Draft Experimental Plan for Commercial SNF Degradation in Repository Environments with a Focus on Fuel Matrix Degradation

Spent Fuel and Waste Disposition

Prepared for
US Department of Energy
Spent Fuel and Waste Science and Technology

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SUMMARY

This report documents work performed supporting US Department of Energy (DOE) Nuclear Energy Spent Fuel and Waste Disposition, Spent Fuel and Waste Science and Technology, under work breakdown structure element 1.08.01.03.04, "Inventory and Waste Form Characteristics and Performance." In particular, this report fulfills the M3 milestone, M3SF-21OR010309072 "Draft Experimental Plan for Commercial SNF Degradation in Repository Environments" as described in work package SF-21OR01030907 "SNF Degradation Testing- ORNL".

This experimental plan is a collaboration between Oak Ridge, Sandia, Argonne, Los Alamos, and Pacific Northwest national laboratories. It is intended to support source-term model development and testing of spent fuel degradation in a range of generic repository conditions. Potential testing and experiments to support validation of the Fuel Matrix Degradation Model (FMDM) and the overall development/ validation of repository source-term modeling are discussed.

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CONTENTS

LIST OF FIGURES

LIST OF TABLES

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ACRONYMS

ANL	Argonne National Laboratory
ASME	American Society of Mechanical Engineers
ASTM	American Society for Testing and Materials
ATF	accident-tolerant fuel
BSS	borated stainless steel
BWR	boiling water reactor
CoC	NRC Certificate of Compliance
CFR	US Code of Federal Regulations
CSNF	commercial spent nuclear fuel
DCRA	disposal control rod assembly
DOE	US Department of Energy
DPC	dual-purpose canister
DSS	dry storage system
EBS	engineered barrier system
EPRI	Electric Power Research Institute
FEPs	features, events, or processes
FMD	fuel matrix degradation
FMDM	fuel matrix degradation model
GDSA	geologic disposal safety assessment
HBS	high burnup structure
HLW	high-level radioactive waste
IRF	instantaneous release fraction
ISG	interim staff guidance
ISO	International Organization for Standardization
LANL	Los Alamos National Laboratory
MIC	microbially influenced corrosion
MOX	mixed oxide [fuel]
NAS	National Academy of Sciences
NCT	normal conditions of transport
NE	DOE Office of Nuclear Energy
NEI	Nuclear Energy Institute
NEUP/IRP	Nuclear Energy University Program / Integrated Research Program
NQA	Nuclear Quality Assurance
NRC	US Nuclear Regulatory Commission
ORNL	Oak Ridge National Laboratory
PCSA	preclosure safety analysis
PNNL	Pacific Northwest National Laboratory
PWR	pressurized water reactor
R&D	research and development
RCCA	rod cluster control assembly
RG	regulatory guide
SALVI	system for analysis at the liquid vacuum interface
SCC	stress corrosion cracking
SER	NRC Safety Evaluation Report
SFST	spent fuel storage and transportation
SFWD	DOE Office of Spent Fuel and Waste Disposition (NE-8)
SFWST	DOE Office of Spent Fuel and Waste Science and Technology (NE-81)

SKB	Swedish Nuclear Fuel and Waste Management Company
SNF	spent nuclear fuel
SNFA	spent nuclear fuel assembly
SNL	Sandia National Laboratories
SPFT	single pass flowthrough
SRP	standard review plan
SSC	structures, systems, and components
SS	stainless steel
TSPA	total system [postclosure] performance assessment
UNS	unified numbering system (for metal alloys)
WF	waste form
WP	waste package
YM	Yucca Mountain
YMP	Yucca Mountain Project

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ABBREVIATIONS AND MEASUREMENT UNITS

°C	degrees centigrade
°F	degrees Fahrenheit
Amd	amendment (to an International Organization for Standardization [ISO] standard)
cm^2	square centimeter
Cor	corrigendum (to an ISO standard)
dpm	disintegrations per minute
g	acceleration due to gravity
GWd	gigawatt-day
in.	inch
kg	kilogram
lb	pound
m	meter
MT	metric tons
MTIHM	metric tons of initial heavy metal
MTU	metric tons of uranium
Rev	Revision
W	watt
wt%	weight percent

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1 Purpose

This document describes an experimental plan for evaluating commercial spent nuclear fuel (SNF) degradation in a range of repository disposal environments. The test program will produce data for validation of commercial spent nuclear fuel (CSNF) degradation models for geologic disposal safety assessments (GDSAs) that are used to evaluate the performance of nuclear waste disposal systems in geologic media. The program will also produce data to evaluate post-closure SNF behavior and to further constrain the source term. The strategic prioritization of testing/experimental work provides the Spent Fuel Waste Disposition (SFWD) program with a foundation for staged progression of data collection. The full range of testing will likely include a combination of unirradiated and irradiated materials.

2 Scope

The DOE SFWD research and development (R&D) roadmap (Sevougian et al., 2019) identifies SNF degradation testing as a medium-high priority R&D activity. This work includes electrochemical experiments and test plan development for collection of SNF degradation model validation data, as well as testing related to other aspects of SNF performance in the post-closure environment. The experimental plan established in this document will be comprehensive and will support the technical bases for GDSAs associated with generic repository environments. The highest priority tests and experiments are designed to primarily validate the Fuel Matrix Degradation Model (FMDM) consistent with the priorities for source-term development noted by Sevougian et al. (2019). The FMDM predicts the degradation rate of SNF. This plan also includes data collection strategies for evaluation of previously established SNF features, events, or processes (FEPs) for further testing of source-term processes. For example, FEP 2.1.02.06, SNF Cladding Degradation and Failure, and FEP 2.1.02.01, SNF Degradation -Commercial, may be addressed through experiments designed to evaluate the following aspects of SNF degradation:

- Cladding degradation
- Exposed surface area of SNF
- Alteration/Phase separation
- Radionuclide release

Additionally, FEP 2.1.09.52, Diffusion of Dissolved Radionuclides in Engineered Barrier Systems (EBSs), is also supported in part by the overall experimental program.

This draft experimental plan describes the proposed work, as well as ongoing tests, at various national laboratories within the scope of the work described above. There is considerable flexibility in the priority of the sequence, quantity, and duration of testing based on the maturity level of the SNF degradation models, overall disposal R&D activities, and the variety of existing techniques at the various national laboratories participating in the Spent Fuel and Waste Science and Technology (SFWST) campaign. The testing program will evolve to support changing priorities throughout the SFWST R&D campaign. As such, the experimental plan will be reviewed periodically and updated as appropriate.

3 Background

3.1 General Description of the Light Water Reactor Spent Fuel System

Nearly all existing commercial SNF is composed of UO_2 ceramic pellets inserted into zirconium-based alloy cladding tubes. The tubes filled with UO_2 are arranged in a square array, and the structure of the fuel assembly includes upper and lower nozzles connected by guide tubes. Although a small fraction of the older SNF assemblies include stainless-steel rod cladding, the predominant cladding materials are Zircaloy-4, Zircaloy-2 (usually includes a zirconium sponge liner), ZIRLO and Optimized ZIRLO (proprietary Zircaloy-based alloys developed by Westinghouse), and M5 (a proprietary Zircaloy-based alloy developed by Framatome). Figure 1 shows a typical pressurized water reactor (PWR) fuel assembly. The different reactor types in the United States and the evolution in fuel assembly designs and reactor operating conditions have led to some variation in the characteristics (e.g., assembly and cladding materials, assembly structure, initial enrichment, discharge burnup, burnable poison types, and irradiation exposure conditions) of the current SNF inventory. Variation in these parameters may impact cladding corrosion products, distribution/ concentration of transuranic radionuclides and fission products in SNF fuel pellets, or structural changes in SNF fuel pellets. These variations can impact overall fuel dissolution rate in a hypothetical repository significantly (See Section 3.3.4 for a description of key mechanisms impacting fuel dissolution rate).

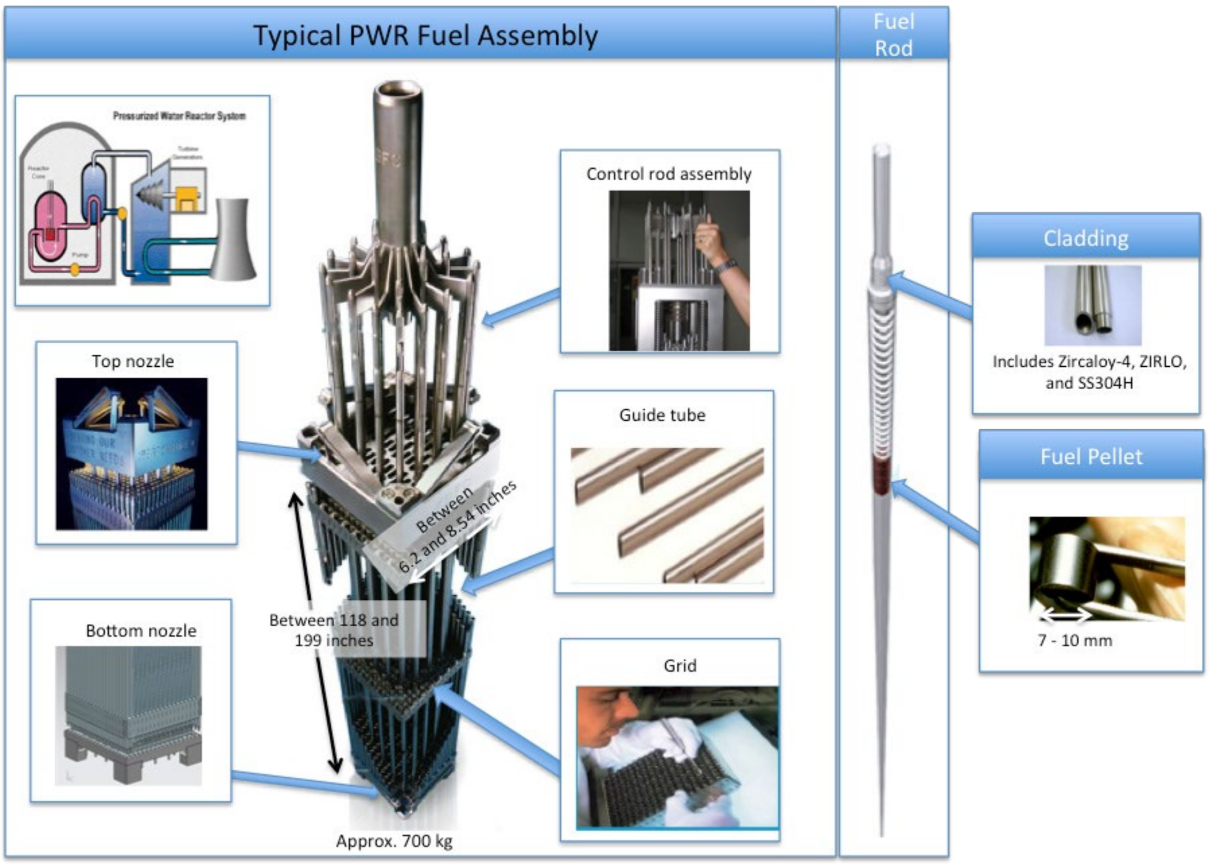

Figure 1. Typical PWR fuel assembly (Wagner et al., 2012).

In addition, accident-tolerant fuel (ATF) and cladding designs are being investigated for eventual use in commercial reactors. ATF and cladding designs currently being irradiated or actively analyzed for batch fuel production include the following:

- Chromia/alumina doped low-enriched uranium UO_2 fuel pellets
- High assay low-enriched uranium (HALEU) fuel enriched between 5 and 20 wt% ^{235}U
- Chromium-coated cladding
- FeCrAl cladding

While these designs use the current commercial fuel configuration, material changes may influence their performance in a repository environment. The current experiment plan nominally considers ATF designs, and future revisions of the plan should further incorporate experiments to address any knowledge gaps related to the ATF designs as they are moved into batch production.

3.2 Possible Repository Environment Conditions

This experiment plan proposes tests to evaluate SNF performance in a range of repository conditions, given that associated degradation will vary widely based on repository physical and chemical characteristics. Table 1 summarizes the general characteristics of repository media that remain under consideration as GDSA reference cases (Mariner et al., 2015; 2016; 2017)

Table 1. Repository Media Characteristics*

Repository Media	General media characteristics	Groundwater Redox potential	Groundwater pH	Repository Backfill/Buffer
Clay/Shale	Low permeability, plastic deformation	Reducing	Neutral to slightly alkaline	Bentonite, crushed rock, swelling clay
Saturated Crystalline	Low permeability, brittle, high structural strength	Oxidizing or reducing, though reducing conditions preferred.	Neutral	Crushed rock and bentonite
Unsaturated Crystalline	Selected for stability	Oxidizing	Depends on source of water flowing through the formation, though generally neutral	No planned backfill
Salt	Low permeability, visco-plastic	Controlled by MgO and backfill material	Acidic to neutral	crushed salt

*Sources: Mariner et al 2015, 2016, 2017; Rechard et al., 2011; U. S. DOE, 2008; Miller, 2002.

3.3 GDSA Model Framework

As noted above, commercial SNF degradation modeling encompasses cladding degradation, instantaneous release fraction (IRF), dissolution of the UO_2 ceramic pellets, and diffusion of dissolved radionuclides in the EBS. This model is integrated into the GDSA framework (Figure 2), which is used to evaluate disposal system performance of nuclear waste in geologic media such as those listed in Section 3.2 above. The remainder of this section provides a brief summary of the state of knowledge associated with key fuel degradation mechanisms- namely waste package (WP) degradation, cladding degradation, IRF, and fuel dissolution.

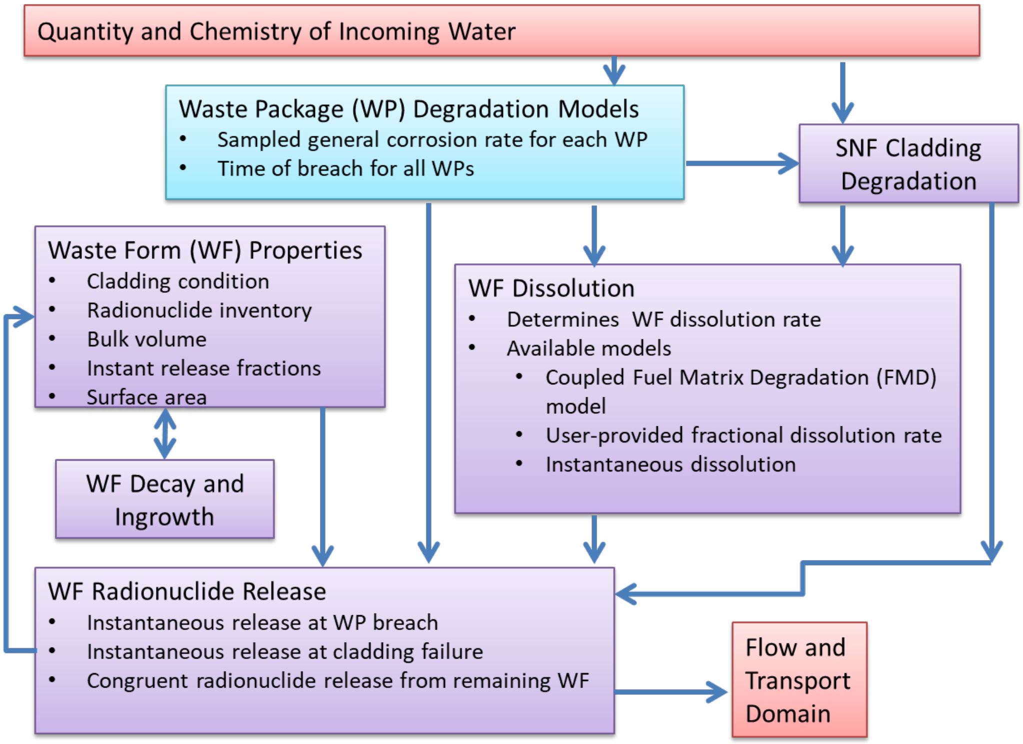

Figure 2. Conceptual framework for source-term processes in GDSA (Adapted from Mariner et al., 2019).

3.3.1 Waste Package Degradation and Breach

Although performance predictions of degradation and breach of the WP are important for assessment of the overall system in a repository, this experimental plan is focused on the SNF interaction with the repository environment after package breach occurs. Currently, the proposed suite of experiments described herein does not propose to actively study the effects of the repository environment on the WP. However, existing WP degradation work that could impact fuel degradation (e.g., WP corrosion leading to hydrogen production) may be leveraged when designing experiments.

3.3.2 Cladding Degradation Processes

The main functions of the SNF cladding are to serve as primary containment and to prevent water or moist air from contacting the fuel pellets. At manufacture, the fuel rod cladding is completely sealed. A small fraction of SNF may exhibit operational cladding failures that were identified once assemblies were removed from the reactor core. The majority of the SNF is expected to be sealed when it reaches the repository, and SNF having failed cladding is expected to be treated differently for the purposes of repository emplacement. Additional failures are not expected due to interim dry storage or transportation (Montgomery & Bevard, 2020).

The potential role of fuel cladding in hermetically isolating the fuel from the environment is addressed in FEP 2.1.02.06.

In the repository, SNF cladding may degrade as a result of oxidation by residual water in the disposal package. After an initial cladding breach, water intrusion into the fuel rod will result in fuel oxidation, pellet swelling, and additional significant cladding breach. Cladding may also degrade as a result of several external corrosion processes, including general corrosion, microbially-influenced corrosion (MIC), and localized corrosion (including pitting and crevice corrosion), though the likelihood that these mechanisms result in cladding failure may vary (Brady & Hanson, 2020).

3.3.3 Instantaneous Release Fraction (IRF)

Waste form (WF) degradation and radionuclide release in a hypothetical repository occurs after failure of the WP and cladding. The radionuclide release rate includes the combined effect of two phenomena: the short-term release of mobile or soluble isotopes and the longer-term dissolution of less mobile isotopes, along with the uranium dioxide matrix. The latter phenomenon is discussed in Section 3.3.4 and is the primary focus of this draft experimental plan.

The IRF is the rapid release of volatile fission products from accessible grain boundaries and the gap region between the fuel pellets and cladding. In a hypothetical repository, the instantaneous release occurs shortly after fuel rod cladding breach and associated groundwater intrusion. In general, it is difficult to delineate between accessible and inaccessible grain boundaries for the purposes of quantifying IRF. Therefore, IRF models often assume that the entire inventory of certain radionuclides is released instantaneously upon fuel cladding breach (Johnson, Ferry, Poinssot, & Lovera, 2005; Bechtel SAIC Company, LLC, 2004).

Previous reports (De Pablo et al., 2008 and 2009; Serrano-Purroy et al., 2012; Johnson et al., 2012) suggest that SNF pellet structural changes associated with high-burnup fuel may affect the availability of volatile fission products for release as part of the IRF. For instance, previous work has evaluated the impact of the high burnup structure (HBS) on IRF, showing that HBS has a finer-grained, higher closed-porosity layer containing fission gases. This layer has been observed on the outermost portion of high burnup fuel pellets. Studies (De Pablo et al., 2008 and 2009; Serrano-Purroy et al., 2012) concluded that HBS samples release volatile fission product inventory more slowly than other samples and one study (Johnson et al., 2012) indicated that the HBS may provide some protection against IRF for radionuclides contained within the HBS ring, despite having a higher specific surface area of exposure.

3.3.4 UO_2 Dissolution Rate and SNF Alteration

Longer-term radionuclide release following WP/cladding failure is controlled by the rate of UO_2 dissolution. Available data suggest that the SNF degradation rate under anoxic or reducing conditions will be far slower than that expected under oxidizing disposal conditions (Shoesmith, 2007). The UO_2 dissolution rate is complex and is dependent on multiple repository environmental conditions such as groundwater chemistry, repository temperatures, and SNF activity. Repository environmental conditions can widely affect redox conditions at the surface of the fuel: for example, oxidizing species at the fuel surface are strongly influenced by SNF radioactivity and alpha particle-induced radiolysis of groundwater (Jerden Jr., Frey, & Ebert, 2015). Dissolved hydrogen in the groundwater is heavily influenced by WP corrosion and has been observed to greatly reduce SNF dissolution, mainly due to reduction of oxidized UO_2 (Shukla et al., 2015). This reduction may be catalyzed via nanometer-size clusters of noble-metal fission products called *ε-particles*. Finally, various groundwater constituents can either inhibit, promote, or not affect the UO_2 dissolution rate due to their influence on radiolytic products such as H_2O_2 that enable the oxidative dissolution of UO_2 (Amme, 2002).

Efforts to model the UO_2 dissolution rate in the conceptual framework shown in Figure 2 above have centered on the FMDM (Jerden, Thomas, Lee, Gattu, & Ebert, 2020) which is summarized in Figure 3, below.

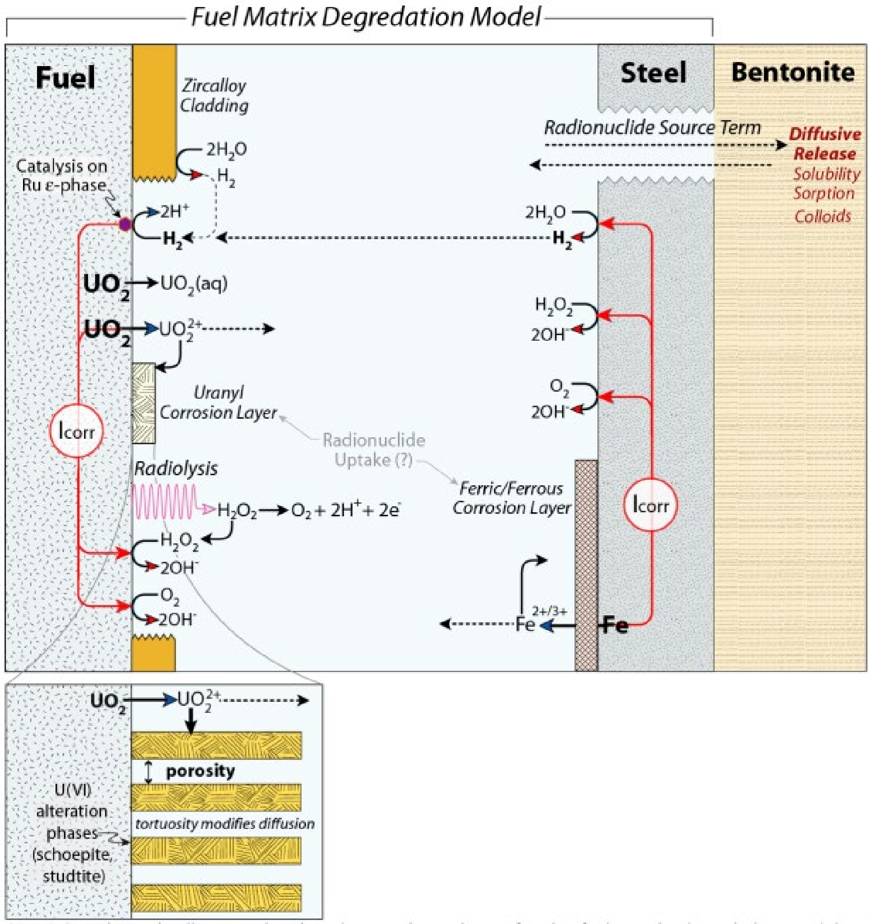

Figure 3. Schematic highlighting basic interactions captured in the FMDM (Jerden, Lee, Kumar Gattu, & Ebert, 2019).

The FMDM employs fundamental redox kinetics and thermodynamics to predict fuel dissolution rate as a function of multiple variables. Specifically, it addresses key dissolution phenomena such as WP corrosion-induced hydrogen production, alpha radiolysis/radiolytic oxidant generation, corrosion phase growth at the surface of the fuel, temperature variations of reaction rates, and bulk solution reactions (e.g., oxidation of ferrous iron by oxygen and radiolytic hydrogen peroxide). As noted in Section 2, above, priority in this draft experimental plan is given to those tests that validate/expand upon the FMDM. Experiments in this category of priority should be designed to complement the FMDM; for instance, experiments should focus on the impact of alpha irradiation on the production of radiolytic oxidants.

4 Proposed Test Plans

This draft plan serves to identify national laboratory capabilities and proposed experiments. The highest priority testing and experiments are those which validate/expand upon the FMDM. Additional testing activities support SNF behavior more broadly, including modeling of cladding degradation, IRF, and diffusion of dissolved radionuclides into the EBS. Proposed experiments are detailed in Appendix A. Ongoing experiments that fall within this scope of work are also described in Appendix A. To the extent that the information is currently known, Appendix A includes testing scope, relation to GDSA, and testing duration. As plans mature, the experiments proposed in Appendix A will be prioritized, and this document will be updated accordingly.

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Appendix A

Table A-1 outlines ongoing experimental work that serves to validate the FMDM and to capture SNF behavior for the purposes of GDSA. The next two tables outline proposed experimental work intended to further evaluate/validate the FMDM (Table A-2) and to capture SNF behavior for the purposes of GDSA (Table A-3).

As noted in Section 2.0 above, the testing program will evolve to support the changing priorities throughout the SFWST R&D campaign. Although each individual test does not have a specific priority, proposed work to validate the FMDM shown in Table A-2 should be prioritized over the proposed work in Table A-3, consistent with priorities included in the DOE SFWST Campaign R&D roadmap (Sevougian et al., 2019). Because knowledge of proposed test duration and rough order of magnitude (ROM) cost is limited, associated columns are included here as placeholders pending future planning discussions. This experimental plan will be reviewed periodically and updated as appropriate.

Table A-1. Ongoing Experimental Work

	GDSA Relevance	Experimental Work	Status of work	Laboratory
1	Study the impact of noble metal catalysts on UO_2 degradation to update FMDM-proof of concept	Electrochemical tests with simulated fuel samples (UO_2 + lanthanides; UO_2 +lanthanides + noble metals)	Ongoing: planned completion in FY2022	Argonne National Laboratory (ANL)
2	Determine hydrogen generation rates for inclusion in FMDM	Evaluate hydrogen generation rate from corrosion of waste package alloys	Ongoing: planned completion in FY2023	ANL
3	Develop reliable thermodynamic experimental data at elevated temperatures	Evaluate uranium speciation in aqueous solutions at high temperatures	2018-ongoing	Los Alamos National Laboratory (LANL)
4	Study influence of peroxide, hydrogen concentration on corrosion potential for the purposes of FMDM validation	UO_2 microchemical experiments using the system for analysis at the liquid vacuum interface (SALVI) cell	2018-ongoing	Pacific Northwest National Laboratory (PNNL)
5	Study simulated fuel dissolution in oxidizing conditions to populate the FMDM	Development of single pass flowthrough (SPFT) system for use with simulated fuel[1]	SPFT system built and testing started	PNNL

1. Admussen & Hanson, 2020.

Table A-2. Proposed Experimental Work Supporting FMDM validation/model development

	GDSA Relevance	Notes	Proposed Experimental Work	Proposed Test Duration	ROM Cost
1	Provide dissolution rate data at a range of repository conditions for FMDM validation	High burnup fuel may include pellet structural changes such as HBS	Study information gaps to refine test conditions. Corrosion tests of SNF at a range of repository conditions (e.g., vary parameters such as temperature, pH, oxidizing conditions) using high burnup fuel. Consider SIMFUEL control to evaluate differences in dissolution rate	Up to 2 years	TBD
2	Evaluate threshold concentrations of hydrogen needed to inhibit oxidative dissolution under short- and long-term repository breach conditions	Concentrations of radiolytic oxidants in high burnup fuels may change sensitivity of fuel degradation rate to H_2 concentration if WP breach occurs less than 1,000 years out of the reactor	Leverage existing WP breach modeling for corrosion potential/dissolution testing	TBD	TBD
3	Further evaluate mechanism by which H_2 inhibits oxidative dissolution	Phenomenon is currently modeled as a couple between UO_2 oxidation and catalytic oxidation of H_2 on ε-particles. Other potential processes to consider: destruction of radiolytic oxidants by H_2	Use of high-burnup fuel or fabricated RADFUEL/SIMFUEL to study other mechanisms by which H_2 inhibits the oxidative dissolution of SNF	TBD	TBD
4	Refine modeling of ε-particle as a catalyst for reduction of oxidized UO_2 in fuel matrix dissolution	It is possible that the FMDM overemphasizes the effect of the ε-particle on inhibition of fuel dissolution	Use of high-burnup fuel or fabricated RADFUEL/SIMFUEL to study effect of ε-particle on inhibition of fuel dissolution	TBD	TBD

	GDSA Relevance	Notes	Proposed Experimental Work	Proposed Test Duration	ROM Cost
5	Refine modeling of ε-particle as a catalyst for reduction of oxidized UO_2 in fuel matrix dissolution	Variations in ε-particle composition, distribution, and size have been observed in SNF[1]	Use of high-burnup fuel or fabricated RADFUEL/SIMFUEL to evaluate the impact of ε-particle variation on inhibition of oxidative fuel matrix dissolution	TBD	TBD
6	Update FMDM to include any processes that counteract the mechanism by which H_2 suppresses oxidative fuel degradation	Some evidence that halides may poison the catalytic properties of ε-particles, which could diminish the inhibiting effect of H_2	Electrochemical tests may be used to evaluate these counteractive processes	TBD	TBD
7	Update FMDM to include processes that consume H_2	Two key processes to consider: microbial redox effects and chemical conversion of H_2 to sulfur species	Experimentally evaluate these processes	TBD	TBD
8	Evaluate corrosion rates for galvanically coupled alloys (e.g., cladding)	N/A	Measure corrosion rates using zero resistance ammeter	TBD	TBD
10	Integrate dissolution data associated with novel LWR fuel forms into FMDM	Initial data suggest that MOX and ATF dissolution rates may differ from those observed for UO_2[2]	Measure degradation rates/corrosion potential of ATF and mixed oxide fuels (MOX)	TBD	TBD
11	Expand upon available and reliable thermodynamic data at elevated temperatures	N/A	Evaluate uranium speciation in aqueous solutions at high temperatures in conjunction with waste package/EBS materials	TBD	TBD

1. Cui et al., 2012 and Buck, Mausolf, McNamara, Soderquist, & Schwantes, 2015
2. Morris & Bauer, 2005 and Demkowicz et al., 2004

Table A-3. Proposed Experimental Work Supporting GDSA modeling of SNF Behavior

	GDSA Relevance	Notes	Proposed Experimental Work	Proposed Test Duration	ROM Cost
1	Proof-of-concept measurement of dissolution rates with high burnup SNF	Work may address impacts from pellet structural changes such as HBS for future integration into models such as the FMDM	Dynamic leach tests at a range of repository conditions (e.g., vary parameters such as temperature, pH, oxidizing conditions)	TBD	TBD
2	Proof-of-concept measurement of corrosion potential with high burnup SNF	Work may address impacts from pellet structural changes such as HBS for future integration into models such as the FMDM	Electrochemical tests	TBD	TBD
3	Diffusion of dissolved radionuclides through EBS	Could use high burnup fuel to address impacts from pellet structural changes such as HBS	Leach testing of SNF through engineered barrier system/near-field materials (e.g., bentonite clay)	Up to 2 years	TBD
4	Dissolution behavior at different points within the SNF pellet (e.g., pellet centerline, HBS)	Smaller samples may simplify handling requirements	Microscale leaching and electrochemical tests with high-burnup fuel samples	TBD	TBD
5	Refine assumptions and reduce conservatism associated with IRF release; accessible surface area	Sample preparation must be carefully considered. Consider using high burnup fuel	Measure the fraction of accessible and inaccessible grain boundary in SNF using imaging techniques (e.g., metallography)	TBD	TBD
6	Different sizes of cladding breaches may impact overall fuel dissolution rate and IRF	Cladding may be more robust than previously evaluated/assumed.	Evaluate fuel dissolution with various sizes of cladding breaches in anoxic/reducing conditions	TBD	TBD
7	Existing boundary conditions within FMDM may need to be reviewed to ensure they are sufficiently bounding for repository environments	This could also be done via analysis	Evaluate SNF dissolution behavior at conditions identified to be beyond what is currently assumed in the FMDM	TBD	TBD