SAND2020-6615R

Revision 5
01/15/2018

APPENDIX E
NTRD DOCUMENT COVER SHEET[1]

Name/Title of Deliverable/Milestone/Revision No.: Permanent Criticality Termination Processes in Disposed DPCs

Work Package Title and Number: Probabilistic Postclosure DPC Criticality Consequence Analyses, SF-20SN01030506

Work Package WBS Number: M4SF-20SN010305063

Responsible Work Package Manager: Laura Price Laura L Price
(Name/Signature)

Date Submitted November 22, 2019

Quality Rigor Level for Deliverable/Milestone[2]	☐ QRL-1 ☐ Nuclear Data	☐ QRL-2	☒ QRL-3	☐ QRL-4 Lab QA Program[3]

This deliverable was prepared in accordance with Sandia National Laboratories
(Participant/National Laboratory Name)

QA program which meets the requirements of
☒ DOE Order 414.1 ☐ NQA-1 ☐ Other

This Deliverable was subjected to:

☒ Technical Review	☐ Peer Review
Technical Review (TR)	**Peer Review (PR)**
Review Documentation Provided	**Review Documentation Provided**
☐ Signed TR Report or,	☐ Signed PR Report or,
☐ Signed TR Concurrence Sheet or,	☐ Signed PR Concurrence Sheet or,
☒ Signature of TR Reviewer(s) below	☐ Signature of PR Reviewer(s) below

Name and Signature of Reviewers
Laura Price Laura L Price

NOTE 1: *Appendix E should be filled out and submitted with the deliverable. Or, if the PICS:NE system permits, completely enter all applicable information in the PICS:NE Deliverable Form. The requirement is to ensure that all applicable information is entered either in the PICS:NE system or by using the NTRD Document Cover Sheet.*

- *In some cases there may be a milestone where an item is being fabricated, maintenance is being performed on a facility, or a document is being issued through a formal document control process where it specifically calls out a formal review of the document. In these cases, documentation (e.g., inspection report, maintenance request, work planning package documentation or the documented review of the issued document through the document control process) of the completion of the activity, along with the Document Cover Sheet, is sufficient to demonstrate achieving the milestone.*

NOTE 2: *If QRL 1, 2, or 3 is not assigned, then the QRL 4 box must be checked, and the work is understood to be performed using laboratory QA requirements. This includes any deliverable developed in conformance with the respective National Laboratory / Participant, DOE or NNSA-approved QA Program.*

NOTE 3: *If the lab has an NQA-1 program and the work to be conducted requires an NQA-1 program, then the QRL-1 box must be checked in the work Package and on the Appendix E cover sheet and the work must be performed in accordance with the Lab's NQA-1 program. The QRL-4 box should not be checked.*

Permanent Criticality Termination Processes in Disposed DPCs

Spent Fuel and Waste Disposition

Prepared for
U.S. Department of Energy
Spent Fuel and Waste Science and Technology

Abdelhalim Alsaed
Enviro Nuclear Services, LLC

June 12, 2020
M4SF-20SN010305063

CONTEXT FOR THIS STUDY

This is a technical paper that does not take into account contractual limitations or obligations under the Standard Contract for Disposal of Spent Nuclear Fuel and/or High-Level Radioactive Waste (Standard Contract) (10 CFR Part 961). For example, under the provisions of the Standard Contract, spent nuclear fuel in multi-assembly canisters is not an acceptable waste form, absent a mutually agreed to contract amendment.

To the extent discussions or recommendations in this paper conflict with the provisions of the Standard Contract, the Standard Contract governs the obligations of the parties, and this paper in no manner supersedes, overrides, or amends the Standard Contract.

This paper reflects technical work which could support future decision making by DOE. No inferences should be drawn from this paper regarding future actions by DOE, which are limited both by the terms of the Standard Contract and a lack of Congressional appropriations for the Department to fulfill its obligations under the Nuclear Waste Policy Act including licensing and construction of a spent nuclear fuel repository.

Sandia National Laboratories

Sandia National Laboratories is a multimission laboratory managed and operated by National Technology and Engineering Solutions of Sandia LLC, a wholly owned subsidiary of Honeywell International Inc. for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-NA0003525. Approved for Unclassified, Unlimited Release.

This page is intentionally left blank.

SUMMARY

This report aims at answering what, how, and when spent nuclear fuel (SNF) or dual-purpose canister (DPC) characteristics could be impacted by disposal events and processes, including decay, corrosion, dissolution, and criticality, such that the potential for criticality initiation or continuation in disposed DPCs becomes permanently significantly diminished. This report uses the term "permanent termination of criticality" to denote the significant diminishment of criticality potential, not absolute prevention.

The occurrence of disposal processes and events is a direct function of disposal time. For fundamental processes (e.g., decay), time is absolute; however, for other processes (e.g., corrosion), time is relative because it is driven by a combination of DPC characteristics (e.g., fuel conditions, basket composition), geologic parameters (e.g., infiltration rate), engineered barrier design, and other processes and events that impact in-package chemistry.

Required reactivity change that can effect permanent termination of criticality for a specific configuration is equivalent to the excess reactivity associated with the DPC disposal analysis basis configuration of closely packed fuel assemblies without any credit for neutron absorbers or baskets. This report concludes that the only perturbations to criticality control parameters that could effect permanent termination of criticality in disposed DPCs are those that relate to fuel compositional changes due to decay, corrosion / dissolution, and additional burnup from a steady-state criticality during disposal.

Although perturbations related to assembly geometry (focusing primarily on pin pitch reduction) would provide for significant negative reactivity, they could not form a defensible basis for permanent criticality termination within the first few hundred thousand years (and possibly over 1 million years) of disposal without detailed repository-dependent cladding and grid spacer corrosion analyses. Representative and defensible corrosion modeling of cladding and grid spacers based on the anticipated range of environments and chemical conditions is warranted since the conservative bases for modeling radionuclide release run counter to those required for postclosure criticality analysis. The fuel cladding could corrode and release fuel constituents prior to corrosion of grid spacers. Fuel corrosion and dissolution would not definitively support permanent criticality termination, and may increase the potential for criticality if the fuel failure and in-package chemistry result in preferential release of key absorber isotopes (i.e., Nd-143, Eu-151, Gd-155, Sm-149, and Rh-103) from the fuel.

Fuel compositional changes due to decay and additional burnup are the only predictable and quantifiable parameters that could effect permanent criticality termination in disposed DPCs. For illustration, the report provides various reactivity, burnup, decay, and enrichment equivalencies using pressurized water reactor (PWR) SNF at an initial enrichment of 4.5 wt.% U-235 and a burnup of 40 GWd/MTU. For example, an additional burnup of 20 GWd/MTU would be required to permanently terminate criticality in a DPC with excess reactivity of ~0.1 for the disposal analysies basis configuration. This DPC could sustain a steady-state criticality at a power level of 4 kW (potentially representative of a criticality in a saturated repository at elevated pressure) for 200,000 years, assuming no changes to fuel configuration.

It is important to note that the analyses and specific values provided in this report are not intended to be absolute with any level of precision. They are presented to provide perspective and to guide follow on analyses using specialized codes with detailed criticality state models based on potential material compositions taking into account decay, depletion, and dissolution.

This page is intentionally left blank.

ACKNOWLEDGEMENTS

Laura Price from Sandia National Laboratories provided valuable guidance in the development of this report.

This page is intentionally left blank.

CONTENTS

LIST OF FIGURES

LIST OF TABLES

ACRONYMS

ANS	American Nuclear Society
ANSI	American National Standards Institute
BWR	Boiling Water Reactor
BSC	Bechtel SAIC Company
DABC	disposal analysis basis configuration
DB	design basis
DOE	Department of Energy
DPC	dual-purpose canister
GWd/MTU	Gigawatt-days per metric ton of uranium
IG	Igneous
LEU	Low-Enriched Uranium
MTU	Metric Tons of Uranium
NE	Nuclear Energy
PWR	pressurized water reactor
SFWST	Spent Fuel and Waste Science and Technology
SNF	spent nuclear fuel
SNL	Sandia National Laboratories
TAD	Transportation, Aging, and Disposal.

SPENT FUEL AND WASTE SCIENCE AND TECHNOLOGY

PERMANENT CRITICALITY TERMINATION PROCESSES IN DISPOSED DPCS

1. INTRODUCTION

The Department of Energy (DOE) Office of Nuclear Energy Spent Fuel and Waste Science and Technology is evaluating the technical feasibility of direct disposal of spent nuclear fuel (SNF) in dual-purpose canisters (DPCs) in various geologies. As part of the ongoing research and development, the impact of potential postclosure criticality events on repository performance is being studied based on modeling postclosure criticality events as either quasi-steady-state (low-power and long-duration) or transient (high-power and short duration) (Price et al. 2019). During disposal, the potential for criticality in a DPC could diminish if certain processes or characteristics cause permanent reduction in DPC reactivity. The purpose of this report is to evaluate DPC characteristics and geologic disposal processes that could render the potential for criticality in a DPC permanently insignificant, thereby not warranting further consideration or analysis. This report aims at answering the following question:

> What, how, and when could SNF or DPC characteristics be impacted by disposal events and processes, including decay, corrosion, dissolution, and criticality, such that the potential for criticality initiation or continuation becomes permanently significantly diminished?

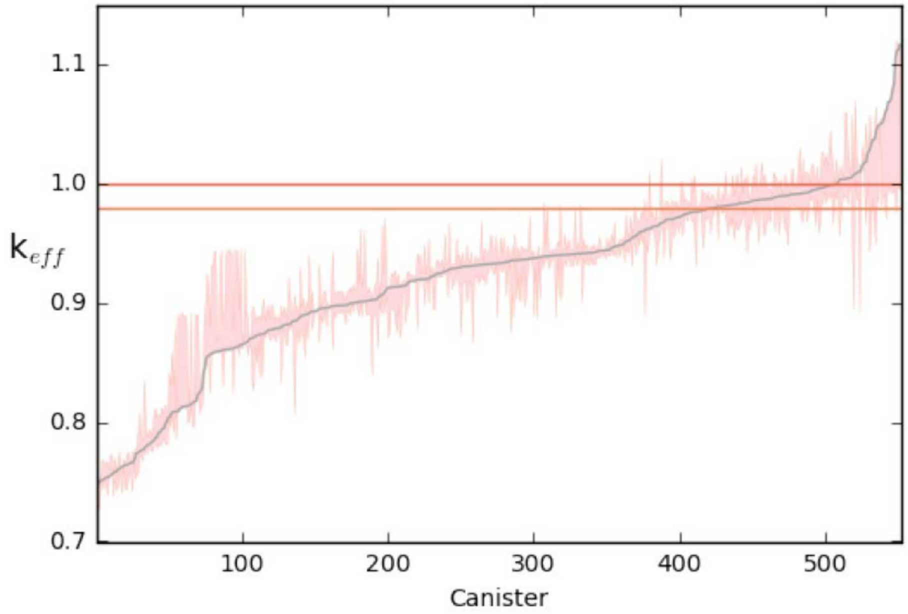
This report uses the term “permanent termination of criticality” to denote the significant diminishment of criticality potential, but not to imply absolute prevention of criticality potential. The minimum critical enrichment for uranium solutions is 0.93 wt.% U-235 and the minimum critical mass of Pu-239 is 450 grams (ANSI/ANS-8.1, 2014). These minimum values are associated with optimum geometry, optimum moderation, and no absorbers, which is certainly not the case for SNF in a DPC (i.e., absorbers are integral to the SNF, geometry changes during disposal, and moderation is limited). Although, the “equivalent” U-235 enrichment of SNF in a typical DPC could exceed 2.5 wt.% and the total mass of Pu-239 could exceed 100 kg, the potential for criticality in a DPC can become very low (as explored in this report), but cannot be completely ruled out.

It is important to note that the analyses and specific values provided in this report are not intended to be absolute with any level of precision. They are presented to provide perspective and to guide follow on analyses using specialized codes with detailed criticality state models based on potential material compositions taking into account decay, depletion, and dissolution.

This report fulfills the Spent Fuel and Waste Science and Technology Campaign deliverable M4SF-20SN010305063.

1.1 Background

Criticality states (k_{eff}) for several hundred DPCs were analyzed based on as-loaded configurations with full burnup credit in a disposal analysis basis configuration (DABC), which represents intact SNF assemblies without any credit for fixed neutron absorber plates (i.e., absorber plates are corroded, constituents dissolved, and removed from DPC) and no credit for baskets for most DPC designs (e.g., egg crate, METAMIC) (Liljenfeldt 2017). Based on these calculations, the DPCs that could support a critical configuration have a range of excess reactivity from essentially 0 (i.e., k_{eff} =1) to over 0.1 (i.e., k_{eff}> 1.1) as shown in Figure 1-1. The figure also presents a "band" for DPC criticality state changes assuming SNF rearrangement in the fuel baskets (i.e., the same fuel assemblies placed in different basket locations). The peak for each "band" represents the highest criticality state for the SNF inventory in a DPC. Although the figure demonstrates that the majority of analyzed DPCs thus far would be subcritical for the DABC, these are mostly older lower reactivity DPCs.



Note: The two orange lines represent critical limits of 1.0 and 0.98, which assumes a 2% bias and bias uncertainty.

Figure 1-2. k_{eff} for Disposal Analysis Configuration of DPCs.

1.2 Approach

The occurrence of disposal processes and events is a direct function of disposal time. For fundamental processes (e.g., decay), time is absolute; however, for other processes (e.g., corrosion), time is relative because it is driven by a combination of DPC characteristics (e.g., fuel condition, basket composition), geologic parameters (e.g., infiltration rate), engineered barrier design, and other processes and events that impact in-package chemistry.

This report evaluates perturbations to criticality control parameters that could impact the reactivity and criticality potential of SNF disposed of in a DPC. A discussion of the extent of change required to reduce the potential for criticality to be insignificant is provided for each of the pertinent parameters. The following is a listing of typical criticality control parameters and the basis for selecting those of relevance to criticality potential in disposed DPCs:

- **Reflection—**Reflection is often bounded in criticality analyses based on the potential materials that could be present. Because DPCs are relatively large systems and the potential for criticality with the depleted low-enriched uranium (LEU) fuel can exist only at thermal neutron energies, reflection plays a secondary role. Therefore, reflection does not influence the determination of permanent termination of criticality potential in disposed DPCs.
- **Leakage—**Similar to reflection, because the DPCs are large thermal systems, leakage plays a minor role in determination of criticality state. Once the system is fully degraded, leakage impacts due to increase in surface to volume ratio could significantly reduce system reactivity, however, these systems are much less reactive than the DABC for DPCs. Therefore, leakage is not evaluated as a separate mechanism for permanent termination of criticality potential in disposed DPCs.
- **Neutron Absorption—**There are several neutron absorbers that could be present in a DPC, including fixed neutron absorber plates/rods, soluble absorbers in incoming water, and absorbers integral to the SNF. Absorber plates are not credited (i.e., assumed not present) in the DABC. Dissolved species in incoming water are repository dependent. Retention of neutron absorbers from corrosion products is repository and time-dependent (i.e., not permanent). Therefore, the only absorption parameter evaluated in this report is that associated with the SNF composition based on the burnup credit analysis.
- **Geometry—**The primary geometry parameters relate to baskets that maintain separation between the fuel assemblies, grid spacers that maintain separation between the fuel rods, and cladding/fuel that maintain the lumped configuration (i.e., most reactive configuration for LEU systems). DPCs have a variety of basket designs. The DABC does not credit baskets (i.e., assumed not present) for most DPC designs (e.g., egg crate, METAMIC). Therefore, the geometry parameters evaluated in this report are the grid spacers, which maintain fuel pin separation, and fuel cladding.
- **Fissile Material Mass/Concentration/Enrichment—**These parameters have a direct influence on permanent criticality termination and, thus, are evaluated collectively in this report taking into account decay, depletion, ingrowth, and dissolution.
- **Interaction—**Neutronic coupling between the SNF disposed in neighboring DPCs is not meaningful for the large DPCs. Neutronic coupling between the fuel assemblies in the same DPC is an inherent assumption for criticality analysis. Materials that were intended to reduce interaction between fuel assemblies (e.g., neutron absorber palates) will eventually corrode; therefore, changes in interaction could not lead to permanent termination of criticality potential in disposed DPCs.
- **Temperature—**Temperature could impact the oscillatory behavior of criticality; however, it cannot lead to permanent termination of criticality.
- **Moderation—**Moderation is the most important parameter in establishing the potential for criticality in DPCs. Availability of water inside a disposed DPC is highly dependent on repository geology (e.g., unsaturated tuff vs. saturated shale), and engineering barrier design. If

water pooling inside a DPC can be permanently prevented, then there is no potential for criticality with intact fuel configurations. Evaluating the ability of the waste package to contain sufficient moderator over long time scales is beyond the scope of this report; therefore, it is assumed for the purposes of this study that sufficient moderator is present.

Required reactivity change that can effect permanent termination of criticality for a specific configuration can be expressed as follows:

Reactivity change = DPC criticality state for a specific configuration – 1.0

Based on this delta, the specific perturbation for each parameter of relevance, as discussed above, that can effect permanent termination of criticality can be determined. For example, the basis for permanent criticality termination for an intact fuel configuration would be different than for the degraded fuel configuration. This report uses the DABC for the perturbation evaluation.

Note that this report does not evaluate the potential for criticality external to the DPC due to dissolution, release and re-concentration of fissile material in the geologic formation. Evaluation of external criticality is a function of the geology and is not highly dependent on whether the SNF is in a DPC or a disposal-specialized canister.

1.3 Report Structure

This report is structured as follows:

- Evaluation of bounding SNF disposal configurations is provided in Section 2.
- Reactivity perturbations due to fuel geometry changes are evaluated in Section 3. These changes are a function of fuel design, condition at discharge, in-package chemistry, and disruptive events.
- Reactivity perturbations due to burnup are evaluated in Section 4. These changes are associated with burnup due to a steady-state criticality in a disposed DPC, not reactor depletion that formed the basis for the initial fuel composition.
- Reactivity perturbations due to decay are evaluated in Section 5. These changes are a function of time starting at fuel discharge and are not dependent on any other parameter.
- Reactivity perturbations due to compositional changes from corrosion and dissolution are evaluated in Section 6. These changes are a function of in-package chemistry, which is dependent on many natural barrier and engineered barrier parameters that could change with other processes. For a specific environment, corrosion is a function of time and starts at the point the engineered barrier sufficiently fails to allow for introduction of water.

2. EVALUATION OF BOUNDING SNF CONFIGURATIONS

Parametric analyses for various commercial SNF degraded configurations are documented in *Commercial Spent Nuclear Fuel Igneous Scenario Criticality Evaluation* (SNL 2007). These calculations demonstrate that degraded fuel configurations, even when the fuel degrades into self-moderated schoepite and the remaining porosity in the fuel and DPC void space is filled with water, are less reactive than configurations with an intact fuel lattice, as long as the fuel composition remains unchanged. Table 2-1 (excerpted from SNL 2007, Table 6-5) presents the criticality state (k_{eff}) values for a base case representing intact pressurized water reactor SNF in a transportation, aging, and disposal (TAD) canister and for a bounding potentially credible degraded configuration.

Table 2-1. Comparison of k_{eff} Values for PWR SNF Configurations

Case description	k_{eff}	Delta from Base Case
Base Case – PWR SNF assemblies at 4 wt.%, 30 gigawatt-days per metric ton of uranium (GWd/MTU) in a TAD canister in a tightly packaged geometry (assemblies separated by 6 mm of borated stainless steel plates). No basket.	0.954	N/A
Degraded Configuration - PWR SNF assemblies at 4 wt.%, 30 GWd/MTU in a TAD canister with the fuel represented as fully hydrated schoepite with 15% porosity, which is the stable form of schoepite that maximizes the amount of hydrogen present. Similar to the Base Case, the degraded fuel assemblies are separated by 6 mm of borated stainless steel plates and no basket.	0.858	0.096

Table A-12 of SNL 2007 documents a large number of sensitivity cases with fresh fuel comparing intact configurations to various configurations of schoepite at a range of porosities and water saturation levels. These sensitivity analyses demonstrate that, although some “theoretical” combination of parameters for degraded configurations could approach the reactivity of intact SNF, none would exceed it. Therefore, the most reactive configuration for commercial SNF in a DPC is intact fuel lattice with closely grouped assemblies and without a fuel basket or neutron absorber plates, which is denoted in this report as the DABC.

To determine the point at which criticality potential for a DPC is permanently terminated, the change in reactivity due perturbations during disposal must be greater than the excess reactivity for the DABC of a specific DPC. The following sections explore the potential magnitudes of permanent reactivity changes due to irreversible geometry change, burnup, decay, or compositional change due to corrosion/dissolution to determine the fraction of DPCs for which criticality can be permanently terminated and associated processes/timing of termination.

3. REACTIVITY PERTURBATIONS DUE TO FUEL GEOMETRY CHANGES

Given that the most reactive credible geometry for commercial SNF is tightly packed assemblies (i.e., no basket or neutron absorbers), the reactivity perturbations due to fuel geometry changes are based on pin pitch (i.e., separation between fuel pins) changes only. Commercial SNF is designed to be under-moderated; therefore, a slight pin pitch expansion would increase reactivity; however, uniform pin pitch expansion in the fuel assemblies is not considered credible for disposal configurations. Figure 3-1 presents normalized k_{eff} as a function of pin pitch for single reflected fully moderated PWR and boiling water reactor (BWR) assemblies as a function of pin pitch. These figures are adapted from BSC 2005, Tables 6.3-4 and 6.3-6.

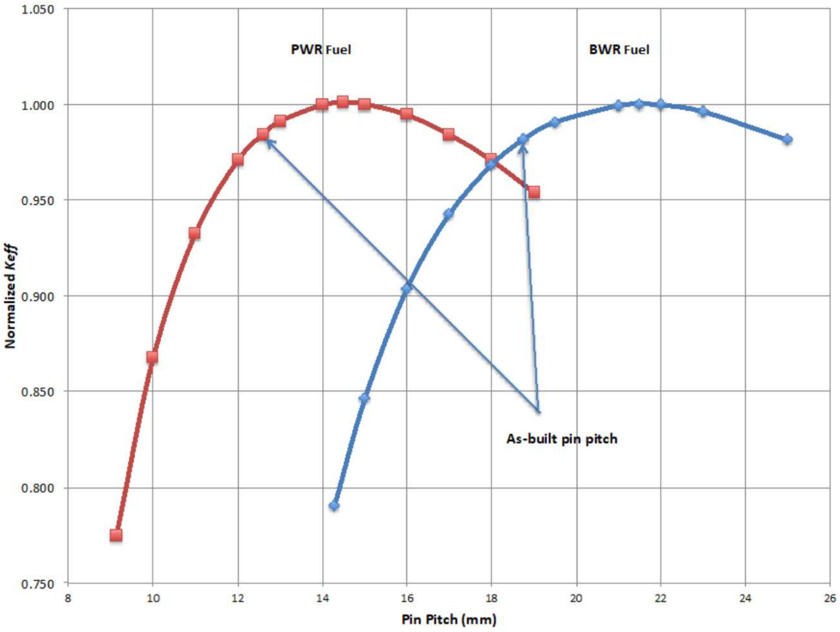


Figure 3-1. k_{eff} as a Function of Pin Pitch.

As shown in Figure 3-1, pin pitch reduction has a significant impact on reducing reactivity. For the modeled PWR fresh fuel, reducing the pin pitch by 1 mm from as-manufactured conditions reduces k_{eff} by 0.023. The reduction in reactivity becomes more significant for additional pin pitch reduction. A uniform pin pitch reduction of ~ 3 mm would likely be sufficient to render the higher reactivity DPCs (with excess DABC reactivity of 0.1) permanently subcritical, since reversal of pin pitch reduction is not plausible during disposal.

SNF assembly geometry changes can be induced by either corrosion or disruptive events. Corrosion is time-based once initiated, whereas disruptive events are less predictable. Fuel grid spacers vary in design, but generally use zircaloy as the primary material of construction of grid straps and dimples and either zircaloy or inconel for the springs. Figure 3-2 provides an illustration of a grid spacer. A typical thickness of grid spacer straps and dimples is ~0.5 mm (Jiang et al. 2016). Cladding thickness for a typical PWR fuel is 0.57 mm and for a typical BWR fuel is 0.74 mm (BSC 2004).

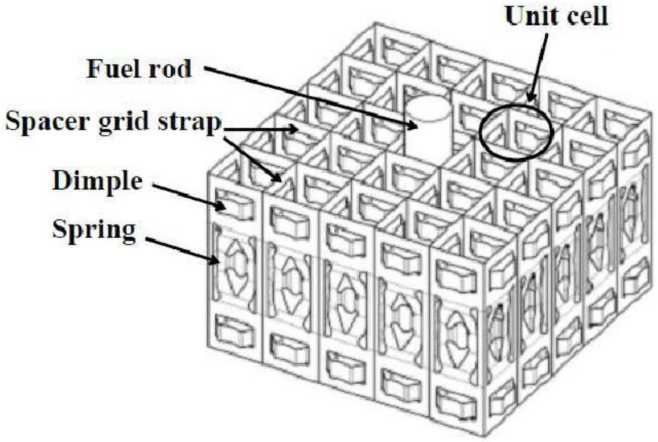


Figure 3-2. Example Grid Spacer.

If the in-package chemistry is conducive to zircaloy corrosion, it is plausible that the grid spacers would degrade at a slower rate than fuel cladding primarily because the grid spacers experience less degradation during irradiation. Primary contributors to cladding degradation are oxidation, creep, and hydride formation/reorientation, several of which are a direct function of hoop stress. Commercial SNF rods are subject to relatively significant hoop stresses, whereas there are no such stresses on grid spacers. These mechanisms tend to impact select fuel rods or even segments of fuel rods (e.g., those with hairline cracks or high peak burnup). Figure 3-3 shows typical fuel unzipping, which does not seem to result in significant fuel releases. The impact of failure of a few fuel rods on system reactivity is difficult to predict because, depending on the location and what the removed fuel is replaced with, localized removal could result in either a positive or a negative reactivity change. Replacing a few fuel rods with water generally increases reactivity because it increases moderation for the normally under-moderated assemblies (see Figure 3-1), whereas replacing a few fuel rods with corrosion products could slightly decrease reactivity (or result in no change).

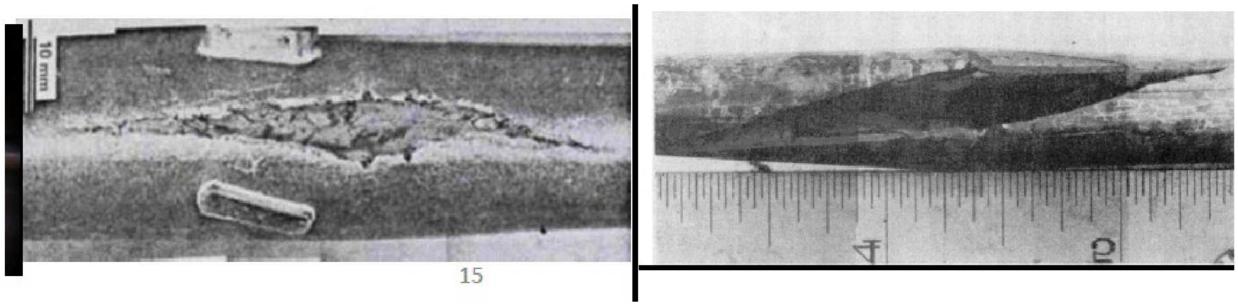


Figure 3-3. Example Cladding Unzipping.

The corrosion rate of zircaloy is highly dependent on the composition of the specific zircaloy alloy, its condition (e.g., oxide layer, hydrides), and in-package chemistry. In-package chemistry varies widely based on repository geology (e.g., unsaturated tuff, saturated shale, salt), in-package components (DPC internal designs vary significantly), radiolysis, and evaporation/concentration (due to decay heat or criticality events). Therefore, predicting a specific timeframe for when the grid spacers would degrade after water intrusion is highly uncertain without a detailed performance assessment. No credit was taken for the cladding for the unsaturated tuff Yucca Mountain Repository (BSC 2008, Section 2.3.7.6); the licensing performance assessment assumed that cladding has failed and the fuel is exposed upon waste package failure. This assumption was conservative from a release standpoint; however, for the purposes of this study, the assumption is not necessarily conservative. Therefore, representative and defensible corrosion modeling of cladding and grid spacers based on the anticipated range of environments and chemical conditions is warranted since the conservative bases for radionuclide release run counter to those required for postclosure criticality analysis. Without significant corrosion modeling of the cladding and grid spacers, a licensing basis of geometry-based permanent termination of criticality would likely not be defensible within the first few hundred thousand years post water intrusion.

4. REACTIVITY PERTURBATIONS DUE TO BURNUP

Burnup changes the composition of the fuel from a reactivity perspective in two ways as follows:

- **Depletion of fissile material**—The only fissile isotope in fresh fuel is U-235, which starts depleting rapidly with burnup. For SNF with relatively high burnup, there are several fissile isotopes that contribute to fission and power generation including Pu-239, Pu-241 and Am-243m. Figure 4-1 provides the concentration of the various fissile isotopes as a function of burnup for PWR SNF with an initial enrichment of 4.5 wt.% U-235. Figure 4-1 (adapted from BSC 2003, Table 17) illustrates that with increased burnup, the primary fissile isotope becomes Pu-239 and essentially reaches an equilibrium concentration at ~30 GWd/MTU (i.e., the generation rate is essentially equivalent to the depletion rate).

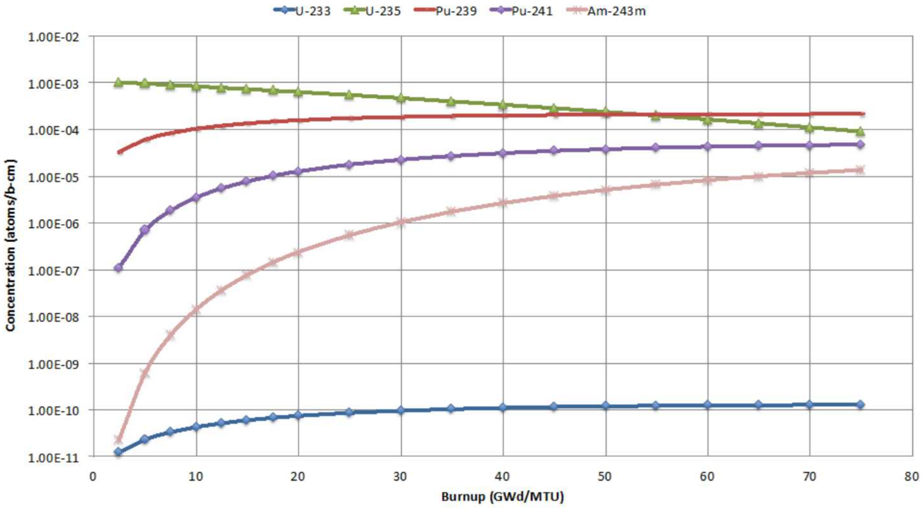


Figure 4-1. Concentration of Fissile Isotopes as a Function of PWR SNF Burnup.

- **Generation of Neutron Absorbers**—Fuel depletion generates neutron absorbers through either fission (i.e., fission products) or parasitic neutron absorptions (i.e., higher actinides). The primary neutron absorbers with importance to long-term disposal criticality are Nd-143, Eu-151, Gd-155, Sm-149, and Rh-103 (See Section 6). Figure 4-2 (adapted from BSC 2003, Table 17) provides the concentrations of these absorbers or their parent nuclides (in the case of Eu-151, the daughter of Sm-151) as a function of burnup for PWR SNF with an initial enrichment of 4.5 wt.% U-235. Figure 4-2 illustrates that fission products concentration growth rates drop with burnup because of their relatively high absorption cross sections, nonetheless with the exception of Sm-149, their generation rate remains higher than their depletion rate.

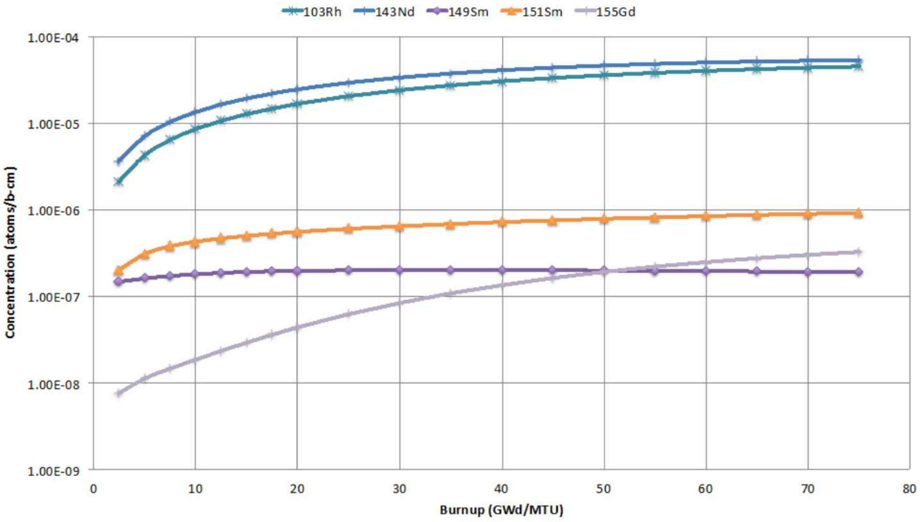
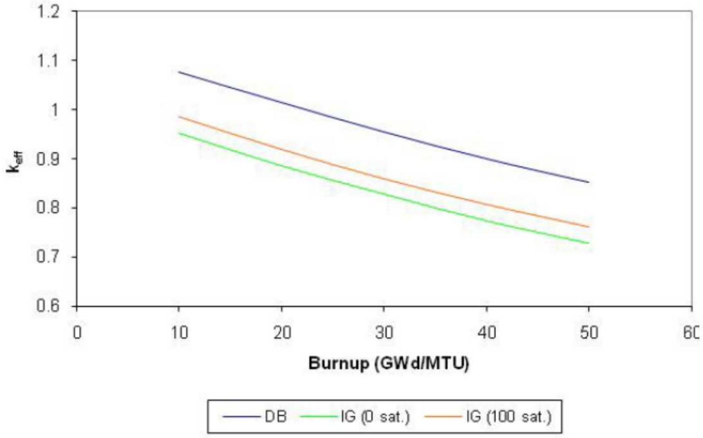
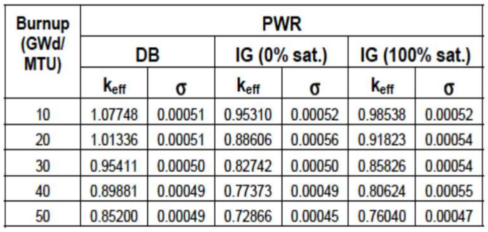


Figure 4-2. Concentration of Neutron Absorber Isotopes as a Function of PWR SNF Burnup.

Because burnup-induced composition changes are strongly correlated to the level of burnup with different generation/depletion rates for the various isotopes, their impacts on reactivity are treated collectively in this report. Figure 4-3 (adapted from SNL 2007) provides k_{eff} as a function of burnup for both intact and fully degraded configurations. The intact configuration represents closely packed SNF assemblies with an initial enrichment of 4.0 wt.%, separated with 6 mm of borated stainless steel (i.e., Yucca Mountain Repository TAD canister design basis [DB] configuration). For the degraded igneous configurations (denoted IG in Figure 4-3), the fuel is represented as fully hydrated schoepite with 15% porosity. Figure 4-3 demonstrates that k_{eff} essentially decreases linearly with burnup, with an average rate of change of approximately -0.005 Δk_{eff} per GWd/MTU for PWR SNF. For example, a DPC with excess reactivity of 0.05 for the DABC would require an additional burnup of ~9 GWd/MTU to become permanently subcritical. Note that this is based on uniform burnup of SNF within the canister. For actual DPCs loaded with SNF with a range of burnup values, the reactivity to burnup ratio would likely be different.





Burnup (GWd/ MTU)	PWR					
	DB		IG (0% sat.)		IG (100% sat.)	
	k_{eff}	σ	k_{eff}	σ	k_{eff}	σ
10	1.07748	0.00051	0.95310	0.00052	0.98538	0.00052
20	1.01336	0.00051	0.88606	0.00056	0.91823	0.00054
30	0.95411	0.00050	0.82742	0.00050	0.85826	0.00054
40	0.89881	0.00049	0.77373	0.00049	0.80624	0.00055
50	0.85200	0.00049	0.72866	0.00045	0.76040	0.00047

Figure 4-3. k_{eff} as a Function of PWR SNF Burnup for the TAD Design Basis Configuration (DB) and Degraded Igneous Configurations (IG) with Varying Levels of Saturation.

Figure 4-4 (SNL 2008, Figure 7-1) presents the PWR loading curve that was developed for the Yucca Mountain TAD canister. The various points on the loading curve for each burnup/enrichment pair have an equivalent criticality state in the assumed design basis configuration (SNF assemblies separated by 6 mm of borated stainless steel). Based on the near linear relationship between burnup, enrichment, and reactivity change, as illustrated by the loading curve, an average burnup/enrichment equivalency of ~13 GWd/MTU per wt.% U-235 can be deduced. Note that this equivalency is more applicable to canisters with relatively uniform burnup in the primary region of importance in a canister (i.e., central region). For the heterogeneously-loaded DPCs, not only would the value be different, but also the overall impact on DPC reactivity if the criticality event is regional with assemblies experiencing depletion while surrounding ones experiencing some growth (i.e., acting as blanket assemblies).

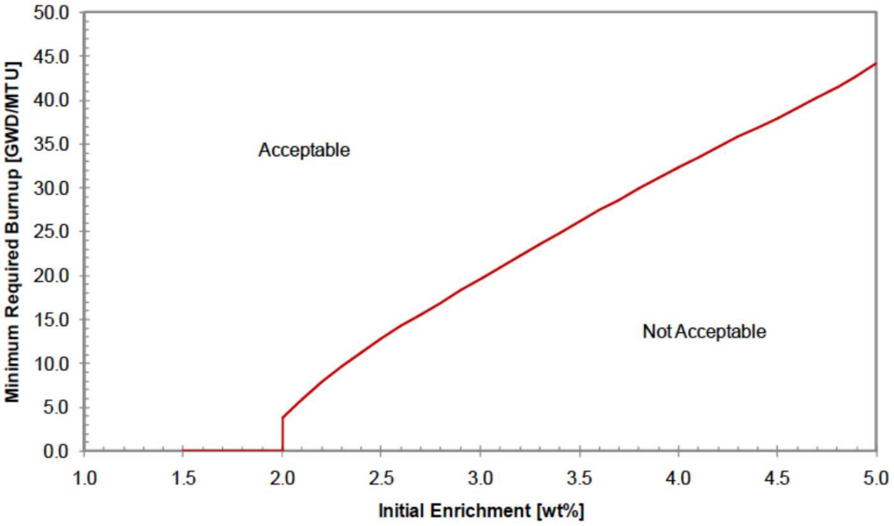


Figure 4-4. PWR Loading Curve for the Yucca Mountain Repository.

A quasi-steady-state criticality during disposal would result in some level of additional burnup. For example, a steady-state criticality at a power level of 400 watts (potentially representative of a criticality in an unsaturated repository at atmospheric pressure) lasting for 10,000 years would result in an additional 0.1 GWd/MTU average burnup in a typical DPC. A steady-state criticality at a power level of 4 kW (potentially representative of a criticality in a saturated repository at elevated pressure) lasting for 10,000 years would result in an additional 1 GWd/MTU average burnup in a typical DPC. This incremental burnup would result in insignificant changes to the fuel composition and DPC reactivity.

For permanent termination of criticality, Figures 4-5 and 4-6 provide the minimum required level of additional burnup and associated equivalent enrichment reduction as a function of initial excess reactivity in a DPC. For example, based on the average rate of change of approximately -0.005 Δk_{eff} per GWd/MTU discussed above, a DPC with excess reactivity of 0.05 would require an additional burnup of ~9 GWd/MTU to become permanently subcritical. Based on the limited burnup that could be achieved during a potential criticality event, even it if lasts for tens of thousands of years, only DPCs with small initial excess reactivity could become permanently subcritical due to the additional burnup. Note that permanent termination would apply to the intact fuel configuration as well as potential degraded configurations, as long as there is no preferential dissolution of fuel constituents.

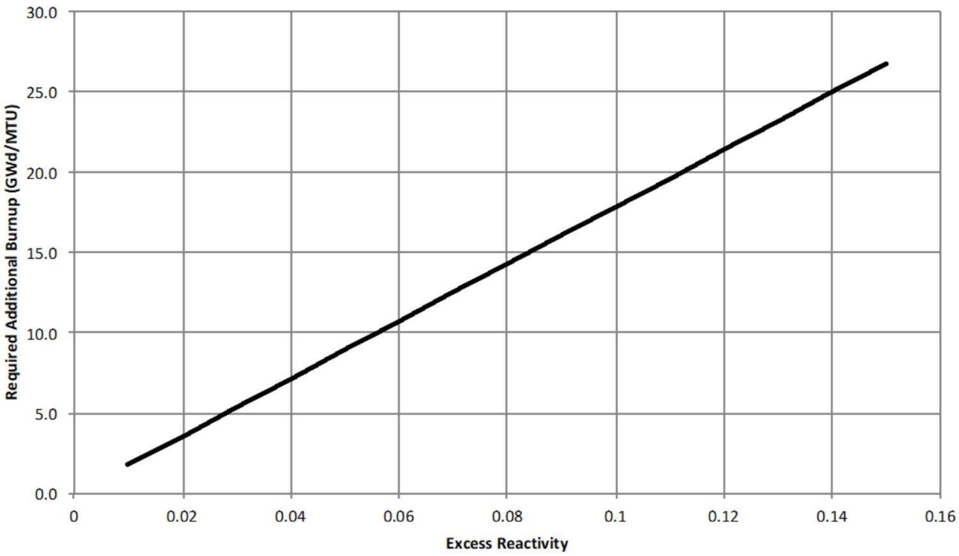


Figure 4-5. Required Additional Burnup for Permanent Criticality Termination as a Function of Excess Reactivity.

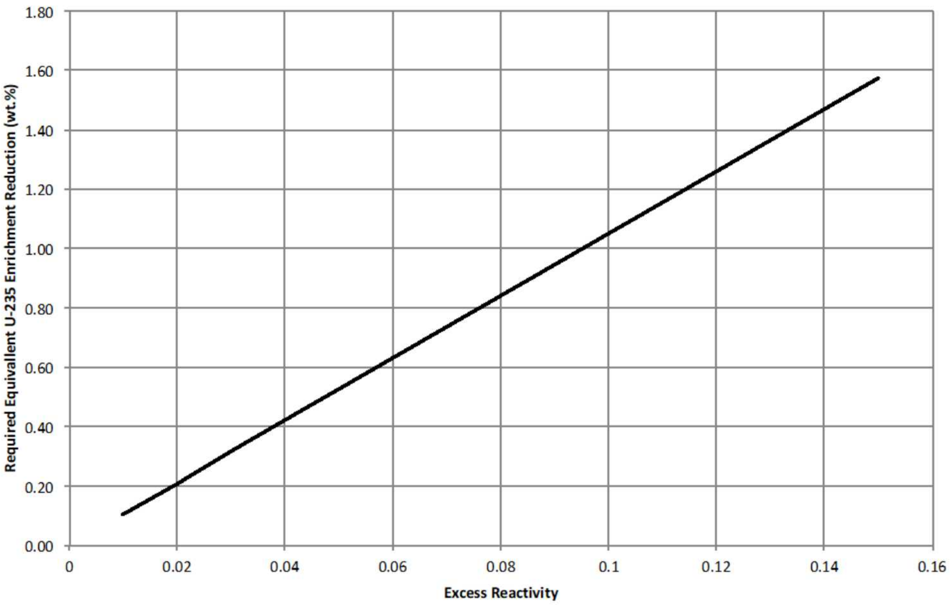


Figure 4-6. Required Equivalent U-235 Enrichment Reduction for Permanent Criticality Termination as a Function of Excess Reactivity.

5. REACTIVITY CHANGES DUE TO DECAY

The reactivity of commercial SNF changes as a function of decay time because several of the fissionable isotopes and absorbers considered in the burnup credit analysis are radioactive with a range of half-lives from tens of years to millions of years. The relative reactivity change as a function of decay highly depends on the SNF composition used in the model to determine the criticality state. Figure 5-1 provides k_{eff} as a function of time for a generic 32-PWR cask with fuel enriched to 4 wt.% U-235 and a burnup of 40 GWd/MTU (Wagner and Parks 2003, Figure 3). Because the DPC disposal criticality analysis is based on full burnup credit, the lowest reactivity line in the figure would be more representative of the reactivity changes as a function of time.

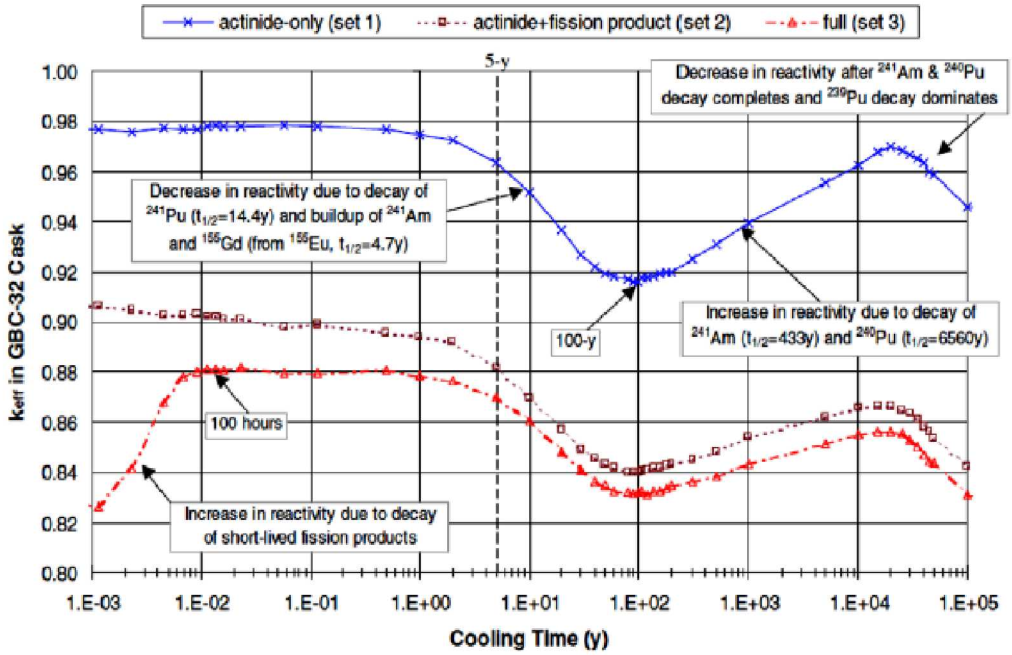


Figure 5-1. k_{eff} as a Function of Decay Time for a Generic 32 PWR Canister.

Figure 5-1 is limited to 100,000 years of decay and demonstrates that this decay duration accounts for 0.05 Δk_{eff} from initial peak reactivity at 100 hours, 0.04 Δk_{eff} from reactivity at 5 years, and ~0.03 Δk_{eff} from the second reactivity peak at ~ 20,000 years. Therefore, barring any other changes, a DPC DABC with excess reactivity less 0.03 would no longer have the potential for criticality at 100,000 years, as long as there is no preferential separation between the various fuel constituents. To determine the impact of longer decay times and the value of the various isotopes considered in the modeling of the criticality state, an equivalency analysis is performed in Appendix A for a representative burnup enrichment pair on the Yucca Mountain PWR loading curve (4.5 wt.% enrichment and 40 GWd/MTU burnup) (BSC 2003, Table 17). The equivalency analysis represents the 29 principal isotopes considered in the burnup credit analysis by a single fissile isotope using fissile material equivalency (Lakes and Ashley 2017, Table B-2) and a single absorber isotope using neutron cross section ratios.

Appendix A provides the fuel composition at 5 years decay with pertinent parameters for each isotope including half-life (if radioactive) and its ingrowth sources and decay daughters. The Appendix evaluates the composition at 1 million years. Durations past 1 million years are not considered meaningful for repository assessments. Appendix A provides the fissile isotopes equivalency in terms of U-235 and the neutron absorber equivalency in terms of the stable Sm-150 at 5 years and 1 million years. The absorption equivalency is based on thermal neutron absorption cross sections; this is considered a reasonable approximation since only thermal criticality events are possible for the low-enriched commercial SNF. Table 5-1 provides a summary of the equivalencies. The equivalent U-235 initial enrichment drops by 28% due to decay from 5 years to 1 million years, whereas, the equivalent absorption drops by 26% for the same decay duration. The relative reduction in equivalent absorber nuclide concentration after 1 million years of decay is comparable to the relative reduction in equivalent fissile nuclide concentration.

Table 5-1. Fissile and Absorber Material Equivalency at 5 years and 1 Million Years for PWR SNF (4.5 wt.% U-235 and 40 GWd/MTU).

Equivalent Isotope	5 Year (Initial)		~ 1 Million Years (Final)		%Change (Final – Initial / Initial)
	Equivalent Concentration (atoms/b-cm)	Equivalent Enrichment at 5 years (wt.%)	Equivalent Concentration (atoms/b-cm)	Equivalent Enrichment (wt.%)	
U-235	7.64E-4	3.33 wt.%	5.47E-4	2.39 wt.%	-28%
Sm-150	2.42E-3	N/A	1.79E-3	N/A	-26%

Figure 5-2 (NEA 2012, Figure 1) shows that isotopic composition changes important to criticality occur within the first 100,000 years, after which the only potentially relevant change is growth of U-233 from decay of Np-237 with a half-life of 2.1 million years. As estimated in Appendix A, the total concentration of U-233 after 1 million years of decay is equivalent to ~1% of the total U-235 concentration and thus would likely have a relatively insignificant impact on DPC reactivity. Therefore, the reactivity of SNF would likely experience relatively minor changes due to decay after 100,000 years based on the 29 principal isotope burnup credit analysis.

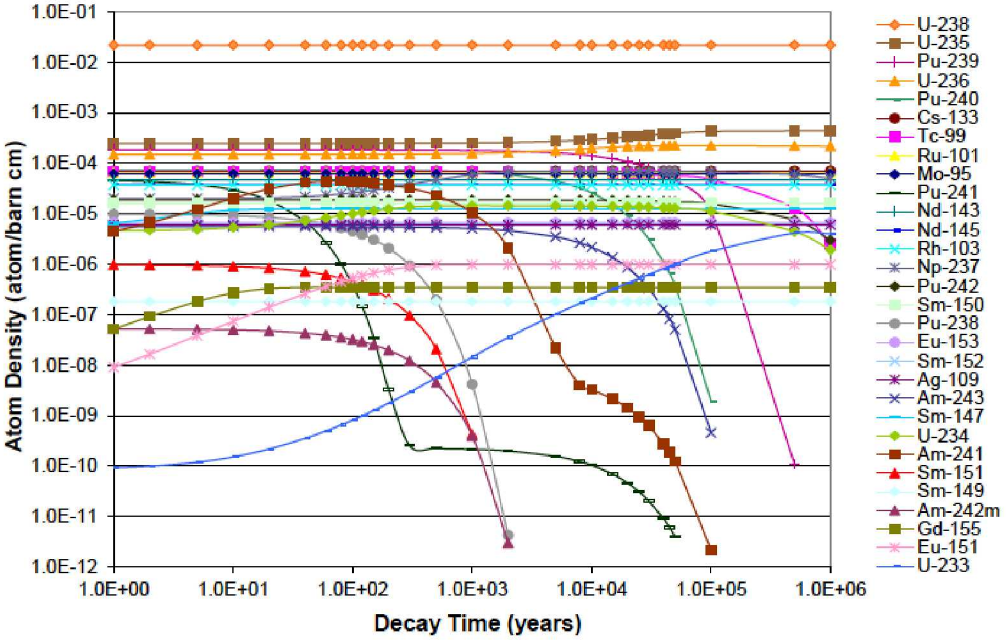


Figure 5-2. Isotopic Concentrations of Isotopes Important to Criticality.

6. REACTIVITY PERTURBATIONS DUE TO COMPOSITIONAL CHANGES FROM CORROSION / DISSOLUTION

The relative reactivity worth (i.e., contribution to fission and/or absorption) of the various isotopes of relevance to criticality state evaluations based on their concentrations at 5 years and 10 million years, fissile equivalency, and thermal neutron absorption cross sections is developed in Appendix A and summarized in Table 6-1.

Table 6-1. Fissile and Absorber Material Equivalency at 5 years and 1 Million Years for PWR SNF (4.5 wt.% U-235 and 40 GWd/MTU).

Isotope	**Absorption**		**Fission**	
	Relative Worth at 5 years	**Relative Worth at 1 million years**	**Relative Worth at 5 years**	**Relative Worth at 1 million years**
16O	0.00%	0.00%		
95Mo	0.31%	0.42%		
99Tc	0.52%	0.00%		
Ru-99	0.00%	0.22%		
101Ru	0.11%	0.15%		
103Rh	1.83%	2.48%		
109Ag	0.19%	0.25%		
143Nd	5.55%	7.53%		
145Nd	0.66%	0.89%		
147Sm	0.22%	0.30%		
149Sm	3.36%	4.56%		
150Sm	0.56%	0.76%		
151Sm	4.56%	0.00%		
152Sm	0.46%	0.63%		
151Eu	0.11%	3.90%		
153Eu	0.66%	0.90%		
155Gd	3.42%	9.26%		
233U	0.00%	0.08%	0.00%	0.93%
234U	0.23%	0.00%		
235U	13.78%	29.87%	44.24%	98.89%
236U	0.29%	0.56%		
238U	24.43%	33.14%		
237Np	1.17%	4.09%	0.05%	0.18%
238Pu	1.29%	0.00%	0.13%	0.00%
239Pu	22.22%	0.00%	40.71%	0.00%
240Pu	6.78%	0.00%	0.26%	0.00%
241Pu	4.66%	0.00%	14.29%	0.00%
242Pu	0.08%	0.00%	0.02%	0.00%
241Am	2.45%	0.00%	0.04%	0.00%
242mAm	0.01%	0.00%	0.26%	0.00%
243Am	0.08%	0.00%	0.01%	0.00%

Dissolution and removal of uranium would not change the relative enrichment of the fuel, only the total mass. Because of the large mass of uranium in a DPC, uniform removal of uranium would likely have a small impact on system reactivity. The impact of localized removal of uranium (e.g., from select failed rods) on system reactivity is difficult to predict because, depending on the location and what the removed uranium is replaced with, localized removal could result in either a positive or a negative reactivity change. Replacing a few fuel rods with water generally increases reactivity because it increases moderation for the normally under-moderated assemblies (see Figure 3-1), whereas replacing a few fuel rods with corrosion products would slightly decrease reactivity. Therefore, system reactivity changes due to preferential removal of uranium, assuming such a mechanism is possible, would likely not result in significant reactivity reduction. Preferential removal of neutron poisons, on the other hand, could increase system reactivity, potentially significantly. Therefore, corrosion, dissolution, and releases evaluations are more meaningful for removal of the following isotopes:

- Removal of plutonium from the fuel could significantly reduce reactivity if it were to occur prior to decay of Pu-239, which has a half-life of 2.41E4 years. As shown in Table 6-1, Pu-239 makes up ~40% of the fissile material at 5 years. Its contribution would be ~20% at ~25,000 years. Therefore, if the package chemistry does not allow for preferential plutonium dissolution from the fuel, or a significant amount could not be removed from the fuel within 100,000 years, then plutonium dissolution would not be a factor in permanent criticality termination.
- Removal of absorber isotopes would negatively impact the permanent criticality termination argument. Therefore, in order to ensure that if criticality is considered terminated based on the demonstration that the DABC is subcritical due to decay, burnup, or geometry change, a defensible argument must be developed to demonstrate that the isotopes listed in Table 6-1 cannot be preferentially separated from the fuel based on their relative importance. The primary non-plutonium and uranium absorbers are Nd-143, Eu-151, Gd-155, Sm-149, and Rh-103.

7. SUMMARY AND CONCLUSIONS

This report aims at answering what, how, and when SNF or DPC characteristics could be impacted by disposal events and processes, including decay, corrosion, dissolution, and criticality, such that the potential for criticality initiation or continuation in disposed DPCs becomes permanently significantly diminished. This report uses the term "permanent termination of criticality" to denote the significant diminishment of criticality potential, not absolute prevention.

The occurrence of disposal processes and events is a direct function of disposal time. For fundamental processes (e.g., decay), time is absolute; however, for other processes (e.g., corrosion), time is relative because it is driven by a combination of DPC characteristics (e.g., fuel conditions, basket composition), geologic parameters (e.g., infiltration rate), engineered barrier design, and other processes and events that impact in-package chemistry.

Required reactivity change that can effect permanent termination of criticality for a specific configuration is equivalent to the excess reactivity associated with the DPC DABC of closely packed fuel assemblies without any credit for neutron absorbers or baskets. Of the various criticality control parameters, this report concludes that the only perturbations to criticality control parameters that could effect permanent termination of criticality in disposed DPCs are those that relate to fuel compositional changes due to decay, corrosion / dissolution, and additional burnup from a steady-state criticality during disposal.

Although perturbations related to assembly geometry (focusing primarily on pin pitch reduction) would provide for significant negative reactivity, they could not form a defensible basis for permanent criticality termination within the first few hundred thousand years (and possibly over 1 million years) of disposal without detailed repository-dependent cladding and grid spacer corrosion analyses. Representative and defensible corrosion modeling of cladding and grid spacers based on the anticipated range of environments and chemical conditions is warranted since the conservative bases for modeling radionuclide release run counter to those required for postclosure criticality analysis. The fuel cladding could corrode and release fuel constituents prior to corrosion of grid spacers. Fuel corrosion and dissolution would not definitively support permanent criticality termination, and may increase the potential for criticality if the fuel failure and in-package chemistry result in preferential release of key absorber isotopes (i.e., Nd-143, Eu-151, Gd-155, Sm-149, and Rh-103) from the fuel.

Fuel compositional changes due to decay and additional burnup are the only predictable and quantifiable parameters that could effect permanent criticality termination in disposed DPCs. Long-term decay (from 5 years to ~100 thousand years) could reduce reactivity by ~0.04. The impact of additional burnup due to a steady-state disposal criticality event could reduce DPC k_{eff} by 0.005 per GWd/MTU, as discussed in Section 4. For a DPC with excess reactivity of ~0.1, an additional burnup of 20 GWd/MTU would be required to effect permanent criticality termination. This DPC could sustain a steady-state criticality at a power level of 4 kW (potentially representative of a criticality in a saturated repository at elevated pressure) for 200,000 years, assuming no changes to fuel configuration.

It is important to note that the analyses and specific values provided in this report are not intended to be absolute with any level of precision. They are presented to provide perspective and to guide follow on analyses using specialized codes with detailed criticality state models based on potential material compositions taking into account decay, depletion, and dissolution.

8. REFERENCES

ANSI (American National Standards Institute) /ANS (American Nuclear Society) 8.1-2014, Nuclear Criticality Safety in Operations with Fissionable Materials outside Reactors. American Nuclear Society, La Grange Park, Illinois.

BSC (Bechtel SAIC Company) 2003. *Isotopic Generation and Confirmation of the PWR Application Model*. CAL-DSU-NU-000004 REV 00A. Bechtel SAIC Company, Las Vegas, Nevada.

BSC 2005. *Dry Transfer Facility Criticality Safety Calculations*. 100-00C-WHS0-00100-000-00C. Bechtel SAIC Company, Las Vegas, NV.

Jiang, H., J. Qu, R. Lu, and J. Wang 2016. *Grid-to-rod flow-induced impact study for PWR fuel in reactor*. Progress in Nuclear Energy, 2016, 91: 355–361.

Lakes M. E. and R. J. Ashley 2017. *Hanford Site Solid Waste Acceptance Criteria*. HNF-EP-0063, Rev. 17. CH2M HILL Plateau Remediation Company, Richland, Washington.

Liljenfeldt, H., K, Banerjee, J, Clarity, J, Scaglione, R, Jubin, V, Sobes, R. Howard, E. Hardin, L. Price, E. Kalinina, T. Hadgu, A. Ilgen, C. Bryan, J. Carter, T. Severynse and F. Perry 2017. *Summary of Investigations on Technical Feasibility of Direct Disposal of Dual-Purpose Canisters*. SFWD-SFWST-2017-000045. U.S. Department of Energy, Office of Spent Fuel and Waste Science and Technology.

NEA (Nuclear Energy Agency) 2012. *Burn-Up Credit Criticality Safety Benchmark—Phase VII: UO2 Fuel: Study of Spent Fuel Compositions for Long-Term Disposal*. NEA No. 6998. Nuclear Energy Agency, Paris, France.

SNL (Sandia National Laboratories) 2007. *Commercial Spent Nuclear Fuel Igneous Scenario Criticality Evaluation*. ANL-EBS-NU-000009 REV 00. Sandia National Laboratories, Las Vegas, Nevada.

SNL 2008. *CSNF Loading Curve Sensitivity Analysis*. ANL–EBS–NU–000010. Rev. 00. Sandia National Laboratories, Las Vegas, Nevada.

Wagner, J.C. and C.V. Parks 2003. *Recommendations on the Credit for Cooling Time in PWR Burnup Credit Analyses*. NUREG/CR-6781 (ORNL/TM-2001/272). Oak Ridge National Laboratory, Oak Ridge, TN.

Appendix A—ISOTOPIC EQUIVALENCY ANALYSIS FOR PWR SNF

Table A-1. Isotopic Concentrations at 5 Years and 1 Million Years for PWR SNF (4.5 wt.% U-235 and 40 GWd/MTU).

Isotope	Half Life (years)	Parent Isotope	Thermal Neutron Absorption Cross Section (b)	Thermal Neutron Fission Cross Section (b)	Decay Daughter	Concentration at 5 years (atoms/b-cm)	Concentration at 1 m years (atoms/b-cm)	Delta	Ratio
16O	S	N/A	1.90E-04	N/A	N/A	4.65E-02	4.65E-02	0.00E+00	1.00E+00
95Mo	S	N/A	1.34E+01	N/A	N/A	5.55E-05	5.55E-05	0.00E+00	1.00E+00
99Tc	2.11E+05	N/A	2.28E+01	N/A	Ru-99	5.52E-05	0.00E+00	-5.52E-05	0.00E+00
101Ru	S	N/A	5.20E+00	N/A	N/A	5.18E-05	5.18E-05	0.00E+00	1.00E+00
103Rh	S	N/A	1.44E+02	N/A	N/A	3.09E-05	3.09E-05	0.00E+00	1.00E+00
109Ag	S	N/A	9.10E+01	N/A	N/A	5.00E-06	5.00E-06	0.00E+00	1.00E+00
143Nd	S	N/A	3.25E+02	N/A	N/A	4.14E-05	4.14E-05	0.00E+00	1.00E+00
145Nd	S	N/A	5.00E+01	N/A	N/A	3.20E-05	3.20E-05	0.00E+00	1.00E+00
147Sm	S	N/A	5.70E+01	N/A	N/A	9.48E-06	9.48E-06	0.00E+00	1.00E+00
149Sm	S	N/A	4.01E+04	N/A	N/A	2.03E-07	2.03E-07	0.00E+00	1.00E+00
150Sm	S	N/A	1.00E+02	N/A	N/A	1.35E-05	1.35E-05	0.00E+00	1.00E+00
151Sm	8.88E+01	N/A	1.52E+04	N/A	Eu-151	7.29E-07	0	-7.29E-07	0.00E+00
152Sm	S	N/A	2.06E+02	N/A	N/A	5.43E-06	5.43E-06	0.00E+00	1.00E+00
151Eu	S	Sm-151	9.20E+03	N/A	N/A	2.98E-08	7.59E-07	7.29E-07	2.55E+01
153Eu	S	N/A	3.12E+02	N/A	N/A	5.15E-06	5.15E-06	0.00E+00	1.00E+00
155Gd	S	Eu-155	6.09E+04	N/A	N/A	1.36E-07	2.72E-07	1.36E-07	2.00E+00
233U	1.59E+05	Np-237	4.55E+01	5.29E+02	Bi-209	1.12E-10	3.27E-06	3.27E-06	2.92E+04
234U	2.46E+05	Pu-238	9.98E+01	6.70E-02	Pb-206 or Pb-208	5.51E-06	0	-5.51E-06	0.00E+00
235U	7.04E+08	Pu-239	9.88E+01	5.83E+02	N/A	3.38E-04	5.41E-04	2.03E-04	1.60E+00
236U	2.35E+07	Pu-240	5.09E+00	N/A	N/A	1.39E-04	1.96E-04	5.68E-05	1.41E+00
238U	4.50E+09	Pu-242	2.68E+00	N/A	N/A	2.21E-02	2.21E-02	1.11E-05	1.00E+00
237Np	2.14E+06	Am-241	1.76E+02	2.00E-02	U-233	1.61E-05	4.15E-05	2.54E-05	2.58E+00
238Pu	8.77E+01	N/A	5.40E+02	1.79E+01	U-234	5.81E-06	0	-5.81E-06	0.00E+00
239Pu	2.41E+04	Am-243	2.69E+02	7.48E+02	U-235	2.00E-04	0	-2.00E-04	0.00E+00
240Pu	6.56E+03	N/A	2.90E+02	5.60E-02	U-236	5.68E-05	0	-5.68E-05	0.00E+00
241Pu	1.43E+01	N/A	3.62E+02	1.01E+03	Am-241	3.12E-05	0	-3.12E-05	0.00E+00
242Pu	3.75E+05	N/A	1.85E+01	2.00E-01	U-238	1.11E-05	0	-1.11E-05	0.00E+00
241Am	4.32E+02	Pu-241	5.87E+02	3.20E+00	Np-237	1.01E-05	0	-1.01E-05	0.00E+00
242mAm	1.41E+02	N/A	3.30E+02	2.10E+03	Pu-238	3.68E-08	0	-3.68E-08	0.00E+00
243Am	7.37E+03	N/A	7.51E+01	1.98E-01	Pu-239	2.66E-06	0	-2.66E-06	0.00E+00

Table A-2. Fission Equivalency Evaluation at 5 Years and 1 Million Years for PWR SNF (4.5 wt.% U-235 and 40 GWd/MTU).

Isotope	Fissile Equivalency Raito	U-235 Equivalency At 5 years					U-235 Equivalency At 1 Million Years					Change due to Decay
		Concentration (atoms/b-cm)	Concentration (atoms/b-cm)	Enrichment	Ratio to Original Concentration	Contribution Fraction	Concentration (atoms/b-cm)	Concentration (atoms/b-cm)	Enrichment	Ratio to Original Concentration	Contribution Fraction	
16O	N/A	N/A					N/A					
95Mo	N/A	N/A					N/A					
99Tc	N/A	N/A					N/A					
101Ru	N/A	N/A					N/A					
103Rh	N/A	N/A					N/A					
109Ag	N/A	N/A					N/A					
143Nd	N/A	N/A					N/A					
145Nd	N/A	N/A					N/A					
147Sm	N/A	N/A					N/A					
149Sm	N/A	N/A					N/A					
150Sm	N/A	N/A					N/A					
151Sm	N/A	N/A					N/A					
152Sm	N/A	N/A					N/A					
151Eu	N/A	N/A					N/A					
153Eu	N/A	N/A					N/A					
155Gd	N/A	N/A					N/A					
233U	1.56	1.74E-10				0.00%	3.27E-05				0.93%	
234U	N/A	N/A					N/A					
235U	1.00	3.38E-04	7.64E-04	3.33%	2.26E+00	44.24%	5.41E-04	5.47E-04	2.39%	1.01E+00	98.89%	7.16E-01
236U	N/A	N/A					N/A					
238U	N/A	N/A					N/A					
237Np	0.02	3.76E-07				0.05%	9.69E-07				0.18%	
238Pu	0.18	1.02E-06				0.13%	0.00E+00				0.00%	
239Pu	1.56	3.11E-04				40.71%	0.00E+00				0.00%	
240Pu	0.03	1.99E-06				0.26%	0.00E+00				0.00%	
241Pu	3.50	1.09E-04				14.29%	0.00E+00				0.00%	
242Pu	0.01	1.29E-07				0.02%	0.00E+00				0.00%	
241Am	0.03	2.95E-07				0.04%	0.00E+00				0.00%	
242mAm	53.81	1.98E-06				0.26%	0.00E+00				0.00%	
243Am	0.02	5.34E-08				0.01%	0.00E+00				0.00%	

Table A-3. Absorption Equivalency Evaluation at 5 Years and 1 Million Years for PWR SNF (4.5 wt.% U-235 and 40 GWd/MTU).

Isotope	**Absorption Ratio Relative to Sm-150**	**Sm-150 Equivalency at 5 Years**				**Sm-150 Equivalency at 1 Million Years**				**Change Due to Decay**
		Concentration (Atoms/b-cm)	**Concentration (Atoms/b-cm)**	**Ratio to Original Concentration**	**Contribution Fraction**	**Concentration (Atoms/b-cm)**	**Concentration (Atoms/b-cm)**	**Ratio to Original Concentration**	**Contribution Fraction**	
16O	1.90E-06	8.84E-08			0.00%	8.84E-08			0.00%	
95Mo	1.34E-01	7.44E-06			0.31%	7.44E-06			0.42%	
99Tc	2.28E-01	1.26E-05			0.52%	0.00E+00			0.00%	
99Ru	7.24E-02	0.00			0.00	4.00E-6			0.22%	
101Ru	5.20E-02	2.69E-06			0.11%	2.69E-06			0.15%	
103Rh	1.44E+00	4.43E-05			1.83%	4.43E-05			2.48%	
109Ag	9.10E-01	4.55E-06			0.19%	4.55E-06			0.25%	
143Nd	3.25E+00	1.35E-04			5.55%	1.35E-04			7.53%	
145Nd	5.00E-01	1.60E-05			0.66%	1.60E-05			0.89%	
147Sm	5.70E-01	5.40E-06			0.22%	5.40E-06			0.30%	
149Sm	4.01E+02	8.15E-05			3.36%	8.15E-05			4.56%	
150Sm	1.00E+00	1.35E-05	2.42E-03	1.80E+02	0.56%	1.35E-05	1.79E-03	1.32E+02	0.76%	7.38E-01
151Sm	1.52E+02	1.11E-04			4.56%	0.00E+00			0.00%	
152Sm	2.06E+00	1.12E-05			0.46%	1.12E-05			0.63%	
151Eu	9.20E+01	2.74E-06			0.11%	6.98E-05			3.90%	
153Eu	3.12E+00	1.61E-05			0.66%	1.61E-05			0.90%	
155Gd	6.09E+02	8.28E-05			3.42%	1.66E-04			9.26%	
233U	4.55E-01	5.10E-11			0.00%	1.49E-06			0.08%	
234U	9.98E-01	5.50E-06			0.23%	0.00E+00			0.00%	
235U	9.88E-01	3.34E-04			13.78%	5.34E-04			29.87%	
236U	5.09E-02	7.08E-06			0.29%	9.97E-06			0.56%	
238U	2.68E-02	5.92E-04			24.43%	5.93E-04			33.14%	
237Np	1.76E+00	2.83E-05			1.17%	7.31E-05			4.09%	
238Pu	5.40E+00	3.14E-05			1.29%	0.00E+00			0.00%	
239Pu	2.69E+00	5.39E-04			22.22%	0.00E+00			0.00%	
240Pu	2.90E+00	1.64E-04			6.78%	0.00E+00			0.00%	
241Pu	3.62E+00	1.13E-04			4.66%	0.00E+00			0.00%	
242Pu	1.85E-01	2.05E-06			0.08%	0.00E+00			0.00%	
241Am	5.87E+00	5.93E-05			2.45%	0.00E+00			0.00%	
242mAm	3.30E+00	1.21E-07			0.01%	0.00E+00			0.00%	
243Am	7.51E-01	2.00E-06			0.08%	0.00E+00			0.00%	