QA: QA

ANL-WIS-MD-000004 REV 04

November 2004

DSNF and Other Waste Form Degradation Abstraction

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ANL-WIS-MD-000004 REV 04

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OCRWM	MODEL SIGNATURE PAGE/CHANGE HISTORY	Page iii 1. Total Pages: 66

2. Type of Mathematical Model

☐ Process Model ☒ Abstraction Model ☐ System Model

Describe Intended Use of Model

Describe degradation of the DSNF inventory in the Total System Performance Assessment for the License Application using an approach that does not differentiate between codisposal waste packages containing different DSNF types.

3. Title

DSNF and Other Waste Form Degradation Abstraction

4. DI (including Rev. No., if applicable):

ANL-WIS-MD-000004 REV 04

5. Total Appendices	6. No. of Pages in Each Appendix
None	Not Applicable

	Printed Name	Signature	Date
7. Originator	Jim Cunnane	Neil K. Brown for	11/19/04
8. Independent Technical Reviewer	Pasu Pasupathi	Pasupathi	11/19/04
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10. QER	Charlie Warren		11/19/04
11. Responsible Manager/Lead	Dennis Thomas		11/19/04
12. Responsible Manager	Neil Brown		11/19/04

13. Remarks

Change History

14. Revision No.	15. Description of Change
00	Initial Issue
01	Extensive changes to model documentation. Clarify recommended model validation requirements, potential test work in support of model validation, and treatment of potential colloidal release forms. Reword assumptions concerning the dissolution kinetics of naval SNF to clarify that the CSNF dissolution model will bound the dissolution kinetics of the naval SNF. Revise immobilized Pu ceramic conservative degradation model. Revise HLW glass degradation model recommended for immobilized ceramic plutonium waste form to conform to that reported in ANL-EBS-MD-000016 REV00. Revise U-metal SNF degradation model to include results of National Spent Nuclear Fuel Program report on oxidation kinetics. Revise CSNF model recommended as representing naval SNF to conform to that finally given in ANL-EBS-MD-000015 REV00. Add (Section 6.4.3 and Table 3b) discussion of exposed surface areas for the various DSNF and immobilized Pu ceramic waste forms. Add discussion of the potential impact of unresolved TBVs in the Conclusions Section.

OCRWM	MODEL CHANGE HISTORY (CONTINUED)	Page iv
		1. Total Pages: 66

3. Title
DSNF and Other Waste Form Degradation Abstraction
4. DI (including Rev. No., if applicable):
ANL-WIS-MD-000004 REV 04

Change History (Continued)

14. Revision No.	15. Description of Change
01 / 01	Replace temporary references and/or Input transmittals with final reports in text, in the References section, and on the DIRS report. Adjust status of some DIRS items to reflect AP-3.15 Rev.2 ICN 0 requirements. Re-designate Inputs from Section 4, which have TBVs associated with them as Assumptions and modify the associated DIRS report accordingly.
02	Extensive modifications to model documentation; revisions are too extensive for the use of change bars. Added an Attachment addressing included FEPs related to DSNF degradation in the TSPA, and an Attachment providing the description of Included FEPs supported by the Model Report. Added a section (8.4) addressing the use of the Model Report in resolving KTI agreements. Added a section (8.3) addressing uncertainties in the recommended degradation model and expanded the discussion of model validation (7). Reformatted/renumbered Section 7.2 to become Section 8 (Conclusions) in response to REV 01 ICN00 of AP-SIII.10Q. A revision to modeling report. *Waste Form Colloid-Associated Concentrations Limits: Abstraction and Summary.* ANL-WIS-MD-000012 REV 01A is in check and review in parallel with this Model Report and its usage in this Model Report has been given TBV-4746. Change the status of all naval SNF-related DIRS items to "N/A – Naval Reactor Program" per the status descriptions in AP-3.15Q, Rev.3, ICN 4 Attachment 4. Clarify discussion in Section 6.3.1 of the origin of the naval SNF release source term. Changed the status of DIRS items per AP-3.15Q Rev 4. Use NSNFP final oxidation report (DOE 2000d), updated DOE SNF Information report (DOE 2002a), the Gray and Einziger (1998) report, and the fact that there is only a very small quantity of non-N-reactor SNF metallic uranium-based SNF, to document a rationale for removing TBV-4983 and TBV-4984 (assumptions that the degradation rate of uranium metal-based SNF may be represented by a simple enhancement of the oxidation rate in water of unirradiated and uncorroded uranium metal, and that the dissolution behavior of all metallic uranium-based (Group 7) SNF may be represented by that of the N reactor SNF. Update SNF Group 9 (aluminum-based SNF) model in Section 6.3.9 to incorporate results presented in DOE (2000d) (NSNF oxidation rate study) for the aqueous corrosion rates of aluminum. Update SNF Group 2 Pu/U alloy degradation models based on updated NSNF DOE SNF Information report (DOE 2002a). Use analysis in Section 6.3.12 to document a rationale for removing TBV-4985 and its associated assumption - that the degradation behavior of the immobilized Pu ceramic is typical of that of a titanate-based pyrochlore ceramic. Correct factor of two conversion factor error in column four and footnote (c) of Table 9. Recommend specific surface area (7.0×10^{-5} m^2/g [m^2 of waste form surface area per gram of heavy metal]) to be used in analyses of waste packages containing N reactor SNF and recommend previous DSNF specific surface area (0.03 m^2/g) for use only on non-N reactor SNF. Include option in Section 8 to use the Group 7 conservative model as being equivalent to the upper limit model for practical TSPA time steps. The approval of this Model Report supports the closure of TER-02-0063.

OCRWM	MODEL CHANGE HISTORY (CONTINUED)	Page v
		1. Total Pages: 66

3. Title
DSNF and Other Waste Form Degradation Abstraction
4. DI (including Rev. No., if applicable):
ANL-WIS-MD-000004 REV 04

Change History (Continued)

14. Revision No.	15. Description of Change
03	Complete revision to address issues raised during regulatory integration. Change bars were not used because the changes were too extensive to track.
04	Revised Model Validation section and included Acknowledgement page.

This report was developed through the contributions of Thomas Thornton, BSC, who was the major author of prior revisions of this report.

EXECUTIVE SUMMARY

Several hundred distinct types of DOE-owned spent nuclear fuel (DSNF) may potentially be disposed in the Yucca Mountain repository. These fuel types represent many more types than can be viably individually examined for their effect on the Total System Performance Assessment for the License Application (TSPA-LA). Additionally, for most of these fuel types, there is no known direct experimental test data for the degradation and dissolution of the waste form in repository groundwaters. The approach used in the TSPA-LA model is, therefore, to assess available information on each of 11 groups of DSNF, and to identify a model that can be used in the TSPA-LA model without differentiating between individual codisposal waste packages containing different DSNF types.

The purpose of this report is to examine the available data and information concerning the dissolution kinetics of DSNF matrices for the purpose of abstracting a degradation model suitable for use in describing degradation of the DSNF inventory in the Total System Performance Assessment for the License Application. The data and information and associated degradation models were examined for the following types of DSNF:

- Group 1 - Naval spent nuclear fuel
- Group 2 - Plutonium/uranium alloy (Fermi 1 SNF)
- Group 3 - Plutonium/uranium carbide (Fast Flux Test Facility-Test Fuel Assembly SNF)
- Group 4 - Mixed oxide and plutonium oxide (Fast Flux Test Facility-Demonstration Fuel Assembly/Fast Flux Test Facility-Test Demonstration Fuel Assembly SNF)
- Group 5 - Thorium/uranium carbide (Fort St. Vrain SNF)
- Group 6 - Thorium/uranium oxide (Shippingport light water breeder reactor SNF)
- Group 7 - Uranium metal (N Reactor SNF)
- Group 8 - Uranium oxide (Three Mile Island-2 core debris)
- Group 9 - Aluminum-based SNF (Foreign Research Reactor SNF)
- Group 10 - Miscellaneous Fuel
- Group 11 - Uranium-zirconium hydride (Training Research Isotopes–General Atomics SNF).

The analyses contained in this document provide an “upper-limit” (i.e., instantaneous degradation) model for use in the TSPA-LA model. “Best-estimate” models for the degradation of the fuels in each of the DSNF groups are discussed to provide a basis for selecting the upper limit model for use in the TSPA-LA model. The instantaneous degradation model is chosen for use in the TSPA-LA model because the available information shows that the degradation rate of the N Reactor fuel (which constitutes most of the DSNF inventory) is very high and because the available qualified information is insufficient to justify use of a less conservative approach.

The commercial spent nuclear fuel model will be used for naval spent nuclear fuel because it has been shown to be conservative for representing naval spent nuclear fuel.

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CONTENTS

Page

CONTENTS (Continued)

Page

TABLES

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ACRONYMS

ANL	Argonne National Laboratory
ASTM	American Society for Testing and Materials
DBE	Design Basis Event
DOE	U.S. Department of Energy
DSNF	DOE-owned spent nuclear fuel
LLNL	Lawrence Livermore National Laboratory
MTHM	metric tons heavy metal
MWd	megawatt days
NNPP	Naval Nuclear Propulsion Program
NSNFP	National Spent Nuclear Fuel Program
OCRWM	Office of Civilian Radioactive Waste Management
PNNL	Pacific Northwest National Laboratory
SNF	spent nuclear fuel
TSPA	Total System Performance Assessment
TSPA-LA	Total System Performance Assessment for the License Application
TSPA-SR	Total System Performance Assessment for the Site Recommendation
TSPA-VA	Total System Performance Assessment for the Viability Assessment

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1. PURPOSE

1.1 PURPOSE OF THE MODEL REPORT

The purpose of this model report is to abstract a model that is suitable for use in the TSPA-LA model to describe the degradation rate of spent nuclear fuel (SNF) in the possession of the U.S. Department of Energy (DOE). This nuclear fuel is referred to as DOE-owned spent nuclear fuel (DSNF).

1.2 BACKGROUND

Several hundred distinct types of DSNF may eventually be emplaced in the repository (DOE 2002 [DIRS 158405], Section 5.2 and Appendix D). It is not practical to attempt to determine the impact of each individual type on any resulting site dose. Eleven general groups have been established to represent the entire inventory for the TSPA-LA model. To enable analyses of a limited number of types to represent, or bound, the behavior of all DSNF disposed in the repository, the DOE Office of Civilian Radioactive Waste Management (OCRWM) and the National Spent Nuclear Fuel Program (NSNFP) collaborated to identify the DSNF groups. These groups are individual categories of DSNF for TSPA analysis. The description of, and justification for, these DSNF groupings for repository criticality, design basis events, and TSPA analysis purposes are in the NSNFP report, *DOE Spent Nuclear Fuel Grouping in Support of Criticality, DBE, TSPA-LA* (DOE 2000 [DIRS 118968]). Also, during the Site Recommendation period, the NSNFP compiled a report, *DOE Spent Nuclear Fuel Information in Support of TSPA-SR* (DOE 2002 [DIRS 158405]), which contains postclosure performance characteristics of the groups and suggests models for describing the degradation rates of waste-forms in each group.

The TSPA-LA groups of DSNF contained in the NSNFP documents and described in Sections 6.1.1 through 6.3.12 of this document follow, along with a typical type of DSNF in each group are:

- Group 1 - Naval SNF
- Group 2 - Plutonium/uranium alloy (Fermi 1 SNF)
- Group 3 - Plutonium/uranium carbide (Fast Flux Test Facility-Test Fuel Assembly SNF)
- Group 4 - Mixed oxide and plutonium oxide (Fast Flux Test Facility-Demonstration Fuel Assembly/Fast Flux Test Facility-Test Demonstration Fuel Assembly SNF)
- Group 5 - Thorium/uranium carbide (Fort St. Vrain SNF)
- Group 6 - Thorium/uranium oxide (Shippingport light water breeder reactor SNF)
- Group 7 - Uranium metal (N Reactor SNF)
- Group 8 - Uranium oxide (Three Mile Island-2 core debris)
- Group 9 - Aluminum-based SNF (Foreign Research Reactor SNF)
- Group 10 - Miscellaneous Fuel
- Group 11 - Uranium-zirconium hydride (Training Research Isotopes–General Atomics SNF).

1.3 OBJECTIVE

The objective of this model report is to use the group-related studies, published analyses, and results of experimental degradation tests to abstract a model for the degradation rate of the DSNF inventory based on consideration of available information for the fuels in the DSNF groups. The model is applicable to the conditions after waste package breach when the waste forms are exposed to water or water vapor and air. The model is also to be suitable for use in a TSPA-LA approach that does not differentiate between codisposal waste packages containing different DSNF types.

This activity is covered under *Technical Work Plan for: Regulatory Integration Modeling and Analysis of the Waste Form and Waste Package* (BSC 2004 [DIRS 171583]).

This model report does not use input from any other analysis reports or model reports. This model report provides direct input to the TSPA-LA model by specifying that all DSNF (with the exception of naval DSNF) should be modeled as degrading instantaneously. Naval DSNF should be modeled using the degradation rate of commercial spent nuclear fuel (CSNF).

2. QUALITY ASSURANCE

Abstraction of the DSNF model and the supporting activities have been determined to be subject to *Quality Assurance Requirements Description* (DOE 2004 [DIRS 171539]), in accordance with Section 8 of *Technical Work Plan for: Regulatory Integration Modeling and Analysis of the Waste Form and Waste Package* (BSC 2004 [DIRS 171583]). This report does not address items or barriers on the Q-List.

Control of the electronic management of data was accomplished in accordance with the controls specified by Appendix A of *Technical Work Plan for: Regulatory Integration Modeling and Analysis of the Waste Form and Waste Package* (BSC 2004 [DIRS 171583]).

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3. USE OF SOFTWARE

No computer software was used in the development of the abstracted upper-limit DSNF model described in this report.

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4. INPUTS

4.1 DIRECT INPUT

There are no direct inputs to the abstraction of the DSNF upper-limit model developed in this model report. Information and experimental data from sources in the open literature are used as indirect input in Section 6 to describe the basis for the abstracted model.

No parameter values are used as direct input to this model report.

4.2 CRITERIA

Section 3.4 of *Project Requirements Document* (Canori and Leitner 2003 [DIRS 166275]) contains requirements relevant to repository postclosure modeling. The key requirements (referred to by their PRD requirement identifier) relevant to this model report are:

1. PRD-002/T-014 (Canori and Leitner 2003 [DIRS 166275]) "Performance Objectives for the Geologic Repository After Permanent Closure."

   This specifies the repository performance objectives that must be met following permanent closure. It includes a requirement for multiple barriers and limits on radiological exposure.

2. PRD-002/T-015 (Canori and Leitner 2003 [DIRS 166275]) "Requirements for Performance Assessment."

   This specifies the technical requirements to be used in performing a performance assessment. It includes requirements for calculations, including data related to site geology, hydrology, variability in the models, and deterioration or degradation processes, including waste form degradation.

3. PRD-002/T-016 (Canori and Leitner 2003 [DIRS 166275]) "Requirements for Multiple Barriers."

   This specifies the requirements for the identification of the repository multiple barriers, describing the capabilities of the barriers to isolate waste, and provide the technical bases for the capability descriptions.

The technical work plan for this model report (BSC 2004 [DIRS 171583], Table 3-1) contains a table of acceptance criteria based on the requirements in *Yucca Mountain Review Plan, Final Report* (NRC 2003 [DIRS 163274]). Applicable Acceptance Criteria are presented below. Section 8.3 quotes the full text that pertains to the specific criteria with pointers to the information within this report that pertains to the criteria.

System Description and Demonstration of Multiple Barriers Acceptance Criteria

The following Acceptance Criteria are from Section 2.2.1.1.3 of *Yucca Mountain Review Plan, Final Report* (NRC 2003 [DIRS 163274]).

- Acceptance Criterion 1—Identification of Barriers Is Adequate
- Acceptance Criterion 2—Description of Barrier Capability to Isolate Waste Is Acceptable
- Acceptance Criterion 3—Technical Basis for Barrier Capability Is Adequately Presented.

Degradation of Engineered Barriers

The following Acceptance Criteria are from Section 2.2.1.3.1.3 of *Yucca Mountain Review Plan, Final Report* (NRC 2003 [DIRS 163274]).

- Acceptance Criterion 1—System Description and Model Integration are Adequate
- Acceptance Criterion 2—Data Are Sufficient for Model Justification
- Acceptance Criterion 3—Data Uncertainty is Characterized and Propagated through the Model Abstraction
- Acceptance Criterion 4—Model Uncertainty is Characterized and Propagated Through the Model Abstraction
- Acceptance Criterion 5—Model Abstraction Output Is Supported By Objective Comparisons.

Radionuclide Release Rates and Solubility Limits Acceptance Criteria

The following Acceptance Criteria are from Section 2.2.1.3.4.3 of *Yucca Mountain Review Plan, Final Report* (NRC 2003 [DIRS 163274]).

- Acceptance Criterion 1—System Description and Model Integration Are Adequate
- Acceptance Criterion 2—Data Are Sufficient for Model Justification
- Acceptance Criterion 3—Data Uncertainty Is Characterized and Propagated Through the Model Abstraction
- Acceptance Criterion 4—Model Uncertainty Is Characterized and Propagated Through the Model Abstraction
- Acceptance Criterion 5—Model Abstraction Output Is Supported by Objective Comparisons.

4.3 CODES AND STANDARDS

American Society for Testing and Materials (ASTM) Standard C 1174-97 [DIRS 105725], *Standard Practice for Prediction of the Long-Term Behavior of Materials, Including Waste Forms, Used in Engineered Barrier Systems (EBS) for Geologic Disposal of High-Level Radioactive Waste*, is used to support the degradation model development methodology.

5. ASSUMPTIONS

One assumption is used in this document to support the output.

5.1 COMMERCIAL SNF AS SURROGATE FOR NAVAL SNF

Assumption: It is assumed that commercial light water reactor SNF can be used as a surrogate for naval SNF under the range of expected repository environmental conditions.

Rationale: To provide a conservative simplification for the TSPA-LA model, the commercial SNF radionuclide release model is used to model release rates from naval SNF. Naval SNF represents a very small fraction of the SNF MTHM inventory (DOE 2002 [DIRS 158405], Table D-1). Releases from naval SNF waste packages are expected to be lower than for commercial SNF (BSC 2001 [DIRS 152059], Section 6.5). Because of its design, the radionuclide releases from naval SNF waste packages are considerably less than releases from commercial light water reactor SNF waste packages; accordingly, this assumption is conservative. This assertion is supported by the demonstration by the Naval Nuclear Propulsion Program (NNPP) that doses from the radionuclides released from naval SNF are less than the doses from radionuclides released from commercial SNF. There are 300 waste packages containing naval SNF compared to the commercial spent nuclear fuel (CSNF) inventory of approximately 7,500 waste packages. A comparison with an equivalent amount of Zircaloy-clad CSNF indicates that the total dose for the first one million years from the TSPA simulation, using the commercial-fuel equivalent, is more than four orders of magnitude higher than the total dose from the source-term simulation for naval SNF (BSC 2001 [DIRS 152059], p. 36 and Figure 6.1-2). Therefore, it is conservative to model naval SNF as CSNF.

Confirmation Status: This assumption will be appropriately confirmed by the naval SNF disposition program in a planned classified Naval Nuclear Propulsion Program Addendum to the License Application.

Use in the Model: This assumption is used in Section 6 to support the use of light water reactor SNF as a conservative surrogate for the naval SNF.

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6. MODEL DISCUSSION

10 CFR Part 63 [DIRS 156605] requires the DOE to perform a systematic analysis of features, events, and processes that could potentially affect the performance of the geologic repository. These analyses provide the technical basis for the inclusion or exclusion of specific features, events, and processes in the TSPA. The evaluation methodology provides an approach for considering the possible future states of the repository system, and begins with identification of features, events, and processes, followed by a rigorous screening that identifies those features, events, and processes that would be directly addressed in the TSPA-LA model.

The development of a comprehensive list of features, events, and processes potentially relevant to postclosure performance of the potential Yucca Mountain repository is an ongoing, iterative process based on site-specific information, design, and regulations. The approach for developing an initial list in support of *Total System Performance Assessment for the Site Recommendation* (CRWMS M&O 2000 [DIRS 153246]) was documented in *The Development of Information Catalogued in REV00 of the YMP FEP Database* (Freeze et al. 2001 [DIRS 154365]). The initial list contained 328 features, events, and processes, of which 176 were included in TSPA-SR models (CRWMS M&2000 [DIRS 153246], Tables B-9 through B-17). To support the TSPA-LA model, the list was reevaluated in accordance with *The Enhanced Plan for Features, Events, and Processes (FEPs) at Yucca Mountain* (BSC 2002 [DIRS 158966], Section 3.2). Table 6-1 provides a list of features, events, and processes included in TSPA-LA models that are described in this model report. The table also summarizes their implementation in the TSPA-LA model and provides specific references to sections within this document**.**

Two features, events, and processes are primarily addressed in this report: FEP 2.1.02.01.0A, DSNF Degradation (Alteration, Dissolution, and Radionuclide Release), and FEP 2.1.02.28.0A, Grouping of DSNF Types into Categories.

Table 6-1. Included Features, Events, and Processes addressed in this Model Report

FEP No.	FEP Name	Section Where the FEP is Addressed
2.1.02.01.0A	DSNF Degradation (Alteration, Dissolution, and Radionuclide Release)	6.1
2.1.02.28.0A	Grouping of DSNF Waste Types into Categories	1.2

Because an upper-limit bounding model is to be used to describe the rate of degradation of DSNF in the TSPA-LA model (Section 8), the effects of potential interactions between codisposed waste on the DSNF degradation rate (i.e., the part of FEP 2.1.01.02.0B that pertains to this report) are not explicitly considered. However, the DSNF instantaneous degradation model, which is to be used in the TSPA-LA model, conservatively bounds the effects of such interactions on the DSNF degradation rate.

For each DSNF category or group this section examines two types of DSNF degradation models for possible TSPA-LA application purposes: best-estimate and upper limit.

Preliminary analyses of the DSNF in the repository (Thornton 1998 [DIRS 125082]; Thornton 1998 [DIRS 107796]) had indicated that for large incremental time steps (>1,000 years), the dose at the compliance point on the site boundary resulting from the failure of waste

packages containing DSNF is very insensitive to the release rates from the DSNF waste form. Subsequent, more detailed, analyses (CRWMS M&O 2000 [DIRS 125038], Section 7, bullets 11 to 16; CRWMS M&O 2000 [DIRS 153595], Section 6; BSC 2003 [DIRS 168796], Section 3.3.6) have supported the conclusion that even complete and instantaneous degradation of the DSNF waste results in calculated doses at the compliance point on the site boundary that are below established limits. Also, as discussed below, examination of the available experimental or analytical degradation rate behavior data for the DSNF groups shows that, in general, insufficient qualified data exists to formulate or abstract separate models for each group.

6.1 MODEL SELECTION AND ABSTRACTION

In the following sections, the best-estimate model for each of the DSNF groups is examined to assess both the model's defensibility and the barrier capability of DSNF, as indicated by the best-estimate model. The degradation modes of interest are oxidation, corrosion, and dissolution of the waste forms in air, water vapor, and liquid water. These modes, as well as their associated radionuclide release rates, are encompassed under the general heading of degradation model. The radionuclide release rate is related to the degradation rate models for each waste form through the simplification that all components of the DSNF waste form are released congruently with the degradation of the matrix. The release rate of radionuclide i (R_i) is related to the rate of waste form degradation as follows:

$$R_i = D \times A \times F \quad \text{(Eq. 1)}$$

where

D = degradation rate (mg/m^2·day)
A = total surface area of waste form exposed to water (m^2)
F = mass fraction of species i

In using the best-estimate models, the release rate of radionuclides from the DSNF is calculated by multiplying the dissolution rate provided by the model(s) by the total surface area of the DSNF exposed to water and by the inventory of the radionuclides.

A list of information sources used to provide the rationale for using an upper-bound instantaneous release model for DSNF degradation in the TSPA-LA model follows:

- Several reports (Thornton 1998 [DIRS 125082]; Thornton 1998 [DIRS 107796]; CRWMS M&O 2000 [DIRS 125038], Sections 6.7.1 and 7, Bullets 11-16; BSC 2003 [DIRS 168796], Section 3.3.6) provide analyses demonstrating that the contribution of the DSNF to the postclosure site dose is negligible, even when extremely conservative degradation and pyrophoric behavior of the DSNF is used. *Performance Assessment Sensitivity Analyses of Selected U.S. Department of Energy Spent Fuels* (CRWMS M&O 2000 [DIRS 125038], Section 6.7.1 and 7, Bullets 11 to 16) provides analyses that show that the dose rates at the site boundary resulting from the failure of DSNF-containing waste packages are insensitive to the degradation rates of the DSNF waste forms. Input values for the degradation rate used in the sensitivity analyses did not require qualification because it is a parametric study that used the maximum conceivable degradation rates of the waste form to perform the analysis. The analyses in

these documents do not require qualification because they do not provide direct input to the abstraction of any of the degradation models.

- *DOE Spent Nuclear Fuel Grouping in Support of Criticality, DBE, TSPA-LA* (DOE 2000 [DIRS 118968]) establishes and justifies the DSNF groups to be used for TSPA-LA analysis purposes and proposes that the degradation behavior of each DSNF group can be adequately represented by the behavior of a surrogate SNF type within the group. The conclusions are supported by TSPA analyses (Thornton 1998 [DIRS 125082]; Thornton 1998 [DIRS 107796]; CRWMS M&O 2000 [DIRS 125038], Sections 6.7.1 and 7, Bullets 11 to 16; CRWMS M&O 2000 [DIRS 153595], Section 6; BSC 2003 [DIRS 168796], Section 3.3.6). These analyses show that the postcontainment site-boundary dose is insensitive to the degradation rate of the DSNF or individual components of the DSNF. This information does not require qualification because no parameter is directly used in selecting or abstracting degradation models. Appendix D of *DOE Spent Nuclear Fuel Grouping in Support of Criticality, DBE, TSPA-LA* (DOE 2000 [DIRS 118968]) contains the equivalent metric tons of heavy metal (MTHM) inventory for each of the DSNF groups in the Total System Performance Assessment for the Site Recommendation (TSPA-SR). The data used in this model report compares the relative importance of each group in formulating a composite degradation model for all DSNF. While the data are required to be qualified for other documents supporting the TSPA-LA model, they are used here to indicate the relative quantities of the DSNF groups only. Therefore, they do not require qualification for use in abstracting group dissolution models in this report.

- DOE reports on SNF (DOE 1999 [DIRS 107790], DOE 2000 [DIRS 152658], DOE 2002 [DIRS 158405]) contain proposed oxidation rate and dissolution models and MTHM inventory for each of the DSNF TSPA groups identified in *DOE Spent Nuclear Fuel Grouping in Support of Criticality, DBE, TSPA-LA* (DOE 2000 [DIRS 118968]). The suggested models were evaluated in this model report. *DOE Spent Nuclear Fuel Information in Support of TSPA-SR* (DOE 1999 [DIRS 107790], Appendix B) and *DOE Spent Nuclear Fuel Information in Support of TSPA-SR* (DOE 2002 [DIRS 158405], Appendix B) contain information on the surface area for each of the TSPA-LA DSNF groups, and also contain the equivalent MTHM inventory. The information in *DOE Spent Nuclear Fuel Information in Support of TSPA-SR* (DOE 1999 [DIRS 107790]) is used for comparative purposes only, to indicate the conservatism inherent in the upper limit dissolution model analyzed in this report.

The following sections describe the upper-limit and best-estimate dissolution models developed for each DSNF group using the available information sources. Table 6-2 contains a summary of these models. This table shows the two types of models for each DSNF group, the surrogate material for which the model was developed, and the literature or document source reference for the model. A discussion of the individual models for each fuel group follows.

Table 6-2. DSNF, Naval SNF, Plutonium Disposition Release/Degradation Models

	Upper-Limit Model			Best-Estimate Model		
DSNF Group	**Surrogate**	**Model**	**Ref**	**Surrogate**	**Model**	**Ref.**
1. Naval	N/A	Section 6.3.1	N/A	Section 6.3.1	Section 6.3.1	N/A
2. Plutonium / Uranium Alloy	N/A	Instantaneous release upon exposure to groundwater	N/A	uranium - molybdenum	(semi-empirical) rate (mg metal/cm^2/h) = 1.15 × 10^8 exp{(-66,500 ± 12,200 J/mol)/RT} [100–178°C] rate (mg metal/cm^2/h) = 1.58 × 10^6 exp{(-80,500 ± 10,600 J/mol)/RT} [304-440°C] (Linear interpolation between 178°C and 304°C)	DOE 2002 [DIRS 158405], Section 6.2, Equations 6-1 and 6-2
3. Plutonium / Uranium Carbide	N/A	Instantaneous release upon exposure to groundwater	N/A	uranium metal	100 × Unirradiated uranium metal best-estimate: k (mg/m^2-day) = 100 × {1.21 × 10^{15} exp(-66.4 ± 2.0 kJ/mol /RT)}	DOE 2002 [DIRS 158405], Section 6.7, Equation 6-11
4. Mixed Oxide and Plutonium Oxide	N/A	Instantaneous release upon exposure to groundwater	N/A	light water reactor SNF	(semi-empirical) uranium oxide best-estimate model	DOE 1999 [DIRS 107790], Section 6.4
5. Thorium / Uranium Carbide	N/A	Instantaneous release upon exposure to groundwater	N/A	SiC	(semi-empirical) R (kg/m^2-s) = 0.6 × 10^{-12}	DOE 1999 [DIRS 107790], Section 6.5 and Figure 6-3
6. Thorium / Uranium Oxide	N/A	Instantaneous release upon exposure to groundwater	N/A	Synroc	(semi-empirical) k (mg/m^2·day) = 82.0 × 10$^{(-1,000/TK)}$	(1-3)
7. Uranium Metal-Based	N/A	Instantaneous release upon exposure to groundwater	N/A	N Reactor	(semi-empirical) 2.52 × 10^{10}exp (-66,400/RT) mg/cm^2-hr R = 8.314 J/mol-K	Section 6.7; DOE 2002 [DIRS 158405], Section 6.7, Equation 6-11
8a. Intact Uranium Oxide	N/A	Instantaneous release upon exposure to groundwater	N/A	light water reactor SNF	(semi-empirical) uranium oxide best-estimate model	DOE 1999 [DIRS 107790], Sections 6.4 and 6.8

Table 6-2. DSNF, Naval SNF, Plutonium Disposition Release/Degradation Models (Continued)

	Upper-Limit Model			Best-Estimate Model		
DSNF Group	**Surrogate**	**Model**	**Ref**	**Surrogate**	**Model**	**Ref.**
8b. Damaged Uranium Oxide	N/A	Instantaneous release upon exposure to groundwater		Three Mile Island-2 debris	(surface area enhancement factor of 100 is based on professional judgment) 100 × uranium oxide best-estimate	Section 6.3.7
9. Aluminum-based	N/A	Instantaneous release upon exposure to groundwater	N/A	Savannah River Site uranium/ aluminum SNF in J-13 well water	(empirical) 1.38 mg metal/m^2·day at 25°C 13.80 mg metal/m^2·day at 90°C	Wiersma and Mickalonis 1998 [DIRS 107984], Tables 3 and 4
10. Miscellaneous	N/A	Instantaneous release upon exposure to groundwater	N/A	N/A	(empirical) rate (mg metal/cm^2/h) = 1.15×10^8 exp{(-66,500 ± 12,200 J/mol)/RT} [100–178°C] rate (mg metal/cm^2/h) = 1.58×10^6 exp{(-80,500 ± 10,600 J/mol)/RT} [304°C to 440°C]	DOE 2002 [DIRS 158405], Section 6.2, Equations 6-1 and 6-2
11. Uranium-Zirconium Hydride	N/A	Instantaneous release upon exposure to groundwater	N/A	Training Research Isotopes–General Atomic	(empirical) 0.1 × uranium oxide best estimate	DOE 1999 [DIRS 107790], Section 6.11; LMITCO 1997 [DIRS 125091], Section 2.1.8

6.1.1 DSNF Group 1 (Naval SNF)

The Naval Nuclear Propulsion Program has demonstrated that the dissolution rate of naval SNF is lower than the dissolution rate of commercial SNF (BSC 2001 [DIRS 152059], Section 6.5). A planned classified Naval Nuclear Propulsion Program Addendum to the License Application will provide the details of this evaluation. To provide a conservative simplification for the TSPA-LA model, the commercial SNF dissolution model will be used for the dissolution of the naval SNF (Assumption 5.1).

6.1.2 DSNF Group 2 (Plutonium/Uranium Alloy)

There are several individual types of plutonium/uranium alloy-based DSNF primarily comprised of uranium-molybdenum, although smaller quantities of uranium-thorium alloy are part of this group (DOE 2002 [DIRS 158405], Sections 5.3.1.2 and 6.2). These alloy fuels are generally clad in zirconium alloy, but some small quantities have aluminum, stainless steel, or tantalum alloy cladding. The largest single fuel type in this group, comprising slightly over 90% by weight of uranium of the total Group 2 inventory, are the zirconium-clad uranium-molybdenum Fermi Unit 1 SNF. Studies of the dissolution behavior of uranium-molybdenum and uranium-zirconium alloys, reported in Section 6.2 of *DOE Spent Nuclear Fuel Information in Support of TSPA-SR* (DOE 2002 [DIRS 158405]), give dissolution rates for the alloy that depend on the amount of alloying molybdenum and zirconium. The corrosion behavior of uranium-molybdenum and uranium-zirconium alloys reported in *DOE Spent Nuclear Fuel Information in Support of TSPA-SR* (DOE 2002 [DIRS 158405]) supported the following correlations for the corrosion rate (in mg metal/$cm^2 \cdot h$) of Group 2 SNF based on uranium-molybdenum alloy:

For the range of temperatures between 100°C and 178°C:

$$\text{rate} = 1.15 \times 10^8 \exp[(-66{,}500 \pm 12{,}200 \text{ J/mol})/RT_k] \quad \text{(Eq. 2)}$$

For the range of temperatures between 304°C and 440°C:

$$\text{rate} = 1.58 \times 10^6 \exp[(-80{,}500 \pm 10{,}600 \text{ J/mol})/RT_k] \quad \text{(Eq. 3)}$$

Equation 3 covers a temperature range higher than that expected for waste form temperature in codisposal packages. However, it can be used to interpolate rates in the interval between 178°C and 304°C for waste package temperatures in that range. These equations are based on uranium-molybdenum alloys with, respectively, molybdenum less than 8% by weight and molybdenum greater than 8% by weight. They predict corrosion rates approximately a factor of 40 lower than rates predicted for unirradiated metallic uranium and a factor of 200 lower than the rates predicted for N Reactor SNF. Section 6.2 of *DOE Spent Nuclear Fuel Information in Support of TSPA-SR* (DOE 2002 [DIRS 158405]) also noted that the observed corrosion rate for uranium-molybdenum alloys was 1.5 to 5 orders of magnitude lower than that of bare uranium metal (Group 7). The information in Figures 6-1 and 6-2 of *DOE Spent Nuclear Fuel Information in Support of TSPA-SR* (DOE 2002 [DIRS 158405]) showed that the corrosion rate of uranium-zirconium alloys is over two orders of magnitude lower than that of the uranium-molybdenum alloy from 75°C to 200°C.

Equations 2 and 3 should be used for the best-estimate models for the Group 2 SNF. It should also be noted that the total inventory of this SNF waste form is small compared to most other DSNF types (Table 6-3). These equations were derived from data in the open literature for the corrosion of unirradiated metallic uranium and uranium alloy, and the suggested best-estimate degradation model for Group 2 SNF for unirradiated uranium alloys was obtained from Section 6.2 of *DOE Spent Nuclear Fuel Information in Support of TSPA-SR* (DOE 2002 [DIRS 158405]). However, the uranium-molybdenum and uranium-zirconium corrosion data used for the best-estimate model did not include irradiation effects, for which there are no known data. Because the data upon which the models in this reference are based is from open literature sources, it is from uncontrolled sources and, thus, would need to be qualified to support direct use of these models in the TSPA-LA model.

Table 6-3. Approximate Dissolution Rates at pH 8.5, 0.002 Molar CO_3^{2-}, and 0.20 Atmospheres Oxygen Calculated from the Best-Estimate Models

DSNF GROUP	Approximate Fuel Matrix Density,ρ [a] in g/cm^3 ($\rho/\rho_{Group\ 7}$)	Inventory (MTHM)[b]	Best-Estimate Corrosion Rate [c] at 50°C (mg/m^2·day) (adjusted for density)
Group 2 - Plutonium/Uranium Alloy	19.05[d] (1)	8.5	492
Group 3 - Plutonium/Uranium Carbide	11.28 (0.59)	0.1	1.09×10^6
Group 4 - Mixed Oxide and Plutonium Oxide	11.03 (0.58)	11.59	5.41
Group 5 - Thorium/Uranium Carbide	9.35 (0.49)	24.52	0.025
Group 6 - Thorium/Uranium Oxide	9.87 (0.52)	46.98	0.034
Group 7 - Uranium Metal-Based	19.05 (1)	1,984.81	1.1×10^5
Group 8a - Intact Uranium Oxide [e]	9.86[f] (0.52)	166.2 (8a + 8b)	4.83
Group 8b - Damaged Uranium Oxide	9.86 (0.52)	—	483
Group 9 - Aluminum-Based	2.70[g] (0.14)	19.54	0.19 at 25°C
Group 10 - Miscellaneous SNF	19.05[d] (1)	4.24	492
Group 11 - Uranium-Zirconium Hydride	6.88 (0.36)	1.51	0.33

NOTES: [a] DOE 2002 [DIRS 158405], Appendix C, Attachment A.
[b] DOE 2002 [DIRS 158405], Appendix D, Radionuclide Inventory Summary of DOE SNF.
[c] The tabulated rates are adjusted for density as follows rate = rategroup i × (ρi/ρgroup 7).
[d] Conservatively taken to be that of metallic uranium.
[e] Using Equation 7 with nominal burnup of 40 MWd/kgU.
[f] Estimating that the actual density after irradiation of the oxide fuel is 90% of the theoretical density of 10.96 g/cm^3.
[g] For the purpose of this comparison the dissolution rate of the aluminum matrix is used.

6.1.3 DSNF Group 3 (Plutonium/Uranium Carbide SNF)

Group 3 SNF consists primarily of fuel from the Fast Flux Test Facility-Test Fuel Assembly with most of the balance from the sodium reactor experiment. Both consist of mixed-carbide-fissile fuel particles in a nongraphite matrix. Only a very limited amount of information concerning the chemical reactivity of the DSNF in this group is available. Because of the lack of specific information concerning degradation behavior and the indication that the fissile particles could be very reactive, the best-estimate rate models should be a rate that is very high, arbitrarily taken as

100 times the dissolution rate of the uranium metal SNF (DOE 2002 [DIRS 158405], Section 6.3). In applying the model, the rate of dissolution should be taken as the congruent dissolution of the composite carbide matrix and fuel particles with the radionuclide content of its particles released as the composite degrades.

The degradation model for Group 3 SNF was obtained from Section 6.3 of *DOE Spent Nuclear Fuel Information in Support of TSPA-SR* (DOE 2002 [DIRS 158405]). The model in this reference is based on sparse data or information from old open literature sources and would need to be qualified to support direct use of these models in the TSPA-LA model.

6.1.4 DSNF Group 4 (Mixed Oxide and Plutonium Oxide SNF)

Group 4 SNF is composed of a mixture of uranium and plutonium oxides with various cladding materials. Although several dozen SNF types are in this category, the predominant types are the Fast Flux Test Facility, Demonstration Fuel Assembly, and Test Demonstration Fuel Assembly fuel, contributing over 83% of the mass of heavy metal (DOE 2002 [DIRS 158405], Section 5.3.1.4). Since the fuel material is either uranium oxide or plutonium oxide, the dissolution kinetics of the fuel form is not expected to be materially different from that for commercial light water reactor SNF. The best-estimate models abstracted are those for the light water reactor SNF.

The degradation model for Group 4 SNF is abstracted in *CSNF Waste Form Degradation: Summary Abstraction* (CRWMS M&O 2000 [DIRS 136060]) for commercial UO_2 fuel. The data or information upon which the suggested model in this reference is based is qualified data.

6.1.5 DSNF Group 5 (Thorium/Uranium Carbide SNF)

This SNF group consists primarily of thorium or uranium-carbide particles coated with pyrolytic carbon or silicon carbide embedded in a carbonaceous matrix. Over 90% by weight of MTHM of this group is Fort St. Vrain SNF, with the remainder being Peach Bottom Unit 1 SNF (DOE 2002 [DIRS 158405], Section 5.3.1.5). The Peach Bottom fuel may be more damaged than the Fort St. Vrain fuel, although there is little qualified information concerning the condition of either. Fuel in this group, whose protective silicon carbide coatings and matrix are intact, would be expected to have dissolution kinetics similar to those of pure silicon carbide, and this would provide the basis for the best-estimate model for this group. Spent nuclear fuel with a damaged coating or matrix would be expected to have dissolution kinetics similar to uranium carbide (DOE 2002 [DIRS 158405], Section 6.5; CRWMS M&O 1998 [DIRS 100362], Section 6.3.2.1), and this should provide the basis for a conservative model for this group.

The suggested degradation model for Group 5 SNF was obtained from Section 6.5 of *DOE Spent Nuclear Fuel Information in Support of TSPA-SR* (DOE 2002 [DIRS 158405]). The data or information upon which the suggested model in this reference is based is from open literature sources and would need to be qualified to support direct use of these models in the TSPA-LA model. The specific surface area used should be taken as 0.022 m^2/g (DOE 2002 [DIRS 158405], Appendix B).

6.1.6 DSNF Group 6 (Thorium/Uranium Oxide SNF)

Thorium/uranium oxide SNF in Group 6 primarily consists of the Shippingport light-water breeder reactor reflector SNF with the remainder from the Dresden Unit 1 and the Experimental Research Reactor thorium/uranium oxide SNF. The Shippingport fuel was clad in zirconium alloy, and the Dresden and Experimental Research Reactor fuel was clad in stainless steel. The thorium/uranium oxide fuel consisted of sintered pellets similar to commercial light water reactor fuel pellets.

Several reports discussed in *DOE Spent Nuclear Fuel Information in Support of TSPA-SR* (DOE 1999 [DIRS 107790]) and Section 6.6 of *DOE Spent Nuclear Fuel Information in Support of TSPA-SR* (DOE 2002 [DIRS 158405]) indicate that the mixed thorium uranium was more corrosion resistant than pure uranium dioxide. The approach taken here, for the best estimate model, is to use a ceramic Synroc™ release model that conservatively bounded data for unirradiated light water breeder reactor fuel (DOE 2002 [DIRS 158405], Section 6.6). This model and the supporting data would need to be qualified to support direct use of the model in the TSPA-LA model

6.1.7 DSNF Group 7 (Uranium Metal SNF)

The zirconium-clad N Reactor SNF constitutes over 95% of the mass of this group, with small quantities of aluminum-clad-single-pass reactor and experimental breeder reactor metallic uranium SNF (DOE 2002 [DIRS 158405], Section 5.3.1.7). Due to the small quantity of other uranium metal-based DSNF compared to the N Reactor SNF, the degradation of the other SNF may be taken as similar to the N Reactor SNF. A significant fraction of the N Reactor fuel is visibly damaged, and much of the rest could have small pinholes/cracks in the cladding. The exposed uranium metal surfaces of the N Reactor fuel elements show extensive corrosion resulting from the many years of direct exposure to the K-basin water (Abrefah et al. 1995 [DIRS 151125], Section 3.1 and Figures 3.1-3.5; Abrefah et al. 1999 [DIRS 151226], Figure 3.1; Welsh et al. 1997 [DIRS 151225], Section 1, Figures 3.3 to 3.7). N Reactor SNF comprises approximately 85% by weight in MTHM of the total quantity of DSNF (DOE 2002 [DIRS 158405], Appendix D).

Experimental studies have been conducted in the past on unirradiated and uncorroded uranium metal and uranium-metal alloy, and some of this work is summarized in Section 6.7 of *DOE Spent Nuclear Fuel Information in Support of TSPA-SR* (DOE 2002 [DIRS 158405]). Uranium metal degradation behavior is obtained from the following references:

- The information in *DOE Spent Nuclear Fuel Information in Support of TSPA-SR* (DOE 2002 [DIRS 158405], Section 6.7) is used to abstract a temperature dependence for the Arrhenius form of the Group 7 dissolution model. The analyses of the dissolution or oxidation reaction kinetics of uranium metal found in *DOE Spent Nuclear Fuel Information in Support of TSPA-SR* (DOE 2002 [DIRS 158405]) include a comprehensive evaluation of the available literature concerning the temperature dependence of the oxidation and dissolution rates of bare uranium metal. These analyses were conducted under an NSNFP quality assurance program similar to *Quality Assurance Requirements and Description* (DOE 2004 [DIRS 171539]). The temperature

dependence developed in the report is used to abstract the temperature dependence for the dissolution of the Group 7 DSNF. The temperature dependence of these reactions is developed in *DOE Spent Nuclear Fuel Information in Support of TSPA-SR* (DOE 2002 [DIRS 158405]). This NSNFP report meets the definition of product output for use in this model report in Attachment 3 of AP-3.15Q, *Managing Technical Product Inputs*.

- Gray and Einziger (1998 [DIRS 109691]) report the results of dissolution testing of irradiated N Reactor SNF.

- Abrefah et al. (1996 [DIRS 162037], Section 3.4), Abrefah, Buchanan, and Marschman (1998 [DIRS 159159], Section 3.2), Abrefah et al. (1999 [DIRS 151226]), Abrefah and Sell (1999 [DIRS 118528], Section 4), Abrefah et al. (2000 [DIRS 153644]), Goldberg (2000 [DIRS 159482]), Kaminski (2001 [DIRS 161679]), Totemeier et al. (1999 [DIRS 159408]), and Tyfield (1988 [DIRS 159597]), along with *DOE Spent Nuclear Fuel Information in Support of TSPA-SR* (DOE 2002 [DIRS 158405]), provide information on the oxidation rate in moist air and the aqueous dissolution rate and chemical reactivity of uranium metal-based fuel, both unirradiated and irradiated.

- "Corrosion of Reactor Grade Uranium in Aqueous Solutions Relevant to Storage and Transport." (Tyfield 1988 [DIRS 159597]) contains the results of a study of the dissolution kinetics of Magnox SNF in aqueous environments under various pH conditions.

Tests by Tyfield (1988 [DIRS 159597]) were performed on samples of Magnox SNF, a nuclear fuel design similar to the N Reactor fuel, except that the Magnox cladding was aluminum, rather than zirconium, alloy. The Magnox SNF did not exhibit the kind of physical damage that much of the N Reactor SNF showed, and therefore the uranium metal did not have the prolonged direct exposure to water that the N Reactor SNF has had. This fuel had burnups ranging from 1,100 to 9,850 MWd/t (reasonably typical of the N Reactor SNF), but had displayed more extensive localized irradiation swelling (ranging from 5% to 50%) compared to swelling of approximately a few percent observed on the N Reactor SNF (Abrefah et al. 1995 [DIRS 151125], Section 3.2.1). The tests in the study by Tyfield (1988 [DIRS 159597]) also used samples with initially uncorroded surfaces, and were conducted in both fluoride-free and 1 kg/m^3 fluoride-doped water solutions. Similarly, the studies by Abrefah, Buchanan, and Marschman (1998 [DIRS 159159]) and Abrefah and Sell (1999 [DIRS 118528]) were performed on irradiated N Reactor fuel samples taken from a section of the assembly that had not been in direct contact with the K-basin water, and therefore did not have corroded uranium metal surfaces. The study by Kaminski (2001 [DIRS 161679]) was performed in EJ-13 water on samples of unirradiated N Reactor fuel obtained from archive materials. And unlike the studies by Tyfield (1988 [DIRS 159597]), Abrefah, Buchanan, and Marschman (1998 [DIRS 159159]) and Abrefah and Sell (1999 [DIRS 118528]), the Kaminski (2001 [DIRS 161679]) tests were performed under anoxic water conditions, whereas the repository environment is expected to be oxic. Other studies had indicated that the rate of degradation of uranium metal in an anoxic water environment is greater than in an oxic environment (DOE 2000 [DIRS 152658], Figures 2-9 and 2-10).

Additionally, the Tyfield (1988 [DIRS 159597]) study reported an activation energy for oxidation of 60 kJ/mol over the temperature range of 75°C to 145°C. The study by Abrefah, Buchanan, and Marschman (1998 [DIRS 159159], Section 3.2) had indicated an activation energy for oxidation in moist air of 67 kJ/mol, and Abrefah and Sell (1999 [DIRS 118528], Equation 4.3) had indicated an activation energy for oxidation in moist helium of 13.3 kcal/mol (55.7 kJ/mol). These values are very comparable with the value of 66.4 ± 2 kJ/mol (DOE 2002 [DIRS 158405]) for the Group 7 models discussed below.

The degradation of all uranium metal-based DSNF is taken to be similar to that observed by Gray and Einziger (1998 [DIRS 109691]) for N Reactor SNF specimens. This dissolution rate is further supported by a study by Abrefah et al. (2000 [DIRS 153644]) concerning the dissolution behavior of unirradiated and uncorroded N Reactor fuel in which they concluded that the dissolution behavior of the uranium metal in the fuel was qualitatively similar to, but significantly less than, that for the N Reactor SNF observed by Gray and Einziger (1998 [DIRS 109691]). The data contained in the report were generated under Pacific Northwest National Laboratory's (PNNL's) quality assurance requirements and have been widely accepted and used in the analysis of N Reactor SNF behavior, in particular by the NSNFP (DOE 2002 [DIRS 158405], Section 6.7; DOE 2000 [DIRS 152658], Section 2.3). Although there is an extensive database for the dissolution of metallic uranium in aqueous environments, the data contained in this report are the only known experimental data specifically for the dissolution behavior of N Reactor SNF. The database for the oxidative dissolution of uranium metal was analyzed by the NSNFP and summarized in (DOE 2000 [DIRS 152658]), and it was concluded that the basic form of the dissolution model, taking account of specific surface area differences, would apply generally to metallic uranium-based SNF inventory. Additionally, the fact that the amount of metallic uranium-based SNF that is not N Reactor fuel is very small (DOE 2002 [DIRS 158405], Section 5.3.1.7) also supports the use of the N Reactor SNF model for all uranium metal-based SNF. Additionally, a study (Tyfield 1988 [DIRS 159597]) concerning the dissolution behavior of Magnox SNF, a metallic uranium-based nuclear fuel similar to the N Reactor SNF, showed similar dissolution kinetics.

The degradation rate for uranium metal-based (i.e., N Reactor) DSNF in water can be regarded as a simple enhancement of the oxidation rate of unirradiated and uncorroded uranium metal given in NSNFP reports (DOE 2000 [DIRS 152658], Section 2.3; DOE 2002 [DIRS 158405], Section 6.7). This rate is supported by test results reported by Abrefah et al. (2000 [DIRS 153644]) concerning the dissolution behavior of unirradiated and uncorroded N Reactor fuel. From the testing results they concluded that the dissolution behavior of the uranium metal in the fuel was qualitatively similar to, but significantly less than, that for the N Reactor SNF observed by Gray and Einziger (1998 [DIRS 109691]). *Review of Oxidation Rates of DOE Spent Nuclear Fuel, Part 1: Metallic Fuel* (DOE 2000 [DIRS 152658]) and *DOE Spent Nuclear Fuel Information in Support of TSPA-SR* (DOE 2002 [DIRS 158405]) are reports generated by the NSNFP that contain analyses of the oxidation rates of metallic uranium in dry air, humid air, saturated water vapor, and liquid water. The data analyzed in the NSNFP reports represent a comprehensive compilation of the available literature on uranium metal oxidation. The basis for using this rate is the similarity of the temperature dependence for the oxidation given in the DOE report and that for N Reactor SNF shown in Gray and Einziger (1998 [DIRS 109691]). The NSNFP reports compile reference data from numerous publications that have been used extensively in the past for the analysis of the behavior of metallic uranium. The reports also

provide a correlation for the oxidation rates of metallic uranium based on literature data for unirradiated and uncorroded uranium metal. The correlation provided in Section 6.7 of *DOE Spent Nuclear Fuel Information in Support of TSPA-SR* (DOE 2002 [DIRS 158405]) forms a partial basis for the best-estimate dissolution rate expression for N Reactor SNF used in this report.

The NSNFP has conducted an analysis of the kinetics of uranium metal oxidation from several literature sources (DOE 2000 [DIRS 152658], Sections 2.2.3 and 2.2.4; DOE 2002 [DIRS 158405], Equation 6-11; DTN: MO0210SPAOXIDA.001 [DIRS 160443]) from which the Equation 4 expression for the dissolution kinetics of unirradiated and uncorroded uranium metal in water was derived from Equation 6-11 of *DOE Spent Nuclear Fuel Information in Support of TSPA-SR* (DOE 2002 [DIRS 158405]):

$$k\ (\text{mg U/cm}^2\cdot\text{hr}) = 5.03 \times 10^9 \exp(-A/RT), \text{ or}$$

$$k\ (\text{mg U/m}^2\cdot\text{day}) = 1.21 \times 10^{15} \exp(-A/RT)\ (20°C \text{ to } 200°C) \qquad \text{(Eq. 4)}$$

where

k = the corrosion rate
A = the activation energy for dissolution (66.4 ± 2.0 kJ/mol)
R = the gas constant (8.314 J/mol-K)
T = the temperature in kelvins

The Arrhenius temperature dependence of these reactions is developed in *DOE Spent Nuclear Fuel Information in Support of TSPA-SR* (DOE 2002 [DIRS 158405]). Additionally, PNNL has conducted flow-through dissolution tests on samples of the N Reactor SNF. These tests generally indicate that the rates of dissolution of this SNF exceed that of the unirradiated or uncorroded uranium metal or alloy. The PNNL experimental work was performed on N Reactor SNF samples cut from an undamaged/uncorroded area of an N Reactor fuel element (Gray and Einziger 1998 [DIRS 109691], Section 2.2 and Figure 1). The test results indicated that there were two stages in the dissolution behavior of the N Reactor SNF samples tested: an initial Stage 1 rate and, after an incubation period, a significantly higher Stage 2 dissolution rate.

This Stage 1 dissolution of the matrix was correlated with the Equation 5 expression (Gray and Einziger 1998 [DIRS 109691], Equation 3):

$$\log_{10}(R)\ [\text{mg/m}^2\cdot\text{day}] = 8.52 + 0.347 \log_{10}([CO_3^{2-}]) + 0.088\ \text{pH} - 1929/T \qquad \text{(Eq. 5)}$$

where

R = the dissolution rate expressed in $\text{mg/m}^2\cdot\text{day}$,
$[CO_3^{2-}]$ = the molar concentration of carbonate in the contacting solution,
T = the absolute temperature.

Unsaturated testing of metallic uranium showed that alteration phases form that can retard the release of solutes from the N Reactor SNF (Goldberg 2000 [DIRS 159482], unnumbered paragraphs labeled “Unsaturated Corrosion Tests” and “Discussion and Summary”). This may explain the lower Stage 1 rates.

During Stage 2, a sludge-like corrosion product material was formed consisting primarily of U_4O_9 and the mineral form schoepite (UO_3 $2H_2O$) (Gray and Einziger 1998 [DIRS 109691], Section 4.2). The rates of dissolution that were observed for Stage 2 reached 13,000 and 180,000 mg/m^2·day at 25°C and 75°C, respectively (Gray and Einziger 1998 [DIRS 109691], Section 4.3). The temperature dependence of the dissolution was similar to that for unirradiated and uncorroded uranium metal in Section 2.2 of *Review of Oxidation Rates of DOE Spent Nuclear Fuel, Part 1: Metallic Fuel* (DOE 2000 [DIRS 152658]), although the absolute rates were higher for the N Reactor SNF (see Table 6-4). Table 6-5 gives values for the Stage 1 dissolution rate as calculated from Equation 5 and for the experimental Stage 2 dissolution rates of the PNNL study. In most of the cases where Stage 2 dissolution was initiated, it began around 60 days into the test, a very short period in terms of TSPA analyses.

Table 6-4. Comparison of Unirradiated/Uncorroded Uranium Metal and N Reactor SNF Corrosion Rates

Temp (°C)	N Reactor SNF[a] (mg/m^2·day)	Uranium Metal[b] (mg/m^2·day)	Uranium Metal[c] (mg/m^2·day)	5× Uranium Metal[b] (mg/m^2·day)	25× Uranium Metal[b] (mg/m^2·day)
25	1.3×10^4	2.8×10^3	6.2×10^3	1.4×10^4	6.9×10^4
75	1.8×10^5	1.3×10^5	2.6×10^5	6.5×10^5	3.3×10^6

NOTES: [a] From Gray and Einziger 1998 [DIRS 109691], Section 4.3.
[b] From Equation 4 using the nominal activation energy of 66.4 kJ/mol.
[c] From Equation 4 using the 1σ minimum activation energy of 64.4 kJ/mol.

Table 6-5. Summary and Comparison of N Reactor SNF Stage 1 and Stage 2 Dissolution Rates

T(°C)	CO_3^{2-} (molar)	pH	HNO_3 (molar)	J-13 well water	Measured[a] Stage 1 Rate (mg/m^2·day)	Calculated[b] Stage 1 Rate (mg/m^2·day)	Measured[c] Maximum Stage 2 Rate (mg/m^2·day)
25	2×10^{-2}	5	—	—	63	79	—[e]
25	2×10^{-2}	8	—	—	160	145	220
25	2×10^{-4}	10	—	—	50	44	100
75	2×10^{-2}	10	—	—	2,100	1,851	4,000
75	2×10^{-4}	5	—	—	200	142	23,000
75	2×10^{-4}	8	—	—	150	250	180,000
25	N/A	5	1×10^{-5}	—	38	—	—[e]
25	N/A	3	1×10^{-3}	—	130	—	—[e]
25	2×10^{-3}	8.5	—	X	—[d]	72	13,000
75	2×10^{-3}	8.5	—	X	—[d]	614	52,000

NOTES: [a] As given in Table 1 of Gray and Einziger 1998 [DIRS 109691].
[b] Calculated using Equation 5: log rate (mg/m^2·day) = 8.52 + 0.347 log[CO_3^{2-}] + 0.088 pH – 1929/TK.
[c] As estimated from the cesium release plots from Figures 3 through 15 of Gray and Einziger 1998 [DIRS 109691].
[d] These samples showed only Stage 2 dissolution behavior.
[e] These samples showed only Stage 1 dissolution behavior.

A significant fraction of the N Reactor fuel has been stored in a damaged condition under water at the K-basins at Hanford, and the metallic uranium has been exposed to the water environment. Therefore, the degradation models for unirradiated and uncorroded uranium metal derived in Section 6.7 of *DOE Spent Nuclear Fuel Information in Support of TSPA-SR* (DOE 2002 [DIRS

158405]) are not recommended for the N Reactor SNF that forms the basis for Group 7. Although the PNNL experimental data is limited, it indicates that Stage 2 dissolution kinetics should be used to predict the repository performance of the N Reactor SNF. Based on these analyses, the abstracted best-estimate degradation rate model for the uranium metal SNF is selected so as to encompass the highest directly observed dissolution rates based on cesium release in the PNNL studies of 1.8×10^5 mg/m^2·day (0.75 mg/cm^2·hr) at 75°C and 13,000 mg/m^2·day (0.05 mg/cm^2·hr) at 25°C (Gray and Einziger 1998 [DIRS 109691], Section 4.3; DTN: MO0210SPAOXIDA.001 [DIRS 160443]). The samples of N Reactor SNF tested in the flow-through testing at PNNL were from an undamaged and uncorroded portion of an N Reactor fuel element. No dissolution testing has been conducted on samples of damaged or corroded N Reactor SNF, although this condition would presumably be the condition of exposure after breach of the waste package in the repository. In other studies at PNNL concerning the ignition properties (Abrefah et al. 1999 [DIRS 151226], Table S.1) and oxidation kinetics (Abrefah et al. 1998 [DIRS 151126], Summary) of N Reactor SNF, and the dissolution behavior of unirradiated N Reactor fuel (Abrefah et al. 2000 [DIRS 153644]), samples of N Reactor SNF taken from the damaged and corroded areas of the N Reactor fuel elements showed higher oxidation rates than undamaged and uncorroded samples.

Table 6-4 compares values for Stage 2 dissolution rates of N Reactor SNF obtained experimentally by Gray and Einziger (1998 [DIRS 109691]) with rates calculated from the correlation for water dissolution of uranium metal given by Equation 4, and 5 times and 25 times this value. Note that the measured reaction rates given for the N Reactor SNF are approximately a factor of two to five higher than those given using Equation 4. Note also that use of the lower (64.4 kJ/mol) one standard deviation (1σ) uncertainty given in the Equation 4 expression for the activation energy (66.4 ± 2.0 kJ/mol) would result in another factor of 2 to 3 in the reaction rate in this temperature range. Thus, the combination of these two factors would result in approximately a factor of 5 increase on the nominal Equation 4 reaction rate, which would represent the most defensible best-estimate model kinetics.

In the PNNL N Reactor SNF experimental work (Gray and Einziger 1998 [DIRS 109691], Section 4.3; DTN: MO0210SPAOXIDA.001 [DIRS 160443]), the highest dissolution rate inferred from cesium releases and corrected for the cesium that was found in the corrosion product was 290,000 mg/m^2·day (1.21 mg/cm^2·hr). This is a factor of 1.6 higher than the maximum value of 180,000 mg/m^2·day observed for dissolution rates based solely on cesium releases to solution. Combining this inferred rate for the N Reactor SNF and a 2σ uncertainty (95%) lower activation energy of 62.4 kJ/mol would result in a higher dissolution rate by factor of 5 over the best-estimate dissolution rate for use in the conservative model.

From the above discussion, the dissolution rate for the best-estimate model for N Reactor SNF and the much smaller quantity of other uranium metal-based DSNF should be taken as 5 times, and a conservative model be taken as 25 times, the model represented by Equation 4 (DOE 2000 [DIRS 152658], Section 2.2; DOE 2002 [DIRS 158405], Section 6.7). Input parameter values used in these models are shown in Table 6-6.

Table 6-6. Input Parameters

Parameter Name	Parameter Source	DTN	Parameter Value	Units
Maximum N Reactor SNF dissolution rate observed at 75°C	Gray and Einziger 1998 [DIRS 109691]	MO0210SPAOXIDA.001 [DIRS 160443]	1.8×10^{5}	$mg/m^2 \cdot day$
Maximum N Reactor SNF dissolution rate observed at 25°C	Gray and Einziger 1998 [DIRS 109691]	MO0210SPAOXIDA.001 [DIRS 160443]	1.3×10^{4}	$mg/m^2 \cdot day$
Arrhenius activation energy for the oxic aqueous dissolution of uranium metal	DOE 2002 [DIRS 158405], Section 6.7	MO0210SPAOXIDA.001 [DIRS 160443]	66.4	kJ/mol
Effective geometric specific surface area and volume (in a waste package) for N Reactor SNF	DOE 2002 [DIRS 158405], Appendices B and C	N/A	7.0×10^{-5} 1.0	m^2/g m^3/pkg

The Arrhenius temperature dependence of the corrosion of uranium metal was the primary information used in developing and corroborating this model. This data set (DTN: MO0210SPAOXIDA.001 [DIRS 160443]) was developed by the NSNFP, and bounding values for the dissolution rate of N Reactor SNF were developed at PNNL.

The corroborative information consists of:

1. Data from Tyfield (1988 [DIRS 159597]) on the corrosion of irradiated uranium metal-based Magnox fuel

2. Data from Kaminski (2001 [DIRS 161679]) on the 90°C batch test corrosion of unirradiated uranium metal in EJ-13 water under anoxic conditions

3. Activation energies for the moist air oxidation and moist helium oxidation of N Reactor SNF from Abrefah, Buchanan, and Marschman (1998 [DIRS 159159], Section 3.2) and Abrefah and Sell (1999 [DIRS 118528], Section 4)

4. The results of dissolution testing on unirradiated N Reactor fuel samples by Abrefah et al. (2000 [DIRS 153644])

5. Qualitative observations and examinations of uranium hydride inclusions under the corroded exposed uranium surfaces of the N Reactor SNF (Abrefah et al. 1996 [DIRS 162037], Section 3.4).

Table 6-7 provides a comparison between the best-estimate model and the models by Tyfield (1988 [DIRS 159597]) and Kaminski (2001 [DIRS 161679]). The Tyfield and best-estimate values are similar at both 30°C and 90°C. Both these correlations relate to irradiated uranium metal dissolution. The Kaminski (2001 [DIRS 161679]) value, which applies to anoxic conditions, is lower by about an order of magnitude.

Table 6-7. Comparison of Irradiated and Unirradiated Uranium Metal SNF Dissolution Rates

Degradation Rate $mgU/cm^2 \cdot hr$	Tyfield 1988 [DIRS 159597][a]	Kaminski 2001 [DIRS 161679][b]	Conservative Model [c]	Best-Estimate Model [c]
30°C, pH 7, fluoride-free	0.045	—	0.18	0.037
30°C, pH 11.5, fluoride-free	0.016	—	0.18	0.037
30°C, pH 7, fluoride-dosed	0.081	—	—	—
30°C, pH 11.5, fluoride-dosed	0.055	—	—	—
90°C, pH~6, EJ-13 water	2.31	0.34	35.1	7.02

NOTES: [a] These data points are approximated from Figure 6 in Tyfield 1988 [DIRS 159597]. The degradation rate values in that figure are given in units of $mgU/m^2 \cdot hr$ and are converted to $mgU/cm^2 \cdot hr$ by multiplying by 10^{-4}. The 90°C (363 K) value is extrapolated from the 30°C (303 K) pH 7 value using the reported Arrhenius activation energy of 60 kJ/mol as follows: k2/k1 = exp{(-60,000 · J/mol/R)(1/T2-1/T1)} = exp{(-60,000 J/mol/8.314 J/mol-K)(1/363 -1/303)} = 51.25 so, k2 = 0.045 × 51.25 = 2.31 $mgU/cm^2 \cdot hr$.

[b] The 90°C pH ~6 data point is obtained from the uranium corrosion rate of 1.9 mg U/day and sample surface area of 23.2 mm^2 (0.232 cm^2) given in Kaminski (2001 [DIRS 161679], pp. 13 and 14). The normalized rate therefore is (1.9 mg U/day)(1/24 day/hr)/(0.232 cm^2) = 0.34 mg $U/cm^2 \cdot hr$.

[c] Table 6-2, Group 7 models.

It has been observed in other analyses of N Reactor SNF that samples having corroded surfaces (similar to the uranium surfaces that would be in contact with an air/water environment typical of repository conditions) exhibit enhanced chemical reactivity-related properties exceeding those of irradiated, but uncorroded, samples (Abrefah et al. 1999 [DIRS 151226]; Abrefah et al. 2000 [DIRS 153644]). Enhanced chemical effects such as these are generally attributed in the literature to the formation of uranium hydride precipitates in the uranium metal during corrosion. The presence of these hydrides has been shown to significantly enhance such aspects of chemical reactivity as oxidation rate and pyrophoricity (e.g., ignition temperature) of corroded uranium surfaces (Abrefah et al. 1999 [DIRS 151226]; Totemeier et al. 1999 [DIRS 159408]).

Metallographic examinations of the corroded uranium metal areas of N Reactor SNF have revealed significant formations of uranium hydride precipitates in the metallic uranium matrix (Abrefah et al. 1996 [DIRS 162037], Section 3.4.1; Abrefah et al. 1999 [DIRS 151226], Section 2.4; Goldberg 2000 [DIRS 159482]). These precipitates were not mentioned in the samples of N Reactor SNF used in the dissolution tests by Gray and Einziger (1998 [DIRS 109691]), which were taken from an area of an N Reactor SNF element far removed from the corroded end of the element. The presence of uranium hydride in the N Reactor SNF samples suggests that the reaction proceeds through uranium hydride and may explain the slow reaction in Stage 1, since the formation of uranium hydride is known to have an induction period that would be consistent with these results.

The studies by Tyfield (1988 [DIRS 159597]), Abrefah et al. (2000 [DIRS 153644]), and Kaminski (2001 [DIRS 161679]) used herein did not include tests with samples of N Reactor SNF with highly corroded metallic uranium surfaces, and only the Tyfield (1988 [DIRS 159597]) study included the effect of pH on the dissolution of DSNF. Abrefah et al. (2000 [DIRS 153644]) testing on unirradiated N Reactor fuel samples included tests at pH values from pH 3 to pH 8, and various water chemistry conditions (Abrefah et al. 2000 [DIRS 153644], Table 1), but the water chemistry effect was minor compared to the SNF results of Gray and Einziger (1998 [DIRS 109691]).

The best-estimate N Reactor SNF release-rate model (as adjusted for the relative density of the particular SNF with respect to that of metallic uranium) is:

$$\text{Rate in units of mg/cm}^2\cdot\text{hr} = 2.52 \times 10^{10} \exp(-66{,}400/RT)(\rho_{matrix}/\rho_{U\text{-}metal})$$

$$\text{Rate in units of mg/m}^2\cdot\text{day} = 6.05 \times 10^{15} \exp(-66{,}400/RT)(\rho_{matrix}/\rho_{U\text{-}metal}) \qquad \text{(Eq. 6)}$$

where

R = the gas constant (8.314 J/mol-K)
ρ_{matrix} = the density of the SNF matrix
$\rho_{U\text{-}metal}$ = the density of uranium metal

Best-estimate calculation of the overall release of radionuclides from a waste package containing N Reactor SNF (i.e., multicanister overpacks), should use the specific surface area for N Reactor SNF (7.0×10^{-5} m^2/g). In TSPA-LA analyses that do not differentiate between codisposal waste packages containing N Reactor SNF and other DSNF types, the waste form specific area of the N Reactor SNF (7.0×10^{-5} m^2/g) should be used. The volume of DSNF in a codisposal waste package should be taken to be the initial volume of N Reactor SNF, or approximately 1.0 m^3/pkg (DOE 2002 [DIRS 158405], Appendix C, Section 5.2).

6.1.8 DSNF Group 8 (Uranium Oxide SNF)

TSPA Group 8 consists of uranium dioxide-based SNF removed from commercial light water reactors or similar SNF from test reactors. About half of the total inventory of approximately 162 MTHM of Group 8 DSNF comes from the Three Mile Island-2 reactor and, therefore, is not in the form of intact fuel rods or assemblies. The other half is substantially undamaged SNF from other commercial reactors. A model (Equation 7) for the dissolution kinetics of light water reactor SNF under neutral-to-alkaline conditions (pH >7) has been developed in *CSNF Waste Form Degradation: Summary Abstraction* (CRWMS M&O 2000 [DIRS 136060] Equation 11, Table 14) based on a functional form of the model originally proposed in Section 6.3.1.3.2 of *Total System Performance Assessment-Viability Assessment (TSPA-VA) Analyses Technical Basis Document* (CRWMS M&O 1998 [DIRS 100362]). The model was derived from qualified flow-through test data for the dissolution kinetics of sintered UO_2 SNF over the pH range 7 to 12. A model (Equation 8) for the dissolution of light water reactor SNF under acidic conditions (pH ≤ 7) was also developed (CRWMS M&O 2000 [DIRS 136060], Equation 18), but from a much more limited data set than the alkaline model.

For pH >7:

$$\log_{10} R = 5.479057 - 2457.050662(1/T) - 1.510878\log_{10}([CO_3^{2-}]) + 1.729906 \log_{10}(O_2) + 0.234718(pH) - 0.799526\log_{10}(BU) - 400.755947\{\log_{10}(O_2)\}\{1/T\} + 780.806133\{\log_{10}(BU)\}\{1/T\} + 0.172305\{\log_{10}(BU)\}\{-\log_{10}([CO_3^{2-}])\} + 0.174428\{\log_{10}(BU)\}\{-\log_{10}(O_2)\} - 0.271203\{\log_{10}(BU)\}\{pH\} - 0.339535\{-\log_{10}([CO_3^{2-}])\}^2 \quad \text{(Eq. 7)}$$

For pH ≤7:

$$\log_{10}(R) = 7.13 - 1{,}085/T - 0.32\,(O_2) - 0.41\,pH \quad \text{(Eq. 8)}$$

where

R is the dissolution rate in $mg/m^2 \cdot day$,
T is the absolute temperature,
$[CO_3^{2-}]$ is the molar concentration of carbonate ion in the liquid phase,
BU is the SNF burnup in megawatt days (MWd)/kgU, and
O_2 is the oxygen partial pressure in atmospheres in the gas phase.

Section 6.8 of *DOE Spent Nuclear Fuel Information in Support of TSPA-SR* (DOE 2002 [DIRS 158405]) suggests a surface-area enhancement factor of 100 for a release model representing the damaged Group 8 SNF. Thus, the abstracted best-estimate degradation model for intact uranium dioxide based Group 8 SNF should be the same as the degradation model for commercial light water reactor SNF (Equations 7 and 8) and the abstracted best-estimate model for the nonintact Group 8 SNF is 100× the intact fuel best-estimate model.

6.1.9 DSNF Group 9 (Aluminum-based SNF)

This group consists of fuels based on a uranium aluminide, uranium silicide, or uranium oxide particle phase dispersed in a continuous aluminum alloy matrix. The fission product radionuclides generally remain in the dispersed phase; therefore, the dissolution of the dispersed phase material is the parameter most germane to the release of the radionuclides upon contact with groundwater. The dissolution rate of interest is expressed in terms of $mgU/m^2 \cdot day$ or $mg\ metal/m^2 \cdot day$.

Approximately 36% of this DSNF is from foreign research reactor sources, with the balance from domestic research reactors such as the Advanced Test Reactor (DOE 2002 [DIRS 158405], Section 5.3.1.9). The Savannah River Site has conducted dissolution studies on Savannah River Site reactor UAl_x, UAl, U_3O_8, and U_3Si_2 dispersion SNF samples at 25°C in J-13 well water and bicarbonate solutions (Wiersma and Mickalonis 1998 [DIRS 107984], Table 3) and on unirradiated samples at 25°C and 90°C (Wiersma and Mickalonis 1998 [DIRS 107984], Table 4). Flow-through dissolution tests at 25°C subsequently performed at PNNL on similar samples gave results similar to the Savannah River Site study (Gray 2000 [DIRS 151876], Table 7). The Savannah River Site data showed dissolution rates for the uranium in the irradiated SNF of 0.14 to 0.22 $mgU/m^2 \cdot day$ in J-13 well water and 22 to 36 $mgU/m^2 \cdot day$ in bicarbonate solution.

The NSNFP has conducted an analysis (DOE 2000 [DIRS 152658], Sections 4.2.1 and 4.3.2) of the available literature concerning the corrosion rates in water of aluminum alloys and unirradiated UAl_x fuel typical of the aluminum-based SNF. As a result of this analysis, the NSNFP recommended the following correlations for the aqueous corrosion of the aluminum alloys and UAl_x:

Aluminum alloy: rate (mg metal/cm^2·hr) = 4.29 exp[(-32,800 ± 1,800 J/mol)/RT]

or rate(mg metal /m^2·day) = 1.03×10^6 exp[(-32,800 ± 1,800 J/mol)/RT] (25-360°C)

(Eq. 9)

UAl_x: rate (mg metal/cm^2·hr) = 5.20 exp[(-34,900 ± 2,300 J/mol)/RT]

or rate (mg metal/m^2·day) = 1.25×10^6 exp[(-34,900 ± 2,300 J/mol)/RT] (178-350°C)

(Eq. 10)

where R = 8.314 J/mol/K and T is in kelvins.

Table 6-8 compares the uranium dissolution results obtained experimentally for irradiated and unirradiated Savannah River Site fuels with the results using the NSNFP correlations. For the purpose of this comparison, the UAl_x correlation, Equation 10, is extrapolated to lower temperatures than the data from which it was derived (DOE 2000 [DIRS 152658], Figure 4-5). This can be done because the aluminum alloy correlation, Equation 9, shows the same log-linear relationship with temperature over a greater temperature range (DOE 2000 [DIRS 152658], Figure 4-4), and the oxidation rates of both the aluminum alloy and the UAl_x is considered to be controlled by the oxidation of the aluminum metal matrix (DOE 2000 [DIRS 152658], Section 4.1). The NSNFP correlations predict an increase in the uranium dissolution rate at 90°C of approximately a factor of 10 over the rate at 25°C, and this is supported by the data for the unirradiated uranium/aluminum alloy fuel samples in J-13 water in the Savannah River Site study.

The best-estimate dissolution model for the aluminum-based SNF is based on values of 0.19 mgU/m^2·day at 25°C and ten times that dissolution rate at 90°C for a nominal uranium content of 13.8% (Wiersma and Mickalonis 1998 [DIRS 107984], Tables 1 and 3). These values translate to a total matrix dissolution rate of 1.38 mg metal/m^2·day at 25°C and 13.80 mg metal/m^2·day at 90°C. This model is primarily based on the Savannah River Site test results for irradiated UAl_x fuel in J-13 water because (1) when normalized to uranium content the UAl_x fuel at 25°C dissolved more rapidly than the U-Al, U_3Si_2, or U_3O_8 dispersion fuel (Wiersma and Mickalonis 1998 [DIRS 107984], Table 3), (2) the groundwater chemistry at the time of waste-package failure is expected to be approximately that of the J-13 well water, rather than the other very low and high pH conditions tested, and (3) the factor of 10 enhancement in the dissolution rate at 90°C is supported by both the unirradiated uranium-aluminum Savannah River Site test results and the NSNFP UAl_x dissolution correlations. In implementing the model, the dissolution rate for a single component should be taken as the total matrix rate multiplied by the inventory fraction of the component.

Table 6-8. Comparison of Measured versus Calculated Uranium Dissolution Rates for Aluminum-Based SNF (in units of mg/m^2·day)

Fuel Type	Uranium Content Fraction	25°C				90°C			
		Calculated	J-13	Low pH	High pH	Calculated	J-13	Low pH	High pH
UAl_x (irradiated)	0.138[a]	0.13[b]	0.19[c]	28[c]	22[c]	1.6[b]	—	—	—
U-Al (irradiated)	0.144[a]	0.27[d]	0.17[c]	99[c]	33[c]	2.8[d]	—	—	—
U-Al (unirradiated)	0.132[e]	0.24[d]	1.7[b]	139[b]	—	2.6[d]	24.2[e]	215[e]	86[e]
U_3O_8 dispersion	0.134[a]	—	0.14[c]	31[c]	33[c]	—	—	—	—
U_3Si_2 dispersion	0.449[a]	—	0.22[c]	36[c]	36[c]	—	—	—	—

NOTES: [a] From Wiersma and Mickalonis 1998 [DIRS 107984], Table 1.
[b] Using Equation 10 with the uranium fraction multiplier.
[c] From Wiersma and Mickalonis 1998 [DIRS 107984], Table 3.
[d] Using Equation 9 with the uranium fraction multiplier.
[e] From Wiersma and Mickalonis 1998 [DIRS 107984], Table 4.

The data for the dissolution of UAl_x in bicarbonate solution documented by Wiersma and Mickalonis (1998 [DIRS 107984], Table 3), and the factor of 10 increase between 25°C and 90°C should be used as the basis for a conservative dissolution model. These values are 22 mgU/m^2·day at 25°C and 220 mgU/m^2·day at 90°C for a nominal uranium content of 13.8% (Wiersma and Mickalonis 1998 [DIRS 107984], Tables 1 and 3). These values translate to a total matrix dissolution rate of about 160 mg metal/m^2·day at 25°C and 1,600 mg metal/m^2·day at 90°C, with linear interpolation of these values for temperatures between 25°C and 90°C.

6.1.10 DSNF Group 10 (Miscellaneous SNF)

Group 10 SNF consists of a small amount of uranium nitride SNF and fuel with unknown matrices (DOE 1999 [DIRS 107790], Section 6.10; DOE 2002 [DIRS 158405], Section 6.10). This group consists of only about 0.2% of the total inventory of DSNF in MTHM. Because of the unknown fuel matrix material, there are no degradation data or analyses specific to this SNF upon which to base a degradation model, and because of the small inventory the impact of the dissolution kinetics on the overall TSPA-LA model is very low. In the absence of information concerning this DSNF type, the dissolution kinetics of Group 10 DSNF is likely to be conservatively represented by the corrosion rate of unirradiated uranium metal. Therefore, the best-estimate and conservative models for this group should be taken as the same as for the Group 2 SNF.

6.1.11 DSNF Group 11 (Uranium-Zirconium Hydride)

Test reactor fuel from Training Research Isotopes–General Atomic comprises approximately 97% of the total Group 11 SNF inventory of approximately 1.5 MTHM (DOE 2002 [DIRS 158405], Section 5.3.1.11 and Appendix D). The Training Research Isotopes–General Atomic fuel consists of a dispersion of fine particles of metallic uranium dispersed in a zirconium hydride matrix (U-Zr-H_x). The fuel matrix is clad in aluminum, stainless steel, or Alloy 800 (LMITCO 1997 [DIRS 125091], Table 2-1). Uranium loadings varied from approximately 8% to 45% by weight (LMITCO 1997 [DIRS 125091], Section 1).

Unirradiated uranium-zirconium hydride fuel has been shown to have good elevated temperature corrosion resistance, but there is no known qualified data for the dissolution rate of this material in repository-relevant water and temperature conditions. However, *Uranium-Zirconium Hydride Fuels for TRIGA Reactors* (LMITCO 1997 [DIRS 125091], p. 2-5) reports a corrosion penetration depth of 2 mils (0.05 mm) for the zirconium hydride matrix after 400 hours in pressurized water at 570°F (300°C). This corresponds to a penetration rate of approximately 1 mm/yr at 300°C. When multiplied by the density of uranium-zirconium hydride of approximately 6.0 g/cm^3 (LMITCO 1997 [DIRS 125091], Section 2.1.5), a penetration rate of 1 mm/yr corresponds to a mass corrosion rate of approximately 16,400 $mg/m^2 \cdot day$. In comparison, the Metals Handbook (ASM International 1987 [DIRS 103753], section on the "Corrosion of Zirconium and Hafnium," Figure 1) gives an oxygen weight-gain corrosion rate for zirconium metal in pressurized water or steam at 315°C of about 2 $g(oxygen)/m^2 \cdot yr$. Accounting for the ZrO_2 stoichiometry of the corrosion product and using 91.2 as the atomic weight of zirconium, the Metals Handbook corrosion rate converts to [2 × (91/16)] or 5.7 $g(Zr)/m^2 \cdot yr$ or approximately 14.5 $mg(Zr)/m^2 \cdot day$. This would indicate that the corrosion rate of the uranium-zirconium hydride is significantly greater than that of pure zirconium.

By way of further qualitative comparison, the corrosion rate of zirconium alloys in various chemical media (such as sodium hydroxide, potassium hydroxide, nitric acid, etc.) at temperatures above approximately 100°C is reported in the Metals Handbook (ASM International 1987 [DIRS 103753], section on the "Corrosion of Zirconium and Hafnium," Table 6) to be generally less than 0.025 mm/yr. A penetration rate of 0.025 mm/yr converts to a corrosion rate of approximately 450 $mg/m^2 \cdot day$. Only very aggressive media (not typical of the expected repository environment) such as highly concentrated acids show corrosion layer penetration rates greater than 0.1 mm/yr.

The NSNFP DSNF information report (DOE 2002 [DIRS 158405], Section 6.11) recommends that the dissolution rate for this DSNF form should be taken as 0.1 times the uranium oxide SNF dissolution rate based on inspections of the fuel assemblies that had shown no observable corrosion or cladding breach, and the judgment that the zirconium fuel matrix should be more stable than uranium oxide.

The NSNFP-recommended value of 0.1 times the dissolution rate of the Group 8 (uranium oxide) degradation rate is used as the best-estimate model, since it more realistically represents the condition of intact cladding, and because the low total inventory in MTHM of the uranium-zirconium hydride SNF makes the dose consequence to the reasonably maximally exposed individual insensitive to the degradation model for this waste form.

The single value rate of 16,400 $mg/m^2 \cdot day$ should be used for a conservative model for the Group 11 DSNF. This value is chosen because it reflects the corrosion data from *Uranium-Zirconium Hydride Fuels for TRIGA Reactors* (LMITCO 1997 [DIRS 125091]), because it may be expected to conservatively bound the expected temperature and chemical environment ranges of the repository, and because it would better represent a breached cladding condition in which water contacts the uranium-zirconium hydride fuel matrix.

6.1.12 Surface Area of DSNF Groups

The best-estimate models for the degradation rates of the DSNF groups are generally expressed in terms of the weight of material released per unit surface area of SNF exposed to water per unit time, typically in units of $mg/m^2 \cdot day$. In order to use the best-estimate models abstracted in this report to calculate the actual release rate from a waste form exposed to water after failure of a waste package, it is, therefore, necessary to use an estimate for the exposed surface area of the fuel. It is not necessary to use a value for the surface area for the upper-limit model since this model, which is total release of the inventory, is not dependent on exposed surface area.

The N Reactor SNF, represented in Group 7, constitutes approximately 85% by weight of the total DSNF inventory, and is known to have extensively damaged cladding, in terms of both gross damage and cracking. Appendix B of *DOE Spent Nuclear Fuel Information in Support of TSPA-SR* (DOE 2002 [DIRS 158405]) calculated the specific exposed metallic uranium surface area for the N Reactor SNF to be 7.0×10^{-5} m^2/g. Appendix B of *DOE Spent Nuclear Fuel Information in Support of TSPA-SR* (DOE 2002 [DIRS 158405]) calculated the geometric surface area of potentially exposed fuel based on the geometric characteristics of representative fuels within the groups. Where the fuel group contained damaged fuel, or fuel suspected to be damaged, a roughness factor for the surface area of 5 was applied. The result is the estimate of exposed surface area per gram of fuel group given in the third column of Table 6-9, and the estimate of the total exposed surface area for each group in the fourth column of the same table. When the best-estimate degradation models for each DSNF group are used, those models should use the estimated surface area for the individual groups in calculating overall release rates for waste packages containing those groups. Due to the great preponderance of the N Reactor SNF in the DSNF inventory—the specific surface area of the N Reactor SNF (7.0×10^{-5} m^2/g) should be used in analyses that do not differentiate between waste packages containing N Reactor SNF and those containing other types of DSNF. The total surface area of DSNF available for reaction in each TSPA-LA time step should be taken to decrease in proportion to the mass of DSNF that has degraded in all previous time steps.

6.2 ABSTRACTION SUMMARY - DSNF DEGRADATION MODELS

A summary of the selected/abstracted group-specific upper-limit, and best-estimate degradation models is given in Table 6-2. Table 6-3 gives a comparison of the dissolution rates calculated by the best-estimate models for each fuel group at 50°C (328 K) and for pH 8, 0.002 molar CO_3^{2-} in the contacting water and 0.20 atmospheres of oxygen in the gas phase. Also included in this table is the approximate inventory in MTHM of each of the DSNF groups. Table 6-9 further gives an example of the mass fractional degradation rate at 50°C that demonstrates the high corrosion rates of these waste forms compared to the long time frames of the TSPA-LA representation. Also, as shown in Table 6-9, the degradation rates of the DSNF types (with the exception of groups 5, 6, 9, and 11, for which the total inventories are small compared to group 7) are high enough to be effectively instantaneous.

The TSPA-LA model requires that the DSNF groups be treated as a single waste type. Therefore, for all 11 DSNF groups, other than the naval DSNF group, the upper-limit model to be used in TSPA-LA analyses is instantaneous degradation of the waste form upon exposure of the waste form to groundwater. This upper-limit model is chosen because most of the best-

estimate models (other than the Group 7 and the naval SNF models) are currently based on limited and unqualified corrosion, dissolution, or oxidation data, and because, as discussed in the preceding paragraph, the degradation of most of the DSNF inventory is effectively instantaneous. The upper-limit model is also chosen in part because earlier TSPA analyses performed for the DSNF (Thornton 1998 [DIRS 125082]; Thornton 1998 [DIRS 107796]) have shown that the overall effect of the failure of DSNF-containing waste packages does not significantly contribute to the dose at the compliance point on the repository site boundary.

Table 6-9. Fractional DSNF Waste Form Dissolution Rates at 50°C, pH 8.5, 0.002 Molar CO_3^{2-}, and 0.20 Atmospheres Oxygen Calculated for Best-Estimate Models

DSNF Group	Best-Estimate Release Rate ($mg/m^2 \cdot day$) [d]	Exposed Specific Surface Area (m^2/g) [a]	Exposed Total Surface Area (m^2) [b]	Fractional Corrosion Rate (d^{-1})
Group 2 - Plutonium/Uranium Alloy	492	1.2×10^{-3}	1.02×10^{4}	5.9×10^{-4}
Group 3 - Plutonium/Uranium Carbide	1.09×10^{5}	2.7×10^{-3}	2.61×10^{2}	0.174
Group 4 - Mixed Oxide and Plutonium Oxide	5.41	4.0×10^{-3} [d]	4.63×10^{4}	2.2×10^{-5}
Group 5 - Thorium/Uranium Carbide	0.025	2.2×10^{-2}	0.54×10^{5}	5.5×10^{-7}
Group 6 - Thorium/Uranium Oxide	0.034	3.6×10^{-4}	1.69×10^{4}	1.2×10^{-8}
Group 7 - Uranium Metal-Based	1.1×10^{5}	7.0×10^{-5}	1.39×10^{5}	7.7×10^{-3}
Group 8a - Intact Uranium Oxide	4.83	4.0×10^{-3}	6.65×10^{5}	1.9×10^{-5}
Group 8b - Damaged Uranium Oxide	483	4.0×10^{-1}	—	0.19
Group 9 - Aluminum-Based	0.19 at 25°C	6.5×10^{-3}	1.27×10^{5}	1.2×10^{-6}
Group 10—Miscellaneous SNF	492	4.0×10^{-1}	1.69×10^{6}	0.2
Group 11—Uranium-Zirconium Hydride	0.33	1.0×10^{-4} [c]	1.51×10^{2}	3.3×10^{-8}

NOTES: [a] DOE 2002 [DIRS 158405], Appendix B, Attachment A and pages 125-126.
[b] Obtained by multiplying the specific surface area given in column 3 in m^2 per gram by the inventory of the DSNF group given in Table 6-3; using 1 MTHM = 1,000 kg = 1,000,000 g. The total MTHM from column 3 of Table 6-3 is approximately 2,333 MTHM or 2.33×10^9 g. Thus the weighted average specific surface area is 2.71×10^6 m^2/ 2.33×10^9 g $\cong$ 0.001 m^2/g.
[c] Conservatively taken to be the same as that of damaged uranium-oxide.
[d] The tabulated rates are adjusted for density as follows rate = rategroup i × (ρi/ρgroup 7).

Commercial light water reactor SNF will be used as the surrogate for naval SNF in repository performance analyses. Expected releases from naval SNF waste packages were provided in *Performance Assessment of U.S. Department of Energy Spent Fuels in Support of Site Recommendation* (BSC 2001 [DIRS 152059]). Because of the robust design of naval SNF, the radionuclide releases from naval SNF waste packages are considerably less than releases from commercial light water reactor SNF waste packages; accordingly, the use of this surrogate is conservative. Naval SNF represents approximately one-tenth of one percent of the SNF MTHM inventory. The information contained in *Revised Source Term for Naval Spent Nuclear Fuel and Special Case Waste Packages* (Mowbray 2000 [DIRS 152077]) is qualified data.

Model Uncertainties—The application of the degradation models abstracted in this document involves the extrapolation of data over periods of time that are orders of magnitude greater than the experimental test periods used to generate the data. The American Society for Testing and Materials recommends that uncertainties in the extrapolation of such models be minimized through the use of models whose mathematical forms are as mechanistic as possible (ASTM C1174-97 1998 [DIRS 105725], Section 24). However, it can be seen from the abstractions above that the lack of any directly relevant experimental dissolution/degradation

data for many of the DSNF waste forms, and the relatively small amount of data for those that have been tested, makes the generation of mechanistic models problematic. Additionally, uncertainties in the data used to generate the models—such as in the surface area measurements used to calculate normalized dissolution rates (Gray and Einziger 1998 [DIRS 109691], Section 2.5)—produce significant uncertainties even in the short-term application of the models. For this reason, and because earlier TSPA analyses (Thornton 1998 [DIRS 125082]; Thornton 1998 [DIRS 107796]) have shown that performance of the repository is very insensitive to the degradation rate of the DSNF, a general upper-limit or bounding degradation model, applicable to all but the naval DSNF, is used in the TSPA-LA analyses. Because the instantaneous release model to be used in the TSPA-LA model—for all groups except Group 1 (i.e., naval SNF)—represents the maximum possible degradation rate of the DSNF inventory, there is no uncertainty associated with its use as a conservative upper-limit model.

6.3 ALTERNATIVE CONCEPTUAL MODELS

The individual DSNF group best-estimate submodels examined in Section 6.1 are themselves alternative conceptual models for the DSNF degradation. Each model could be applied separately in the TSPA-LA model to waste packages containing that particular type of DSNF. A comparison of the corrosion rates predicted from each best-estimate model at 50°C is given in Table 6-3. These alternative models are not used in the TSPA-LA model because insufficient qualified data on the corrosion rates and the surface areas of the fuel in each group are available to justify their use.

6.4 DESCRIPTION OF BARRIER CAPABILITY

The DSNF waste forms provide a barrier to the release of radionuclides because the fuel matrices must degrade before radionuclides are made available to be transported by groundwater. The DNSF waste form will immobilize radionuclides until the waste form is contacted by water and begins to degrade. The DSNF degradation models developed in this model report bound the rate at which radionuclides are made available for transport as either dissolved or colloidal species. The primary focus of this model report is the development of an upper bound model; best estimate mathematical models are used to calculate the degradation rate for DSNF exposed to humid or aqueous exposure conditions and to show that the choice of an upper-bound model is a reasonable model.

Best-estimate degradation rates indicate that the degradation rate of the N Reactor DSNF, which constitutes most DSNF inventory, is very high. On this basis, most of the DSNF inventory is expected to have very limited barrier capability after the fuel is exposed to the repository environment. This, together with the uncertainties in the best-estimate models, leads to the choice of the upper-limit model for use in the TSPA-LA model. Use of this model conservatively indicates that the DSNF does not provide a barrier to the release of radionuclides.

7. VALIDATION

The technical work plan (BSC 2004 [DIRS 171583], Table 2-1) states that "This model does not require validation or confirmatory testing because it represents essentially the instantaneous mobilization of all the available radionuclides from the DSNF within a waste package, and thus the model is a conceptual model not implemented in a mathematical model." However, the instantaneous release model is mathematically implemented within TSPA and, therefore, it is appropriate to validate the model and an exception is taken to the technical work plan (BSC 2004 [DIRS 171583]).

The model is validated per the method described in Section 5.3.2 (c)(2) of AP-SIII.10Q, *Models*. This validation method consists of corroborating model results with alternative mathematical models. The mathematical models developed in Section 6.1, which result in the fractional dissolution rates in Table 6-9, show that Group 7 uranium metal-based fuels are expected to be completely degraded in less than one year of exposure. Group 7 comprises approximately 85% of the total DSNF MTHM and is, thus, representative of the DSNF. Because one year is an insignificant timeframe relative to geologic timeframes, the instantaneous release model is bounding, yet reasonable compared to the alternative mathematical model and is, therefore, validated through corroboration.

Because the technical work plan (BSC 2004 [DIRS 171583]) did not require validation of the model, it also does not provide for an acceptability criterion. For the other Level 1 models, the technical work plan (BSC 2004 [DIRS 171583]) specified a validation metric of "corroborating data must match qualitatively and must be bounded by model predictions." Therefore, the validation criterion is formed by substituting "comparison to alternative mathematical models" for "corroborating data" in the technical work plan (BSC 2004 [DIRS 171583]) criterion for Level 1 models. This results in the applicable criterion: 'alternative mathematical models must match qualitatively and must be bounded by model predictions.' As shown above, this criterion is met and the model is considered to be fully validated.

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8. CONCLUSIONS

8.1 RECOMMENDED DSNF MODEL FOR THE TSPA-LA MODEL

This document assessed the data and models that are available for modeling the degradation rates and associated radionuclide release rates from DSNF. Based on this assessment it is concluded that a conservative bounding approach should be implemented in the TSPA-LA model. The recommended model is an upper-limit model for each DSNF group of instantaneous degradation and complete release of radionuclides upon exposure to groundwater. For Group 7 (metallic uranium SNF), which represents most of the DSNF inventory, the degradation and radionuclide release rates are effectively instantaneous (i.e., occur within a TSPA time step).

Based on the discussion in Section 6.1.12, the volume of DSNF in a codisposal waste package is taken to be the initial volume of N Reactor SNF, or approximately 1.0 m^3/pkg (DOE 2002 [DIRS 158405], Appendix C, Section 5.2).

The outputs from this report are summarized in Table 8-1.

Naval SNF represents approximately one-tenth of one percent of the SNF MTHM inventory. Because of its robust design, the radionuclide releases from naval SNF waste packages would be considerably less than releases from commercial light water reactor SNF waste packages. For this reason commercial light water reactor SNF is used as a conservative surrogate for naval SNF in repository performance analyses.

Table 8-1. Summary of Outputs

Output	Description
Degradation and Radionuclide Release Model for DSNF Groups 2-11	An upper-limit model is the instantaneous release of radionuclides upon exposure to groundwater is used in the TSPA-LA model for each DSNF group, and for the DSNF inventory as a whole.
The volume of DSNF in a codisposal waste package	The volume of DSNF in a codisposal waste package is taken to be the initial volume of N Reactor SNF, or approximately 1.0 m^3/pkg (DOE 2002 [DIRS 158405], Appendix C Section 5.2).

8.2 MODEL AND PARAMETER UNCERTAINTIES

The emphasis in this document is on the basis for using an instantaneous upper-limit degradation model—applicable to all but the naval DSNF—for use in the TSPA-LA model analyses. Because the instantaneous release of the DSNF inventory used in the TSPA-LA model for all groups except Group 1 (i.e., naval SNF) represents the maximum possible rate there is no uncertainty associated with the use of the instantaneous release model as a conservative upper-limit model.

8.3 KEY TECHNICAL ISSUE RESOLUTION AND ACCEPTANCE CRITERIA

The NRC Acceptance Criteria (NRC 2003 [DIRS 163274]) applicable to this model report are identified in Table 3-1 of *Technical Work Plan for: Regulatory Integration Modeling and Analysis of the Waste Form and Waste Package* (BSC 2004 [DIRS 171583]). For each applicable criterion, the criterion is quoted in italics, followed by pointers to where within the model report information addressing the criterion can be found. In some cases, the criterion is

only partially addressed in this report. A demonstration of full compliance requires a review of multiple reports.

8.3.1 System Description and Demonstration of Multiple Barriers

The acceptance criteria in this section are found in Section 2.2.1.1.3 of *Yucca Mountain Review Plan, Final Report* (NRC 2003 [DIRS 163274]).

Acceptance Criterion 1—Identification of Barriers Is Adequate.

> *Barriers relied on to achieve compliance with 10 CFR 63.113(b), as demonstrated in the total system performance assessment, are adequately identified, and are clearly linked to their capability.*

As described in Sections 6.4 and 8.1, the TSPA-LA DSNF model conservatively indicates that the DSNF (with the exception of naval SNF) provides no barrier function in limiting radionuclide release. The basis for modeling the naval SNF is provided elsewhere.

Acceptance Criterion 2—Description of Barrier Capability to Isolate Waste Is Acceptable.

> *The capability of the identified barriers to prevent or substantially reduce the rate of movement of water or radionuclides from the Yucca Mountain repository to the accessible environment, or prevent the release or substantially reduce the release rate of radionuclides from the waste is adequately identified and described:*
>
> *(1) The information on the time period over which each barrier performs its intended function, including any changes during the compliance period, is provided;*
>
> *(2) The uncertainty associated with barrier capabilities is adequately described;*
>
> *(3) The described capabilities are consistent with the results from the total system performance assessment; and*
>
> *(4) The described capabilities are consistent with the definition of a barrier at 10 CFR 63.2.*

As described in Sections 6.4 and 8.1, the TSPA-LA DSNF model conservatively indicates that the DSNF (with the exception of naval SNF) provides no barrier function in limiting radionuclide release. The basis for modeling the naval SNF is provided elsewhere.

Acceptance Criterion 3—Technical Basis for Barrier Capability Is Adequately Presented.

> *The technical bases are consistent with the technical basis for the performance assessment. The technical basis for assertions of barrier capability is*

commensurate with the importance of each barrier's capability and the associated uncertainties.

The technical bases for evaluating the barrier capabilities of the DSNF groups are presented in Section 6.

8.3.2 Degradation of Engineered Barriers

The acceptance criteria in this section are found in Section 2.2.1.3.1.3 of *Yucca Mountain Review Plan, Final Report* (NRC 2003 [DIRS 163274]).

Acceptance Criterion 1—System Description and Model Integration are Adequate

(1) TSPA adequately incorporates important design features, physical phenomena and couplings and uses consistent assumptions throughout the degradation of engineered barriers abstraction process;

The appropriate design features and applicable physical phenomena, as well as environmental factors are discussed in Section 6.

(2) Abstraction uses assumptions, technical bases, data and models that are appropriate and consistent with [those used] in other abstractions.

This model report uses one assumption that is to be confirmed by the naval SNF disposition program. The technical bases for using an upper-limit model in the TSPA-LA model are described in Section 6.

(3) The descriptions of the engineered barriers, design features, degradation processes, physical phenomena, and couplings that may affect the degradation of the engineered barriers are adequate.

Descriptions of the relevant DSNF design features and degradation processes (Sections 6.1.1 through 6.1.12), and the physical phenomena and their couplings that may affect the DSNF degradation are provided.

(4) Initial and boundary conditions are propagated consistently throughout the abstraction process.

The DSNF initial (Sections 6.1.1 through 6.1.12) and relevant environmental boundary conditions are described and used as a basis for selecting an upper-limit bounding (instantaneous degradation) model for use in the TSPA-LA model for all DSNF groups except Group 1.

(5) Sufficient technical basis for the inclusion and exclusion of FEPs are provided;

The features, events, and processes (FEPs) relevant to assessing DSNF degradation and radionuclide release are identified and discussed in Section 6.

(7) Guidance in NUREG 1297 and NUREG 1298 [re: Expert Elicitation] are followed.

Not applicable to this model report because expert elicitation is not used.

Acceptance Criterion 2—Data Are Sufficient for Model Justification

(1) Parameters used to evaluate the degradation of EBS are adequately justified;

Input data are not needed to justify use of the instantaneous degradation model for use in the TSPA-LA model. The basis for modeling the naval SNF is provided elsewhere.

(2) Sufficient data have been collected to establish initial and boundary conditions;

Input data are not needed to justify use of the instantaneous degradation model for use in the TSPA-LA model. The basis for modeling the naval SNF is provided elsewhere.

(3) Data on the degradation of the engineered barriers (e.g. general and localized corrosion, microbially induced corrosion, galvanic interactions, hydrogen embrittlement and phase stability) are based on laboratory measurements, site-specific field measurements, industrial and/or natural analogs and tests designed to replicate anticipated conditions. As appropriate, sensitivity or uncertainty analyses are provided and are shown to be adequate.

Data related to the various potential degradation modes for the DSNF waste form are discussed in detail in Section 6.

Acceptance Criterion 3—Data Uncertainty is Characterized and Propagated through the Model Abstraction

(1) Models use parameter values, assumed ranges, probability distributions and/or bounding assumptions that are technically defensible, reasonably account for uncertainties and variabilities, and do not result in under-representation of the risk estimate.

Not applicable to this report; a conservative upper-limit model is to be used in the TSPA-LA model.

(2) Appropriate parameters, based on techniques that may include laboratory experiments, field measurements, and industrial analogs are used.

Not applicable to this report; a conservative upper-limit model is to be used in the TSPA-LA model.

(3) Assumed range of values and probability distributions for parameters used in conceptual and process-level models are not likely to underestimate the actual degradation and failure of engineered barriers.

Not applicable to this report; a conservative upper-limit model is to be used in the TSPA-LA model.

(5) Where sufficient data do not exist, the definition of parameter values and conceptual models is based on appropriate use of other sources, such as expert elicitation.

Not applicable to this report; a conservative upper-limit model is to be used in the TSPA-LA model.

Acceptance Criterion 4—Model Uncertainty is Characterized and Propagated Through the Model Abstraction

(1) Alternative modeling approaches are considered and are consistent with available data and current scientific understanding.

Alternative modeling approaches that are consistent with available data and current scientific understanding are considered and discussed in Section 6.3. Alternative models are screened out for use in the TSPA-LA model.

(2) Consideration of conceptual model uncertainty is consistent with available site characterization data, laboratory experiments, ... and the treatment of uncertainty does not result in under-estimation of the risk estimate.

Consideration of conceptual and other uncertainties is addressed by using a conservative upper-limit model is to be used in the TSPA-LA model.

(3) Alternative modeling approaches, consistent with available data and current scientific understanding, are used and the modeling results are evaluated using tests that are sensitive to the processes modeled.

Alternative modeling approaches that are consistent with available data and current scientific understanding are considered and discussed in Section 6.3. Alternative models are screened out for use in the TSPA-LA model.

Acceptance Criterion 5—Model Abstraction Output Is Supported By Objective Comparisons

(1) Models implemented in this total system performance assessment abstraction provide results consistent with output from detailed process-level models and or empirical observations (laboratory and field testing, and/or natural analogs).

Not applicable to this report; a conservative upper-limit model is to be used in the TSPA-LA model.

(3) Evidence is sufficient to show that models will not underestimate the actual degradation and failure of engineered barriers.

A conservative upper-limit (instantaneous degradation) model is to be used in the TSPA-LA model.

(4) Mathematical degradation models are based on the same environmental parameters, material factors, assumptions and approximations shown to be appropriate for closely analogous applications.

Not applicable to this report; a conservative upper-limit model is to be used in the TSPA-LA model.

(5) Accepted and well documented procedures are used to construct and test the numerical models that simulate the EB chemical environment and degradation of EB;

Not applicable to this report.

8.3.3 Radionuclide Release Rates and Solubility Limits

The acceptance criteria in this section are found in Section 2.2.1.3.4.3 of *Yucca Mountain Review Plan, Final Report* (NRC 2003 [DIRS 163274]).

Acceptance Criterion 1—System Description and Model Integration Are Adequate

(2) The abstraction of radionuclide release rates and solubility limits uses assumptions, technical bases, data, and models that are appropriate and consistent with other related U.S. Department of Energy abstractions. For example, the assumptions used for this model abstraction are consistent with the abstractions of "Degradation of Engineered Barriers" (Section 2.2.1.3.1); "Mechanical Disruption of Waste Packages" (Section 2.2.1.3.2); "Quantity and Chemistry of Water Contacting Waste Packages and Waste Forms" (Section 2.2.1.3.3); "Climate and Infiltration" (Section 2.2.1.3.5); and "Flow Paths in the Unsaturated Zone" (Section 2.2.1.3.6). The descriptions and technical bases provide transparent and traceable support for the abstraction of radionuclide release rates and solubility limits;

A conservative upper-limit model is to be used in the TSPA-LA model to describe the DSNF degradation and associated radionuclide release rate for all DSNF groups other than Group 1. The basis for modeling the naval SNF is provided elsewhere.

(3) The abstraction of radionuclide release rates and solubility limits provides sufficient, consistent design information on waste packages and engineered barrier systems. For example, inventory calculations and selected radionuclides are based on the detailed information provided on the distribution (both spatially and by compositional phase) of the radionuclide inventory, within the various types of high-level radioactive waste;

The DSNF degradation model reflects available information on DSNF types (Section 6).

> *(4) The U.S. Department of Energy reasonably accounts for the range of environmental conditions expected inside breached waste packages and in the engineered barrier environment surrounding the waste package. For example, the U.S. Department of Energy should provide a description and sufficient technical bases for its abstraction of changes in hydrologic properties in the near field, caused by coupled thermal-hydrologicmechanical- chemical processes;*

Not applicable to this report; a conservative upper-limit model is to be used in the TSPA-LA model.

> *(5) The description of process-level conceptual and mathematical models is sufficiently complete, with respect to thermal-hydrologic processes affecting radionuclide release from the emplacement drifts. For example, if the U.S. Department of Energy uncouples coupled processes, the demonstration that uncoupled model results bound predictions of fully coupled results is adequate;*

Not applicable to this report; a conservative upper-limit model is to be used in the TSPA-LA model.

> *(6) Technical bases for inclusion of any thermal-hydrologic-mechanical-chemical couplings and features, events, and processes in the radionuclide release rates and solubility limits model abstraction are adequate. For example, technical bases may include activities, such as independent modeling, laboratory or field data, or sensitivity studies;*

Not applicable to this report; a conservative upper-limit model is to be used in the TSPA-LA model.

Acceptance Criterion 2—Data Are Sufficient for Model Justification

> *(1) Geological, hydrological, and geochemical values used in the license application are adequately justified. Adequate description of how the data were used, interpreted, and appropriately synthesized into the parameters is provided;*

Not applicable to this report; a conservative upper-limit model is to be used in the TSPA-LA model.

> *(2) Sufficient data have been collected on the characteristics of the natural system and engineered materials to establish initial and boundary conditions for conceptual models and simulations of thermal-hydrologic-chemical coupled processes. For example, sufficient data should be provided on design features, such as the type, quantity, and reactivity of materials, that may affect radionuclide release for this abstraction;*

Not applicable to this report; a conservative upper-limit model is to be used in the TSPA-LA model.

(4) The corrosion and radionuclide release testing program for high-level radioactive waste forms intended for disposal provides consistent, sufficient, and suitable data for the in-package and in-drift chemistry used in the abstraction of radionuclide release rates and solubility limits. For expected environmental conditions, the U.S. Department of Energy provides sufficient justification for the use of test results, not specifically collected from the Yucca Mountain site, for engineered barrier components, such as high-level radioactive waste forms, drip shield, and backfill.

Not applicable to this report; a conservative upper-limit model is to be used in the TSPA-LA model.

Acceptance Criterion 3—Data Uncertainty Is Characterized and Propagated Through the Model Abstraction

(1) Models use parameter values, assumed ranges, probability distributions, and bounding assumptions that are technically defensible, reasonably account for uncertainties and variabilities, and do not result in an under-representation of the risk estimate;

Not applicable to this report; a conservative upper-limit model is to be used in the TSPA-LA model.

(2) Parameter values, assumed ranges, probability distributions, and bounding assumptions used in the abstractions of radionuclide release rates and solubility limits in the total system performance assessment are technically defensible and reasonable based on data from the Yucca Mountain region, laboratory tests, and natural analogs. For example, parameter values, assumed ranges, probability distributions, and bounding assumptions adequately reflect the range of environmental conditions expected inside breached waste packages;

Not applicable to this report; a conservative upper-limit model is to be used in the TSPA-LA model.

(3) The U.S. Department of Energy uses reasonable or conservative ranges of parameters or functional relations to determine effects of coupled thermal-hydrologic-chemical processes on radionuclide release. These values are consistent with the initial and boundary conditions and the assumptions for the conceptual models and design concepts for natural and engineered barriers at the Yucca Mountain site. If any correlations between the input values exist, they are adequately established in the total system performance assessment. For example, estimations are based on a thermal loading and ventilation strategy; engineered barrier system design (including drift liner, backfill, and drip-shield); and natural system masses and fluxes that are consistent with those used in other abstractions;

Not applicable to this report; a conservative upper-limit model is to be used in the TSPA-LA model.

(4) Uncertainty is adequately represented in parameter development for conceptual models, process models, and alternative conceptual models considered in developing the abstraction of radionuclide release rates and solubility limits, either through sensitivity analyses or use of bounding analyses;

Uncertainty is addressed in Section 8.2. A conservative upper-limit model is to be used in the TSPA-LA model to account for uncertainties in the degradation performance of the DSNF inventory. As indicated in Section 8.2, there is no uncertainty associated with this model.

(8) The U.S. Department of Energy adequately considers the uncertainties, in the characteristics of the natural system and engineered materials, such as the type, quantity, and reactivity of material, in establishing initial and boundary conditions for conceptual models and simulations of thermal-hydrologic-chemical coupled processes that affect radionuclide release; and

Uncertainty is addressed in Section 8.2. A conservative upper-limit model is to be used in the TSPA-LA model to account for uncertainties in the degradation performance of the DSNF inventory. As indicated in Section 8.2, there is no uncertainty associated with this model.

Acceptance Criterion 4—Model Uncertainty Is Characterized and Propagated Through the Model Abstraction

(1) Alternative modeling approaches of features, events, and processes are considered and are consistent with available data and current scientific understanding, and the results and limitations are appropriately considered in the abstraction;

Uncertainty is addressed in Section 8.2. A conservative upper-limit model is to be used in the TSPA-LA model to account for uncertainties in the degradation performance of the DSNF inventory. As indicated in Section 8.2, there is no uncertainty associated with this model.

(2) In considering alternative conceptual models for radionuclide release rates and solubility limits, the U.S. Department of Energy uses appropriate models, tests, and analyses that are sensitive to the processes modeled for both natural and engineering systems. Conceptual model uncertainties are adequately defined and documented, and effects on conclusions regarding performance are properly assessed. For example, in modeling flow and radionuclide release from the drifts, the U.S. Department of Energy represents significant discrete features, such as fault zones, separately, or demonstrates that their inclusion in the equivalent continuum model produces a conservative effect on calculated performance; and

Alternative modeling approaches that are consistent with available data and current scientific understanding are considered and discussed in Section 6.3. It is shown that the base model is more appropriate than the alternative conceptual models considered.

(3) Consideration of conceptual model uncertainty is consistent with available site characterization data, laboratory experiments, field measurements, natural analog information and process-level modeling studies; and the treatment of

conceptual model uncertainty does not result in an under-representation of the risk estimate; and

Not applicable to this model report; a conservative upper-limit model is to be used in the TSPA-LA model.

(4) The effects of thermal-hydrologic-chemical coupled processes that may occur in the natural setting, or from interactions with engineered materials, or their alteration products, on radionuclide release, are appropriately considered.

Not applicable to this model report; a conservative upper-limit model is to be used in the TSPA-LA model.

Acceptance Criterion 5—Model Abstraction Output Is Supported by Objective Comparisons

(1) The models implemented in this total system performance assessment abstraction provide results consistent with output from detailed process-level models and/or empirical observations (laboratory and field testings and/or natural analogs);

Not applicable to this model report; a conservative upper-limit model is to be used in the TSPA-LA model.

(3) The U.S. Department of Energy adopts well-documented procedures that have been accepted by the scientific community to construct and test the numerical models, used to simulate coupled thermal-hydrologic-chemical effects on radionuclide release. For example, the U.S. Department of Energy demonstrates that the numerical models used for high-level radioactive waste degradation and dissolution, and radionuclide release from the engineered barrier system, are adequate representations; include consideration of uncertainties; and are not likely to underestimate radiological exposures to the reasonably maximally exposed individual and releases of radionuclides into the accessible environment; and

The DSNF degradation model is developed using ASTM C1174-97 [DIRS 105725].

9. INPUTS AND REFERENCES

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9.3 SOURCE DATA, LISTED BY DATA TRACKING NUMBER

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