume (Lotts et al., 1992). In the U.S., graphite waste classified as Class C LLW can be disposed in a surface or shallow repository. Hence, it is critical to keep graphite impurities low and to track neutron exposure to preclude the creation of greater than Class C waste to insure the surface repository disposal option remains viable (IAEA, 2010).

2.2 Waste Forms from Spent Metal Fuels and Disposal Options

Because sodium-bonded metal fuel reacts vigorously with water and the enrichment level varies within a wide range (0.1 to 93 percent U^{235}), spent metal fuel has not been studied for direct disposal in any geologic repository. As discussed in Hall et al. (2019a), the U.S. Department of Energy (DOE) has chemically treated some spent metal fuels and plans to continue to treat the remaining spent driver metal fuel to (i) change the highly reactive metallic sodium to nonreactive ionic sodium, (ii) recover the highly enriched uranium, and (iii) convert cladding and remaining materials to other HLW forms. The highly enriched uranium recovered from the spent fuel is diluted with depleted uranium to create a low-enriched uranium product for other uses (NWTRB, 2017). The HLW forms include salt waste and metallic waste. Salt waste can be treated further to generate ceramic HLW. These converted HLW forms could have viable disposal options in a geologic repository. The sodium in the spent blanket fuel will be separated, likely by a melt, drain, evaporate, and carbonate process, leaving the metal fuels for disposal. As a result, there are three possible waste categories for disposal:



EXTERNAL DIMENSIONS OF YUCCA MOUNTAIN REFERENCE CONTAINER (6 fuel blocks, total)

CONTAINER FOR 3 FUEL BLOCKS PER LEVEL (18 fuel blocks, total)

CONTAINER FOR 7 FUEL BLOCKS PER LEVEL (42 fuel blocks, total)

Figure 2-3. Example waste package design for spent coated particle fuel (Lotts et al., 1992)

(i) sodium-bonded spent driver metal fuel,
(ii) converted salt waste, ceramic HLW, and metallic HLW without metallic sodium, and
(iii) spent blanket fuel without metallic sodium.

Based on DOE research, available information is not sufficient to evaluate the performance of direct disposal of sodium-bonded spent driver metal fuel in any geologic disposal concept. Due to the challenges associated with disposing the extremely reactive metallic sodium, this waste form may require treatment regardless of the disposal approach. As a result, direct disposal of sodium-bonded spent metal fuel [category (i)] is not a feasible disposal option. Waste forms from categories (ii) and (iii) are feasible disposal options in a geologic repository because they do not contain the reactive metallic sodium.

2.2.1 Salt High Level Waste Form

As mentioned in Hall et al. (2019a), the chemical treatment of sodium-bonded spent metal fuel generates two separate HLW streams: (i) salt waste and (ii) metallic waste. The majority of the salt is LiCl and KCl, followed by NaCl (Wang et al., 2011). Other salts include CsCl, $SrCl_2$, $BaCl_2$, $LaCl_3$, $CeCl_3$, $PrCl_3$, $NdCl_3$, $SmCl_3$, UCl_3, and $PuCl_3$ (Karlsson et al., 2019). Many of the actinides and fission products from the spent metal fuel would be dispersed in the salts (Rechard et al., 2017a). The Pu concentration is higher than commercial LWR SNF and defense related HLW. The $PuCl_3$ content in these salt waste forms can be 3 percent and 6 percent from spent driver and blanket fuels, respectively, and may present challenges for nuclear criticality issues in disposal environments. In one concept for direct disposal of salt waste, 43 kg [95 lb] of the waste would be contained in a 50.5 cm [19.9 in]-long Type 316 stainless steel canister with a diameter of 25 cm [9.8 in]. A larger stainless steel canister with a diameter of 27 cm [11 in] and a length of 155 cm [61.0 in] would be used to hold three smaller canisters. The large canister is loaded into an overpack closed with a welded lid to complete the waste package. It is expected that 25 waste packages will be needed to contain all of the existing and projected salt waste if it is not turned into a ceramic HLW form. The heat output from the salt waste is expected to be lower than from LWR SNF (Rechard et al., 2017a).

Rechard et al. (2017a) discussed three possible disposal options for the salt waste: (i) deep borehole disposal in crystalline basement rock, (ii) the Waste Isolation Pilot Plant (WIPP) located in bedded salt in New Mexico, and (iii) a mined repository for commercial and/or defense waste. The specific packaging selection depends on the storage environment of the repository.

In the deep borehole disposal concept, the radioactive waste would be placed in a container in the bottom 1–2 km [0.6–1.2 mi] of the approximately 5 km [3.1 mi] deep borehole in the crystalline basement rock. According to Rechard et al. (2017a), the advantages of this disposal concept include (i) restricted migration of radionuclides due to low permeability in deep crystalline rocks, (ii) little interaction between deep and shallow fluids, (iii) limited solubility and enhanced sorption of radionuclides in deep reducing conditions, and (iv) further restricted mobility of radionuclides by the high salt content of the waste. Rechard et al. (2017a) conducted a thermal-hydrologic analysis of this disposal concept assuming that the salt waste would be contained and disposed in the same package as the transportation cask RH-TRU 72-B described in Hall et al. (2019b). Figure 2-4 shows the thermal and salt concentration profiles as a function of time for salt water at 3,000-m [1.9 mi] depth in the borehole. The temperature was computed to reach a peak of 87.5 °C [190 °F] in several years, then decay to 85 °C [185 °F] in hundreds of years. The salt concentration reached a small peak at around 250 years, then reached larger concentrations at about 10,000 years. The released salt is expected to move downward because of the larger density of saline water. This disposal concept could be a feasible option for salt waste because it relies less on waste form and waste package integrity to demonstrate repository performance.

Rechard et al. (2017b) described a roadmap for disposing salt waste as remote-handled transuranic waste at WIPP. There are 11 tasks for implementing this alternative disposal pathway, including modifying the WIPP Hazardous Waste Facility Permit, updating the environmental impact assessment on disposition of the salt waste, and developing a shipping cask. WIPP sets limits on remote-handled transuranic waste volume, total activity, and activity concentration. Rechard et al. (2017b) commented that accepting salt waste at WIPP is expected to fall within the limits because the salt waste only accounts for a small part of the total waste that WIPP can accept. However, performance assessments would be needed to demonstrate the safety and viability of this option.

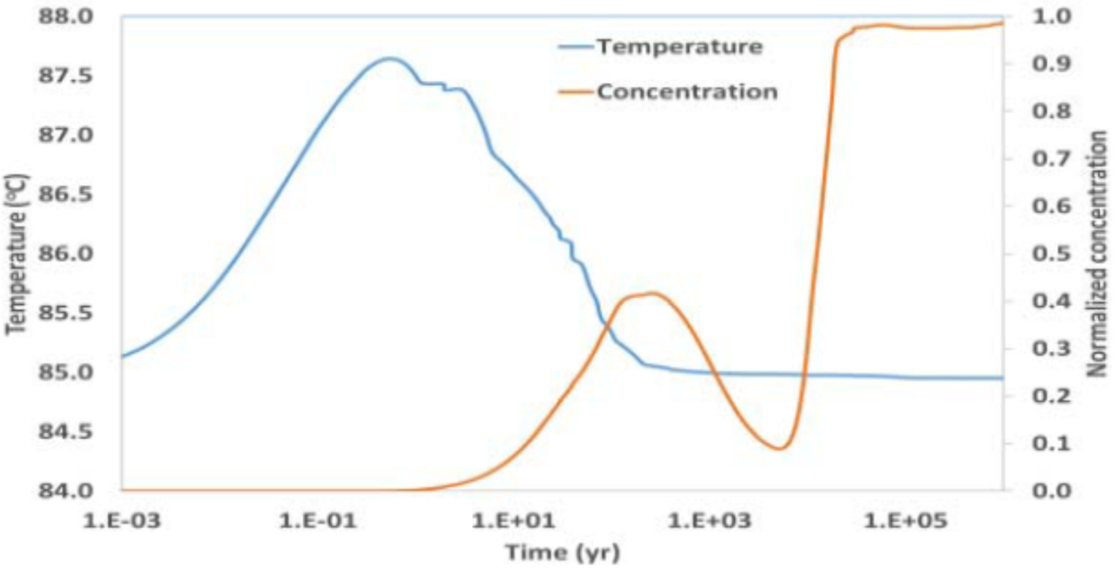


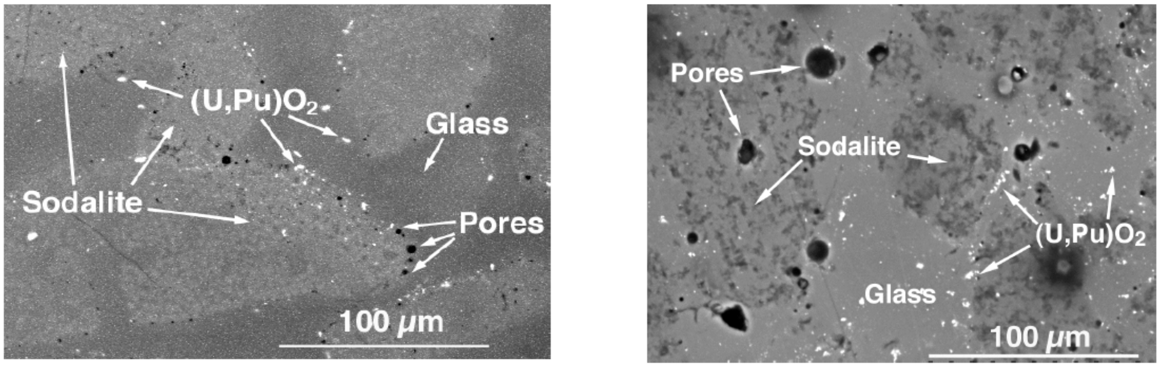
Figure 2-4. Computed thermal and salt concentration as a function of time for salt water at 3,000-m [1.9 mi] depth in a disposal borehole (Rechard et al., 2017a)

2.2.2 Ceramic High-Level Waste Form

Because of its high salt concentration, salt waste cannot be processed into HLW glass, which is a commonly engineered HLW form from defense waste. Instead, DOE blended the salt waste with zeolite and glass and then processed the salt waste into a glass-bonded sodalite [$Na_8(AlSiO_4)_6Cl_2$] composite ceramic HLW form (Hall et al., 2019a). Compared to salt waste, this process is more complex and is a more expensive path to treat spent metal fuel because (i) it involves additional steps and (ii) it increases the volume of HLW to be disposed of, given the addition of glass and zeolite during treatment. However, the ceramic HLW form immobilizes the radionuclides in the ceramic structure and dilutes the Pu concentration in the waste form to mitigate potential criticality issues associated with the salt waste. Furthermore, the chloride salts are incorporated into the cages of the zeolite structure, forming salt loaded sodalite, making the salt much less soluble and less corrosive when in contact with residual water (Richardson, 1997).

This ceramic HLW form could contain 0.2 to 0.6 mass percent low enriched uranium (LEU) and up to about 0.5 mass percent plutonium, which is lower than in salt waste. The ceramic is made into a 1-m [39- in] long cylinder with a diameter of 0.5 cm [0.2 in]. Based on current designs, there would be 64 HLW canisters for disposal of ceramic waste, amounting to approximately 51,000 kg [110,000 lb] of waste from treating both EBR-II and FFTF fuels. Based on projected radionuclide content, the thermal output of a canister with two cylinders is calculated to be 2.24, 1.73, and 1.25 kW/canister for 6-, 10-, and 20-year-old fuel wastes, respectively. ^{90}Sr, ^{137}Cs, ^{239}Pu, ^{240}Pu, ^{241}Pu, and ^{241}Am are projected to be the primary potential dose contributors.

The ceramic HLW form is a composite material that comprises several phases, including approximately 70 mass percent sodalite, 25 mass percent glass, and 5 mass percent of a mixture of halite and inclusions within the glass. The inclusions comprise most of the radionuclides, including actinides and rare earth element fission products that are immobilized as insoluble oxides in the glass. Small amounts of radionuclides are dissolved in the glass or incorporated into other phases. Figure 2-5 is photomicrographs showing of the microstructures of U-, Pu-doped ceramic HLW forms produced from hot isostatic press and pressure-less consolidation methods (Ebert, 2005), in which the glass binds and encapsulates sodalities (Ebert, 2005). Most of the radionuclides formed as inclusions within the binder glass. Small amounts of radionuclides are dissolved in the glass or incorporated into the sodalite and halite phases.



(a) (b)

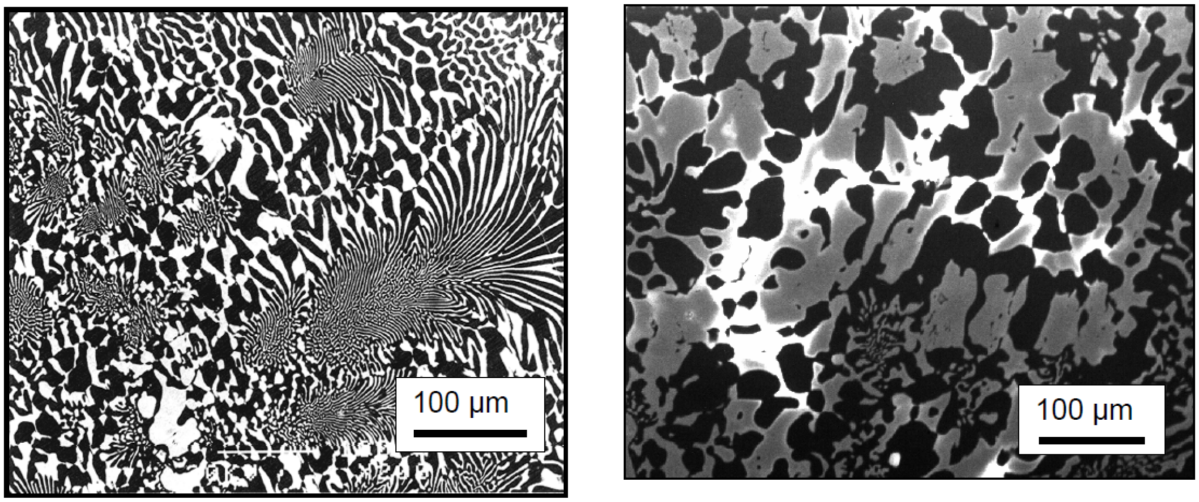
Figure 2-5. Photomicrographs showing microstructures of U, Pu-doped ceramic HLW forms produced from (a) hot isostatic press and (b) pressure-less consolidation (Ebert, 2005)

2.2.3 Metallic High-Level Waste Form

The metallic waste stream from treating spent driver metal fuel is immobilized by melting in an induction furnace at about 1,600 °C [2,912 °F] with added zirconium and depleted uranium to alloy the radionuclides and reduce the enrichment level to 20 mass percent ^{235}U (Ebert, 2005).

Zirconium is usually added to make some radionuclides less soluble and therefore less likely to be released. ^{234}U and ^{99}Tc are the primary dose contributors for metallic HLW forms. The metallic form is cast as disk-shaped 13 cm [5.0 in]-thick ingots, which each are about 36–41 cm [14–16 in] in diameter and weigh about 12 kg [26 lb]. The total amount of the metallic HLW form is estimated to be 5,900 kg [12,900 lb] from treating both EBR-II and FFTF fuels. DOE also plans to package these metallic ingots in the same type of canister used for packaging the ceramic HLW form for disposal or mix one metallic HLW form ingot with two ceramic HLW cylinders in the same package for co-disposal.

The predominant composition (about 90 percent) of the metallic HLW form is irradiated stainless steel cladding. The metallic HLW form has two main phases with nearly equal volumetric amounts interspersed in a microscopic scale: (i) an intermetallic $Zr(Fe,Cr,Ni)_{2+x}$ phase and (ii) an iron solid solution phase. The latter may contain ferrite or a mixture of ferrite and austenite, depending on the type of original stainless steel cladding. Figure 2-6(a) shows an SEM photomicrograph of a metallic HLW form with 15 mass percent Zr. The dark regions are the stainless steel phase and the light regions are the intermetallic phase. The phases are present in almost equal amounts. Most of the radionuclides (uranium isotopes being the most abundant) are contained in the intermetallic phase and small amounts of Mo and Tc are contained in the other phase. In Figure 2-6(b), the visibly brighter regions of the intermetallic phase correspond to areas in which U and other actinide elements are more concentrated. Because the amount of heat-generating radionuclides is very small, the thermal output of the metallic HLW is negligible.



(a)

(b)

Figure 2-6. (a) Microstructure of a metallic waste form with 15 mass percent Zr containing about 50 volume percent stainless steel (dark regions) and 50 volume percent intermetallic phase (light regions) (b) regions in intermetallic phase enriched in uranium (brighter regions) (Ebert, 2005)

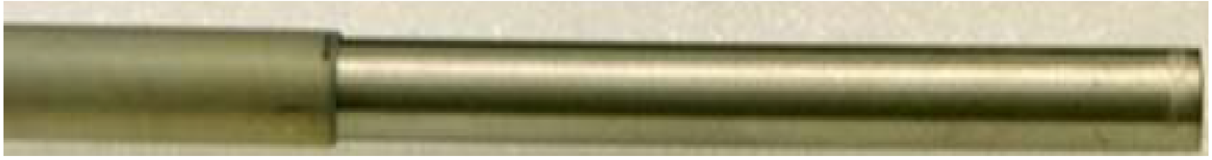


Figure 2-7. Fermi-1 blanket fuel rod after removing sodium by melt-drain-evaporate-carbonate process (INL, 2007)

2.2.1 Spent Blanket Metal Fuel

According to the DOE plan for disposal of spent blanket metal fuels, some of these fuels, such as those from Fermi-1, will be disposed directly in a geologic repository after removing the metallic sodium (INL, 2007). Disposal of this fuel in a deep geologic repository is possible because sodium diffusion into the fuel is minimal. Figure 2-7 shows a Fermi-1 blanket fuel rod after removing sodium (INL, 2007). The blanket fuel contains mostly depleted uranium (less than 0.35 percent U^{235}) and experienced much less fission than driver fuel during reactor operation. For Fermi-1 blanket fuel, a maximum burnup of 0.4 atomic percent was achieved. Because of the low neutron exposure, the Fermi-1 blanket fuel contains only about 0.02 weight percent plutonium. As such, the thermal load and radionuclides are very limited for this waste form in a disposal system.

3 DEGRADATION MECHANISMS OF WASTE PACKAGES AND WASTE FORMS

3.1 Waste Packages and Degradation Mechanisms

The metals or alloys that may be used for the outer container (e.g., for containment of both spent TRISO and HLW forms converted from spent metal fuel) would vary depending on whether the surrounding environment is mostly reducing or oxidizing, and based on the major corroding species that would be present. The location of the repository with respect to the water table has a significant impact on the corrosivity of the environment and the selection of waste package materials. Carbon steel and Cu are two of the candidate metallic materials for waste packages in a deep geologic repository where the environment is mostly reducing, and some titanium (Ti) alloys are candidate materials for waste containment in salt formations. Carbon steel is being considered in different ways as a candidate material for waste packages in HLW geologic disposal systems in several countries (Martin et al., 2014; Kursten et al., 2011; Shibata et al., 2014; King et al., 2014; Boyle and Meguid, 2015). In the Belgian supercontainer concept, the carbon steel overpack is surrounded by a thick concrete buffer (Kursten et al., 2011). Some nations, such as Canada, are looking at Cu-coated steel packages, and propose steel as an internal structural material for instances where steel is not in direct contact with solution (Boyle and Meguid, 2015).

There are important environmental, thermal, mechanical, and radiological factors that affect materials degradation mechanisms and rates of the outer container material, including temperature, groundwater or porewater chemistry, pH, microbial activity, gamma radiation level, the degree of saturation, the level of residual stress, and external load. The following degradation mechanisms have been evaluated and continue to be evaluated for waste package materials under different metallurgical conditions such as mill annealed, welded, and thermally aged:

- General corrosion
- Localized corrosion
- Galvanic corrosion
- Microbiologically influenced corrosion
- Stress corrosion cracking
- Hydrogen induced cracking
- Creep
- Fatigue
- Radiation embrittlement

Copper, titanium, and carbon steels have been characterized for the reducing repositories and Alloy 22 and Ti Gr 7 have been characterized for the mostly dry oxidizing environment of the proposed U.S. repository. In 2014, DOE evaluated options for permanent geologic disposal of SNF and HLW in support of a comprehensive national nuclear fuel cycle strategy (DOE, 2014). In particular, DOE considered disposal of SNF from commercial, defense, and research reactors and existing and projected HLW in mined repositories in three geologic media (salt, clay/shale rocks, and crystalline rocks) and deep boreholes in crystalline rocks. Based on the possible waste forms, the degradation mechanisms listed before this paragraph would be applicable and other potentially applicable mechanisms have not been identified. Because it is uncertain what the waste package material and environmental conditions would be in any future repository designs, the degradation mechanisms of waste package materials are not discussed in detail in this report.

3.2 Coated Particle Fuel

A number of coated particle fuel failure mechanisms identified under in-reactor or postulated accident conditions were reviewed in Hall et al. (2019a). For disposal of the spent coated particle fuel in a geologic repository, numerous studies have been reported on failure mechanisms and degradation rates for TRISO-coated particles and performance assessments have been performed for disposal of spent TRISO fuel in various repository environments (Gelbard and Sassani, 2018; Van den Akker and Ahn, 2013; Peterson and Dunzik-Gougar, 2011; Fachinger et al., 2006; Morris and Bauer, 2005; Richards, 2002). In geologic disposal, the following failure mechanisms are considered for potential release of fission products from TRISO-coated particles:

- pressure vessel (i.e., the SiC layer) failure caused by internal gas pressure buildup
- aqueous corrosion of the SiC layer
- diffusive release through the SiC layer

Since the SiC layer is the primary structural component of the TRISO-coated particle fuel, the effectiveness of the SiC layer as a barrier is critical to overall fuel performance. Ruptures of the SiC layer due to internal pressure buildup from gaseous fission and decay products would permit faster pathways for radionuclide release, which may substantially decrease its barrier function for containment. If groundwater penetrates the waste package, aqueous corrosion reduces the SiC layer thickness, which may reduce the effective tensile strength to retain pressure within the TRISO-coated particle and facilitate SiC layer rupture. Although not formally considered structural failure, there are situations in which the SiC layer becomes functionally failed and is no longer retentive of fission products. Two cases are generally considered: (a) diffusive release through intact SiC and (b) degradation of the SiC layer, resulting in measurable SiC permeability to fission products (IAEA, 2010).

Peterson and Dunzik-Gougar (2011) developed a TRISO fuel degradation model to predict the time to failure of the fuel in a repository environment. The model considered the SiC layer as a pressure vessel within the TRISO fuel that contains fission products. The TRISO-coated particle was assumed to fail after the complete aqueous corrosion of the outer pyrolytic carbon (OPyC) layer, followed by failure of the SiC layer from corrosion and pressure buildup. The authors performed a sensitivity analysis using this model to determine which TRISO fuel parameters would have the most impact on the overall performance of the fuel. Results of the analysis indicate that the corrosion rates, temperature evolution over time, and thicknesses of the OPyC and SiC layers had a significant effect on the estimated time to particle failure. The thicknesses of the kernel, porous carbon buffer, and inner pyrolytic carbon layers, the strength of the SiC layer, and the pressure in the TRISO-coated particle did not significantly alter time to particle failure. This study concluded that a better understanding of the corrosion rates of the OPyC and SiC layers, along with increasing the quality control of the OPyC and SiC layer thicknesses, can significantly reduce uncertainty in estimates of the time to failure of spent TRISO fuel in a repository environment.

Fachinger et al., (2006) conducted experiments to measure corrosion rates for the pyrolytic carbon and SiC (nonirradiated and irradiated) layers of TRISO-coated particles in a number of aqueous solutions (i.e., $MgCl_2$ brine, clay pore water, and granitic groundwater) at various temperatures. The corrosion rates for the irradiated SiC layer ranged from 1.39×10^{-6} g/m^2/day in granitic water at 25 °C [77 °F] to 8.34×10^{-5} g/m^2/day in clay pore water at 90 °C [194 °F]. Based on the corrosion rates of the SiC layer and neglecting the protective effects of the OPyC layer, the lifetime of the TRISO-coated particles was estimated to be between 3,100 and 190,000 years.

Gelbard and Sassani (2018) assessed radionuclide releases from TRISO-coated particles by considering the coupled processes of SiC layer corrosion, diffusion of fission products through a corroding layer, and radioactive decay. A range of fission product diffusivities and corrosion rates for the SiC layer were analyzed. Based on the corrosion rates reported by Fachinger et al. (2006) for irradiated SiC at various temperatures for different brines, the average lifetime of a 35-μm [0.0014-in] thick SiC layer varied between 6,780 years and 104,000 years for the average corrosion rate of 4.48×10^{-5} g/m^2/day at 90 °C [194 °F] and 2.93×10^{-6} g/m^2/day at 25 °C [77 °F], respectively (Figure 3-1). In addition, the corrosion rate of pyrolytic carbon in an $MgCl_2$ brine under air atmosphere at 90 °C [194 °F] is 2.2×10^{-7} g/m^2/day, which would lead to a lifetime of one million years for the OPyC layer. Thus, if the OPyC layer remains intact, it may provide substantial protection of the SiC layer to corrosion by aqueous solutions such that the SiC layer may have a lifetime of up to two million years. The authors also found that fission product releases may occur before the SiC layer is removed by corrosion if the diffusivity of those fission products is more than about 10^{-20} m^2/s (Figure 3-2). Although effective diffusion coefficients have been established for the fission products in SiC under in-reactor conditions, the bulk diffusivities in SiC of the radionuclides of interest at repository temperatures are not available in the literature. As such, quantitative evaluation of diffusion in the compact, porous graphite matrix and the TRISO coating layers at repository condition is warranted.

3.3 High Level Waste Forms Converted from Spent Metal Fuel

The chemical composition, physical size, fissile material concentration, and radioactivity of HLW forms derived from spent metal fuels affect degradation processes that occur in a repository and contribute to the risk of radionuclide releases to the environment.

3.3.1 Salt High Level Waste Form

Salts contained in the waste are chloride-rich and soluble and dissolved chloride is known to induce corrosion, especially pitting corrosion and stress corrosion cracking, in many metals such as stainless steel and nickel alloys. The waste would be subject to possible dissolution if residual water in the waste package is present, forming chloride-rich electrolyte. This electrolyte can corrode the waste package materials internally. If the waste package is breached, the waste can be potentially highly corrosive to other metallic barriers or other waste forms in a disposal system.

The compatibility of salt waste with a repository depends on its location. For example, salt waste is compatible with a repository in a mined salt dome; radionuclides could be less soluble and mobile in salt. As a result, it is a feasible option to dispose salt waste in a salt formation. If the mined repository is not in a salt formation, such as crystalline rock and volcanic tuff, the salt waste would be less compatible with the environment. In non-salt formations, it would be beneficial for the internal environment to remain dry and the waste package to be robust to maintain confinement integrity. It may also be required to separate these salt waste-containing waste packages from others. The performance of the engineered barrier system would need to be assessed and verified with performance assessment analyses for scenarios in which enclosed salts are released from the waste packages.

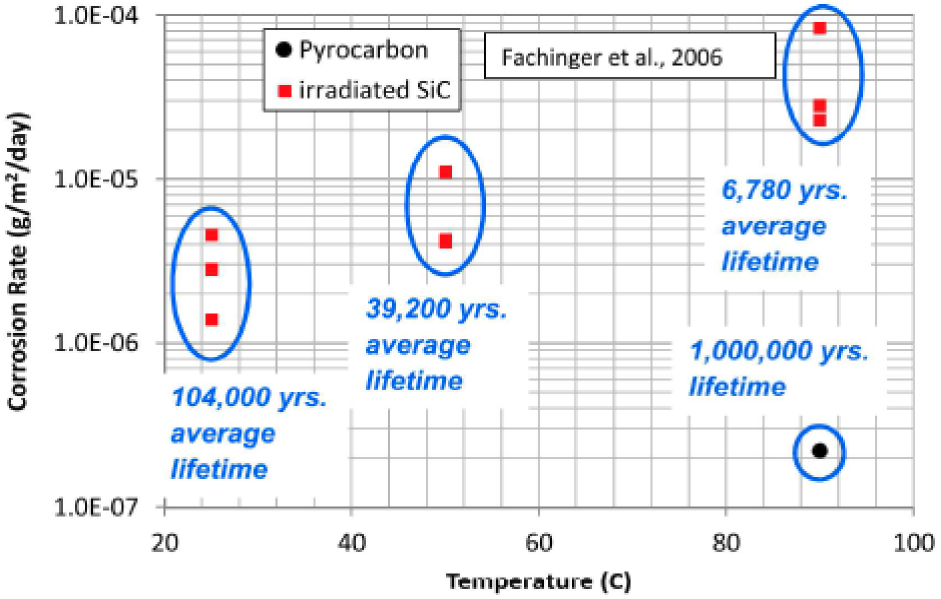


Figure 3-1. Estimated SiC layer lifetimes by corrosion of SiC at various temperatures for different brines (Gelbard and Sassani, 2018)

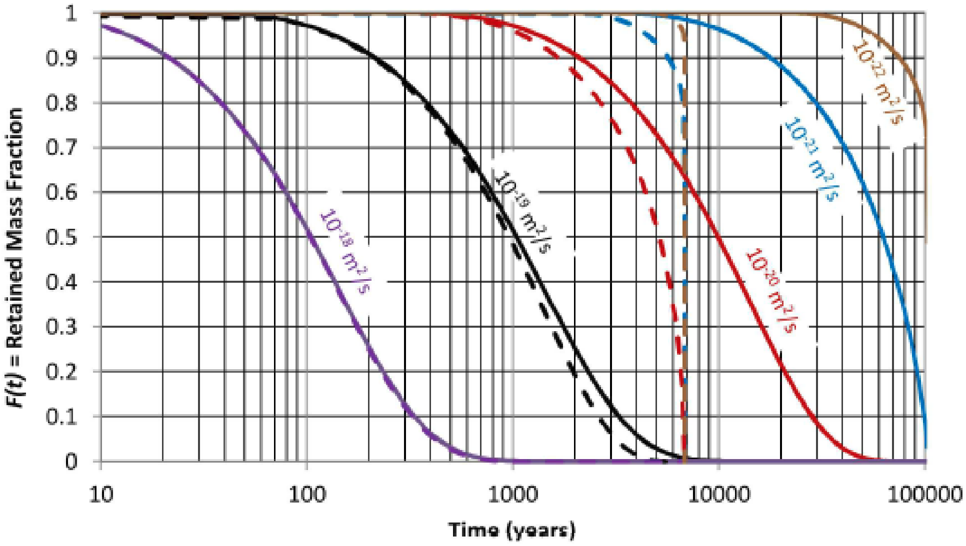


Figure 3-2. Fraction of fission product mass remaining within the TRISO fuel as a function of the diffusivity and corrosion rate of the SiC layer. Solid lines and dashed lines correspond to corrosion lifetimes of 100,000 years and 7,000 years, respectively. (Gelbard and Sassani, 2018)

3.3.2 Ceramic High-Level Waste Form

The degradation mechanisms of ceramic HLW forms depend on the environmental moisture content and the radiation level. If the environment is dry, the only possible degradation process would be radiation embrittlement. If wet, the HLW form can exhibit preferential dissolution of some components, release of occluded salt from sodalite, and hydrolysis. Any of these degradation mechanisms may result in the release of radionuclides from the waste form in a repository. Glass and some materials in the ceramic are hygroscopic and will absorb water vapor even at a very low relative humidity. The initial water uptake would start from dissolution of halite inclusion phases exposed at the surface. Because salt is diffused into the zeolite cages during the loading process, it is expected that salt can diffuse out of the cages when the salt-loaded zeolite is contacted by water vapor, although it should be at a much slower rate than salt released from salt waste form (Section 3.3.1). Figure 3-3 shows images of the surface and cross section of a ceramic HLW sample reacted with humid air at 150 °C [302 °F] for 105 days (Ebert, 2005). The glass binding the sodalite dissolved preferentially and many pits formed in the binder glass. The release rate depends on the temperature, composition of the water solution, the type of zeolite, and the salt loading process. The release of halites would generate chloride solutions. The deliquescent nature of these chloride-rich solutions can lead to water accumulation if the waste package is breached and, subsequently, to waste form dissolution and radionuclide release.

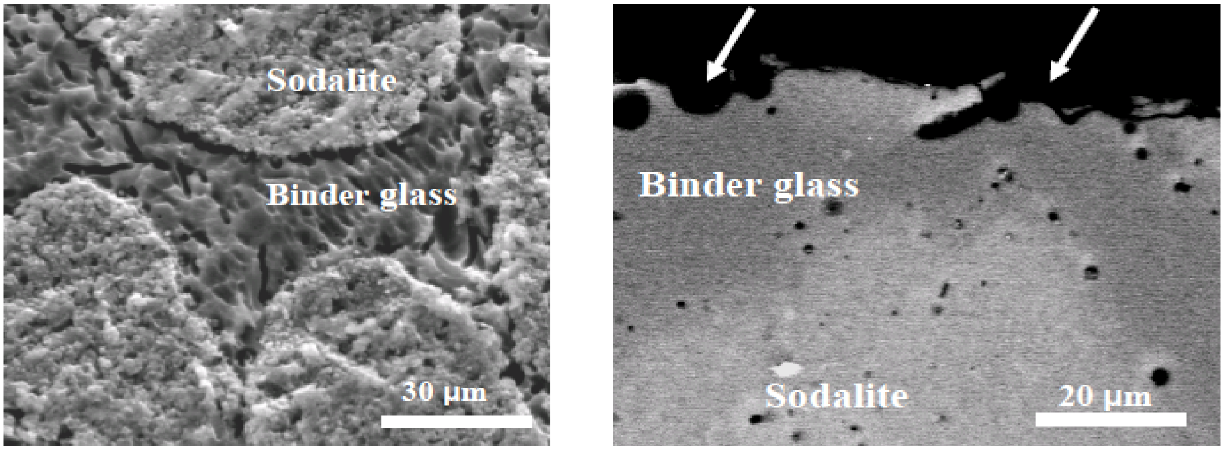
Ceramic HLW form can dissolve more if immersed in water than if exposed to water vapor. Figure 3-4 shows photomicrographs of the microstructures of U and Pu-doped hot isostatic press and pressure-less consolidated ceramic HLW forms after an immersion test in 110 ppm radionuclides from the ceramic HLW form is controlled by the dissolution rate of the glass binding the sodalite. Separate from HLW glass dissolution, the release of salt in contact with H_4SiO_4 solution at 90 °C [194 °F] (Ebert, 2005). Both ceramics showed signs of dissolution, but the dissolution process differed between the two types. The ceramic produced by hot isostatic press showed preferential dissolution at the boundaries between sodalite and glass, and the ceramic produced by pressure-less consolidation showed more uniform dissolution at the surface (Ebert, 2005). Furthermore, as shown in Figure 3-5 (Ebert, 2005), the release of moisture or water is expected to be another process that affects the performance of the ceramic HLW form in a disposal system. The relative effects of these two processes would need to be assessed further.

Bourcier and Sickafus (2004) showed that radiation, especially alpha decay, can degrade ceramic waste forms. Radiation damage could lead to swelling and cracking of the material, with potential volume changes of 15 percent or more. The accumulated radiation damage may break the ceramic along grain boundaries and increase the exposed surface area, ultimately enhancing radionuclide release. Wald and Weber (1984) and Weber et al. (1998) showed that materials damaged by alpha decay corroded ten to a hundred times faster than undamaged materials. However, the magnitude of the effects of radiation damage determined from laboratory tests may not be representative of actual ceramic waste forms, because of the contrasts between laboratory-induced and long-term radiation-induced damage mechanisms. The extent of ceramic HLW form degradation from radiation would need to be assessed further for an actual disposal environment.

3.3.3 Metallic High-Level Waste Form

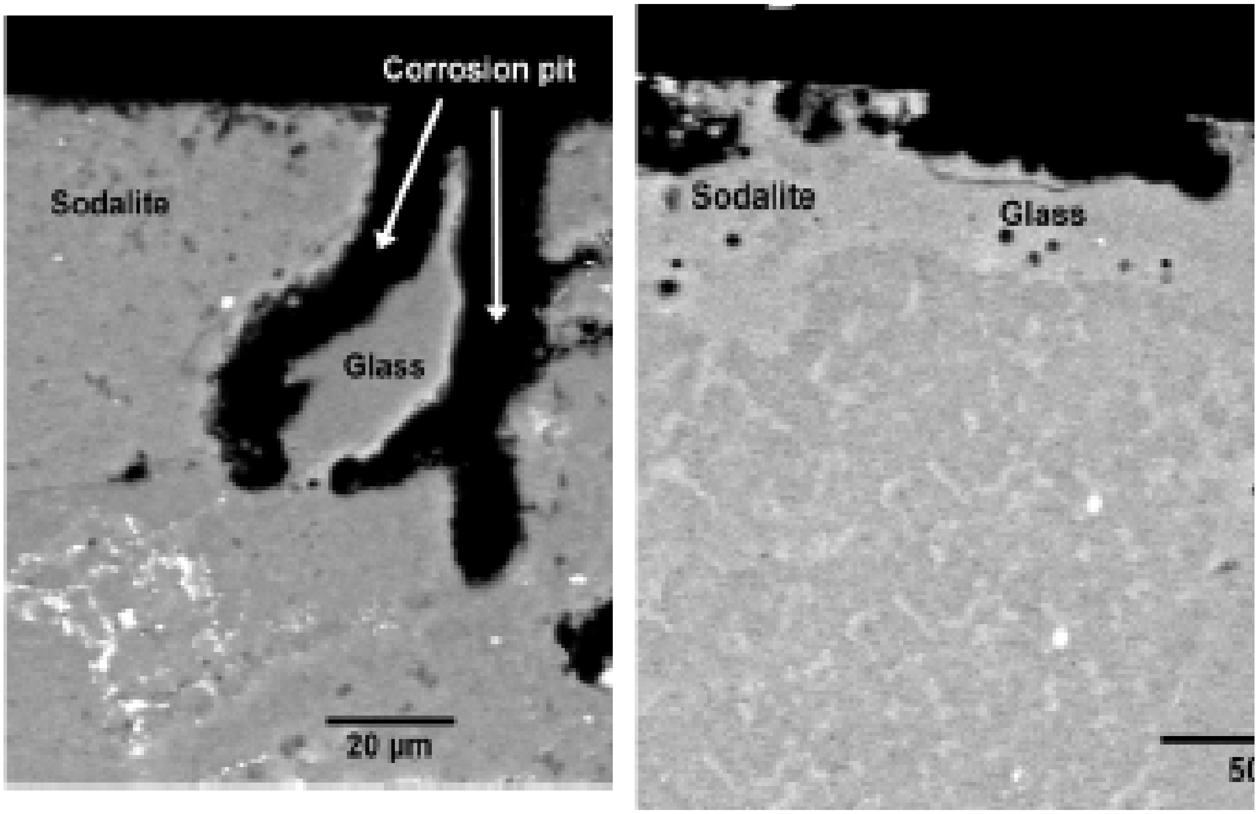
Similar to the ceramic HLW forms, the degradation mechanisms of metallic HLW forms depend on the internal environment and the radiation level. For a disposal system, if most of the air and moisture is evacuated from the container, the only possible dominant degradation process would be radiation embrittlement. In the presence of air and moisture, there are three main degradation mechanisms for metallic HLW form:

i. Oxidation/general corrosion
ii. Localized corrosion (pitting and crevice corrosion)
iii. Galvanic corrosion



(a) (b)

Figure 3-3. Photomicrographs showing (a) surface and (b) cross-section of ceramic HLW form reacted with humid air for 105 days at 150 °C [302°F] (Ebert, 2005)



(c)

(d)

Figure 3-4. **Photomicrographs showing the structures U and Pu-doped (a) hot isostatic press and (b) pressure-less consolidation ceramic HLW forms after immersion test in 110 ppm H_4SiO_4 solution at 90°C [194 °F] (Ebert, 2005)**

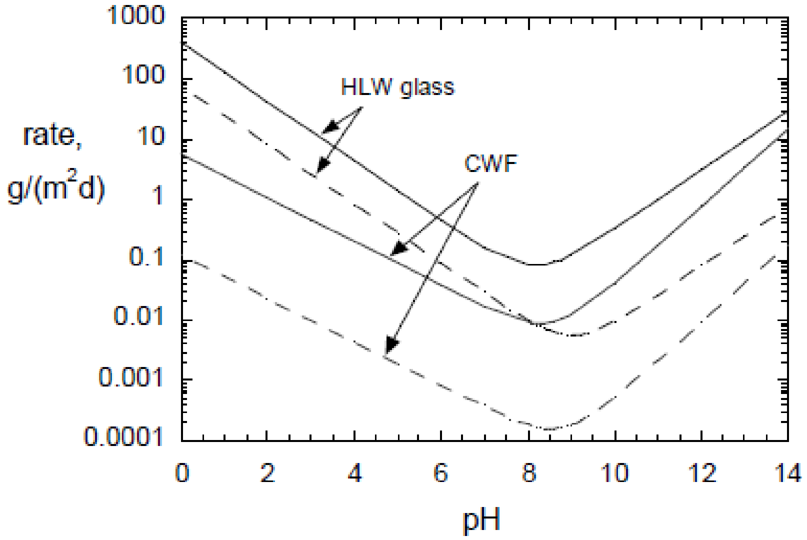
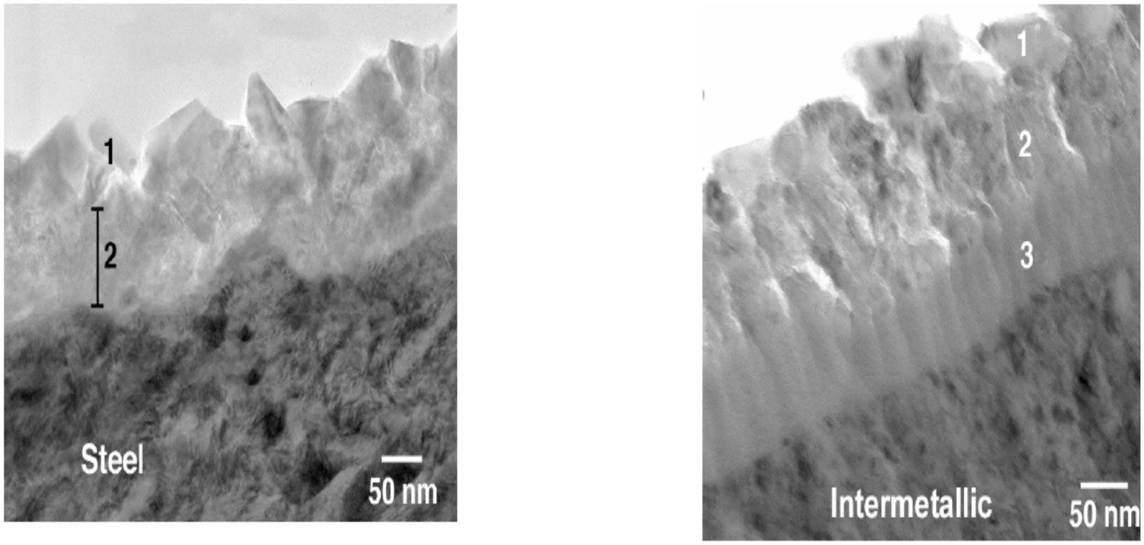


Figure 3-5. **Dissolution rates for ceramic HLW form compared to HLW glass (Ebert, 2005)**



(a)

(b)

Figure 3-6. **Transmission electron microscope photomicrographs of (a) two labeled oxide layers with different morphologies over stainless steel phase and (b) three labeled oxide layers with different morphologies over intermetallic phase of metallic HLW form reacted at 200 °C [392 °F] for 91 days in water vapor (Ebert, 2005)**

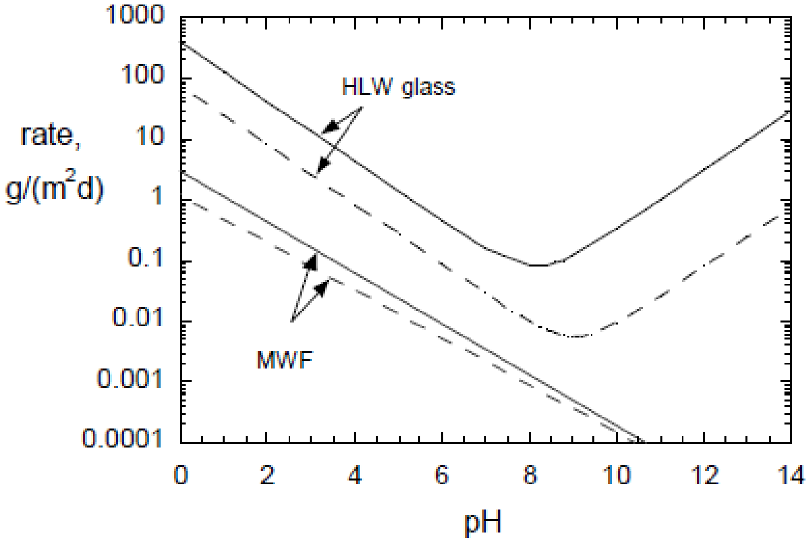


Figure 3-7. **Dissolution rates for metallic HLW form and high-level waste glass (Ebert, 2005)**

Similar to regular stainless steel and other passive metals, the metallic HLW form can be oxidized in air or water, forming an oxide layer on the surface. Radiolysis could result in a more oxidizing environment by producing radiolytic oxidants, such as H_2O_2. Figure 3-6 shows transmission electron microscope photomicrographs of the cross section of one metallic HLW form reacted at 200 °C [392 °F] for 91 days in water vapor (Ebert, 2005). Oxide layers are clearly visible over the stainless steel and intermetallic phases. General corrosion and release of radionuclides are expected to proceed through the dissolution of these outer oxide layers. Figure 3-7 shows that the dissolution rates for metallic HLW form are pH dependent and a couple of orders of magnitude lower than rates for HLW glass (Ebert, 2005). Ebert (2005) showed that U releases preferentially to Tc under most conditions and radionuclide release is bounded by HLW glass. Radiolytic effects on radionuclide release from metallic HLW forms would need further consideration.

Similar to regular stainless steel, localized corrosion of metallic HLW could occur if chloride is present in the disposal environment. The occurrence and extent of degradation would depend on composition of the metallic HLW, temperature, and chloride concentration. Ebert (2005) showed that metallic HLW can be nobler than Alloy 22 waste package material. If the waste form is in contact with different metals such as carbon steel, galvanic corrosion is possible. The likelihood of occurrence and magnitude of this degradation mechanism would depend on the metal that the metallic HLW contacts, the temperature of the canister environment, and the presence and quantity of water.

3.3.4 Spent Blanket Metal Fuel

Similar to other HLW forms, the degradation mechanisms of spent blanket metal fuel depend on the internal environment. If air and moisture are evacuated from the container, degradation of the fuel is not expected to occur. If the internal environment remains ambient with air and moisture at closure, some corrosion processes would occur. General corrosion and hydriding can be the predominant degradation mechanisms.

Similar to some passive metals, uranium forms a surface oxide film under exposure to air or water. The uranium oxides, the presence and concentration of O_2, and alloy elements play a large role in corrosion resistance. General corrosion is slower in the presence of O_2 because of the formation of a protective oxide film. For example, at 50 °C [120 °F], the corrosion rate of unalloyed uranium is about 300 µm/yr [12 mils/yr] under exposure to water saturated with H_2, but is about 8 µm/yr [0.3 mil/yr] in water saturated with air (Lillard and Hanrahan, 2005).

The general corrosion rate and corrosion products of uranium also depend on solution pH, with higher corrosion rates at lower pH. In addition, the corrosion rate increases with temperature and humidity. The blanket metal fuel is usually a uranium alloy, such as the U-10Mo alloy for Fermi-1 fuel. The alloy elements such as titanium, niobium, molybdenum, zirconium, and vanadium are reported to decrease the corrosion rate by enhancing passivity (Lillard and Hanrahan, 2005).

In the absence of O_2, uranium reacts with water, forming uranium oxides and hydrogen. Some of the hydrogen will be accumulated in the system as H_2 gas and some will be absorbed into the metal, forming uranium hydrides such as UH_3 (Lillard and Hanrahan, 2005). Uranium hydrides are pyrophoric materials, which means they can spontaneously ignite in the presence of air. Uranium hydrides also react with water under anoxic conditions to generate hydrogen gas. Pyrophoricity of spent nuclear metal fuel stored under poor conditions has been observed and reported (Guenther et al., 1996). After repository closure, a pyrophoric event could produce an adverse effect on repository performance by producing heat and increasing waste form degradation and radionuclide release rates. As such, it would be necessary to ensure that the disposal system for spent metal fuel is dried to certain level and backfilled with dry and inert gas to mitigate corrosion.

4 SUMMARY AND CONCLUSIONS

To date there is no domestic or international disposal experience for irradiated ARF and associated waste forms; however, repository performance has been well characterized for a variety of geologic formations including salt, crystalline rock, or clay/shale. Additional characterization performed for waste packages, including waste forms, canister materials, and other engineered features, can provide insights into specific waste package performance in disposal environments.

Chemical and physical characteristics of spent coated particle fuel discharged from advanced reactors in both pebble-bed and prismatic block designs are advantageous for disposal in a geologic repository due to the multiple coating layers on the fuel particles, which provide better containment than LWR fuel. Spent coated particle fuels are expected to degrade more slowly than LWR fuels in oxidizing and reducing repository settings, primarily due to the protective SiC layer (which is an additional barrier to water intrusion). Possible options for disposal of spent coated particle fuel include direct disposal and removal of graphite from the spent prismatic block fuel, allowing graphite to be disposed as Class C LLW. For solid coated particle fuel, estimates of the time to failure in a repository environment are highly dependent on the thickness, strength, and post-irradiated condition of the SiC and OPyC layers. Fission product diffusivities and corrosion rates are important factors to characterize the performance of solid coated particle fuel in hypothetical disposal environments. The longevity of solid coated particle fuel is dependent on corrosion rates for the OPyC and SiC layers, other TRISO fuel parameters, and environmental factors such as temperature. Bulk diffusivities in SiC of the radionuclides of interest at repository temperatures are not available in the literature. Quantitative evaluation of diffusion in the compact, porous graphite matrix and the TRISO coating layers at repository conditions would provide important insights into disposal performance for spent ARF.

Direct disposal of spent driver metal fuel is not a feasible option due to the reactive nature of the metallic sodium fused into the fuel. Chemical treatment of sodium bonded driver fuel recovers the highly enriched uranium and converts the remaining material into less chemically reactive forms such as salt waste, ceramic waste, and metallic waste. These waste forms would be packaged in containers, which are filled with inert gas and placed within a sealed over-pack prior to disposal in a deep geologic repository or bore hole. These waste forms converted from spent nuclear metal fuel have been characterized to understand degradation mechanisms in disposal environments. DOE research has concluded that ceramic and metallic waste forms converted from spent metal fuel are acceptable for repository disposal (Ebert 2005). For these waste forms, radionuclide release rates would depend on widely variable repository and fuel conditions, which include temperature, fluid chemistry, and radiation level.

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