QA: QA

Civilian Radioactive Waste Management System
Management & Operating Contractor

Waste Form Degradation Process Model Report

TDR-WIS-MD-000001 REV 00 ICN 01

July 2000

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ACKNOWLEDGMENTS

The *Waste Form Degradation Process Model Report* (PMR) is a summary of 19 Analysis and Model Reports (AMRs) and 87 primary entries in the Features, Events, and Processes (FEPs) Database. As such, the authors of these AMRs and FEPs have produced the underlying text for this PMR. In addition, these same authors and others have contributed by summarizing this information for inclusion in the PMR. These contributors include C.D. Leigh, Sandia National Laboratories (SNL), Section 3.1 (Radioisotope Inventory); P.V. Brady, SNL, Sections 3.2, 3.3, 3.6, and 3.7 (In-Package Chemistry, Commercial Spent Nuclear Fuel [CSNF] Matrix Degradation, High-Level Waste [HLW] Degradation, and Radioisotope Dissolved Concentration); E.R. Siegmann, Duke Engineering Services, Section 3.4 (CSNF Cladding Degradation); T.A. Thornton, Framatome Cogema Fuels (FCF), Section 3.5 (DSNF Degradation); H.W. Papenguth, SNL, Section 3.8 (Colloidal Concentrations). In addition, J. Cunnane, Argonne National Laboratories, served as general technical editor of the text. J.K. McCoy, FCF, coordinated development of the ICN. The references were input and checked by E. James and N. Garcia of Science Applications International Corporation. Regulatory inputs were provided by D.M. Franks, Duke Engineering & Services, and K.R. Iyengar, Science and Engineering Associates. The compliance checker for this document was P. VanDillen, FCF. A.S. Mobasheran, FCF, was the Product Checking Group reviewer. Production support was provided by Critical Document Production.

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EXECUTIVE SUMMARY

The *Waste Form Degradation Process Model Report* (Waste Form Degradation PMR) is one of nine PMRs that have the shared objective of describing the technical information that was used in the total system performance assessment (TSPA) conducted to evaluate the postclosure performance of a potential monitored geologic repository at Yucca Mountain. The TSPA-SR (site recommendation) will be used in the preparation of a document for a Secretarial decision on whether to recommend the Yucca Mountain site for development as a repository. The Waste Form Degradation PMR summarizes the results of investigations on the degradation of the radioactive spent nuclear fuel (SNF), high-level waste (HLW), dissolved radionuclides, and colloidal radionuclides.

The culmination of these investigations was the construction of the Waste Form Degradation Model to predict the dissolved or colloidal radionuclides available for transport in the TSPA for SR (TSPA-SR). The Waste Form Degradation Model consists of eight major modeling/analysis components: (1) Radioisotope Inventory, (2) In-Package Chemistry, (3) Commercial Spent Nuclear Fuel (CSNF) Degradation, (4) CSNF Cladding Degradation (5) U.S. Department of Energy (DOE) Spent Nuclear Fuel (DSNF) Degradation, (6) HLW Degradation, (7) Radioisotope Dissolved Concentration (solubility), and (8) Radioisotope Colloidal Concentration. These eight components are generally connected sequentially starting with the radioisotope inventory as input and ending with projected radioisotope dissolved and colloidal concentration.

As a result of further investigations and in response to extensive comments by, for example, internal reviews and the U.S. Nuclear Regulatory Commission (NRC) through its Issue Resolution Status Reports (IRSRs), these eight components of the Waste Form Degradation Model have evolved substantially from those used in previous analyses as described below.

1. The Radioisotope Inventory Component developed for the TSPA-SR uses updated values for the inventory. The sources of information are based on internal documents for CSNF, reports from the National Spent Nuclear Fuel Program (NSNFP) for DSNF, and the Environmental Impact Statement (EIS) for HLW. An updated screening was conducted and resulted in 27 radionuclides identified as important (24 based on human dose and 3 mandated by the Groundwater Protection Requirements of the proposed U.S. Environmental Protection Agency [EPA] standard 40 CFR 197 [64 FR 46976]).

2. The In-Package Chemistry Component developed for the TSPA-SR couples the seepage rate of water into the package and degradation of the steel, aluminum, DSNF, CSNF, and HLW inside the package to evaluate water chemistry parameters such as hydronium ion concentration, total carbonate concentration, dissolved partial pressures of carbon dioxide and oxygen, ionic strength, and fluoride and chloride ion concentrations (i.e., pH, $p[CO_3]_T$, P_{CO_2}, P_{O_2}, [I], [F], [Cl]). The analysis is performed using the EQ3/6 geochemical code. The sources of information for the EQ3/6 geochemical simulations include a chemical thermodynamics database and information on the in-package environment. These results influence degradation of the CSNF cladding and matrix, HLW degradation, radionuclide solubility, and colloid

availability and stability. The degradation of the CSNF matrix and HLW, in turn, influence the In-Package Chemistry Component.

3. The CSNF Matrix Degradation Component uses two linear regression equations based on pH, $p[CO_3]_T$, P_{O_2}, and temperature (T) to evaluate matrix degradation over time; one equation is applicable for pH less than or equal to 7 and the other is applicable for pH greater than 7. The regression variables used, pH, $p[CO_3]_T$ and P_{O_2}, are coupled to in-package chemistry to account for uncertainty. The sources of information include flow through, static, batch reactor, and drip tests involving the dissolution of fresh and spent reactor fuels under both saturated and unsaturated conditions.

4. The CSNF Cladding Degradation Component evaluates degradation of the cladding and is coupled to the CSNF Matrix Degradation and In-Package Chemistry components developed for the TSPA-SR. The sources of information include failure data from reactor operation, pool and dry storage, and transportation. To better evaluate the performance of the cladding, two steps of degradation are included: perforation and matrix exposure. Perforation mechanisms include cladding damage during reactor use, creep failures at high temperatures during dry storage, transportation, or initial disposal, mechanical failure from earthquakes, and localized corrosion after disposal. The process of exposing the matrix and releasing radionuclides is through potential unzipping of the cladding caused by expansion of the fuel matrix as the uranium dioxide (UO_2) forms secondary minerals.

5. The DSNF Degradation Component developed for TSPA-SR uses a constant, bounding degradation rate based upon experimental data for N-Reactor SNF for all the DSNF waste types with the exception of naval SNF. However, the radioisotope inventory is the weighted mass average of DSNF waste types. Naval SNF degradation behavior is bounded by that of the CSNF.

6. The HLW Degradation Component developed for TSPA-SR uses bounds on parameters of a phenomenological model to develop a simplified (Arrhenius-type) rate equation of degradation that is dependent only upon pH and temperature. Conservative estimates of the model parameters are based upon experimental data for the degradation of borosilicate glass.

7. In the Dissolved Radioisotope Concentration Component, the solubilities of important radionuclides (as determined by updated screening) were reevaluated. The sources of information include EQ3/6 simulations of in-package chemistry for three categories of radionuclides. Three radioisotope solubilities were abstracted as a direct function of in-package chemistry (Np, U, Am) and three radionuclides solubilities (Ac, Cm, Sm) set equal to that of Am. Four additional radioisotope solubilities were defined by probability distributions (Pu, Pb, Pa, Ni). All other radioisotope solubilities were set at bounding values. The distributions and bounding values were based on results of the process modeling of the in-package chemistry.

8. For the Colloidal Radioisotope Concentration Component, the conceptualization directly used YMP-relevant experimental results from YMP-specific work and from the published literature. The conceptualization identified the availability and the stability of three categories of colloids: (1) existing colloids in the groundwater, (2) colloids generated during degradation of the waste, and (3) colloids generated during degradation of the disposal container.

Based on previous sensitivity analysis, the solubility of Np in the Dissolved Radioisotope Concentration Component has a strong influence on human dose and thus is a principal factor of the repository safety strategy.

Concurrent with model development, numerous (FEPs) have been formally developed to ensure that no important and/or influential FEP has been omitted in Waste Form Degradation Model. This formal process assigned 87 FEPs to the Waste Form Degradation PMR for consideration, and the vast majority are included in the Waste Form Degradation Model. This formal process has influenced the CSNF Cladding Degradation Component more than other components since it is a rather new component in comparison to others that have been under study over the past 20 years.

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ACRONYMS AND ABBREVIATIONS

ACNW	Advisory Committee on Nuclear Waste
AMR	Analysis and Model Report
ANL	Argonne National Laboratory
AP	Administrative Procedure
ASTM	American Society for Testing and Materials
ATM	Approved Testing Material
BU	burnup
BWR	boiling water reactor
CCDF	complementary cumulative distribution function
CDF	cumulative distribution function
CFR	Code of Federal Regulations
CLST	Container Life and Source Term (NRC KTI IRSR)
CRWMS	Civilian Radioactive Waste Management System Management
CSCI	Computer Software Configuration Item
CSNF	commercial spent nuclear fuel
DCCG	diffusion controlled cavity growth
DEIS	Draft Environmental Impact Statement
DHC	delayed hydride cracking
DI	document identifier
DIRS	Document Input Reference System
DOE	U.S. Department of Energy
DSNF	DOE spent nuclear fuel
DST	Drift-Scale Test
EBS	engineered barrier system
EIS	Environmental Impact Statement
ENFE	evolution of the near field environment
EPA	U.S. Environmental Protection Agency
FCF	Framatome Cogema Fuels
FEP	feature, event, and process
FEIS	Final Environmental Impact Statement
FFTF	Fast Flux Test Facility
FR	Federal Register
HLW	high-level waste
HR	Hanford Reservation
ICN	Interim Change Notice
INEEL	Idaho National Engineering and Environmental Laboratory
IPA	Iterative Performance Assessment
IRSR	issue resolution status report

ACRONYMS AND ABBREVIATIONS (Continued)

KTI	key technical issue
LA	license application
LANL	Los Alamos National Laboratory
LBU	log burnup
LWR	light water reactor
M&O	management and operating contractor
MCO	multi-canister overpack
MIC	microbially influenced corrosion
MOX	mixed oxide fuel
MTHM	metric tons of heavy metal
MWd	megawatt days
NEA	Nuclear Energy Agency
NFE	near-field environment
NRC	U.S. Nuclear Regulatory Commission
NSNFP	U.S. National Spent Nuclear Fuel Program
NWTRB	Nuclear Waste Technical Review Board
OCRWM	Office of Civilian Radioactive Waste Management (DOE)
OECD	Organization for Economic Cooperation and Development
PA	performance assessment
PARP	Total System Performance Assessment Peer Review Panel
PCT-A	Product Consistency Test, Type A
PMR	process model report
PWR	pressurized water reactor
QA	quality assurance
REV	Revision
RIP	Repository Integration Program
RN	radionuclide
RSS	Repository Safety Strategy
RW	radioactive waste
SCC	stress corrosion cracking
SCP	Site Characterization Plan
SNF	spent nuclear fuel
SNL	Sandia National Laboratories
SR	site recommendation
SRCR	Site Recommendation Consideration Report
SRS	Savannah River Site
SS	stainless steel

ACRONYMS AND ABBREVIATIONS (Continued)

SZ	saturated zone
THC	thermal-hydrologic-chemical
TRIGA	Training, Research, Isotopes, General Atomics
TSPA	total system performance assessment
TSPAI	Total System Performance Assessment and Integration (NRC IRSR)
UZ	unsaturated zone
VA	viability assessment
WF	waste form
WIPP	Waste Isolation Pilot Plant
WP	waste package
WVDP	West Valley Demonstration Project
YM	Yucca Mountain
YMP	Yucca Mountain Site Characterization Project
YMSCO	Yucca Mountain Site Characterization Office (OCRWM)

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1. INTRODUCTION

To evaluate the postclosure performance of a potential monitored geologic repository at Yucca Mountain, a total system performance assessment (TSPA) will be conducted. A set of nine process model reports (PMRs), of which this document is one, is being developed to summarize the technical basis for each of the process models supporting the TSPA model. These reports cover the following areas:

- Integrated Site Model
- Unsaturated Zone Flow and Transport
- Near Field Environment
- Engineered Barrier System Degradation, Flow, and Transport
- Waste Package Degradation
- Waste Form Degradation
- Saturated Zone Flow and Transport
- Biosphere
- Disruptive Events.

These PMRs are supported by analysis model reports (AMRs) that contain the more detailed technical information to be input into each PMR and the TSPA. This technical information consist of data, analyses, models, software, and supporting documentation that will be used to defend the applicability of each process model for its intended purpose of evaluating the postclosure performance of the potential Yucca Mountain repository system. The methodology used to develop AMRs and PMRs is designed to ensure traceability of information. Its final use in the TSPA is intended to ensure that the data are the best available when the TSPA is performed.

1.1 OBJECTIVE

The degradation and potential release of the radioactive waste placed into the potential Yucca Mountain repository are dependent on numerous features[1], events[2], and processes[3] (FEPs) of the disposal system. Because of the different spatial and time scales and the large number of FEPs, they have been grouped into several modeling systems and components for modeling and discussion in reports. The overall objective of the *Waste Form Degradation Process Modeling Report* (Waste Form Degradation PMR) is to summarize the technical basis of the Waste Form Degradation model. The degradation of the waste eventually leads to mobilization of radionuclides and release into other components of the engineered barrier system (EBS) in the TSPA. Because a small fraction of the radioisotope in the waste can be transported through the EBS and the natural rock barrier, the source concentrations of radionuclides evaluated by the Waste Form Degradation Model can influence results of the TSPA analysis when the waste

[1] An object, structure, or condition that has a potential to affect performance of the disposal system.
[2] A natural or anthropogenic phenomenon that has a potential to affect performance of the disposal system and that occurs during an interval that is short compared to the period of performance.
[3] A natural or anthropogenic phenomenon that has a potential to affect performance of the disposal system and that occurs during all or a significant part of the period of performance.

package (WP) is breached. This TSPA analysis, in turn, is an important basis of the Site Recommendation Consideration Report (SRCR) and, thus, designated as TSPA-SR in this report.

1.2 PURPOSE AND SCOPE

Nineteen AMRs were written to document (1) various experimental results, (2) conceptual models and detailed numerical models, and (3) further abstraction of the experiments or numerical models and to define consistent model parameters pertinent to degradation of the waste form. The purpose and scope of the Waste Form Degradation PMR is to summarize the most important aspects of these 19 AMRs to provide a coherent view of the components of the Waste Form Degradation Model. More specifically, in order to satisfy several important requirements for the analysis for the TSPA-SR according to the DOE interim guidance (Dyer 1999) to the proposed U.S. Nuclear Regulatory Commission (NRC) regulation (proposed DOE 10 CFR 63.114 Interim Guidance [Dyer 1999]), Waste Form Degradation PMR purpose and scope are to briefly summarize:

1. The technical basis for either including or excluding FEPs influencing the degradation, deterioration, and alteration of the waste form

2. The technical basis for the models (usually the experimental foundation but occasionally comparison with more detailed modeling or natural analogs) used to evaluate FEPs thought to significantly influence the degradation of the waste form

3. The technical basis for bounding values or probability distributions of parameters of the corresponding components

4. The relationship of the FEPs to particular key technical issues (KTIs) that have been raised by the NRC in their IRSRs–particularly the Container Life and Source Term (CLST) IRSR.

The numerical results of the Waste Form Degradation Model as implemented in the analysis for the TSPA-SR are not discussed here. These results will be found in documentation of the analysis for the TSPA-SR. Hence, the features of the waste form that are definitely considered barriers important to waste isolation are not identified here. Rather these identified features will be found in the TSPA-SR, the SRCR, and future versions of the Repository Safety Strategy (RSS) (CRWMS M&O 2000a).

1.3 QUALITY ASSURANCE

The Quality Assurance (QA) program applies to this PMR and the AMRs that support it. The development of this analysis is conducted under activity evaluation *Waste Form Analysis & Models* (CRWMS M&O 2000b), which was prepared per AP-2.16Q, *Activity Evaluation*. The results of that evaluation were that the activity is subject to the *Quality Assurance Requirements and Description* (DOE 2000) requirements.

This report has a formal planning document (CRWMS M&O 2000c). Development of this report did not involve the electronic management of data. The purpose of this ICN is to address DOE comments.

This report was prepared in accordance with AP-3.11Q, *Technical Reports* and reviewed in accordance with AP-2.14Q, *Review of Technical Products*. The Waste Form Degradation PMR does not serve as a primary source for data, models, and codes described in the AMRs related to waste form degradation; this function is provided by the AMRs themselves. The AMRs supporting the Waste Form Degradation PMR were prepared in accordance with AP-3.10Q, *Analyses and Models*, including AP-3.15Q, *Managing Technical Product Inputs*, for data verification, AP-SIII.2Q, *Qualification of Unqualified Data and the Documentation of Rationale for Accepted Data*, for data qualification and AP-SI.1Q, *Software Management*, for management of software. The quality assurance documentation of the models, codes, and data is deferred to the AMRs cited in this document. The current quality status of data and software can be found in the Document Input Reference System (DIRS).

This PMR includes the results from software codes and routines used in the supporting AMRs. However no software codes were used in the development of this PMR. The following statements are presented for information only. The only software code used in the AMRs was the EQ3/6 package, Version 7.2b, which was approved for QA work by LLNL and is identified as Computer Software Configuration Item (CSCI): UCRL-MA-110662 V 7.2b (CRWMS M&O 1998a) and its addendum (CRWMS M&O 1999c). The package was used both for chemical equilibrium calculations, and reaction path calculations (see Sections 3.2 and 3.7).

1.4 RELATIONSHIP TO OTHER PROJECT REPORTS

The Waste Form Degradation PMR describes how information is used in the TSPA-SR analysis by summarizing the modeling components of the Waste Form Degradation Model. Although not directly coupled, the Waste Form Degradation Model, in turn, is related to the Waste Package Degradation Model and Engineered Barrier System Model and their corresponding PMRs: Waste Package Degradation PMR (CRWMS M&O 2000d) and Engineered Barrier System PMR (CRWMS M&O 2000y). The Waste Form Degradation PMR is also related to the Disruptive Events PMR (CRWMS M&O 2000an) since it requires waste inventory information.

Eight summary/abstraction AMRs specifically support the Waste Form Degradation PMR (Figure 1.4-1). Other documents underlying these primary areas are mentioned within Chapter 3.

Abstraction Level AMRs:

- *Inventory Abstraction*, ANL-WIS-MD-000006 (CRWMS M&O 2000f)
- *In-Package Chemistry Abstraction*, ANL-EBS-MD-000037 (CRWMS M&O 2000g)
- *CSNF Waste Form Degradation: Summary Abstraction*, ANL-EBS-MD-000015 (CRWMS M&O 2000h)
- *Clad Degradation-Summary and Abstraction*, ANL-WIS-MD-000007 (CRWMS M&O 2000i)
- *DSNF and Other Waste Form Degradation Abstraction*, ANL-WIS-MD-000004 (CRWMS M&O 2000j)

- *Defense High Level Waste Glass Degradation*, ANL-EBS-MD-000016 (CRWMS M&O 2000k)

- *Summary of Dissolved Concentration Limits*, ANL-WIS-MD-000010 (CRWMS M&O 2000l)

- *Waste Form Colloid-Associated Concentrations Limits: Abstraction and Summary*, ANL-WIS-MD-000012 (CRWMS M&O 2000m).

Process Level AMRs:

- *Miscellaneous Waste-Form FEPs*, ANL-WIS-MD-000009 (CRWMS M&O 2000n)

- *Summary of In-Package Chemistry for Waste Forms*, ANL-EBS-MD-000050 (CRWMS M&O 2000o)

- *Initial Cladding Condition*, ANL-EBS-MD-000048 (CRWMS M&O 2000p)

- *Clad Degradation-Local Corrosion of Zirconium and Its Alloys under Repository Conditions*, ANL-EBS-MD-000012 (CRWMS M&O 2000ag)

- *Hydride-Related Degradation of SNF Cladding Under Repository Conditions*, ANL-EBS-MD-000011 (CRWMS M&O 2000r)

- *Clad Degradation-FEPs Screening Arguments*, ANL-WIS-MD-000008 (CRWMS M&O 2000s)

- *Clad Degradation-Wet Unzipping*, ANL-EBS-MD-000014 (CRWMS M&O 2000t)

- *Clad Degradation-Dry Unzipping*, ANL-EBS-MD-000013 (CRWMS M&O 2000u)

- *Pure Phase Solubility Limits-LANL*, ANL-EBS-MD-000017 (CRWMS M&O 2000v)

- *Secondary Uranium-Phase Paragenesis and Incorporation of Radionuclides into Secondary Phases*, ANL-EBS-MD-000019 (CRWMS M&O 2000w)

- *Colloid-Associated Radionuclide Concentration Limits: ANL*, ANL-EBS-MD-000020 (CRWMS M&O 2000x).

1.5 OVERVIEW OF WASTE FORM DEGRADATION MODEL

The analysis for the TSPA-SR requires numerous models and analyses. Many of these models are components of larger models that, in turn, are models to system-level models. Rather than call all the parts "models," this report uses terms that identify the hierarchy of the models to help orient the reader. The system-level model, which uses total system simulation software, is referred to as the "TSPA-SR system-level model" or "TSPA-SR." The major parts of TSPA-SR are referred to as "models." The Waste Form Degradation Model is one such model. The models and analyses that are parts of the Waste Form Degradation Model are referred to as

"components." (A possible alternative term is "submodel." However, not all the parts are models in the sense used by YMP but rather are analyses, so the term "components" was selected.) Although the Waste Form Degradation PMR has several broad purposes already listed, one important purpose is to describe the Waste Form Degradation Model in the TSPA-SR and its underlying components.

1.5.1 Function of Waste Form Degradation Model

Within the analysis for the TSPA-SR analysis, the function of the Waste Form Degradation Model is to determine three outputs over time: (1) dissolved concentration, (2) reversible colloidal concentration, and (3) irreversible colloidal concentration of radionuclides (Figure 1.5-1).

Numerous inputs are required for the Waste Form Degradation Model. Several inputs are intimately tied with the waste form degradation models such as initial cladding condition and thermodynamic data for radionuclides. They are summarized in this report. Other inputs are summarized in other PMRs. For example, the time dependent seepage flow into the WP and WP surface temperatures are summarized in the EBS PMR (CRWMS M&O 2000y). Corrosion rates of the inner stainless steel container and supports for waste inside the waste package are summarized in the Waste Package Degradation PMR (CRWMS M&O 2000d).

1.5.2 Components of Waste Form Degradation Model

To determine the three output radioisotope concentrations, the Waste Form Degradation Model uses eight major modeling/analysis components to evaluate the degradation, deterioration, or alteration of the waste: (1) Radioisotope Inventory (described in CRWMS M&O 2000f), (2) In-Package Chemistry (described in CRWMS M&O 2000g), (3) Commercial Spent Nuclear Fuel (CSNF) Matrix Degradation (described in CRWMS M&O 2000h), (4) CSNF Cladding Degradation (described in CRWMS M&O 2000i), (5) DOE Spent Nuclear Fuel (DSNF) Degradation (described in CRWMS M&O 2000j), (6) High-Level Waste (HLW) Degradation (described in CRWMS M&O 2000k), (7) Radioisotope Dissolved Concentration (described in CRWMS M&O 2000l), and (8) Radioisotope Colloidal Concentration (described in CRWMS M&O 2000m) (Figure 1.4-1).

The mathematical description of these eight components is one specific purpose of the Waste Form Degradation PMR. Another important purpose is to summarize the technical bases of the eight components. Generally, the primary technical bases or "foundations" of the eight components of the waste form degradation models are the CSNF, DSNF, and HLW degradation experiments described throughout the report.

1.5.3 Major Models Communicating with Waste Form Degradation Models within TSPA-SR Analysis

For TSPA-SR analysis, the major model communication with the Waste Form Degradation Model is from the EBS abstraction models and the TSPA (Figure 1.5-2). The EBS abstraction models determine the water seepage quantity onto the waste form that is used by both the In-Package Chemistry Component and the Dissolved Radioisotope Concentration Components. In turn, the Waste Form Degradation Model determines dissolved and colloidal concentrations of

radionuclides for the EBS. The Waste Form Degradation Model is also uncoupled from the Waste Package Degradation Model. Although conceptually, the Waste Package Degradation Model could calculate the rate iron oxides or other corrosion products are produced by the degradation of the WP and pass this information onto the Waste Form Degradation Model. Instead of this communication, the underlying in-package process model evaluated the resulting chemistry from a variety of corrosion rates and expressed this as uncertainty in the final in-package Chemistry Component.

1.5.4 Linkage of Components in Waste Form Degradation Model

The components of the Waste Form Degradation Model are generally connected sequentially starting with the Radioisotope Inventory Component and ending with the Radioisotope Colloidal Concentration Component; however, some complications occur. First, the rates of degradation of the three general categories of waste forms modeled (CSNF, DSNF, and HLW) are not coupled, and so three different paths through the Waste Form Degradation Model are possible (Figure 1.5-3). Furthermore, because the in-package chemistry (primarily pH) is dependent upon the amount of CSNF exposed and the rate of alteration of the borosilicate glass of the HLW, some minor feedback does occur. Because the feedback is minor (i.e., secondary effect), the feedback lags by one timestep (i.e., the Waste Form Degradation Model does not iterate during the timestep).

1.6 PRINCIPAL AND OTHER FACTORS CONSIDERED

The magnitude of the YMP and the complexities associated with both the natural and EBSs dictate that the YMP prioritize its activities and focus on the factors most important to performance, hereafter named the principal factors. The Repository Safety Strategy (RSS) (CRWMS M&O 2000a, Table 3-1) has identified seven principal factors and 18 other factors of lesser importance. The selection of the principal factors has been based on preliminary TSPA analyses and expert judgment, which identified these factors as likely to provide sufficient confidence for the safety case. The other factors were deemed to be less important to the safety case, nevertheless requiring representation in the current methodology (CRWMS M&O 2000a, Section 3.1). Table 1.6-1 lists the seven principal factors, the 18 other factors, and the PMRs that address each. The Waste Form Degradation PMR addresses the principal factor Solubility Limits of Dissolved Radionuclides.

Solubility Limits of Dissolved Radionuclides is a principal factor because it describes the limitation to the mobilization of the relatively immobile radionuclides because of their limited solubilities in the water at Yucca Mountain. Some of these radionuclides present significant risk potential because of the long half-life and large dose conversion factor. In many cases, the solubilities of these radionuclides are so low they present no significant issue for the potential repository system. However, there are a few cases, notably neptunium, plutonium, and uranium isotopes, for which solubility limits could be very important to the postclosure safety case (CRWMS M&O 2000a, Section 3.2.4).

Table 1.6-1. Principal Factors, Other Factors, and the Process Model Reports Where Addressed

	Factor	Process Model Report
Principal Factors (Nominal Scenario)	Seepage into drifts	Unsaturated Zone Flow and Transport
	Performance of the drip shield	Waste Package Degradation
	Performance of the waste package barriers	Waste Package Degradation
	Solubility limits of dissolved radionuclides	Waste Form Degradation
	Retardation of radionuclide migration in the unsaturated zone	Unsaturated Zone Flow and Transport
	Retardation of radionuclide migration in the saturated zone	Saturated Zone Flow and Transport
	Dilution of radionuclide concentrations during migration	Saturated Zone Flow and Transport
Other Factors (Nominal Scenario)	Climate, net infiltration into the mountain, and unsaturated zone flow above the repository	Unsaturated Zone Flow and Transport
	Coupled processes – effects on unsaturated zone flow	Unsaturated Zone Flow and Transport
	Coupled processes – effects on seepage	Near-Field Environment
	Environments on the drip shield	Engineered Barrier System Degradation, Flow, and Transport
	Environments on the waste package	Engineered Barrier System Degradation, Flow, and Transport
	Environments within the waste package	Waste Form Degradation
	CSNF waste form performance	Waste Form Degradation
	HLW glass waste form performance	Waste Form Degradation
	DSNF, naval SNF, Pu-disposition waste form performance	Waste Form Degradation
	Colloid-associated radionuclide concentrations	Waste Form Degradation
	In-package radionuclide transport	Waste Form Degradation
	Transport through the drift invert	Engineered Barrier System Degradation, Flow, and Transport
	Advective pathways in the unsaturated zone	Unsaturated Zone Flow and Transport
	Colloid-facilitated transport in the unsaturated zone	Unsaturated Zone Flow and Transport
	Coupled processes – effects on unsaturated zone transport	Unsaturated Zone Flow and Transport
	Advective pathways in the saturated zone	Saturated Zone Flow and Transport
	Colloid-facilitated transport in the saturated zone	Saturated Zone Flow and Transport
	Biosphere transport and uptake	Biosphere
Factors for Disruptive Event Scenarios	To be determined. See CRWMS M&O (2000a, Section 3.5) for preliminary considerations	Disruptive Events

Source: Derived from CRWMS M&O 2000a, Table 3-3

In addition, the following six other factors are within the scope of the Waste Form Degradation PMR.

1. Environment within the WP (Section 3.2)
2. CSNF waste form performance (Section 3.3 and 3.4)
3. HLW glass waste form performance (Section 3.5)
4. DSNF, naval SNF, Pu-disposition waste form performance (Section 3.6)
5. Colloid-associated radionuclide concentrations (Section 3.8)
6. In-package radionuclide transport (Section 3.2.1.2).

The general guidelines for addressing these other factors were to use conservative bounding analysis, when possible. Principal factors are studied and evaluated more thoroughly, using both rigorous, realistic numerical evaluations and also conservative bounding analyses if warranted by the overall site performance.

1.7 ORGANIZATION OF REPORT

Chapter 2 discusses the evolution of the Waste Form Degradation Model from past TSPA analysis and the formal screening of FEPs to include in the current model. The major portion of this report, Chapter 3, discusses the inner workings and technical basis of the eight main components of the Waste Form Degradation Model used in the TSPA-SR analysis. The order of discussion for the eight components reflects the direction of data flow between components (Figure 1.5-3). Each section of Chapter 3 begins with an introduction to the component. This introduction includes a listing of the AMRs and calculations that provide the detailed technical basis for that component, and an overview of the conceptual model as appropriate. The description sub-section, which follows, includes summaries of (1) the descriptions of the analysis, models, and/or abstractions, (2) relevant data and uncertainties, (3) assumptions and basis, and (4) model results. The amount of detail provided varies with each component. For many components, the results are simply the models and parameters provided to the TSPA-SR and are, thus, included in the model/analysis abstraction description. In addition, many times, the relevant data and uncertainties are only briefly summarized, and the reader is referred to the supporting AMRs for detailed discussion. The Confidence/Limitations/Validation sub-section includes a discussion of model limitations, the justification and confidence, and validation that the component is appropriate for the intended use (i.e., appropriate as support for making a recommendation on the suitability of the Yucca Mountain site for a repository). Each component section ends with a sub-section, which summarizes alternative models and/or addresses known issues posed by groups that have project oversight, regulatory oversight, or stakeholder interfaces with the YMP. The last section of Chapter 3, Section 3.9, tabulates these issues for all eight components, and provides cross-references to the component sections. Chapter 4 of the report summarizes the relationship of the topics of this report with the NRC IRSRs, primarily the CLST KTIs. Chapter 5 is a brief summary of the report.

2. EVOLUTION OF WASTE FORM DEGRADATION MODEL

The waste form degradation models have generally evolved over the years along with the overall TSPA model of the disposal system. Thus, this section first presents a short summary of the evolution of the TSPAs of the Yucca Mountain disposal system by the Yucca Mountain Project (YMP), followed by a summary of the evolution of various components in the Waste Form Degradation Model. More detail is presented along with each major section of the various components.

2.1 GENERAL HISTORY OF ASSESSMENTS OF YUCCA MOUNTAIN PERFORMANCE

As summarized by Rechard (1999), simple analytic calculations to determine the relative importance of various phenomena postulated to occur at Yucca Mountain were conducted in 1984 (which identified ^{99}Tc, ^{129}I, and ^{237}Np as important radionuclides for evaluating compliance) (Sinnock et al. 1984) and 1988 (performed in conjunction with the Site Characterization Plan (SCP) (DOE 1988, Section 8.3.5.13). The first large-scale analysis of water movement through the unsaturated zone occurred in 1990 (Prindle and Hopkins 1990). Shortly thereafter, a series of deterministic calculations using best estimates for model parameters were run by several organizations—Sandia National Laboratories, Pacific Northwest National Laboratory, and Los Alamos National Laboratory—to simulate the radioisotope transport in the unsaturated zone (Barnard and Dockery 1991).

In 1992, the YMP completed the first probabilistic TSPA of the potential Yucca Mountain disposal system to evaluate releases to a 5-km boundary. Two different organizations conducted total system performance analysis: Sandia National Laboratories (TSPA-91-SNL) (Barnard et al. 1992) and Pacific Northwest National Laboratory (TSPA-91-PNNL) (Eslinger et al. 1993). For the first time, gaseous flow of ^{14}C was included. In this first probabilistic assessment, the YMP was at a relatively early stage in conceptual model development; thus, parameter values and distributions were determined primarily by individual PA analysts.

In 1993, a second iteration of the SNL TSPA (TSPA-93-SNL) (Wilson et al. 1994) was started that included an improved Waste Form Degradation Model (called source-term model) and a saturated zone model. The analysis also greatly expanded the data used for defining geochemical parameters. The formality increased as well in that distributions for many more parameters were developed and more often based on the consensus of several PA analysts, accompanied by input from site characterization scientists.

In 1993, the newly assigned CRWMS M&O also conducted a TSPA (TSPA-93-Duke) (CRWMS M&O 1994) using the Repository Integration Program (RIP) modeling system intended to rapidly simulate the behavior of the disposal system to evaluate design systems. The system used a variety of techniques such as curve fits to previous results and selection of distributions for particular data to incorporate previous results. This simplified modeling style, called abstraction, had been originally proposed in the 1988 SCP (DOE 1988, Chapter 8, Section 8.3.5.13) as the culmination of sensitivity analysis on process models. The analyses using RIP were the only TSPAs performed by CRWMS M&O after 1993. The basic information on parameter distributions reported in TSPA-93-SNL (Wilson et al. 1994) was used for

TSPA-93-Duke (CRWMS M&O 1994) and subsequent TSPAs in 1995, 1996, and 1997 (CRWMS M&O 1995; CRWMS M&O 1996a; CRWMS M&O 1996b; CRWMS M&O 1997a); although some differences did occur for radionuclide inventory, and other parameter values were sometimes changed for parametric sensitivity analysis.

Besides TSPAs conducted specifically by CRWMS M&O for the Yucca Mountain Site Characterization Office of DOE (DOE/RW/YMSCO), TSPAs conducted specifically for the National Spent Nuclear Fuel Program (NSNFP) of the Office of Environmental Management of DOE (DOE/EM) by SNL in 1993, 1995, and 1998 examined the behavior of DSNF to test the viability of direct disposal of the waste in salt, granite, and tuff (DSNF-TSPA-93-SNL) (Rechard 1993), DSNF-TSPA-94-SNL (Rechard 1995), DSNF-TSPA-98-SNL) (DOE 1998a; Rechard 1993, 1995). The analysis included the effects of cladding since this was an important feature that distinguished ~250 types of DSNF and CSNF. The NSNFP also contracted with CRWMS M&O to conduct similar analysis in 1997 for DSNF, DSNF-TSPA-97-Duke (CRWMS M&O 1997a). The CRWMS M&O had done a similar sensitivity study the year before on the disposal of excess weapons and plutonium (CRWMS M&O 1996b). In 1998, NSNFP supported CRWMS M&O such that DSNF was included as a sensitivity study in the TSPA-VA (DOE 1998b) (viability assessment) technical basis document discussed below (CRWMS M&O 1998b).

In 1997, Congress mandated in its energy appropriation bill that the YMP provide a viability assessment that (along with a preliminary design and costs estimates for constructing and operating that design) would include a TSPA describing the probable behavior of the potential Yucca Mountain disposal system (Energy and Water Development Appropriations Act 1997). A TSPA-VA was thus initiated and completed in November 1998 (DOE 1998b). For TSPA-VA, numerous changes and additions were made to the TSPA-95 (CRWMS M&O 1995) models, including the addition of more phenomena. Some of these changes included the influence of the Zircaloy cladding on CSNF, evaluation and inclusion of geochemistry changes near the waste package, colloid formation and transport, and a factor of 100 reduction in solubility of Np (DOE 1998b; CRWMS M&O 1997b; CRWMS M&O 1998b).

2.2 EVOLUTION OF EACH MODEL COMPONENT

A brief history of the evolution of the waste form models and components is discussed.

1. Radioisotope Inventory Component (Section 3.1)

   - Many of the early assessments in the 1990s used the inventory for CSNF developed around the time of the 1988 SCP: the DOE Characteristics Data Base of 1987.

   - TSPA-93-Duke (CRWMS M&O 1994) used the DOE Characteristics Data Base of 1987 to evaluate the inventory of CSNF and HLW and screen radionuclides.

   - TSPA-95 (CRWMS M&O 1995) used the DOE Characteristics Data Base of 1992; the weighted average burnup (BU) was slightly lower than TSPA-93-Duke (CRWMS M&O 1994) (36,666 versus 39,075 megawatt days [MWd]/metric tons

of heavy metal [MTHM]); nine radionuclides were tracked in TSPA-95. The screening criteria for the radioisotope selection were based on the maximum release rates allowed in 10 CFR 60.

- TSPA-VA (DOE 1998b) used the same inventory information as TSPA-95 (CRWMS M&O 1995). Inventories for sixteen DSNF groups (supplied by NSNFP), naval fuel (supplied by the Naval Nuclear Propulsion Program), and immobilized plutonium from the disposition of excess weapons were not included in the base case analysis but were considered in separate sensitivity analysis (CRWMS M&O 1998b, Section 6.2.1). Releases from naval SNF were found to be insignificant.

- TSPA-SR analysis uses updated values (CRWMS M&O 2000f) for the CSNF inventory based on internal documents, DSNF inventory based on a report of eleven groups supplied by NSNFP (DOE 1999b), HLW inventory based on the draft EIS (DOE 1999a, Section A.2.1), and immobilized plutonium based on the draft EIS (DOE 1999a, Section A.2.3). This evaluation includes the trend toward higher burnup fuels in utility operation.

2. In-Package Chemistry Component (Section 3.2)

- Through 1997, the effects of in-package chemistry on degradation of the waste form and solubility of radionuclides were incorporated through judgement when assigning a parameter value or distribution, or through use of conservatively bounded models. This resulted in wide distribution ranges for many parameters. Sensitivity analyses suggested that application of chemically dependent models could reduce uncertainty and conservatism.

- TSPA-VA (DOE 1998b) evaluated chemistry of the water potentially entering a waste package based on the water flux and presence of a degraded concrete liner for several input conditions. The results were then collapsed into two time periods and used to guide the selection of solubility for a few radionuclides (CRWMS M&O 1998b). This “two group” chemistry was dominated by the concrete liner, but allowed further sensitivity assessment for the effect of chemical conditions within the EBS.

- For TSPA-SR analysis, the Waste Form Degradation Model has an In-Package Chemistry Component that was coupled with the seepage rate of water into the package and degradation of the steel, aluminum, SNF, and HLW inside the package. The TSPA-SR analysis design does not have a concrete liner that dominates the chemistry, therefore processes at the surface of waste package materials and waste forms control the range of chemistry. The estimated minimum pH after breach of the container was lower than was considered in previous TSPAs. Results from the In-Package Chemistry influenced degradation of the CSNF cladding and matrix, solubility, and colloid availability and stability. The degradation of HLW and exposed amount of CSNF, in turn, influenced the In-Package Chemistry (Figure 1.5-3).

3. CSNF Matrix Degradation Component (Section 3.3)

- Early performance assessments bound CSNF oxidation and dissolution rates by experimental observations. Testing of UO_2 and spent fuel has included elevated temperature air oxidation, aqueous dissolution at high water flow rates, and in aqueous dissolution batch reactors. Because of the possibility for early waste package failure, high temperatures, and no modeling of cladding performance, dry oxidation of the fuel matrix to more soluble forms was an important consideration in early assessments. Aqueous dissolution is observed to be temperature and pH dependent. Carbonate ions can influence the surface reaction rate, and at high rates and limited oxygen availability the rate may be bounded by P_{O_2}. Burnup dependence includes a combination of effects from radiation, fission products and thermal alteration of the matrix.

- Since TSPA-93-Duke (CRWMS M&O 1994), empirical dissolution rates have been based on experimental data.

- For TSPA-95, a simplified regression equation based on T, pH, PO2, [CO3]T was used, based on high flow rate experiments to provide a upper bound on dissolution rate.

- After TSPA-95, the fuel test program was expanded to include aqueous dissolution at very low 'drip rates' to represent the slow seepage conditions indicated as most likely to occur in an unsaturated repository.

- For TSPA-VA, an eleven term regression equation dependent on temperature (T), pH, total carbonate concentration [CO3]T , oxygen partial pressure (as percent of atmosphere) (PO2), burnup of the fuel, and several cross-product terms was used. This equation was based on high flow rate dissolution experiments, and thus represents an upper bound. The longevity of the VA waste package and inclusion of cladding performance resulted in protection of the fuel matrix until temperatures were low enough that dry oxidation was no longer an important process. A more detailed chemistry dependence was included to allow sensitivity analyses to evaluate the value of tracking chemical condition within the EBS. The collection of radionuclides in secondary phases, as seen in the drip tests was considered in a sensitivity study.

- For TSPA-SR analysis, a simplified regression equation based only on T, pH, P_{O_2}, $[CO_3]_T$ was used and these controlling variables were coupled to in-package chemistry to account for uncertainty. The model is based on data from the high flow rate experiments and represents an upper bound for the dissolution rate.

4. CSNF Cladding Degradation Component (Section 3.4)

- While fuel cladding is very corrosion resistant, it is thin and used in a harsh reactor environment. Because of these concerns, cladding was not considered in early assessments. Cladding degradation testing and modeling has evolved and is used

as a basis for extending the period of wet and dry storage and for licensing shipping casks for spent fuel. This information has accumulated in the literature. Development of long lived waste packages that protect the fuel during the early thermal transient, along with the accumulated literature, has allowed consideration of cladding in repository performance.

- Cladding degradation was first considered in DSNF-TSPA-93-SNL (Rechard 1993) and DSNF-TSPA-94-SNL (Rechard 1995) to distinguish performance of various SNF (Rechard 1993, 1995; DOE 1998a); the cladding was divided into two classes, failed and intact cladding; the degradation of intact cladding was based on Arrhenius type rate equation and a reduction factor to account for localized corrosion.

- For TSPA-95, cladding degradation from creep rupture was discussed but not used in the analysis, and the fuel was considered to be bare.

- For TSPA-VA, several cladding degradation modes (initial reactor failures, creep rupture, failures from rockfall, and localized corrosion as estimated by a zirconium model within the waste package degradation model, WAPDEG) were combined into a time-varying function of failed cladding. Other cladding modes such as cladding unzipping, stress corrosion cracking, and hydride failures (hydride embrittlement, delayed hydride cracking [DHC] and hydride reorientation) were not considered likely to occur and were omitted from consideration.

- For TSPA-SR analysis, cladding degradation was coupled to CSNF Matrix Degradation and In-Package Chemistry; furthermore, two steps of degradation were included: perforation and unzipping. The modes of cladding degradation were formally evaluated during FEP screening; hydride failures were still omitted from consideration.

5. DSNF Degradation Component (Section 3.5)

- DSNF was first considered in DSNF-TSPA-93-SNL (Rechard 1993) and DSNF-TSPA-94-SNL (Rechard 1995). An Arrhenius-type rate equation was used for degradation except that each of the various types of DSNF used different model parameters.

- CRWMS M&O studied the sensitivity of DSNF in 1997.

- TSPA-VA (DOE 1998b) studied the sensitivity of DSNF in 1998; the degradation rate of N-Reactor SNF was used to bound the response of all other waste types; the rate equation was that developed for DSNF-TSPA-94-SNL (Rechard 1995).

- In an analysis to support TSPA-SR, DSNF (with the exception of naval SNF) was included using N-Reactor SNF to represent the physical degradation rate; however, the inventory was the weighted sum of all waste types included in the DSNF allocation category. The degradation rate of N-Reactor SNF was a bound on the maximum degradation rate observed to date in on-going experiments (Gray and Einziger 1998).

6. HLW Degradation Component (Section 3.6)

   - Early assessments were based on bounds of observed glass dissolution rates. Initially short-term, aqueous batch testing was done for developing glass mixtures and evaluating product consistency. Later, long term batch and high flow rate testing was done to obtain long term reaction rate.

   - A separate degradation rate for HLW was first included in TSPA-93-SNL and TSPA-93-Duke (CRWMS M&O 1994) and afterwards.

   - For TSPA-VA, a phenomenological model of degradation was used where several parameters were based on experimental data; a few variables were set at fixed values (e.g., silica concentration). Development of long lived waste packages reduced the importance of higher temperature processes.

   - For TSPA-SR analysis, bounds on parameters of a phenomenological model were used to develop a simplified (Arrhenius-type) rate equation of degradation. The parameter values in this model are based on laboratory test results for a range of reference glass compositions and are designed to conservatively bound the rate of glass corrosion under the range of conditions (immersion, humid air and dripping groundwater) to which it may be exposed in the repository. Specifically, the model is now designed to bound the higher corrosion rates observed after extensive reaction progress in some laboratory tests and also bounds the corrosion rates observed under vapor hydration and dripping groundwater conditions.

7. Dissolved Radioisotope Concentration Component (Section 3.7)

   - Early assessments assumed conservative upper bounds on aqueous solubility.

   - For TSPA-93-SNL, an elicitation of project experts was used in 1993 to determine solubility of important radionuclides.

   - TSPA-95 (CRWMS M&O 1995) generally used the values of TSPA-93-SNL; however, some values were modified based on experimental observations and controlling processes and phase assumptions.

   - TSPA-VA (DOE 1998b) used the values of TSPA-95 (CRWMS M&O 1995) except Np solubility was reduced by factor of 100 after reevaluating previous experiments.

- In TSPA-SR analysis, solubility of important radionuclides (as determined by updated screening) was reevaluated. Three radioisotope solubilities were abstracted as a direct function of in-package chemistry (Np, U, Am) to better account for the uncertainty. Four radioisotope solubilities were defined by a probability distribution, and all others were set at bounding values.

8. Colloidal Radioisotope Concentration Component (Section 3.8)

   - Early assessments used wide ranges on solubility that bounded the potential effect of colloidal transport of specific radionuclides since experimental data and colloid formation, stability and transport models were limited.

   - Colloids first incorporated as a radioisotope transport mechanism in TSPA-VA. Colloid formation, stability, and transport properties were inferred from glass and fuel dissolution tests, field observations, and laboratory experiments.

   - Colloid model incorporates new details for TSPA-SR.

2.3 SCREENING OF FEATURES, EVENTS, AND PROCESSES

2.3.1 Screening Criteria

Throughout the 20 years of analysis of the potential Yucca Mountain disposal system, various FEPs have been identified and their influence on the disposal system evaluated. The FEPs have been identified by a variety of methods, such as hypothesis by scientists and engineers working on the project or through review of their work. The variety of project participants and reviewers have helped ensure that a wide variety of FEPs has been considered. These hypotheses and reviews have been the impetus for the changes and evolution of the models discussed above. An initial set of FEPs was created for TSPA-SR analysis by combining three general lists of FEPs: 82 from various YMP workshops in 1998 and 1999, 292 FEPs gathered from YMP literature and site studies, and 1,412 FEPs from a draft report of the Nuclear Energy Agency (NEA) of the Organization for Economic Cooperation and Development (OECD) for a total of 1,786 FEPs. The latter list is a compilation of FEPs from seven geologic repository programs in other countries and, thus, the most complete attempt internationally to develop a comprehensive list of FEPs relevant to radioactive waste disposal.

To ensure the list was comprehensive, all potentially relevant FEPs identified have been included; however, this list has led to considerable redundancy. Consequently, late in 1998, the FEPs were classified as either primary or secondary. The 310 primary FEPs are those FEPs for which detailed screening arguments are developed. The remaining 1,476 secondary FEPs are either completely redundant or can be reasonably aggregated and mapped into a single primary FEP through modification of the description the primary FEP.

To develop screening arguments, the primary FEPs have been assigned to applicable PMRs. However, since a FEP can effect many facets of the disposal system, a FEP may be assigned to several PMRs. For example, some FEPs that affect the degradation of waste form also influence

the waste package degradation and the evolution of the Engineered Barrier System and, thus, have been assigned to all three corresponding PMRs.

The 87 primary waste form FEPs assigned to the Waste Form Degradation PMR have been evaluated by subject-matter experts and discussed in three different AMRs: 57 primary waste form FEPs are discussed in *Miscellaneous Waste-Form FEPs* (CRWMS M&O 2000n, Table 1), 15 are discussed in *Waste Form Colloid-Associated Concentrations Limits: Abstraction and Summary* (CRWMS M&O 2000m, Attachments I through XV), and 16 are discussed in *Clad Degradation—FEPs Screening Arguments* (CRWMS M&O 2000s). One FEP (Mutation) appears in two AMRs (CRWMS M&O 2000n, CRWMS M&O 2000m).

The decision to include or exclude a FEP related to the waste form was based on two criteria in the NRC's proposed rule 10 CFR 63.114 (Dyer 1999) .[4]

- "Consider only events that have at least one chance in 10,000 of occurring over 10,000 years" (Section 63.114(d)).

- "Provide the technical basis for either inclusion or exclusion of degradation, deterioration, or alteration processes of engineered barriers in the performance assessment, including those processes that would adversely affect the performance of natural barriers. Degradation, deterioration, or alteration processes of engineered barriers must be evaluated in detail if the magnitude and time of the resulting expected annual dose would be significantly changed by their omission" (Section 63.114(f)).

In accordance with its proposed rule (Dyer 1999), the NRC provides additional guidance in the TSPAI IRSR (NRC 2000a) on the screening process to follow to exclude FEPs. The guidance states four criteria that will be used (NRC 2000a, Section 4.2.3), with the first three criteria dealing with evaluating the probability of the FEP:

- "Criterion T1: Categories of processes and events that are not credible for the YM repository because of waste characteristics ... are identified and sufficient justification is provided for DOE's conclusions."

- "Criterion T2: The probability assigned to each category of processes and events not screened based on criterion T1 or criterion T2 is consistent with site information, well documented, and appropriately considers uncertainty."

- "Criterion T3: DOE has demonstrated that processes and events screened from the PA on the basis of their probability of occurrence, have a probability of less that one chance in 10,000 of occurring over 10,000 years."

[4] The proposed EPA Standard 40 CFR 197 (Dyer 1999; 64 FR 46976) has a similar approach in Section 197.40.

- "Criterion T4: DOE has demonstrated that categories of processes and events omitted from the PA on the basis that their omission would not significantly change the calculated expected annual dose, do not significantly change the calculated expected annual dose."

In summary, the first criterion permits an exclusion argument for FEPs that are not credible because of waste characteristics, repository design, or site characteristics. The second and third criteria deal with the assignment of probabilities to FEPs and demonstrating that the probabilities of excluded FEPs are below the quantitative probability limit of 10^{-4} over 10^{4} yr. The fourth criterion permits exclusion of FEPs that do not significantly change the calculated expected annual dose. These exclusions must be adequately justified in accompanying discussions or calculations, including the use of either bounding or representative estimates. In applying the fourth criterion, calculations of intermediate results are often used instead of dose calculations. For example, if a FEP does not affect release from the waste packages, it will not affect dose, so it is sufficient to consider release alone.

Based on the four criteria stated above, a statement on whether to Include or Exclude the FEP is provided. If the screening decision is Include, the FEP is considered either in the nominal scenario (i.e., the scenario that contains all expected FEPs and no disruptive[5] FEPs), in a disruptive scenario (i.e., any scenario that contains all expected FEPs and one or more disruptive FEPs), or in the human intrusion scenario.

Because the primary FEPs are the coarsest aggregate suitable for analysis, situations may result in which a given primary FEP contains some secondary FEPs that are Include and some that are Exclude. In these situations, the screening decision specifies which elements are Include and which are Exclude.

2.3.2 Screening Decisions

Most of the waste form FEPs that have been excluded from further consideration were excluded based on low consequence. Several waste form FEPs have been excluded based on the FEP not being credible for the waste characteristics and repository design at Yucca Mountain.

For the discussion herein, the 87 FEPs have been grouped and tabulated according to the eight major modeling components of the waste degradation model (Figure 2.3-1). Each of the eight tables provides the YMP FEP number, the short title, the decision as to whether to include or exclude the FEP, the PMRs where the FEP was discussed, the AMR that provides the full argument for the screening decision related to the waste form, and a brief synopsis of the screening argument. The synopsis only pertains to the Waste Form Degradation PMR. The reasoning to include or exclude a FEP in relation to other PMRs is not given but can be found in those other PMRs or the underlying AMR.

Several waste form FEPs have been excluded because the FEP is not credible for the characteristics of the waste proposed for disposal at Yucca Mountain (i.e., first criterion, T1).

[5] A disruption FEP is a retained FEP that has a probability of occurrence less than 1.0 during the period of performance but greater than the cutoff of $10^{-4}/10^{4}$ yr defined by the NRC as proposed 10 CFR 63.114(e) (Dyer 1999).

The succinct description for this reasoning is "Excluded based on low probability (credibility)." For example, cellulosic material is not present in CSNF, DSNF, or HLW that would be disposed of at Yucca Mountain; furthermore, organic material will be excluded based on current waste acceptance criteria. Therefore, FEP 2.1.02.10.00 in Table 2.3-2 that discusses this feature of the waste is excluded based on a low probability (credibility) argument.

No FEPs were excluded based on quantitatively evaluating the probability of a FEP (i.e., criteria T2 and T3). Rather, most of the excluded FEPs related to the waste form were excluded based on the fourth criterion (T4), "no significant change in the expected annual dose." FEPs excluded based on this criterion are succinctly described as "Excluded based on low consequence." For example, generation of H_2 is credible in an anoxic environment when steel corrodes; yet, it is unlikely that much will be produced since the drifts at Yucca Mountain will be mostly oxic. Only in small pores and crevices would an anoxic environment be found in the waste. Because evaluating a probability for this phenomenon is very difficult, the low consequences of this phenomenon were used to exclude this FEP. H_2, if produced, is very mobile and kinetically unreactive at low temperatures, so it is expected to leave the drift before it reacts with any of the emplaced materials. In addition, oxidation of hydronium ion to H_2 instead of sulfur to sulfate results in less pH suppression.

The latter reason is a minor aspect of the argument to exclude the process, but in other FEPs it can be a major aspect of the argument because bounding estimates are often used in the TSPA-SR. That is, the argument for exclusion is often that the exclusion of the FEP provides a bound on the expected annual dose (i.e., inclusion of the FEP would only decrease the expected annual dose). For this case, the succinct description of the FEP reasoning used in the summary tables is "Excluded based on low consequence (conservative bound)." For example, a few radioisotopes, such as Np, may be incorporated into the structure of phases of other minerals (primarily uranium minerals) that form during degradation of the waste. For chemically similar elements such as Np and U, the effective solubility of Np as a minor constituent of a uranium mineral is less than the solubility of the analogues pure Np mineral. However, this FEP, "secondary phase effects on dissolved radionuclide concentrations at the waste form" in Table 2.3-7, was conservatively excluded in TSPA-SR. The degree of conservatism is not quantified since the collection of data to justify the model would require either too much time or too many resources for the surmised potential benefit. The exclusion of FEPs that have a potential beneficial consequence is a conservatism of the FEPs screening process. This conservatism is in addition to the many conservative approaches used to include FEPs as more fully discussed in Chapter 3.

2.3.2.1 FEPs Related to the Radioisotope Inventory Component

Five FEPs relate to the Radioisotope Inventory Component (Table 2.3-1). The waste inventory was reconsidered and 27 important radionuclides identified. The radioisotopic gas inventory was omitted because the screening analysis showed that gases were not significant contributors to dose. However, one radioisotopic gas, ^{14}C, is included as a dissolved species. Not all isotopes of uranium and plutonium are included in the 27 important radionuclides, and so the proper proportion ("dilution") of the included uranium and plutonium isotopes is evaluated for the Waste Form Degradation Model when evaluating solubility. For the TSPA-SR analysis, radioactive decay (and ingrowth[6]) is modeled, and the heterogeneity of the inventory in the waste form is included using three waste forms (CSNF, DSNF, and HLW).

Table 2.3-1. FEPs Related to the Radioisotope Inventory Component

FEP Number	FEP Title	Include/ Exclude	Process Model Report (PMR)	AMR Document Identifier (DI)	Reason for Include/Exclude Decision
2.1.01.01.00	Waste inventory	Include	WF	ANL-WIS-MD-000009 ANL-WIS-MD-000006	Included: Average and bounding inventories for 27 radioisotopes for 3 waste allocation categories (CSNF, DSNF, and HLW) have been developed; 24 isotopes from screening arguments based on human dose, two isotopes mandated by the Groundwater Protection Requirement of the proposed 40 CFR 197 and one isotope (^{235}U) to properly perform radioactive decay; different subsets of the isotopes are used for the various scenarios: volcanic release (16), nominal groundwater release (21), and human intrusion (23). This FEP is the topic of AMR, *Inventory Abstraction*.
2.1.01.03.00	Heterogeneity of waste forms	Include	WF	ANL-WIS-MD-000009	Heterogeneity of waste forms, is included by handling CSNF, DSNF, and HLW separately. Heterogeneity is included within these three categories only to the extent that it is used to determine the average or bounding inventory.

[6] Ingrowth is the further production of a radioisotope as its parent decays, e.g., initial inventory of ^{237}Np increases as the parent ^{241}Am decays.

Table 2.3-1. FEPs Related to the Radioisotope Inventory Component (Continued)

FEP Number	FEP Title	Include/ Exclude	Process Model Report (PMR)	AMR Document Identifier (DI)	Reason for Include/Exclude Decision
2.1.12.07.00	Radioactive gases in waste and EBS	Exclude	WF, EBS	ANL-WIS-MD-000009	Excluded based on low consequence: The potential human dose from inhalation of radioactive gases was low relative to other doses and, thus they were screened out except for dissolved ^{14}C. Other influences of gases (radioactive or not) are also excluded as discussed in other FEPs: Biological activity in the waste is screened out (colloid FEP 2.1.10.01.00) and so generation of microbial gases such as CO_2, CH_4, H_2S are also screened out (in-package chemistry FEP 2.1.12.04.00). The chemical influence of H_2 is conservatively neglected (in-package chemistry FEP 2.1.12.03.00). Generally, the physical aspects of gas generation such as displacement of oxygen and pressurization of the disposal drifts by He and H_2 (in-package chemistry FEPs 2.1.12.02.00 and 2.1.12.03.00, respectively) have been excluded since gas would readily dissipate from the repository over geologic time (i.e., because fractures and fault zones are unsaturated, the relative permeability to gas is large). (Note: the relative permeability of the fractures and fault zones to water is lower than for air and is included in the unsaturated flow model—see FEPs related to unsaturated flow model.) Included: See cladding FEP 2.1.02.20.00 where the pressurization of CSNF cladding is included.
3.1.01.01.00	Radioactive decay and ingrowth	Include	WF, UZ, SZ	ANL-WIS-MD-000009	Included: Radioactive decay and ingrowth (i.e., further production of a radioisotope as its parent decays) is either incorporated in the computer codes used for the TSPA-SR total system model, or the inventory of a daughter is conservatively increased when generating the inventory in cases where decay and ingrowth are not explicitly modeled.
3.2.07.01.00	Isotopic dilution	Include/ Exclude	WF, SZ	ANL-WIS-MD-000009	Included: Mixing or dilution of specific radionuclides of the same element within the WP is accomplished by assuming that the dissolved isotopic mass at a given elemental concentration is proportional to the isotopic inventory at that time. Exclude outside waste package based on low consequence (conservative bound). Isotopic dilution during transport away from the WP due to stable and/or naturally occurring isotopes of the same element; dilution only decreases adverse radiological effects.

2.3.2.2 FEPs Related to the In-Package Chemistry Component

Twenty-five FEPs relate to the In-Package Chemistry Component (Table 2.3-2). Prior to TSPA-SR analysis, uncertainty in the in-package chemistry was evaluated through scientific judgment; but objectively demonstrating that the uncertainty from these 25 FEPs was included was difficult; an in-package chemistry component was developed for TSPA-SR analysis to explicitly include many of these FEPs. Furthermore, a few FEPs dealt with the coupling of the in-package chemistry with other model components; these FEPs can now be included in the current TSPA-SR analysis.

Table 2.3-2. FEPs Related to the In-Package Chemistry Component

FEP Number	FEP Title	Include/ Exclude	Process Model Report (PMR)	AMR Document Identifier (DI)	Reason for Include/Exclude Decision
2.1.01.02.00	Codisposal/ colocation of waste	Include/ Exclude	WF	ANL-WIS-MD-000009	Included: The chemical interaction between DSNF and HLW in one codisposal package is explicitly included in the In-Package Chemistry Component. Codisposal/colocation of DSNF and HLW is also included in the TSPA-SR by separately modeling waste form degradation of both categories within a codisposal waste package (see DSNF FEP 2.1.02.01.00 and HLW FEP 2.1.02.03.00). Colocation: outside WF scope.

Table 2.3-2. FEPs Related to the In-Package Chemistry Component (Continued)

FEP Number	FEP Title	Include/ Exclude	Process Model Report (PMR)	AMR Document Identifier (DI)	Reason for Include/Exclude Decision
2.1.01.04.00	Spatial heterogeneity of emplaced waste	Exclude/ Include	WF	ANL-WIS-MD-000009	Excluded based on low consequence: The time scale of convective transport and diffusion of aqueous species in the fully flooded waste package is rapid in relation to alteration of the contents (waste form, cladding, and support material); consequently, the contents are assumed to be uniformly mixed and spatial heterogeneity in the waste to evaluate in-package chemistry is excluded. Spatial heterogeneity of the two types of waste packages modeled is also excluded based on low consequence. WPs of CSNF and codisposed DSNF and HLW are to be closely packed together so that variations in temperature (which can influence aspects of waste form degradation and in-package chemistry) of individual WPs will not vary significantly. Included: Spatial heterogeneity of temperatures in a waste package are evaluated to determine perforation through creep rupture (cladding FEP 2.1.02.19.00) and stress corrosion cracking (cladding FEP 2.1.02.21.00).
2.1.02.09.00	Void space (in waste package)	Include/ Exclude	WF	ANL-WIS-MD-000009	Included: For determining the radioisotope concentration, the Dissolved Radioisotope Concentration Component uses a time varying void volume of the altered waste form (see Solubility FEP 2.1.09.12.00). In addition, in the process model for evaluating the In-Package Chemistry Component, the mixing cell volume is equal to the fixed void volume in the waste package; hence, void space is indirectly included in the In-Package Chemistry Component. Excluded based on low consequence. The potentially time varying void space available and its influence on determining the in-package chemistry was excluded based on low consequence. Rather, the uncertainty in the in-package chemistry that might be caused by different void volumes would be swamped by the use of various water inflow rates into the waste package that were used to define the various simulations and, thereby, the uncertainty used in the In-Package Chemistry Component.
2.1.02.10.00	Cellulosic degradation	Exclude	WF	ANL-WIS-MD-000009	Excluded because of low probability (credibility). According to preliminary waste acceptance criteria (DOE 1999c, Section 4.2.3.A.3), no cellulosic materials will be included as part of the waste in the potential Yucca Mountain repository.

Table 2.3-2. FEPs Related to the In-Package Chemistry Component (Continued)

FEP Number	FEP Title	Include/ Exclude	Process Model Report (PMR)	AMR Document Identifier (DI)	Reason for Include/Exclude Decision
2.1.03.06.00	Internal corrosion of waste container	Include/ Exclude	WF, WP	ANL-WIS-MD-000009	Included: After breach of the WP, the corrosion of the inner structural stainless steel is important to determining the in-package chemistry (provided a high sulfur steel is used) and is, thus, included in the process model and as part of the uncertainty of the In-Package Chemistry Component (see also in-package chemistry FEP 2.1.09.02.00). Excluded based on low consequence: Prior to WP breach, internal corrosion will not be significant. The current design specifies placement of inert gases in the package and limited water allowed in the CSNF (see cladding FEP 2.1.02.11.00) and HLW glass packages. Some canisters containing DSNF such as those containing N-Reactor fuel, may have more water, but this water would be scavenged by the waste form itself.
2.1.08.07.00	Pathways for unsaturated flow and transport in the waste and EBS	Include/ Exclude	WF, EBS	ANL-WIS-MD-000009	Include through the use of a series of linked one-dimensional flow paths and mixing cells through the EBS, drip shield, waste package, and into the invert. Exclude based on low consequence (conservative bound): transport pathways inside container excluded; rather, container assumed to be a mixing cell for In-Package Chemistry Component. Inclusion of the pathways would delay release of radionuclides and thus they are conservatively neglected.
2.1.08.08.00	Induced hydrological changes in the waste and EBS	Include/ Exclude	WF, EBS	ANL-WIS-MD-000009	Included: Changes in flow volume into the WP as it degrades are included. Changes in the exposed fuel area as CSNF cladding fails are also included (see cladding unzipping FEP 2.1.02.23.00). Excluded based on low consequence: Changes to hydrologic properties of the waste form and invert are excluded since the slight changes in hydrologic properties are small in relation to the overall uncertainty of the in-package chemistry predictions.
2.1.08.10.00	Desaturation/ dewatering of the repository	Include	WF, NFE	ANL-WIS-MD-000009	Included through unsaturated flow modeling in the TSPA thermal hydrologic models which, in turn, affects the volume of water entering the disposal WPs.

Table 2.3-2. FEPs Related to the In-Package Chemistry Component (Continued)

FEP Number	FEP Title	Include/ Exclude	Process Model Report (PMR)	AMR Document Identifier (DI)	Reason for Include/Exclude Decision
2.1.09.01.00	Properties of the potential carrier plume in the waste and EBS	Include/ Exclude	WF	ANL-WIS-MD-000009	Included: Water entering the package is assumed to have chemical properties of J-13 well water in the process models of the in-package chemistry. Water chemistry was then altered based on waste type, cladding failure, seepage flux, and waste degradation rate. Furthermore, corrosion of steel is directly used to establish an uncertainty range for the water chemistry. Excluded based on low consequence: the changing properties of incoming water, as evaluated by EBS. Although the changing properties of the incoming water as evaluated by EBS are not coupled to these process calculations, slight changes in the incoming water over time are swamped by the dramatic changes that are predicted in the in-package chemistry because of the waste and internal parts of the waste package. That is, the range of uncertainty used for the In-Package Chemistry Component is not changed by the minor changes of the initial chemistry of the water entering the package. This fact is especially true now that a concrete tunnel liner is not used.
2.1.09.02.00	Interaction with corrosion products	Exclude/ Include	WF, EBS	ANL-WIS-MD-000009	Included: The chemical effects of the corrosion of steel (e.g., release of sulfur) on the in-package water chemistry have been included (see also in-package chemistry FEP 2.1.03.06.00). Several other interactions are also included and discussed in other FEPs: the presence of corrosion product is included in determining the availability of water for radioisotope dissolution (solubility FEP 2.1.09.12.00); the volume increase of the corrosion products and the unzipping of the cladding is included (cladding FEP 2.1.09.03.00); and the sorption on mobile colloids is included (colloid FEP 2.1.09.05.00). Coupling of related processes was also included (see in-package chemistry coupled processes FEP 2.1.11.04.00). Excluded based on low consequence (conservative bound): The potential beneficial effects from corrosion products—which include sorption on immobile corrosion products (colloid FEP 2.1.09.05.00), and the decreased ability for advective and diffusive transport of radioisotopes—have been conservatively excluded.

Table 2.3-2. FEPs Related to the In-Package Chemistry Component (Continued)

FEP Number	FEP Title	Include/ Exclude	Process Model Report (PMR)	AMR Document Identifier (DI)	Reason for Include/Exclude Decision
2.1.09.06.00	Reduction-oxidation potential in waste and EBS	Include	WF, EBS	ANL-WIS-MD-000009	In developing the regression equation of pH for the In-Package Chemistry Component, which is based on the EQ3/6 equilibrium process model, the fluid is assumed equilibrated with the atmosphere to ensure maximum plausible O_2 and CO_2 conditions. Furthermore, to match these assumptions, the In-Package Chemistry Component sets the O_2 and CO_2 conditions at atmospheric conditions. These conditions are then used by the CSNF Matrix Degradation Component (see CSNF FEP 2.1.02.02.00) and Dissolved Radioisotope Concentration Component (see Solubility FEP 2.1.09.04.00).
2.1.09.07.00	Reaction kinetics in waste and EBS	Include/ Exclude	WF, EBS	ANL-WIS-MD-000009	Included reaction kinetics through quasi-equilibrium analysis. The equilibrium model evaluates the pH over time as a function of several rates of degradation of the cladding, SNF matrix, HLW glass, and internal components of the disposal package (stainless steel and aluminum); the resulting variance in the pH is then used to set bounds on the uncertainty of the in-package water chemistry. Excluded based on low consequence of the reaction transients between time steps: The in-package chemistry process model, EQ3/6, assumes instantaneous equilibrium between changes in amounts of corrosion products available. Small enough time steps are taken to avoid errors exceeding uncertainty in thermodynamic parameters.
2.1.09.08.00	Chemical gradients / enhanced diffusion in waste and EBS	Exclude	WF, EBS	ANL-WIS-MD-000009	Excluded based on low consequence. The WP is assumed to be a mixing cell without chemical gradients in the In-Package Chemistry Component. Provided the WP is fully saturated, the rate of the flow of water into and out of the package is slow enough that no long-term gradient would be present and equilibrium would occur. Furthermore, rather than include diffusive release of radionuclides out of the CSNF perforated cladding, DSNF perforated cladding, or cracked HLW glass, a more conservative conceptual model is developed that subsumes enhanced release from diffusion caused by chemical gradients, cladding unzipping for CSNF (cladding unzipping FEP 2.1.02.23.00), 100% failed cladding for DSNF except naval SNF (DSNF cladding FEP 2.1.02.25.00), and very high reactive surface area for HLW glass (HLW surface area FEP 2.1.02.05.00). Performance analysis of naval SNF will account for the performance of the cladding.

Table 2.3-2. FEPs Related to the In-Package Chemistry Component (Continued)

FEP Number	FEP Title	Include/ Exclude	Process Model Report (PMR)	AMR Document Identifier (DI)	Reason for Include/Exclude Decision
2.1.09.09.00	Electrochemical effects (electrophore-sis, galvanic coupling) in waste and EBS	Exclude	WF	ANL-WIS-MD-000009	Excluded based on low consequence. The influence of fluid flow through a failed container on the in-package chemistry is much greater than any effect on the degradation of the SNF or HLW matrix that can be created by electrophoresis or electro-osmosis.
2.1.09.11.00	Waste-rock contact	Include/ Exclude	WF, EBS	ANL-WIS-MD-000009	Included: The indirect influence of waste-rock contact on in-package chemistry is included through the use of the J-13 well water (in-package chemistry FEP 2.2.08.12.00). The perforation of cladding from shaking in a severe earthquake is included as a disruptive event (cladding FEP 2.1.02.24.00). Excluded based on low consequence: Because of the drip shield and the long-lived waste package, rock is not expected to come in contact with the waste in the first 10,000 yr. Furthermore, even if some contact were to occur, the overall result would be little or no involvement of the rock minerals in chemical reaction due to their dissolution kinetics. The perforation of CSNF cladding from rock fall is also excluded in the first 10,000 yr (cladding FEP 2.1.07.01.00).
2.1.11.04.00	Temperature effects / coupled processes in waste and EBS	Include/ Exclude	WF, EBS	ANL-WIS-MD-000009	Included: Many of the processes of the Waste Form Degradation model are coupled. CSNF matrix degradation (CSNF FEP 2.1.02.02.00), HLW degradation (HLW FEP 2.1.02.03.00), and Dissolved Radioisotope Concentration (solubility FEP 2.1.09.04.00) are coupled to the chemical conditions calculated by the In-Package Chemistry Component (this FEP). Cladding unzipping (Cladding FEP 2.1.02.23.00) is coupled to CSNF matrix degradation (see also in-package chemistry FEP 2.1.09.02.00). In turn, weak feedback to the In-Package Chemistry Component occurs from HLW degradation and cladding degradation. Temperature is also coupled to CSNF matrix degradation (CSNF FEP 2.1.11.05.00), CSNF cladding degradation (cladding FEP 2.1.11.07.00), and HLW degradation (HLW FEP 2.1.11.01.00), and solubility of uranium (solubility FEP 2.1.09.04.00). Excluded based on low consequence: because the primary effects of temperature on waste form degradation are included directly, the secondary effects of temperature on in-package chemistry such as pH are of low additional consequence (also see in-package chemistry FEP 2.1.11.08.00).

Table 2.3-2. FEPs Related to the In-Package Chemistry Component (Continued)

FEP Number	FEP Title	Include/ Exclude	Process Model Report (PMR)	AMR Document Identifier (DI)	Reason for Include/Exclude Decision
2.1.11.08.00	Thermal effects: chemical and microbiological changes in the waste and EBS	Include/ Exclude	WF, NFE, EBS	ANL-WIS-MD-000009	Included: temperature effects are included directly in various components (e.g., cladding degradation rate [cladding FEP 2.1.11.07.00], CSNF matrix degradation rate [CSNF FEP 2.1.02.02.00], HLW degradation rate [HLW FEP 2.1.02.03.00], and solubility of uranium [solubility FEP 2.1.09.04.00]). Excluded based on low consequence: Because the primary effects of temperature on waste form degradation are included directly, the secondary effects of temperature on in-package chemistry such as pH are of low additional consequence. Furthermore, the use of a cooler repository design, drip shield, and long-lived WP implies WP breach occurs when temperatures are near ambient temperature where thermodynamic data has been collected. Thus, the collection of thermodynamic data as a function of temperature is not necessary. Thermal effects on microbiological activity are excluded since microbiological FEP was screened out as a process that effects waste form degradation (colloid FEP 2.1.10.01.00).
2.1.11.09.00	Thermal effects on liquid or two-phase fluid flow in the waste and EBS	Include/ Exclude	WF, EBS, NFE	ANL-WIS-MD-000009	Included: thermal effects on fluid flow are included indirectly to the extent that they influence the seepage flux into the drift. Excluded based on low consequence. Thermal effects on flow within the waste form are excluded because temperature within and outside the package will be fairly uniform by the time WPs fail, and thus thermal gradients will not significantly influence flow. Furthermore, temperature will be too low to promote two-phase flow (see in-package chemistry FEPs 2.1.11.10.00 and 2.1.09.08.00 on thermal and chemical diffusion processes, which are also excluded).
2.1.11.10.00	Thermal effects on diffusion (Soret effect) in waste and EBS	Exclude	WF, EBS, NFE	ANL-WIS-MD-000009	Excluded based on low consequence. Bounding analyses indicate diffusion due to temperature gradients within the waste form is insignificant (see cladding gap release FEP 2.1.02.07.00).

Table 2.3-2. FEPs Related to the In-Package Chemistry Component (Continued)

FEP Number	FEP Title	Include/ Exclude	Process Model Report (PMR)	AMR Document Identifier (DI)	Reason for Include/Exclude Decision
2.1.12.01.00	Gas generation	Exclude	WF, EBS, UZ	ANL-WIS-MD-000009	Excluded based on low consequence: Prior to breach of a WP, gas generation will be insignificant (see in-package chemistry FEP 2.1.03.06.00 on internal corrosion of waste container). After breach of a WP, the chemical influence of gas generation of H_2 has been conservatively excluded since it moderates the low pH values encountered from sulfur release from steel (see in-package FEP 2.1.12.03.00). Generation of radioactive gases in waste is also excluded since the inhalation dose is low relative to ingestion doses (inventory FEP 2.1.12.07.00). In addition, most physical aspects of gas generation such as displacement of oxygen and pressurization of the disposal drifts by He and H_2 (in-package chemistry FEPs 2.1.12.02.00 and 2.1.12.03.00, respectively) have been excluded since gas would readily dissipate from the repository over geologic time (i.e., because fractures and fault zones are unsaturated, the relative permeability to gas is large). (Note: The relative permeability of the fractures and fault zones to water is lower than air and included in the unsaturated flow model—see FEPs related to unsaturated flow model.) Only the physical aspect of pressurization of CSNF cladding is indirectly included (cladding FEP 2.1.02.20.00).
2.1.12.02.00	Gas generation (He) from fuel decay	Exclude	WF, EBS	ANL-WIS-MD-000009	Excluded based on low consequence. Radioactive alpha decay of actinides such as Pu, U, Th, etc., could result in the creation of additional He gas within the waste form; however, studies done for TSPA-VA shows increases in internal pressures from He to be small relative to fission gas pressures. Internal gas pressure inside of CSNF cladding is a driving force for cladding creep and stress corrosion cracking and is included in 2.1.01.20.00.
2.1.12.03.00	Gas generation (H_2) from metal corrosion	Exclude	WF, EBS, WP	ANL-WIS-MD-000009	Excluded based on low consequence. The amount of hydrogen that could be produced from metal corrosion will be small since the repository is likely to be primarily oxic since it is well connected to the surface. Also, because the repository is connected to the surface, any gas produced is expected to escape. The influence of H_2 gas on in-package chemistry is negligible. It is conservative to exclude H_2 evolution from steel corrosion as this limits pH depression. See cladding FEP 2.1.02.22.00 for effect of H_2 on cladding degradation.

Table 2.3-2. FEPs Related to the In-Package Chemistry Component (Continued)

FEP Number	FEP Title	Include/ Exclude	Process Model Report (PMR)	AMR Document Identifier (DI)	Reason for Include/Exclude Decision
2.1.12.04.00	Gas generation (CO_2, CH_4, H_2S) from microbial degradation	Exclude	WF, EBS	ANL-WIS-MD-000009	Excluded based on low probability (credibility) and consequence. Biological activity inside the WP is excluded (colloid FEP 2.1.10.01.00) since no organic material will be allowed inside the WP based on preliminary waste acceptance criteria (DOE 1999c, Section 4.2.3.A.3). Therefore, the effects of biological gases on the in-package chemistry are also expected to be negligible.
2.1.12.06.00	Gas transport in waste and EBS	Exclude	WF, EBS	ANL-WIS-MD-000009 ANL-WIS-MD-000006	Excluded based on low consequence: Screening studies conclude that if most gaseous radioisotopes, except ^{14}C, were to be released through an atmospheric pathway, the potential dose relative to the ingestion dose would be low thus most gaseous radioisotopes were screened out (waste inventory FEP 2.1.01.01.00). However, in TSPA-SR, rather than transport ^{14}C as a gas (which would normally be incorporated in CO_2), ^{14}C was transported to a receptor entirely in the aqueous phase (i.e., gas transport of ^{14}C is included indirectly). This approach is conservative since less dilution occurs in aqueous transport).
2.2.08.12.00	Use of J-13 well water as a surrogate for water flowing into the EBS and waste	Include	WF	ANL-WIS-MD-000009	J-13 well water is used to set initial water chemistry inside the WP. On-going work will verify or improve this assumption.

2.3.2.3 FEPs Related to the CSNF Matrix Degradation Component

Seven FEPs relate to the CSNF Matrix Degradation Component (Table 2.3-3). The process of CSNF dissolution and radioisotope release has been included in TSPA analysis for a number of years through an regression equation of experimental results on both irradiated and unirradiated fuel. The use of samples of irradiated fuel implies that the potential enhanced dissolution from radiolysis, radiation damage, and alpha recoil has indirectly been included in the dissolution rates. The small differences in results and theoretical arguments, however, show that these latter effects are actually insignificant and can be excluded from consideration. While an unimportant distinction for the CSNF since experimental results are available that include these effects, enhanced dissolution from radiolysis, radiation damage, and alpha recoil are excluded from DSNF and HLW degradation as discussed below. The process of magma interacting with the waste has formally been included this year in the analysis for the TSPA-SR and is described in the Disruptive Events PMR.

Table 2.3-3. FEPs Related to the CSNF Matrix Degradation Component

FEP Number	FEP Title	Include/ Exclude	Process Model Report (PMR)	AMR Document Identifier (DI)	Reason for Include/Exclude Decision
1.2.04.04.00	Magma interacts with waste	Include	WF, Tec, WP	ANL-WIS-MD-000009	Included: Direct volcanic disruption of the repository is included as a process in TSPA-SR. Hence, magmatic interaction with the waste is plausible and included also. This FEP sets plausible particle sizes for the fragmented waste. Direct volcanic effects (i.e., radioisotopes carried by ash plumes from volcanic eruptions) are modeled as described in the Disruptive Events Report.
2.1.02.02.00	CSNF alteration, dissolution, and radionuclide release	Include	WF	ANL-WIS-MD-000009	CSNF degradation is included in the TSPA-SR analysis as a source term for the mobilization of contaminants. A kinetic rate equation, assuming saturated conditions, is used where terms of the coefficients have been evaluated through regression analysis on experimental data obtained over a range of temperatures and water chemistry.
2.1.02.04.00	Alpha recoil enhances dissolution	Exclude	WF	ANL-WIS-MD-000009	Excluded based on low consequence. The radioactive decay processes that directly increase waste matrix corrosion are bounded by alpha-recoil rates. Even when assuming that all radioactive decays result in alpha-recoils, analysis shows that it will not cause significant increases to the degradation rate of the different waste forms (CSNF, DSNF, or HLW).
2.1.08.07.05	Waste-form and backfill consolidation	Exclude	WF	ANL-WIS-MD-000009	Excluded based on consequence (low conservative bound) since backfill and waste form consolidation would tend to decrease the available reactive surface area. The potential deleterious effect of maintaining water contact with the waste form is already conservatively bounded (with or without backfill) by assuming the waste package is fully flooded with water.
2.1.11.05.00	Differing thermal expansion of repository component	Include	WF	ANL-WIS-MD-00009 ANL-EBS-MD-000015	Degradation of CSNF is a function of temperature in the CSNF Matrix Degradation Component. This FEP is the topic of *CSNF Waste Form Degradation: Summary and Abstraction*; influence of temperature is also discussed in FEPs 2.1.11.07.00 on cladding and 2.1.11.01.00 on HLW.

Table 2.3-3. FEPs Related to the CSNF Matrix Degradation Component (Continued)

FEP Number	FEP Title	Include/ Exclude	Process Model Report (PMR)	AMR Document Identifier (DI)	Reason for Include/Exclude Decision
2.1.13.01.00	Radiolysis	Exclude	WF, EBS, WP	ANL-WIS-MD-000009	Excluded based on low consequence. Fluid is not expected to contact the waste (CSNF, DSNF, or HLW) until gamma radiolysis has become negligible. Specifically for CSNF and HLW, alpha radiolysis is included since irradiated fuel and active HLW was used for some of the experiments from which regression equations were developed. It is, therefore, not necessary to include radiolysis explicitly in the models.
2.1.13.02.00	Radiation damage in waste and EBS	Exclude	WF, WP, EBS	ANL-WIS-MD-000009 ANL-EBS-MD-000015 ANL-WIS-MD-000010	Excluded based on low consequence. Experimental data show little influence of burnup on degradation rate of CSNF as described in AMR, *CSNF Waste Form Degradation Summary Abstraction*. However, radiation damage was conservatively assumed for evaluating the solubility of Pu, Pa, and Cm when the controlling solid was unknown; that is, an amorphous controlling solid was used, thereby indirectly assuming radiation damage to the crystal lattice of the unknown controlling solid.

2.3.2.4 FEPs Related to the CSNF Cladding Degradation Component

Twenty FEPs relate to the CSNF Cladding Degradation Component (Table 2.3-4). These 20 FEPs were developed based on comments received on early TSPAs (e.g., DSNF-TSPA-93-SNL [Rechard 1993] and DSNF-TSPA-94-SNL [Rechard 1995] and the TSPA-VA [DOE 1998b]).

Several perforation mechanisms have been included in the CSNF Cladding Degradation Component: initial perforation in reactor before receipt of the waste, creep perforation, stress corrosion cracking, mechanical failure due to severe earthquakes, and localized corrosion. The influence of temperature on creep and stress corrosion cracking are also included. Localized crevice corrosion, and diffusion-controlled cavity growth, have been excluded based on FEP arguments. Based on arguments in an AMR dealing solely with this topic, hydride perforation mechanisms such as hydride embrittlement and DHC have also been excluded.

Complete failure of the cladding after perforation is modeled through unzipping of the cladding in an aqueous environment[7]. Unzipping in a very hot, dry environment where the UO_2 is rapidly oxidized to U_3O_8 is excluded based on low consequence since the required high temperature conditions do not exist when WP failure typically occurs.

[7] The unzipping in a wet environment is hypothetical since it has not been observed experimentally or in storage pools at reactors. The unzipping process conservatively bounds the slow release of radionuclides through pinhole perforations. This is important for release of radionuclides, such as ^{99}Tc, which are not controlled by solubility.

Table 2.3-4. FEPs Related to the CSNF Cladding Degradation Component

FEP Number	FEP Title	Include/ Exclude	Process Model Report (PMR)	AMR Document Identifier (DI)	Reason for Include/Exclude Decision
2.1.02.07.00	Gap and grain release of radionuclides after cladding perforation	Include/ Exclude	WF	ANL-WIS-MD-000009 ANL-WIS-MD-000007	Included: The inventory of radionuclides located at gap and grain boundaries is included. This inventory is released either when the WP fails (for cladding that is already perforated) or when the cladding is first perforated. Excluded based on low consequence: Generation of additional gap and grain-boundary inventory after emplacement in the repository since temperatures are not high enough to promote sufficient diffusion. Interaction with impurities in the waste matrix which mitigate the release of the gap and grain boundary inventory is also excluded both because the reactions are thought to be limited and because it is conservative to do so.
2.1.02.11.00	Waterlogged rods	Exclude	WF	ANL-WIS-MD-000008	Excluded based on low consequence. The moisture in a dried fuel rod using standard drying processes is insufficient to cause further degradation of spent fuel cladding or structural stainless steel of container.
2.1.02.12.00	Cladding degradation before YMP receives it	Include	WF	ANL-WIS-MD-000008 ANL-EBS-MD-000048	The CSNF Cladding Degradation Component includes a distribution with a median of 0.1% of fuel rods perforated during reactor operations. The minor additional failures during less harsh storage and transportation conditions are also included but do not substantially change the distribution of perforations. This FEP is the topic of AMR– *Initial Cladding Condition*.
2.1.02.13.00	General corrosion of cladding	Exclude	WF	ANL-WIS-MD-000008	Excluded based on low consequence. General corrosion of Zircaloy cladding is very slow and will not be important in the first 10,000 yrs or even 100,000 yr.
2.1.02.14.00	Microbially influenced corrosion (MIC) of cladding	Exclude/ Include	WF	ANL-WIS-MD-000008	While microbial activity has been screened out as a significant bulk process at YMP (colloid FEP 2.1.10.01.00), it has not been excluded for localized effects. There is no experimental evidence of enhanced MIC of zirconium metal or alloys, however, the possibility of local depression of pH by microbes is included in the conservative local corrosion model.
2.1.02.15.00	Acid corrosion of cladding from radiolysis	Exclude/ Include	WF	ANL-WIS-MD-000008	Excluded based on low consequence. Zirconium is used for fuel cladding because of its resistance to corrosion in highly acidic environments including those local environments in high radiation fields and studies show that zirconium has excellent corrosion resistance to nitric acids and concentrated hydrogen peroxide. However, this process is has not been ruled out as one of the possible contributors to local suppression of pH resulting in local corrosion of cladding and is thus included in the local corrosion model.

Table 2.3-4. FEPs Related to the CSNF Cladding Degradation Component (Continued)

FEP Number	FEP Title	Include/ Exclude	Process Model Report (PMR)	AMR Document Identifier (DI)	Reason for Include/Exclude Decision
2.1.02.16.00	Localized corrosion of cladding through pitting	Exclude/ Include	WF	ANL-WIS-MD-000008 ANL-EBS-MD-000012	Include: While the conditions for pitting corrosion of the cladding (very low pH and high anion concentration) are not predicted for the bulk solution within the WP, they have not yet been ruled out for localized regions. Thus, a conservative localized corrosion model has been included in the TSPA-SR as discussed in cladding FEP 2.1.02.27.00. This FEP is the topic of AMR, *Clad Degradation-Local Corrosion of Zirconium and its Alloys Under Repository Conditions*
2.1.02.17.00	Localized corrosion (crevice corrosion) of cladding	Exclude	WF	ANL-WIS-MD-000008 ANL-EBS-MD-000012	Excluded based on low probability (credibility). The CSNF Cladding Degradation Component excludes a component that accounts for localized corrosion of the cladding through crevice corrosion because zirconium does not corrode in this manner. This FEP is the topic of AMR, *Clad Degradation-Local Corrosion of Zirconium and its Alloys Under Repository Conditions.*
2.1.02.18.00	High dissolved silica content of waters enhances corrosion of cladding	Exclude	WF	ANL-WIS-MD-000008	Excluded based on low consequence. Silica is stable and is not corrosive to most materials, including Zircaloy.
2.1.02.19.00	Creep rupture of cladding	Include	WF	ANL-WIS-MD-000008 ANL-WIS-MD-000007	Perforation of Zircaloy cladding from creep rupture at high temperature is included in the CSNF Cladding Degradation Component as described in detail in AMR, *Clad Degradation – Summary and Abstraction*. The creep model conservatively bounds recent experiments on irradiated cladding.
2.1.02.20.00	Pressurization from He production causes cladding failure	Include	WF	ANL-WIS-MD-000008	Both DHC and creep perforation are driven by the cladding stress caused by the internal gas (including initial fill gas, fission product gases, and helium gas from alpha decay) and, thus indirectly, pressurization is included in the CSNF Cladding Degradation Component.
2.1.02.21.00	Stress corrosion cracking (SCC) of cladding	Include	WF	ANL-WIS-MD-000008 ANL-WIS-MD-000007	Stress corrosion cracking is modeled as a perforation mechanism for rods with high stresses. SCC is primarily relevant to iodine since SCC from other halogens such as chlorides is generally not observed. However, fluoride is considered independently as a localized corrosion mechanism (cladding FEP 2.1.02.16.00).
2.1.02.22.00	Hydride embrittlement of cladding	Exclude	WF	ANL-WIS-MD-000008 ANL-EBS-MD-000011	Excluded based on low consequence. Experimental data indicate that the in-package environment and cladding stresses are not conducive to hydride cracking and embrittlement. This FEP is the subject of AMR, *Hydride-Related Degradation of SNF Cladding under Repository Conditions*.

Table 2.3-4. FEPs Related to the CSNF Cladding Degradation Component (Continued)

FEP Number	FEP Title	Include/ Exclude	Process Model Report (PMR)	AMR Document Identifier (DI)	Reason for Include/Exclude Decision
2.1.02.23.00	Cladding unzipping	Include/ Exclude	WF	ANL-WIS-MD-000008 ANL-EBS-MD-000013 ANL-EBS-MD-000014	Included: Unzipping of cladding in an aqueous environment is included as the key element in exposing and dissolving fuel after perforation of the cladding. This mechanism conservatively bounds the slow diffusive release of radionuclides through pinholes of the fuel cladding. "Wet unzipping" is the topic of AMR, *Clad Degradation - Wet Unzipping*. Excluded: Unzipping of cladding in a dry environment is excluded based on low consequence. In the environment of the repository only a very small fraction of the fuel would undergo dry unzipping, and then only if the disposal container was breached in the first few hundred years, and the cladding was already perforated such that the UO_2 would be rapidly oxidized to U_3O_8. Dry unzipping is the topic of AMR, *Clad Degradation - Dry Unzipping*.
2.1.02.24.00	Mechanical failure of cladding	Include	WF	ANL-WIS-MD-000008	The CSNF Cladding Degradation Component includes perforation of cladding from severe earthquakes (once per million years).
2.1.02.27.00	Localized corrosion perforation of cladding by fluoride	Include	WF	ANL-WIS-MD-000008 ANL-EBS-MD-000012	The CSNF Cladding Degradation Component includes a component that accounts for localized corrosion of the cladding through corrosion by fluorides flowing into the WP. This FEP is the topic of AMR, *Clad Degradation Local Corrosion of Zirconium and its Alloys Under Repository Conditions*.
2.1.02.28.00	Diffusion-controlled cavity growth (DCCG) concerns	Exclude	WF	ANL-WIS-MD-000008 ANL-WIS-MD-000007	Excluded based on low probability. Diffusion-controlled cavity growth as a mechanism to fail Zircaloy cladding has not been observed experimentally. Excluded based on low probability (credibility): NRC once required license applicants for dry storage of CSNF to assume DCCG to evaluate dry storage designs. Recent literature does not support the use of this specific creep mechanism for zirconium materials since it has not been observed experimentally (voids and cavities are rarely seen in irradiated Zircaloy). Current NRC Interim Staff Guidance permits license applicants to use other creep models. Although DCCG is excluded as a specific type of creep rupture mechanism, creep rupture, as a general cladding perforation process, is included (see cladding FEP 2.1.02.19.00).

Table 2.3-4. FEPs Related to the CSNF Cladding Degradation Component (Continued)

FEP Number	FEP Title	Include/ Exclude	Process Model Report (PMR)	AMR Document Identifier (DI)	Reason for Include/Exclude Decision
2.1.07.01.00	Rockfall (Large Block)	Exclude	WF	ANL-WIS-MD-000009 ANL-WIS-MD-000008	Excluded based on low consequence. Cladding perforation from the collapse of the WP with a large block rockfall is not considered since cladding perforation from other modes occurs before the WP can collapse (see also FEP 2.1.02.24.00).
2.1.09.03.00	Volume increase of corrosion products	Include/ Exclude	WF, WP	ANL-WIS-MD-000009	Included: The underlying driving mechanism assumed for wet unzipping is the volume increase as the UO_2 forms secondary minerals, and thus this FEP is included in the CSNF Cladding Degradation Component. Excluded based on low consequence. Dry oxidation of CSNF is not expected to be a significant contributor to the results since it can only occur when both the WP and cladding fail in the first 300 years (see cladding FEP 2.1.02.23.00).
2.1.11.07.00	Thermally-induced stress changes in waste and EBS	Include	EBS, WF	ANL-WIS-MD-000009 ANL-WIS-MD-000007	Included: Thermally induced stress changes in the CSNF cladding influences creep rupture and SCC perforations in the CSNF Cladding Degradation Component. Furthermore, temperature differences on different fuel assemblies in the cross-section of the WP are considered in the first 1000 yr. Degradation of the waste matrix of CSNF and HLW is also a function of temperature (CSNF matrix FEP 2.1.11.05.00 and HLW FEP 2.1.11.01.00).

2.3.2.5 FEPs Related to the DSNF Degradation Component

Three of the FEPs related to CSNF Degradation Component (Section 2.3.2.3) also apply here (radiolysis, alpha recoil, and radiation damage). These three radiation effects are included by conservatively bounding the degradation of rate of experiments on irradiated N-Reactor fuel. Six additional FEPs specifically related to the DSNF Degradation Component are listed here (Table 2.3-5). Four of these FEPs relate to the potentially increased reactivity of some types of the DSNF. However, the additional reactivity is of low consequence and so these four FEPs are screened out. One FEP relates to the degradation of cladding for DSNF, but is conservatively excluded by not including cladding performance for DSNF except for the naval SNF, whose analysis will account for the performance of the cladding.

Table 2.3-5. FEPs Related to the DSNF Degradation Component

FEP Number	FEP Title	Include/ Exclude	Process Model Report (PMR)	AMR Document Identifier (DI)	Reason for Include/Exclude Decision
2.1.02.01.00	DSNF degradation, alteration, and dissolution	Include/ Exclude	WF	ANL-WIS-MD-000009 ANL-WIS-MD-000004	Included: DSNF and its degradation are included in TSPA-SR. The largest component of the DSNF is the metallic N-Reactor fuel. A constant rate that conservatively bounds the temperature varying rate of metallic uranium is used since the corrosion characteristics of uranium metal, in turn, conservatively bound the degradation of all other DSNF. (The inventory, however, is the weighted mass average of all the ~250 types of DSNF [waste inventory FEP 2.1.01.01.00].) Excluded based on low consequence: Time dependence of available surface area for degradation or release of radioisotopes (see DSNF cladding FEP 2.1.02.25.00).
2.1.02.08.00	Pyrophoricity	Exclude	WF	ANL-WIS-MD-000009	Excluded based on low consequence. The performance on the DSNF waste form itself is not affected since (except for naval SNF) no credit for cladding is assumed and the degradation rate of the matrix is already conservatively bounded.
2.1.02.08.04	Flammable gas generation from DSNF	Exclude	WF	ANL-WIS-MD-000009	Excluded based on low consequence. The chemical reaction of U/Th carbide with water could produce CO_2 and hydrocarbon gases. The major constituent of the gas is methane; minor constituents are ethane, ethylene, and acetylene. Only DSNF from the Peach-Bottom Core 1 has uncoated U/Th carbide pellets and thus the potential to generate flammable gases over a short period of time. However, the Peach Bottom core 1 is in only ~100 WPs out of ~3900 DSNF WPs and of ~11,800 WPs. Also, relatively good connection with the surface through fractures and fault zones would eventually disperse any flammable gas in the repository before explosive concentrations could be obtained (see in-package chemistry gas generation FEP 2.1.12.01.00).
2.1.02.25.00	DSNF cladding degradation	Exclude	WF	ANL-WIS-MD-000009	Excluded based on low consequence (conservative bound): No credit is taken for DSNF cladding (with the exception of naval SNF cladding). More than 80% of the DSNF is from the N-Reactor and is in poor condition; up to 50% may already be perforated; hence, all the cladding is conservatively assumed to be perforated. The cladding is conservatively neglected since the bounding alteration rate of the DSNF fuel matrix is so high and the radioisotope inventory is so small. The rapid degradation of DSNF fuel influences the in-package chemistry pH in the first 1000 yr (in-package chemistry codisposal FEP 2.1.01.02.00).

Table 2.3-5. FEPs Related to the DSNF Degradation Component (Continued)

FEP Number	FEP Title	Include/ Exclude	Process Model Report (PMR)	AMR Document Identifier (DI)	Reason for Include/Exclude Decision
2.1.11.03.00	Exothermic reactions and other thermal effects in waste and EBS	Exclude	EBS, WF	ANL-WIS-MD-000009	Excluded based on low consequence. The possible temperature rise in a disposal container from exothermic degradation of DSNF is inconsequential in comparison to heat generated by radioactive decay. The fixed, conservative rate bounds by at least an order of magnitude the maximum degradation rate observed and thus also bounds any thermal effects on waste degradation.
2.1.12.08.00	Gas explosions	Exclude	EBS, WF	ANL-WIS-MD-000009	Excluded based on low probability. Because of the absence of O_2 and water, there is a very low probability of explosion within the canister until the WP and DSNF canister is breached. After the canister is breached, generation of flammable gases has been excluded (see DSNF FEP 2.1.02.08.04). Furthermore, any gas produced will disperse into the drift. The permeability of Yucca Mountain provides adequate connection to the surfaces such that flammable/explosive gases would be diluted and/or dispersed before they could reach explosive concentrations (see in-package chemistry gas generation FEP 2.1.12.01.00)

2.3.2.6 FEPs Related to the HLW Degradation Component

Four FEPs specifically relate to the HLW Degradation Component (Table 2.3-6). One primary FEP and its underlying secondary FEPs deal with the enhanced degradation that may occur if extensive phase separation or glass recrystallization occur in the encapsulating glass; however, production controls will prevent significant initial phase separation. Furthermore, temperatures in the repository are low relative to the melting point of the glass such that extensive recrystallization will not occur in the glass. The HLW Degradation Component explicitly includes the effect of temperature on HLW glass degradation. Another FEP deals with the proper reactive surface area to use when evaluating the degradation rate of the glass. The HLW Degradation Component bound the reactive surface area by multiplying the geometric surface area by a very conservative, but constant cracking factor of 20.

Table 2.3-6. FEPs Related to the HLW Degradation Component

FEP Number	FEP Title	Include/ Exclude	Process Model Report (PMR)	AMR Document Identifier (DI)	Reason for Include/Exclude Decision
2.1.02.03.00	Glass degradation, alteration, and dissolution	Include/ Exclude	WF	ANL-WIS-MD-000009	Included: HLW degradation is included in the TSPA-SR analysis as a source term for the congruent mobilization of contaminants. Degradation rate is dependent on pH of in-package water chemistry. The effects of minor phase separation are subsumed in the degradation model since parameters for the model conservatively bound dissolution experiments on samples where minor phase separation might have occurred. Excluded based on low probability (credibility): Extensive phase separation (precipitation/coprecipitates/solid solution phase) is excluded because phase separation is controlled during production. Excluded based on low consequence: Extensive selective leaching is excluded since the rate of degradation used for the glass matrix is an upper bound on the actual rate of release and, therefore, bounds any additional consequence due to selective leaching.
2.1.02.05.00	Glass cracking and surface area	Include	WF	ANL-WIS-MD-000009 ANL-EBS-MD-000016	The thermal stress during cooling increases the surface area accessible to water through cracking beyond the geometric surface area; a robust container and cool temperatures, relative to the transition temperature of glass, prevent extensive increases in this initial surface area during disposal. Nonetheless, a very conservative cracking surface area-enhancement factor of 20 is used to set the surface area accessible to water for degradation.
2.1.02.06.00	Glass recrystalli-zation	Exclude	WF	ANL-WIS-MD-000009	Extensive recrystallization excluded based on low probability (credibility). Controls are to be implemented as part of the waste production to ensure that extensive glass recrystallization and phase separation will not occur.
2.1.11.01.00	Heat output / temperature in waste and EBS	Include	WF, EBS, NFE	ANL-WIS-MD-000009	Included: Decay heat is a major issue in repository design, where high loading densities and high temperatures (~96°C) are intended to be part of the waste isolation scheme. Degradation of HLW is a function of temperature. See also CSNF FEP 2.1.11.05.00 and Cladding FEP 2.1.11.07.00.

2.3.2.7 FEPs Related to the Dissolved Radioisotope Concentration Component

Many processes influence the dissolved concentration of radionuclides in the Dissolved Radioisotope Concentration Component. However, most of those processes have been discussed separately in the In-Package Chemistry Component since these processes also affect other model components. Here four FEPs are listed that have a more direct connection with the dissolved radionuclide concentration (Table 2.3-7). First, solubility control based on pure phases has been and continues to be included in the TSPA-SR analysis. Although a few radionuclides such as Np may be incorporated into the structure of phases of other minerals (primarily uranium) that form during degradation of the waste such that the solubility of these other minerals control the concentration of the radioisotope, these effects have been conservatively excluded. Complexation of radionuclides by organic ligands has been excluded because organic material is generally prohibited from the repository and the incidental amounts of organic material that may inadvertently occur cannot alter the solubility of significant amounts of radionuclides.

Table 2.3-7. FEPs Related to the Dissolved Radioisotope Concentration Component

FEP Number	FEP Title	Include/ Exclude	Process Model Report (PMR)	AMR Document Identifier (DI)	Reason for Include/Exclude Decision
2.1.09.04.00	Radionuclide solubility, solubility limits, and speciation in the waste form and EBS	Include	WF	ANL-WIS-MD-000009	A controlling phase is used to develop a dissolved concentration limit for each of the transported radioactive elements. These limits are expressed as an empirical equation, distribution, or bounding constant, depending on the element and available data. The concentration limit is used to bound the amount of a radionuclide that can be mobilized by the Dissolved Radioisotope Concentration Component.
2.1.09.10.00	Secondary phase effects on dissolved radionuclide concentrations at the waste form	Exclude	WF	ANL-WIS-MD-000009 ANL-EBS-MD-000019	Excluded based on beneficial consequences (conservative bound). The solubility of many radionuclides depends only on pure phases; however, a few radionuclides such as Np may be incorporated into other minerals (primarily uranium minerals) that form during degradation of the waste. These other minerals control the concentration of the radioisotope. These effects have been conservatively excluded; hence solubility of Np is somewhat greater than in TSPA-VA.
2.1.09.12.00	Rind (altered zone) formation in waste, EBS, and adjacent rock	Include/ Exclude	EBS, NFE WF	ANL-WIS-MD-000009	Included: For determining the radioisotope concentration, the Dissolved Radioisotope Concentration Component assumes that the volume of water available for dissolution in the waste form cell is equal to the pore space of the rind of alteration products that forms as the UO_2 is converted into secondary minerals. Excluded (conservative bound) based on beneficial consequences: Rind or altered zone may tend to prevent advective flow past the waste and thus reduce release of radionuclides. This effect has been conservatively excluded.

Table 2.3-7. FEPs Related to the Dissolved Radioisotope Concentration Component (Continued)

FEP Number	FEP Title	Include/ Exclude	Process Model Report (PMR)	AMR Document Identifier (DI)	Reason for Include/Exclude Decision
2.1.09.13.00	Complexation by organics in waste and EBS	Exclude	EBS WF	ANL-WIS-MD-000009	Excluded based on low probability (credibility). Based on the preliminary waste acceptance criteria (DOE 1999c, Section 4.2.3.A.3), organic material will be excluded from the radioactive waste. Hence, sources of organic material will not be present in waste; only small amounts could occur and even then it will be isolated incidents. For example, organic complexes have not been associated with Yucca Mountain waters, now or in the past. Furthermore, drift temperatures are sufficient to drive off many volatile organics, should they occur.

2.3.2.8 FEPs Related to the Colloidal Radioisotope Concentration Component

Sixteen FEPs relate to the Colloidal Radioisotope Concentration Component (Table 2.3-8). Three types of colloids were included: those already present in the groundwater, those formed during corrosion of the WP, and those formed during degradation of the waste. Condensed Pu polymer colloids were excluded since they have not been observed in fuel or glass dissolution tests. Biological activity was screened out, and so microbial colloids were also excluded. Transport and filtration of colloids inside the WP were conservatively excluded.

Table 2.3-8. FEPs Related to the Colloidal Radioisotope Concentration Component

FEP Number	FEP Title	Include/ Exclude	Process Model Report (PMR)	AMR Document Identifier (DI)	Reason for Include/Exclude Decision
2.1.09.05.00	In Package Sorption	Include/ Exclude	WF	ANL-WIS-MD-000009 ANL-WIS-MD-000012	Included: Sorption on mobile colloidal material is included. Excluded based on low consequence (conservative bound): Sorption on stationary material inside the waste package is excluded. This is a conservative exclusion since sorption decreases the release rate of radioisotopes.
2.1.09.14.00	Colloid formation in waste and EBS	Include	WF, EBS	ANL-WIS-MD-000012	A colloid source term has been developed for the Waste Form Degradation Model of the TSPA-SR analysis using experimental data produced from YMP investigations.
2.1.09.15.00	Formation of true (real) colloids in waste and EBS	Exclude	WF, EBS	ANL-WIS-MD-000012	Excluded based on low consequence: true colloids would be expected to dissolve when the solution is diluted.
2.1.09.16.00	Formation of pseudo-colloids (natural) in waste and EBS	Include	WF, EBS	ANL-WIS-MD-000012	The Colloidal Radioisotope Concentration Component assumes pseudo-colloids may form from groundwater colloids and can reversibly sorb Am, Pu, and other radionuclides.

Table 2.3-8.FEPs Related to the Colloidal Radioisotope Concentration Component (Continued)

FEP Number	FEP Title	Include/ Exclude	Process Model Report (PMR)	AMR Document Identifier (DI)	Reason for Include/Exclude Decision
2.1.09.16.01	Colloidal phases produced by coprecipitation in the waste package or EBS.	Include	WF	ANL-WIS-MD-000012	The Colloidal Radioisotope Concentration Component assumes colloids produced from degradation of HLW glass incorporate Am and Pu radioisotopes irreversibly. The assumption of irreversibility is a bounding approximation.
2.1.09.17.00	Formation of pseudo-colloids (corrosion products) in waste and EBS	Include	WF, EBS	ANL-WIS-MD-000012	The Colloidal Radioisotope Concentration Component assumes colloids are produced from corrosion of the disposal container and other material inside the container and reversibly sorb Am and Pu and other radionuclides. These colloids are conservatively assumed to be similar to natural colloids (see colloid FEP 2.1.09.16.00).
2.1.09.18.00	Microbial colloid formation in the waste and transport in EBS.	Exclude	WF, EBS	ANL-WIS-MD-000012	Excluded based on low consequence (conservative bound). Microbial activity has been screened out as a FEP as concerns the waste form (colloid FEP 2.1.10.01.00). However, even if microbial activity were present, it would tend to increase colloid size over time, which would result in increased gravitational settling and filtration.
2.1.09.19.00	Colloid sorption and transport in the waste and EBS	Exclude	WF, EBS	ANL-WIS-MD-000012	Exclude based on low consequence (conservative bound). The Colloidal Radioisotope Concentration Component assumes colloids reversibly sorb or irreversibly incorporate radionuclides. However, the modeling component only acts as a source-term and conservatively neglects the transport of these colloids inside the WP. Filtration is also excluded (see colloid FEP 2.1.09.20.00).
2.1.09.20.00	Colloid filtration	Exclude	WF, EBS	ANL-WIS-MD-000012	Exclude based on low consequence (conservative bound). The conservative assumption is made that all colloids produced within the WP (the calculated colloid source term) exit the WP and enter the drift/EBS. Colloid transport time is also excluded (see colloid FEP 2.1.09.19.00).
2.1.09.21.00	Suspensions of particles larger than colloids	Exclude	EBS, SZ	ANL-WIS-MD-000012	Excluded based on low consequence. It is unlikely that larger-than-colloid particles will have access to a sufficient number of vertical or subvertical fractures whose apertures permit their passage in significant quantities. The relatively small quantity of particles that may make it through the UZ would encounter low groundwater velocities in the SZ, which would likely result in the particles settling and becoming immobile.
2.1.09.22.00	Colloidal sorption at the groundwater interface	Exclude	WF	ANL-WIS-MD-000012	Excluded based on low consequence (conservative bound). Although colloids may be retained at the air-water interface in unsaturated conditions, neglecting this phenomena is conservative; thus, the conceptual model of waste package uses a fully saturated mixing cell.

Table 2.3-8.FEPs Related to the Colloidal Radioisotope Concentration Component (Continued)

FEP Number	FEP Title	Include/ Exclude	Process Model Report (PMR)	AMR Document Identifier (DI)	Reason for Include/Exclude Decision
2.1.09.23.00	Colloidal Stability and concentration dependence on aqueous chemistry	Include	WF	ANL-WIS-MD-000012	The Colloidal Radioisotope Concentration Component develops (based on experimental data) stability curves as a function of pH and ionic strength.
2.1.09.24.00	Colloidal diffusion	Include	WF	ANL-WIS-MD-000012	The Colloidal Radioisotope Concentration Component uses a diffusion coefficient of 10^{-2} i.e., the diffusion coefficient of colloids is a factor of 100 less than the diffusion coefficient of dissolved radionuclides.
2.1.09.25.00	Colloidal gravitational settling	Exclude	WF	ANL-WIS-MD-000012	Excluded based on low consequence (conservative bound). The conservative assumption is made that gravitational settling of radionuclide-bearing colloids does not occur, but instead all of the colloids (if stable) leave a breached waste package and can reach the accessible environment. Most likely, some percent of the colloids will settle out during the transport process and reduce the amount of radionuclides reaching the accessible environment.
2.1.10.01.00	Biological activity in waste and EBS	Exclude	WF, EBS, WP	ANL-WIS-MD-000012	Excluded based on low probability (credibility) and low consequence. Since organic matter is excluded from the waste based on the preliminary waste acceptance criteria (DOE 1999c, Section 4.2.3.A.3), preliminary analysis shows that sufficient quantities of microbes will not be available to promote colloid mobility or adversely accelerate corrosion rates significantly.
2.1.13.03.00	Mutation	Exclude	EBS	ANL-WIS-MD-000009	Excluded based on low consequence. General principles of population genetics indicate that most mutations are either neutral or deleterious to the fitness of an organism and, in the absence of strong natural selection, are unlikely to produce any definite change in the phenotypes of the organisms.

2.3.3 Relationship of Model Components, FEPs, and Factors

The model components listed in Section 2.2 are closely aligned with the principal and other factors that affect repository performance. Table 2.3-9 shows the correspondence. Note that FEPs 2.1.02.09.00 and 2.1.09.02.00 are related to two different factors.

Table 2.3-9. Relationship of Model Components to Factors

Model Component	Factor
Radioisotope Inventory (all FEPs)	(none)
In-Package Chemistry (FEPs 2.1.01.02.00, 2.1.02.09.00, 2.1.02.10.00, 2.1.03.06.00, 2.1.08.10.00, 2.1.09.01.00, 2.1.09.02.00, 2.1.09.06.00, 2.1.09.07.00, 2.1.09.11.00, 2.1.11.04.00, 2.1.11.08.00, 2.1.12.01.00, 2.1.12.02.00, 2.1.12.03.00, 2.1.12.04.00, 2.2.08.12.00)	Environments within the waste package
In-Package Chemistry (FEPs 2.1.01.04.00, 2.1.02.09.00, 2.1.08.07.00, 2.1.08.08.00, 2.1.09.02.00, 2.1.09.08.00, 2.1.09.09.00, 2.1.11.09.00, 2.1.11.10.00, 2.1.12.06.00)	In-package radionuclide transport
CSNF Matrix Degradation (all FEPs)	CSNF waste form performance
CSNF Cladding Degradation (all FEPs)	CSNF waste form performance
DSNF Degradation (all FEPs)	DSNF, naval SNF[a], Pu-disposition waste form performance
HLW Degradation (all FEPs)	HLW glass waste form performance
Dissolved Radioisotope (all FEPs)	Solubility limits of dissolved radionuclides
Colloidal Radioisotope Concentration (all FEPs)	Colloid-associated radionuclide concentrations

NOTE: [a]For purposes of performance assessment, Naval SNF will be represented by CSNF.

2.4 INFLUENCE OF DESIGN CHANGES ON WASTE FORM DEGRADATION MODEL

The design of the repository continues to evolve in preparation for the future license application. In general, these design changes can directly influence the screening arguments for FEPs and their inclusion in or exclusion from the waste form degradation model. The elimination of backfill is an important design change that has been made after the development of the current Waste Form Degradation Model. The primary effect of the elimination of backfill is the decrease of peak temperatures inside the WP, which is beneficial. For example, the temperature decrease (1) reduces the chance for creep rupture and stress corrosion cracking of CSNF cladding; (2) reduces the degradation rates of the CSNF and HLW matrices; (3) improves the applicability of the current data for in-package chemistry; and (4) decreases the solubility of uranium. However, design change to eliminate backfill does not affect the Waste Form Degradation Model because temperature is explicitly included as a model variable. That is, the model is an explicit function of the surface temperature of the waste package (Figure 1.5-1), so any changes in the surface temperature of the waste package because of the design change are automatically included.

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3. MODELS AND ABSTRACTIONS

3.1 RADIONUCLIDE INVENTORY COMPONENT

The function of the radioisotope inventory abstraction component is to estimate the inventory of those radionuclides most important to human dose. The inventory abstraction component is input for the waste form degradation models and is developed from a series of steps that starts with radioisotope inventories of various spent nuclear fuel assemblies and HLW then estimates the radioisotope inventory when packaged in disposal containers. Three important aspects of the radionuclide inventory are (1) selecting the most important radionuclides for human dose out of the few hundred found within the waste, (2) obtaining the radioisotope inventories of various wastes, and (3) grouping the fuels into the waste packages selected for modeling in the TSPA-SR analysis.

Using radionuclide activities for CSNF assemblies, DSNF canisters, and HLW canisters, the radionuclide inventory component provides an estimate for activities in containers destined for disposal in the potential Yucca Mountain repository as shown in Figure 3.1-1.

The inventory abstraction is described in the *Inventory Abstraction* AMR (CRWMS M&O 2000f) and the eleven supporting calculations (Figure 3.1-2). Three sources were used for inventory data: the commercial utilities for CSNF (CRWMS M&O 1999d), the DOE NSNFP for DSNF (DOE 1999b), and the Yucca Mountain EIS program for the HLW, mixed oxide fuel (MOX), and plutonium ceramic (DOE 1999a). From this radioisotope inventory, the most important radionuclides for human dose were evaluated. Arrival scenarios were developed for CSNF, and the inventory was assigned to WP configurations. Average and bounding inventories were developed for each package configuration recommended for the repository. Then, the package-specific radionuclide activities were combined using the number of WPs in each group as a weighting factor to get the radionuclide activities in each allocation category.

3.1.1 Description

3.1.1.1 Radionuclides Important for Total System Performance Assessment

The relative importance of individual radionuclides for human inhalation and ingestion doses was evaluated for several waste types, time frames, and release scenarios. In this evaluation, the effects of inventory abundance, radionuclide longevity, element solubility, and element transport affinity were considered. To address inventory abundance, eight waste types were examined.[9] To address radionuclide longevity, the fuels were evaluated between 100 and 1 million years after repository closure. The elements were evaluated in two solubility groups, the relatively soluble and the relatively insoluble (Am, Cm, Zr, Th, Nb, Pa, and Sn) and three transport affinity groups: (1) highly sorbing, (2) moderately sorbing, and (3) slightly to non-sorbing. The isotopes within each group were compared to one another for relative importance. Three release scenarios were considered: nominal case, human intrusion, and direct volcanic release. Two time frames were considered: 100 to 10,000 years and 100 to 1 million years.

[9] Average and bounding types for each of PWR, BWR, DSNF, and HLW

The set of important isotopes was different for each scenario and time frame. For the first 10,000 years, ^{99}Tc and ^{129}I are normally the primary contributors to dose (DOE 1998c, Figure 4-29). For a direct release from a disruptive event scenario out to 10,000 years, ^{90}Sr, ^{137}Cs, ^{227}Ac, ^{229}Th, ^{231}Pa, ^{232}U, ^{233}U, ^{234}U, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Am, ^{243}Am were screened in. These are the isotopes that contribute most to the dose when release is not mitigated by either solubility or transport. For a nominal release and human intrusion scenario out to 10,000 years ^{14}C, ^{99}Tc, ^{129}I, ^{227}Ac, ^{229}Th, ^{232}U, ^{233}U, ^{234}U, ^{236}U, ^{238}U, ^{237}Np, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Am, ^{243}Am were screened in. By modeling the plutonium isotopes (^{238}Pu, ^{239}Pu, ^{240}Pu), the americium isotopes (^{241}Am, ^{243}Am), ^{229}Th, and ^{227}Ac, doses that could result from colloidal transport of radioactive material to the biosphere will be adequately represented. By modeling ^{14}C, ^{99}Tc, ^{129}I, the uranium isotopes (^{232}U, ^{233}U, ^{234}U, ^{236}U, ^{238}U), and ^{237}Np, doses that could result from transport of solutes, either by fracture flow or matrix diffusion, will be adequately represented. In addition, ^{63}Ni, ^{90}Sr and ^{137}Cs are screened in for a human intrusion scenario because the event could occur as early as 100 years after repository closure. For the time period from 10,000 to 1 million years, ^{210}Pb, ^{226}Ra, ^{230}Th, and ^{242}Pu are screened in for all of the scenarios, and ^{231}Pa is screened in for the human intrusion and nominal release scenarios. The result is 24 isotopes recommended for modeling in the TSPA-SR analysis and TSPA-FEIS. The 24 isotopes are shown in Table 3.1-1, along with the isotope selection from previous performance assessments.

For TSPA 1993 (Wilson et al. 1994) and 1995 (CRWMS M&O 1995), all radionuclides contributing to 99.99 percent of the potential dose at any time from 1,000 to 1 million years were retained for the inventories. For TSPA-SR analysis, all radionuclides contributing to 95 percent of the potential dose at any time from 100 to 1 million years were retained for the inventories. Thus, radionuclides that contribute to the last five percent of the dose estimate were eliminated from consideration in the TSPA-SR analysis.

For the TSPA-VA (CRWMS M&O 1998b), a smaller number of representative radionuclides were chosen. A radionuclide was included in the TSPA-VA (DOE 1998b) if it had: (1) a high solubility, (2) a low sorption affinity, (3) a significant inventory,[10] (4) a high dose conversion factor, and (5) a long half-life. In addition, some radionuclides were included based on previous experience (1993 TSPA calculations [Wilson et al. 1994], 1995 TSPA calculations [CRWMS M&O 1995], and scoping calculations) and the experience of other organizations (NRC performance assessments [Wescott et al. 1995]).

[10] Including the inventory generated by decay of a parent radionuclide

Table 3.1-1. Isotope Selection

Isotope	TSPA-SR[a] & Final EIS 2000	TSPA 1993[b]	TSPA 1995[c]	TSPA-VA 1998[d]	NRC Iterative Performance Assessment 1995[e]
^{227}Ac	X	X	X		
^{108m}Ag		X			
^{241}Am	X	X	X		X
^{242m}Am		X	X		
^{243}Am	X	X	X		X
^{14}C	X	X	X	X	X
^{36}Cl		X	X		
^{243}Cm		X			
^{244}Cm		X	X		
^{245}Cm		X	X		X
^{246}Cm		X	X		X
^{135}Cs		X	X		X
^{137}Cs	X	X			X
^{129}I	X	X	X	X	X
^{93}Mo		X			
^{93m}Nb			X		
^{94}Nb		X	X		X
^{59}Ni		X	X		X
^{63}Ni	X	X	X		
^{237}Np	X	X	X	X	X
^{231}Pa	X	X	X	X	
^{210}Pb	X	X	X		X
^{107}Pd		X	X		
^{238}Pu	X	X	X		
^{239}Pu	X	X	X	X	X
^{240}Pu	X	X	X		X
^{241}Pu		X	X		
^{242}Pu	X	X	X	X	
^{226}Ra	X	X	X		X
^{228}Ra			X		
^{79}Se		X	X	X	X
^{151}Sm		X	X		
^{121m}Sn		X			
^{126}Sn		X	X		
^{90}Sr	X	X			
^{99}Tc	X	X	X	X	X
^{229}Th	X	X	X		
^{230}Th	X	X	X		X
^{232}Th			X		
^{232}U	X	X			
^{233}U	X	X	X		
^{234}U	X	X	X	X	X
^{235}U		X	X		
^{236}U	X	X	X		
^{238}U	X	X	X		X
^{93}Zr		X	X		

Sources: [a] CRWMS M&O 2000f, Section 7.1
[b] Wilson et al. 1994
[c] CRWMS M&O 1995
[d] DOE 1998b
[e] Wescott et al. 1995

For the Iterative Performance Assessment (IPA) (Wescott et al. 1995), the NRC screened radionuclides to include only the major contributors to cumulative release and dose. A radionuclide was retained in the inventory if, in preliminary calculations, it contributed more than one percent of the EPA cumulative release limit for that radionuclide.[11] The screening analysis also checked the maximum dose to a farm family to see if any of the radionuclides that might have been screened out on the basis of cumulative release should have been kept on the basis of dose.

The differences between the isotope selection in these PAs are primarily due to: (1) the changing regulations (dose - cumulative release - groundwater protection; 10,000 years - 1 million years, groundwater - human intrusion - volcanic releases), (2) inventory data relied upon, and (3) the screening techniques.

3.1.1.2 Commercial Spent Nuclear Fuel Inventory

Commercial nuclear power plants use a variety of fuels and fuel configurations in their reactor cores to generate power. The predominant fuel is enriched uranium dioxide, but a plutonium/uranium mix is also planned for use in commercial reactors to dispose of excess plutonium from the government inventory. Fuel pellets are packed into fuel rods (which vary in size depending on the application), and fuel rods (clad in Zircaloy or stainless steel) are bundled into assemblies. The number of fuel rods per assembly and the number of assemblies in a reactor core vary depending on the reactor design.

Once a reactor fuel has been irradiated to the extent that it can no longer effectively sustain a chain reaction, it is removed from the reactor and becomes spent nuclear fuel. Its isotopic composition at that point depends on the initial enrichment of the fuel, the reactor configuration (pellet size, fuel rod size, assembly layout, and other parameters), and the extent of irradiation (called burnup). Once removed from the core, the fuel is placed in storage and it ages.

Almost 230,000 CSNF assemblies will need to be disposed, and each assembly, depending on the reactor configuration, initial fuel enrichment, burnup, and the age of the waste (time in storage), will have a unique isotopic composition. The radionuclide inventory abstraction collects this information and synthesizes it into a form that can be modeled in TSPA-SR analysis.

A 1995 data submittal from the commercial utilities provided the basic information from which the TSPA-SR analysis inventory for CSNF was developed. In 1995, the utilities supplied historical information about reactor assembly discharges up through December 1995, and they provided five-cycle forecasts for assembly discharges from their reactors. With this information, a design basis waste stream was developed (CRWMS M&O 1999d), and forecasts for assembly discharges over the lifetime of each commercial power reactor were developed.

For the base case repository design of 70,000 MTHM, three alternative schedules were developed for moving the CSNF assemblies out of storage and shipping the assemblies to Yucca Mountain. The schedules, called arrival scenarios, include the year of receipt for each shipment, the number of assemblies in each shipment, the type of fuel in each shipment (pressurized water

[11] From 40 CFR 191 (now obsolete)

reactor (PWR) versus boiling water reactor (BWR), and the enrichment/burnup characteristics of the fuel.

Radionuclide activities for each assembly in the waste stream were estimated, and the WP configuration that could accommodate the assembly based on its potential criticality level was determined. The result was a grouping of the 230,000 CSNF assemblies into five proposed WP configurations. For each group, average and bounding assembly radionuclide activities were calculated.

3.1.1.3 U.S. Department of Energy Spent Nuclear Fuel and High-Level Waste

For the most part, DSNF (with the exception of the naval SNF) and HLW are planned to be disposed together in codisposal waste packages. Therefore, they are discussed together here.

The DSNF consists of more than 250 distinct types of spent fuel, and much like CSNF, radionuclide inventories for defense fuels will vary widely depending on the history of the fuel. The NSNFP grouped the fuels into 11 groups (DOE 1999b). DOE fuels will be packaged in three types of canisters before they are shipped to Yucca Mountain; short, long, and multi-container overpack (MCO). Similarly, the naval fuels will be packaged in two types of canisters: naval long and naval short (Dirkmaat 1997, Appendix F, Attachment, p. 1). For analysis, the fuels are grouped by canister type and average and bounding per canister radionuclide inventories were calculated.

The HLW in storage at DOE sites is the result of reprocessing SNFs (some CSNF and some DSNF). The proposed technology for immobilization of HLW is vitrification in a borosilicate glass. The vitrified waste will be placed in one of two canister types (long and short). A small amount of HLW glass has been produced at the West Valley Demonstration Project (WVDP) in New York. Production of HLW has started at the Savannah River Site (SRS) and HLW will eventually be produced and stored at two other sites—the Hanford Reservation (HR) and the Idaho National Engineering and Environmental Laboratory (INEEL). Because the fuels reprocessed at each of these sites differ, the radionuclide inventory of the HLW and resultant glass product will vary slightly among the sites.

The data source for the HLW source terms, which include radionuclide inventory, decay heat, and radiation sources due to gamma rays and neutrons, is the response to the EIS data call as described in DOE (1999a). The sites included in the data call are the HR, SRS, INEEL, and WVDP. HLW fuels will be packaged in three types of canisters before they are shipped to Yucca Mountain: short, long, and Pu disposition. The information from the data call was used to calculate average and bounding per canister inventories.

The average radionuclide activity from DSNF for one of these WPs is the number of canisters times the average per canister radionuclide activity calculated. The average radionuclide activity from HLW for one of these WPs is the number of canisters times the average per canister radionuclide activity calculated. The radionuclide activity for the DSNF allocation group is an average, weighted by the number of packages in each group, over the six WP configurations. Similarly, the radionuclide activity for the HLW allocation category is an average, weighted by the number of packages in each group.

The naval SNF, by agreement, is conservatively treated separately using average CSNF inventory and degradation characteristics.

3.1.1.4 Radionuclide Masses in Allocation Categories for Total System Performance Assessment for Site Recommendation

The waste types, allocations, and waste packages for commercial spent nuclear fuel, DOE spent nuclear fuel, high-level radioactive waste, and plutonium disposition waste are shown in Figure 3.1-1. In the TSPA model, over 250 types of DOE spent nuclear fuels and HLWs are represented as being packaged in the ten types of canisters listed in Table 3.1-2. These canisters and the CSNF assemblies are, in turn, represented as being emplaced in ten types of waste packages, which are also listed in Table 3.1-3. The waste packages and canisters combine to give a total of thirteen waste package configurations. The waste package configurations and the number of packages with each configuration for TSPA modeling purposes are listed in Table 3.1-3.

Table 3.1-2. Waste Package and Canister Designs

WP Design	WP Design Code
21 PWR absorber plate	W1
21 PWR control rod	W2
12 PWR	W3
44 BWR	W4
24 BWR	W5
Codisposal short	W6
Codisposal long	W7
2-MCO/2-HLW	W8
Naval short	W9
Naval long	W10

Canister Design	Canister Design Code
Naval short	C1
Naval long	C2
DSNF short	C3
DSNF long	C4
DSNF short, wide	C5
DSNF long, wide	C6
HLW short	C7
HLW long	C8
HLW short with Pu-ceramic	C9
MCO	C10

Source: CRWMS M&O 2000ao, Section 4

Table 3.1-3a. Waste Configurations Used in the Inventory Abstraction

Configuration	WP Design Code	Number	DSNF Canisters per Waste Package	HLW Canisters per Waste Package
21 PWR absorber plate	W1	4500[a]	---	---
21 PWR control rod	W2	100[a]	---	---
12 PWR	W3	170[a]	---	---
44 BWR	W4	3000[a]	---	---
24 BWR	W5	90[a]	---	---
Total CSNF		**7860**		

Table 3.1-3a. Waste Configurations Used in the Inventory Abstraction (Continued)

Configuration	WP Design Code	Number	DSNF Canisters per Waste Package	HLW Canisters per Waste Package
Pu-ceramic in HLW	W6	100[a]	---	Five C9 (short)
Codisposal short	W6	1100[a]	One C3 or C5 (short)	Five C7 (short)
Codisposal long	W7	1500[a]	One C4 or C6 (long)	Five C8 (long)
Codisposal mixed	W7	130[a]	One C3 or C5 (short)	Five C8 (long)
HLW-only	W7	600[a]	---	Five C8 (long)
2-MCO/2-HLW	W8	160[a]	Two C10	Two C8 (long)
Total Codisposal		**3590**		
Naval short	W9	200[b]	One C1 (short)	---
Naval long	W10	100[b]	One C2 (long)	---
Total Naval		**300**		

Sources: [a]Stroupe 2000, Att. 1
[b]Dirkmaat 1997, Appendix F, Attachment, p. 6

These waste package numbers were specified by management edict (Stroupe 2000), and are somewhat different from the "Proposed Action" numbers used in the DEIS (DOE 1999a) or the "truncated case" in recent input transmittals (CRWMS M&O 2000aj and 2000ae). The management specified numbers are "not to exceed" numbers and are rounded up from the truncated case, in order to convey flexibility in the design. The truncated case has more HLW canisters than the DEIS, in order to use the "ideal waste emplacement scenario" for codisposal. Neither the management case nor the truncated case may be emplaced without a redefinition of "HLW MTHM equivalency" or the elimination of the 70,000 MTHM limit specified in the Nuclear Waste Policy Act of 1982. Using the historical definition of 0.5 MTHM per HLW canister, these cases contain more than 4,667 MTHM HLW equivalent. Table 3.1-3b compares some of the canister and assembly numbers for the various scenarios.

Table 3.1-3b Canister and Assembly Numbers for the Various Scenarios

Total	Management Case[a]	Truncated Case[b]	Full Inventory Case[b]	DEIS Proposed Action[c]	DEIS Module 1[c]
HLW canisters (including Pu)	17,470	16,613	22,153	8,315[d]	22,300[d]
DSNF canisters (excluding naval SNF)	3,050	2,882	3,842	3,470[e]	3,721[f]
CSNF assemblies	232,800	220,810	291,792	218,700[g]	359,963[h]

NOTES: [a]Attachment 1 of Stroupe 2000 as used in TSPA-SR
[b]from waste package counts in CRWMS M&O 2000aj and 2000ae
[c]DOE 1999a
[d]Table J-8 of DOE 1999a
[e]Table J-7 of DOE 1999a
[f]Table A-21 of DOE 1999a
[g]Table A-6 of DOE 1999a
[h]Table A-7 of DOE 1999a

There will be 7860 CSNF packages and 3590 codisposal packages in the TSPA-SR analysis model. The 300 naval packages were grouped with the codisposal packages in the first version of CRWMS M&O 2000f, but were regrouped with CSNF packages in the subsequent input transmittal (CRWMS M&O 2000ak). Naval spent fuel is expected to perform very well within the repository, and it is better represented by commercial packages than by codisposal packages in the TSPA. Accordingly, the naval fuel inventory was removed from the high level waste and DOE spent nuclear fuel averages, and the averages were recalculated for the input transmittal (CRWMS M&O 2000ak). The impact analysis of this change is expected to show that the old values were conservative, and the total TSPA values are not significantly affected. The radionuclide inventories in grams for the selected isotopes are shown in Table 3.1-4.

Three radionuclides appear in Table 3.1-4 that were not listed in Table 3.1-1. ^{228}Ra, ^{232}Th, and ^{235}U were not identified as important contributors to dose for the direct release scenario, the human intrusion scenario, or the nominal release scenario. However, ^{228}Ra and ^{232}Th will be modeled in TSPA for the groundwater protection scenario and therefore were included in the inventory abstraction. ^{235}U will be modeled in the TSPA because it is a parent to ^{231}Pa. To get accurate estimates of the dose from ^{231}Pa, TSPA must track the transport of ^{235}U.

Table 3.1-4. Abstracted Inventory

Isotope	Specific Activity (Ci/g)	Grams in TSPA-SR CSNF Packages	Grams in TSPA-SR Codisposal Packages	
			From Spent Fuel	From HLW glass
^{227}Ac	7.24E+01	3.09E–06	1.13E–04	4.67E–04
^{241}Am[a]	3.44E+00	1.09E+04	1.18E+02	6.51E+01
^{243}Am	2.00E–01	1.29E+03	1.49E+00	5.09E–01
^{14}C	4.46E+00	1.37E+00	4.96E–02	6.43E–03
^{137}Cs	8.65E+01	5.34E+03	1.12E+02	4.50E+02
^{129}I	1.73E–04	1.80E+03	2.51E+01	4.80E+01
^{63}Ni	5.68E+01	5.53E+01	4.44E–01	3.39E–01
^{237}Np	7.05E–04	4.74E+03	4.79E+01	8.28E+01
^{231}Pa	4.72E–02	9.87E–03	3.25E–01	7.96E–01
^{210}Pb	7.64E+01	0.00E+00	1.40E–08	1.44E–07
^{238}Pu	1.71E+01	1.51E+03	6.33E+00	9.04E+01
^{239}Pu	6.21E–02	4.38E+04	2.30E+03	3.89E+03
^{240}Pu	2.27E–01	2.09E+04	4.89E+02	3.80E+02
^{242}Pu	3.93E–03	5.41E+03	1.11E+01	7.66E+00
^{226}Ra	9.89E–01	0.00E+00	1.87E–06	1.67E–05
^{228}Ra	2.72E+02	0.00E+00	6.98E–06	3.37E–06
^{90}Sr	1.37E+02	2.24E+03	5.54E+01	2.88E+02
^{99}Tc	1.70E–02	7.68E+03	1.15E+02	7.31E+02
^{229}Th	2.14E–01	0.00E+00	2.66E–02	4.08E–03
^{230}Th	2.06E–02	1.84E–01	1.06E–02	7.79E–03

Table 3.1-4. Abstracted Inventory (Continued)

Isotope	Specific Activity (Ci/g)	Grams in TSPA-SR CSNF Packages	Grams in TSPA-SR Codisposal Packages	
			From Spent Fuel	From HLW glass
^{232}Th	1.10E–07	0.00E+00	1.49E+04	7.77E+03
^{232}U	2.20E+01	1.01E–02	1.47E–01	8.23E–04
^{233}U	9.66E–03	7.00E–02	2.14E+02	1.11E+01
^{234}U	6.24E–03	1.83E+03	5.72E+01	4.63E+01
^{235}U	2.16E–06	6.28E+04	8.31E+03	1.70E+03
^{236}U	6.47E–05	3.92E+04	8.53E+02	3.96E+01
^{238}U	3.36E–07	7.92E+06	5.09E+05	2.66E+05

Source: From Chapter 7 and Table I-5 of the *Inventory Abstraction* AMR (CRWMS M&O 2000f) and a subsequent input transmittal (CRWMS M&O 2000ak).

NOTE: [a]Part of the ^{241}Am inventory is due to decay of ^{241}Pu and ^{245}Cm.

3.1.2 Uncertainties, Limitations, and Conservatisms

The screening analysis is conservative because all fuel types, bounding and average, all time periods, and all transport groups were examined. Any changes that might be expected in the wastes that may be disposed at Yucca Mountain will not change the radionuclides that were screened in for modeling in TSPA.

The representative inventories were derived from unqualified projections of future waste streams. The actual waste streams will be known only at the time of actual repository loading. The projected waste streams could differ from the actual waste streams in their fuel burnups, fuel ages, fuel enrichments, and utility efficiencies. However, changes that might be expected in the waste stream will produce only minimal (less than 20 percent) changes in the radionuclide activities in the fuels. Given this understanding, the values chosen for initial inventories in CSNF and codisposal waste packages are reasonable representations of the inventory that may be disposed at Yucca Mountain (CRWMS M&O 2000f).

3.1.3 Other Views

Alternative approaches for developing the radionuclide inventory have been explored in previous TSPAs (Wilson et al. 1994; CRWMS M&O 1995; CRWMS M&O 1998b) and in the DEIS (DOE 1999a). In previous TSPAs, (Wilson et al. 1994; CRWMS M&O 1995; CRWMS M&O 1998b), radionuclide activities for CSNF were developed by assuming an average set of fuel characteristics (enrichment, burnup, and age of the waste) and using the DOE's Characteristics Database to determine radionuclide activities for a fuel having the specified characteristics. Radionuclide activities for DSNF were developed by assuming that the defense fuels can be grouped into a few representative groups (based on their chemical characteristics) and calculating inventories for a representative fuel from each group. Radionuclide inventories for HLW were developed by assuming that an average of glass waste from the Savannah River Vitrification Plant and processed waste from Hanford, INEEL, and the West Valley Facility can represent HLW.

The TSPA-SR analysis screening method used a qualified process to ensure traceability. It looked at both bounding and average fuels, DSNF and HLW as well as CSNF, time periods from 100 to 1 million years, and both inhalation and ingestion doses. By using two solubility groups and three different transport groups and comparing the radionuclides within the groups, better discrimination was achieved. For example, if all isotopes were lumped together, plutonium would dominate the dose. But this is true only if Pu is readily transported to the accessible environment. The full TSPA-SR analysis and its colloid models will predict the likelihood of significant Pu transport. The screening could not rely on TSPA output and, thus, did not rely on any assumptions about solubility or transport affinity.

The TSPA-SR inventory analysis is more detailed and flexible than previous analyses and is tied to the waste stream. Changes in WP configuration or waste stream are more easily reflected in the per package inventory of representative WPs. An example of such a change is seen in the recent regrouping of naval spent nuclear fuel.

3.2 IN-PACKAGE CHEMISTRY COMPONENT

The function of the in-package chemistry model component is to estimate the fluid chemistry inside the WP over time after the initial breach of the disposal container. This chemistry is then used by the several other model components (see Figure 1.5-3) since the rate of degradation of the matrix of waste, the resulting dissolved concentration of radionuclides, the stability of any colloids, and degradation of cladding are all dependent on the chemistry of fluids within the WP. The rate of degradation of the waste matrix and inner stainless steel container, in turn, influences the fluid chemistry and so there is a coupling between all the chemically interacting components of the system.

The *In-Package Chemistry Abstraction* AMR (CRWMS M&O 2000g) is the primary document describing the regression equations used for evaluating the in-package chemistry. This document, in effect, abstracts the process models that are developed in *Summary of In-Package Chemistry for Waste Forms* (CRWMS M&O 2000o). The *In-Package Chemistry Abstraction* (CRWMS M&O 2000g) also relies on a few of the FEP screening arguments in *Miscellaneous Waste-Form FEPs* (CRWMS M&O 2000n). While abstraction of process models is numerically convenient, it is technically defensible only if the abstracted model conservatively bounds the predictions of process-level models. In this case, dissolved radionuclide levels predicted with the abstracted model must be equal to, or greater than, those predicted by the underlying process-level models. The most reliable way to assure technical reliability of the abstraction is to force it to be mechanistically identical to the process-level model. This was done for the solubilities of the majority of the primary radionuclides by building abstractions of in-package chemistry and solubility expressions on equilibrium speciation output of EQ3NR.

3.2.1 Description

The fluid chemistry inside the package (in-package chemistry) is dependent upon the initial chemistry of the water entering the breached package, the volume of water flowing through the package, the amount of water remaining within the package, and the amount of time that inflows into the waste package occur. The inflows were assumed to have the composition of J-13 groundwater. Early breach of a WP would almost certainly entail chemical interaction under

substantially higher temperatures. Under such a scenario, evaporative concentration of reacting fluids would be expected to result in in-package fluids that diverge substantially from the compositions calculated under the 25°C, zero-evaporation limiting case considered here (see below). Various breach scenarios can be envisioned for the container, but as explained later, the current model assumes a fully flooded container (bathtub scenario) for the volume of water present in the container at all times. In addition, the in-package chemistry is dependent upon the degradation rates of the contents of the package. Two representative WPs were modeled, a CSNF package and a DSNF/HLW codisposal package (Figure 3.2-1). In both cases, there is an inner stainless steel disposal container, but the basket materials and waste forms are different and influence the fluid chemistry at least for short time periods.

Direct use of a complex equilibrium code within the TSPA-SR analysis calculation was not practical; rather, a limited number of simulations were run with the complex equilibrium code, for a variety of input conditions and degradation rates of the contents. Regression equations were then developed for use directly in the TSPA-SR analysis calculations as explained below (Figure 3.2-2).

3.2.1.1 In-Drift Fluids

The calculations for revision 00 of the *Summary of In-Package Chemistry for Waste Forms AMR* (CRWMS M&O 2000o) were performed in parallel with the testing and analysis of the effects of the near-field environment (NFE) on the water that may contact and enter the waste package. This parallel work is summarized in the NFE (CRWMS M&O 2000z), EBS (CRWMS M&O 2000y), and waste package degradation (CRWMS M&O 2000d) process model reports. However, for these first calculations, the J-13 well water composition was used for water entering the waste package (Harrar et al. 1990, Section 11). J-13 well water is quite dilute and its composition is not expected to significantly affect the in-package chemistry. On-going work will verify this assumption, or provide better information.

J-13 well water is used as a surrogate for the groundwaters passing through the repository and possibly into breached WPs several thousand years after the repository has closed. The most abundant dissolved constituents in J-13 well water include Na and C, along with Si, Ca, K, F, Cl, N, and Mg. Dissolved Na, K, and Ca came from dissolution of feldspars into down-flowing meteoric water. Si was contributed by dissolution of feldspars and Si polymorphs. C and N came from equilibrium with soil and atmospheric gases encountered in transit. J-13 well water has undergone cation exchange with zeolitic rocks (causing increased Na/Ca) compared to UZ pore waters at the repository horizon. This is important for understanding the overall chemical budget of repository waters. Because Na and Ca play non-specific roles in the overall degradation process, ion exchange is less important to predicting what occurs once J-13 comes into contact with WPs. J-13 well water has a pH of ~8 and is thought to be close to equilibrium with carbon dioxide at levels slightly higher than that present in the atmosphere. The slightly higher than neutral pH of the solution is a direct result of alkalinity production that occurs when primary phase silicates are weathered. Waste package alteration scenarios are less sensitive to the exact composition of the water entering the package than they are to dissolution rates of a number of waste package components because: (1) The rates of WP component dissolution are not overly sensitive to minor variations in these parameters and (2) WP components rapidly come to dominate the dissolved phase after onset of reaction. Calculations have been started for

a new version of the *Summary of In-Package Chemistry for Waste Forms* analyses and models report to test the sensitivity of the in-package chemistry to incoming water composition. It is expected that the incoming water will consist of the water found in the unsaturated zone subsequently modified by evaporation and/or condensation and fluid-rock interaction. Accordingly, sensitivity calculations are being done using a range of waters including the compositions in Table 3.2-1 as well as 100-fold evaporated J-13 water.

Table 3.2-1 provides the major element composition of J-13 well water, Drift-Scale Test (DST) water, and the output of THC modeling. DST compositions represent an average of waters collected from Alcove 5 near the DST and were used as input compositions for modeling of thermal-hydrologic-chemical coupling in the NFE. DST water is thought to have moved appreciably through fractures and interacted with the rock matrix. The modeling output for waters expected to seep into the drifts is shown for two time periods, the transitional cool-down period (1,000-2,000 years) and the extended cool-down period of 2,000-100,000 years. By using these water compositions as well as 100-fold evaporated J-13 well water, the sensitivity calculations are covering a broad range of chemistries and should give a good indication of the sensitivity of the resulting in-package chemistry on the input water composition.

Table 3.2-1. Major Element Composition of Input Waters Considered in Sensitivity Studies

Component	J-13 Well Water[a]	DST[b]	Modeling[c] 1000-2000 years	Modeling[c] 2000-100,000 years
pH	7.41	8.32	7.8	7.3
Na (ppm)	45.8	61.3	60	60
$SiO_{2(aq)}$ (ppm)	61	70.5	126	72
Ca (ppm)	13.0	101	40	72
K (ppm)	5.04	8.0	12	3.9
Mg (ppm)	2.01	17	0.04	0.19
F (ppm)	2.18	0.86	0.85	0.85
Cl (ppm)	7.14	117	113	117
SO_4(ppm)	18.4	116	115	115
HCO_3 (ppm)	128.9	200	18.3	128

Sources: [a]From Harrar et al. 1990 Table 4.1 with ppm of Si converted to ppm of SiO_2.
[b]Units converted from Table 3 of CRWMS M&O 2000af.
[c]Units converted from Table 3 of CRWMS M&O 2000ai.

The in-package chemical reaction calculations used input fluid flows of 1.5 to 150 L/yr. The temperature of the simulations was set at 25°C to represent the conditions that will occur several thousand years after WP emplacement, when the original thermal pulse has passed and temperatures have returned to near ambient levels. Waste form degradation may occur at temperatures up to 100°C. It is assumed that the process can be modeled adequately with the 25°C thermodynamic database. The calculations represent what occurs at times > 10,000 years, after the thermal pulse has passed and package temperatures are at, or below, 100°C. The justification for using 25°C thermodynamic data to model processes that might occur at somewhat higher temperatures is that many of the input thermodynamic parameters are not strongly sensitive to temperature over the range of 25 to 100°C, hence the broad scale features of

the output fluid compositions are deemed independent of temperature. Dissolution rates of WP internal components would be greater at temperatures greater than 25°C. Typically, dissolution rates of minerals roughly double with every 10°C increase in temperature. In the absence of high temperature dissolution rates, this is probably a reasonable approximation for the behavior of metallic and oxide WP internal components as well. The net effect of temperatures greater than 25°C would therefore be a more rapid release of WP components into solution than what is reported here. Note that the existing calculations tend to err in this direction already because they use very high rates as input.

Higher temperatures would also affect the boundary conditions of the calculation by decreasing the solubilities of oxygen and carbon dioxide in the fluids reacting with the WP. A more important impact of temperature on in-package chemistry would almost certainly be the evaporative concentration of inflowing and reacting solutions, which could conceivably change the concentrations of a number of important chemical species by orders of magnitude. Evaporation of J-13 water before interaction with WP components would result in a high ionic strength, alkaline solution. For example, 200-fold open-system concentration of J-13 water produces a pH 10, 1 mol/L ionic strength solution, which has 200 times as much chloride and total carbonate as J-13. Evaporation of J-13 type water is expected to lead to the formation of calcite, chalcedony, fluorite, and possibly sepiolite and other less abundant minerals. The important consequences of evaporation for fluid influx chemistry are increases in chloride, pH, and bicarbonate. Increased chloride impacts WP internal component corrosion. Glass and mineral dissolution rates tend to double for every unit increase in pH above 8. Many radionuclides are more soluble in the presence of bicarbonate. The sensitivity calculations will provide an indication of how important evaporation may be on in-package chemistry.

The partial pressure of carbon dioxide and oxygen were set to, respectively, $10^{-3.0}$ and $10^{-0.7}$ bar (DOE 1998c, Figure 3-36). The latter represents equilibrium with oxygen at atmospheric levels. Reaction with rock bolts (and steel supports) was neglected because the interaction of the fluid with waste package components would greatly outweigh any transient contact of the fluid with the rock bolts and the steel ground support. The total surface area of rock bolts likely to be encountered by seepage before contact with waste package components is small. It is likewise difficult to envision substantial interaction with the support material before seepage into the waste package.

3.2.1.2 Water Contact Scenarios

As described in the *Summary of In-Package Chemistry for Waste Forms* Process Model CRWMS M&O (2000o), a simple degradation scenario for the WP entails breaching the WP (presumably through stress corrosion cracking and/or mechanical failure); filling of the WP void volume with seepage fluids from the drift; reaction within the WP; and releasing contaminated fluids at the same rate at which fresh fluids entered. This simple breach scenario was adopted and each breached WP was modeled as a continually stirred, fixed volume vessel. Specifically, the calculations used the solid-centered, flow-through mode of EQ3/6; in this mode, an increment of fresh fluid solution is added to the WP system and a like amount of solution is removed at each time step. The void volumes considered were a constant 4,507 L for the CSNF WP and a constant 5,811 L for the DSNF WP.

The assumption of a continually stirred vessel is a simple and common approach to estimating the nature and extent of chemical reaction. The basis of the assumption is that either the fluid is continually stirred, or equivalently, the residence time of the fluids in the vessel is sufficient for diffusion to eliminate chemical gradients. In general, the latter basis is valid because of the geologic time scale of seepage and chemical reaction involved.

Further justification is reasoned as follows. If wetting of WP components were complete, but void filling was not, the reduced fluid/solid ratio would tend to maximize the amount of dissolved solids in the effluent. On the other hand, the overall chemical reaction would be less for fluids that found themselves in "dead-end" channels, or if portions of the WP were physically inaccessible to fresh fluids. Furthermore, fluids that found "fast paths" that short-circuited interaction with WP solids would bear a much fainter signature of chemical reaction.

Evaporation of fluids by residual heat could increase the salt content (i.e., ionic strength), and, thereby, increase some radioisotope solubilities, and possibly increase the rate of WF or WP degradation. The increased salt content, however, is not expected to significantly alter the overall reaction path (CRWMS M&O 2000o, Section 7) and was examined in a FEP on the use of J-13 well water (Section 2.3.2.2).

This breach scenario provides an upper bound for in-package radionuclide transport and release. It does not include any time delay for water that enters the package; it does not include any retardation of radionuclides into immobile secondary phases by coprecipitation or sorption; it does not allow for radionuclides to be held-up in stagnant areas of the WP, and it maximizes contact between oxidizing fluids and WF and WP materials.

The analyses using the in-package chemistry results also used bounding assumptions or large uncertainty (see Section 3.3, CSNF Matrix Degradation; Section 3.4, CSNF Cladding Degradation, Section 3.6, HLW Degradation, and Section 3.7, Dissolved Radioisotope Concentration). In particular, the possible effects of localized chemistry are included in the CSNF cladding degradation model.

3.2.1.3 Waste Package Contents

In-drift solutions seeping into a breached WP would encounter a number of kinetically reactive solids whose reaction rates are only known within orders of magnitude (Table 3.2-2). Fluids intruding into WPs containing CSNF would encounter UO_2 fuel wrapped in Zircaloy cladding, Al alloy, 316 stainless steel (with and without neutron absorbers, such as boron or $GdPO_4$), and A516 low carbon steel. A range of degradation rates was used for each. Dissolution of the fuel, and release of radionuclides, occurs only after degradation of some of the cladding. General corrosion of cladding is likely to be insignificant under the geochemical conditions expected inside reacting WPs. Uncertainty in the initial failures, localized corrosion, mechanical damage, and other degradation mechanisms are addressed in the cladding degradation AMRs and were not available when the in-package calculations were started, so a range of clad damage and CSNF fuel exposure was investigated (100 percent, 20 percent, and 1 percent). The WP configuration used for the codisposal package calculations was that of the Fast Flux Test Facility (FFTF) DSNF with five HLW glass logs.

Table 3.2-2. Chemical Characteristics of CSNF and Codisposal Waste Package Components

Material	Amount (mol/L)	Degradation Rate ($mol/cm^2 \cdot s$) (25°C)	Surface Area (cm^2/L)
CSNF WP Parameters			
Al alloy	2.36	2.28×10^{-13}	96.424
A516	11.948	2.487×10^{-11} (high) 8.706×10^{-12} (low)	504.160
316 SS	23.75	2.528×10^{-13} (high) 2.528×10^{-14} (low)	52.500
316 SS/B	7.856	1.169×10^{-13} (high) 1.169×10^{-14} (low)	156.010
CSNF	27.21	$4.420 \times 10^{-14} + 10^{-10.59 - 0.5 \cdot pH}$	96200
Codisposal WP Parameters			
304L	6.384 0.065	2.516×10^{-13} (high) 2.516×10^{-14} (low)	150.804 6.661
A516	10.140		129.615
HLW glass	24.821	$1.983 \times 10^{-19}[H^+]^{-0.4721} + 6.144 \times 10^{-12}[H^+]^{0.6381}$ (low) $1.983 \times 10^{-18}[H^+]^{-0.4721} + 6.144 \times 10^{-11}[H^+]^{0.6381}$ (high)	1915.142
316 SS (various parts)	0.946 20.573 0.423 26.35		24.134 27.267 257.913 56.5
316 SS/ $GdPO_4$	0.653	2.528×10^{-14}	16.732
U oxide	0.018	$4.420 \times 10^{-14} + 10^{-10.59 - 0.5 \cdot pH}$	72.072
MOX	0.393	$4.420 \times 10^{-14} + 10^{-10.59 - 0.5 \cdot pH}$	1559.382

NOTE: Table 1 of *Summary of In-Package Chemistry for Waste Forms* ANL-EBS-MD-000050 (CRWMS M&O 2000o). Values for 304L have been corrected; each had an extraneous "2" added to the beginning.

3.2.1.4 EQ3/6 Reaction Path Modeling

As described in the *Summary of In-Package Chemistry for Waste Forms* AMR (CRWMS M&O 2000o) simulations of WP alteration by ambient groundwater were done using the qualified reaction path code EQ3/6 (CRWMS M&O 1998a; CRWMS M&O 1999c), which titrates masses of WP components at a rate determined by input reaction rates and fluid influxes into the breached WP. The software package, EQ3/6, Version 7.2b, was approved for QA work by LLNL and is identified as Computer Software Configuration Item (CSCI): UCRL-MA-110662 V 7.2b. EQ3/6 periodically assesses the chemical equilibrium state of the solution and removes newly saturated, secondary mineral or gas phases from the fluid. In addition to kinetic inputs (e.g., rates, compositions, and masses of reactants) EQ3/6 relies on a thermodynamic database describing the chemical stability of minerals, aqueous species, and gases. When the reacting solution becomes saturated with respect to solids or gasses, EQ3/6 converts dissolved components into the respective phase and then allows the latter to act as a reservoir of the respective components for use at subsequent times. In this way, EQ3/6 tracks the elemental composition of the reacting fluid for the duration of the reaction path calculation while at the same time providing estimates of the nature and masses of secondary phases that are predicted to

form. The code, however, does not provide for the kinetic inhibition for phase formation, which is, therefore, the responsibility of the analyst. Generally, the assessment of kinetic inhibition must be made on the basis of there being an absence of the particular phase in low-temperature (~25°C) environments. For example, dolomite ($CaMg(CO_3)_2$) is predicted to form in a number of runs due to the accumulation of Ca, Mg, and alkalinity upon reaction with WP components. Dolomite, though thermodynamically favored to grow in a number of low temperature natural solutions such as seawater, apparently must overcome severe kinetic obstacles to actually form and is actually observed growing only in highly evaporated brines. These kinetic considerations are then the basis for suppressing formation of dolomite in the reaction path calculations. Similar arguments are used to suppress the formation of a number of oxide and silicate minerals that are typically observed to form only under high temperature conditions.

3.2.1.5 Results of Waste Package and Waste Form Alteration Modeling

Table 3.2-3 summarizes the range of in-package fluid compositions predicted to occur for both the CSNF and codisposal packages (see Section 3.2.1.6 for further discussion of this chemistry and Section 3.2.2 for limitations).

Table 3.2-3. Range of In-Package Fluid Compositions (mol/L for all except pH and Eh [volts])

Variable	CSNF	Codisposal
pH	3.6 - 8.1	4.8 - 10.0
Eh (V)	0.7 - 1.0	0.6 - 0.9
Ionic Strength	0.003 - 1.7	0.003 - 5.8
Ca	3.9×10^{-6} - 0.00032	3.0×10^{-7} - 0.02
C	2.8×10^{-5} - 0.002	3.5×10^{-5} - 0.5
Si	3.7×10^{-10} - 1.9×10^{-4}	5.3×10^{-10} - 1.9×10^{-4}

Source: Table 3 of *Summary of In-Package Chemistry for Waste Forms* ANL-EBS-MD-000050 (CRWMS M&O 2000o)

CSNF WPs–The majority of WPs in the repository will contain spent fuel rods from commercial reactors. The primary reactions that occur upon breach of CSNF WPs and subsequent alteration by intruding fluids are: (1) a rapid decrease in pH caused by dissolution of low-carbon steel and (2) a subsequent rise in pH caused by CSNF dissolution and oxidation of Al alloy (Figure 3.2-3). The latter rise is more pronounced under conditions of high clad failure. By the same token, the lowest predicted pHs are those calculated for the least amount of clad failure. Feedback between the pH and clad corrosion rate was not included within the calculations, as the pH is not expected to go low enough to cause significant increases in clad failure rates (see Section 3.4.2.4). Under the assumed fixed CO_2 conditions of the simulations, carbonate levels are relatively high at high pH and low at low pH. Because both U and Pu form soluble complexes with carbonate, the dissolved levels of each tend to increase at high pH and decrease at low pH. However, the lowest pHs occur in the early stages of the reaction paths before appreciable U or Pu has been dissolved from the CSNF matrix.

Minerals predicted to form during alteration of CSNF WPs include: copper minerals, silicates (clays, uranium silicates), oxides (metal oxides, actinide oxides), carbonates, phosphates, sulfates, and fluorides. Although the inputs to the calculation are normalized to a liter of fluid, and a liter volume is assumed to persist, the large volumes of lower density corrosion products (mainly hematite and $UO_3 \cdot 2H_2O$) suggest that alteration products might actually fill the void space and seal off portions of the WPs to further fluid influx. This is further discussed in Section 3.4.

Codisposal WPs–In all codisposal runs (Figure 3.2-4), the input pH of ~8.1 prevails initially. Then the system pH progresses towards a minimum, primarily due to the oxidation of the A516 carbon steel. The high specific rate and relatively high surface area of the latter mean that its dissolution tends to dominate the whole reaction path, at least as long as it remains. Oxidation of sulfur in the steel to sulfate is the primary proton-producing reaction. After the steel is exhausted, dissolution of base cation-containing HLW glass leads to increased pHs. pHs as high as 10 and ionic strengths as high as ~5.8 mol/L were predicted for codisposal WPs under conditions of high glass dissolution and low flow rates.

Minerals predicted to form during alteration of codisposal WPs typically include the same minerals formed during CSNF alteration, though borates are also predicted to form, and the uranium silicates are more important.

3.2.1.6 Abstraction of In-Package Chemistry

The predicted compositions of in-package fluids are of particular importance for estimating releases of radionuclides to the biosphere. The *Summary of In-Package Chemistry for Waste Forms* (CRWMS M&O 2000o, Section 6) provides reaction-path modeling assuming steady-state CO_2 and O_2 concentrations in the drift and breached WPs. The analysis indicates the broad range of effluent compositions that might be expected from a breached WP and provides a basis from which to establish abstractions describing radioisotope solubilities.

The analysis of calculated in-package fluid compositions possesses four broad features. Carbonate alkalinity increases with pH due to the assumed fixed partial pressure of CO_2. System Eh decreases with pH due to the assumed constant partial pressure of O_2. Low early pHs are only seen with low cladding failures, and alkalinities typically correlate with high ionic strengths associated with high glass dissolution and low flow rates.

The EQ3/6 simulations calculate the fluid chemistries that are operative over long periods of time (> 1,000 years). The simulations over this time period are typically different than those that prevail over shorter periods of time. Recognition of this change in process is important for estimating long-term behavior. Typically reaction times of less than 1,000 years result in relatively minor degradation of WP components; hence, the concentrations of many radionuclides are often well below their respective solubility limits. In effect their concentrations depend directly on the dissolution rates of WP components. At much greater time spans, many, but not all, radionuclides have reached saturation with at least one mineral phase. Once this has occurred, dissolved levels of the particular radionuclides will depend on the thermodynamics of secondary phase stability and much less directly upon the dissolution rates of the WP components.

The abstraction of in-package processes is based upon a series of multiple linear regression analyses of the output from the EQ3/6 simulations as presented in the *In-Package Chemistry Abstraction* (CRWMS M&O 2000g, Attachment I). The multiple linear regression analyses take explicit account of the change in process control with time and treats results for times less than 1,000 years after waste package breach separately from those for later times. The specific relationships between pH, Eh, clad failure, and component degradation are illustrated in Figure 3.2-2. The predictions from the multiple regression analyses, and the output from the EQ3/6 simulations are presented in the abstraction AMR (CRWMS M&O 2000g) and summarized here.

Because it is difficult to track the failures of individual packages in TSPA, the in-package chemistry abstraction was implemented in an approximate way for a group of packages. Let t be the current time and let t_{fail} be the average time of failure for all the breached waste packages. The short-time abstraction would apply for $t - t_{fail} < 1000$ years, and the long-time abstraction would apply for $t - t_{fail} > 1000$ years.

The pH abstraction follows two lines of reasoning based on the WP type (CSNF or codisposal), and the difference in kinetic rate laws between the two. The rate law for CSNF is proportional to the hydrogen ion activity, i.e., proportional to pH, such that at low pH the dissolution rate increases (CRWMS M&O 2000o, Section 6.1.1). For HLW the rate law is "U" shaped with the minimum at pH 7 and the rate increasing above and below pH 7 (CRWMS M&O 2000o, Section 6.1.1). Therefore, conservative assumptions for one waste form may not be conservative for the other. In the case of the CSNF (and naval SNF), for example, assuming the lowest observed pH for the first 1000 years after breach, and the averaged pH thereafter would be the most conservative, while still honoring the pH–time history. However, for the codisposal package using the lowest observed pH would be conservative for the first 1000 years after breach but not for later times, when use of the maximum pH is conservative. The difference in the rate laws between HLW and CSNF and the difference in their pH–time profiles for the two waste forms predicates the use of different assumptions in the abstractions.

For the CSNF, at times less than 1,000 years after waste package breach, minimum pH values for each flow/cladding/corrosion scenario were regressed to produce two abstractions of pH; one for low WP corrosion rates and the other for high WP corrosion rates, where the axes were x = cladding coverage, y = water flow (Q), and z = pH. This process was repeated for times greater than 1,000 years after breach, where the average pH for the entire modeled duration (0 to 10,000 years after breach) was used to calculate the average pH. The average pH for the entire duration is conservative because it includes the low pH values at early times, which tends to lower the average value compared to averaging over the late time information (1,000 to 10,000 years after breach).

The resulting four data matrices were used to generate four response surfaces describing pH at less than and greater than 1,000 years after breach. These response surfaces were provided for TSPA to use in pH calculations. Table 3.2-4 provides the equations of the planes that bound the pH parameter space for CSNF, and Figures 3.2-5 and 3.2-6 show these planes. Each plane represents either the low or high corrosion rate data for the WP materials, 316 stainless steel, aluminum alloy, A516 carbon steel, and borated stainless steel. An uncertainty range of ±1 pH unit was recommended for TSPA.

Table 3.2-4. Response Surface Parameters for CSNF pH

$z = y_o + ax + by$							
Time After Breach (Years)	**Corrosion Rate**	**z**	**y_o**	**a**	**b**	**Adj. R^2**	**Sigma (pH)**
< 1,000	Low	Min. pH in 0 - 1,000 years	3.4916	–1.0918	0.4571	0.993	0.066
< 1,000	High	Min. pH in 0 - 1,000 years	3.3977	–0.7468	0.3515	0.998	0.027
> 1,000	Low	Ave. pH in 0 – 10,000 years	6.0668	–0.5395	4.0479	0.774	0.257
> 1,000	High	Ave. pH in 0 – 10,000 years	6.0913	–0.3057	1.2913	0.77	0.13

Source: *In-Package Chemistry Abstraction*. ANL-EBS-MD-000037. (CRWMS M&O 2000g, Table 6 and Attachment I)

NOTE: z = pH a range of ± 1 pH unit was applied to all calculated pH values. x = $\log_{10}$ (cladding coverage), y = water flow (m^3/yr) (Q)

Table 3.2-5 and Figures 3.2-7 and 3.2-8 show the equations describing minimum and maximum pH parameter space for codisposal WPs. The balance of the acid production from the Alloy 516 carbon steel and base production from the glass determine the pH. At early times (< 1,000 years after breach), acid production dominates, but as the steel is consumed, base production overcomes the acid production. In most cases, this is not calculated to occur until considerably after 10,000 years after breach. To obtain conservative glass degradation rates, the maximum pH in the 100,000 year simulations was used.

Table 3.2-5. Response Surface Parameters for Codisposal pH

$z = y_o + ax + by$							
Time After Breach (Years)	**Corrosion Rate**	**z**	**y_o**	**a**	**b**	**Adj. R^2**	**Sigma (pH)**
< 1,000	Low	Min. pH in 0 - 1,000 years	5.1257	2.6692	0.0764	0.94	0.11
< 1,000	High	Min. pH in 0 - 1,000 years	4.7324	0.7307	0.0837	0.99	0.04
> 1,000	Low	Max. pH in 100,000 years	8.4247	–3.4173	0.1403	0.78	0.37
> 1,000	High	Max. pH in 100,000 years	9.2554	–3.1280	–0.0418	–0.31	0.76

Source: *In-Package Chemistry Abstraction*. ANL-EBS-MD-00037. (CRWMS M&O 2000g, Table 12 and Attachment I)

NOTE: z = pH, a range of ± 1 pH unit was applied to all calculated pH values. x = water flow (m^3/yr) (Q), y = relative glass dissolution rate

Due to the simplicity of the mathematical expressions used to calculate pH, it is possible, by increasing the water flow Q, to calculate unrealistic pH values. Therefore, it was necessary to set limits for the maximum allowable Q. Since the calculated pH from all of the expressions in Tables 3.2-4 and 3.2-5 are directly proportional to water flow through the WP and as the water flow increases the potential for reaction of the through-flowing water with the WP/waste form materials decreases, the pH of the water exiting the WP should approach that of J-13 well water. Therefore, above some maximum Q-value, the water exiting the WP should have the same pH as the water entering the WP. This critical value of Q was calculated by solving the expressions in Tables 3.2-4 and 3.2-5 for Q and using the input, J-13 well water value for pH and assuming linear behavior.

For CSNF at time less than 1,000 years after breach, the pH expressions (Table 3.2-4) are valid within the range of $0 < Q < 5.3$ (m^3/yr). Likewise, when $Q \geq 5.3$ m^3/yr, then the pH is constant at 8.1 (±1). For time greater than 1,000 years after breach, the pH expression (Table 3.2-4) is valid for $0 < Q < 0.24$ (m^3/yr), and when $Q \geq 0.24$ m^3/yr the pH is constant at 8.1 (±1). Similarly for codisposal waste packages, the pH expressions (Table 3.2-5) are valid within the range of $0 < Q < 1.1$ (m^3/yr) at time less than 1,000 years after breach and $0 < Q < 0.36$ (m^3/yr) for time greater than 1,000 years after breach. Above these values, the pH is constant at 8.1 (±1).

Total dissolved carbonate affects the solubilities of a number of actinide phases as well as the degradation rate of CSNF. The total dissolved carbonate is abstracted as a function of pH by assuming a partial pressure of atmospheric $CO_2 = 10^{-3}$ atm and equilibrium between the carbonate, bicarbonate, and carbonic acid in solution:

$$\text{Total C (mol/L)} = 10^{-4.47} + (10^{-10.82})/10^{-pH} + (10^{-21.15})/10^{-2pH} \qquad \text{(Eq. 3.2-1)}$$

The system Eh is important in particular for determining the solubilities of many of the actinides. In aqueous systems in equilibrium with the atmosphere, the Eh may be abstracted directly from the pH and the assumed partial pressure of oxygen (0.20 atm):

$$Eh = 1.217 - 0.059 \cdot pH \qquad \text{(Eq. 3.2-2)}$$

where Eh is in volts. The ionic strength was used in determining the colloid content of the water exiting the NFE. For the period before 1,000 years after breach, the minimum ionic strength was calculated for each flow/cladding scenario (beta distribution from 2.757×10^{-3} to 2.922×10^{-3} mol/L) and glass rate/corrosion rate scenario (beta distribution from 2.537×10^{-3} to 3.479×10^{-3} mol/L). At times greater than 1,000 years after breach, the average value of ionic strength was used from each scenario to get the ionic strength range used in the abstraction (beta distributions from 2.827×10^{-3} to 3.943×10^{-1} mol/L for CSNF and from 7.86×10^{-3} to 1.353 mol/L for codisposal). This is a conservative approach compared to using the maximum value because low ionic strength is conducive to colloid formation, and using the minimum value would also create the worst case, which is not reflective of the actual in-package conditions.

Fluoride was considered in the cladding degradation model. Fluoride was only pertinent for the CSNF and naval SNF because there was no cladding protection included in the codisposal package modeling. The CSNF calculations indicate that the fluoride concentration is controlled by fluorapatite. The abstracted distribution was normal between 1.148×10^{-4} and 9.114×10^{-4} mol/kg with a standard deviation of 2.2×10^{-4} mol/ kg. Chloride levels remain constant at 2.0×10^{-4} mol/kg and therefore are too low to influence the models that rely on bulk chemistry.

3.2.2 Confidence/Limitations/Validation

This work represents the first attempt by the DOE to model the chemical interactions that occur within the WP for use within the TSPA calculations. Only limited sensitivity runs have been completed to study the importance of issues such as:

- **Codisposal fuel configurations** (type and amount of DSNF, type and amount of basket materials) (Sections 6 of CRWMS M&O 2000aa, CRWMS M&O 2000ab, CRWMS M&O 2000ac).

- **Feedback between chemistry and corrosion rates (built-in or iterations)**–The substantial accumulation of alteration products might "choke off" subsequent corrosion. At the same time, sharp shifts in fluid chemistry associated with WP alteration might also accelerate corrosion. At present, there is little feedback, either negative or positive, within the modeling. Iterations or more involved rate laws describing chemical corrosion would allow the more important feedbacks to be assessed. For example, for a flooded WP, the development of an alteration rind, even 1 cm thick, could prevent widespread oxidizing conditions at the sites of waste form degradation, which could substantially lower actinide solubilities (Section 6.3 of CRWMS M&O 2000ad). The uncertainties associated with predictions of fluid flow and solid exposure in the WP are formidable. By assuming full exposure of WP components, the calculation is conservative since it maximizes the potential for chemical degradation of the WP and for release of radionuclides.

- **Water contact scenario**–Predictions of specific degradation geometries and reduction in the uncertainty due to water contact scenario are difficult. The water contact scenario in the TSPA-SR analysis used to calculate in-package chemistries is merely the simplest of approaches. Only scoping calculations to determine the sensitivity of the system are justified at this time.

- **Assumptions of fixed O_2, CO_2**–Assuming oxidized, carbonated conditions in the WP tends to maximize the solubilities of many of the most important radionuclides. Reaction paths calculated under less oxidized, and/or less carbonated conditions would tend to predict lower dissolved levels of Pu and U (Section 6.3 of CRWMS M&O 2000ad).

- **Hydrogen evolution**–The assumption was made that H_2 gas was not evolved in modeling carbon steel corrosion. This is conservative in allowing for the maximum pH reduction.

- **Radiolysis**–Radiolysis was neglected in the present analysis. The probability of WP breach is quite low before about 1,000 years, and the likelihood of significant radiolysis is much lower at later times. In theory the formation of NO_2 by radiolysis might conceivably cause in-package pHs to decrease by forming nitric acid. Two features that would tend to work against such a scenario are the inherent buffer capacity of the assemblage and outgassing of NO_2 from the breached WP and into the drift. Radiolysis of water and off-gassing of H_2 may also increase the local effective Eh; however, at late

times this is also not expected to be significant (CRWMS M&O 2000s, Section 6.2; 2000h, Section 6.1; 2000n, Section 6.2.52). Highly localized energy deposition from alpha-decay might have some effect on dissolution rates, but these effects are also not expected to be significant (CRWMS M&O 2000h).

- **Database**
  - **Temperature**–Although the thermodynamic database used in the EQ3/6 calculations extends to 300°C, for many species, the data are most accurate at 25°C, the temperature where the data were originally measured.
  - **Ionic strength**–Note that calculations for solutions with ionic strengths exceeding ~0.7 mol/L are less accurate than dilute systems, but the uncertainties are small relative to the other uncertainties within the system.

The above limitations and uncertainties were considered as the modeling was performed (see CRWMS M&O 2000ad, Section 5.1 for discussion of ionic strength effects on actinide carbonate complexes and some uncertainties in the databases [Pu stability constants, U-silicate stability constants]). The range of predicted chemistry is quite large and conservative, and further work is expected to only fine-tune the scenario and time dependent in-package chemistry. The calculated range of in-package chemistries is believed to represent the widest possible range of conditions that can confidently be expected. Indeed, a number of natural processes would tend to prevent more extreme chemistries than predicted here. Substantial reservoirs of freely exchangeable carbon dioxide would tend to prevent excursions to hyperalkaline conditions. Dissolution of solid components in the WP can buffer pH as well. The fact that free oxygen is likely to prevail in the drifts sets limits on how reduced WP fluids can become. Moreover, the accumulation of reactive corrosion products formed during WP degradation will also tend to buffer in-package solution chemistries.

Based on the considerations discussed above, this model is appropriate for its intended use.

In summary, the in-package geochemistry model was based on a large number of reaction path calculations covering a range of fluid fluxes, WP component degradation rates, and clad exposure scenarios. The calculated range of solution compositions is believed to encompass the most reasonable range of compositions of fluids that ultimately exit the WP. Nevertheless, the model outputs are valid only under the conditions stated above, and by necessity require a number of assumptions about the amount of WP components contacted by fluids, feedback between chemical compositions and corrosion, radiolysis, in-package evaporation, and the validity of the thermodynamic database. Wherever possible, these inputs have been used in a conservative fashion. Nevertheless, because there exist no complete long-term measurements of WP degradation, and because natural analogs for such a process are not known to exist, it is not possible to fully validate the model at this time, or to assign absolute confidence limits to in-package chemistry outputs. Model outputs would be expected to be less valid if temperatures greatly exceed 25°C, if in-package evaporation causes ionic strengths to exceed 1 mol/L, or if input solutions contact only certain parts of the WP rather than the entire WP.

3.2.3 Other Views

Chemistry inside of the WP affects the degradation of the waste form and the ability to support cladding credit. The TSPA Peer Review report (Budnitz et al. 1999, Section II.E) cited the TSPA-VA (DOE 1998b, p. 3-102) in noting that "…the chemistry within the WP, hence the long term performance of Zircaloy cladding, is not well understood and has considerable uncertainty." It also noted (Budnitz et al. 1999, Section II.G) that hydrogen production internal to the WP was not addressed adequately in the TSPA-VA (DOE 1998b). The peer review report (Budnitz et al. 1999, Section IV.E) also cites an Nuclear Waste Technical Review Board (NWTRB) report to Congress (NWTRB 1998, p. 34) calling for theoretical and experimental work to predict the ranges of local environmental conditions that could exist inside a WP and identifies the need for more explicit definition of the geochemical environment during potential corrosion events, particularly within the WP (Budnitz et al. 1999, Section II.E).

For the TSPA-SR analysis, the Waste Form Degradation Model now explicitly contains an In-Package Chemistry Component (a regression model based on process modeling) to predict ranges of chemical conditions inside the WP to better characterize the uncertainty in the degradation of the cladding and waste form. Ongoing sensitivity studies will provide more information. For the TSPA-SR analysis, the production of hydrogen was examined; however, the process was screened out in a FEP argument.

3.3 CSNF DEGRADATION COMPONENT

Under oxidizing conditions in the presence of liquid or vapor water, the UO_2 in CSNF is not stable and alters. Alteration of the UO_2 matrix can liberate radionuclides important to human dose.

The main function of the CSNF degradation model component is to determine the rate of degradation of the CSNF fuel as a function of temperature and water chemistry (specifically, pH, and partial pressures of O_2 and CO_2) as shown in Figure 3.3-1. This degradation rate function is combined with the in-package chemistry to determine a rate, which is then directly used by the Cladding Degradation Model Component (Section 3.4) to determine the rate that the CSNF cladding splits open and exposes more of the fuel matrix. In addition, the CSNF degradation model examined the distribution of radionuclides within the fuel and established a gap fraction for the more volatile radionuclides.

One AMR is the primary document supporting the CSNF Matrix Degradation Model Component, *CSNF Waste Form Degradation: Summary Abstraction* (CRWMS M&O 2000h) (see Figure 3.3-2). This, in turn, depends on the data reports of the major sets of experiments conducted on CSNF. A minor document supporting this AMR is *Miscellaneous Waste-Form FEPs* (CRWMS M&O 2000n).

3.3.1 Model Description

To determine the rate and consequences of CSNF degradation, dissolution of a number of fresh and spent fuels was examined under both saturated and unsaturated conditions using flow-through reactors as well as static, batch reactors and drip tests. Flow-through experiments involve passing fluids past a chamber containing CSNF fragments and allow rapid measurement

of the dependency of CSNF forward dissolution rates on variables such as solution pH and pCO_3. Batch tests involve placing a few grams of spent fuel in various configurations in less than a liter of synthetic J-13 groundwater, periodically sampling the solution, and analyzing for the appropriate radionuclides. These data provide information on the spent fuel dissolution rates for saturated conditions such as may occur if water pools within the WP. Unsaturated drip tests, in which the fuel is exposed to 100 percent relative humidity and various amounts of injected simulated groundwater, provide information on the degradation of the fuel in WPs that do not have pooling of water. The batch and unsaturated tests also give empirical information on secondary phase production, colloids, and effective solubility limits for the more insoluble radionuclides.

From the many experiments performed, an understanding of the mechanisms of spent fuel dissolution has emerged. Commercial nuclear fuel is composed primarily of uranium dioxide which, when exposed to mildly oxidizing solutions, is oxidatively dissolved to uranyl ion, UO_2^{+2}. The rate of UO_2 oxidation depends on the interaction of specific surface species that control the rate-determining dissolution step. The increase in CSNF degradation rates with decreasing pH (at low pH), and with increasing carbonate levels (at high pH), suggests that adsorbed protons and/or carbonate ions control the dissolution reaction under flow-through conditions. Indeed, Grandstaff (1976) showed that the uraninite dissolution rate dependence on aqueous carbonate concentrations could be explained by using a Langmuir adsorption isotherm relating coverage of the surface by carbonate ions to carbonate solution concentrations.

Important aqueous species that might also affect dissolution rates are calcium and silicon ions, which can form stable corrosion products possessing low solubilities. Electrochemical studies have shown fast reduction in dissolution currents when thin layers of corrosion products form, but an increase of UO_2 reactivity as the applied potential is increased (Shoesmith 1999, Section 3.1.1). At very low applied potentials (< –400 mV vs. saturated calomel) very slow oxidation begins (possibly concentrating at grain boundaries); up to –100 mV, dissolution becomes discernible. As the applied potential increases to +300 mV oxidation, dissolution and corrosion product formation are significant; above +300 mV dissolution is rapid because corrosion products are not formed on the solid surface. The likely electrochemical reaction sequence is:

$$UO_2 \Rightarrow UO_{2+x} \overset{\text{surface oxidation}}{\Rightarrow} UO_{2.33} \overset{\text{bulk oxidation}}{\Rightarrow} \{UO_2^{2+}\}_{surface} \overset{\text{dissolution}}{\Rightarrow} [UO_2^{2+}]_{bulk} \Rightarrow [UO_2^{2+}]_{soln} \Rightarrow \text{Secondary Phases}$$

The intrinsic dissolution rates of UO_{2+x} and a number of spent fuels were determined by using a single-pass flow-through reactor that allows UO_2 dissolution to be measured far from solution saturation (no precipitation of dissolved products). Note that the forward dissolution rate bounds the release rate, the latter being the material loss rate from the fuel + alteration products. Tables 3.3-1 and 3.3-2 show the tests and results from which the dissolution model for alkaline conditions (pH > 7) were regressed (CRWMS M&O 2000h, Equation 16):

$$\text{Log Rate} = 4.69 + [-1085/T] - 0.12 \cdot p[CO_3]_T - 0.32 \cdot pO_2 \qquad \text{(Eq. 3.3-1)}$$

Rate has units of $mg/m^2 \cdot day$. T is temperature (kelvin); p denotes the negative logarithm of either the carbonate concentration (mol/L) or the oxygen partial pressure (atm).

Table 3.3-1. Test Parameters and Dissolution Measurement Results for YMP Spent-Fuel Studies

Run	BU[a]	LBU	T (°C)	T (K)	1/T (K)	Total CO_3	$p[CO_3]_T$	O_2	pO_2	$[H^+]$ (mol/L))	pH	DR	LDR
1	30	1.48	50	323	3.09E–03	0.002	2.70	0.2	0.7	1E–09	9	6.34	0.802
2	30	1.48	50	323	3.09E–03	0.002	2.70	0.2	0.7	1E–09	9	7.05	0.848
3	30	1.48	50	323	3.09E–03	0.002	2.70	0.2	0.7	1E–09	9	5.07	0.705
4	30	1.48	22	295	3.39E–03	0.020	1.70	0.2	0.7	1E–08	8	3.45	0.538
5	30	1.48	74	347	2.88E–03	0.020	1.70	0.2	0.7	1E–10	10	14.20	1.152
6	30	1.48	74	347	2.88E–03	0.0002	3.70	0.2	0.7	1E–08	8	8.60	0.934
7	30	1.48	21	294	3.40E–03	0.0002	3.70	0.2	0.7	1E–10	10	0.63	–0.201
8	30	1.48	22	295	3.39E–03	0.02	1.70	0.2	0.7	1E–09	9	2.83	0.452
9	30	1.48	22	295	3.39E–03	0.002	2.70	0.2	0.7	1E–10	10	2.04	0.310
10	30	1.48	27	300	3.33E–03	0.0002	3.70	0.02	1.7	1E–08	8	1.79	0.253
11	30	1.48	78	351	2.85E–03	0.0002	3.70	0.02	1.7	1E–10	10	1.49	0.173
12	30	1.48	25	298	3.35E–03	0.02	1.70	0.02	1.7	1E–10	10	2.05	0.312
13	30	1.48	77	350	2.86E–03	0.02	1.70	0.02	1.7	1E–08	8	2.89	0.461
14	30	1.48	23	296	3.38E–03	0.02	1.70	0.003	2.5	1E–08	8	2.83	0.452
15	30	1.48	74	347	2.88E–03	0.02	1.70	0.003	2.5	1E–10	10	0.69	–0.16
16	30	1.48	78	351	2.85E–03	0.0002	3.70	0.003	2.5	1E–08	8	1.98	0.297
17	30	1.48	19	292	3.42E–03	0.0002	3.70	0.003	2.5	1E–10	10	0.51	–0.29
18	30	1.48	50	323	3.09E–03	0.02	1.70	0.003	2.5	1E–10	10	1.04	0.017
19	30	1.48	21	294	3.40E–03	0.002	2.70	0.003	2.5	1E–09	9	1.87	0.272
20	30	1.48	75	347	2.88E–03	0.02	1.70	0.02	1.7	1E–10	10	4.75	0.677
21	31	1.49	50	323	3.10E–03	0.002	2.70	0.2	0.7	1E–09	9	6.60	0.82
22	50	1.70	25	298	3.39E–03	0.02	1.70	0.2	0.7	1E–08	8	1.50	0.18
23	31	1.49	25	298	3.36E–03	0.02	1.70	0.2	0.7	1E–08	8	4.00	0.60
24	31	1.49	75	348	2.87E–03	0.02	1.70	0.2	0.7	1E–08	8	9.10	0.96
25	31	1.49	25	298	3.36E–03	0.0002	3.70	0.2	0.7	1E–08	8	2.60	0.41
26	31	1.49	75	348	2.87E–03	0.0002	3.70	0.2	0.7	1E–08	8	11.00	1.04
27	44	1.64	25	298	3.36E–03	0.02	1.70	0.2	0.7	1E–08	8	3.50	0.54
28	50	1.70	25	298	3.36E–03	0.02	1.70	0.2	0.7	1E–08	8	3.80	0.58
29	50	1.70	75	348	2.87E–03	0.02	1.70	0.2	0.7	1E–08	8	6.90	0.84
30	50	1.70	25	298	3.36E–03	0.0002	3.70	0.2	0.7	1E–08	8	2.90	0.46
31	50	1.70	75	348	2.87E–03	0.0002	3.70	0.2	0.7	1E–08	8	9.50	0.98
32	50	1.70	25	298	3.36E–03	0.02	1.70	0.002	2.7	1E–08	8	4.1	0.61
33	50	1.70	75	348	2.87E–03	0.02	1.70	0.002	2.7	1E–08	8	1.4	0.15
34	50	1.70	25	298	3.36E–03	0.0002	3.70	0.002	2.7	1E–08	8	1.9	0.28
35	50	1.70	75	348	2.87E–03	0.0002	3.70	0.002	2.7	1E–08	8	3.5	0.54
36	15	1.18	25	298	3.36E–03	0.02	1.70	0.20	0.7	1E–08	8	3.2	0.51

Table 3.3-1. Test Parameters and Dissolution Measurement Results for YMP Spent-Fuel Studies (Continued)

37	15	1.18	75	348	2.87E–03	0.02	1.70	0.20	0.7	1E–08	8	11.9	1.08
38	15	1.18	25	298	3.36E–03	0.0002	3.70	0.20	0.7	1E–08	8	3.7	0.57

Source: *CSNF Waste Form Degradation: Summary Abstraction*. ANL-EBS-MD-00005 (CRWMS M&O 2000h, Table 1). The source document lists four additional runs; since they were not used for model development, they are not included here.

NOTES: [a]BU = burnup MWd/kgU, LBU = $\log_{10}$ (BU), T = temperature,
Total CO_3 = [HCO_3^-] + [CO_3^{-2}] in molar, $p[CO_3]_T = -\log_{10}$ (Total CO_3)
O_2 = oxygen pressure in atmospheres, $pO_2 = -\log_{10}(O_2)$
DR = dissolution rate in mg/(m^2·day), LDR = $\log_{10}$ (DR)

Table 3.3-2. Test Parameters and Dissolution Measurement Results for YMP UO_2 Studies

Run	BU[a]	LBU	T (°C)	T (K)	1/T (K)	Total CO_3	$p[CO_3]_T$	O_2	pO_2	$[H^+]$ (mol/L)	pH	DR	LDR
39	0	b	50	323	3.09E–03	0.002	2.70	0.02	1.7	1E–09	9	12.30	1.090
40	0	b	50	323	3.09E–03	0.002	2.70	0.02	1.7	1E–09	9	7.96	0.901
41	0	b	50	323	3.09E–03	0.002	2.70	0.02	1.7	1E–09	9	10.4	1.015
42	0	b	25	298	3.35E–03	0.02	1.70	0.2	0.7	1E–08	8	2.42	0.384
43	0	b	75	348	2.87E–03	0.02	1.70	0.2	0.7	1E–10	10	77.38	1.889
44	0	b	75	348	2.87E–03	0.0002	3.70	0.2	0.7	1E–08	8	10.9	1.036
45	0	b	25	298	3.35E–03	0.0002	3.70	0.2	0.7	1E–10	10	2.55	0.407
46	0	b	25	298	3.35E–03	0.02	1.70	0.002	2.7	1E–08	8	0.22	–0.666
47	0	b	75	348	2.87E–03	0.02	1.70	0.002	2.7	1E–10	10	5.61	0.749
48	0	b	75	348	2.87E–03	0.0002	3.70	0.002	2.7	1E–08	8	0.51	–0.292
49	0	b	26	299	3.34E–03	0.0002	3.70	0.002	2.7	1E–10	10	0.23	–0.633
50	0	b	26	299	3.34E–03	0.0002	3.70	0.02	1.7	1E–08	8	0.12	–0.922
51	0	b	75	348	2.87E–03	0.0002	3.70	0.02	1.7	1E–10	10	9.21	0.964
52	0	b	26	299	3.34E–03	0.02	1.70	0.02	1.7	1E–10	10	1.87	0.272
53	0	b	75	348	2.87E–03	0.02	1.70	0.02	1.7	1E–08	8	5.11	0.709
54	0	b	50	323	3.09E–03	0.02	1.70	0.002	2.7	1E–10	10	4.60	0.663
55	0	b	25	298	3.35E–03	0.02	1.70	0.2	0.7	1E–09	9	6.72	0.827
56	0	b	25	298	3.35E–03	0.002	2.70	0.2	0.7	1E–10	10	9.34	0.970
57	0	b	26	299	3.34E–03	0.002	2.70	0.002	2.7	1E–09	9	1.52	0.180
58	0	b	75	348	2.87E–03	0.0002	3.70	0.2	0.7	1E–10	10	6.48	0.812
59	0	b	75	348	2.87E–03	0.002	2.70	0.2	0.7	1E–09	9	23.3	1.367
60	0	b	75	348	2.87E–03	0.02	1.70	0.2	0.7	1E–08	8	54.0	1.700

Source: *CSNF Waste Form Degradation: Summary Abstraction*. ANL-EBS-MD-000015. (CRWMS M&O 2000h, Table 1)

NOTES: [a]BU = burnup MWd/kgU. LBU = $\log_{10}$ (BU),
[b] $\log_{10}$ (0) is undefined, a LBU of 0 is used for modeling UO_2.
T = temperature, Total CO_3 = [HCO_3^-] + [$CO_3^=$] in molar, $p[CO_3]_T = -\log_{10}$ (Total CO_3)
O_2 = oxygen pressure in atmospheres, $pO_2 = -\log_{10}(O_2)$
DR = dissolution rate in mg/(m^2·day), LDR = $\log_{10}$ (DR)

To construct the abstracted acid dissolution model, the temperature and oxygen rate dependencies derived under alkaline conditions were assumed to hold for acid conditions. The pH 7 alkaline rate was then assumed to be common to the alkaline and acid pH rate laws (within the uncertainty of both rate laws). A qualified data point (pH = 3) was used to define the low pH end of the acid pH rate law. The latter, which describes rates at pH ≤ 7, is given below (CRWMS M&O 2000h, Equation 18):

$$\text{Log Rate} = 7.13 + [-1085/T] - 0.32 \cdot pO_2 - 0.41 \cdot pH \qquad \text{(Eq. 3.3-2)}$$

The combined abstracted model is shown in Figure 3.3-3.

As discussed in the AMR (CRWMS M&O 2000h, Section 6.2.2.2), pH dependence is weak under alkaline conditions and rate and pH are controlled by carbonate and oxygen. Under acidic conditions pH is a more significant factor, while $p[CO_3]_T$ is less significant as (H^+) protons dominate the surface reaction sites.

3.3.1.1 Gap and Grain Boundary Inventory

As summarized in CRWMS M&O (2000h, Section 6.5) some fission-produced radionuclides, ^{14}C, ^{135}Cs, ^{137}Cs, ^{129}I, ^{99}Tc, and ^{79}Se, migrate from the UO_2 matrix of light water reactor (LWR) spent fuels at the high fuel temperatures of reactor operation and deposit onto the cooler grain boundaries and fuel/cladding gap surfaces. At waste package and cladding breach, these radionuclides may be released faster than those within the fuel matrix. Volatile cesium and iodine, in addition to the fission gases, are the most conspicuous elements in this category. Recent performance assessments (CRWMS M&O 1998b, Section 6.3.1.3.1) of the potential repository at Yucca Mountain used gap and grain-boundary fractions of 2 percent of the total inventories of ^{135}Cs, ^{137}Cs, ^{129}I, and ^{99}Tc. The gap fraction was modeled to dissolve rapidly if the spent fuel were to be contacted by groundwater (CRWMS M&O 1998b, Section 6.3.1.3.2). Laboratory measurements of a few LWR spent fuels show that both the gap and grain boundary inventories of ^{99}Tc and ^{90}Sr were near the detection limits of the methods used, less than 0.2 percent of the total inventories of these elements. However, some of the ^{99}Tc may reside at the grain boundaries in the form of relatively insoluble metallic particles and not be detected by these tests. Measured combined gap and grain-boundary inventories of ^{129}I approximately equal the fission gas release fractions. For ^{137}Cs, the combined gap and grain-boundary inventories were approximately one third of the fission gas release fractions. For the TSPA-SR analysis, the fast release fraction consists of an immediate release of 4.2 percent of the total iodine inventory, and 1.4 percent of the cesium inventory. In addition, 0.2 percent (range 0 to 0.4 percent, uniformly distributed) of all the radionuclides including additional cesium and iodine is immediately released from the grain boundaries and fuel and gap surfaces.

3.3.2 Confidence/Limitations/Validation

The abstracted CSNF dissolution model was based on a large set of qualified flow-through experiments. It is valid from pH 3 to 10, oxygen pressure from 0.002 to 0.2 atmospheres, and total carbon concentrations from 2×10^{-4} to 2×10^{-2} molar. At pHs less than or equal to 7, this model is valid at CO_2 pressures of 10^{-3} atmospheres. From an analysis of the fit of the model to the data, and from further consideration of the uncertainty of application of data from young

spent fuel (< 30 years out of reactor) and unburned UO_2 toward the prediction of long term (> 1,000 year) performance of spent fuel, the model was estimated to be valid to within 1.5 orders of magnitude. This model was compared to unsaturated drip tests, batch tests, and a range of literature results. The model and uncertainty range adequately accounted for, or overestimated, all dissolution rate data. In addition, a comparison of the phases produced in the unsaturated drip tests compare well with that of natural analogs.

Based on the considerations discussed below, this model is appropriate for its intended use.

3.3.2.1 Comparison to Unsaturated Drip Tests

Long-term drip testing of CSNF under hydrologically unsaturated conditions, i.e., limited water and an oxidizing atmosphere, has been done over the past six years to determine the relationship between the rate of CSNF alteration, i.e., dissolution and secondary phase formation, and the release rate of radionuclides. Small samples of two commercial PWR spent nuclear fuels, Approved Testing Material-103 and Approved Testing Material-106, are subjected to three experimental conditions: vapor injection, low-drip-rate injection, and high-drip-rate injection, all at 90°C (CRWMS M&O 2000h, Section 6.3). All drip samples were completely wetted. The nominal drip rate is 0.75 mL every 3.5 days in the high-drip-rate tests and 0.075 mL every 3.5 days in the low-drip-rate-tests. In the vapor tests, minimum water is available for transport, and the absence of added cations and anions limits the type of alteration products (secondary phases) that may form. A CSNF intrinsic dissolution/corrosion rate is estimated from the release of highly soluble ^{99}Tc and ^{90}Sr which are assumed to be homogeneously distributed in the spent fuel matrix (^{129}I and ^{137}Cs are concentrated at the fuel grain boundaries and fuel-clad gap).

In the high-drip-rate tests iodine release rates decreased about an order of magnitude after 1.3 years of reaction. Tc release remained relatively constant over time and appears to depend on the initial burnup of the fuel. Less than 1 percent of the reacted uranium was released into solution after 1.6 years. Most of the neptunium and plutonium release occurred in the first year. Thereafter, nearly all of the ^{237}Np (96.5 - 99 percent) and ^{239}Pu (99.5 - 99.6 percent) was retained. Transmission-electron microscope examination (Finn et. al. 1998) indicates that the Pu is retained as a residue at the fuel surface. Of the 0.01 percent of the Pu that is released into the leachate, greater than 50 percent is sorbed on the walls of the stainless steel vessel (the remainder is colloidal). After 4.8 years of reaction, ^{129}I and ^{99}Tc release rates are similar.

In the low-drip-rate tests, ^{99}Tc release rates increased by 2.5 orders of magnitude when the fuel was immersed in leachate at 3.1 years, suggesting that the ^{99}Tc that was originally dissolved had become loosely bound to the corrosion products before immersion. Apparently pertechnetate, TcO_4^-, precipitates as a soluble salt as the liquid in the low-drip-rate test becomes concentrated. When excess water is introduced as in an immersion test, the salts dissolve and Tc, Cs, and Sr are released.

In the vapor hydration tests, material transport may occur by one of two pathways: thin-film flow or spallation of unwashed fuel fines. Radioisotope release rates were observed to vary substantially between the different fuel types that were considered. The cause of this remains unclear.

If a roughness factor (the ratio of the measured surface area to the geometrically estimated surface area) of three is assumed, the high-drip-rate dissolution rates are estimated to be 17 and 38 mg/(m^2·d), compared against a predicted value of 22 mg/(m^2·d) at pH 8, 0.001 mol/L carbonate, and 90°C for a fuel with a burnup of 30 MWd/kgU. Therefore, the agreement between the rate model above and ^{99}Tc release rates is good. The calculations for the other isotopes show reduced release rates because the isotopes are retained in corrosion products.

3.3.2.2 Comparison to Batch Tests and Literature

The rate models can be compared against batch tests, published data cited in reviews by Grambow (1989) and McKenzie (1992) of pre-1992 literature, as well as more recent, flow-through measurements of de Pablo et al. (1997), Torrero et al. (1997), and Tait and Luht (1997). Batch dissolution rates are lower than predicted rates but within a factor of 6. The rate model above does not have as strong a carbonate dependency as that seen by de Pablo et al.'s (1997) study. The alkaline pH model predicts 0.0002 mol/L total carbonate rates that are consistently higher than Torrero et al.'s (1997) carbonate-free rates. The difference in carbonate levels though makes direct comparison impossible. Torrero's acidic data can be as low as 1/20th of the predicted value, yet Torrero's acidic data overall seem low compared to other work (see Grambow 1989). In general, the present model predicts Tait and Luht's (1997) results well at the various sample, temperature, oxygen, and water-chemistry conditions. Moreover, the trends in the model and data with temperature, carbonate, and oxygen are the same. The models underpredict Tait and Luht's (1997) data at bounding conditions, such as 75°C, and 0.1 mol/L bicarbonate by at most 0.88 orders of magnitude, which is within the uncertainty range of the model.

3.3.2.3 Natural Analogs

To establish confidence that experimental CSNF alteration pathways effectively represent likely natural processes, it is useful to compare the alteration mineralogy observed in experiments with natural occurrences of altered uranium oxides. Uranium dioxide occurs in nature as the mineral uraninite which is structurally similar to CSNF. Nopal I, a uranium mining site at Peña Blanca, Mexico, contains substantial quantities of uraninite and is arguably one of the best natural analogs for CSNF degradation in Yucca Mountain as it possesses geologic, geochemical, and hydrogeologic characteristics most similar to those at Yucca Mountain (Murphy 1995). The volcanic (tuffaceous) host rock at Nopal I, the youngest of the studied sites, has been relatively oxidizing for tens of thousands of year, though uraninite, containing U^{4+}, was originally formed several million years ago.

In general, uraninite has been oxidized and hydrated at Nopal I. In the presence of silicate and alkali ions, such as calcium and sodium, various alkaline uranyl silicate hydrates such as Na-boltwoodite and β-uranophane dominates alteration mineralogy at long times.

In the vapor tests a relatively simple combination of uranyl oxy-hydroxide alteration phases is observed–dehydrated schoepite $(UO_2)O_{0.25-x}(OH)_{1.5+2x}$ ($0 \leq x \leq 0.15$) and metaschoepite (CRWMS M&O 2000h, Section 6.6.2. (A minor phase is Cs-Ba-Mo-uranate, which incorporates two fission products, cesium and molybdenum.) Sodium and silicon are among the most abundant elements in J-13 well water, and, not surprisingly, the most abundant alteration product

in the high-drip rate tests is the Na- and Si-bearing U^{6+} compound Na-boltwoodite; the Ca- and Si-bearing U^{6+} compound β-uranophane has also been identified in solid samples from these tests. The higher-drip-rate tests exhibit two uranophane-group silicates, β-uranophane [$Ca(UO_2)_2(SiO_3OH)_2(H_2O)_5$] and Na-boltwoodite $(Na,K)(UO_2)(SiO_3OH)(H_2O)$. Samples from low-drip-rate tests include metaschoepite, a Na-uranyl oxy-hydroxide tentatively identified as Na-compreignacite, and soddyite. Again, Cs-Ba-Mo-uranate is a minor constituent. In one sample, soddyite appears to replace Na-compreignacite. Also, a few isolated crystals of Na-boltwoodite were first detected in one low-drip-rate test. The groundwater at Nopal I is richer in calcium than J-13 (Pearcy et al. 1994) but poorer in sodium and potassium. This could explain the dominance of β-uranophane at the natural site as well as the limited soddyite and weeksite occurrence. There is substantial calcite at Yucca Mountain. In time this may make repository alteration products conform more to the Nopal I sequence than that seen in the laboratory, which produces β-uranophane at long times. Overall, the phase assemblage observed at Nopal I is similar to that derived experimentally in the CSNF alteration drip tests.

The general agreement between the observed alteration products in the various tests, the natural analogs, and the geochemical modeling provide confidence that the mechanisms of spent fuel corrosion are well understood, and that the forward dissolution model is bounding for long term prediction of CSNF degradation rates.

3.3.3 Other Views

The TSPA Peer Review Panel Report (Budnitz et al. 1999, Executive Summary, Section E, Data and Research Needs) stressed the importance of advancing the spent fuel corrosion model beyond its VA representation, stating that a considerable knowledge of spent fuel corrosion is needed. It noted that the databases used in the VA for the corrosion of spent fuel were not adequate (Budnitz et al. 1999, Section II.A). It also noted that the processes that ensue among the WP internals following penetration of the package were not addressed in any depth within the TSPA-VA (Budnitz et al. 1999, Section II.F). The panel also questions the conservatism of the related base case conditions. It suggested that steps be taken to develop better methods for analyzing the movement of radionuclides from the WPs. Substantial discussion was dedicated to discussion of weaknesses in the VA spent fuel corrosion model (Budnitz et al. 1999, Section IV.F).

Weaknesses in the VA spent fuel corrosion model and documentation were identified by the peer review panel. These were addressed in the development of the SR model and documentation as discussed in Section 3.3 and CRWMS M&O 2000h. In particular, the mechanistic basis was discussed, a simplified model was developed, and model validation using an expanded database was performed. Removing the conservatism of modeling of water-waste contact and radionuclide transport within the package was not given priority for SR, because in-package radionuclide transport was not determined to be a principal factor for the safety case. However, the cladding degradation and the waste matrix degradation have been coupled as explained in Section 3.4 and the degradation of the waste matrix was also coupled to the in-package chemistry to better characterize uncertainty as explained in Sections 3.2 and 3.3.

3.4 CSNF CLADDING DEGRADATION COMPONENT

3.4.1 Introduction

Since the 1950s, most CSNF has been clad with less than 1 mm (usually between 600 through 900 μm) of Zircaloy, an alloy that is about 98 percent zirconium with small amounts of tin, iron, nickel, and chromium. The Zircaloy cladding is not a designed engineered barrier of the Yucca Mountain disposal system, but rather is an existing characteristic of the CSNF that is important to determining the rate of release of radionuclides once engineered barriers such as the waste packaged have breached. Zircaloy is very resistant to corrosion, and cladding failure is expected to be minimal in the first 10,000 years. However, while Zircaloy provides excellent protective properties, characterization of the uncertainty in its performance is important. This characterization is possible since data have been collected on its performance over the past 40 years by the nuclear industry and by others in several different harsh environments.

3.4.1.1 Function of Cladding Degradation Component

The cladding degradation component of the Waste Form Degradation Model predicts the rate that the CSNF matrix is exposed and altered based on the number of rods with perforated cladding at any one time.

3.4.1.2 Summary of Conceptual Model of CSNF Cladding Degradation

The degradation of CSNF cladding is assumed to proceed through two distinct steps: (1) rod failure (perforation of the cladding) and (2) progressive exposure of UO_2 spent fuel matrix (see Figure 3.4-1). The number of failed rods is based on the observed rod failures while in the reactor and during storage, potential creep failures because of high disposal temperatures, stress corrosion cracking from high stresses and the presence of halide ions, hypothetical physical failures from seismic loads, and hypothetical localized corrosion inside the WP. In all cases, the perforation is assumed to occur at the center of the rod because this ensures the fastest exposure of the waste matrix. After the initial perforation, further cracking and splitting of the cladding is assumed to occur, which exposes the CSNF matrix as it progresses. The splitting of the cladding (unzipping) is calculated as a function of the CSNF matrix alteration rate, which was described in the previous section, Section 3.3.

3.4.1.3 Modes for Initiating Cladding Perforations

Cladding perforation is the formation of small cracks or holes in the cladding. Perforation of the cladding may occur because (1) the cladding initially fails within the reactor or during storage, (2) the cladding fails from creep or stress corrosion cracking, (3) an earthquake severely shakes and severs the rods, or (4) the cladding fails from localized corrosion (Figure 3.4-2). While other mechanisms of initiating cladding perforations such as diffusion controlled cavity growth (DCCG) or DHC were explored, they were screened out based on low consequence as described later.

3.4.1.4 Cladding Failure after Perforation

After the cladding has been perforated, the fuel matrix can react with moisture or air. The reaction is assumed to unzip the fuel rod because the fuel volume increases during oxidation of the UO_2, (i.e., the oxidation of the UO_2 to secondary phases of uranium is assumed to cause enough pressure by volume expansion to burst the cladding from within) (Figure 3.4-2). This mechanism for increasing the exposed surface area of the matrix is sensitive to environmental conditions such as temperature. Based on observed behavior within storage pools at reactors, unzipping in a wet environment does not occur in observed time periods of 40 years, once the Zircaloy cladding has been perforated. However, because unzipping in a wet environment could not be entirely ruled out and because complete exposure of the matrix bounds diffusive releases of radionuclides through the pinhole perforations, the project included the possibility of the clad unzipping in a wet environment (Figure 3.4-2). A small percentage of the inventory of iodine, cesium, and a few other radionuclides resides in the gap between the cladding and waste matrix. The release of the gap inventory is assumed to be instantaneous when the cladding is perforated and, thus, independent of the cladding unzipping. Cladding that was initially perforated is predicted to release its gap inventory when the WP is breached.

3.4.1.5 Documents Supporting Model Development

To carefully examine and describe the uncertainty about the expected protective capability of the cladding, eight AMRs and two calculations were prepared. One AMR dealt strictly with FEPs related to cladding to more carefully focus attention on hypothetical failure modes. Three AMRs dealt with specific modes of potentially perforating the cladding, an AMR and calculation dealt with evaluating temperatures of the cladding for creep calculations, and two AMRs dealt with further exposure of the cladding through unzipping (Figure 3.4-3).

3.4.2 Description

3.4.2.1 Initial Cladding Condition

3.4.2.1.1 Initial Cladding Perforations

Two aspects of initial cladding condition are important (Figure 3.4-2). The first aspect is the number of rods of CSNF that arrive for disposal with cladding perforations and is discussed in this subsection. The second aspect is the condition and characteristics of the unperforated cladding (e.g., the internal pressure and strain history of the cladding) and is discussed in the next subsection. Because of the propensity of stainless steel to stress corrosion crack (SCC), all of the stainless steel cladding (~1.1 percent of the CSNF) is considered as a separate category of CSNF and is assumed to be initially perforated. In the Zircaloy cladding category, the percent of rods arriving at the repository with perforations is dependent on the number of rods that perforate while (a) in the reactor (including fuel handling), (b) in pool storage or dry storage (including fuel handling), and (c) in transportation from storage. Data exist for all three aspects. Based on the data, the largest percentage of rods perforate when first placed in the harsh reactor environment (CRWMS M&O 2000p, Section 7, Table 13) (Table 3.4-1). Few perforations occur in the second and third cycles of the reactor operation or during storage and transportation. Some data for reactor perforations extend over the operating history of the commercial nuclear

industry. The data for perforations used for TSPA-SR span the period 1969 through 1995. As described in CRWMS M&O 2000p, between 1969 and 1985, a contractor report for NRC is used; between 1985 and 1995 updates as reported in the literature are used. The data on perforations are based on ~65,000 BWR assemblies with ~4 million rods and ~47,000 PWR assemblies with ~10 million rods (CRWMS M&O 2000p, Attachment I).

Currently most fuel is stored in pools at reactors but in the future much may be placed in dry storage. Of these two methods of storage, dry storage has higher temperatures and thus greater propensity to cause perforations. While not extensive, data have been collected and estimates made on future cladding perforations while in dry storage in studies done by the DOE at the INEEL and Pacific Northwest National Laboratory. Cladding failures during transportation have also been estimated in order to license transportation casks for the NRC (CRWMS M&O 2000p, Section 6.11).

Table 3.4-1. Percent and Cause of Rods Failed in a Waste Package

Rod Perforation Mode	Percent of Rods Perforated/WP
Reactor Operation including handling perforations (mean)	0.109; Range: 0.0 to 5.23
Pool Storage	0.0
Dry Storage (including handling)	0.045
Dry Storage & Transportation from DHC	0.0
Transportation (vibration, impact)	0.01
Total	**0.164; Range: 0.055 to 5.28**

Source: *Initial Cladding Condition*. ANL-EBS-MD-000048. (CRWMS M&O 2000p, Table 13).

To include uncertainty in TSPA-SR, a distribution of initial rod perforations in a WP, expressed as a complementary cumulative distribution function (CCDF), was developed from the available data on cladding perforations. When developing the distribution of the perforations per WP, however, the type of blending assumed in the WP can be important since the performance of cladding has greatly improved over the years; in the mid-1970s, Zircaloy cladding perforations were much greater (still less than 5 percent) than observed today (less than 0.1 percent). The SNF of the 1970s has lower initial burnup and has also cooled longer and so this CSNF fuel will likely be blended with newer, hotter CSNF of today to keep the waste packages at a more uniform temperature.

Conservatively assuming, however, that no blending occurs, a CCDF can be developed that is the sum of the CCDF of Zircaloy cladding perforations during reactor operations and the perforations during storage and transportation. The small number of perforations that occur during storage and transportation (Table 3.4-1) does not have the ability to significantly affect the shape of the distribution for initial perforations; consequently, no ranges or distributions were assigned to the data on storage and transportation. A reasonable upper bound to this CCDF can be developed by assuming that whenever a rod was damaged (say from handling), all four rods in a square cluster were damaged. The distribution was assumed to be symmetric and so the lower bound was developed by dividing by 4 (Figure 3.4-4).

Potential perforation from creep rupture or stress corrosion cracking is not included as part of the initial perforation in Table 3.4-1. Rather this was treated as incipient perforation information and

became part of the initial condition of unperforated cladding discussed in the next subsection. The creep rupture analysis was integrated because the rod damage from one period must be carried forward to the next temperature period to calculate the total strain and thus potential for creep rupture. Stress corrosion cracking is also addressed in the same manner because the same highly stressed rods are susceptible to failure from either mechanism.

3.4.2.1.2 Initial Condition ("Pressure and Strain") of Unperforated Cladding

The fact that few cladding perforations occur after the first cycle in a reactor suggests that there are few rods almost ready to perforate (i.e., "incipient" perforations). Nonetheless, in order to analyze the potential perforation of the Zircaloy cladding from creep rupture or stress corrosion cracking after disposal, the initial condition of the cladding (e.g., internal pressure and strain history) was also estimated and uncertainty included (Figure 3.4-2) (CRWMS M&O 2000p, Section 6.7). Table 3.4-2 summarizes the distributions of the various properties developed for TSPA-SR. The CCDFs for the various properties are not independent; consequently, they were generated using a single burnup distribution based on PWR cladding to maintain consistency. PWR cladding was modeled rather than BWR cladding because BWR cladding is thicker, BWR fuel is enclosed in flow channels which offer additional protection to the cladding, and BWR fuel is discharged with lower burnup and thus lower cladding stress.

Table 3.4-2. CCDFs Describing Expected Fuel Stream into YMP

Property	Mean Value	5 Percent CCDF Value
Burnup	44.1 MWd/kgU	63.3 MWd/kgU
Helium fill Pressure	3.71 MPa	4.99 MPa
Fission Gas Release	4.2%	10.8%
Free Volume	17.7 cm^3	14.1 cm^3
Internal Pressure	4.8 MPa	7.34 MPa
Oxide Thickness	54.2 µm	112 µm
Hydride Content	358 ppm	738 ppm
Crack Size	19 µm	57.1 µm
Cladding Thickness	522 µm	475 µm
Stress	38.4 MPa	61.8 MPa
Stress Intensity Factor (K_I)	0.47 $MPa \cdot m^{0.5}$	1.08 $MPa \cdot m^{0.5}$

Source: *Initial Cladding Condition* (CRWMS M&O 2000p, Table 12)

3.4.2.2 Temperature History of Cladding and Fuel Matrix

In order to calculate the strain for creep rupture and hoop stress for stress corrosion cracking, the temperature history during storage and transportation is needed. The analysis assumed all Zircaloy cladding was placed in dry storage at the design temperature of the Castor Mark V WP. Dry storage and the Castor Mark V package with 55 MWd/kgU burnup fuel was selected to be conservative. For transportation, the cladding was conservatively assumed to be placed in a shipping cask for 3 weeks at a temperature of 350°C. This is conservative since most rods will actually be exposed to lower temperatures for a shorter time frame.

In addition to the temperature history of the unperforated cladding prior to disposal, the temperature of the cladding during disposal is needed (Figure 3.4-2). (Only hoop stress is considered since thermal stresses from differences in temperature are negligible because of the uniform temperatures in the WPs.) For TSPA-SR, a functional relationship between average surface temperature of the waste package and the temperature of the cladding was developed (Figure 3.4-5); the functional relationship was based on a heat conduction model. Because the temperature decreases from the center to the outside of the waste package, the waste package was discretized into six zones and a separate temperature profile is developed for each zone. Evaluation of cladding and matrix temperatures from the waste package surface temperature (Figure 3.4-6) gives the cladding temperature for the average center rod in a WP. In the repository, the average center rod cladding temperature peaks at 308°C; there is a ±13 percent uncertainty distribution included. The uncertainty addresses both the uncertainty in WP internal temperature distribution and the uncertainty in predicting the WP surface temperature. In the statistical analysis of 12,000 fuel rods, the full range of temperature uncertainties is sampled. After 1,000 years the WP surface temperature and internal temperatures are practically identical and so the WP surface temperature is used. The calculation of internal temperatures is fairly independent of the design of the engineered barrier since the WP surface temperature is treated as an independent variable in this approach. Only the WP surface temperature is needed to correctly account for the engineered barrier design.

3.4.2.3 Creep Strain and Stress Corrosion Cracking Perforation

From the statistical expression of the initial internal pressures, temperature history during transportation and storage, and the temperatures during disposal, the perforations from creep and stress corrosion cracking are calculated in the CSNF Cladding Degradation Component (Figure 3.4-2).

This approach is recommended in both the Pescatore and Cowgill (1994, p. 85) review of creep analysis and a recent NRC Interim Staff Guidance for analysis of creep damage during dry storage (NRC 2000b, p. 85). It is the methodology used in Germany (Peehs 1998, Figures 11 to 13) for licensing dry storage designs. Uncertainties are established based on both experimental observations and analysis. This analysis is appropriate for the TSPA since it uses both accepted methodology, and sufficient uncertainties.

Murty's creep strain correlation (Henningson 1998, p. 57), with appropriate uncertainties, was used for creep rupture for TSPA-SR.

Table 3.4-3 compares six different creep rupture correlations to 503 data points reported in five different sets of experiments. It should be noted that the stresses and temperatures are appropriate for higher stressed rods in a hot repository. While the Matsuo correlation (Matsuo 1987, p. 23) has a slightly smaller relative error, Murty's correlation was selected because it explicitly models Coble creep, a type of creep that could be important at lower stresses and temperatures. The uncertainty in the creep correlation was set at ±0.80, which was the maximum relative error range for Murty's correlation. The uncertainty was assumed to be uniformly distributed. Both the data and correlations are for unirradiated cladding. Irradiated cladding has a creep rate of about 1/3 that of unirradiated material (Peehs 1998, Figure 10). Most of the data, correlations, and earlier creep analyses were for unirradiated cladding. The use of an

unirradiated creep correlation introduces a conservatism that could be removed with additional creep modeling.

Table 3.4-3. Comparison of Relative Error of Creep Correlations for All Data Points

		Creep Correlations					
Reference[b]	Total Data Points[a]	Matsuo	Murty	Mayuzumi	Limback	Spilker	Peehs
Spilker, T2	192	0.758	0.727	0.648	0.560	0.344	0.726
Spilker, T3	240	0.340	0.482	0.901	1.779	2.256	0.546
Matsuo	21	0.135	0.392	0.640	0.557	1.994	0.521
Mayuzumi	31	0.282	0.283	0.121	0.477	1.291	0.579
Limback	19	0.334	0.417	0.142	0.145	1.354	0.596
Average		0.487	0.557	0.717	1.121	1.431	0.606

Source: *Clad Degradation-Summary and Abstraction*. (CRWMS M&O 2000i, Table 3)

NOTE: Values are: Absolute Value ([Calculated–Measured]/Measured)
[a]Total number of points : 503
[b]Tests: unirradiated material, over the range 250 ≤ T (°C) ≤ 385 and 55 ≤ Stress (MPa) ≤ 120

A CCDF of the strain required for creep failure was constructed from a series of tests by Chung et al. (1987, pp. 780, 781). Chung et al. did a series of 20 slow burst and mandrel tests with irradiated BWR and PWR cladding. Half the tests were for over 200 hours. They measured an average strain at failure. The average failure strain was 3.3 percent with a range of 0.4 percent to 11.7 percent (18 data points reported). The lower values of this range are for BWR rods, which burst at lower strains. The creep-failure analysis covers the full range of observed failures. CRWMS M&O (2000p, Section 6.10.1), compares this failure criterion to four (4) different failure criteria developed by four different sources and shows that this criterion is conservative.

Figure 3.4-7 gives the CCDF for the hoop stress (at room temperature) in the rods expected to be received at YMP. A sampling of 2,000 rods with stresses that define this distribution is used for the stress variation. One rod (total of 12,000 rods analyzed, 2,000 rods located in each of 6 regions) with each of these stresses is assumed to be in each of the six regions across the WP. This rod represents a specific fraction of the total WP inventory. Each rod is exposed to a temperature profile represented by Figure 3.4-6 (including dry storage and shipping) and a repository temperature history normalized by a WP radial-shaping factor. A uniform temperature uncertainty of ±13.5 percent is included. The creep strain is then calculated for the rod using the equations given in the AMR for *Clad Degradation-Summary and Abstraction* (CRWMS M&O 2000i, Section 6.2.4) (including a uniform uncertainty of ±80 percent). This strain is compared to a random sampling of the strain limit CCDF. If the rod creep strain exceeds the strain limit, it is assumed that the fraction of rods represented by that radial zone in the WP has become perforated and is available for cladding unzipping. This procedure is repeated for 12,000 rods in an Excel spreadsheet and the fraction of the WP inventory perforated is calculated. In TSPA-SR, for a typical WP, the fraction of rods failed by creep ranges from 1.1 percent to 19 percent. The best estimate is 2.4 percent. A triangular distribution is sampled between these limits. These failures are predicted for dry storage and transportation.

The above analysis has not been directly incorporated into the TSPA, but an abstraction has been included. For this abstraction, the independent variable is the peak WP surface temperature. The above analysis is performed for peak WP surface temperatures that vary from 177°C to 412°C. The cladding temperature history while in the WP (right side of Figure 3.4-6) is linearly shifted upward or downward by the difference between the base case and new peak WP surface temperature. The change in temperature across the WP is then calculated.

Figure 3.4-8 is a graphical representation of the abstraction used in TSPA. The plateau at the lower WP surface temperatures represents the rods that are failed in dry storage and shipping. These rods can only fail once and, therefore, these rods cannot fail again in the repository. Rods that have not failed have undergone some creep during dry storage and shipping and start the additional creeping from that point. Figure 3.4-8 shows that at a peak WP surface temperature above about 300°C, the best estimate fraction of rods perforated from creep increases dramatically. The upper limit represents the fraction of rods failed if the failure criterion is 0.4 percent creep, the minimum value in the failure criteria CCDF. The minimum values represent the fraction of rods that would fail if the maximum failure criterion of 11.7 percent were applied. The best estimate value represents a random sampling of the failure criteria CCDF. In the TSPA, a triangular distribution is used between the upper limit, best estimate and lower limit in each realization to incorporate uncertainties in the WP peak surface temperature.

Stress corrosion cracking (SCC) is considered along with creep rupture as a perforation mechanism since both require evaluating cladding stresses (Figure 3.4-2) (CRWMS M&O 2000i, Section 6.2.4). SCC requires a susceptible material, an aggressive chemical environment, and a high stress level (CRWMS M&O 2000ag, Section 6.1.11). Zircaloy is not susceptible to SCC in NaCl, HCl, $MgCl_2$, and H_2S solutions (Yau 1992, p. 299-311). The environments known to cause SCC in zirconium include $FeCl_3$, $CuCl_2$, halogen or halide-containing methanol, concentrated HNO_3, liquid mercury or cesium (Yau 1992, p. 303), and 64 to 69% H_2SO_4 (Fitzgerald and Yau 1993). The extremely oxidizing acidic conditions are not predicted for the bulk in-package chemistry but the possibility of such conditions occurring locally and transiently, are included in the local corrosion model. However, iodine has been identified by several investigators as a cause of SCC in Zircaloy. Iodine stress corrosion cracking (I-SCC) requires an iodine concentration in the fuel-cladding gap greater than 5×10^{-6} g/cm^2. Below this threshold of free iodine concentration, Zircaloy cracking due to I-SCC has not been observed. Free iodine concentrations are expected to be negligible in SNF. For the TSPA-SR it was conservatively assumed that a sufficient amount of iodine was present on the cladding, the stress was high, and that the duration of elevated temperatures was sufficiently long that once cracking started, there was sufficient time to propagate through the cladding. The gas pressure inside the cladding and the resulting stress were evaluated (CRWMS M&O 2000p, Section 6.7). It was assumed that any fuel rod with a stress above 180 MPa perforated by SCC. This threshold was reported by Tasooji et al. (1984, Figure 3, p. 600). Few rods have such high stresses and the same rods that fail from SCC are also prone to perforation from creep rupture. The calculations were combined, to avoid double counting perforated rods. The inclusion of SCC increases the failure from creep alone from 0.49 percent to 1.1 percent for dry storage and transportation (left side of Figure 3.4-6). At higher repository temperatures, the difference disappears.

3.4.2.4 Localized Corrosion Perforation

Local corrosion is also considered as a perforation mechanism in the CSNF Cladding Degradation Component (Figure 3.4-2). Corrosion of zirconium has been observed in concentrated fluoride or chloride solutions at very low pHs or very high oxidation potential (CRWMS M&O 2000ag, Section 4.1). These conditions are not predicted to occur in the bulk solution (see Section 3.2), but have not yet been ruled out for localized and/or non-equilibrium effects such as MIC, galvanic coupling, radiolysis in a humid environment, and extreme concentration by evaporation. Each of these mechanisms may locally depress the pH or increase the concentration of corrosive species such as fluoride or chloride, at least temporarily. None of these conditions are expected to impact cladding performance under repository conditions. However, until they can be ruled out, or shown to be too transitory to have negative consequences on cladding, a conservative model has been adopted to bound the uncertainty.

This conservative model uses two severe assumptions. The first assumption is that there is no filling or flushing of waste package by water. This is the worst case for concentration of aggressive species and breach of cladding. It is assumed that ground water entry occurs while there is still significant heat and/or radiation. The water flow rate must nearly match the evaporation rate within the package. If the flow is too high, the package will fill and overflow. The scenario of a filled, well-mixed, overflowing package is covered by the in-package chemistry model, which shows only moderate pH reduction (CRWMS M&O 2000g, Section 6.1). The scenario with flushing is the worst for transport of species out of the package, but is not the worst for cladding performance. Filling the package with water displaces the nitrogen that otherwise might be radiolytically converted to nitric acid. Overflow will flush out aggressive species, and prevent ground water from concentrating to the point of promoting galvanic corrosion of the carbon steel basket materials. Oxygen influx is also greatly reduced in a flooded waste package, so the corrosion potential and corrosion rates are reduced, as are many radionuclide solubilities.

The second assumption is that aggressive species concentrate in the worst possible way. Because fluoride and chloride are consumed by reaction with Zircaloy (CRWMS M&O 2000ag, Section 6.2.2.3.2) and carbon steel (McCright 1998, Section 2.1.8), respectively, it is assumed that aggressive species from incoming water will concentrate where they can do the most damage. For fluoride, this would be nearly total evaporation of incoming drops on a drop-width section of a single fuel rod. For chloride, this would be in a crevice between Zircaloy and carbon steel basket materials. It is assumed that the incoming water species are directed to a single rod, until they have breached that rod. Then it is assumed that new aggressive species are directed to another single rod until it is breached, and so on. In this approach, the fraction of cladding perforated is linearly dependent on the water inflow, which generally increases with time as the waste package degrades. Inflow, however, is also a function of other factors such as climate, location within the repository, etc. The likelihood of significant concentration of aggressive species decreases with time, as decay heat and radioactivity decrease, likelihood of good galvanic connection to the Zircaloy decreases, and amount of reduced iron as an energy source for microbes decreases. The trend toward less aggressive conditions is ignored, however, as it is assumed that the aggressive species concentrate in the worst possible way for all time.

Figure 3.4-9 shows an example: with 50 liters/year of water entering the WP, 20 percent of the rods would fail by this mechanism in 10,000 years of water flow.

3.4.2.5 Physical Perforation

The CSNF Cladding Degradation Component also considers physical perforation of the Zircaloy cladding from mechanical and seismic loads (Figure 3.4-2). Rubble (from the drift wall) falling onto the cladding when the WP collapses can perforate the cladding but was not modeled since WP collapse takes hundreds of thousands of years to occur. During this time, other modes of perforating the cladding (e.g., localized corrosion) are predicted to occur.

Studies of rock fall (CRWMS M&O 2000ah, Section 6) have concluded that credible rock falls will not be sufficient to make the drip shield contact the waste package. Therefore, rock falls will not cause cladding perforation. Even an earthquake so strong that it occurs only once in 100,000 years would not perforate the CSNF cladding. Only severe seismic events with a frequency of 1.1×10^{-6}/year can perforate the cladding and were used in CSNF Cladding Degradation Component. This event perforates all the cladding and makes it available for unzipping.

3.4.2.6 Other Perforation Mechanisms Evaluated (Features, Events, and Processes)

Other cladding degradation mechanisms were reviewed using FEPs screening arguments (CRWMS M&O 2000s) (see Section 2.3). The results are summarized below.

Hydride Failures–A review of the various hydride degradation mechanisms was performed for TSPA-SR (CRWMS M&O 2000p, Section 6.10.2). DHC of existing cracks was analyzed using the distribution of stresses and crack sizes summarized in Table 3.4-2. Stress intensity factors were calculated and have a mean of 0.44 $MPa \cdot m^{0.5}$ (range 0.002 to 2.7 $MPa \cdot m^{0.5}$); this intensity factor is below the usual threshold, which ranges from 5 to 12 $MPa \cdot m^{0.5}$. Therefore, crack propagation by DHC was not expected and was screened out based on low consequence. The calculated stress intensities are also below those needed to produce embrittlement failures ($K_I < K_{IC}$) and so embrittlement was screened out based on low consequence. Perforation of the cladding by hydride reorientation is also unlikely and has not been included in the abstraction for TSPA-SR because of the its inability to cause significant consequences. Stresses and temperatures are too low for reorientation to occur, and the material will maintain sufficient strength even if reorientation did occur, so failure would not be expected (CRWMS M&O 2000r, Section 6.4).

Hydride Embrittlement of Cladding is Unlikely–Hydrogen will be generated in the WP as the WP internals corrode. This hydrogen is not absorbed because the H_2 molecules do not migrate through the high-density zirconium oxide layer. Available data on zirconium hydriding indicate that corrosion of WP internals will not result in hydriding of fuel cladding.

Although hydriding as a result of galvanic corrosion has been observed, cladding alloys resist such corrosion on contact with carbon steel, and, if such an effect should occur, corrosion of the carbon steel would quickly break the electrical contact between the materials. Hydriding from

Inconel contact requires smearing the Inconel into the zirconium metal matrix and then occurs only for a short time.

Waterlogged Rods–The fraction of fuel rods with breached cladding is small (~0.16 percent). Most moisture can be removed from defective rods by standard drying processes. When drying the handling can be minimized. The impact of any residual amount of moisture is insufficient to cause significant corrosion of the internal structural portion of the WP and further degradation of the cladding. Furthermore, any moisture available would likely be consumed first by any CSNF exposed by the initially perforated cladding; therefore, the issue is excluded based on low consequence.

General Corrosion of Cladding–General corrosion is synonymous with zirconium oxidation. The outer surface of the cladding becomes coated with oxide (ZrO_2). Because this ZrO_2 is of high density and adheres to the surface, it slows down further oxidation. The oxidation can be from O_2 consumption (dry oxidation) or H_2O consumption (wet oxidation). Once the fuel assemblies are placed in the waste package, further oxidation cannot occur until the WP is penetrated. Wet oxidation has been studied for over forty years because of the importance of cladding performance in reactor operation. Furthermore, the metallurgical condition of cladding while in storage has been investigated. There was no significant degradation of the Zircaloy cladding. Various analyses have concluded that cladding oxidation under repository conditions would not lead to perforation within 10,000 or even 100,000 years. The conclusion from this analysis is that general corrosion will not perforate the cladding during the regulatory period (CRWMS M&O 2000s, Section 6.2.3).

Microbially Influenced Corrosion of Cladding–There is no experimental evidence that microbially influenced corrosion (MIC) occurs on zirconium metal or alloys. The two major forms of MIC for materials being considered for containment vessels in mined geologic repositories are (1) sulfide attack through the action of sulfate reducing bacteria and (2) corrosion induced by organic acids secreted from certain bacteria. With respect to these forms of MIC, (1) sulfate reducing bacteria do not affect zirconium, and (2) corrosion induced by organic acids is most unlikely because of zirconium's tolerance to organic acids and a wide range of pHs. However, MIC was not excluded as a mechanism for producing locally very acid conditions and is thus included in the conservative localized corrosion model.

Acid Corrosion from Radiolysis –Zirconium is used for fuel cladding because of its high resistance to corrosion in highly acidic environments including those local environments in high radiation fields. Studies show that zirconium has excellent corrosion resistance to nitric acids and concentrated hydrogen peroxide. However, this process was not excluded as a mechanism for producing locally very acid conditions and is thus included in the conservative localized corrosion model.

High Dissolved Silica Content of Waters Enhances Corrosion of Cladding–Silica is stable and is not corrosive to most materials, including Zircaloy. Therefore, this mechanism is excluded. The corrosion of Zircaloy in the presence of fluoride in silica saturated water (i.e., the formation of fluosilicic acid) is not considered to be a plausible failure mechanism because of the predicted concentration of fluorides is low and the pH is above 3.6.

3.4.2.7 Matrix Exposure Model

The preceding subsections of Section 3.4 discuss the process of perforating the cladding. This subsection addresses the tearing open (unzipping) of the cladding and the release of radionuclides. The release of radionuclides does not occur until after the WP fails and steam or water can enter the WP. The release is predicted to occur in two stages, (1) fast release and (2) wet unzipping (tearing of the cladding).

Fast Release–The fast release refers to the inventory of radionuclides that are in the gap between the fuel pellets and the cladding (gap inventory) as summarized in Section 3.3.1.1 and the radionuclides at the grain boundaries of the spent fuel matrix. The gap inventory of iodine and cesium is predicted to be released in proportion to the fission gas release fractions (CRWMS M&O 2000h, p. 82). That is, the release of iodine is 4.2 percent. The cesium gap inventory is 1/3 of the fission gas release fraction or 1.4 percent.

The inventory of the radionuclides in the grain boundaries is also released quickly. The amount released is determined as follows. As discussed in CRWMS M&O (2000i, Section 6.5.2), Wilson (1990, Section 3) reports releases of various radionuclides from fuel rod samples. The samples included intact and defected fuel rod sections and bare fuel. Wilson exposed the samples to water and measured the amounts of various radionuclides that were released during that period. After the first measurement in about 200 days, the samples were again exposed, and the measurements were repeated after an additional period of comparable length. The fast release from the uranium pellet through slits and holes in the cladding is estimated by calculating the release rate from Wilson's eight experiments and extrapolating this release rate until the larger gaps are closed by secondary phases (approximately 5 years). Figure 3.4-10 gives the cumulative distribution function (CDF) for the fast release of radionuclides from the fuel matrix generated from Wilson's eight experiments. Also shown is a uniform distribution of release fraction between 0 and 0.004 (mean and median value = 0.002). This linear fit is conservative, overpredicting the release values for the lower CDF samplings. The cesium and iodine gap inventories discussed above are added to obtain the total fuel matrix fast release fraction. In the TSPA abstraction, the fast release grain boundary fraction is uniformly distributed between 0 and 0.4 percent for all radionuclides.

Wet and Dry Unzipping–Fuel rods with perforated cladding are expected to remain intact until the WP fails and permits air and moisture to enter. While the humidity is low, dry unzipping could occur. In this mechanism, the UO_2 of the CSNF rapidly oxidizes to U_3O_8. However, the WP is expected to last for the regulatory period for the nominal scenario, thus the fuel temperatures will be too low for dry unzipping to occur. Wet unzipping is modeled to start at WP failure for rods already perforated at WP failure or start when rod perforation occurs, if after WP failure. The fuel matrix is modeled to dissolve at the intrinsic dissolution rate that is evaluated at the temperature and in-package chemistry. The dissolved UO_2 precipitates locally as metaschoepite. This secondary phase isolates most of the fuel from the moisture and increases volume compared to UO_2. In time, the cladding in the reaction region may be torn as the reaction continues. This reaction region is modeled to be cone shaped based on experimental observations of dry unzipping. The unzipping propagates along the rod at a rate approximately 40 times (range 1 to 240 times) as fast as the intrinsic dissolution rate of the CSNF evaluated in the CSNF Matrix Degradation Component. It is assumed that the cladding perforation is in the

center of the rod. This maximizes the release rate. Figure 3.4-11 gives the time to unzip a rod as a function of temperature for a fixed in-package chemistry. In the TSPA-SR, the intrinsic dissolution velocity is calculated for each time step as a function of local chemistry, pH, and temperature. The unzipping velocity is then calculated using a triangular distribution with the minimum, best estimate, and maximum unzipping velocity being 1, 40, 240 times the intrinsic dissolution velocity, respectively. The fraction of fuel dissolved and exposed is then calculated for each time step.

The bases for matrix exposure analysis are measurements of gap inventories, experimental measurements of the release of radionuclides from damaged cladding and intrinsic dissolution measurements. Although unzipping has not been observed for rods in storage pools for the past 40 years, the unzipping analysis is consistent with dry unzipping experiments. Uncertainties have been defined that are based on both experiments and extremes in the analysis. Therefore this analysis is appropriate for TSPA-SR. Although it is conservative, it is not as conservative as the assumption of instantaneous unzipping to expose the CSNF that was used in previous TSPAs.

3.4.3 Confidence/Limitations/Validation

There is good reason to have confidence in the cladding degradation analysis. The cladding degradation analysis is based on over 40 years of experiments and observations of cladding behavior. The analysis of initial cladding conditions is based on reactor fuel performance reports that have been published since the start of the industry. Creep, SCC and DHC analysis is supported by an extensive experimental base. Zirconium alloys were originally developed for use in the chemical industry to handle very corrosive fluids such as hydrochloric acid. In water environments, continuous corrosion experiments have been performed for 27.5 years. Fuel has been exposed in spent fuel pools for over 25 years and in dry storage research programs. The models, including ranges and uncertainties, that have been developed for TSPA are based primarily on experimental observations.

There are constraints, caveats, and limitations to this analysis. This cladding degradation analysis is only applicable to commercial PWR fuel with Zircaloy cladding. Fuel reliability from reactor operation is for both PWR and BWRs. It is also limited to fuel exposed to normal operation and anticipated operational events (events which are anticipated to occur within a reactor lifetime), and not fuel that has been exposed to severe accidents. (For example, fuel from the Three Mile Island reactor accident is included in the DSNF fuel category, which, with the exception of naval SNF, does not account for the performance of the cladding.) Fuel burnup projections have been limited to the current licensing environment with restrictions on fuel enrichment, oxide coating thickness, and rod plenum pressures. Cladding degradation from YMP surface facility handling and operation was not considered. Ranges of uncertainties have been established and conservatism was used in developing this analysis.

Based on the considerations discussed above, this analysis is appropriate for its intended use.

3.4.4 Other Views

3.4.4.1 Past Modeling of Cladding Degradation

Before TSPA-VA (DOE 1998b), YMP performance assessments did not consider protection of the fuel by cladding and all the fuel in a drip zone was available to dissolve. This rapid dissolution was then limited by the solubility of the various radionuclides.

In TSPA-VA, the failure of PWR cladding was assumed to bound the failure of BWR cladding because the PWR cladding is typically thinner than BWR cladding. For PWR cladding, the initial cladding failure percentage was set at 1.25 percent; the major portion of this failure (1.15 percent) was stainless steel cladding, which was uniformly distributed in all WPs throughout the repository. PWR cladding failure from rockfall rubble was assumed to begin after the WP was sufficiently weakened (which was set in all cases at 10^5 years after initial breach of the WP). The cladding failure caused by rubble then increased as a power function (linearly on a log scale) thereafter from 0.2 percent at 10^5 years after breach to a sampled value at 10^6 years after breach; the sample range varied between 0.2 percent and 11 percent. PWR cladding failure from localized corrosion was evaluated using WAPDEG; the localized corrosion rate was assumed to be 10 to 1,000 times smaller than the corrosion rate of Alloy 22. Based on these assumptions, the amount of fuel exposed at 10^6 years varied between 0.3 and 40 percent (mean of 7.8 percent).

In TSPA-VA, the fuel in rods that were failed before emplacement were assumed to be completely exposed for dissolution. The fuel in an area below the rock or corrosion patch was available for dissolution but the remaining ends of the rods were not. This model was not necessarily conservative.

The method of estimating the performance of the Zircaloy cladding on the CSNF has changed substantially from TSPA-VA. Two specific phases of cladding degradation are modeled: (1) perforation (such as existing perforations from use in the reactor and creep failures from overheating the cladding during storage, etc.) and (2) exposure of the SNF matrix over time (here evaluated by assuming the Zircaloy cladding unzips). Many mechanisms of perforation were studied and many dismissed in FEPs. Yet, in response to comments discussed in the following sections, an extremely conservative estimate of the number of cladding perforations over time is used to acknowledge uncertainty. Also in response to comments, the exposure of the SNF is evaluated through a hypothetical but bounding rate of unzipping of the cladding such that the entire inventory of radionuclides can eventually be exposed over sufficient time.

3.4.4.2 Comments About Including the CSNF Cladding in Waste Form Degradation Model

Concerns have been expressed in meetings of the Advisory Committee on Nuclear Waste (ACNW) and NWTRB as well as in documented reports of the TSPA Peer Review Panel and in the IRSR on CLST related to DOE's taking credit for cladding of SNF and the performance of the cladding if included in the TSPA. One issue is whether taking credit for cladding is practical and the other issue is whether sufficient information has been considered related to clad degradation.

Comments

During the 106th meeting of the ACNW, the NRC challenged DOE's heavy reliance on cladding credit, considering that the cladding has to remain intact for very long periods of time under conditions that it was not designed for and questioning DOE's ability to gather data to support related assumptions. During the 110th meeting of the ACNW, the NRC contended that the model that the NRC uses for spent nuclear fuel dissolution is better than the DOE models because it relied upon slower CSNF dissolution rates and not cladding credit. The TSPA Peer Review Report (Budnitz et al. 1999, Section II.E) questioned the extent of credit taken for cladding in the TSPA-VA, noting that the credit taken for spent nuclear fuel cladding may be optimistic.

Response

NRC purposefully adopted a TSPA approach such that realistic estimates could be made on the performance of a disposal system in order to make wise policy decisions (60 FR 42622, Section IV; Rechard 1999, p. 777). The Zircaloy cladding is an important characteristic of the CSNF. The Zircaloy cladding on the CSNF is in good condition and has been handled carefully at the nuclear reactors. Furthermore, Zircaloy cladding readily resists corrosion, and the modes of failure in various environments have been studied for 30 years. Therefore, it is believed that sufficient data exist for the inclusion of Zircaloy degradation. The Waste Form Degradation Model includes the existence of the Zircaloy cladding on the CSNF and naval SNF to provide for a realistic estimate of the behavior of these fuels in the TSPA. The NRC fuel dissolution model takes credit for the combination of calcium and silicon to suppress the dissolution of the UO_2. This effect is not observed in drip tests on fuel and, therefore, not used in the TSPA-SR. The cladding degradation model used in TSPA-VA was criticized as possibly being optimistic. The model used in TSPA-SR is more conservative.

3.4.4.3 Comments on Modeling Cladding Performance

Comments

Questions have been raised related to DOE's ability to sufficiently establish the performance of cladding. NRC noted during the 110th meeting of the ACNW that the initial damage and condition of cladding are important to determining the effect of cladding. The TSPA Peer Review Panel (Budnitz et al. 1999, Executive Summary Sections B and E, Section II.E) noted that studies needed to take credit for cladding are not available and identified the quality of data from the industry regarding cladding initial state as a vulnerability to taking cladding credit. The TSPA Peer Review Panel also stated that the estimates of performance in the TSPA-VA (DOE 1998b) rest in large part on potentially optimistic, or at least undemonstrated, assumptions of the behavior or performance of cladding including understatement of related uncertainties (Budnitz et al. 1999, Executive Summary Sections B and E, Section II.E). Insufficient analysis in the TSPA-VA (DOE 1998b) was cited related to hydrogen embrittlement of zirconium cladding, and suggestions were made related to further investigations to identify additional potential failure mechanisms (e.g., pitting and crevice corrosion, unzipping due to secondary phase formation, stress corrosion cracking, long-term localized corrosion in a radiolysis-induced acidic condition) (Budnitz et al. 1999, Section II.G, IV.E). It also suggested that data needs related to cladding performance had not been met (Budnitz et al. 1999, Executive Summary, Section E). The

NWTRB (NWTRB 1998, Chapter 3, Section V.B.2) expressed concern that combined interactions of corrosion products from the WP liner and outer walls, radiolysis, water, and elevated temperatures could produce a corrosive environment inside WPs that could affect Zircaloy cladding and had not yet been fully investigated. In the IRSR on CLST (NRC 1999a, Section 5.3.5), NRC noted a lack of direct comparison of DCCG model prediction against experimental data and lack of experimental evidence of DCCG in Zircaloy cladding. It also noted disagreements on some constraints in the models and on the validity of extrapolating certain creep model results to lower stresses and temperatures.

Response

To evaluate all possible behavior of a potential disposal system at Yucca Mountain, DOE undertook a formal approach to develop pertinent features, events, and processes (FEPs) for the disposal system. This approach also included evaluating FEPs related to perforating the cladding. The DOE has proposed 20 FEPs including localized corrosion (general pitting corrosion, crevice corrosion, ferric chloride pitting), creep rupture, hydride failures (e.g., DHC and embrittlement), mechanical failure due to rock fall, stress corrosion cracking (SCC), cladding degradation before YMP receives it, MIC, acid corrosion from radiolysis, DCCG, future pressurization of the rods during disposal, and general cladding oxidation (as already mentioned and tabulated in Section 2.3.2.4).

In summary, the DOE has reviewed all potential perforation mechanisms and has taken a conservative view for those mechanisms retained. Specifically, the project has included a conservative model for localized corrosion due to some unspecified mechanism. This action was taken after a review of corrosion mechanisms (CRWMS M&O 2000ag, Section 7) identified no plausible mechanism for accelerated corrosion under repository conditions. This conservative approach should address the NWTRB concerns. For localized corrosion, the NRC also postulates that chloride attack in pits is possible given the presence of ferric ions. However, when the waste packages fail late in life, the potential for achieving aggressive ferric chloride chemistries is very low. Hence, this mechanism is not included. The NRC is currently re-evaluating its approach to estimating creep failures for reactor service. However, in regard to creep in the repository, the NRC states that "there is general consensus that cladding failure by creep rupture is unlikely under repository conditions." Thus, creep is not an issue. Again, there is consensus regarding delayed hydride attack in that the NRC that believes that this mechanism is not important under repository conditions. The project has used a realistic treatment for mechanical failure due to rock fall and the NRC agrees with this approach. The NRC notes that Zircaloy is susceptible to SCC in the presence of tensile stresses lower than the yield stress. The DOE agrees that SCC is possible and has provided a conservative model for SCC failure. General cladding oxidation is not likely to be of concern due to the long life of the waste packages and the resultant low temperatures when the waste packages do breach. Because of the long life of the waste packages, the likelihood of aggressive conditions from radiolytic acids inside the waste package are small. Although it was not noted in the NRC's IRSR on CLST, there has been concern, noted above by the ACNW, that the initial condition of the cladding is uncertain. This has been thoroughly reviewed in the AMR on initial cladding condition and been found to be only a minor contributor to failure of the cladding.

To evaluate the slow release of radionuclides from perforated cladding, the DOE has assumed an unzipping mechanism that conservatively bounds the possible diffusive release out of pinholes in the cladding.

3.5 DEPARTMENT OF ENERGY SPENT NUCLEAR FUEL DEGRADATION COMPONENT

The DSNF Degradation Model Component predicts the rate of degradation of the DSNF waste category and of the immobilized plutonium ceramic waste. The component was developed using the logic shown in Figure 3 of *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* (ASTM C1174-97).

The primary document supporting the DSNF Degradation Model Component was the *DSNF and Other Waste Form Degradation Abstraction* AMR (CRWMS M&O 2000j), which in turn was supported to a limited extent by *Miscellaneous Waste-Form FEPs* (CRWMS M&O 2000n).

3.5.1 Model Description

The degradation rates of all waste forms other than commercial spent nuclear fuel and high level waste were analyzed in *DSNF and Other Waste Form Degradation Abstraction* (CRWMS M&O 2000j). Over 250 distinct types of DOE spent nuclear fuel may be disposed of in the potential repository at Yucca Mountain. The Office of Civilian Radioactive Waste Management and the National Spent Nuclear Fuel Program have collaborated in the identification of spent nuclear fuel "groups" to simplify the analysis of their effects on repository preclosure safety analyses or for postclosure TSPA.

The DOE spent nuclear fuel groups are:

- Group 1–Naval spent nuclear fuel
- Group 2–Plutonium/uranium alloy
- Group 3–Plutonium/uranium carbide
- Group 4–Mixed oxide and plutonium oxide fuels
- Group 5–Thorium/uranium carbide
- Group 6–Thorium/uranium oxide
- Group 7–Uranium metal
- Group 8–Uranium oxide
- Group 9–Aluminum-based spent nuclear fuel
- Group 10–Unknown
- Group 11–Uranium-zirconium-hydride.

In addition, the immobilized ceramic plutonium waste form was also evaluated. This waste form will consist of disks of a plutonium-containing, titanium dioxide-based ceramic that will be enclosed in stainless steel cans, which in turn will be encased in a borosilicate glass matrix within the high level waste canisters.

DSNF and Other Waste Form Degradation Abstraction (CRWMS M&O 2000j) provided three types of degradation models for the DOE spent nuclear fuel and plutonium-ceramic waste forms: upper-limit, conservative, and best-estimate, to provide the user of the models appropriate flexibility in their application to any particular postclosure performance scenario. A summary of the upper-limit, conservative, and best-estimate dissolution models for each DSNF group is shown in Table 3.5-1. This table shows the three types of models for each DSNF group and the immobilized ceramic Pu-disposition form, the surrogate material for which the model was developed, the literature/document source reference for the model, and the ASTM C1174-97 classification of the model. An upper-limit model predicts release rates always well in excess of actual dissolution rate data. The conservative degradation model provides an estimate of dissolution rate that reflects the higher rate end of available dissolution data or the SNF groups or similar materials. A best-estimate model is appropriate only when sufficient dissolution data exists and the characteristics of the waste form can be shown to correspond to the characteristics of the materials that provided the dissolution database. For the conservative and best-estimate models, various surrogate spent nuclear fuels were evaluated for degradation behavior.

A discussion of the individual models for each fuel group is provided in CRWMS M&O (2000j). The uranium fuel accounts for the biggest portion of the MTHM of DSNF. In general, the degradation rates of the metallic uranium or carbide fuels are much higher than the oxide fuels. Because the metallic uranium and carbide fuels degrade very rapidly in TSPA time scales, the TSPA results have been very insensitive to these rates. In addition, TSPA analyses have shown that the overall performance of the potential repository is driven primarily by the commercial spent nuclear fuel and high-level radioactive waste inventories. For these reasons, the conservative Hanford N Reactor fuel model (Equation 3.5-1) was recommended as the surrogate to bound all the DOE spent nuclear fuel groups 2 through 11. Figure 3.5-1 provides a schematic description of how the degradation model is implemented for these groups.

$$\text{Rate} = 1.75 \times 10^5 \, (\rho_{matrix}/\rho_{U\text{-}metal}) \text{ mg/m}^2\cdot\text{day} \qquad \text{(Eq. 3.5-1)}$$

where ρ_{matrix} is the density of the spent fuel matrix, and $\rho_{U\text{-}metal}$ is the density of uranium metal.

The Navy will provide a best-estimate model for the group 1 naval fuel, but pending this, the naval SNF will be represented by commercial spent nuclear fuel in TSPA.

The best-estimate for the degradation of the immobilized ceramic plutonium is given by a titanate ceramic model, with an exposed surface area corresponding to the geometric surface area of the plutonium-containing ceramic disks. For TSPA, however, the immobilized plutonium inventory was averaged into the high level waste inventory, and the higher rates for high level waste degradation (Section 3.6) were used.

Table 3.5-1. DSNF, Naval SNF, Pu-Disposition Release/Degradation Models

Waste Type	DSNF Group	Upper-limit Model		Conservative Model		Best-estimate Model	
		Surrogate	Model	Surrogate	Model	Surrogate	Model
DSNF	1. Naval SNF	CSNF	Commercial LWR SNF dissolution model[a]	CSNF	Commercial LWR SNF dissolution model[a]	CSNF	Commercial LWR SNF dissolution model[a]
DSNF	2. Pu /U Alloy	Fermi	Full release over TSPA time step	U - 6% Mo	(bounding) 10 × Unirradiated U-metal best-estimate: Wet oxic conditions: R (kg/m^2·s) = 1.88 × 10^4 exp (–7970/T_K) Humid oxic conditions: R (kg/m^2·s) = 0.27 × 10^3 exp (–7240/T_K)	U - 8% Mo	(semi-empirical) Unirradiated U-metal best-estimate: Wet oxic conditions: R (kg/m^2·s) = 1.88 × 10^3 exp (–7970/T_K) Humid oxic conditions: R (kg/m^2·s) = 0.27 × 10^2 exp (–7240/T_K)
DSNF	3. Pu/U Carbide	FFTF	Full release over TSPA time step	UC_2	(bounding) 100 × Unirradiated U-metal best-estimate: R (kg/m^2·s) = 100 × [1.88 x 10^3 exp (–7970/T_K)]	UC_2	(bounding) 100 × Unirradiated U-metal best-estimate: R (kg/m^2·s) = 100 × [1.88 × 10^3 exp (–7970/T_K)]
DSNF	4. MOX and Pu Oxide	LWR SNF	Full release over TSPA time step	LWR SNF	(bounding) 100 × Intact U Oxide best-estimate	UO_2	(semi-empirical) U Oxide best-estimate model
DSNF	5. Th/U Carbide	UC_2	Full release over TSPA time step	UC_2	(bounding) 10 × Unirradiated U-metal best-estimate	SiC	(semi-empirical) R (kg/m^2·s) = 0.6 × 10^{-12}
DSNF	6. Th/U Oxide	Fort St. Vrain	Full release over TSPA time step	ThO_2 - 2 wt% UO_2	(bounding) 1000 × Best-estimate	Synroc	(semi-empirical) R (mg/m^2·d) = 82.0 × 10$^{(-1000/T_K)}$
DSNF	7. U-Metal-Based	N-Reactor	Full release over TSPA time step	N-Reactor	(semi-empirical) 1.75 × 10^6 mg/m^2·d	N-Reactor	(semi-empirical) 1.75 × 10^5 mg/m^2·d

Table 3.5-1. DSNF, Naval SNF, Pu Disposition Release/Degradation Models (Continued)

Waste Type	DSNF Group	Upper-limit Model		Conservative Model		Best-estimate Model	
		Surrogate	Model	Surrogate	Model	Surrogate	Model
DSNF	8a. Intact U Oxide	LWR SNF	Full release over TSPA time step	LWR SNF	(bounding) 100 × Intact U Oxide best-estimate	LWR SNF	(semi-empirical) Commercial LWR SNF dissolution model[a]
DSNF	8b. Damaged U Oxide	Three Mile Island	Full release over TSPA time step	Three Mile Island	(bounding) 100 × Intact U Oxide best-estimate	Three Mile Island	(bounding) 100 × Intact U Oxide best-estimate
DSNF	9. Al-based	Foreign Research Reactors, Advanced Test Reactor	Full release over TSPA time step	U-Al alloy in bicarbonate solution	(bounding) 36 $mgU/m^2 \cdot d$ @ 25°C 360 $mgU/m^2 \cdot d$ @ 90°C	SRS U-Al SNF in J-13 well water	(empirical) 0.22 $mgU/m^2 \cdot d$ @ 25°C 2.20 $mgU/m^2 \cdot d$ @ 90°C
DSNF	10. Unknown	N-Reactor	Full release over TSPA time step	N-Reactor	(bounding) 10 × Unirradiated U-metal best-estimate	N-Reactor	(empirical) Unirradiated U-metal best-estimate
DSNF	11. U-Zr-Hx	TRIGA	Full release over TSPA time step	U-Zr-Hx	(bounding) 0.1 × U-oxide best-estimate	U-Zr-Hx	(empirical)[b] 0.1 × U-oxide best-estimate
Pu	N/A	N/A	Full release over TSPA time step	HLW Glass	HLW glass dissolution model[c] with effective surface area taken as 20 × the geometric surface area of the exposed glass matrix	Multi-phase titanate ceramic	(semi-empirical) pH-dependent titanate ceramic dissolution equations[d]

Source: DSNF and Other Waste Form Degradation Abstraction. ANL-WIS-MD-000004. (CRWMS M&O 2000j, Table 1)

NOTES: (bounding)–indicates a bounding model per the materials behavior modeling logic of ASTM C 1174-97
(empirical)–indicates an empirical model per the materials behavior modeling logic of ASTM C 1174-97
(semi-empirical)–indicates a semi-empirical model per the materials behavior modeling logic of ASTM C 1174-97
(mechanistic)–indicates a mechanistic model per the materials behavior logic of ASTM C 1174-97
[a]Commercial LWR SNF Dissolution Model, see Equations 3.3-3 and 3.3-4
[b] See CRWMS M&O 2000j, Section 6.3.11 for reference to empirical data and recommended model
[c]HLW Glass Dissolution Model, see Equations 3.6-4, 3.6-5 and 3.6-6
[d]See CRWMS M&O 2000j, Section 6.3.12 for equations

3.5.2 Confidence/Limitations/Validation

The application of the DSNF and immobilized ceramic Pu degradation models involves the extrapolation of the models over long periods of time, which are orders of magnitude greater than the experimental test periods used to generate the data used to derive the models. ASTM C1174-97 (Section 24), recommends that uncertainties in the extrapolation of such models be minimized through the use of models whose mathematical forms are as mechanistic as possible. 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 small amount of data for those which have been tested, precludes the development of a mechanistic model at this time. Additionally, uncertainties in the data used to generate the models–such as in the surface area measurements used to calculate normalized dissolution rates–produce significant uncertainties even in the short-term application of the models. For this reason and because preliminary TSPA analyses have shown that the overall performance of the repository is very insensitive to the degradation rate of the DSNF, upper-limit or bounding degradation models will be used.

The initial results of TSPA sensitivity analyses for DSNF (CRWMS M&O 1999e, Section 7) indicate that the performance of the repository is very insensitive to the DSNF degradation kinetics. That is, the use of the upper-limit model for the DSNF in the TSPA performed in that study still resulted in a calculated boundary dose well within requirements. Use of a less conservative model for the DSNF in the TSPA boundary dose calculation would not significantly lower the calculated boundary dose, because even with the upper limit, model releases due to DSNF are significantly lower than those due to HLW and CSNF.

If because of this insensitivity the upper-limit model is the only one used for TSPA analyses, then validation of the other models would be unnecessary. Since the upper limit release model is that of instantaneous release of all radionuclides, i.e., the maximum rate conceivable, it does not require validation. It depends only on the total inventory of radionuclides in the DSNF. The upper limit model proposed for the DSNF and Pu disposal waste forms are, thus, impacted primarily by the total inventory of radionuclides that are present. The conservative and best estimate models for the DSNF waste forms are primarily impacted by the validity of the uranium metal-based SNF dissolution models.

Based on the considerations discussed above, this model is appropriate for its intended use.

3.5.3 Other Views

For TSPA-VA, the model for the degradation of metallic uranium was a classic Arrhenius kinetic rate equation using parameters from assessments of SNF and HLW (Rechard 1995, p. 11-22; DOE 1998a, p. 7-32; see also CRWMS M&O 1998b, p. 6-69). The overall degradation rate was the Arrhenius rate times the effective surface area. In TSPA-VA, the effective surface area was five times the geometric surface area. Because the degradation rate estimated by the Arrhenius equation was so rapid, the sensitivity of the results to varying the multiplier for the geometric surface area between 0.1 and 100 (thus, the overall degradation rate) was small.

3.6 HIGH-LEVEL WASTE DEGRADATION COMPONENT

The function of the HLW Degradation Component is to provide a conservative model for calculating the rate of degradation of borosilicate glass for the range of conditions (immersion, humid air, and dripping water) to which it may be exposed after the WPs fail.

The HLW model will be implemented in the TSPA-SR analysis to calculate the rate of glass degradation. The rate of radionuclide release from the HLW matrix will be calculated by multiplying the glass degradation rate by the mass fraction of the radionuclide in the glass. This approach for calculating the radionuclide release rate is based on the conservative assumption that the release of radionuclides is congruent with the degradation of borosilicate glass.

The model is implemented in the form of an analytical expression containing four parameters (η, E_a, S, and k_{eff}; see Figure 3.6-1 and Equation 3.6-2) and two variables (i.e., temperature and pH). The model parameters account for the pH, temperature, surface area, and the combined effects of glass composition and solution composition on the rate of glass corrosion. Conservative estimates of the values for the model parameters are provided based on experimental data. Implementation of the model for the TSPA-SR analysis requires input of temperature and pH data.

Consistent with the conceptual model for in-package chemistry, degradation of borosilicate glass is assumed to occur as if the glass were fully immersed although it is expected that much of the glass will be exposed to humid air or dripping water conditions. This assumption is based on a comparison of the model that was developed for immersion (see Equations 3.6-4 and 3.6-5) to the model developed for glass degradation in humid air or dripping groundwater conditions (see Equation 3.6-7) CRWMS M&O (2000k, Section 6.3.2). This comparison showed that the rate of glass corrosion under humid air and dripping water conditions was conservatively bounded by the dissolution rate under immersion conditions.

The primary document supporting the HLW Degradation Model Component was the *Defense High Level Waste Glass Degradation* (CRWMS M&O 2000k), which in turn was supported to a limited extent by *Miscellaneous Waste-Form FEPs* (CRWMS M&O 2000n, Section 6.2.8).

3.6.1 Model Description

Available experimental data concerning the degradation behavior of borosilicate-based glasses were analyzed to develop models for the dissolution and consequent radioisotope release from HLW glass under aqueous immersion conditions in the repository environment. The mechanism and rate expression for aqueous dissolution are sufficiently well understood to designate the HLW model as a semi-empirical model as described in ASTM Standard C1174-97 (1997, Section 19.3.4).

The basic form of rate expression that describes dissolution of waste glass immersed in water is (CRWMS M&O 2000k, Section 6.1.1):

$$\text{Rate} = S \{ k_o \cdot 10^{\eta \cdot pH} \cdot \exp(-E_a/RT) \cdot [1 - (Q/K)] \} \qquad \text{(Eq. 3.6-1)}$$

where

Rate = the dissolution rate of the glass, in units of mass/time
S = the surface area of glass immersed in water, in units of area
k_o = the intrinsic dissolution rate, which depends only on glass composition, in units of mass/(area · time)
η = the pH dependence coefficient, which is dimensionless
E_a = the effective activation energy, in units of kJ/mol
R = the gas constant, which is 8.314 kJ/(mol · K) (accepted value)
T = the temperature in kelvins
Q = the concentration of dissolved silica in the solution, in units of mass/volume
K = a quasi-thermodynamic fitting parameter equal to the apparent silica saturation concentration for the glass, in units of mass/volume

This rate expression contains two main parts: the forward rate, $k_o \cdot 10^{\eta \cdot pH} \cdot \exp(-E_a/RT)$, which represents the dissolution rate in the absence of feedback effects of dissolved silica (and other aqueous species), and the reaction affinity term (1–Q/K), which quantifies the feedback effects. Because the value of Q can range between zero and K, the value of the affinity term is mathematically constrained to values between one and zero. A glass will, therefore, dissolve at the highest rate possible (at a given temperature and pH value) when the value of the affinity term is one (i.e., when Q = 0). The dissolution rate will decrease as the value of the affinity term decreases (i.e., as the value of Q approaches K) until a minimum rate is reached.

Use of the form of the rate expression shown above would require knowledge of several parameter values that depend on the glass (k_0, η, E_a, S, K) and of several variables related to the exposure conditions (pH, T, and Q). Evaluation of Q would be complex and subject to considerable uncertainty. A simpler bounding approach was, therefore, adopted. In this approach the affinity term (1 – Q/K) was combined with the intrinsic dissolution rate constant k_o to generate the following abstraction for aqueous degradation of HLW for the TSPA-SR analysis:

$$\text{Rate}/S = k_{eff} \cdot 10^{\eta \cdot pH} \cdot \exp(-E_a/RT) \qquad \text{(Eq. 3.6-2)}$$

where,

$$k_{eff} = k_0 \cdot (1 - Q/K)$$

Although k_{eff} is a complex function of glass composition and solution variables, the approach adopted in the abstraction is to provide a constant value for k_{eff} that conservatively bounds the anticipated range of the function $k_0 \cdot (1 - Q/K)$. This approach reduces the abstracted model to an equation involving four parameters (η, E_a, S, and k_{eff}) and two variables (pH and T). Available experimental data were used to obtain conservative estimates of each of these parameters (CRWMS M&O 2000k, Section 6).

3.6.1.1 Estimation of the Model Parameters

The approach used to estimate the model parameters can be broken into three steps.

The first step was to analyze the dependence of the forward rate on the glass composition, pH, and temperature. These dependencies were measured under flow-through test conditions in

which the value of the affinity term is maintained near one. In these tests, the dependence of the forward rate on glass composition, pH, and temperature was measured under test conditions in which two of the variables (e.g., glass composition and pH) were held constant and the other (e.g., temperature) varied. On the basis of the experimental results, the pH and temperature dependence of rates were found to be independent of the glass composition, within the composition range of the glasses tested, so the same values for the η, and E_a parameters are used for all waste glasses. The values of these parameters do however depend on pH. Analysis of the available data (CRWMS M&O 2000k, Section 6.2) resulted in the empirical estimates for values of the η and E_a parameters for acidic and basic conditions (see Equations 3.6-4 and 3.6-5).

In the second step, a bounding value for the k_{eff} parameter was determined based on experimental observations of the behavior of the affinity term for test conditions that spanned a broad range of reaction progress. Experimental data show that the behavior of the corrosion rate with reaction progress can be characterized as proceeding through three stages. Stage I occurs when the value of the affinity term is one and glass dissolves at the forward rate for the specific temperature and pH conditions involved. Stage I will not occur when glass is contacted by groundwater containing high-levels of dissolved silica (from dissolution of minerals present in tuff and from the dissolution of the glass itself). Stage II occurs as the value of the affinity term decreases significantly due to the accumulation of glass components particularly dissolved silica, in solution. The value of the affinity term (and thus the dissolution rate) cannot become zero because glass is thermodynamically unstable and cannot equilibrate with the solution. Stage III occurs when the dissolution rate increases due to the formation of alteration phases, effectively removing dissolved silica from the solution. The formation of alteration phases is believed to cause a decrease in the value of Q due to the consumption of dissolved silica as these silica-bearing phases form (CRWMS M&O 2000k, Section 6.2.2). This Stage III behavior has not been observed for all glasses or all experimental conditions and the mechanisms involved remain unclear. It is not possible, therefore, to predict when Stage III occurs or to unambiguously predict the value of $k_0 \cdot (1 - Q/K)$ i.e., k_{eff} for use in performance assessment calculations. For these reasons a conservative bounding approach was adopted in evaluating k_{eff}. After evaluating several options, an approach based on using the ASTM Product Consistency Test, Type A, (PCT-A) (ASTM C1285-97, *Standard Test Methods for Determining Chemical Durability of Nuclear, Hazardous, and Mixed Waste Glasses: The Product Consistency Test (PCT)*) test results was selected (CRWMS M&O 2000k, Section 6.2.3). This approach involved using the relatively short-term PCT-A dissolution rate test results to obtain bounding values for k_{eff} that would conservatively encompass all three potential stages of dissolution behavior. The bounding values estimated for k_{eff} are shown in the leading coefficients of Equations 3.6-4 and 3.6-5.

The third step involves estimating the surface area parameter S. The glass surface area that is available for corrosion in the disposal environment is a difficult parameter to estimate precisely. The logs of waste glass will crack within the pour canister due to thermal and mechanical stresses generated as the glass cools and as the waste form is handled. Cracking of the glass log will result in surfaces that may be contacted by water. The reactivity of surfaces in the tight fracture cracks that result from thermal or mechanical stresses will be determined by the glass composition, temperature, and the chemistry of the water that fills the crack in the same way that these factors affect the dissolution rate of free surfaces. The key difference between the dissolution rates of the glass within cracks and at the outer surface is the transport rates of reactant into the crack and reaction products out of the crack. Short-term dissolution tests have

shown that, while more glass dissolved in tests with samples of fractured glass than in tests with samples of a glass that was not fractured, the difference was less than the estimated increase in the surface area. It had been concluded from the results of tests in which various crack widths were simulated with platinum wire spacers that "the assumption that crack surfaces leach as readily as the external surface is unduly conservative" (Perez and Westsik 1981, page 168). However, since sufficient information is not available to quantify either the fraction of cracks that are accessible to water or the difference between the corrosion rates in cracks and at an outer surface, the conservative assumption that all surfaces corrode at the same rate when exposed to water was adopted. The value of the parameter S is, therefore, based on an estimate of the total surface area of the glass (i.e., the geometric surface area together with the fracture surface area). This area has been estimated to be approximately 20 times the geometric surface area (CRWMS M&O 1998b, pp. 6-79, 6-80). An expression to calculate the initial surface area of a fractured cylindrical glass log is:

$$S_o = 20 \cdot (2\pi r_o^2 + 2\pi r_o L_o) \qquad \text{(Eq. 3.6-3)}$$

where,

S_o = the initial exposed surface area of a glass log due to fracturing
r_o = the initial radius of a glass log
L_o = the initial length of a glass log

It is conservatively assumed that the surface area parameter (S) remains constant as the glass corrodes. This approach is similar to that used in the TSPA-VA (CRWMS M&O 1998b, p. 6-79).

3.6.1.2 The Abstracted Model

The bounding rate expressions are obtained when the estimated value for the four model parameters are substituted into Equation 3.6-2. The result is as follows (CRWMS M&O 2000k, Section 6.2.3.3):

For the low pH leg ($pH < pH_m$),

$$\text{Rate/S } (g/(m^2 \cdot day) = 10^{(9\pm1)} \cdot 10^{(-0.6\pm0.1)\cdot pH} \cdot \exp((-58\pm15)/RT) \qquad \text{(Eq. 3.6-4)}$$

For the high pH leg ($pH \geq pH_m$),

$$\text{Rate/S } (g/(m^2 \cdot day) = 10^{(6.9\pm0.5)} \cdot 10^{(0.4\pm0.1)\cdot pH} \cdot \exp((-80\pm10)/RT) \qquad \text{(Eq. 3.6-5)}$$

where,

R = the gas constant, which is 8.314×10^{-3} kJ/(mol·K) (accepted value)
T = the temperature, in kelvins
pH = the negative base-10 logarithm of the hydrogen ion concentration in the water
pH_m = the pH at which the minimum dissolution rate occurs = 2.1 + 1149/T

$$pH_m = 2.1 + 1149/T \qquad \text{(Eq. 3.6-6)}$$

as shown in Figure 3.6-2.

3.6.2 Confidence/Limitations/Validation

The abstracted model is designed to provide a conservative estimate of the rate (i.e., an upper bound or overestimate of the likely rate) at which waste glass will corrode when immersed in groundwater or exposed to humid air and/or dripping water in the repository. The general algebraic form of the proposed model is widely accepted and used in the literature of waste glass corrosion (e.g., Strachan et al. 1994, House et al. 2000). Because each of the model parameter values were evaluated using a conservative bounding approach (CRWMS M&O 2000k, Section 6.4), there is confidence that the abstracted model is likely to bound the long-term HLW degradation under neutral to basic conditions. The dissolution rates calculated using Equation 3.6-5 can be compared directly with the dissolution rate of basalt glass recovered from the sea bed. The dissolution rates of several basalt glass samples were calculated based on the thickness of the layer of palagonite that forms as an alteration phase and the age of the basalt (Grambow et al. 1986, Table 2, Figure 3, pp. 268 to 269). The dissolution rates for basalts covered in sediment and exposed to Si-saturated seawater are about 0.1 μm/1000 yr, which is equivalent to 6×10^{-7} g/(m^2·day). The dissolution rates calculated using Equations 3.6-5 at 3°C are 3.6×10^{-6}, 9.1×10^{-6} and 2.3×10^{-5} g/(m^2·day) at pH values of 7, 8, and 9, respectively, which is the typical pH range for seawater. Thus, the rate expression to be used for basic conditions (i.e., Equation 3.6-5) is conservative with regard to the long-term dissolution rate of basalt glass.

If further model refinement is required in the future, this will be accomplished by confirmation testing as described in Section 17 of ASTM C1174-97 and/or by analysis of other appropriate experimental data that become available.

Based on the consideration discussed above, this model is appropriate for its intended use.

3.6.3 Other Views and Alternative Models

When a disposal container breaches, the glass logs may be exposed to humid air or dripping groundwater. A model was developed for the rate of corrosion of waste glass under humid air and dripping water conditions, and it is summarized below. The TSPA-SR analysis will not include a separate alteration model for these conditions because use of the immersion model is conservative.

In the unsaturated environment of the Yucca Mountain site, it is likely that waste glass will be contacted initially by humid air. When glass is exposed to humid air, molecular water will sorb onto specific sites on the glass surface, primarily silanol sites, and alkali metal sites. The amount of liquid water that sorbs on the glass will depend on the relative humidity of the air, temperature of the glass surface, and hygroscopicity of the glass surface. The first monolayer forms at a relative humidity of only a few percent. Subsequent monolayers form as water vapor bonds with sorbed water to form beads of water on the glass surface. At relative humidities above about 80 percent, a sufficient amount of water has condensed to coalesce into a thin film covering the entire surface. The glass will react with these sorbed water layers just as it does when immersed in water. Initial reactions will likely result in the dissolution of alkali metals into the film of water. This will decrease the equilibrium vapor pressure of the film of water and cause more water to condense in the film. After the initial film of water is sorbed, the amount of water on the glass will likely be determined by the salinity of the water.

In an open system such as the disposal system, water vapor will continually condense in the film of saline water on the exposed waste glass as the glass corrodes. For the various configurations in which fractured glass may be contacted by humid air or dripping water, water may drip or flow away from the glass or may accumulate over time while contacting the glass. Once alkali metals are released into the film of water, the hygroscopicity of the film will result in continuous condensation of water vapor. Continuous exposure to water-saturated air will result in a process of vapor condensation, flow across the sample, and dripping, wherein dissolved species can be transported away from the glass as solution drips from the glass, and fresh water vapor condenses. The corrosion rate of the glass under these conditions will be affected by the rates at which water vapor condenses in the film, and solution drips from the sample. These processes will affect the glass dissolution rate through their effects on the solution chemistry of the film. The effects of the condensation, flow, and drip rates on the glass dissolution rate are taken into account in the model by the use of experimental data measured under test conditions designed to replicate the phenomena discussed above.

The corrosion behavior of glass contacted by a thin film of water is modeled as a special case of aqueous corrosion that is described with the rate expression given below (Equation 3.6-7). Analysis of the available information (CRWMS M&O 2000k, Section 6.3.2) indicates that the same mechanistic rate expression used for the case of water immersion can be used for glass

contacted by humid air and dripping water. The abstracted rate expression for exposure to dripping water or to humid air at a relative humidity of 80 percent or higher:

$$\text{Rate} = S_{tf} \cdot k_{tf} \exp(-E_{tf}/RT) \qquad \text{(Eq. 3.6-7)}$$

where S_{tf} is the surface area contacted by humid air or dripping water. In this abstraction, the intrinsic dissolution rate (k_0), the pH dependence, and the affinity term ($1 - Q/K$) are combined into a single term called the thin film rate, k_{tf}. The use of this rate expression requires knowledge of the surface area that is contacted by a thin film of water due to exposure to humid air or dripping water and the temperature. The parameter values for the bounding rate are:

For relative humidity > 80 percent or exposure to dripping water,

$$\log k_{tf} = 9.2 \pm 0.2 \text{ g/(m}^2\cdot\text{day)}$$

$$E_{tf} = 80 \pm 10 \text{ kJ/mol}$$

Comparison of the model for the corrosion rate under immersion conditions with that summarized above for exposure to humid air and dripping water showed that for pH values greater than 5.75, use of the rate expression for immersion for all contact modes provides a conservative estimate for the glass dissolution rate. At pH values lower than 5.75, the rate expression for the acid leg for immersion should also be used for contact by humid air and dripping water. However, it is important to note that since laboratory tests results for contact by humid air and dripping water are in near neutral or alkaline solutions, no database exists to evaluate dissolution in acidic solutions under humid air or dripping conditions.

In the IRSR on CLST (NRC 1999a, Section 5.4.5), the NRC identified that DOE has not properly considered data on naturally occurring glasses, combined with experimental data and models on dissolution of HLW glasses, to demonstrate that long-term dissolution behavior under repository conditions can be represented by extrapolation of results from short-term laboratory tests. As stated more fully in Chapter 4, the bounding formulation of the HLW Degradation Component, discussed in Section 3.6, included the consideration of experimental data developed by the CRWMS M&O and other relevant data including information from natural analogs. Natural analog information is used for corroboration (see Section 3.6.2 and Cunnane et al. 1994).

3.7 DISSOLVED RADIOISOTOPE CONCENTRATION COMPONENT

The dissolved concentration component evaluates the dissolved concentration of radionuclides (or parents of radionuclides) that are important to human dose as determined by the radioisotope screening described in the inventory Section (3.1) (Figure 3.7-1).

Doses calculated for groundwater pathways from the repository to the environment depend critically on the concentrations of radionuclides in fluids issuing from breached WPs. While dissolution of radioisotope-containing SNF rods and/or HLW glass solids into incoming fluids provides the primary source term, the formation of secondary phases often limits the amounts of radionuclides available for subsequent groundwater transport.

The primary document supporting the radioisotope concentration model component is *Summary of Dissolved Concentration Limits* AMR (CRWMS M&O 2000l) (Figure 3.7-2). An important component of this AMR was the analysis with EQ3NR (a component of the qualified EQ3/6 Version 7.2b package, CSCI: UCRL-MA-110662; CRWMS M&O 1998a). Ultimately though, the values described in the radioisotope concentration model reflect a newly revised thermodynamic database developed in CRWMS M&O (2000v). New measurements of radionuclide solubilities (Efurd et al. 1998; Kaszuba and Runde 1999) were combined with a critical review of recent thermodynamic parameterizations of Np, U, Pu, and Am phase stabilities that are being used by the international waste disposal community (NEA OECD) at large.

Important components of this effort included the following.

- Measurement and critical analysis of Np solubility in J-13 groundwater (see Table 3.7.1 below). This led to revision of the formation free energy of $Np_2O_5 \cdot xH_2O$, an important Np-limiting phase, and Np speciation data (see Table 3.7.2 below).

- Measurement and critical analysis of Pu solubility in J-13 groundwater.

- Critical analysis of technetium and uranium silicate thermodynamic data

3.7.1 Models Description

The dissolved concentration limits calculation builds upon three primary feeds: (1) estimates of in-package fluid major element composition (pH, Eh, ionic strength, carbonate levels), (2) measured (and estimated) thermodynamic parameters describing the stabilities of aqueous species and solid radioisotope phases, and (3) a determination of the likely solubility-controlling phases for the specific radionuclides of concern. The in-package chemistry is described in Section 3.2 of this PMR. The thermodynamic databases that were used are described in the supporting AMRs (CRWMS M&O 2000l, 2000v).

The chosen solubility-controlling phase can affect the calculated radionuclide concentrations by orders of magnitude and is one of the sources for uncertainty in the predicted concentration. In nature, the controlling phase may either be a pure radionuclide solid phase with the radioisotope as the dominant element or a solid phase with trace amounts of the radionuclide as can occur with coprecipitation. For TSPA-SR analysis the pure phases were chosen because in general they yield higher dissolved concentrations. See CRWMS M&O (2000w) for discussion of the potential effects of secondary phases on radionuclide concentration. The phases were chosen based upon geologic and/or experimental observations or crystallochemical arguments. For example, uranium solids observed at ore bodies and mine tailings are used to provide some clues as to the phases likely to form when fuel elements dissolve. For non-naturally occurring radioelements such as Pu and Np, experimental observations must be relied upon. Where no information can be gleaned from field observations or experimental results, it is conservative to simply assume that the most amorphous and hydrated (hence, most soluble) oxide of the particular radioelement forms. Typically, the secondary phases that form first in low temperature (T ~25°C), natural systems are poorly-crystalline oxides and hydroxides whose solubility tends to be relatively high. Over time, secondary alteration minerals tend to become

more crystalline and less hydrous. Consequently, the dissolved limits decrease. Some exceptions to this trend exist. Crystalline plutonium oxides can lose their crystallinity and become amorphous due to structural damage to the crystal caused by the ejection of alpha particles. These issues are discussed in CRWMS M&O (2000l) for U, Np, Pu, and Am radioisotopes.

The amount of thermodynamic data available for the radionuclides, the sensitivity of the radionuclides solubilities on in-package chemistry, and the importance of the radionuclides to total system performance are quite uneven. For example, there is a large database for U, Np, and Am as a function of many chemical environments but very little for Pa. For these reasons, the implementation of solubility within the TSPA-SR analysis ranged from: (1) multi-termed functions of chemistry for U, Np, and Am, (2) distributions for Pu, Pa, Pb, and Ni, and (3) constant bounding values for Tc, I, Th, Cs, Sr, Cl, C, Nb, Zr, Ra, and Sn. For the multi-termed functions, the solubility of the radionuclide was calculated as a function of time for each simulation. For the distribution type, a value for the simulation was sampled from the distribution and used for the entire simulation. For the last type, the same bounding value was used for all times in all simulations (see Table 3.7-5).

The multi-termed functions describing U, Np, and Am solubilities, while developed from a statistical fit to predicted solubilities, possess some deterministic meaning as well. Specifically, the reactions affecting the solubilities of U, Np, and Am–carbonate complexation and hydrolysis–were identified beforehand. Hence, the solubilities can be resolved into, respectively, a P_{CO_2} and pH dependence. For many of the other radionuclides, where little data are available, use of a high value bounding solubilities does not result in high doses because of the low inventory or other factor. Detailed discussions for each radionuclide are provided in CRWMS M&O (2000l). Some highlights are provided here.

3.7.1.1 Uranium

Uranyl (UO_2^{2+}) minerals are expected to precipitate under the oxidizing conditions likely to prevail when breached WPs are initially altered by incoming fluids. Laboratory experiments and field observations show that the most common secondary uranyl minerals likely to form in the repository are schoepite, soddyite, uranophane, and/or Na-boltwoodite. For TSPA-SR analysis, schoepite was assumed to be the solubility-controlling mineral because: (1) schoepite is the first mineral to be formed during spent fuel corrosion; (2) field observations and modeling study indicate that schoepite can persist for more than 10,000 years; (3) schoepite is the most soluble of the common secondary phases; and (4) the temperature dependence of schoepite solubility is known.

EQ3NR calculations were performed with pHs in half units from 5 to 9, temperatures 30, 50, 70, and 90 Celsius, and log($f_{CO2[bar]}$) of −2.5, −3.0, −3.5, and −4.0. The calculated U solubility ranges from 5×10^{-7} to 4×10^{-4} mol/L, with an average of 2×10^{-5} mol/L. The following regression equation provides the best fit ($R^2 = 0.924$) to the 128 calculated points:

$$\log[U] = 7.9946 - 2.6963pH + 0.4292pH^2 - 1.6286\log f_{CO2} + 0.0095T + 0.4161pH \cdot \log f_{CO2} - 0.0051pH \cdot T - 0.0022\log f_{CO2} \cdot T \quad \text{(Eq. 3.7-1)}$$

where [U] is the total concentration of uranium in mg/L, f_{CO2} is the fugacity of CO_2 (bar), T is the temperature in kelvins (CRWMS M&O 2000l, Sections 6.3.5 and 6.3.7).

3.7.1.2 Neptunium

NpO_2 is expected to be the most stable Np-bearing phase under in-package conditions. However, NpO_2 has only rarely been observed to precipitate in laboratory experiments, presumably because kinetic obstacles prevent its nucleation and/or growth over short periods of time. Instead, the more soluble Np_2O_5 (or $Np[OH]_4$[am] under reducing conditions) are assumed to be the solubility-controlling minerals.

Table 3.7-1 shows measured Np solubilities in J-13 groundwater from which the new free energy of $Np_2O_5 \cdot xH_2O$ was derived. Data are measured as a function of pH and temperature from oversaturation (after 450 days equilibration) and undersaturation (after 151 days equilibration) (Efurd et al. 1998, p. 3896) and compared with literature data (Nitsche et al. 1993, p. 2). Uncertainties are given as one standard deviation.

Table 3.7-1. Averaged ^{237}Np Concentrations in mol/L in J-13 Groundwater

		pH 6	pH 7	pH 8.5
25°C	**Oversaturation**	$(6.5\pm1.1)\times10^{-4}$	$(3.1\pm0.2)\times10^{-5}$	$(1.5\pm0.6)\times10^{-5}$
	Undersaturation	$(6.5\pm1.0)\times10^{-4}$	$(2.9\pm0.7)\times10^{-5}$	$(1.5\pm0.3)\times10^{-5}$
	[Lit.]	$(5.3\pm0.3)\times10^{-3}$	$(1.3\pm0.2)\times10^{-4}$	$(4.4\pm0.7)\times10^{-5}$
60°C	**Oversaturation**	$(9.4\pm1.2)\times10^{-4}$	$(1.6\pm0.6)\times10^{-5}$	$(1.7\pm0.9)\times10^{-5}$
	Undersaturation	–	–	–
	[Lit.]	$(6.4\pm0.4)\times10^{-3}$	$(9.8\pm1.0)\times10^{-4}$	$(1.0\pm0.1)\times10^{-4}$
90°C	**Oversaturation**	$(9.1\pm1.8)\times10^{-4}$	$(8.6\pm2.3)\times10^{-6}$	$(5.8\pm2.5)\times10^{-6}$
	Undersaturation	$(8.7\pm0.9)\times10^{-4}$	$(9.3\pm1.9)\times10^{-6}$	$(5.9\pm2.1)\times10^{-6}$
	[Lit.]	$(1.2\pm0.1)\times10^{-3}$	$(1.5\pm0.4)\times10^{-4}$	$(8.9\pm0.4)\times10^{-5}$

Source: *Pure Phase Solubility Limits-LANL* ANL-EBS-MD-000017. (CRWMS M&O 2000v, Table 5)

Table 3.7-2 describes the output, updated thermodynamic parameters that were subsequently used in *Summary of Dissolved Concentration Limits* AMR (CRWMS M&O 2000l).

Table 3.7-2. Updated Thermodynamic Data for Np(IV,V) Species

Reaction or Species	$\Delta_f G°_{298}$ (kJ mol^{-1})
NpO_2^+	–907.90 ± 5.77
Np^{4+}	–491.10 ± 9.49
$NpO_2^+ + OH^-$ = **$NpO_2OH(aq)$**	–1080.61 ± 5.88
$NpO_2^+ + 2\ OH^-$ = **$NpO_2(OH)_2^-$**	–1247.33 ± 5.84
$NpO_2^+ + 2\ OH^- + CO_3^{2-}$ = **$NpO_2(OH)_2(CO_3)^{3-}$**	–1791.01 ± 7.36
$NpO_2^+ + OH^- + 2\ CO_3^{2-}$ = **$NpO_2(OH)(CO_3)_2^{4-}$**	–2155.41 ± 6.71
$NpO_2^+ + CO_3^{2-}$ = **$NpO_2CO_3^-$**	–1463.34 ± 5.84
$NpO_2^+ + 2\ CO_3^{2-}$ = **$NpO_2(CO_3)_2^{3-}$**	–2001.25 ± 5.92
$NpO_2^+ + 3\ CO_3^{2-}$ = **$NpO_2(CO_3)_3^{5-}$**	–2523.46 ± 5.80
$NpO_2^+ + F^-$ = **$NpO_2F(aq)$**	–1197.07 ± 8.55
$NaNpO_2CO_3{\cdot}3.5H_2O(s)$ = $Na^+ + NpO_2^+ + CO_3^{2-} + 3.5H_2O$	–2591.02 ± 5.86
$Na_3NpO_2(CO_3)_2(s)$ = $3\ Na^+ + NpO_2^+ + 2\ CO_3^{2-}$	–2831.01 ± 5.94
$KNpO_2CO_3(s)$ = $K^+ + NpO_2^+ + CO_3^{2-}$	–1795.97 ± 5.80
$K_3NpO_2(CO_3)_2(s)$ = $3\ K^+ + NpO_2^+ + 2\ CO_3^{2-}$	–2902.00 ± 5.81
$NpO_2(OH)\ (am)$ = $NpO_2^+ + OH^-$	–1115.26 ± 5.79
$NpO_2(OH)\ (cr)$ = $NpO_2^+ + OH^-$	–1119.31 ± 5.84
$Np_2O_5\ (s)$ + $2\ H^+ = 2\ NpO_2^+ + H_2O$	–2023.30 ± 12.41
$Np^{4+} + H_2O$ = **$NpOH^{3+}$** + H^+	–726.00 ± 10.30
$Np^{4+} + 4\ H_2O$ = **$Np(OH)_4(aq)$** + $4\ H^+$	–1382.74 ± 11.08
$Np(OH)_4(am)$ + $4\ H^+ = Np^{4+} + 4\ H_2O$	–1431.26 ± 9.65
$NpO_2(s)$ + $4\ H^+ = Np^{4+} + 2\ H_2O$	–1021.80 ± 2.5
$Np^{4+} + 5\ CO_3^{2-}$ = **$Np(CO_3)_5^{6-}$**	–3324.64 ± 11.26
$Np^{4+} + 4\ CO_3^{2-}$ = **$Np(CO_3)_4^{4-}$**	–2803.14 ± 11.36
$Np^{4+} + F^-$ = **NpF^{3+}**	–823.94 ± 9.55
$Np^{4+} + 2\ F^-$ = **NpF_2^{2+}**	–1144.10 ± 9.74

Source: *Pure Phase Solubility Limits-LANL* ANL-EBS-MD-000017. (CRWMS M&O 2000v, Table 2)

NOTE: Bolded species are those for which $\Delta_f G°_{298}$ is given.

Scoping calculations (CRWMS M&O 2000l, Section 6.4.3) indicated that the only important independent variables were pH and f_{CO2}. Calculations were performed with pHs in half units from 4.5 to 8.5, and log ($f_{CO2[bar]}$) of –2.5, –3.0, –3.5, and –4.0. The 32 calculated solubilities ranged from 2×10^{-6} to 1.5×10^{-2} mol/L, with an average of 2.3×10^{-3} mol/L. The best fit expression for Np solubility is:

$$[Np] = 7.538 \times 10^{-8} + 1.086 \times 10^{8-pH}, \; R^2 = 0.9997 \qquad \text{(Eq. 3.7-2)}$$

where [Np] is the total concentration of neptunium in mg/L. Since f_{CO2} was significant only when pH was above the range of interest for in-package chemistry, it was not included in Equation 3.7-2. Figure 3.7-3 shows a comparison of the abstraction (Equation 3.7-2) and the under saturation data for 25°C as listed in Table 3.7-1.

3.7.1.3 Plutonium

The primary candidates for solubility-controlling Pu phases are PuO_2 and $Pu(OH)_4$(am) (or $PuO_2 \cdot xH_2O$, with x varying from 2 to zero). Since the crystalline phase forms within laboratory time scale, it is reasonable to assume that over geological time, plutonium hydroxides will convert to PuO_2, and it should be used as the solubility-controlling mineral for Pu for the repository. However, α-decay of Pu isotopes has been observed to decrease the crystallinity of PuO_2, while at the same time Pu hydroxide can gradually convert to anhydrous, though poorly crystalline, material. In order to be conservative, $Pu(OH)_4$(am) was chosen as the controlling phase. The recommended thermodynamic data for Pu are shown in Table 3.7-3.

Table 3.7-3. Recommended Thermodynamic Data for Pu(III,IV,V,VI) Species

Reaction or Species	$\Delta_f G^o_{298}$ (kJ mol^{-1})
PuO_2^{2+}	–767.0 ± 6.6
PuO_2^{+}	–857.5 ± 6.7
Pu^{4+}	–477.8 ± 3.4
Pu^{3+}	–578.6 ± 3.3
$PuO_2^{2+} + OH^- =$ **PuO_2OH^+**	–970.5 ± 6.6
$PuO_2^{2+} + 2\ OH^- =$ **$PuO_2(OH)_2(aq)$**	–1162 ± 6.6
$PuO_2^{2+} + CO_3^{2-} =$ **$PuO_2CO_3\ (aq)$**	–1348.6 ± 6.8
$PuO_2^{2+} + 2\ CO_3^{2-} =$ **$PuO_2(CO_3)_2^{2-}$**	–1906.9 ± 6.8
$PuO_2^{2+} + 3\ CO_3^{2-} =$ **$PuO_2(CO_3)_3^{4-}$**	–2449.1 ± 6.8
$PuO_2^{2+} + F^- =$ **PuO_2F^+**	–1075.1 ± 6.8
$PuO_2^{2+} + 2\ F^- =$ **$PuO_2F_2(aq)$**	–1371.6 ± 7.3
$PuO_2^{+} + H_2O =$ **$PuO_2OH(aq)$** $+ H^+$	–1039.1 ± 6.7
$PuO_2^{+} + CO_3^{2-} =$ **$PuO_2CO_3^-$**	–1414.7 ± 6.7
$PuO_2^{+} + 2\ CO_3^{2-} =$ **$PuO_2(CO_3)_2^{3-}$**	–1951.2 ± 8.8
$PuO_2^{+} + 3\ CO_3^{2-} =$ **$PuO_2(CO_3)_3^{5-}$**	–2469.4 ± 7.8
$Pu^{4+} + H_2O =$ **$PuOH^{3+}$** $+ H^+$	–712.2 ± 4.1
$Pu^{4+} + 4\ H_2O =$ **$Pu(OH)_4$** $+ 4\ H^+$	–1375 ± 7
$Pu^{4+} + 5\ CO_3^{2-} =$ **$Pu(CO_3)_5^{6-}$**	–3109.7 ± 3.5
$Pu^{4+} + F^- =$ **PuF^{3+}**	–809.9 ± 6.5
$Pu^{4+} + 2\ F^- =$ **PuF_2^{2+}**	–1131.2 ± 6.7
$Pu^{3+} + H_2O =$ **$PuOH^{2+}$** $+ H^+$	–775.8 ± 4.4
$Pu(OH)_3(s)$ $= Pu^{3+} + 3\ OH^-$	–1200.1 ± 5.7
$PuO_2{\cdot}2H_2O$ $= Pu^{4+} + 4\ OH^-$ or **$Pu(OH)_4(am)$** $= Pu^{4+} + 4\ OH^-$	–1432 ± 17
$PuO_2(cr)$ $+ 2\ H_2O = Pu^{4+} + 4\ OH^-$	–976.2 ± 3.6
$PuO_2(OH)_2(s)$ $= PuO_2^{2+} + 2\ OH^-$	–1211.2 ± 9.1
$PuO_2(OH)(am)$ $= PuO_2^{+} + OH^-$	–1065.6 ± 7.5
$PuO_2CO_3(s)$ $= PuO_2^{2+} + CO_3^{2-}$	1376.1 ± 6.8

Source: *Pure Phase Solubility Limits-LANL* ANL-EBS-MD-000017 (CRWMS M&O 2000v, Table 3)

NOTE: Bolded species are those for which $\Delta_f G°_{298}$ is given. Bolding was absent from the source document but was added in consultation with the author.

EQ3NR calculations were performed with pHs in half units from 4 to 8, Eh (V) of 0.34, 0.55, and 0.76, and log ($f_{CO2[bar]}$) of −3.0, and −3.5. The calculated Pu solubility ranged from 10^{-10} mol/L at pH 8 and Eh 0.34 V to 2×10^{-4} mol/L at pH 8 and Eh 0.76 V. Because the data points were more uniformly distributed on a log than a linear scale, a log-uniform distribution was assigned (Table 3.7-5).

A recent study (Haschke et al. 2000) on the reaction of PuO_2 with water has found that in the presence of water and oxygen, PuO_2 may be metastable and can be converted into PuO_{2+x}. In other words, Pu may exist as Pu(VI) in the solid, and consequently in the solution. This new finding has raised the concern that Pu may be more soluble than previously believed, just as the conversion of U(IV) into U(VI) increases U solubility. This concern, however, is mitigated by the use of the much more soluble $Pu(OH)_2$(am) instead of PuO_2(c) as the solubility controlling solid. While PuO_{2+x} may be more soluble than PuO_2(c), it is not clear that PuO_{2+x} is more soluble than $Pu(OH)_2$(am). Also, the recent identification of PuO_{2+x} does not preclude its presence in previous PuO_2 solubility experiments, some of which were conducted for several hundred days. Only further study of PuO_{2+x} and other possible solubility controlling solids will provide definitive answers to these questions. Because $Pu(OH)_2$(am) solubility is quite high relative to dissolved Pu concentrations observed in spent fuel tests, this model is believed to be conservative.

3.7.1.4 Americium, Actinium, Curium, and Samarium

$AmOHCO_3$ was chosen as the conservative solubility controlling phase (CRWMS M&O 2000l, Section 6.6 for discussion) for Am. Am solubility for 46 calculations ranged from about 2×10^{-8} to 0.19 mol/kg as a function of pH and f_{CO_2}:

$$\log[Am] = 58.0335 - 18.9422 \cdot pH + 1.4744 \cdot pH^2 - 6.0032 \cdot \log(f_{CO_2}) - 0.7005 \cdot (\log f_{CO_2})^2 + 0.1162 \cdot pH^2 \cdot \log(f_{CO_2}) + 0.1146 \cdot pH \cdot (\log f_{CO_2})^2 \quad \text{(Eq. 3.7-3)}$$

where [Am] is the concentration of americium in units of mg/L (Figure 3.7-4).

Because of chemical similarities, the response functions for Ac, Cm, and Sm were set identical to that of Am (with adjustments for atomic weight).

3.7.1.5 Technetium, Carbon, Chlorine, Iodine, Cesium, and Strontium

Under the conservative assumption of oxidizing repository conditions, no solids are predicted to form to limit the solubility of Tc, C, Cl, I, and Cs. Therefore, the solubility of each is set to 1.0 mol/L, which will let the waste inventory control their respective release. The recommended thermodynamic data for Tc are shown in Table 3.7-4.

Strontium compounds are potentially quite soluble. Under repository conditions, strontium may coprecipitate with uranyl solids, or it may precipitate as carbonate or sulfate. In lieu of a more involved treatment, the Sr solubility is set to 1.0 mol/L, which will let the waste inventory control Sr release.

Table 3.7-4. Recommended Thermodynamic Data for Tc Species

Reaction or Species	$\Delta_f G°_{298}$ (kJ mol^{-1})
TcO_4^-	–637.2 ± 7.6
Tc(s) + 1.75 O_2(aq) + 0.5 H_2O = TcO_4^- + H^+	0
Tc^{3+} + O_2(aq) + 2 H_2O = TcO_4^- + 4 H^+	92.4 ± 10.5
TcO^{2+} + 0.75 O_2(aq) + 1.5 H_2O = TcO_4^- + 3 H^+	–114.0 ± 10.5
TcO_4^{2-} + H^+ + 0.25 O_2(aq) = 0.5 H_2O + TcO_4^-	–578.3 ± 10.0
TcO_4^{3-} + 2 H^+ + 0.5 O_2(aq) = 1.0 H_2O + TcO_4^-	–521.4 ± 11.2
$HTcO_4$(aq) = TcO_4^- + H^+	–637.1
$(TcO(OH)_2)_2$(aq) + 4 H^+ = 2 TcO^{2+} + 4 H_2O	–1172.2 ± 14.2
H_2TcO_4(aq) = TcO_4^{2-} + 2 H^+	–629.7 ± 12.1
$HTcO_4^-$ = TcO_4^{2-} + H^+	–628.0 ± 10.5
$TcO(OH)_2$(aq) + 2 H^+ = TcO^{2+} + 2 H_2O	–567.5 ± 7.1
$TcO(OH)^+$ + H^+ = TcO^{2+} + H_2O	–344.7 ± 10.5
$TcO(OH)_3^-$ + H^+ = $TcO(OH)_2$(aq) + H_2O	–742.52 ± 7.10
$Tc(OH)_2CO_3$(aq) = $TcO(OH)_2$(aq) + CO_2(g)	–667.91 ± 7.20
$Tc(OH)_3CO_3^-$ + H^+ = $TcO(OH)_2$(aq) + CO_2(g) + H_2O	–857.95 ± 7.20
$HTcO_4$(s) = TcO_4^- + H^+	–603.2 ± 10.5
$KTcO_4$(s) = TcO_4^- + K^+	–932.7 ± 7.6
$NaTcO_4$(s) = TcO_4^- + Na^+	–890.4 ± 10.1
$CsTcO_4$(s) = TcO_4^- + Cs^+	–949.8 ± 9.8
$Tc(OH)_2$(s) + 3 H^+ + 0.25 O_2(aq) = Tc^{3+} + 2.5 H_2O	–460.3 ± 3.8
$Tc(OH)_3$(s) + 3 H^+ = Tc^{3+} + 3 H_2O	–656.6 ± 3.8
Tc_2O_7(s) + H_2O = 2 TcO_4^- + 2 H^+	–943.2 ± 15.0
Tc_3O_4(s) + 9 H^+ + 0.25 O_2(aq) = 3 Tc^{3+} + 4.5 H_2O	–861.4 ± 3.8
Tc_4O_7(s) + 10 H^+ = 2 Tc^{3+} + 2 TcO^{2+} + 5 H_2O	–1278.4 ± 21.0
$TcO_2 \cdot 2H_2O$(am) + 2 H^+ = TcO^{2+} + 3 H_2O	–849.7 ± 7.0
TcOH(s) + 3 H^+ + 0.5 O_2(aq) = Tc^{3+} + 2 H_2O	–233.4 ± 3.8
TcS_2(s) + H_2O = TcO^{2+} + 2 HS^-	–229
TcS_3(s) + 4 H_2O = TcO_4^{2-} + 3 HS^- + 5 H^+	–276
Tc_2S_7(s) + 8 H_2O = 2 TcO_4^- + 7 HS^- + 9 H^+	–594
TcO_2(cr)	–404±8

Source: *Pure Phase Solubility Limits-LANL* ANL-EBS-MD-000017 (CRWMS M&O 2000v, Table 4)

NOTE: Bolded species are those for which $\Delta_f G°_{298}$ is given. Bolding was absent from the source document but was added in consultation with the author.

3.7.1.6 Nickel, Protactinium, and Lead

A brief EQ3NR calculation carried out in the AMR titled *Pure Phase Solubility Limits–LANL* (CRWMS M&O 2000v, Section 6.3.6) gives the range of Ni solubility for Yucca Mountain waters as 1.4×10^{-6} to 3.1 mol/L, provided that bunsenite (NiO) is the solubility controlling mineral. No effort has been made to generate the distribution type for Ni solubility. A log-uniform distribution is proposed for Ni solubility for TSPA-SR, with a minimum of 1.4×10^{-6} and a maximum of 3.1 mol/L.

Due to the lack of thermodynamic data for Pa, no data compilation has been carried out in the *Pure Phase Solubility Limits-LANL* (CRWMS M&O 2000v). Thus, it is not possible to conduct EQ3NR calculations for Pa solubility. The process AMR suggests a solubility range of 1.0×10^{-5} to 1.0×10^{-10} mol/L. This range is the same as Pa solubility values used in TSPA-93, which was obtained by the Project experts elicitation process (Wilson et al. 1994, Table 9-2b). The project experts further suggested a log-uniform distribution over that range. Therefore, for TSPA-SR, a log-uniform distribution for Pa solubility is recommended, with a minimum of 1.0×10^{-10}, a maximum of 1.0×10^{-5} (CRWMS M&O 2000l, Table 19), and a mean of 3.2×10^{-8} mol/L.

In carbonate containing waters, $Pb_3(CO_3)_2(OH)_2$ is believed to be a major important solid phase for constraining Pb solubility (CRWMS M&O 2000v, Section 6.3.11). Unfortunately, EQ3/6 databases do not contain thermodynamic data for this solid. The process AMR (CRWMS M&O 2000v, Section 6.3.11) claims that, without thermodynamic data for that solid, calculated Pb(II) solubility is suspect. Based on reported Pb(II) solubility range in carbonate containing waters, the process AMR suggests a solubility range of 1.0×10^{-10} to 1.0×10^{-5} mol/L in Yucca Mountain waters.

The above range is very close to the solubility range of Pb (1.0×10^{-8} to 1.0×10^{-5} mol/L, log-beta distribution) used in the TSPA-93 (Wilson et al. 1994, Table 9-2b), which is suggested by the Project experts.

It is recommended that TSPA-SR use a log-uniform distribution, with a minimum of 1.0×10^{-10}, and a maximum of 1.0×10^{-5} mol/L (CRWMS M&O 2000l, Table 19), to constrain Pb solubility.

3.7.1.7 Niobium, Radium, Tin, Thorium, and Zirconium

A study of Nb solubility has been conducted and documented in *Pure Phase Solubility Limits-LANL* (CRWMS M&O 2000v, Section 6.3.5) using EQ3NR. It suggests that Nb solubility ranges from 1.0×10^{-15} to 1.0×10^{-7} mol/L. It is recommended that TSPA-SR use a constant solubility of 1.0×10^{-7} mol/L for Nb. This abstraction is conservative and thus, adequate for use in TSPA-SR.

Radium solubility has also been studied briefly in *Pure Phase Solubility Limits–LANL* (CRWMS M&O 2000v, Section 6.3.7). For Yucca Mountain waters, Ra solubility ranges from 1.1×10^{-6} to 2.3×10^{-6} mol/L. A constant solubility of 2.3×10^{-6} mol/L is recommended for Ra.

The calculated solubility range of tin reported in *Pure Phase Solubility Limits-LANL* (CRWMS M&O 2000v, Section 6.3.9) for Yucca Mountain waters is very narrow, from 4.8×10^{-8} to 4.9×10^{-8} mol/L. Thus, a constant solubility of 5.0×10^{-8} mol/L for Sn is suggested for TSPA-SR.

The AMR titled *Pure Phase Solubility Limits–LANL* (CRWMS M&O 2000v) does not provide a Th solubility range. The major reason for this omission is that the EQ3/6 current databases do not include thorium-carbonate species. As experiments have demonstrated that Th solubility may increase dramatically if the concentration of carbonate is high enough (Rai et al. 1995), model calculations without thorium-carbonate species are not meaningful.

As a work around solution, a constant solubility of 1.0×10^{-5} mol/L is proposed for Th (CRWMS M&O 2000l, Table 19). A small amount of corroborating data support this proposed value: 1) the 1993 Expert Elicitation of the Project gives the Th solubility range of 10^{-10} to 10^{-7} mol/L; 2) NRC's total-system performance assessment code version 3.2 (Center for Nuclear Waste Regulatory Analyses 1998, p. A-38) uses 2.3×10^{-4} kg/m^3 = 10^{-6} mol/L as the solubility limit for Th, with consideration of thorium-carbonate species. In fact, this proposed value is one to two orders of magnitude higher than these corroborating data, and thus, is conservative and adequate for use in TSPA-SR.

The Th thermodynamic data are being updated. Th carbonate complex data have been entered into a new input file (data0.ymp.R0). A qualification plan for this input file using the technical assessment method has been prepared. Once the data file is qualified by the Project, modeling calculations for Th solubility will be conducted and new values of Th solubility limits will be generated.

Another brief EQ3NR calculation documented in the AMR titled *Pure Phase Solubility Limits-LANL* (CRWMS M&O 2000v, Section 6.3.10) generates a Zr solubility for Yucca Mountain waters of 6.8×10^{-10} mol/L (6.20×10^{-5} mg/L). This value is recommended for TSPA-SR.

Table 3.7-5. Dissolved Concentration Limits for TSPA-SR

Element	Distribution Type	Min (mol/L)	Max (mol/L)
Am	Function	~$10^{-7.7}$	~0.19
Ac	Function	~$10^{-7.7}$	~0.19
Sm	Function	~$10^{-7.7}$	~0.19
Cm	Function	~$10^{-7.7}$	~0.19
C	Constant	1	1
Cl	Constant	1	1
Cs	Constant	1	1
I	Constant	1	1
Nb	Constant	10^{-7}	10^{-7}
Ni	Log-Uniform	$10^{-5.9}$	$10^{0.5}$
Np	Function	~$10^{-5.7}$	~$10^{-1.8}$

Table 3.7-5. Dissolved Concentration Limits for TSPA-SR (Continued)

Element	Distribution Type	Min (mol/L)	Max (mol/L)
Pa	Log-Uniform	10^{-10}	10^{-5}
Pb	Log-Uniform	10^{-10}	10^{-5}
Pu	Log-Uniform	10^{-10}	$10^{-3.7}$
Ra	Constant	$10^{-5.6}$	$10^{-5.6}$
Sn	Constant	$10^{-7.3}$	$10^{-7.3}$
Sr	Constant	1	1
Tc	Constant	1	1
Th	Constant	10^{-5}	10^{-5}
U	Function	$\sim 10^{-6.3}$	$\sim 10^{-3.4}$
Zr	Constant	$10^{-9.2}$	$10^{-9.2}$

Source: Excerpted from AMR: *Summary of Dissolved Concentration Limits* (CRWMS M&O 2000l, Table 19) plus Sections 3.7.1.1 and 3.7.1.4 of this report

3.7.2 Confidence/Limitations/Validation

A systematic review of thermodynamic data and controlling phases has been performed for a large range of chemical conditions. When uncertainties were encountered, choices were made that would result in higher predicted solubilities. Inherent limitations within the databases can lead to an uncertainty up to a factor of 2 when the ionic strength exceeds about 0.7 mol/L. However, this uncertainty is small relative to other uncertainties within the system. The net result of these efforts is that predicted radionuclide levels closely approximate values measured in the laboratory. Equations for Pu solubility predict 25°C pH 6 to 8.5 levels between 1×10^{-10} and 2×10^{-4} (CRWMS M&O 2000l, Section 6.5.4) compared to measured values of 9.4×10^{-9} to 1.1×10^{-6} (CRWMS M&O 2000v, Table 6). Parameterizations for Np solubility at pH < 8.1 (see CRWMS M&O 2000l, Section 6.4) predict 25°C pH 6 and 7 values of respectively 4.6×10^{-4} and 4.6×10^{-5} mol/L of Np. For comparison, measured Np values (CRWMS M&O 2000v, Table 5) are respectively 6.5×10^{-4} and 3.1×10^{-5}.

Based on the considerations discussed above, this model is appropriate for its intended use.

3.7.3 Other Views

The TSPA peer review panel (Budnitz et al. 1999, Executive Summary, Section E, Data Research Needs) identified that there were several areas for which the need for data had not been met including identifying that solubility limit distributions for several key radionuclides come from a limited experimental database and suggesting further experimental study. It also identified difficulty in applying the conceptual model used to define solubility-limited concentrations (Budnitz et al. 1999, Section IV.G). In the VA (DOE 1998b, Section 6.5.2.7) DOE acknowledged the need for additional research in the areas of neptunium solubility and technetium sorption on degraded WP materials. It noted comments from the peer review panel that identified the need for experimental data to confirm the processes that control neptunium

solubility (DOE 1998b, Section 6.5.2.2). DOE also acknowledged (DOE 1998b, Section 6.5.3.9) the need to support the basis for neptunium solubility with applicable measurements under suitable conditions.

Several substantial changes were made in the Dissolved Radioisotope Concentration Component to address these concerns. Thermodynamic data were thoroughly reviewed for Np, and a regression model was built such that the solubility limit could be coupled to the estimated in package chemical environment as discussed in Section 3.2. Because experiments of the incorporation of Np into secondary phase minerals of U were not conclusive, the mechanism to potentially reduce solubility was not incorporated into the Dissolved Radioisotope Concentration Component (CRWMS M&O 2000w). The detailed process modeling to develop a regression model was also applied to U and Am.

The distributions of three other key radionuclides (Pu, Th, and Ni) were carefully reevaluated in conjunction with some process modeling and knowledge of expected in-package chemical conditions. Finally, values for other key radionuclides (Tc, I, C, Nb, Zr, Pa, Pb, Ra, Sn, Cs, and Cl) were reevaluated through literature review process model calculations and were fixed at bounding values. The solubility values for radionuclides determined by the informal expert elicitation conducted in 1993 (Wilson et al. 1994) were used only to corroborate the newly evaluated distributions and fixed values.

3.8 COLLOIDAL RADIONUCLIDE CONCENTRATION COMPONENT

3.8.1 Introduction

3.8.1.1 Function of Colloidal Concentration Component

The function of the colloidal radioisotope concentration component is to calculate the concentration of colloid-associated radionuclides that may be transported from the waste package. The approach described herein is consistent with the approach used in the EBS PMR (CRWMS M&O 2000y).

3.8.1.2 Radionuclides Susceptible to Colloidal Transport

Colloid transport is potentially important for radionuclide elements that have low solubility and can be entrained in, or sorbed onto, waste form, engineered barrier, or geologic barrier materials that form colloidal particle substrates. Of these radionuclides, only those that are a major part of the waste inventory and have potentially large dose conversion factors are of potential importance to the performance of the disposal system. Considering these screening criteria, plutonium is the dominant radionuclide and was treated in TSPA-VA (CRWMS M&O 1998b, p. 6-97). Americium is also considered for the TSPA-SR analysis.

3.8.1.3 Colloid Types

The radionuclide-bearing colloids are formed by radionuclide entrainment as discrete phases in, or by radionuclide sorption onto, colloidal substrates. They are, therefore, classified into “types” based on the source of the substrate material and can be further classified based on how the radionuclides are attached to the substrates. Three major types of colloids are recognized to be

important for the colloidal radionuclide concentration component based on the source of the colloid substrate material:

1. Waste-form colloids formed during degradation of HLW glass (Note: these colloids are further classified into "reversibly attached" and "irreversibly attached" radionuclide types)

2. Corrosion-product colloids formed during corrosion of iron-containing waste packages.

3. Groundwater colloids present in the waste-form area.

Based on laboratory evidence, waste-form colloids are produced from degradation of borosilicate HLW glasses. Fewer colloids have been observed in laboratory testing of CSNF, and experimental data are not yet available for defense spent-nuclear fuel DSNF and other waste forms, although testing is underway at NSNFP. The waste-form colloids that have been observed consist principally of smectite-type clay minerals. The laboratory evidence suggests that as HLW glass degrades, colloids are generated and often contain "entrained" plutonium. The engulfed solid inclusions of plutonium may be isolated from the aqueous system by the colloid itself and so may not be in equilibrium with the surrounding aqueous environment. Therefore, the engulfed plutonium in the waste-form colloids is conservatively assumed to be irreversibly associated with the waste form colloids.

Corrosion-product colloids consist of ferric iron-(hydr)oxide minerals, primarily goethite (FeOOH) and hematite (Fe_2O_3) that are assumed to form as a result of corrosion of the WP materials (CRWMS M&O 2000m, Section 6.1.1).

Groundwater colloids potentially present in the repository include those already present in the geosphere such as microbes, organic macromolecules (humic substances, including humic and fulvic acids), and mineral colloids (primarily clay minerals, silica, and iron-[hydr]oxide minerals). Microbe-facilitated radionuclide transport is disregarded because the relatively large size of microbes makes them susceptible to filtration. An assessment of the concentration of humic substances in groundwaters collected near the Yucca Mountain was conducted by Minai et al. (1992). In that study, humic substances were extracted from J-13 well water and experiments were conducted to characterize site binding densities and complexation with Am. Considering the presence of calcium in J-13 well water, the authors estimated that the effective complexation capacity of humic substances is about 2.3×10^{-10} eq/L (equivalents per liter) at pH 6.9 and 2.7×10^{-11} eq/L at pH 8.2. Based on this very low complexation capacity of humic substances in this system, they are not considered to be important groundwater colloids. Clays, silica, hematite, and goethite colloids occur in groundwater in the vicinity of Yucca Mountain, and it is assumed that small quantities of these colloids will enter a failed WP and be available to interact with released radionuclides. Consequently, mineral colloids are considered and will, henceforth, be referred to generically as groundwater colloids. These groundwater colloids are assumed to consist of smectite clays, which are fairly strongly sorbing minerals, and have been recognized at Yucca Mountain (Kingston and Whitbeck 1991). This assumption is based on the fact that smectite colloids are the dominant colloid in groundwaters at the Nevada Test Site and

the fact that it is a conservative choice because clay minerals are more sorptive than silica and are comparable to iron-(hydr)oxide minerals.

Radionuclides are attached to colloids as a result of two types of processes. For waste form colloids, radionuclides can be incorporated into the substrate material before it is suspended as colloids. All three colloid types may sorb radionuclides to form pseudocolloids, depending on the affinity of the colloid mineral substrate for a dissolved radionuclide. These two types of attachment processes are referred to as irreversible and reversible attachment, respectively.

As illustrated in Figure 3.8-1, the colloid source term includes contributions from the various types of colloids, discussed above (i.e., waste-form, corrosion-product, and groundwater colloids with reversibly and irreversibly attached radionuclides). The contributions of each colloid type are summed to produce the mobile colloid source term for each important radionuclide.

3.8.1.4 Colloid Stability in the Waste-Form Environment

Like other mineral colloids, the stability of waste form, corrosion-product, and groundwater colloids are controlled by their surface charge and the associated electrostatic forces that influence colloid/colloid particle interactions. These surface charges and electrostatic forces are affected by the solution ionic strength and pH conditions.

Surface charge on metal-(hydr)oxide and phyllosilicate minerals develops as hydroxyl functional groups protonate and deprotonate. At relatively low pH values, (pH's lower than the zero point of charge of the respective mineral) these minerals acquire a positive surface charge, due to protonation. Conversely, at high pH values, (pH's greater than the pH of zero surface charge), these minerals acquire a negative surface charge, due to deprotonation. Other minerals may develop a charge due to reactions of carbonate, phosphate, and silinol surface functional groups. Each mineral has a particular pH where the mineral surface is neutral, called the pH at zero-point of charge (pH_{ZPC}). At the pH_{ZPC}, dispersions of colloidal particles composed of a particular mineral tend to be destabilized.

Colloidal particles are maintained in suspension by Brownian forces when electrostatic forces inhibit colloidal particles from achieving sufficiently close proximity that the particles can stick and agglomerate due to Van Der Waals interactions. However, as the ionic strength of the system is increased to a threshold value (the critical coagulation concentration or c.c.c.), the thickness and effectiveness of the protective electrostatic shell surrounding the particles diminishes, and the particles no longer repel one another during collisions. As a result, the colloidal particles agglomerate and eventually become large enough so that they settle out of suspension due to gravitational forces.

3.8.1.5 Implementation of Colloidal Concentration Component

The colloid concentration component is implemented by summing the contributions of the three colloid types to the colloid radioisotope concentration for each radionuclide.

A model was developed to calculate the colloid radioisotope concentration for each of the three colloid types. The model for the waste form colloids includes the contributions of the engulfed (irreversible attached) plutonium observed in waste glass tests. The models for all three colloid

types include reversibly attached radionuclides. These models are based on the population of each colloid type (expressed in terms of mass of colloids per volume of fluid) and experimental data for the sorption of radionuclides onto the colloid substrate materials involved. The effects of pH and ionic strength on the stability of dispersions of each colloid type are considered. The location of the boundaries between pH and ionic strength regimes in which each type of colloid substrate is stable or unstable are defined together with the colloid mass concentration in each regime.

The model is shown schematically in Figure 3.8-2.

3.8.1.6 Documents Supporting Colloidal Concentration Component

This section is based on the model abstraction AMR *Waste Form Colloid-Associated Concentrations Limits: Abstraction and Summary* (CRWMS M&O 2000m), the model abstraction AMR draws on Yucca Mountain research conducted at Argonne National Laboratory (ANL) (CRWMS M&O 2000x, Section 6) and Los Alamos National Laboratory (LANL).

3.8.2 Model Description

In this section, the technical basis for the model subcomponents, the abstraction, and logic diagrams illustrating the implementation in TSPA are described. In reading the section, Table 3.8-1 should be consulted for parameter definitions and values discussed in the section. The section starts with a description of how radionuclides are attached to colloidal substrates. This is followed by a description of how the colloid radioisotope concentrations are modeled for each colloid type.

3.8.2.1 Radionuclide Uptake Mechanisms

This section addresses the modeling of radionuclide attachment/detachment for colloidal substrates. It is based on the experimental evidence that the attachment/detachment processes can be divided into two categories. The first involves incorporation of the radionuclides into waste form alteration material that subsequently becomes suspended as colloidal material. The second involves attachment of dissolved radionuclides to preexisting colloids. Because attachment of radionuclides by sorption onto the colloid substrate material is important for all three types of colloids, it is discussed first.

Table 3.8-1. Parameter Names, Definitions, and Values

Parameter Name	Parameter Description	Units[a]	Basis for Value/Source	Value and Distribution
Waste-form colloids Figure 3.8-6				
$M_{coll,wf,both}$	Total mass of both types of waste-form colloids per unit volume or mass of water (assumes that the mass contributed by reversibly attached radionuclides is negligible)	mg/L	d	d
$C_{RNcoll,wf,irrev,max}$	Highest observed or expected concentration of radionuclide element RN associated with both types of waste-form colloids	mol/L	CRWMS M&O 2000m, Section 6.2.2	6×10^{-8} mol/L[b]
$C_{RNcoll,wf,irrev,min}$	Lowest observed or expected concentration of radionuclide element RN associated with both types of waste-form colloids	mol/L	CRWMS M&O 2000m, Section 6.2.2	1×10^{-11} mol/L[b]
$M_{coll,wf,both,max}$	Highest observed or expected mass of both types of waste-form colloids per unit volume or mass of water	mg/L	e	5 mg/L[c]
$C_{RNcoll,wf,irrev}$	Concentration of irreversibly attached (or engulfed as a solid inclusion) radionuclide element RN associated with waste-form colloids	mol/L	d	d
$C_{RNcoll,wf,rev}$	Concentration of reversibly attached radionuclide element RN associated with waste-form colloids	mol/L	d	d
$M_{coll,wf,both}$	Mass of waste-form colloids with reversibly sorbed or irreversibly attached (or engulfed) radionuclide element RN per unit volume or mass of water	mol/L	d	d
$I_{lo\text{-}thresh,coll,wf}$	Ionic strength below which waste-form colloids are stable	mol/L	Figure 3.8-4 (Pu); Figure 3.8-5	0.01 mol/L
$I_{hi\text{-}thresh,coll,wf}$	Ionic strength above which waste-form colloids are unstable; also used for groundwater colloids	mol/L	Figure 3.8-4 (Pu); Figure 3.8-5	0.05 mol/L
$K_{d,RNcoll,wf,rev}$	Distribution coefficient for reversible sorption of radionuclide element RN onto waste-form colloids	mL/g colloid	Various sources – refer to Section 3.8.2.2.1	Geometric mean values: Pu = 10,000 Am = 100,000 log-normal distributions; geometric std. dev. = 1 order-of-magnitude
$ST_{coll,wf,is}$	Function relating stability of waste-form colloids to ionic strength, based on results of HLW glass degradation experiments (based on Pu concentration)	mol/L	CRWMS M&O 2000m, Section 6.2.2	[Pu colloid] = $-1.25 \times 10^{-6} \times I + 7.25 \times 10^{-8}$
$ST_{coll,wf,pH}$	Function relating stability of waste-form colloids to ionic strength and pH, over limited pH range, based on results of Montmorillonite stability experiments; also used for groundwater colloids	mol/L	Figure 3.8-5	I = pH/200

Table 3.8-1.Parameter Names, Definitions, and Values (Continued)

Parameter Name	Parameter Description	Units[a]	Basis for Value/Source	Value and Distribution
iron-(hydr)oxide colloids Figure 3.8-8				
$C_{RNcoll,FeOx}$	Concentration of reversibly attached radionuclide element RN associated with iron-(hydr)oxide colloids	mol/L	d	d
$M_{coll,FeOx}$	Mass of iron-(hydr)oxide colloids per unit volume or mass of water	mg/L	d	d
$M_{coll,FeOx,max}$	Highest observed or expected mass of iron-(hydr)oxide colloids per unit volume or mass of water	mg/L	(CRWMS M&O 2000m, Table 1)	1 mg/L
$M_{coll,FeOx,min}$	Lowest observed or expected mass of iron-(hydr)oxide colloids per unit volume or mass of water	mg/L	(CRWMS M&O 2000m, Table 1)	1×10^{-3} mg/L
$I_{lo\text{-}thresh,coll,FeOx}$	Ionic strength below which iron-(hydr)oxide colloids are stable	mol/L	Figure 3.8-7	0.01 mol/L
$I_{hi\text{-}thresh,coll,FeOx}$	Ionic strength above which iron-(hydr)oxide colloids are unstable	mol/L	Figure 3.8-7	0.05 mol/L
$K_{d,RNcoll,FeOx}$	Distribution coefficient for reversible sorption of radionuclide element RN onto iron-(hydr)oxide colloids	mL/g colloid	various sources – refer to Section 3.8.2.1	geometric mean values: Pu, Am = 10,000 log-normal distributions; geometric std. dev. = 1 order-of-magnitude
$ST_{coll,FeOx,pHlo}$	Function relating stability of iron-(hydr)oxide colloids to pH and ionic strength at relatively low pH values	mol/L	Figure 3.8-7	$I = -0.02 \times pH + 0.17$
$ST_{coll,FeOx,pHhi}$	Function relating stability of iron-(hydr)oxide colloids to pH and ionic strength at relatively low pH values	mol/L	Figure 3.8-7	$I = +0.02 \times pH - 0.17$
$ST_{coll,FeOx,pHint}$	Function relating stability of iron-(hydr)oxide colloids to pH and ionic strength at intermediate pH values	boolean	Figure 3.8-7	boolean ($pH \geq 8$ AND $pH \leq 9$)
groundwater colloids Figure 3.8-10				
$C_{RNcoll,gw}$	Concentration of reversibly attached radionuclide element RN associated with groundwater colloids	mol/L	d	d
$M_{coll,gw}$	Mass of groundwater colloids per unit volume or mass of water	mg/L	d	d

Table 3.8-1.Parameter Names, Definitions, and Values (Continued)

Parameter Name	Parameter Description	Units[a]	Basis for Value/Source	Value and Distribution
$M_{coll,gw,max}$	Highest observed or expected mass of groundwater colloids per unit volume or mass of water	mg/L	Figure 3.8-9	3×10^{-2} mg/L
$M_{coll,gw,min}$	Lowest observed or expected mass of groundwater colloids per unit volume or mass of water	mg/L	Figure 3.8-9	3×10^{-6} mg/L
$K_{d,RN,coll,gw}$	Distribution coefficient for reversible sorption of radionuclide element RN onto groundwater colloids	mL/g colloid	various sources – refer to Section 3.8.2.1	geometric mean values: Pu = 10,000 Am = 100,000 log-normal distributions; geometric std. dev. = 1 order-of-magnitude
$I_{lo\text{-}thresh,coll,gw}$	Ionic strength below which groundwater colloids are stable	mol/L	Figure 3.8-9	0.01 mol/L
$I_{hi\text{-}thresh,coll,gw}$	Ionic strength above which groundwater colloids are unstable	mol/L	Figure 3.8-9	0.05 mol/L
$ST_{coll,gw}$	Function relating stability of groundwater colloids to ionic strength at intermediate ionic strengths	mg/L	Figure 3.8-9	CRWMS M&O (2000m, p. 53, equations 4 and 5) for equation
colloid source term			Figure 3.8-11	
C_{RNcoll}	Concentration of mobile colloidal radionuclide element RN per unit volume or mass of water	mol/L	d	d
M_{coll}	Total mass of all mobile colloids per unit volume or mass of water	mg/L	d	d
input parameters			Figures 3.8-6, 3.8-8, 3.8-10, and 3.8-11	
I	Ionic strength	mol/L	f	f
pH	pH	pH units	f	f
C_{RNdiss}	Concentration of dissolved radionuclide element RN per unit volume or mass of water	mol/L	f	f

Source: *Waste Form Colloid-Associated Concentrations Limits: Abstraction and Summary* (CRWMS M&O 2000m, Table 1).

NOTES: [a] mol/L is actually moles per liter of dispersion and is not molarity in the strict sense; L may be replaced with kg, if PA prefers molality
[b] uncertainty is linked to uncertainty for ionic strength, which is an input parameter; no additional uncertainty is added
[c] mass is correlated with irreversible radionuclide concentration
[d] model output
[e] (CRWMS M&O 2000m, Figure 10)
[f] input parameters

The processes (referred to as sorption processes) controlling the attachment and detachment of soluble materials to colloidal substrates are complex. They are, however, similar to the processes that control attachment and detachment of dissolved materials to rock surfaces in the subsurface. The approach adopted to describe interactions between colloid substrate materials and dissolved radionuclides is, therefore, similar to that used for contaminant-rock interactions in the subsurface. The most common approach used in assessing these interactions is the linear isotherm, or K_d, approach based on results of batch sorption experiments. The linear isotherm model relationship is defined as follows:

$$S = K_d \cdot C \qquad \text{(Eq. 3.8-1)}$$

where K_d is the distribution coefficient (mass or surface-area based), S is the mass of a solute sorbed on a unit mass (or surface area) of solid, and C is the concentration of the sorbing solute. The basis for the K_d values used is described in CRWMS M&O (2000m, Section 6.1.3, Attachment XVI). Table 3.8-1 summarizes recommended values, which have been extracted from the LANL data. For Pu and Am, values were developed from the colloid experiments. The maximum K_d values from the ranges for devitrified tuff and iron oxide were used as an analog for colloids consisting of smectite and corrosion-product colloids, respectively. Given the non-exact nature of developing these K_d values, a plus-or-minus one-order-of-magnitude uncertainty band was assigned to each value.

As described in CRWMS M&O (2000m, Section 6.1.3 and Attachment XVI) a large amount of data exists in published literature for sorption. The ranges of K_d values developed from the YMP data are generally consistent with those in the literature.

It is recognized that, because of the complexity of the processes involved, the assumptions implicit in the use of the linear isotherm model may not be satisfied for attachment of actinides to colloid substrates. For example, experimental evidence from plutonium sorption experiments (CRWMS M&O 2000m, Section 6.1.3, Attachment XVI) with colloidal hematite and goethite show that the rate of desorption (backward rate) of Pu is significantly slower than the rate of sorption (forward rate). More importantly, over a significant time period (up to 150 days in some experiments), the extents of desorption is somewhat less than the extent of sorption. To accommodate this non-ideal sorption behavior of plutonium, a larger K_d was selected based on the desorption experiments, pending further results from LANL.

The second type of radionuclide attachment to colloidal substrates involves incorporation of the radionuclide into the colloidal material substrate before it becomes suspended as colloids. Experimental results from HLW glass degradation experiments show that plutonium can be incorporated as discrete phases in smectite colloids generated during the experiments (CRWMS M&O 2000x, Section 6.2). Because the plutonium in these colloids appears to be formed by coprecipitation, along with the substrate materials, it can be envisioned to be engulfed in the colloid matrix. The extent to which these engulfed radionuclides are available for interaction with the solution is uncertain. To accommodate this uncertainty, the model and abstraction accounts for smectite colloids with “engulfed” plutonium and other radionuclides by treating them as a separate colloid subtype and by conservatively assuming that the engulfed radionuclides are an intrinsic part of, and irreversibly attached to, the colloid substrate.

3.8.2.2 Waste-Form Colloids

Three broad categories of waste form are to be placed in the repository. The most abundant waste type is CSNF from commercially owned and operated electric power reactors. DSNF is a diverse collection of waste from reactors at DOE nuclear-complex sites. HLW is a borosilicate glass-based waste form. In the following descriptions, CSNF and DSNF are treated together.

3.8.2.2.1 HLW Glass

In long-term tests (CRWMS M&O 2000x, Section 6.2 and Ebert 1995, Section 9.2.3) experimental evidence indicated that colloids were produced through spallation of altered glass at the glass-water interface. The colloids were primarily smectite clay containing discrete radionuclide-bearing phases incorporated (entrained) into the clay (CRWMS M&O 2000x, Sections 6.2, 6.2.1, Figure 1). Some iron silicate colloids were also observed. Several entrained phases were identified including brockite (thorium calcium orthophosphate) and an amorphous thorium-titanium-iron silicate, similar to thorutite. The phases contained actinides and rare-earth elements. Uranium was detected within the clays and iron silicates in some samples.

Because the experimental evidence indicates that the entrained phases are often engulfed in a smectite clay substrate the colloidal properties of the entrained radionuclide-bearing phases are assumed to be governed by the properties of the smectite. The stability (and therefore mobility) of smectite colloids will control the stability of the colloids. Also, because the extent to which the smectite clay substrate isolates the entrained radionuclide phases from interacting with the solution is uncertain, it is conservatively assumed that the radionuclides are irreversibly attached to the smectite colloid substrate.

It was observed in the static tests that colloids developed and increased in concentration with time, up to a point where the colloids concentration reached a maximum and then became unstable. This was attributed to increase in the ionic strength to a threshold above which the colloids precipitated and flocculated (CRWMS M&O 2000x, Section 6.2.1, Figure 8). These data provide the experimental basis for evaluating the ionic strength regimes in which the waste form colloids are stable or unstable.

3.8.2.2.2 Spent Nuclear Fuel

Very little radionuclide-bearing colloidal material was detected in tests on corrosion of the uranium dioxide-based commercial LWR SNF (CRWMS M&O 2000x, Sections 6.3.2 and 6.4.3, Figures 25 and 30). The available leachate analyses showed that the total Pu concentration (dissolved, colloidal, and sorbed onto the test vessel) was in the range 10^{-8} to 10^{-10} mol/L. However, no colloids with embedded radionuclide phases similar to those observed HLW glass tests were observed in the much fewer number of CSNF tests. Although the data show that some of this plutonium was colloidal, the types of colloids involved are not known. Little information is currently available concerning the quantity or types of colloids formed from DSNF, but preliminary tests indicate that colloid formation from metallic uranium-based DSNF may be significant. DSNF colloid formation and characterization testing has been initiated. For modeling purposes, it is assumed that only colloids with reversibly attached radionuclides are formed in the corrosion of commercial and defense spent nuclear fuels. This assumption is

potentially non-conservative, and will be verified as pertinent results of ongoing confirmatory test become available (CRWMS M&O 2000m, Section 6.4.1). Note, however, that corrosion-product colloids, generated from the degradation of the waste packages, are formed in proximity of all waste types. As metal-(hydr)oxides, corrosion-product colloids will behave similarly to metal-(hydr)oxide colloids generated during SNF degradation, in terms of radionuclides sorption and stability characteristics. The mass concentrations of corrosion-product colloids are reasonable, based on the ranges of colloid concentrations observed in natural waters spanning a wide range of hydrogeochemical conditions.

3.8.2.2.3 Waste-Form-Colloid Concentration Model and Abstraction

The waste form concentration model includes contributions from colloids with reversibly and irreversibly-attached radionuclides.

The model for the concentration of irreversibly attached radionuclide is based on the experimental data for HLW glasses. It was observed experimentally that, as the ionic strength increased, colloid concentration generally decreased and ultimately a threshold value was reached above which the colloids were not observed or were observed in very low quantities (Figure 3.8-3) (CRWMS M&O 2000x, Figure 8). As this threshold was approached, it was also observed for one glass that the colloid size increased significantly due to aggregation of the colloids. The threshold at which flocculation occurred was approximately 0.05 mol/kg.

Colloids formed from HLW glasses in the ANL tests consist predominantly of a smectite clay substrate often containing discrete radionuclide-bearing phases entrained within the clay. For modeling purposes it is assumed that colloids derived from HLW are all radionuclide-bearing smectite. The radionuclides are conservatively assumed to be irreversibly attached to the smectite colloid substrate, pending forthcoming data from LANL on radionuclide attachment and detachment to/from waste form colloids. Figure 3.8-4 represents the abstraction of the ANL experimental results. Based on ionic strength conditions near the waste form, the concentration of irreversibly attached colloidal plutonium is calculated. A flow chart illustrating how these calculations are performed is shown in Figure 3.8-6a. At ionic strengths less than 0.01 mol/kg, colloidal Pu concentration is set to equal the maximum value observed in the experiments (6×10^{-8} mol/L). At ionic strengths greater than or equal to 0.05 mol/kg, colloidal Pu concentration is defined as a minimum observed value (1×10^{-11} mol/L). At intermediate ionic strengths, colloidal Pu concentration is an intermediate value calculated as illustrated in Figure 3.8-6b. The details of these calculations are described more fully in CRWMS M&O (2000m, Section 6.2.2 and Figure 13). Because of changes in methods of analysis, slightly larger Pu concentrations are given in CRWMS M&O (2000x, Figure 8). However, the differences are small enough that they can have, at the most, a minor effect on total system performance.

The pH_{ZPC} of smectite occurs at a pH value of approximately 2. Since a pH this low is not anticipated in the repository, it is a reasonable assumption that smectite colloids will remain in a stable pH range much of the time under anticipated repository conditions (unless ionic strength exceeds a certain threshold). Tombacz et al. (1990) investigated the stability of montmorillonite suspensions as a function of pH and ionic strength in NaCl solutions. They found that suspensions became unstable and flocculated at pH 2, 4, and 8 in 0.01 mol/L, 0.025 mol/L, and 0.04 mol/L NaCl solutions, respectively (Figure 3.8-5a). This relationship suggests that the

stability of smectite colloids becomes increasingly sensitive to ionic strength as pH drops below about 8. This concept is represented by the abstraction shown in Figure 3.8-5b. At combinations of ionic strength and pH falling below the sloped line, waste-form colloids are stable, and the maximum concentration defined in the previous paragraph (i.e., 6×10^{-8} mol/L) is maintained. At combinations of pH and ionic strength falling above the sloped line, waste-form colloids are unstable, and the concentration is adjusted to the minimum value (1×10^{-11} mol/L). This approach assumes that the concentration of irreversibly attached colloidal Pu is directly proportional to the mass of colloids. That assumption is valid based on observations from ANL experiments and descriptions in the AMR (CRWMS M&O 2000x, Figure 24, Section 6.3). The abstraction of the effect of ionic strength on waste-form colloid concentration (Figure 3.8-4) and the effect of the combination of ionic strength and pH (Figure 3.8-5) on colloid stability is illustrated as a flow chart in Figure 3.8-6a and 6b, respectively.

To determine the amount of a radionuclide that can reversibly sorb onto the surface of waste-form colloids, a mobile mass is required. Since the concentration of irreversibly attached colloidal Pu is directly proportional to the mass of colloids, the mobile mass is determined by scaling on the basis of predicted plutonium concentration (determined from Figures 3.8-4 and 3.8-5b). The scaling is based on the ratio of the maximum plutonium concentration (6×10^{-8} mol/L) and the maximum colloid mass concentration (5 mg/L). This approach is illustrated in flow chart format in Figure 3.8-6c. Based on a comparison of colloid mass and plutonium associated with colloids, it was found that a concentration of approximately 1 mg colloids correlated to about 1.2×10^{-8} moles of colloidal plutonium (CRWMS M&O 2000x, Figure 24; CRWMS M&O 2000m, Figure 10). The maximum stable colloid mass concentration was about 5 mg/L (corresponding to 6×10^{-8} mol/L colloidal Pu). These observations are specific to the ANL experiments, but they provide a means for scaling irreversible colloid-associated Pu concentration to ionic strength.

The concentration of reversibly sorbed radionuclide is determined using appropriate K_d values and Equation 3.8-1 (Figure 3.8-6d).

3.8.2.3 Corrosion Product Colloids

Corrosion product colloids are assumed to be generated as a result of the corrosion of iron-bearing components of the WP. In this section, radionuclide uptake of mobile corrosion product colloids, assumed to be iron-(hydr)oxides such as hematite and goethite, is described.

3.8.2.3.1 Corrosion-Product-Colloid Concentration Model and Abstraction

Data are not available on the concentration of corrosion product colloids that may be generated as a result of corrosion of the WP materials. Limited data do exist, however, for concentrations of iron-colloids in natural systems. At the Morro de Ferro natural analog site (Poços de Caldas, Brazil) concentrations of iron-hydroxide [$Fe(OH)_3$] colloids were measured in deep groundwaters adjacent to an iron-rich rock body. The measured concentration, 0.25 mg/L, is similar to concentrations of other inorganic colloids found in natural waters at other deep sites (McCarthy and Degueldre, 1993, Table 6). Consequently, it is conservatively assumed that the concentration of these colloids is comparable to that of the groundwater colloids (all mineralogies) that are observed in natural groundwaters (CRWMS M&O 2000m,

Section 6.1.1.3). Colloid concentrations observed in natural waters represent concentrations developed over a wide range of hydrogeochemical conditions, which provides some assurance that the approach used is reasonable. A maximum value of 1 mg/L (Table 3.8-1) was used in the TSPA calculations, which imparts a four-fold conservatism relative to concentrations observed at the analog site.

The stability behavior of iron-(hydr)oxide colloids with respect to ionic strength and pH is predictable. As shown in Figure 3.8-7a, iron-(hydr)oxide has a pH_{ZPC} of about 8.5. At much lower and higher pH values, iron-(hydr)oxide colloids are affected by ionic strength much like other minerals, in that ionic strengths greater than about 0.05 mol/kg result in destabilization (Figure 3.8-7a). These concepts are represented in the abstraction shown in Figure 3.8-7b. At pH values close to the pH_{ZPC}, the dispersion is determined to be unstable regardless of ionic strength, and a minimum mass concentration is established. The same situation applies if an input pH value is several pH units away from the pH_{ZPC} and ionic strengths are greater than 0.05 mol/kg. At intermediate pH values the combined effect of ionic strength and pH are described by two lines; above the lines, the colloidal dispersion is unstable, and a minimum mass concentration is established. These concepts are illustrated in a flow-chart format in Figure 3.8-8a in which the approach for calculating the mobile mass of iron-(hydr)oxide colloids is defined.

Given the mass concentration of the corrosion product colloids, the concentration of reversibly sorbed radionuclide is determined using appropriate K_d values and Equation 3.8-1 (see Figure 3.8-8).

3.8.2.4 Groundwater Colloids

As described in Section 3.8.1.3 smectite colloids are expected to be in the groundwater that enters the degraded WPs. The basis for modeling the mass concentration and the radioisotope concentration associated with these colloids as they exit the WP is described below.

3.8.2.4.1 Groundwater-Colloid Model and Abstraction

Several workers (for example, McCarthy and Zachara 1989; McCarthy and Degueldre 1993) have studied and compiled the characteristics of colloids in groundwaters from crystalline and sedimentary rocks in saturated and unsaturated hydrologic regimes throughout the world. Data on colloid concentration and ionic strength for these groundwaters (Figure 3.8-9a) show a general inverse correlation above an ionic strength of about 0.01 mol/kg and provide a predictive tool for estimating groundwater colloid mass concentration. This relationship is not precise because there are other factors, such as pH, that can significantly affect concentration.

To be useful for TSPA-SR analysis calculations, these data were converted to mass or surface area per unit volume. For naturally occurring colloids, it has been observed that the number population and colloid size correlate using a power function (CRWMS M&O 2000m, Section 6.2.3). In general, the number of colloids increases by a factor of 1000 as colloid diameter decreases by a factor of 10 (assuming spherical colloids). Expressed mathematically, the colloid population is proportional to the inverse of the colloid diameter cubed. By taking the first derivative of the cumulative distribution, combining with masses of individual colloids

assuming spherical geometry and then integrating the function between a lower and upper size limit, the total colloidal mass is obtained. Colloid masses were calculated using the number populations in Figure 3.8-9a and extrapolated to include colloid sizes between 10 nm and 1 micrometer. A colloid mineral density of 2.5 g/cm^3 was assumed. Details of this abstraction are described in CRWMS M&O (2000m, Section 6.2.3).

Figure 3.8-9 illustrates the abstraction. The upper and lower bounding lines capture the highest and lowest concentrations of colloids shown in Figure 3.8-9a. Between ionic strengths of 0.01 and 0.05 mol/kg, the number and mass concentrations decrease as groundwater colloids are destabilized. The decrease is assumed to be linear in log-log space. The flow chart in Figure 3.8-10 illustrates how the groundwater colloid mass concentration is calculated as a function of ionic strength.

Given the assumption that Yucca Mountain groundwater colloids are clays, the relationship developed above for the effect of ionic strength and pH on montmorillonite can also be used here. The approach for calculating the effect of pH on the groundwater colloid mass concentration is illustrated in the flow chart in Figure 3.8-10b. It is similar to the approach illustrated in Figure 3.8-5a and 5b.

It is assumed that sorption for groundwater colloids is similar to clay minerals (CRWMS M&O 2000m, Section 6.2.3), and the same K_d values used for waste-form colloids are used for groundwater colloids. The concentration of a reversibly-sorbed radionuclide is determined using appropriate K_d values (see Figure 3.8-10c).

3.8.2.5 Mobile Colloidal Radionuclide Source Term

Also illustrated in Figure 3.8-11, the mobile colloidal radionuclide source term is the sum of the radionuclide contribution from waste-form colloids (reversibly and irreversibly attached), corrosion-product colloids, and groundwater colloids. The total colloid mass concentration is also provided for recalculating reversibly attached radionuclide concentrations as dissolved concentrations change during transport.

3.8.3 Confidence/Limitations/Validation

The abstractions are based on laboratory results from waste form corrosion testing and testing of adsorption and desorption properties of Pu and Am on clay and iron-(hydr)oxide colloids. To the extent that the laboratory tests and test conditions represent anticipated repository conditions, the abstraction is valid for calculating the colloid-associated radionuclide concentrations and colloid mass concentrations. The approach for the colloidal-radionuclide source term is based on YM-specific field and laboratory studies and results from YM-relevant studies. The development of the conceptual model and implementation requires consideration of colloid generation, colloid-radionuclide interaction, colloid stability behavior, and to some extent, colloid transport/ retardation behavior. Information used for groundwater colloids, waste-form colloids, and corrosion-product colloids was obtained primarily from YM-specific studies. Consequently, the colloid-concentration model is expected to be representative of YM behavior.

Field evidence of small concentrations of radionuclides associated with colloids migrating considerable distances provides insight into possible colloid behavior at YM. At the Benham

nuclear test site at the Nevada Test Site, rapid transport of colloid-associated Pu appears to have occurred (Kersting et al. 1999). At a point approximately 1.3 km from the blast site, 1×10^{-14} mol/L colloid-associated plutonium was detected 30 years after the detonation. It is difficult to determine what fraction of the transport was due to transport on colloids, injection through fractures by the blast, or transport as dissolved Pu. However, in the cases of other detonations, fracture injections have not been observed to extend beyond a few hundred meters, and dissolved Pu would be expected to sorb strongly to the fracture surfaces. It is plausible that the Pu was transported as Pu irreversibly attached to colloids. This example underscores the potential significance of the irreversibility of radionuclide attachment to smectite colloids observed in the ANL waste form corrosion experiments. Irreversibly attached colloids are included in the abstracted model as a contribution to the colloid-associated radionuclide concentration. As pointed out in 3.8.2.2 the colloids formed from CSNF and DSNF are assumed to have reversibly attached radionuclides; this assumption is potentially non-conservative and remains to be verified when results from ongoing tests become available.

Based on the considerations discussed above, this model is appropriate for its intended use.

3.8.4 Alternative Approaches and Other Views

The approach described herein for the colloidal radionuclide source term is consistent with what was done in the TSPA-VA (DOE 1998b) but is more sophisticated in several ways. The treatment for TSPA-SR analysis includes direct consideration of groundwater chemical conditions (pH and ionic strength) on stability of waste-form and corrosion-product colloids. The effect of ionic strength was considered in the TSPA-VA (DOE 1998b) for groundwater colloids, as it is in the TSPA-SR analysis. In TSPA-VA (DOE 1998b), the concentration of colloids (C_c) was evaluated from a linear empirical relation with ionic strength of the liquid solution. Here, the mass concentration of colloids is not assumed to be constant but varies according to groundwater chemical conditions. This approach provides more realism in the TSPA-SR analysis calculations.

In terms of radionuclide uptake, the treatment herein utilizes laboratory results that describe the irreversible uptake, or engulfing, of plutonium phases in smectitic waste-form colloids created during glass corrosion. In contrast, in the TSPA-VA, the parameter distribution for the fraction of radionuclides irreversibly sorbed on colloids was estimated using preliminary information from the Benham nuclear test area at the Nevada Test Site. The Benham data showed rapid transport of colloid associated plutonium occurred (CRWMS M&O 1998b, p. 6-103). For reversible uptake, a similar approach to what was used in the TSPA-VA (DOE 1998b) is used; experimental results for Pu and Am sorption by colloidal iron-(hydr)oxides, montmorillonite, and silica were used to develop K_d values (CRWMS M&O 1998b, p. 6-101; Lu et al. 1998).

3.8.4.1 Alternative Models

An alternative model for waste-form colloid generation was proposed in CRWMS M&O (2000x) in which the rate of colloid formation was based on the amount of altered waste form as indicated by release of boron for glass and technetium for spent fuel. This model was not used because it is based on limited laboratory data. It may, however, be useful in the future.

To a large extent, the effectiveness of colloids in facilitating contaminant transport is due to their very large surface area available for sorption. Depending on the size distribution of colloids in the groundwater, the impact of choosing a mass-based K_d, or a surface-area-based K_a, may be significant. The greatest variability exists in situations in which an inordinately large number of very small colloids exist, which have a high surface-area-to-mass ratio. Based on experimental measurements and observations of colloid characteristics in Yucca Mountain groundwater, this situation does not exist at Yucca Mountain, and the use of a mass-based K_d is appropriate (CRWMS M&O 2000m, Attachment XVI, Section 1.1).

In the TSPA-VA, a slightly different approach was taken by assuming a constant steady-state mass concentration of colloids in groundwater. The steady-state mass concentration was embedded in a sorption term referred to as K_c. The approach described herein is more comprehensive in that the effect of ionic strength and pH on mass concentration are included. In the waste-form area, the K_d approach adopted here will provide more realism by accounting for the destabilizing effect of high ionic strength conditions and some pH conditions. The K_c approach, however, is well suited for far-field transport, where transients in aqueous chemical conditions are not expected.

3.8.4.2 Issues

The VA (DOE 1998b, Section 6.5.3.13) acknowledged the need to better define the mechanistic basis for plutonium sorption onto colloids, as well as other characteristics associated with these colloids. As described in Section 3.8.2.1, an improved mechanistic understanding has been accomplished, and has been utilized herein. In particular, research was conducted to evaluate the reversibility of plutonium sorption on hematite, goethite, silica, and montmorillonite colloids, rates of sorption/desorption, and applicability of the linear isotherm model. The approach taken for plutonium sorption was to utilize the relatively large and, therefore, conservative desorption K_d in the TSPA model.

The TSPA peer review panel noted that one of the aspects of repository performance with large uncertainties includes actinide transport by colloids and the panel stated that there is no convincing way to estimate the type, amounts, or stability of colloids (Budnitz et al. 1999, Section II.A and II.G). The approach used is a systematic treatment of the colloid source term based on consideration of: (1) the possible types of colloids that could be present or generated, (2) modes of radionuclide uptake, and (3) the stability of those colloid types. Screening considerations were also made based on near-field colloid transport/retardation behavior, which is partly controlled by colloid size, engineered barrier or rock characteristics, colloid surface characteristics (including hydrophobicity).

Because some results from experiments conducted since TSPA-VA (DOE 1998b) on colloidal generation on HLW glass were available, a more defensible component of colloidal generation and transport has been constructed for TSPA-SR analysis (Section 3.8). A mass-balance model for the waste-form colloid component, is used indirectly. Because data for waste-form colloid generation rates are based on a few waste types for a relatively short time period (several years), it is difficult to develop rate data that are applicable to many thousands of years. However, the approach described herein allows tracking of waste-form colloid generation rates, and comparison to laboratory observations outside of the TSPA calculation.

3.9 ISSUES RELATED TO WASTE FORM DEGRADATION

Technical issues have been raised concerning degradation of the waste form and related topics in a number of forums by groups that have project oversight, regulatory oversight, or stakeholder interfaces with the YMP. These groups include the NRC, the ACNW, the NWTRB, and peer review panels. Raising these issues by the review bodies has resulted in further consideration of the issues and, frequently, modifications to project testing and modeling. This section identifies issues related to waste form degradation raised by these groups and presents the PMR's approach to the issues (Table 3.9-1).

Table 3.9-1. Key External Issues for the Waste Form Degradation Process Model Report

Issue	Source	PMR Approach
Cladding Credit. Practicality of taking performance credit for cladding. Comments include: (i) excessive optimism in the credit taken in the VA, (ii) substantial related uncertainties, (iii) quality of data on cladding initial state, (iv) lack of industry studies to support cladding credit, (v) ability of the cladding to remain intact for long periods of time under conditions that it was not designed for, and (vi) ability of DOE to gather data to support related assumptions.	ACNW[a], NWTRB[b], PARP[c]	Cladding models have been heavily revised since VA, so criticisms of the VA models do not apply to SR models. SR models are based on information from various sources, including peer-reviewed technical literature, and the models account for uncertainty in cladding condition. Although the nuclear power industry has not studied cladding credit for disposal, that fact is irrelevant because the nuclear power industry has no responsibility for disposal. Zircaloy has exceptional resistance to a wide variety of corrosive environments, so a long lifetime is to be expected in the relatively benign environment of a repository. DOE has collected a large amount of information on as-irradiated cladding and cladding degradation mechanisms. See Section 3.4.4 for further discussion of this issue.
Performance of Cladding. Ability to sufficiently establish the performance of cladding. Questions raised relate to: (i) the initial damage and condition of cladding, (ii) potentially optimistic or undemonstrated assumptions of the behavior or performance of cladding, (iii) understatement of related uncertainties, (iv) insufficient analysis related to hydrogen embrittlement of Zirconium cladding, and (v) need for investigations to identify additional potential failure mechanisms.	NRC[d], ACNW[a], PARP[c]	The first three items are treated in the discussion of cladding credit above. Hydrogen effects have been treated in detail (CRWMS M&O 2000r). Literature from the nuclear power and chemical industries has been heavily reviewed to assure that all potential mechanisms have been considered. See Section 3.4.4 for further discussion of this issue.
In-Package Chemistry. The level of understanding of and the uncertainty related to the chemistry inside of the WP. Chemistry affects the degradation of the waste form and the ability to support cladding credit.	NWTRB[b], PARP[c]	For TSPA-SR analysis, the Waste Form Degradation Model now explicitly contains an In-Package Chemistry Component (Section 3.2) to better characterize the uncertainty in the degradation of the cladding and waste form. See Section 3.2.3 for further discussion of this issue.

Table 3.9-1. Key External Issues for the Waste Form Degradation Process Model Report (Continued)

Issue	Source	PMR Approach
Waste Form Corrosion. Weaknesses were noted in the VA spent fuel corrosion model related to the adequacy of existing data and to the depth of addressing the processes that ensue among the WP internals following penetration.	PARP[c]	These VA model and documentation weaknesses were addressed in the development of the SR model and documentation as discussed in Section 3.3. In particular, the mechanistic basis was discussed, a simplified model was developed, and model validation was performed. The degradation of the cladding and waste matrix is now coupled as explained in Section 3.4 and the degradation of the waste matrix was also coupled to the in-package chemistry to better characterize uncertainty as explained in Sections 3.2 and 3.3.
Formation of Colloids. The need to better define the mechanistic basis for plutonium sorption onto colloids, as well as other characteristics associated with these colloids, has been identified. Large uncertainties were identified associated with actinide transport by colloids.	NRC,[d] PARP[c]	Results from experiments have resulted in an improved component of colloidal generation and transport. See Section 3.8.4.2 for additional information on this issue.
Radionuclide Solubility. The need for data and additional studies related to solubility limit distributions for several key radionuclides, including neptunium and technetium were identified.	NRC,[d] PARP[c]	Several substantial changes were made in the Dissolved Radioisotope Concentration Component to address these concerns. See Section 3.7.3 for additional information related to this issue.
Screening of Radionuclides. Thorough documentation of the approach used to eliminate unimportant radionuclides from consideration and to demonstrate the potential impacts of not considering the full suite of possible radionuclides present.	NRC[d]	For TSPA-SR analysis a reevaluation of radioisotope screening was conducted. See Section 3.1.3 for further discussion of this issue.
Radionuclide Retardation. Failure to take credit for radionuclide retardation through coprecipitation or sorption by secondary alteration phases.	PARP[c]	TSPA-SR analysis conservatively neglects radioisotope retardation through coprecipitation or sorption inside the WP. This topic is a FEP and mentioned in Table 2.3-8, FEPs Related to the Colloidal Radionuclides Concentration Component.
Natural Analog Studies of High-Level Waste Glasses. Proper consideration of data on naturally occurring glasses, combined with experimental data and models on dissolution of high-level waste glasses, to demonstrate that long-term dissolution behavior under repository conditions can be represented by extrapolation of results from short-term laboratory tests.	NRC[d]	As stated more fully in discussion of related criteria in Chapter 4, the bounding formulation of the HLW Degradation Component, discussed in Section 3.6, included the consideration of experimental and other relevant data, including information from natural analogs. Natural analog information is used for corroboration (see Section 3.6.2 and Cunnane et al. 1994).
Consistency of Modeling Assumptions. Modeling assumptions should be consistent across different process models, unless there is a defensible technical rationale.	NRC[d]	The AMRs that document assumptions have been subjected to thorough interdisciplinary reviews to help ensure consistency among assumptions made in more than one document about a given parameter. In addition, the PMRs that summarize and integrate the results of the AMRs have been subjected to a review by a single review team, one of whose main objectives is to identify inconsistencies among the PMRs.

Table 3.9-1. Key External Issues for the Waste Form Degradation Process Model Report (Continued)

Issue	Source	PMR Approach
Importance of Detailed Documentation of Assumptions. The need for detailed documentation of assumptions used in performance assessments to provide transparency and credibility of TSPAs.	NRC[d]	Procedures have been developed that control development and documentation of analyses and models that are subject to the requirements of the quality assurance program (DOE 2000). The QA program requires identification and detailed documentation of assumptions for both analyses and models (DOE 2000, Section 3.2.2, Section 3.2.3, and Supplement III.2.6). Implementation of the governing procedures fundamentally ensures that analyses and models used for performance assessments are transparent and credible.
Excessive Use of Expert Elicitations. Concern that a substantial part of the knowledge base rests in expert elicitations, e.g., for data on waste form degradation.	PARP[c]	Additional experimental programs in waste form degradation have been conducted. The concern has been alleviated by additional work in the area and by discontinuing the direct use of the expert elicitations.

NOTES: [a] ACNW: Advisory Committee on Nuclear Waste
[b] NWTRB: Nuclear Waste Technical Review Board
[c] PARP: Total System Performance Assessment Peer Review Panel
[d] NRC: Nuclear Regulatory Commission

4. RELATIONSHIP TO NUCLEAR REGULATORY COMMISSION ISSUE RESOLUTION STATUS REPORTS

4.1 SUMMARY OF THE KEY TECHNICAL ISSUES

As part of the review of site characterization activities, the Nuclear Regulatory Commission (NRC) has undertaken an ongoing review of information on Yucca Mountain Site characterization activities to allow early identification and resolution of potential licensing issues. The principal means of achieving this goal is through pre-licensing consultation with DOE. This approach attempts to reduce the number of, and to better define, issues that may be in dispute during the NRC licensing review, by obtaining input and striving for consensus from the technical community, interested parties, and other groups on such issues.

The NRC has focused pre-licensing issue resolution on those topics most critical to the post-closure performance of the potential geologic repository. These topics are called Key Technical Issues (KTIs). Each KTI is subdivided into a number of subissues. The KTIs are:

- Activities Related to Development of the EPA Standard
- Container Lifetime and Source Term
- Evolution of the Near-Field Environment (ENFE)
- Igneous Activity
- Radionuclide Transport
- Repository Design and Thermal-Mechanical Effects
- Structural Deformation and Seismicity
- Thermal Effects on Flow
- Total System Performance Assessment (TSPA) and Integration
- Unsaturated Zone and Saturated Zone Flow Under Isothermal Conditions.

Identifying KTIs, integrating their activities into a risk-informed approach, and evaluating their significance for post-closure performance helps ensure that NRC's attention is focused on technical uncertainties that will have the greatest affect on the assessment of repository safety.

Early feedback among all parties is essential to define what is known, what is not known, and where additional information is likely to make a significant difference in the understanding of future repository safety. The Issue Resolution Status Reports (IRSRs) are the primary mechanism that the NRC staff uses to provide feedback to the DOE on the status of the KTI subissues. IRSRs focus on NRC acceptance criteria for issue resolution and the status of issue resolution, including areas of agreement or when the staff has comment or questions. Open meetings and technical exchanges between NRC and DOE provide additional opportunities to discuss issue resolution, identify areas of agreement and disagreement and plans to resolve any disagreements.

Each KTI is subdivided into a number of subissues. For most subissues, the NRC staff has identified technical acceptance criteria that the NRC may use to evaluate the adequacy of information related to the KTIs. The NRC has also identified two cross-cutting programmatic criteria that apply to all IRSRs related to the implementation of the Quality Assurance Program and the use of expert elicitation.

Chapter 4 documents DOE's approach to addressing the acceptance criteria and work performed that is related to the criteria. The following sections provide a summary level discussion of the related KTIs by subissues and specific NRC acceptance criteria. Table 4.1-1 identifies the IRSRs/KTIs related to the WF PMR and tabulates the subissues identified in those IRSRs that are addressed in this chapter.

Table 4.1-1. Issue Resolution Status Report/Key Technical Issues Related to the Waste Form Degradation Process Model Report

NRC KTIs/ IRSRs	Subissues
Container Life and Source Term	*General Acceptance Criteria* *SUBISSUE 3 – The Rate at Which Radionuclides in Spent Nuclear Fuel Are Released from the Engineered Barrier Subsystem through the Oxidation and Dissolution of Spent Fuel* *SUBISSUE 4 – The Rate at Which Radionuclides in High-Level Waste Glass Are Released from the Engineered Barrier Subsystem* *SUBISSUE 5 – The Effects of In-Package Criticality on Waste Package and Engineered Barrier Subsystem Performance*
Evolution of the Near-Field Environment	*SUBISSUE 3 – Effects of Coupled Thermal-Hydrologic-Chemical Processes on the Chemical Environment for Radionuclide Release* *Data and Model Justification* *Data Uncertainty and Verification* *Model Uncertainty* *Model Verification* *Integration* *Programmatic Criteria*
Total System Performance Assessment and Integration	*Programmatic Acceptance Criteria* *SUBISSUE 1 – System Description and Demonstration of Multiple Barriers* *Transparency and Traceability of the Analysis* *Total System Performance Assessment Documentation Styllow* *SUBISSUE 2 – Scenario Analysis* *Identification of an Initial Set of Processes and Events* *Classification of Processes and Events* *Screening of Processes and Events* *Formation of Scenarios* *Screening of Scenario Classes* *SUBISSUE 3 – Model Abstraction* *Quantity and Chemistry of Water Contacting Waste Packages and Waste Forms* *Radionuclide Release Rates and Solubility Limits*

4.2 RELATION OF THE WASTE FORM DEGRADATION PROCESS MODEL REPORT TO THE KEY TECHNICAL ISSUES

The PMRs address issues and processes that frequently correspond to or overlap those identified in the KTIs and IRSRs. Because of their importance, this chapter identifies the KTIs and the associated IRSRs and subissues that are related to this PMR and its associated AMRs.

Table 4.2-1 identifies the specific acceptance criteria that relate to this PMR, describes the approach to addressing the criteria, and cross-references sections of the PMR or AMRs for additional detail, as appropriate.

4.2.1 Key Technical Issue: Container Life and Source Term

The primary issue of the KTI on Container Life and Source Term (CLST) is adequacy of the EBS design to provide reasonable assurance that containers will be adequately long-lived and that radionuclide releases from the EBS will be sufficiently controlled such that container design and packaging of SNF and HLW glass will make a significant contribution to the overall repository performance. The related IRSR is focused on the containers and waste forms as the primary engineered barriers (NRC 1999a, p. 3).

The IRSR on CLST (NRC 1999a, p. 4) identifies six subissues important to resolution of the KTI. This PMR addresses applicable acceptance criteria of Subissues 3 and 4 as well as the general acceptance criteria that the NRC considers applicable to all of the subissues of the IRSR.

4.2.1.1 General Acceptance Criteria

The IRSR that addresses CLST identifies nine general acceptance criteria that are applicable to all of the CLST subissues (NRC 1999a, pp. 19 to 20) (Table 4.2-1). These criteria deal with aspects of data collection, qualification, and verification; documentation of uncertainties; and limitations in both data and process models. The Waste Form Degradation PMR and supporting AMRs were developed in accordance with project procedures for documenting data, analyses, models, and/or computer codes and preparing and reviewing technical reports. The general acceptance criteria were implemented as part of the overall quality assurance program and implementing procedures and are addressed in Table 4.2-1.

4.2.1.2 Acceptance Criteria for Subissue 3

Subissue 3 of the IRSR on CLST deals with the rate at which radionuclides in SNF are released from the EBS. Each of the nine acceptance criteria identified in the IRSR on CLST for Subissue 3 is related to this PMR (NRC 1999a, pp. 24 to 25). This PMR deals with identification of the radionuclides of concern to performance of the repository and identification of the source term for release from SNF waste packages. These issues are covered in Sections 3.1 through 3.5, 3.7, 3.8 and the AMRs that support them. Acceptance criteria related to this subissue and the PMR approach that addresses the acceptance criteria are provided in Table 4.2-1.

4.2.1.3 Acceptance Criteria for Subissue 4

Subissue 4 of the IRSR on CLST deals with the rate at which radionuclides in HLW glass are released from the EBS. Each of the nine acceptance criteria identified in the IRSR on CLST for Subissue 4 is related to this PMR (NRC 1999a, pp. 26 to 27). This PMR deals with identification of the radionuclides of concern to performance of the repository and identification of the source term for release from HLW glass waste packages and co-disposal waste packages. These issues are covered in Sections 3.1, 3.2, 3.6 through 3.8, and the AMRs that support them. Acceptance criteria related to this subissue and the PMR approach that addresses the acceptance criteria are provided in Table 4.2-1.

4.2.1.4 Acceptance Criteria for Subissue 5

Subissue 5 of the IRSR on CLST addresses the effects of in-package criticality on waste package and engineered barrier subsystem performance. In-package criticality is not addressed in this PMR. However, the related acceptance criteria are addressed in DOE's *Disposal Criticality Analysis Methodology Topical Report* (YMP 1998) and its supporting references. Criticality is also mentioned in the *Disruptive Events Process Model Report* (CRWMS M&O 2000an, p. v).

4.2.2 Key Technical Issue: Evolution of the Near-Field Environment

The primary objective of the KTI on the evolution of the ENFE is to assess all aspects of the evolution of the near-field geochemical environment that have the potential to affect the performance of the potential repository.

The IRSR on ENFE (NRC 1999b, Section 2.0) identifies five subissues important to resolution of the KTI. This PMR addresses applicable acceptance criteria of Subissue 3.

Acceptance Criteria for Subissue 3–Subissue 3 of the IRSR on ENFE deals with the effect of coupled thermal-hydrologic-chemical (THC) processes on the chemical environment for radionuclide release. The WP corrosion, the quantity and chemistry of fluids contacting waste forms, and radionuclide release rates and solubility limits will be affected as a result of the evolution of the near-field geochemical environment (NRC 1999b, Section 3.2.3). Subissue 3 of the IRSR on ENFE further divides its acceptance criteria into subsections (NRC 1999a, Section 4.3.1) as shown in Table 4.2-1. The Waste Form Degradation PMR addresses in-package chemistry, solubility limits, formation of colloids, and the various forms of degradation of the waste as a result of infiltration of fluids after failure of the waste package. These issues are mainly discussed in Section 3.2 and supporting AMRs, but also have less direct effect on the other components of the WF degradation model. Acceptance criteria related to this subissue and the PMR approach that addresses the acceptance criteria are provided in Table 4.2-1.

4.2.3 Key Technical Issue: Total System Performance Assessment and Integration

The primary objective of the KTI on Total System Performance Assessment and Integration (TSPAI) is to describe an acceptable methodology for conducting assessments of repository performance and for using these assessments to demonstrate compliance with the overall performance objective and requirements for multiple barriers. The associated IRSR identifies

four subissues important to resolution of the KTI (NRC 2000a, p. 4). This PMR addresses selected acceptance criteria related to Subissues 2 and 3, as well as the programmatic acceptance criteria identified as relevant to all subissues.

4.2.3.1 Programmatic Acceptance Criteria

The IRSR identifies two programmatic acceptance criteria as directly applicable to Subissues 2 and 3 (NRC 2000a, p. 8). As addressed in the table below, the programmatic criteria are integral requirements of the quality assurance program and are applied to all quality-affecting activities, including development of the PMRs, AMRs, and supporting documents. The approaches to these criteria are addressed in Table 4.2-1.

4.2.3.2 Acceptance Criteria for Subissue 1

Subissue 1, System Description and Demonstration of Multiple Barriers, addresses the demonstration and efficacy of multiple barriers. This includes the identification of natural and engineered barriers important to isolation, the capability of these barriers to isolate waste, and the identification of processes that may degrade the engineered barriers and thus adversely affect performance of the natural barriers (NRC 2000a, p. 4). DOE's TSPA, including scenario analysis (Section 4.2.3.3), model abstractions (Section 4.2.3.4) and transparency and traceability (this section) will support the demonstration of multiple barriers (NRC 2000a, p. 17). Specific technical acceptance criteria have not yet been developed for the demonstration of multiple barriers.

This subissue also addresses the NRC's staff expectation on the contents of DOE's TSPA and supporting documents (NRC 2000a, p. 4). It provides criteria to evaluate the transparency and traceability of the DOE's TSPA, of the identification and classification of features, events, and processes (FEPs), and of the abstractions for TSPA. Finally, this subissue focuses on the transparency, traceability and validity of data used to support TSPA and of code design and the flow of data in the TSPA (NRC 2000a, pp. 10-17).

With respect to transparency and traceability, the YMP is committed to improving the documentation of the technical analyses that support the TSPA. The development of this PMR, and the associated AMRs, focused on improving the transparency and traceability from initial data collection through the analysis and incorporation of data and analyses into the process model for Waste Form Degradation, and to the abstraction of the process models for use in the TSPA. Section 1.3 summarizes the quality assurance status of data and software used in the development of this PMR.

4.2.3.3 Acceptance Criteria for Subissue 2

Subissue 2, Scenario Analysis, involves the identification of processes and events that could affect repository performance and then binning these events and processes into categories. A probability of occurrence is assigned to each of these categories of processes and events that could result in some processes and events being excluded from the performance assessment. Subissue 2 divides its acceptance criteria into five subsections (NRC 2000a, Section 4.2). This

PMR addresses the acceptance criteria in each of those subsections (NRC 2000a, Sections 4.2.1 through 4.2.5) as shown in Table 4.2-1. A systematic method was applied to identify and screen FEPs for waste form degradation. Specific information on FEP screening as related to the 87 primary FEPs related to the Waste Form Degradation PMR is addressed in three AMRs: 57 in *Miscellaneous Waste-Form FEPs* (CRWMS M&O 2000n), 15 in *Waste Form Colloid-Associated Concentrations Limits: Abstraction and Summary* (CRWMS M&O 2000m), and 16 in *Clad Degradation-FEPs Screening Arguments* (CRWMS M&O 2000s). One FEP (Mutation) appears in two AMRs (CRWMS M&O 2000n, CRWMS M&O 2000m). Acceptance criteria related to this subissue and the PMR approach that addresses the acceptance criteria are provided in Table 4.2-1.

Regarding the grouping that addresses formation of scenarios, FEPs related to waste form degradation have not been grouped into scenario classes for further screening. Rather, all included FEPs have been incorporated in the Waste Form Degradation Model, which is used in the nominal scenario that is simulated in the TSPA-SR analysis.

4.2.3.4 Acceptance Criteria for Subissue 3

Subissue 3, Model Abstraction, focuses on the information and technical needs related to the development of abstracted models for TSPA. This PMR addresses the acceptance criteria of this subissue that deal with chemistry of water contacting waste forms (NRC 2000a, Section 4.3.1.1.3) and radionuclide release rates and solubility limits (NRC 2000a, Section 4.3.1.1.4). It includes documentation of data used to develop conceptual or process models for model abstractions, verification of the consistency of the abstractions, and explanation of their integration (such as coupling and dependencies) into the TSPA. These issues are addressed in the EBS PMR (CRWMS M&O 2000y), Sections 2 and 3 of this report, and the AMRs that support them. Acceptance criteria related to this subissue and the PMR approach that addresses the acceptance criteria are provided in Table 4.2-1.

Table 4.2-1. Issue Resolution Status Reports, Subissues, Acceptance Criteria, and Process Model Report Approach (NRC 1999a, 1999b, 2000a)

NRC Technical Acceptance Criteria	PMR Approach and Section Reference
IRSR: CONTAINER LIFE AND SOURCE TERM	
GENERAL ACCEPTANCE CRITERIA	
1. The collection and documentation of data, as well as development and documentation of analyses, methods, models, and codes, were accomplished under approved quality assurance and control procedures and standards.	Activities associated with the development of this PMR and its related AMRs were determined to be subject to the QA program as described in the *Quality Assurance Requirements and Description* (DOE 2000). As such, collection of related data, development of analyses and models, and use and validation of software are subject to the requirements of procedures developed to implement QA program requirements.
2. Expert elicitations, when used, were conducted and documented in accordance with the guidance provided in NUREG-1563 (Kotra et al. 1996) or other acceptable approaches.	Appendix C of the *Quality Assurance Requirements and Description* (DOE 2000) and implementing procedures for expert elicitation were developed using the guidance provided in NUREG-1563 (Kotra et al. 1996). No expert elicitations are directly associated with the development of the Waste Form Degradation PMR and its related AMRs. Only two expert elicitations have indirect bearing on the Waste Form Degradation PMR. An expert elicitation on waste form degradation (CRWMS M&O 1998c) was conducted related to the TSPA-VA (DOE 1998c) and met the requirements of the QA program. An informal elicitation to evaluate solubility limits for important radionuclides (Wilson et al. 1994) was conducted before the provisions of NUREG-1563 were implemented. The results of this elicitation were used only to corroborate more recent solubility estimates for the Waste Form Degradation PMR.
3. Sufficient data (field, laboratory, and natural analog) are available to adequately define relevant parameters for the models used to evaluate performance aspects of the subissues.	The purpose of the Waste Form Degradation PMR is to summarize the technical bases of the parameters of the models. As summarized in Chapter 3, sufficient degradation data were collected by the project or available in the literature to develop defensible bounding components of the Waste Form Degradation Model and corresponding parameters of the components. While continuing testing and model refinement may narrow the ranges of parameters and allow less conservative bounds, the validity of the recommended representations described in this PMR is supported by the data and models described in the AMRs supporting this document.

Table 4.2-1. Issue Resolution Status Reports, Subissues, Acceptance Criteria, and Process Model Report Approach (NRC 1999a, 1999b, 2000a) (Continued)

NRC Technical Acceptance Criteria	PMR Approach and Section Reference
4. Sensitivity and uncertainty analyses (including consideration of alternative conceptual models) were used to determine whether additional data would be needed to better define ranges of input parameters.	For most modeling components within the Waste Form Degradation PMR, a detailed process component model of the phenomena was not developed. Instead, a simplified (abstraction) component was directly developed from the experimental data and information. Hence, sensitivity and uncertainty analyses on a detailed process component model were not appropriate and are not discussed in the Waste Form Degradation PMR. However, the discussion of each waste form degradation model component in Chapter 3 has a section titled “Confidence/Limitations/Validation" where the current confidence of the components is discussed and its suitability for the site recommendation justified. Furthermore, sensitivity and uncertainty analyses on the components of the Waste Form Degradation Model to determine whether additional data are needed are part of the general sensitivity and uncertainty analyses of the TSPA associated with the site recommendation.
5. Parameter values, assumed ranges, test data, probability distributions, and bounding assumptions used in the models are technically defensible and can reasonably account for known uncertainties.	An important purpose of the Waste Form Degradation PMR is to summarize the technical bases of models addressed in this criterion. The level of validation and methods of validation vary from model to model. Sections 3.1 through 3.8 provide the information suggested by this criterion. Further detail is provided within the AMRs.
6. Mathematical model limitations and uncertainties in modeling were defined and documented.	This criterion is reflected as requirements of the QA program (DOE 2000, Section 3.2.2, Section 3.2.3, Supplement III.2.6D); implementing procedures specify adequate documentation of assumptions. An important purpose of the Waste Form Degradation PMR is to summarize the assumptions, limitations, and uncertainties contained in the corresponding AMRs. A subsection in each component section in Chapter 3 of this document summarizes model limitations. Further discussion can be found in the AMRs cited in this report.
7. Primary and alternative modeling approaches consistent with available data and current scientific understanding were investigated and their results and limitations considered in evaluating the subissue.	Each of the sections of this PMR that describes the components of the Waste Form Degradation Model also describes alternative modeling approaches that have been considered or suggested.
8. Model outputs were validated through comparisons with outputs of detailed process models, empirical observations, or both.	Model validation requirements are established in the QA program (DOE 2000, Supplement III.2.6). The Waste Form Degradation PMR summarizes the current technical basis of model validation. Detailed documentation of model validation is included in the appropriate AMRs.
9. The structure and organization of process and abstracted models were found to adequately incorporate important design features, physical phenomena, and coupled processes.	In general, an important purpose of Waste Form Degradation PMR is to summarize the technical basis of models including important design features, physical phenomena, and coupled processes. Specifically, included/excluded FEPs are discussed in three AMRs (CRWMS M&O 2000m, 2000n, and 2000s) that support the Waste Form Degradation PMR. Section 2.3 summarizes the FEP analysis relevant to this PMR.

Table 4.2-1. Issue Resolution Status Reports, Subissues, Acceptance Criteria, and Process Model Report Approach (NRC 1999a, 1999b, 2000a) (Continued)

NRC Technical Acceptance Criteria	PMR Approach and Section Reference
SUBISSUE 3 – THE RATE AT WHICH RADIONUCLIDES IN SPENT NUCLEAR FUEL ARE RELEASED FROM THE ENGINEERED BARRIER SUBSYSTEM THROUGH THE OXIDATION AND DISSOLUTION OF SPENT FUEL	
1. DOE has considered all categories of SNF planned for disposal at the proposed repository.	The types of SNF that have been considered for disposal in the potential repository are discussed in Section 3.1 of the Waste Form Degradation PMR. As discussed in Sections 3.1, 3.3 and 3.5, the majority of radionuclides are represented in CSNF while the wide variety of DSNF is considered in the inventory and (with the exception of naval SNF) conservatively represented in performance by the most rapid release rate component (N-Reactor fuel).
2. DOE has adequately justified the selection of radionuclides tracked in the release models from SNF and their related release parameters.	With the issuance of draft regulations proposed DOE 10 CFR 63 Interim Guidance (Dyer 1999) and 40 CFR 197 (64 FR 46976), the selection of radionuclides was reevaluated and is discussed in Section 3.1 of this PMR. As discussed further in the supporting *Inventory Abstraction* AMR (CRWMS M&O 2000f) the selection of radionuclides is based on considerations of inventory, half-life, mobility, and dose contribution.
3. DOE has identified the range of environmental conditions to be expected inside breached WPs.	The In-Package Chemistry Component discussed in Section 3.2 was specifically added to the Waste Form Degradation Model to capture the range of environmental conditions expected inside the WPs. Examples of environment ranges considered include fully immersed versus flow through models and local chemistries dominated by different WP and WF components. This range of potential conditions is based on experimental observations and process modeling. EBS testing is currently underway to confirm these processes and environmental conditions.
4. DOE has identified and considered likely processes for SNF degradation and the release of radionuclides from the EBS, as follows: dissolution of the irradiated UO_2 matrix, with the consequent formation of secondary minerals and colloids; prompt release of radionuclides; degradation in the dry air environment; degradation and failure of fuel cladding; preferential dissolution of intermetallics in DOE SNF; and release of radionuclides from the WP emplacement drifts.	Section 3.3 of this PMR is devoted to dissolution of the irradiated UO_2 matrix. Another entire section, Section 3.4, is devoted to the improved modeling of the degradation and failure of fuel cladding. Section 3.4 includes the topics of prompt release of radionuclides and degradation of UO_2 and cladding in the dry air environment (this topic is also a FEP). Another entire section, Section 3.5, is devoted to dissolution of DOE SNF. Finally, Section 3.8 discusses the formation of colloids. The part of this criterion that deals with release of radionuclides from the WP emplacement drifts is not discussed in the Waste Form Degradation PMR. That criterion is discussed in the EBS PMR (CRWMS M&O 2000y).

Table 4.2-1. Issue Resolution Status Reports, Subissues, Acceptance Criteria, and Process Model Report Approach (NRC 1999a, 1999b, 2000a) (Continued)

NRC Technical Acceptance Criteria	PMR Approach and Section Reference
5. DOE has demonstrated that the numerical models used for SNF degradation and radionuclide release from the EBS are adequate representations, including consideration of uncertainties, of the expected SNF performance. They are not likely to overestimate the actual performance in the repository environment.	Models in the Waste Form Degradation PMR are regression analyses of experimental data at repository conditions (Section 3.3), bounding representations of experimental data (Sections 3.5 and 3.8), or assume very aggressive degradation to bound uncertainty (Section 3.4). Bounding representations of numerical simulations (Section 3.2) explicitly include a function to represent uncertainty, define an uncertain distribution, or use bounding values for radioisotope solubility (Section 3.7). The models were designed to avoid over estimation of performance.
6. DOE has considered the compatibility of SNF and the internal components of the WP, such as the basket materials, in the evaluation of radionuclide releases. Specifically, the SNF should not compromise the performance of the WP.	Compatibility of internal components, the SNF, and the WP has been considered in selection of materials for the WP and internals (CRWMS M&O 2000al, Section 3.3.2). The corrosion of the internal components was taken into consideration in the In-Package Chemistry Component model summarized in Section 3.2 of the Waste Form Degradation PMR and justified in the corresponding AMRs, *In-Package Chemistry Abstraction* (CRWMS M&O 2000g) and *Summary of In-Package Chemistry for Waste Forms* (CRWMS M&O 2000o).
7. DOE has justified the use of SNF test results not specifically collected for the Yucca Mountain site for the environmental conditions expected to prevail after breaching of the containers at the Yucca Mountain site.	In several cases, the Waste Form Degradation PMR uses test results of the degradation of SNF not specifically collected for the Yucca Mountain site to corroborate the YMP data (e.g., CSNF Matrix Degradation Component, Section 3.3). In the case of the CSNF Cladding Degradation Component, Section 3.4, YMP uses the extensive data collected over the past 30 years on cladding failure mechanisms. This use, however, is evaluated under specific QA procedures that address qualification of unqualified data and appropriateness of data for their intended use.

Table 4.2-1. Issue Resolution Status Reports, Subissues, Acceptance Criteria, and Process Model Report Approach (NRC 1999a, 1999b, 2000a) (Continued)

NRC Technical Acceptance Criteria	PMR Approach and Section Reference
8. DOE has conducted a consistent, sufficient, and suitable SNF corrosion and radionuclide release testing program at the time of the license application submittal. In addition, DOE has identified specific plans for further testing to reduce any significant area (or areas) of uncertainty as part of the performance confirmation program.	As explained in Sections 3.3 and 3.5, respectively, the foundation of the CSNF and DSNF degradation rates for the TSPA-SR analysis is the testing program conducted by YMP and NSNFP. A *Performance Confirmation Plan* (CRWMS M&O 2000am) has been developed that establishes the test and analysis requirements to confirm, with reasonable assurance, that the performance objective for the period after permanent repository closure is met. Postclosure performance has been shown to be insensitive to SNF corrosion and dissolution for credible ranges in applicable parameters. However, laboratory waste form testing has been included in the performance confirmation program in compliance with specific regulatory requirements regarding the waste forms and the in-package environments. These include tests to confirm dissolution rates of waste forms under bounding in-package and repository environments. Additionally, SNF cladding has been identified as a potential additional enhancement to the defense-in-depth strategy, and therefore, cladding degradation tests have also been identified for the pre-emplacement period. See Section 5.3.1.8.2 and Appendix G of the *Performance Confirmation Plan* (CRWMS M&O 2000am).
9. DOE has established an adequate program of monitoring radionuclide release from the WP during the performance confirmation period, to assure that assumptions and calculations of SNF dissolution and radionuclide release from the WP are appropriately substantiated.	A *Performance Confirmation Plan* (CRWMS M&O 2000am) has been developed that establishes the test and analysis requirements to confirm, with reasonable assurance, that the performance objective for the period after permanent repository closure is met. Because of the expected long life of the waste package, it is not possible to establish a meaningful program for monitoring radionuclide release from waste packages. In-drift environments will be monitored during the performance confirmation period to ensure no waste package breaches occur during the preclosure period. Postclosure performance has been shown to be insensitive to SNF corrosion and dissolution for credible ranges in applicable parameters. However, laboratory waste form testing has been included in the performance confirmation program in compliance with specific regulatory requirements regarding the waste forms and the in-package environments. These include tests to confirm dissolution rates of waste forms under bounding in-package and repository environments. Additionally, SNF cladding has been identified as a potential additional enhancement to the defense-in-depth strategy, and therefore, cladding degradation tests have also been identified for the pre-emplacement period. See Sections 5.3.1.8.2 and Appendix G of the *Performance Confirmation Plan* (CRWMS M&O 2000am).

Table 4.2-1. Issue Resolution Status Reports, Subissues, Acceptance Criteria, and Process Model Report Approach (NRC 1999a, 1999b, 2000a) (Continued)

NRC Technical Acceptance Criteria	PMR Approach and Section Reference
SUBISSUE 4 – THE RATE AT WHICH RADIONUCLIDES IN HIGH-LEVEL WASTE GLASS ARE RELEASED FROM THE ENGINEERED BARRIER SUBSYSTEM	
1. DOE has taken into account all types of HLW glass planned for disposal at the repository.	Consideration of the several types of HLW to be placed into the potential Yucca Mountain is discussed in Section 3.6 of the Waste Form Degradation PMR. The model is based on testing of the most representative glass to be disposed. Variations in source term from the range of HLW are expected to be within the conservative bounds of the recommended representative model.
2. DOE has adequately justified the selection of radionuclides tracked in the release models from HLW glass and their related release parameters.	After issuance of draft regulations proposed in DOE 10 CFR 63 Interim Guidance (Dyer 1999) and 40 CFR 197 (64 FR 46976), the selection of radionuclides was reevaluated, as discussed in Section 3.1, formally providing justification for the radionuclides and parameters to be tracked in release models. Further details are provided in the *Inventory Abstraction* AMR (CRWMS M&O 2000f).
3. DOE has identified the range of environmental conditions to be expected inside breached WPs containing HLW glass and, eventually, certain types of SNF as in the codisposal WPs.	The In-Package Chemistry Component discussed in Section 3.2 was specifically added to the Waste Form Degradation Model to capture the range of environmental conditions expected inside the WPs.
4. DOE has identified and considered likely processes for the degradation of HLW glass and the release of radionuclides from the EBS (i.e., dissolution of the primary phase; formation of secondary minerals and colloids; microbial action; and radionuclide releases and transport from the WP emplacement drifts).	Section 3.6 of this PMR is devoted to dissolution of the HLW glass. Section 3.8 directly uses experimented data on the formation of colloids from HLW. Microbial action is the topic of a FEP (Section 2.3.2.8 FEP 2.1.10.01.00) that applies both to SNF and HLW degradation. Only release and transport of radionuclides from the WP emplacement drifts is not discussed in this PMR, since the topic is discussed in the EBS PMR (CRWMS M&O 2000y).

Table 4.2-1. Issue Resolution Status Reports, Subissues, Acceptance Criteria, and Process Model Report Approach (NRC 1999a, 1999b, 2000a) (Continued)

NRC Technical Acceptance Criteria	PMR Approach and Section Reference
5. DOE has demonstrated that the numerical models used for determining the rate of dissolution of HLW glass and the rate of radionuclide release from the EBS are adequate representations, taking into consideration the associated uncertainties of the expected HLW glass performance, and are not likely to underestimate the actual rate of degradation of the HLW glass and the subsequent rate of release in the repository environment.	As summarized in Section 3.6 and the associated *Defense High Level Waste Glass Degradation* AMR (CRWMS M&O 2000k), the degradation model is a bounding representation of experimental results at different pH values. Furthermore, representations of numerical simulations (Section 3.2) explicitly include a function to represent uncertainty of the pH. The HLW model was designed to avoid under estimation of rate of degradation and release. In the discussion of this criterion in Section 5.4.5 of the IRSR (NRC 1999a), NRC expressed a concern that, "DOE has not considered field data on naturally occurring glasses, combined with experimental data and models on dissolution of HLW glasses, to demonstrate that long-term dissolution behavior under repository conditions can be represented by extrapolation of results from short-term laboratory tests." The DOE has considered data on naturally occurring glasses and successfully compared the paragenetic sequence observed to that found for HLW glass. As summarized in Section 3.6, DOE has used a bounding value for a variety of experimental data on dissolution of HLW glass to bolster confidence in the prediction of long-term behavior. Finally, DOE has been conservative in its use of short-term data for modeling of long-term HLW glass dissolution (see Section 3.2 and supporting AMR [CRWMS M&O 2000k]).
6. DOE has assessed the compatibility of HLW glass with internal components of the WP in the evaluation of radionuclide release, taking into consideration codisposal with DSNF in the same WP. Specifically, HLW glass should not compromise the performance of the WP.	The compatibility of internal components, the HLW, and the WP was considered in selection of materials for the WP and internal parts of the WP (CRWMS M&O 2000al, Section 3.3.2). Corrosion of the internal components was taken into consideration in the In-Package Chemistry Component model summarized in Section 3.2 and justified in the corresponding AMRs: Bounding chemistries include those dominated by waste forms such as HLW glass.
7. DOE has justified the use of test results for HLW glass that are not specifically collected for the Yucca Mountain site for environmental conditions expected to prevail after breaching of the containers at the site.	In several cases, the AMRs supporting the Waste Form Degradation PMR use test results of the degradation of HLW not specifically collected for the Yucca Mountain site. This use is evaluated under specific QA procedures that address tracking of information to be verified, qualification of unqualified data, and appropriateness of data for their intended use.

Table 4.2-1. Issue Resolution Status Reports, Subissues, Acceptance Criteria, and Process Model Report Approach (NRC 1999a, 1999b, 2000a) (Continued)

NRC Technical Acceptance Criteria	PMR Approach and Section Reference
8. DOE has conducted a consistent, sufficient, and suitable HLW glass and radionuclide release corrosion testing program at the time of the LA submittal. In addition, DOE has identified specific plans for further testing to reduce any significant area (or areas) of uncertainty as part of the performance confirmation program.	As explained in Section 3.6, the foundation of the HLW degradation rates for the TSPA related to the site recommendation is the testing program conducted by YMP. A *Performance Confirmation Plan* (CRWMS M&O 2000am) has been developed that establishes the test and analysis requirements to confirm, with reasonable assurance, that the performance objective for the period after permanent repository closure is met. Postclosure performance has been shown to be insensitive to HLW corrosion and dissolution for credible ranges in applicable parameters. However, laboratory waste form testing has been included in the performance confirmation program in compliance with specific regulatory requirements regarding the waste forms and the in-package environments. These include tests to confirm dissolution rates of waste forms under bounding in-package and repository environments. See Section 5.3.1.8.2 and Appendix G of the *Performance Confirmation Plan* (CRWMS M&O 2000am).
9. DOE has established an adequate program of monitoring radionuclide release from the WP during the performance confirmation period to assure that assumptions and calculations regarding HLW glass dissolution and radionuclide release are appropriately substantiated.	A *Performance Confirmation Plan* (CRWMS M&O 2000am) has been developed that establishes the test and analysis requirements to confirm, with reasonable assurance, that the performance objective for the period after permanent repository closure is met. Because of the expected long life of the waste package, it is not possible to establish a meaningful program for monitoring radionuclide release from waste packages. In-drift environments will be monitored during the performance confirmation period to ensure no waste package breaches occur during the preclosure period. Postclosure performance has been shown to be insensitive to HLW corrosion and dissolution for credible ranges in applicable parameters. However, laboratory waste form testing has been included in the performance confirmation program in compliance with specific regulatory requirements regarding the waste forms and the in-package environments. These include tests to confirm dissolution rates of waste forms under bounding in-package and repository environments. See Sections 5.3.1.8.2 and Appendix G of the *Performance Confirmation Plan* (CRWMS M&O 2000am).

Table 4.2-1. Issue Resolution Status Reports, Subissues, Acceptance Criteria, and Process Model Report Approach (NRC 1999a, 1999b, 2000a) (Continued)

NRC Technical Acceptance Criteria	PMR Approach and Section Reference
SUBISSUE 5 – THE EFFECT OF IN-PACKAGE CRITICALITY ON WASTE PACKAGE AND EBS PERFORMANCE	
Subissue 5 addresses the effects of in-package criticality on waste package and engineered barrier subsystem performance. In-package criticality discussions are not included in this PMR. However, the related acceptance criteria are addressed in DOE's *Disposal Criticality Analysis Methodology Topical Report* (YMP 1998) and its supporting references.	
IRSR: EVOLUTION OF THE NEAR-FIELD ENVIRONMENT	
SUBISSUE 3 – EFFECTS OF COUPLED THERMAL-HYDROLOGIC-CHEMICAL PROCESSES ON THE CHEMICAL ENVIRONMENT FOR RADIONUCLIDE RELEASE	
Data and Model Justification	
1. Available data relevant to both temporal and spatial variations in conditions affecting coupled THC effects on the chemical environment for radionuclide release were considered.	An important purpose of the Waste Form Degradation PMR is to summarize the technical bases of models. This includes consideration of the coupled effects on the chemical environment. A conservative range of bounding chemistries have been considered as discussed in Section 3.2 and the supporting AMRs *In-Package Chemistry Abstraction* (CRWMS M&O 2000g) and *Summary of In-Package Chemistry for Waste Forms* (CRWMS M&O 2000o). Where adequate THC coupled data and models are not available, conservative representations are used, such as using the minimum pH for CSNF dissolution and the maximum pH for HLW dissolution, each of which is the more conservative bound.
2. DOE's evaluation of coupled THC processes properly considered site characteristics in establishing initial and boundary conditions for conceptual models and simulations of coupled processes that may affect the chemical environment for radionuclide release.	In general, an important purpose of the Waste Form Degradation PMR is to summarize the technical bases of models, which includes the evaluation of coupled processes. As more specifically explained in Section 3.2, the In-Package Chemistry Component justified use of several initial conditions (e.g., use of J-13 water chemistry in a FEP argument) in modeling in-package chemistry. However, the Waste Form Degradation PMR is not the sole source for initial and boundary conditions. Initial and boundary conditions for hydrologic effects and temperature effects are found in the PMR that addresses the EBS (CRWMS M&O 2000y).
3. Sufficient data were collected on 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 THC-coupled processes that affect the chemical environment for radionuclide release.	An important purpose of the Waste Form Degradation PMR is to summarize the technical bases of models. As explained throughout this Waste Form Degradation PMR, sufficient data were collected by the project or were available in the literature to develop defensible bounding models of the chemical environment. Section 3.2 of this PMR and its supporting AMRs summarize the basis of EBS chemistry, although ongoing EBS testing may allow further refinement of these bounded representations.

Table 4.2-1. Issue Resolution Status Reports, Subissues, Acceptance Criteria, and Process Model Report Approach (NRC 1999a, 1999b, 2000a) (Continued)

NRC Technical Acceptance Criteria	PMR Approach and Section Reference
4. A nutrient and energy inventory calculation should be used to determine the potential for microbial activity that could impact radionuclide release.	A nutrient and energy balance was estimated for the repository. However, based on arguments for FEPs related to microbial activity (see Section 2.3), the influence of microbial activity on radionuclide release was screened out and, thus, not considered further as a process within the repository.
5. Should microbial activity be sufficient to potentially affect the chemical environment for radionuclide release, then the time-history of temperature, humidity, and dripping should be used to constrain the probability for microbial effects, such as production of organic by-products that act as complexing ligands for actinides and microbial-enhanced dissolution of the HLW glass form.	Based on arguments for FEPs related to microbial activity, microbial activity was screened out and, thus, not further considered as a process within the repository. The screening (see Sections 2.3.2.7 and 2.3.2.8), as summarized in FEPs 2.1.10.01.00 and 2.1.09.18.00 and 2.1.09.13.00 is based on limited nutrient availability limiting biological activity and the offsetting beneficial effect of microbial activity increasing colloid size and increasing filtration.
6. Sensitivity and uncertainty analyses (including consideration of alternative conceptual models) were used to determine whether additional new data are needed to better define ranges of input parameters.	In the case of the In-Package Chemistry Component and the solubility of neptunium, americium and uranium, a detailed process model was developed and parameter values were varied across a wide range. The numerical results were then used directly through a regression analysis to develop a simple empirical relationship. These models are summarized in Sections 3.2 and 3.7 of the Waste Form Degradation PMR and justified in the corresponding AMRs, *In-Package Chemistry Abstraction* (CRWMS M&O 2000g), *Summary of In-Package Chemistry for Waste Forms* (CRWMS M&O 2000o), and *Summary of Dissolved Concentration Limits* (CRWMS M&O 2000l). As explained in Chapter 1, for most other modeling components within the Waste Form Degradation PMR, a detailed process component model of the phenomena was not developed. Rather, a simplified component (abstraction) was directly developed from the experimental data and information. Hence, sensitivity and uncertainty analyses on a detailed process component model were not appropriate and are not discussed in the Waste Form Degradation PMR. Instead, sensitivity and uncertainty analyses on the simplified components of the Waste Form Degradation Model to determine whether additional data are needed as part of the general sensitivity and uncertainty analyses of the TSPA related to the site recommendation and discussed in its accompanying documentation.

Table 4.2-1. Issue Resolution Status Reports, Subissues, Acceptance Criteria, and Process Model Report Approach (NRC 1999a, 1999b, 2000a) (Continued)

NRC Technical Acceptance Criteria	PMR Approach and Section Reference
7. If the testing program for coupled THC processes on the chemical environment for radionuclide release from the EBS is not complete at the time of license application, or if sensitivity and uncertainty analyses indicate additional data are needed, DOE has identified specific plans to acquire the necessary information as part of the performance confirmation program.	Testing and model refinement for coupled THC processes is ongoing and will be complete or sufficiently bounded to support licensing arguments at the time of the license application. A *Performance Confirmation Plan* (CRWMS M&O 2000am) has been developed that establishes the test and analysis requirements to confirm, with reasonable assurance, that the performance objective for the period after permanent repository closure is met. Coupled processes and their implications for radionuclide release will be measured at several emplacement drift locations across the repository horizon from nearby observation drifts, in addition to specific tests under simulated postclosure conditions. Waste form testing during the pre-emplacement and monitoring periods is also identified under reasonably bounding chemical environments to confirm alteration and dissolution characteristics of HLW glass and SNF waste forms. See sections 5.3.1.5, 5.3.1.6, 5.3.1.8, and 5.3.2, as well as Appendix G of the *Performance Confirmation Plan* (CRWMS M&O 2000am).
Data Uncertainty and Verification	
1. Reasonable or conservative ranges of parameters or functional relations were used to determine effects of coupled THC processes on the chemical environment for radionuclide release. Parameter values, assumed ranges, probability distributions, and bounding assumptions are technically defensible and reasonably account for uncertainties.	Models in the Waste Form Degradation PMR are regression analyses of experimental data at repository conditions (Section 3.3), bounding representations of experimental data (Sections 3.5 and 3.8), or very aggressive assumptions of degradation to bound uncertainty (Section 3.4). Bounding representations of numerical simulations (Section 3.2) explicitly include a function to represent uncertainty, define an uncertain distribution, or use bounding values for radioisotope solubility (Section 3.7). The bounded, or most aggressive conditions are determined from coupled THC calculations.
2. Uncertainty in data due to both temporal and spatial variations in conditions affecting coupled THC effects on the chemical environment for radionuclide release was considered.	The In-Package Chemistry Component discussed in Section 3.2 uses the available data to couple temporal thermal effects (waste temperature), temporal hydrologic effects (seepage into the package), and temporal chemical effects (degradation rates of steel, aluminum, HLW, SNF) to evaluate the chemical environment inside the WP. Although the conceptual model of a fully saturated WP did not assume spatial variations within one package, spatial variations in temperature were considered. Spatial discretization across the repository and temporal discretization of the simulation is not the topic of the Waste Form Degradation PMR; rather, justification of the temporal and spatial discretization is discussed in the TSPA-SR analysis report (under development).

Table 4.2-1. Issue Resolution Status Reports, Subissues, Acceptance Criteria, and Process Model Report Approach (NRC 1999a, 1999b, 2000a) (Continued)

NRC Technical Acceptance Criteria	PMR Approach and Section Reference
3. DOE's evaluation of coupled THC processes properly considered 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 THC coupled processes that affect the chemical environment for radionuclide release.	The In-Package Chemistry Component (Section 3.2) justified use of several initial conditions (e.g., use of J-13 water chemistry in a FEP argument) in modeling in-package chemistry. However, the Waste Form Degradation PMR is not the sole source for initial and boundary conditions. Initial and boundary conditions for hydrologic effects and temperature effects are found in the PMR related to the EBS (CRWMS M&O 2000y).
4. The initial conditions, boundary conditions, and computational domain used in sensitivity analyses involving coupled THC effects on the chemical environment for radionuclide release were consistent with available data.	The In-Package Chemistry Component (Section 3.2) justified use of several initial conditions (e.g., use of J-13 water chemistry in a FEP argument) in modeling in-package chemistry. However, the Waste Form Degradation PMR is not the sole source for initial and boundary conditions. Initial and boundary conditions for hydrologic effects and temperature effects are found in the PMR related to the EBS (CRWMS M&O 2000y).
5. DOE's performance confirmation program should assess whether the natural system and engineered materials are functioning as intended and anticipated with regard to coupled THC effects on the chemical environment for radionuclide release from the EBS.	A *Performance Confirmation Plan* (CRWMS M&O 2000am) has been developed that establishes the test and analysis requirements to confirm, with reasonable assurance, that the performance objective for the period after permanent repository closure is met. Coupled processes and their implications for radionuclide release will be measured at several emplacement drift locations across the repository horizon from nearby observation drifts, in addition to specific tests under simulated postclosure conditions. Waste form testing during the pre-emplacement and monitoring periods is also identified under reasonably bounding chemical environments to confirm alteration and dissolution characteristics of HLW glass and SNF waste forms. See sections 5.3.1.5, 5.3.1.6, 5.3.1.8, and 5.3.2, as well as Appendix G of the *Performance Confirmation Plan* (CRWMS M&O 2000am).
Model Uncertainty	
1. Appropriate models, tests, and analyses were used that are sensitive to the THC couplings under consideration for both natural and engineering systems as described in the following examples. The effects of THC coupled processes that may occur in the natural setting or due to interactions with engineered materials or their alteration products include: (i) thermal-hydrologic effects on gas and water chemistry; (ii) hydrothermally driven geochemical reactions, such as zeolitization of volcanic glass; (iii) dehydration of hydrous phases liberating moisture; (iv) effects of microbial processes; and (v) changes in water chemistry that may result from interactions between cementitious, or WP, materials and groundwater, which, in turn, may affect the chemical environment for radionuclide release.	The In-Package Chemistry Component (Section 3.2) uses the available data to couple temporal thermal effects (waste temperature), temporal hydrologic effects (seepage into the package), and temporal chemical effects (degradation rates of steel, aluminum, HLW, SNF) to evaluate the chemical environment inside the WP. Hydrothermally driven geochemical processes and zeolitization of volcanic glass (item ii) and dehydration of tuff (item iii) occur in the near-field environment; they are outside the scope of this PMR. Based on arguments found in the FEPs discussion, microbial activity (item iv) was screened out (see Section 2.3). Interactions of waters exiting the WP with invert and structural material (item v) are found in the EBS PMR (CRWMS M&O 2000y).

Table 4.2-1. Issue Resolution Status Reports, Subissues, Acceptance Criteria, and Process Model Report Approach (NRC 1999a, 1999b, 2000a) (Continued)

NRC Technical Acceptance Criteria	PMR Approach and Section Reference
2. Alternative modeling approaches consistent with available data and current scientific understanding were investigated, and their results and limitations were appropriately considered.	Each of the sections describing the components of the Waste Form Degradation Model (Sections 3.3, 3.5, 3.6) of this PMR describes alternative modeling approaches that were considered.
3. DOE provided a reasonable description of the mathematical models included in its analyses of coupled THC effects on the chemical environment for radionuclide release. The description should include a discussion of alternative modeling approaches not considered in its final analysis and the limitations and uncertainties of the chosen model.	Thorough descriptions of models are included in the Waste Form Degradation PMR and related AMRs. Additionally, the quality assurance program and implementing procedures require that analyses, models, and calculations provide adequate documentation of assumptions (DOE 2000, Section 3.2.2, Section 3.2.3, and Supplement III.2.6). Procedure AP-3.10Q (Attachment I, item 6) addresses discussion, as applicable, of any alternate methods or models that were not used and the rationale for not selecting them.
Model Verification	
1. The mathematical models for coupled THC effects on the chemical environment for radionuclide release are consistent with conceptual models based on inferences about the near-field environment, field data and natural alteration observed at the site, and expected engineered materials.	Model validation requirements are established in the quality assurance program (DOE 2000, Supplement III.2.6). The Waste Form Degradation PMR summarizes the current technical basis of the CSNF Matrix Degradation Component (Section 3.3), the CSNF Cladding Degradation Component, (Section 3.4), the HLW Degradation Component (Section 3.6), and the Dissolved Radioisotope Concentration Component (Section 3.7), which are all functions of temperature and the chemical effects evaluated in the In-Package Chemistry Component discussed in Section 3.2. Furthermore, the In-Package Chemistry Component is a function of the influx of water (hydrology) into the WP. The justification of and confidence in the components are discussed in sections titled “Confidence/Limitations/ Validation.”
2. DOE appropriately adopted accepted and well-documented procedures to construct and test the numerical models used to simulate coupled THC effects on the chemical environment for radionuclide release.	An important purpose of the Waste Form Degradation PMR is to summarize the assumptions, limitations, and uncertainties of the waste form degradation model. More importantly, the QA procedures for the analyses, models, and calculations require adequate documentation of methods used to construct models. The Waste Form Degradation PMR summarizes the current technical basis of the CSNF Matrix Degradation Component (Section 3.3), the CSNF Cladding Degradation Component, (Section 3.4), the HLW Degradation Component (Section 3.6), and the Dissolved Radioisotope Concentration Component (Section 3.7), which all are a function of temperature and the chemical effects evaluated in the In-Package Chemistry Component discussed in Section 3.2. The validation of the components are discussed in sections titled “Confidence/Limitations/Validation.”

Table 4.2-1. Issue Resolution Status Reports, Subissues, Acceptance Criteria, and Process Model Report Approach (NRC 1999a, 1999b, 2000a) (Continued)

NRC Technical Acceptance Criteria	PMR Approach and Section Reference
3. Abstracted models for coupled THC effects on the chemical environment for radionuclide release were based on the same assumptions and approximations shown to be appropriate for closely analogous natural or experimental systems. Abstracted model results were verified through comparison to outputs of detailed process models and empirical observations. Abstracted model results are compared with different mathematical models to judge robustness of results.	For most modeling components within the Waste Form Degradation PMR, a detailed process component model of the phenomena was not developed. Rather a simplified (abstraction) component was directly developed from the experimental observations and information. For the In-Package Chemistry Component (Section 3.2), a detailed process model was developed and then the numerical results used directly through regression to develop a simple empirical relationship, as summarized in Chapter 3 and justified in the corresponding AMRs, *In-Package Chemistry Abstraction* (CRWMS M&O 2000g) and *Summary of In-Package Chemistry for Waste Forms* (CRWMS M&O 2000o). Other views and alternative models are discussed in each section of Chapter 3 as appropriate.
Integration	
1. DOE has considered the relevant features, events, and processes. The abstracted models adequately incorporated important design features; physical phenomena and couplings; and used consistent and appropriate assumptions throughout.	Specifically included/excluded FEPs are discussed in three AMRs that support the Waste Form Degradation PMR, and many of these are discussed in the Waste Form Degradation PMR itself. Section 2.3 summarizes the FEP screening relevant to this PMR.
2. Models reasonably accounted for known temporal and spatial variations in conditions affecting coupled THC effects on the chemical environment for radionuclide release.	The In-Package Chemistry Component (Section 3.2) uses the available data to couple temporal thermal effects (waste temperature), temporal hydrologic effects (seepage into the package), and temporal chemical effects (degradation rates of steel, aluminum, HLW, SNF) to evaluate the chemical environment inside the WP.
3. Not all THC couplings may be determined to be important to performance, and DOE may adopt assumptions to simplify performance assessment analyses. If potentially important couplings are neglected, DOE should provide a technical basis for doing so. The technical basis can include activities, such as independent modeling, laboratory or field data, or sensitivity studies.	Specifically included/excluded FEPs are discussed in AMRs that support the Waste Form Degradation PMR, and many of these are discussed in the Waste Form Degradation PMR itself. Section 2.3 summarizes the FEP screening relevant to this PMR. Section 3.2 summarizes the coupled process analysis describing chemical conditions in the waste package, and how this is abstracted into conservative bounds for waste form modeling.
4. Where simplifications for modeling coupled THC effects on the chemical environment for radionuclide release were used for performance assessment analyses instead of detailed process models, the bases used for modeling assumptions and approximations were documented and justified.	The QA procedures for analyses, models, and calculations require adequate documentation of modeling assumptions and approximations used to construct models. Bases used for modeling assumptions and approximations are well-documented in supporting AMRs.

Table 4.2-1. Issue Resolution Status Reports, Subissues, Acceptance Criteria, and Process Model Report Approach (NRC 1999a, 1999b, 2000a) (Continued)

NRC Technical Acceptance Criteria	PMR Approach and Section Reference
Programmatic	
1. Data and models were collected, developed, and documented under acceptable QA procedures.	Activities associated with development of this PMR and its related AMRs were determined to be subject to the QA program as described in the *Quality Assurance Requirements and Description* (DOE 2000). As such, collection of related data, development of analyses, calculations, and models, and use and validation of software are subject to the requirements of procedures developed to implement quality assurance program requirements.
2. Deficiency reports concerning data quality on issues related to coupled THC effects on the chemical environment for radionuclide release were closed.	Activities associated with deficiencies and associated corrective action are subject to the QA program as described in the *Quality Assurance Requirements and Description* (DOE 2000). Deficiencies are identified, tracked, resolved, and closed based on implementing procedures. While data quality issues have been well documented in deficiency documents, the corrective action process assures effective resolution.
3. If used, expert elicitations were conducted and documented in accordance with the guidance in NUREG-1563 (Kotra et al. 1996) or other acceptable approaches.	Appendix C of the *Quality Assurance Requirements and Description* (DOE 2000) and implementing procedures for expert elicitation were developed using the guidance provided in NUREG-1563 (Kotra et al 1996). No expert elicitations are directly associated with the development of the Waste Form Degradation PMR and its related AMRs. Only two expert elicitations have indirect bearing on the Waste Form Degradation PMR. An expert elicitation on waste form degradation (CRWMS M&O 1998c) was conducted related to the TSPA-VA and met the requirements of the QA program. An informal elicitation to evaluate solubility limits for important radionuclides (Wilson et al. 1994) was conducted before the provisions of NUREG-1563 (Kotra et al. 1996) were implemented. The results of this elicitation were used only to corroborate more recent solubility estimates for the Waste Form Degradation PMR.

Table 4.2-1. Issue Resolution Status Reports, Subissues, Acceptance Criteria, and Process Model Report Approach (NRC 1999a, 1999b, 2000a) (Continued)

NRC Technical Acceptance Criteria	PMR Approach and Section Reference
IRSR: TOTAL SYSTEM PERFORMANCE ASSESSMENT AND INTEGRATION	
Programmatic	
1. The collection, documentation, and development of data, models, and/or computer codes have been performed under acceptable quality assurance (QA) procedures, or if the data, models and/or computer codes were not subject to an acceptable QA procedure, they have been appropriately qualified.	Activities associated with development of this PMR and its related AMRs were determined to be subject to the quality assurance program as described in the *Quality Assurance Requirements and Description* (DOE 2000). As such, collection of related data, development of analyses and models, and use and validation of software are subject to the requirements of procedures developed to implement quality assurance program requirements. The QA program (DOE 2000) addresses use of data, models, and/or codes not subject to development under a QA program, and these provisions were applied in accordance with related implementing procedures. Detailed documentation of data, models and/or computer codes is in the AMRs, which this PMR summarizes.
2. Formal expert elicitations can be used to support data synthesis and model development for DOE's TSPA, provided that the elicitations are conducted and documented under acceptable procedures.	Appendix C of the *Quality Assurance Requirements and Description* (DOE 2000) and implementing procedures for expert elicitation were developed using the guidance provided in NUREG-1563 (Kotra et al. 1996). No expert elicitations are directly associated with the development of the Waste Form Degradation PMR and its related AMRs. Only two expert elicitations have indirect bearing on the Waste Form Degradation PMR. An expert elicitation on waste form degradation (CRWMS M&O 1998c) was conducted related to the TSPA-VA and met the requirements of the QA program. An informal elicitation to evaluate solubility limits for important radionuclides (Wilson et al. 1994) was conducted before the provisions of NUREG-1563 (Kotra et al. 1996) were implemented. The results of this elicitation were used only to corroborate more recent solubility estimates for the Waste Form Degradation PMR.
SUBISSUE 1 – System Description and Demonstration of Multiple Barriers	
Transparency and Traceability of the Analysis	
Total System Performance Assessment Documentation Style, Structure, and Organization	
1. Documents and reports are complete, clear, and consistent.	The Waste Form Degradation PMR was carefully structured to be complete, clear, and consistent. Reviews of the draft document included checks for completeness, clarity and consistency.
2. Information is amply cross-referenced.	The Waste Form Degradation PMR contains ample references to data sources, codes, assumptions, and conclusions.

Table 4.2-1. Issue Resolution Status Reports, Subissues, Acceptance Criteria, and Process Model Report Approach (NRC 1999a, 1999b, 2000a) (Continued)

NRC Technical Acceptance Criteria	PMR Approach and Section Reference
Features, Events, and Processes Identification and Screening	
1. The screening process by which FEPs were included or excluded from the TSPA is fully described.	Section 2.3 of this PMR summarizes included or excluded FEPs including the rationale for the decisions.
2. Relationships between relevant FEPs are fully described.	Section 2.3 of this PMR describes the relationship between FEPs. The miscellaneous waste form FEPs AMR (CRWMS M&O 2000n), cladding FEPs AMR (CRWMS M&O 2000s), and colloid summary AMR (CRWMS M&O 2000m) provide additional documentation including the TSPA disposition of FEPs, IRSR issues relevant to specific FEPs, and analysis and discussion on specific FEPs.
Abstraction Methodology	
1. The levels and method(s) of abstraction are described starting from assumptions defining the scope of the assessment down to assumptions concerning specific processes and the validity of given data.	For each model in the Waste Form Degradation PMR, descriptions are provided of process models and, if the models are abstracted, descriptions of the abstractions of the models are provided. The description includes a summary of data and assumptions used to construct models. The supporting AMRs describing the models and the abstracted models provide additional details regarding data and assumptions.
2. A mapping (e.g., a road map diagram, a traceability matrix, a cross-reference matrix) is provided to show what conceptual features (e.g., patterns of volcanic events) and processes are represented in the abstracted models, and by what algorithms.	The Waste Form Degradation PMR and supporting AMRs provide descriptions of the basis for the decisions and assumptions that were made during the abstraction process. Each major section of Chapter 3 contains figures showing the abstracted models. In addition, most major sections of Chapter 3 contain figures showing features of the conceptual models.
3. An explicit discussion of uncertainty is provided to identify which issues and factors are of most concern or are key sources of disagreement among experts.	The Waste Form Degradation PMR provides a discussion of uncertainties and limitations for the major process models included in the report. The supporting AMRs describing the abstracted models provide additional details regarding uncertainties and limitations.
Data Use and Validity	
1. The pedigree of data from laboratory tests, natural analogs, and the site is clearly identified.	Section 1.3 and Chapter 3 of this PMR summarize the quality assurance status of the data and software used in the component models.
2. Input parameter development and basis for their selection is described.	The Waste Form Degradation PMR discusses input parameter development and the basis for using the parameters. The supporting AMRs describing the models provide additional details regarding input parameter development and the basis for input selection.

Table 4.2-1. Issue Resolution Status Reports, Subissues, Acceptance Criteria, and Process Model Report Approach (NRC 1999a, 1999b, 2000a) (Continued)

NRC Technical Acceptance Criteria	PMR Approach and Section Reference
3. A thorough description of the method used to identify performance confirmation program parameters [has been developed].	The *Performance Confirmation Plan* (CRWMS M&O 2000am) specifically addresses the methodology for identifying and selecting parameters that are important to performance based upon TSPA sensitivity analyses and the repository safety strategy. Methods used to collect information for each parameter will be described by the performance confirmation plan or relevant supporting documents to support the license application. Performance confirmation test selection and rationale is also described in the plan based upon the significance of the parameter being measured, and the ability of the test to distinguish construction, emplacement, or time dependent changes in the parameter significant to performance.
Assessment Results	
1. PA results (i.e., the peak expected annual dose within the compliance period) can be traced back to applicable analyses that identify the FEPs, assumptions, input parameters, and models in the PA.	The TSPA-SR summarizes features, processes, conceptual models, and their implementation into the TSPA. This discussion will be based in part on information provided by the Waste Form Degradation PMR.
2. The PA results include a presentation of intermediate results that provide insight into the assessment (e.g., results of intermediate calculations of the behavior of individual barriers)	TSPA-SR provides performance analysis results for the total system and will include intermediate results for the components of the system.
Code Design and Data Flow	
1. The flow of information (input and output) between the various modules is clearly described.	TSPA-SR provides a description of information flow between component models including couplings between information and data, conceptual and process-level models, and abstracted models.
2. Supporting documentation (e.g., user's manuals, design documents) clearly describes code structure and relationships between modules.	TSPA-SR describes the TSPA code and provides a reference to supporting documentation such as the user's guide.
SUBISSUE 2 - SCENARIO ANALYSIS	
Identification of an Initial Set of Processes and Events	
1. DOE has identified a comprehensive list of processes and events that: (i) are present or might occur in the Yucca Mountain region, and (ii) includes those processes and events that have the potential to influence repository performance.	Section 2.3 provides a list of the processes and events applicable to this PMR. It summarizes the screening arguments and dispositions for the FEPs. The process for developing and screening FEPs was formalized for TSPA-SR analysis. An overview of the process adopted by DOE is described in *Total System Performance Assessment-Site Recommendation Methods and Assumptions* (CRWMS M&O 1999f). Over 1,786 FEPs were identified that are applicable to the Yucca Mountain region. One hundred fourteen are related in some way to waste form degradation (e.g., criticality), of these, 86 were specifically assigned to the Waste Form Degradation PMR.

Table 4.2-1. Issue Resolution Status Reports, Subissues, Acceptance Criteria, and Process Model Report Approach (NRC 1999a, 1999b, 2000a) (Continued)

NRC Technical Acceptance Criteria	PMR Approach and Section Reference
Classification of Processes and Events	
1. DOE has provided adequate documentation identifying how its initial list of processes and events has been grouped into categories.	Section 2.3 of this PMR describes the manner that included FEPs were incorporated into the Waste Form Degradation Model. Section 2.3 also summarizes justification for screening arguments and TSPA dispositions. For comprehensiveness, traceability is maintained in the FEPs database from the secondary to the related primary FEPs.
2. Categorization of processes and events is compatible with the use of categories during the screening of processes and events.	The NRC staff will review the categorization of processes and events as described in the various PMRs. Section 2.3 of this PMR describe the features, events, and processes applicable to this PMR. The AMRs supporting this section, *Miscellaneous Waste-Form FEPs* (CRWMS M&O 2000n) *Waste Form Colloid-Associated Concentrations Limits: Abstraction and Summary* (CRWMS M&O 2000m), and *Clad Degradation-FEPs Screening Arguments* AMR (CRWMS M&O 2000s), provide documentation and justification for screening arguments and TSPA dispositions. Documentation is maintained of all mapping of FEPs into primary and secondary categories. The PMRs and AMRs describe the categorization of processes and events in sufficient detail to determine that events are not narrowly defined and that the categorization of processes and events is compatible with the use of categories during the screening of processes and events.
Screening of Processes and Events	
1. Categories of processes and events that are not credible for the Yucca Mountain repository because of waste characteristics, repository design, or site characteristics are identified and sufficient justification is provided for DOE's conclusions.	The arguments in the AMRs containing FEPs include a statement of the screening decision for each FEP. Justification is provided for each excluded FEP, including the criterion on which it was excluded and the technical basis for the screening argument.
2. The probability assigned to each category of processes and events not screened based on criterion 1 or 2 is consistent with the site information, well documented, and appropriately considers uncertainty.	Any probability estimates for FEPs related to waste form degradation are generally based on rational arguments as to feasibility of the FEP consistent with site information that considers uncertainty.
3. DOE has demonstrated that processes and events screened from the performance assessment on the basis of their probability of occurrence, have a probability of less than one chance in 10,000 of occurring over 10,000 years.	In the three AMRs that address waste form FEPs (CRWMS M&O 2000m, 2000n, and 2000s), justification is provided for each excluded FEP, including the criterion on which it was excluded and the technical basis for the screening argument. The probability assigned to a FEP may be one of the screening criteria but were only excluded solely based on this criterion if it could be argued to have a probability of occurrence of less than 10^{-8}/year.
4. DOE has demonstrated that categories of processes and events omitted from the performance assessment on the basis that their omission would not significantly change the calculated expected annual dose, do not significantly change the calculated expected annual dose.	All of the three AMRs that address waste form FEPs provide documentation and justification for screening arguments and TSPA disposition. For omitted categories, documentation includes the criteria on which it was excluded and the technical basis for the screening argument.

Table 4.2-1. Issue Resolution Status Reports, Subissues, Acceptance Criteria, and Process Model Report Approach (NRC 1999a, 1999b, 2000a) (Continued)

NRC Technical Acceptance Criteria	PMR Approach and Section Reference
Formation of Scenarios	
1. DOE has provided adequate documentation identifying: (i) whether processes and events have been addressed through consequence model abstraction or scenario analysis and (ii) how the remaining categories of processes and events have been combined into scenario classes.	FEPs that have not been excluded are identified as either expected FEPs or disruptive FEPs. Expected FEPs have been included in the TSPA-SR analysis nominal scenario simulated in the TSPA-SR analysis.
2. The set of scenario classes is mutually exclusive and complete.	Section 2.3 of this PMR describe the features, events, and processes in this PMR. The AMRs supporting this section, *Miscellaneous Waste-Form FEPs,* (CRWMS M&O 2000n) *Waste Form Colloid-Associated Concentrations Limits: Abstraction and Summary* (CRWMS M&O 2000m), and *Clad Degradation-FEPs Screening Arguments* (CRWMS M&O 2000s), provide documentation and justification for screening arguments and TSPA dispositions. No specific scenarios were constructed for the Waste Form Degradation PMR; however, taken together, the entire set of PMRs will provide NRC staff with sufficient documentation to evaluate whether the set of scenario classes developed is mutually exclusive and complete and whether expected FEPs are addressed through model abstraction or through incorporation into scenarios.
Screening of Scenario Classes	
1. Scenario classes that are not credible for the Yucca Mountain repository because of waste characteristics, repository design, or site characteristics—individually or in combination—are identified, and sufficient justification is provided for DOE's conclusions.	In general, scenarios have been screened using the same regulatory, probability, and consequence criteria used for screening individual FEPs. Documentation of this process includes identification of any scenarios that have been screened from the analysis and the technical basis for that screening decision. FEPs related to waste form degradation have not been grouped into scenario classes for further screening. Rather, the FEPs have been included in the nominal scenario simulated in the TSPA-SR analysis.
2. The probability assigned to each scenario class is consistent with site information, well documented, and appropriately considers uncertainty.	FEPs related to waste form degradation have not been grouped into scenario classes for further screening. Rather, the FEPs have been included in the nominal scenario simulated in the TSPA-SR analysis, and have appropriately considered uncertainty.
3. Scenario classes that combine categories of processes and events may be screened from the performance assessment on the basis of their probability of occurrence, provided: (i) the probability used for screening the scenario class is defined from combinations of initiating processes and events, and (ii) DOE has demonstrated that they have a probability of less than one chance in 10,000 of occurring over 10,000 years.	FEPs related to waste form degradation have not been grouped into scenario classes for further screening. Rather, the FEPs have been included in the nominal scenario simulated in the TSPA-SR analysis. Thus, scenario screening based on probability of occurrence is not applicable to FEPs related to this PMR. Inclusion in the nominal TSPA-SR simulation provides an alternative approach that achieves the same objective.

Table 4.2-1. Issue Resolution Status Reports, Subissues, Acceptance Criteria, and Process Model Report Approach (NRC 1999a, 1999b, 2000a) (Continued)

NRC Technical Acceptance Criteria	PMR Approach and Section Reference
4. Scenario classes may be omitted from the performance assessment on the basis that their omission would not significantly change the calculated expected annual dose, provided DOE has demonstrated that excluded categories of processes and events would not significantly change the calculated expected annual dose.	FEPs related to waste form degradation have not been grouped into scenario classes for further screening. Rather, the FEPs have been included in the nominal scenario simulated in the TSPA-SR analysis. Thus omission of scenario classes from the TSPA based on significance to the calculated expected dose is not applicable to FEPs related to this PMR.
SUBISSUE 3 – MODEL ABSTRACTION	
Quantity and Chemistry of Water Contacting Waste Packages and Waste Forms	
1. Sufficient data (field, laboratory, and/or natural analog data) are available to adequately define relevant parameters and conceptual models necessary for developing the quantity and chemistry of water contacting WPs and waste forms abstracted in TSPA. Where adequate data do not exist, other information sources such as expert elicitation have been appropriately incorporated into the TSPA.	An important purpose of Waste Form Degradation PMR is to summarize the technical bases of models and corresponding parameters. As explained throughout this Waste Form Degradation PMR, sufficient data were collected by the project or were available in the literature to develop defensible bounding models of the chemical environment. The specific aspects of the water chemistry are discussed in Section 3.2 and corresponding AMRs, *In-Package Chemistry Abstraction* (CRWMS M&O 2000g) and *Summary of In-Package Chemistry for Waste Forms* (CRWMS M&O 2000o).
2. Parameter values, assumed ranges, probability distributions, and bounding assumptions used in the quantity and chemistry of water contacting WPs and waste forms abstraction, such as the pH, carbonate concentration, chloride concentration, and amount of water flowing in and out of the breached WP, are technically defensible and reasonably account for uncertainties and variability.	Variations in water chemistry and three effects on in-package chemistry are discussed in Section 3.2. In addition, as described in Section 3.4, a specific model component was added to the total system performance assessment for site recommendation to reasonably account for uncertainties and variabilities in chemistry of water contacting the waste forms. The quantity of water entering the WP was evaluated in the PMR on the EBS (CRWMS M&O 2000y).
3. Alternative modeling approaches consistent with available data and current scientific understanding are investigated and results and limitations appropriately factored into the quantity and chemistry of water contacting WPs and waste forms abstraction.	Each of the sections describing the components of the Waste Form Degradation Model of the Waste Form Degradation PMR describes alternative modeling approaches that could have been used.

Table 4.2-1. Issue Resolution Status Reports, Subissues, Acceptance Criteria, and Process Model Report Approach (NRC 1999a, 1999b, 2000a) (Continued)

NRC Technical Acceptance Criteria	PMR Approach and Section Reference
4. Output of quantity and chemistry of water contacting WPs and waste forms abstraction are verified through comparison to output of detailed process models and/or empirical observations (laboratory testings or natural analogs, or both).	The Waste Form Degradation PMR summarizes the current technical basis of models. For most modeling components within the Waste Form Degradation PMR, a detailed process component model of the phenomena was not developed. Rather, a simplified (abstraction) component was directly developed from the experimental observations and information. For the In-Package Chemistry Component, a detailed process model was developed, and then the numerical results used directly through regression to develop a simple empirical relationship, as summarized in Section 3.2 and justified in the corresponding AMRs, *In-Package Chemistry Abstraction* (CRWMS M&O 2000g) and *Summary of In-Package Chemistry for Waste Forms* (CRWMS M&O 2000o).
5. Important design features, physical phenomena and couplings, and consistent and appropriate assumptions are incorporated into the quantity and chemistry of water contacting WPs and waste forms abstraction.	Specifically included/excluded FEPs relevant to the quantity and chemistry of water are discussed in AMRs that support the Waste Form Degradation PMR, and many of these are discussed in the Waste Form Degradation PMR. Section 2.3 summarizes the FEP screening relevant to this PMR. Section 3.2 summarizes the coupled process analysis describing chemical conditions in the waste package and how it is abstracted into conservative bounds for waste form modeling.
Radionuclide Release Rates and Solubility Limits	
1. Sufficient data (field, laboratory, and/or natural analog data) are available to adequately define relevant parameters and conceptual models necessary for developing radionuclide release rates and solubility limits abstracted in TSPA. Where adequate data do not exist, other information sources such as expert elicitation have been appropriately incorporated into the TSPA.	As explained throughout this Waste Form Degradation PMR, sufficient data were collected by the project or available in the literature to develop defensible bounding models of the chemical environment. Specific aspects of the radioisotope release rates are discussed in Sections 3.3, 3.4, 3.5, and 3.6 and corresponding AMRs. Specific aspects of the solubility limits are discussed in Section 3.7 and the corresponding AMRs, *Summary of Dissolved Concentration Limits* (CRWMS M&O 2000l) and *Pure Phase Solubility Limits-LANL* (CRWMS M&O 2000v).
2. Parameter values, assumed ranges, probability distributions, and bounding assumptions used in the radionuclide release rates and solubility limits abstraction, such as the pH, temperature, and amount of liquid contacting the waste forms, are technically defensible and reasonably account for uncertainties and variabilities.	An important purpose of Waste Form Degradation PMR is to summarize the technical bases of model parameters. Specific aspects of the radioisotope release rates are discussed in Sections 3.3, 3.4., 3.5, and 3.6 and the corresponding AMRs. Specific aspects of the solubility limits are discussed in Section 3.7 and the corresponding AMRs. To better characterize the uncertainty in the radionuclide release rates and solubility limits, the corresponding modeling components were directly coupled with In-Package Chemistry Component (Section 3.2). In turn, the In-Package Chemistry Component uses the available data to couple temporal thermal effects (waste temperature), temporal hydrologic effects (seepage into the package), and temporal chemical effects (degradation rates of steel, aluminum, HLW, SNF) to evaluate the chemical environment inside the WP.

Table 4.2-1. Issue Resolution Status Reports, Subissues, Acceptance Criteria, and Process Model Report Approach (NRC 1999a, 1999b, 2000a) (Continued)

NRC Technical Acceptance Criteria	PMR Approach and Section Reference
3. Alternative waste form dissolution and radionuclide release modeling approaches consistent with available data and current scientific understanding are investigated and results and limitations appropriately factored into the radionuclide release rates and solubility limits abstraction.	Each of the sections describing the components of the Waste Form Degradation Model of the Waste Form Degradation PMR describes alternative modeling approaches that were considered. The alternative modeling approaches for the radioisotope release rates and solubility limits are discussed in Sections 3.3, 3.4, 3.5, 3.6, and 3.7.
4. Radionuclide release rates and solubility limits abstraction outputs is supported by comparison to outputs of detailed process models or empirical observations (field, laboratory, and natural analog data).	The Waste Form Degradation PMR summarizes the current technical basis of models. For most modeling components within the Waste Form Degradation PMR, a detailed process component model of the phenomena was not developed. Rather, a simplified (abstraction) component was directly developed from the experimental observations and information. For radionuclides of neptunium, americium, and uranium and the directly related In-Package Chemistry Component (Section 3.2), a detailed process model was developed and then the numerical results used directly through regression to develop a simple empirical relationship, as summarized in Sections 3.2 and 3.7. Although material analog data were not used directly, the paragenetic sequence observed in experiments mirrors natural analogues.
5. Important design features, physical phenomena and couplings, and consistent and appropriate assumptions are incorporated into the radionuclide release rates and solubility limits abstraction.	Specifically included/excluded FEPs are discussed in AMRs that support the Waste Form Degradation PMR and many of these are discussed in the Waste Form Degradation PMR. Section 2.3 summarizes the FEP screening relevant to this PMR. Section 3.2 summarizes the coupled process analysis describing chemical conditions in the waste package, and how this is abstracted into conservative bounds for waste form modeling.

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5. SUMMARY AND CONCLUSIONS

The *Waste Form Degradation Process Model Report* (Waste Form Degradation PMR) is one of nine PMRs that have the shared objective of describing the technical information used in the TSPA-SR that was conducted to evaluate the postclosure performance of a potential monitored geologic repository at Yucca Mountain. The TSPA-SR will be used in the preparation of a document for a Secretarial decision on whether to recommend the Yucca Mountain site for development as a repository. The Waste Form Degradation PMR summarizes the results of investigations on the degradation of radioactive spent nuclear fuel and high-level waste.

The performance of the potential Yucca Mountain repository will depend on the rate of radionuclide release from the waste packages after the containers are eventually breached. After breach, water, or water vapor may enter the packages and contact exposed waste forms. The waste forms will degrade slowly and release radionuclides, as solutes and colloids, into the contacting groundwater as a result of a broad range of processes and events. This Waste Form Degradation PMR, together with the supporting AMRs, describes the approach and scientific basis for modeling. In order to ensure that all relevant processes were included, a detailed analysis of FEPs believed to be important in controlling the rate of radionuclide release was conducted.

The decision to include or exclude each of the waste form FEPs was based on criteria in Section 114(e) of the proposed DOE 10 CFR Part 63 Interim Guidance (Dyer 1999). If the screening decision is "Include", the FEP is considered either in the nominal scenario (i.e., the scenario that contains all expected FEPs and no disruptive FEPs); in the disruptive scenario (i.e., any scenario that contains all expected FEPs and one or more disruptive FEPs); or in the human intrusion scenario. If the screening decision is "Exclude", the basis for the exclusion, such as low probability or low consequence, is detailed.

The 87 waste form FEPs assigned to the Waste Form Degradation PMR have been evaluated by subject-matter experts and discussed in three different AMRs. Fifty-seven of the waste form FEPs are discussed in *Miscellaneous Waste-Form FEPs* (CRWMS M&O 2000n). Fifteen are discussed in *Waste Form Colloid-Associated Concentrations Limits: Abstraction and Summary* (CRWMS M&O 2000m). Sixteen are discussed in *Clad Degradation—FEPs Screening Arguments* AMR (CRWMS M&O 2000s). One FEP (Mutation) appears in two AMRs (CRWMS M&O 2000n, CRWMS M&O 2000m).

The culmination of these investigations on degradation of the waste forms was the construction of the Waste Form Degradation Model to predict the dissolved or colloidal radionuclides available for transport in the TSPA analysis for SR (TSPA-SR). The Waste Form Degradation Model consists of eight major modeling/analysis components: (1) Radioisotope Inventory, (2) In-Package Chemistry, (3) Commercial Spent Nuclear Fuel (CSNF) Degradation, (4) CSNF Cladding Degradation, (5) DOE Spent Nuclear Fuel (DSNF) Degradation, (6) High-Level Waste (HLW) Degradation, (7) Radioisotope Dissolved Concentration (solubility), and (8) Radioisotope Colloidal Concentration. These eight components are generally connected sequentially, starting with the radioisotope inventory as input and ending with projected radioisotope dissolved and colloidal concentrations. The summary and conclusions reached in each of the eight components are given below.

Radioisotope Inventory Component–The function of the Radioisotope Inventory Component is to estimate the inventory of those radionuclides most important to human dose. This component uses updated values for the inventory based on internal documents for CSNF, reports from the National Spent Nuclear Fuel Program (NSNFP) for DSNF, and the Environmental Impact Statement (EIS) for HLW. An updated screening was conducted, which resulted in 27 radionuclides being identified as important (24 based on human dose and 3 mandated by the groundwater protection requirements of the EPA's proposed standard 40 CFR 197 (64 FR 46976). The relative importance of individual radionuclides for human inhalation and ingestion doses was evaluated for several waste types, time frames, and release scenarios. In this evaluation, the effects of inventory abundance, radionuclide longevity, element solubility, and element transport affinity were considered.

In-Package Chemistry Component–The function of the In-package Chemistry Model Component is to estimate the fluid chemistry inside the waste package over time after the initial breach of the waste package. The water chemistry parameters in the model include hydronium ion concentration, dissolved partial pressures of carbon dioxide and oxygen, ionic strength, and fluoride and chloride ion concentrations. This chemistry is used by the several other model components (see Figure 1.5-3) because the rate of degradation of the matrix of waste, the resulting dissolved concentration of radionuclides, the stability of any colloids, and degradation of cladding are all dependent on the chemistry of fluids within the WP. The in-package chemistry component in the TSPA-SR couples the seepage rate of water into the package with the degradation of the steel, aluminum, SNF, and HLW inside the package to evaluate the key water chemistry parameters. These results influence degradation of the CSNF cladding and matrix, HLW degradation, radionuclide solubility, and colloid availability and stability. The degradation of the CSNF cladding and matrix, and HLW, in turn, influence the in-package chemistry component.

CSNF Degradation Component–The main function of the CSNF Degradation Model Component is to determine the rate of degradation of the CSNF matrix as a function of temperature and water chemistry (specifically, pH and partial pressures of O_2 and CO_2). This degradation rate is then directly used by the Cladding Degradation Model Component (Section 3.4) to determine the rate at which the CSNF cladding splits open and exposes more of the fuel matrix. In addition, the CSNF degradation model examines the distribution of radionuclides within the fuel and establishes a gap fraction for the more volatile radionuclides.

Under oxidizing conditions, in the presence of water or water vapor, UO_2 in CSNF is not stable and alters. Alteration of the UO_2 matrix can liberate radionuclides important to human dose. The CSNF matrix degradation component uses two linear regression equations, based on pH, the partial pressures of O_2 and CO_2, and temperature (T), to evaluate matrix degradation over time; one equation is applicable for pH less than or equal to 7 and the other is applicable for pH greater than 7. The regression variables used (pH, P_{CO2}, and P_{O2}) are coupled to in-package chemistry to account for uncertainty.

The CSNF matrix degradation component is based on experimental measurement of the dissolution rate of a number of fresh and spent fuels. These materials were examined under both saturated and unsaturated conditions using flow-through reactors as well as static, batch, and drip reactors. These tests also give empirical information on secondary phase production, colloids,

and effective solubility limits for the more insoluble radionuclides. The secondary phases are similar to those observed in the corrosion of natural uraninite deposits, indicating that the experimental results are indeed relevant to the long-term behavior.

CSNF Cladding Degradation Component–This component evaluates degradation of the cladding and is coupled to the CSNF Matrix Degradation and In-Package Chemistry components in the TSPA-SR. The degradation of CSNF cladding is assumed to proceed through two distinct steps: cladding failure (usually by small perforations), and progressive exposure of the fuel matrix.

Initial breach of the cladding may occur because (1) the cladding fails within the reactor or during storage or transportation, (2) an earthquake severely shakes and severs the rods, (3) the cladding fails from localized corrosion (Figure 3.4-2), or (4) the cladding fails from creep or stress corrosion cracking. While other mechanisms of initiating cladding perforations, such as rock fall or DHC, were explored, they were screened out based on low probability.

After the cladding has initially been breached, it is assumed to unzip because the fuel volume increases during alteration of the UO_2, (i.e., the alteration of the UO_2 to secondary phases of uranium is assumed to cause enough pressure due to volume expansion to burst the cladding from within) (Figure 3.4-2). This mechanism for increasing the exposed surface area of the matrix is sensitive to environmental conditions such as temperature. The anticipated temperature of the fuel matrix is not high enough, nor does it occur for a long enough period, to cause unzipping in a dry environment, so the dry unzipping is not included. Once the Zircaloy cladding has been perforated, unzipping in a wet environment does not occur in time periods of 40 years, based on observed behavior within storage pools at reactors. However, because unzipping in a wet environment could not be entirely ruled out, and because complete exposure of the matrix bounds diffusive releases of radionuclides through the perforation, it is conservatively assumed that clad unzipping in a wet environment can occur. The rate of wet unzipping was conservatively estimated by relating it to the movement of the alteration front.

DSNF Degradation Model Component–This component determines the rate of degradation of the DSNF waste category and of the immobilized plutonium ceramic waste. The degradation behavior of DSNF (excluding the naval SNF) will be represented by the metallic uranium SNF from N-Reactor. The naval SNF degradation behavior will be represented by that of the CSNF. TSPA-SR will use a degradation rate and a corresponding effective surface area that bounds the experimental data on N-Reactor fuel collected over a range of conditions in the last few years. In the TSPA-SR, the DSNF degradation component uses a constant, bounding degradation rate of N-Reactor SNF to bound all the DSNF waste types except naval SNF. However, the radioisotope inventory of the DSNF components is the weighted mass average of all waste types.

HLW Degradation Component–This component provides a conservative model for calculating the rate of degradation of borosilicate glass for the range of conditions (immersion, humid air, and dripping water) to which it may be exposed after the waste packages fail. For TSPA-SR, this component uses bounds on parameters of a phenomenological model to develop a simplified (Arrhenius-type) rate equation of degradation that is dependent only upon pH and temperature.

The model parameters account for the pH, temperature, surface area, and the combined effects of glass composition and solution composition on the rate of glass corrosion. Conservative estimates of the values for the model parameters are provided based on experimental data. Consistent with the conceptual model for in-package chemistry, degradation of borosilicate glass is conservatively assumed to occur as if the glass were fully immersed, although it is expected that much of the glass will be exposed to humid air or dripping water conditions.

Dissolved Radioisotope Concentration Component–This component evaluates the dissolved concentration of radionuclides (or parents of radionuclides) that are important to human dose, as determined by the radioisotope screening described in the inventory section (Section 3.1). In the dissolved radioisotope concentration component, the solubilities of important radionuclides were reevaluated: three radioisotope solubilities were abstracted as a direct function of in-package chemistry (and three more radionuclides equated to one of these); four radioisotope solubilities were defined by a probability distribution; and all others were set at bounding values based on results of the process modeling of the in-package chemistry.

The dissolved concentration limits calculation builds upon three primary feeds: (1) estimates of in-package fluid major element composition (pH, Eh, ionic strength, carbonate levels), (2) measured (and estimated) thermodynamic parameters describing the stabilities of aqueous species and solid radioisotope phases, and (3) a determination of the likely solubility-controlling phases for the specific radionuclides of concern. In nature, the controlling phase may either be a pure radionuclide solid phase with the radioisotope as the dominant element, or a solid phase with trace amounts of the radionuclide as can occur with coprecipitation. For TSPA-SR, the pure phases were chosen because, in general, they yield higher dissolved concentrations. Where insufficient information can be gleaned from field observations or experimental results, it is conservatively assumed that the most amorphous and hydrated (hence, most soluble) oxide of the particular radioelement forms.

A systematic review of thermodynamic data and controlling phases has been performed for a large range of chemical conditions. When uncertainties were encountered, choices were made that would result in higher predicted solubilities. Inherent limitations within the databases can lead to an uncertainty, up to a factor of 2, when the ionic strength exceeds about 0.7 mol/L. However, this uncertainty is small relative to other uncertainties within the system.

Colloidal Radioisotope Concentration Component–The function of this component is to calculate the concentration of colloid-associated radionuclides that may be transported from the waste package. A conceptualization was developed that directly used experimental results. The conceptualization identified the availability and the stability of three categories of colloids: (1) existing colloids in the groundwater, (2) colloids generated during degradation of the waste, and (3) colloids generated during degradation of the disposal container.

Colloid transport is potentially important for radionuclide elements that have low solubility and can be entrained in, or sorbed onto, waste form, engineered barrier, or geologic barrier materials that form colloidal particle substrates. Of these radionuclides, only those that are a major part of the waste inventory and have potentially large dose conversion factors are of potential importance to the performance of the disposal system. Considering these screening criteria, plutonium is the dominant radionuclide. Americium is also considered for the TSPA-SR.

Radionuclides are attached to colloids as a result of two types of processes: irreversible and reversible attachment. For waste form colloids, radionuclides can be incorporated into the substrate material before it is suspended as colloids. All colloid types may sorb radionuclides to form pseudocolloids, depending on the affinity of the colloid mineral substrate for a dissolved radionuclide. The contributions of each colloid type are summed to produce the mobile colloid source term for each important radionuclide.

The abstractions are based on laboratory results from waste form corrosion testing and testing of adsorption and desorption properties of Pu and Am on clay and iron-(hydr)oxide colloids. To the extent that the laboratory tests and test conditions represent anticipated repository conditions, the abstraction is valid for calculating the colloid-associated radionuclide concentrations and colloid mass concentrations.

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6. REFERENCES

6.1 DOCUMENTS CITED

Barnard, R.W. and Dockery, H.A., eds. 1991. *"Nominal Configuration" Hydrogeologic Parameters and Calculational Results.* Volume 1 of *Technical Summary of the Performance Assessment Calculational Exercises for 1990 (PACE-90).* SAND90-2726. Albuquerque, New Mexico: Sandia National Laboratories. ACC: NNA.19910523.0001.

Barnard, R.W.; Wilson, M.L.; Dockery, H.A.; Gauthier, J.H.; Kaplan, P.G.; Eaton, R.R.; Bingham, F.W.; and Robey, T.H. 1992. *TSPA 1991: An Initial Total-System Performance Assessment for Yucca Mountain.* SAND91-2795. Albuquerque, New Mexico: Sandia National Laboratories. ACC: NNA.19920630.0033.

Budnitz, B.; Ewing, R.C.; Moeller, D.W.; Payer, J.; Whipple, C.; and Witherspoon, P.A. 1999. *Peer Review of the Total System Performance Assessment-Viability Assessment Final Report.* Las Vegas, Nevada: Total System Performance Assessment Peer Review Panel. ACC: MOL.19990317.0328.

Center for Nuclear Waste Regulatory Analyses 1998. *Total-System Performance Assessment (TPA) Version 3.2 Code: Module Descriptions and User's Guide.* Draft. San Antonio, Texas: Center for Nuclear Waste Regulatory Analyses. TIC: 248267.

Chung, H.M.; Yaggee, F.L.; and Kassner, T.F. 1987. "Fracture Behavior and Microstructural Characteristics of Irradiated Zircaloy Cladding." *Zirconium in the Nuclear Industry, Seventh International Symposium Sponsored by ASTM Committee B-10 on Reactive and Refractory Metals, Strasbourg, France, 24-27 June 1985.* Adamson, R.B. and Van Swam, L.F.P., eds. Pages 775-801. Philadelphia, Pennsylvania: American Society for Testing and Materials. TIC: 238255.

CRWMS M&O 1994. *Total System Performance Assessment - 1993: An Evaluation of the Potential Yucca Mountain Repository.* B00000000-01717-2200-00099 REV 01. Las Vegas, Nevada: CRWMS M&O. ACC: NNA.19940406.0158.

CRWMS M&O 1995. *Total System Performance Assessment - 1995: An Evaluation of the Potential Yucca Mountain Repository.* B00000000-01717-2200-00136 REV 01. Las Vegas, Nevada: CRWMS M&O. ACC: MOL.19960724.0188.

CRWMS M&O 1996a. *Description of Performance Allocation.* B00000000-01717-2200-00177 REV 00. Las Vegas, Nevada: CRWMS M&O. ACC: MOL.19970116.0052.

CRWMS M&O 1996b. *Total System Performance Assessment of a Geologic Repository Containing Plutonium Waste Forms.* A00000000-01717-5705-00011 REV 00. Las Vegas, Nevada: CRWMS M&O. ACC: MOL.19970109.0229.

CRWMS M&O 1997a. *Total System Performance Assessment Sensitivity Studies of U.S. Department of Energy Spent Nuclear Fuel.* A00000000-01717-5705-00017 REV 01. Las Vegas, Nevada: CRWMS M&O. ACC: MOL.19980618.0474.

CRWMS M&O 1997b. *Constraints on Solubility-Limited Neptunium Concentrations for Use in Performance Assessment Analyses.* B00000000-01717-2200-00191 REV 00. Las Vegas, Nevada: CRWMS M&O. ACC: MOL.19980213.0484.

CRWMS M&O 1998a. *Software Qualification Report (SQR) Addendum to Existing LLNL Document UCRL-MA-110662 PT IV: Implementation of a Solid-Centered Flow-Through Mode for EQ6 Version 7.2B.* CSCI: UCRL-MA-110662 V 7.2b. SCR: LSCR198. Las Vegas, Nevada: CRWMS M&O. ACC: MOL.19990920.0169.

CRWMS M&O 1998b. "Waste Form Degradation, Radionuclide Mobilization, and Transport Through the Engineered Barrier System." Chapter 6 of *Total System Performance Assessment-Viability Assessment (TSPA-VA) Analyses Technical Basis Document.* B00000000-01717-4301-00006 REV 01. Las Vegas, Nevada: CRWMS M&O. ACC: MOL.19981008.0006.

CRWMS M&O 1998c. *Waste Form Degradation and Radionuclide Mobilization Expert Elicitation Project.* Las Vegas, Nevada: CRWMS M&O. ACC: MOL.19980804.0099.

CRWMS M&O 1999a. Not used.

CRWMS M&O 1999b. Not used.

CRWMS M&O 1999c. *Addendum to: EQ6 Computer Program for Theoretical Manual, Users Guide, & Related Documentation.* Software Change Request (SCR) LSCR198. UCRL-MA-110662 PT IV. Las Vegas, Nevada: CRWMS M&O. ACC: MOL.19990305.0112.

CRWMS M&O 1999d. *1999 Design Basis Waste Input Report for Commercial Spent Nuclear Fuel.* B00000000-01717-5700-00041 REV 00. Washington, D.C.: CRWMS M&O. ACC: MOV.19991006.0003.

CRWMS M&O 1999e. *Draft Analysis/Modeling Report - Department of Energy Spent Fuel Performance Assessment.* Input Transmittal WP-WP-99408.T. Las Vegas, Nevada: CRWMS M&O. ACC: MOL.19991209.0105.

CRWMS M&O 1999f. *Total System Performance Assessment-Site Recommendation Methods and Assumptions.* TDR-MGR-MD-000001 REV 01 ICN 01. Las Vegas, Nevada: CRWMS M&O. ACC: MOL.19991104.0570.

CRWMS M&O 2000a. *Repository Safety Strategy: Plan to Prepare the Postclosure Safety Case to Support Yucca Mountain Site Recommendation and Licensing Considerations.* TDR-WIS-RL-000001 REV 03. Las Vegas, Nevada: CRWMS M&O. ACC: MOL.20000119.0189.

CRWMS M&O 2000b. *Waste Form Analysis & Models.* Activity Evaluation, June 13, 2000. Las Vegas, Nevada: CRWMS M&O. ACC: MOL.20000615.0387.

CRWMS M&O 2000c. *Waste Form Degradation Process Model Report.* Development Plan TDP-WIS-MD-000031 REV 00. Las Vegas, Nevada: CRWMS M&O. ACC: MOL.20000202.0180.

CRWMS M&O 2000d. *Waste Package Degradation Process Model Report.* TDR-WIS-MD-000002 REV 00. Las Vegas, Nevada: CRWMS M&O. ACC: MOL.20000328.0322.

CRWMS M&O 2000e. Not used.

CRWMS M&O 2000f. *Inventory Abstraction.* ANL-WIS-MD-000006 REV 00. Las Vegas, Nevada: CRWMS M&O. ACC: MOL.20000414.0643.

CRWMS M&O 2000g. *In-Package Chemistry Abstraction.* ANL-EBS-MD-000037 REV 00. Las Vegas, Nevada: CRWMS M&O. ACC: MOL.20000418.0818.

CRWMS M&O 2000h. *CSNF Waste Form Degradation: Summary Abstraction.* ANL-EBS-MD-000015 REV 00. Las Vegas, Nevada: CRWMS M&O. ACC: MOL.20000121.0161.

CRWMS M&O 2000i. *Clad Degradation - Summary and Abstraction.* ANL-WIS-MD-000007 REV 00. Las Vegas, Nevada: CRWMS M&O. ACC: MOL.20000602.0055.

CRWMS M&O 2000j. *DSNF and Other Waste Form Degradation Abstraction.* ANL-WIS-MD-000004 REV 00. Las Vegas, Nevada: CRWMS M&O. ACC: MOL.20000223.0502.

CRWMS M&O 2000k. *Defense High Level Waste Glass Degradation.* ANL-EBS-MD-000016 REV 00. Las Vegas, Nevada: CRWMS M&O. ACC: MOL.20000329.1183.

CRWMS M&O 2000l. *Summary of Dissolved Concentration Limits.* ANL-WIS-MD-000010 REV 00. Las Vegas, Nevada: CRWMS M&O. ACC: MOL.20000525.0372.

CRWMS M&O 2000m. *Waste Form Colloid-Associated Concentrations Limits: Abstraction and Summary.* ANL-WIS-MD-000012 REV 00. Las Vegas, Nevada: CRWMS M&O. ACC: MOL.20000525.0397.

CRWMS M&O 2000n. *Miscellaneous Waste-Form FEPs.* ANL-WIS-MD-000009 REV 00. Las Vegas, Nevada: CRWMS M&O. ACC: MOL.20000526.0339.

CRWMS M&O 2000o. *Summary of In-Package Chemistry for Waste Forms.* ANL-EBS-MD-000050 REV 00. Las Vegas, Nevada: CRWMS M&O. ACC: MOL.20000217.0217.

CRWMS M&O 2000p. *Initial Cladding Condition.* ANL-EBS-MD-000048 REV 00. Las Vegas, Nevada: CRWMS M&O. ACC: MOL.20000523.0150.

CRWMS M&O 2000q. Not used.

CRWMS M&O 2000r. *Hydride-Related Degradation of SNF Cladding Under Repository Conditions.* ANL-EBS-MD-000011 REV 00. Las Vegas, Nevada: CRWMS M&O. ACC: MOL.20000319.0048.

CRWMS M&O 2000s. *Clad Degradation - FEPs Screening Arguments.* ANL-WIS-MD-000008 REV 00. Las Vegas, Nevada: CRWMS M&O. ACC: MOL.20000525.0378.

CRWMS M&O 2000t. *Clad Degradation-Wet Unzipping.* ANL-EBS-MD-000014 REV 00. Las Vegas, Nevada: CRWMS M&O. ACC: MOL.20000502.0398.

CRWMS M&O 2000u. *Clad Degradation - Dry Unzipping.* ANL-EBS-MD-000013 REV 00. Las Vegas, Nevada: CRWMS M&O. ACC: MOL.20000503.0200.

CRWMS M&O 2000v. *Pure Phase Solubility Limits - LANL.* ANL-EBS-MD-000017 REV 00. Las Vegas, Nevada: CRWMS M&O. ACC: MOL.20000504.0311.

CRWMS M&O 2000w. *Secondary Uranium-Phase Paragenesis and Incorporation of Radionuclides into Secondary Phases.* ANL-EBS-MD-000019 REV 00. Las Vegas, Nevada: CRWMS M&O. ACC: MOL.20000414.0644.

CRWMS M&O 2000x. *Colloid-Associated Radionuclide Concentration Limits: ANL.* ANL-EBS-MD-000020 REV 00. Las Vegas, Nevada: CRWMS M&O. ACC: MOL.20000329.1187.

CRWMS M&O 2000y. *Engineered Barrier System Degradation, Flow, and Transport Process Model Report.* TDR-EBS-MD-000006 REV 00. Las Vegas, Nevada: CRWMS M&O. ACC: MOL.20000324.0558.

CRWMS M&O 2000z. *Near-Field Environment Process Model Report.* TDR-NBS-MD-000001 REV 00. Las Vegas, Nevada: CRWMS M&O. ACC: MOL.20000421.0034.

CRWMS M&O 2000aa. *Evaluation of Codisposal Viability for UZrH (TRIGA) DOE-Owned Fuel.* TDR-EDC-NU-000001 REV 00. Las Vegas, Nevada: CRWMS M&O. ACC: MOL.20000207.0689.

CRWMS M&O 2000ab. *Evaluation of Codisposal Viability for HEU Oxide (Shippingport PWR) DOE-Owned Fuel.* TDR-EDC-NU-000003 REV 00. Las Vegas, Nevada: CRWMS M&O. ACC: MOL.20000227.0240.

CRWMS M&O 2000ac. *Waste Package Related Impacts of Plutonium Disposition Waste Forms in a Geologic Repository.* TDR-EBS-MD-000003 REV 01. Las Vegas, Nevada: CRWMS M&O. ACC: MOL.20000202.0170.

CRWMS M&O 2000ad. *Near-Field Accumulation of Fissile Material from Waste Packages Containing Plutonium Disposition Waste Forms.* Input Transmittal WP-WP-00025.T. Las Vegas, Nevada: CRWMS M&O. ACC: MOL.20000125.0608.

CRWMS M&O 2000ae. *DOE and Commercial Waste Package Inventory: Various Input Scenarios for Design.* Input Transmittal WP-RSO-00023.Ta. Las Vegas, Nevada: CRWMS M&O. ACC: MOL.20000403.0095.

CRWMS M&O 2000af. *Drift-Scale Coupled Processes (DST and THC Seepage) Models.* MDL-NBS-HS-000001 REV 00. Las Vegas, Nevada: CRWMS M&O. ACC: MOL.19990721.0523.

CRWMS M&O 2000ag. *Clad Degradation - Local Corrosion of Zirconium and Its Alloys Under Repository Conditions.* ANL-EBS-MD-000012 REV 00. Las Vegas, Nevada: CRWMS M&O. ACC: MOL.20000405.0479.

CRWMS M&O 2000ah. *Rock Fall on Drip Shield.* CAL-EDS-ME-000001 REV 00. Las Vegas, Nevada: CRWMS M&O. ACC: MOL.20000509.0276.

CRWMS M&O 2000ai. *Abstraction of Drift-Scale Coupled Processes.* ANL-NBS-HS-000029 REV 00. Las Vegas, Nevada: CRWMS M&O. ACC: MOL.20000525.0371.

CRWMS M&O 2000aj. *Waste Package and DOE Canister Inventory.* Input Transmittal RSO-SSR-99360.T. Las Vegas, Nevada: CRWMS M&O. ACC: MOL.20000202.0002.

CRWMS M&O 2000ak. *Revised Grams Per Package for Use in TSPA-SR.* Input Transmittal 00306.T. Las Vegas, Nevada: CRWMS M&O. ACC: MOL.20000628.0043.

CRWMS M&O 2000al. *Waste Package Internal Materials Selection Report.* B00000000-01717-2200-00226 REV 00. Las Vegas, Nevada: CRWMS M&O. ACC: MOL.20000209.0299.

CRWMS M&O 2000am. *Performance Confirmation Plan.* TDR-PCS-SE-000001 REV 01 ICN 01. Las Vegas, Nevada: CRWMS M&O. ACC: MOL.20000601.0196.

CRWMS M&O 2000an. *Disruptive Events Process Model Report.* TDR-NBS-MD-000002 REV 00. Las Vegas, Nevada: CRWMS M&O. ACC: MOL.20000504.0295.

CRWMS M&O 2000ao. *Waste Package Design Sensitivity Report.* TDR-EBS-MD-000008 REV 00. Las Vegas, Nevada: CRWMS M&O. ACC: MOL.20000518.0179.

Cunnane, J.C.; Bates, J.K.; Bradley, C.R.; Buck, E.C.; Ebert, W.L.; Feng, X.; Mazer, J.J.; Wronkiewicz, D.J.; Sproull, J.; Bourcier, W.L.; McGrail, B.P.; and Altenhofen, M.K., eds. 1994. *High-Level Waste Borosilicate Glass: A Compendium of Corrosion Characteristics.* DOE-EM-0177. Volume 1. Washington, D.C.: U.S. Department of Energy. TIC: 242675.

de Pablo, J.; Casas, I.; Gimenez, J.; Molera, M.; and Torrero, M.D. 1997. "Effect of Temperature and Bicarbonate Concentration on the Kinetics of UO2(s) Dissolution Under Oxidizing Conditions." *Scientific Basis for Nuclear Waste Management XX, Symposium held December 2-6, 1996, Boston, Massachusetts.* Gray, W.J. and Triay, I.R., eds. *465,* 535-542. Pittsburgh, Pennsylvania: Materials Research Society. TIC: 238884.

Dirkmaat, P.J. 1997. "Response to Repository Environmental Impact Statement (EIS) Data Call (OPE-SFP-97-230)." Memorandum from P.J. Dirkmaat (DOE) to K.J. Skipper, July 10, 1997, with attachment. ACC: MOL.19970725.0067; MOL.19970725.0068.

DOE (U.S. Department of Energy) 1988. *Site Characterization Plan Yucca Mountain Site, Nevada Research and Development Area, Nevada.* DOE/RW-0199. Nine volumes. Washington, D.C.: U.S. Department of Energy, Office of Civilian Radioactive Waste Management. ACC: HQO.19881201.0002.

DOE (U.S. Department of Energy) 1996. *Title 40 CFR Part 191 Compliance Certification Application for the Waste Isolation Pilot Plant.* DOE/CAO-1996-2184. Carlsbad, New Mexico: U.S. Department of Energy, Carlsbad Area Office. TIC: 240511.

DOE (U.S. Department of Energy) 1998a. *Update to Assessment of Direct Disposal in Unsaturated Tuff of Spent Nuclear Fuel and High-Level Waste Owned by U.S. Department of Energy.* DOE/SNF/REP-015. Washington, D.C.: U.S. Department of Energy. TIC: 243859.

DOE (U.S. Department of Energy) 1998b. *Viability Assessment of a Repository at Yucca Mountain.* DOE/RW-0508. Washington, D.C.: U.S. Department of Energy, Office of Civilian Radioactive Waste Management. ACC: MOL.19981007.0027; MOL.19981007.0028; MOL.19981007.0029; MOL.19981007.0030; MOL.19981007.0031; MOL.19981007.0032.

DOE (U.S. Department of Energy) 1998c. *Total System Performance Assessment.* Volume 3 of *Viability Assessment of a Repository at Yucca Mountain.* DOE/RW-0508. Washington, D.C.: U.S. Department of Energy, Office of Civilian Radioactive Waste Management. ACC: MOL.19981007.0030.

DOE (U.S. Department of Energy) 1999a. *Draft Environmental Impact Statement for a Geologic Repository for the Disposal of Spent Nuclear Fuel and High-Level Radioactive Waste at Yucca Mountain, Nye County, Nevada.* DOE/EIS-0250D. Summary, Volumes I and II. Washington, D.C.: U.S. Department of Energy, Office of Civilian Radioactive Waste Management. ACC: MOL.19990816.0240.

DOE (U.S. Department of Energy) 1999b. *DOE Spent Nuclear Fuel Information in Support of TSPA-SR.* DOE/SNF/REP-0047, Rev. 0. Washington, D.C.: U.S. Department of Energy. TIC: 245482.

DOE (U.S. Department of Energy) 1999c. *Waste Acceptance System Requirements Document.* DOE/RW-0351, Rev. 03. Washington, D.C.: U.S. Department of Energy, Office of Civilian Radioactive Waste Management. ACC: HQO.19990226.0001.

DOE (U.S. Department of Energy) 2000. *Quality Assurance Requirements and Description.* DOE/RW-0333P, Rev. 10. Washington, D.C.: U.S. Department of Energy, Office of Civilian Radioactive Waste Management. ACC: MOL.20000427.0422.

Dyer, J.R. 1999. "Revised Interim Guidance Pending Issuance of New U.S. Nuclear Regulatory Commission (NRC) Regulations (Revision 01, July 22, 1999), for Yucca Mountain, Nevada." Letter from J.R. Dyer (DOE/YMSCO) to D.R. Wilkins (CRWMS M&O), September 3, 1999, OL&RC:SB-1714, with enclosure, "Interim Guidance Pending Issuance of New NRC Regulations for Yucca Mountain (Revision 01)." ACC: MOL.19990910.0079.

Ebert, W.L. 1995. *The Effects of the Glass Surface Area/Solution Volume Ratio on Glass Corrosion: A Critical Review.* ANL-94/34. Argonne, Illinois: Argonne National Laboratory. TIC: 215400.

Efurd, D.W.; Runde, W.; Banar, J.C.; Janecky, D.R.; Kaszuba, J.P.; Palmer, P.D.; Roensch, F.R.; and Tait, D. 1998. "Neptunium and Plutonium Solubilities in a Yucca Mountain Groundwater." *Environmental Science & Technology, 32,* (24), 3893-3900. Easton, Pennsylvania: Environmental Science & Technology. TIC: 243857.

Energy and Water Development Appropriations Act, 1997. Public Law No. 104-206. 110 Stat. 2984. Readily Available

Eslinger, P.W.; Doremus, L.A.; Engel, D.W.; Miley, T.B.; Murphy, M.T.; Nichols, W.E.; White, M.D.; Langford, D.W.; and Ouderkirk, S.J. 1993. *Preliminary Total-System Analysis of a Potential High-Level Nuclear Waste Repository at Yucca Mountain.* PNL-8444. Richland, Washington: Pacific Northwest Laboratory. ACC: HQO.19930219.0001.

Finn, P.A.; Finch, R.; Buck, E.; and Bates, J. 1998. "Corrosion Mechanism of Spent Fuel Under Oxidizing Conditions." *Scientific Basis for Nuclear Waste Management XXI, Symposium held September 28-October 3, 1997, Davos, Switzerland.* McKinley, I.G. and McCombie, C., eds. *506,* 123-131. Warrendale, Pennsylvania: Materials Research Society. TIC: 240702.

Fitzgerald, B.J. and Yau, T. 1993. "The Mechanism and Control of Stress Corrosion Cracking of Zirconium in Sulfuric Acid." *Corrosion Control for Low-Cost Reliability, 12th International Corrosion Congress, Houston, Texas, September 19-24, 1993. 2,* 762-777. Houston, Texas: National Association of Corrosion Engineers. TIC: 247512.

Grambow, B. 1989. *Spent Fuel Dissolution and Oxidation: An Evaluation of Literature Data.* SKB Technical Report 89-13. Stockholm, Sweden: Swedish Nuclear Fuel and Waste Management Company. TIC: 208579.

Grambow, B.; Jercinovic, M.J.; Ewing, R.C.; and Byers, C.D. 1986. "Weathered Basalt Glass: A Natural Analogue for the Effects of Reaction Progress on Nuclear Waste Glass Alteration." *Scientific Basis for Nuclear Waste Management IX, Symposium held September 9-11, 1985 in Stockholm, Sweden.* Werme, L.O., ed. *50,* 263-273. Pittsburgh, Pennsylvania: Materials Research Society. TIC: 206807.

Grandstaff, D.E. 1976. "A Kinetic Study of the Dissolution of Uraninite." *Economic Geology and the Bulletin of The Society of Economic Geologists, 71,* (8), 1493-1506. El Paso, Texas: Economic Geology Publishing Company. TIC: 246339.

Gray, W.J. and Einziger, R.E. 1998. *Initial Results from Dissolution Rate Testing of N-Reactor Spent Fuel Over a Range of Potential Geologic Repository Aqueous Conditions.* DOE/SNF/REP-022, Rev. 0. Washington, D.C.: U.S. Department of Energy. TIC: 245380.

Harrar, J.E.; Carley, J.F.; Isherwood, W.F.; and Raber, E. 1990. *Report of the Committee to Review the Use of J-13 Well Water in Nevada Nuclear Waste Storage Investigations.* UCID-21867. Livermore, California: Lawrence Livermore National Laboratory. ACC: NNA.19910131.0274.

Haschke, J.M.; Allen, T.H.; and Morales L.A. 2000. "Reaction of Plutonium Dioxide with Water: Formation and Properties of PuO2+x." *Science, 287,* ([5451]), 285-287. [Washington, D.C: American Association for the Advancement of Science]. TIC: 248119.

Henningson, P.J. 1998. *Cladding Integrity Under Long Term Disposal.* 51-1267509-00. Lynchburg, Virginia: Framatome Technologies. ACC: MOL.19990310.0103.

House, W.A.; Denison, F.H.; Warwick, M.S.; and Zhmud, B.V. 2000. "Dissolution of Silica and the Development of Concentration Profiles in Freshwater Sediments." *Applied Geochemistry, 15,* (4), 425-438. New York, New York: Elsevier Science. TIC: 247719.

Kaszuba J.P. and Runde W.H. 1999. "The Aqueous Geochemistry of Neptunium: Dynamic Control of Soluble Concentrations with Applications to Nuclear Waste Disposal." *Environmental Science & Technology, 33,* (24), 4427-4433. Washington, D.C.: American Chemical Society. TIC: 246667.

Kersting, A.B.; Efurd, D.W.; Finnegan, D.L.; Rokop, D.J.; Smith, D.K.; and Thompson, J.L. 1999. "Migration of Plutonium in Ground Water at the Nevada Test Site." *Nature, 397,* 56-59. London, England: Macmillan Publishers. TIC: 243597.

Kingston, W.L. and Whitbeck, M. 1991. *Characterization of Colloids Found in Various Groundwater Environments in Central and Southern Nevada.* DOE/NV/10384-36. Las Vegas, Nevada: U.S. Department of Energy. ACC: NNA.19930607.0073.

Kotra, J.P.; Lee, M.P.; Eisenberg, N.A.; and DeWispelare, A.R. 1996. *Branch Technical Position on the Use of Expert Elicitation in the High-Level Radioactive Waste Program.* NUREG-1563. Washington, D.C.: U.S. Nuclear Regulatory Commission. TIC: 226832.

Liang, L. and Morgan, J.J. 1990. "Chemical Aspects of Iron Oxide Coagulation in Water: Laboratory Studies and Implications for Natural Systems." *Aquatic Sciences, 32,* (1), 32-55. Basel, Switzerland: Birkhauser Verlag. TIC: 246125.

Lu, N.; Triay, I.R.; Cotter, C.R.; Kitten, H.D.; and Bentley, J. 1998. *Reversibility of Sorption of Plutonium-239 Onto Colloids of Hematite, Goethite, Smectite, and Silica.* LA-UR-98-3057. Los Alamos, New Mexico: Los Alamos National Laboratory. ACC: MOL.19981030.0202.

Matsuo, Y. 1987. "Thermal Creep of Zircaloy-4 Cladding Under Internal Pressure." *Journal of Nuclear Science and Technology, 24,* (2), 111-119. Tokyo, Japan: Atomic Energy Society of Japan. TIC: 237137.

McCarthy, J.F. and Degueldre, C. 1993. "Sampling and Characterization of Colloids and Particles in Groundwater for Studying Their Role in Contaminant Transport." Chapter 6 of *Environmental Particles.* Buffle, J. and van Leeuwen, H.P., eds. Volume 2. Boca Raton, Florida: Lewis Publishers. TIC: 245905.

McCarthy, J.F. and Zachara, J.M. 1989. "Subsurface Transport of Contaminants." *Environmental Science & Technology, 23,* (5), 496-502. Easton, Pennsylvania: American Chemical Society. TIC: 224876.

McCright, R.D. 1998. *Corrosion Data and Modeling Update for Viability Assessment.* Volume 3 of *Engineered Materials Characterization Report.* UCRL-ID-119564, Rev. 1.1. Livermore, California: Lawrence Livermore National Laboratory. ACC: MOL.19981222.0137.

McKenzie, W.F. 1992. *UO2 Dissolution Rates: A Review.* UCRL-ID-111663. Livermore, California: Lawrence Livermore National Laboratory. TIC: 246408.

Minai, Y.; Choppin, G.R.; and Sisson, D.H. 1992. "Humic Material in Well Water from the Nevada Test Site." *Radiochimica Acta, 56,* 195-199. Munchen, Germany: R. Oldenbourg Verlag. TIC: 238763.

Murphy, W.M. 1995. "Natural Analogs for Yucca Mountain." *Radwaste Magazine, 2,* (6), 44-50. La Grange Park, Illinois: American Nuclear Society. TIC: 237929.

Nitsche, H.; Gatti, R.C.; Standifer, E.M.; Lee, S.C.; Muller, A.; Prussin, T.; Deinhammer, R.S.; Maurer, H.; Becraft, K.; Leung, S.; and Carpenter, S.A. 1993. *Measured Solubilities and Speciations of Neptunium, Plutonium, and Americium in Typical Groundwater (J-13) from the Yucca Mountain Region.* LA-12562-MS. Los Alamos, New Mexico: Los Alamos National Laboratory. ACC: MOL.19950621.0265.

NRC (U.S. Nuclear Regulatory Commission) 1999a. *Issue Resolution Status Report Key Technical Issue: Container Life and Source Term.* Rev. 2. Washington, D.C.: U.S. Nuclear Regulatory Commission. TIC: 245538.

NRC (U.S. Nuclear Regulatory Commission) 1999b. *Issue Resolution Status Report Key Technical Issue: Evolution of the Near-Field Environment.* Rev. 2. Washington, D.C.: U.S. Nuclear Regulatory Commission. ACC: MOL.19990810.0640.

NRC (U.S. Nuclear Regulatory Commission) 2000a. *Issue Resolution Status Report Key Technical Issue: Total System Performance Assessment and Integration.* Rev. 2. Washington, D.C.: U.S. Nuclear Regulatory Commission. TIC: 247614.

NRC (U.S. Nuclear Regulatory Commission) 2000b. *Interim Staff Guidance-11 — Storage of High Burnup Spent Fuel.* Washington, D.C.: U.S. Nuclear Regulatory Commission. TIC: 247227.

Nuclear Waste Policy Act of 1982. 42 U.S.C. 10101 et seq. Readily Available

NWTRB (Nuclear Waste Technical Review Board) 1998. *Report to the U.S. Congress and the U.S. Secretary of Energy, November, 1998.* Arlington, Virginia: U.S. Nuclear Waste Technical Review Board. ACC: MOV.19981201.0015.

Pearcy, E.C.; Prikryl, J.D.; Murphy, W.M.; and Leslie, B.W. 1994. "Alteration of Uraninite from the Nopal I Deposit, Pena Blanca District, Chihuahua, Mexico, Compared to Degradation of Spent Nuclear Fuel in the Proposed U.S. High-Level Nuclear Waste Repository at Yucca Mountain, Nevada." *Applied Geochemistry, 9,* 713-732. New York, New York: Elsevier. TIC: 236934.

Peehs, M. 1998. *Assessment of Dry Storage Performance of Spent LWR Fuel Assemblies with Increasing Burn-Up.* Erlangen, Germany: Bereich Energieerzeugung. TIC: 245171.

Perez, J.M., Jr. and Westsik, J.H., Jr. 1981. "Effects of Cracks on Glass Leaching." *Nuclear and Chemical Waste Management, 2,* 165-168. New York, New York: Pergamon Press. TIC: 246072.

Pescatore, C. and Cowgill, M. 1994. *Temperature Limit Determination for the Inert Dry Storage of Spent Nuclear Fuel.* EPRI TR-103949. Palo Alto, California: Electric Power Research Institute. TIC: 102933.

Prindle, R.W. and Hopkins, P.L. 1990. *On Conditions and Parameters Important to Model Sensitivity for Unsaturated Flow Through Layered, Fractured Tuff: Results of Analyses for HYDROCOIN Level 3 Case 2.* SAND89-0652. Albuquerque, New Mexico: Sandia National Laboratories. ACC: NNA.19900523.0211.

Rai, D.; Felmy, A.R.; Moore, D.A.; and Mason, M.J. 1995. "The Solubility of Th(IV) and U(IV) Hydrous Oxides in Concentrated NaHCO3 and Na2CO3 Solutions." *Scientific Basis for Nuclear Waste Management XVIII, Symposium held October 23-27, 1994, Kyoto, Japan.* Murakami, T. and Ewing, R.C., eds. *353,* 1143-1150. Pittsburgh, Pennsylvania: Materials Research Society. TIC: 216341.

Rechard, R.P. 1999. "Historical Relationship Between Performance Assessment for Radioactive Waste Disposal and Other Types of Risk Assessment." *Risk Analysis, 19,* (5), 763-807. New York, New York: Plenum Press. TIC: 246972.

Rechard, R.P., ed. 1993. *Initial Performance Assessment of the Disposal of Spent Nuclear Fuel and High Level Waste Stored at Idaho National Engineering Laboratory.* SAND93-2330. Volumes 1 and 2. Albuquerque, New Mexico: Sandia National Laboratories. TIC: 247212.

Rechard, R.P., ed. 1995. *Executive Summary.* Volume 1 of *Performance Assessment of the Direct Disposal in Unsaturated Tuff of Spent Nuclear Fuel and High-Level Waste Owned by U.S. Department of Energy.* SAND94-2563/1. Albuquerque, New Mexico: Sandia National Laboratories. TIC: 237101.

Shoesmith, D.W. 1999. *Fuel Corrosion Processes Under Waste Disposal Conditions.* AECL-12034. Pinawa, Manitoba, Canada: Whiteshell Laboratories. TIC: 246006.

Sinnock, S.; Lin, Y.T.; and Brannen, J.P. 1984. *Preliminary Bounds on the Expected Postclosure Performance of the Yucca Mountain Repository Site, Southern Nevada.* SAND84-1492. Albuquerque, New Mexico: Sandia National Laboratories. ACC: NNA.19870519.0076.

Strachan, D.M.; Bourcier, W.L.; and McGrail, B.P. 1994. "Toward a Consistent Model for Glass Dissolution." *Radioactive Waste Management and Environmental Restoration, 19,* (1-3), 129-145. Chur, Switzerland: Harwood. TIC: 247530.

Stroupe, E.P. 2000. "Approach to Implementing the Site Recommendation Design Baseline." Interoffice correspondence from E.P. Stroupe (CRWMS M&O) to D.R. Wilkins, January 26, 2000, LV.RSO.EPS.1/00-004, with attachment. ACC: MOL.20000214.0480.

Tait, J.C. and Luht, J.L. 1997. *Dissolution Rates of Uranium from Unirradiated UO2 and Uranium and Radionuclides from Used CANDU Fuel Using the Single-Pass Flow-Through Apparatus.* 06819-REP-01200-0006 R00. Toronto, Ontario, Canada: Ontario Hydro. TIC: 243164.

Tasooji, A.; Einziger, R.E.; and Miller, A.K. 1984. "Modeling of Zircaloy Stress-Corrosion Cracking: Texture Effects and Dry Storage Spent Fuel Behavior." *Zirconium in the Nuclear Industry, Sixth International Symposium, Vancouver, British Columbia, June 28- July 1, 1982.* Franklin, D.G. and Adamson, R.B., eds. *ASTM STP 824,* 595-626. Philadelphia, Pennsylvania: American Society for Testing and Materials. TIC: 241417.

Tombacz, E.; Abraham, I.; Gilde, M.; and Szanto, F. 1990. "The pH-Dependent Colloidal Stability of Aqueous Montmorillonite Suspensions." *Colloids and Surfaces, 49,* 71-80. Amsterdam, The Netherlands: Elsevier Science. TIC: 246046.

Torrero, M.E.; Baraj, E.; De Pablo, J.; Gimenez, J.; and Casas, I. 1997. "Kinetics of Corrosion and Dissolution of Uranium Dioxide as a Function of pH." *International Journal of Chemical Kinetics, 29,* (4), 261-267. New York, New York: John Wiley & Sons. TIC: 246160.

Wescott, R.G.; Lee, M.P.; Eisenberg, N.A.; McCartin, T.J.; and Baca, R.G., eds. 1995. *NRC Iterative Performance Assessment Phase 2: Development of Capabilities for Review of a Performance Assessment for a High-Level Waste Repository.* NUREG-1464. Washington, D.C.: U.S. Nuclear Regulatory Commission. TIC: 221527.

Wilson, C.N. 1990. *Results from NNWSI Series 3 Spent Fuel Dissolution Tests.* PNL-7170. Richland, Washington: Pacific Northwest Laboratory. ACC: NNA.19900329.0142.

Wilson, M.L.; Gauthier, J.H.; Barnard, R.W.; Barr, G.E.; Dockery, H.A.; Dunn, E.; Eaton, R.R.; Guerin, D.C.; Lu, N.; Martinez, M.J.; Nilson, R.; Rautman, C.A.; Robey, T.H.; Ross, B.; Ryder, E.E.; Schenker, A.R.; Shannon, S.A.; Skinner, L.H.; Halsey, W.G.; Gansemer, J.D.; Lewis, L.C.; Lamont, A.D.; Triay, I.R.; Meijer, A.; and Morris, D.E. 1994. *Total-System Performance Assessment for Yucca Mountain – SNL Second Iteration (TSPA-1993).* SAND93-2675. Executive Summary and two volumes. Albuquerque, New Mexico: Sandia National Laboratories. ACC: NNA.19940112.0123.

Yau, T-L. 1992. "Stress-Corrosion Cracking of Zirconium Alloys." Chapter 11 of *Stress-Corrosion Cracking.* Jones, R.H., ed. Materials Park, Ohio: ASM International. TIC: 246059.

YMP (Yucca Mountain Site Characterization Project) 1998. *Disposal Criticality Analysis Methodology Topical Report.* YMP/TR-004Q, Rev. 0. Las Vegas, Nevada: Yucca Mountain Site Characterization Office. ACC: MOL.19990210.0236.

6.2 CODES, STANDARDS, REGULATIONS, AND PROCEDURES

10 CFR 60. Energy: Disposal of High-Level Radioactive Wastes in Geologic Repositories. Readily Available

40 CFR 191. Protection of Environment: Environmental Radiation Protection Standards for Management and Disposal of Spent Nuclear Fuel, High-Level and Transuranic Radioactive Wastes. Readily Available

60 FR 42622. Use of Probabilistic Risk Assessment Methods in Nuclear Regulatory Activities: Final Policy Statement. Readily Available

64 FR 46976. 40 CFR 197: Environmental Radiation Protection Standards for Yucca Mountain, Nevada; Proposed Rule. Readily Available

AP-2.14Q, Rev.0, ICN 1. *Review of Technical Products.* Washington, D.C.: U.S. Department of Energy, Office of Civilian Radioactive Waste Management. ACC: MOL.20000405.0477.

AP-2.16Q, Rev. 0, ICN 0. *Activity Evaluation.* Washington, D.C.: U.S. Department of Energy, Office of Civilian Radioactive Waste Management. ACC: MOL.20000207.0716.

AP-3.10Q, Rev. 2, ICN 2. *Analyses and Models.* Washington, D.C.: U.S. Department of Energy, Office of Civilian Radioactive Waste Management. ACC: MOL.20000619.0576.

AP-3.11Q, Rev. 1, ICN 0. *Technical Reports.* Washington, D.C.: U.S. Department of Energy, Office of Civilian Radioactive Waste Management. ACC: MOL.20000516.0008.

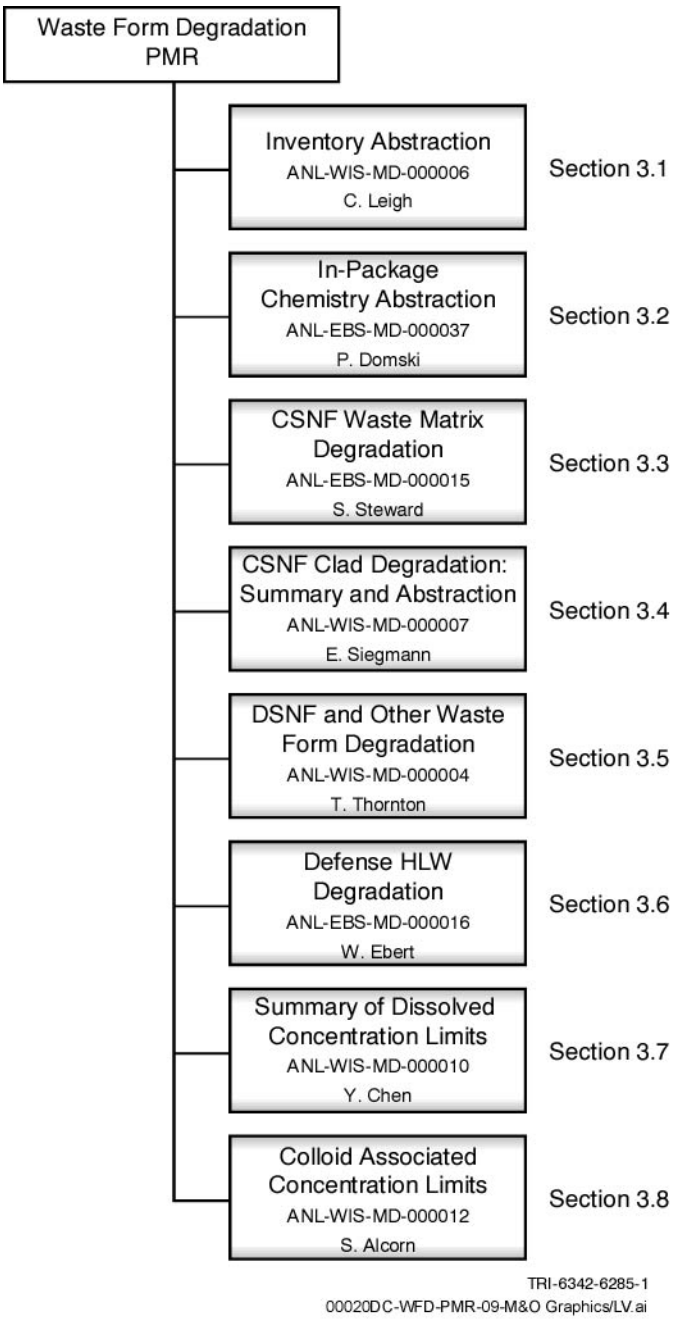
AP-3.15Q, Rev. 1, ICN 1. *Managing Technical Product Inputs.* Washington, D.C.: U.S. Department of Energy, Office of Civilian Radioactive Waste Management. ACC: MOL.20000218.0069.

AP-SI.1Q, Rev. 2, ICN 4. *Software Management.* Washington, D.C.: U.S. Department of Energy, Office of Civilian Radioactive Waste Management. ACC: MOL.20000223.0508.

AP-SIII.2Q, Rev. 0, ICN 2. *Qualification of Unqualified Data and the Documentation of Rationale for Accepted Data.* Washington, D.C.: U.S. Department of Energy, Office of Civilian Radioactive Waste Management. ACC: MOL.19991214.0625.

ASTM C 1174-97. 1997. *Standard Practice for Prediction of the Long-Term Behavior of Materials, Including Waste Forms, Used in Engineered Barrier Systems (EBS) for Geological Disposal of High-Level Radioactive Waste.* West Conshohocken, Pennsylvania: American Society for Testing and Materials. TIC: 246015.

ASTM C1285-97. 1999. *Standard Test Methods for Determining Chemical Durability of Nuclear, Hazardous, and Mixed Waste Glasses: The Product Consistency Test (PCT).* West Conshohocken, Pennsylvania: American Society for Testing and Materials. TIC: 247001.



NOTE: See CRWMS M&O 2000f through 2000m for AMR references for Sections 3.1 through 3.8 respectively.

Figure 1.4-1. Documents Specifically Supporting the Waste Form Degradation PMR

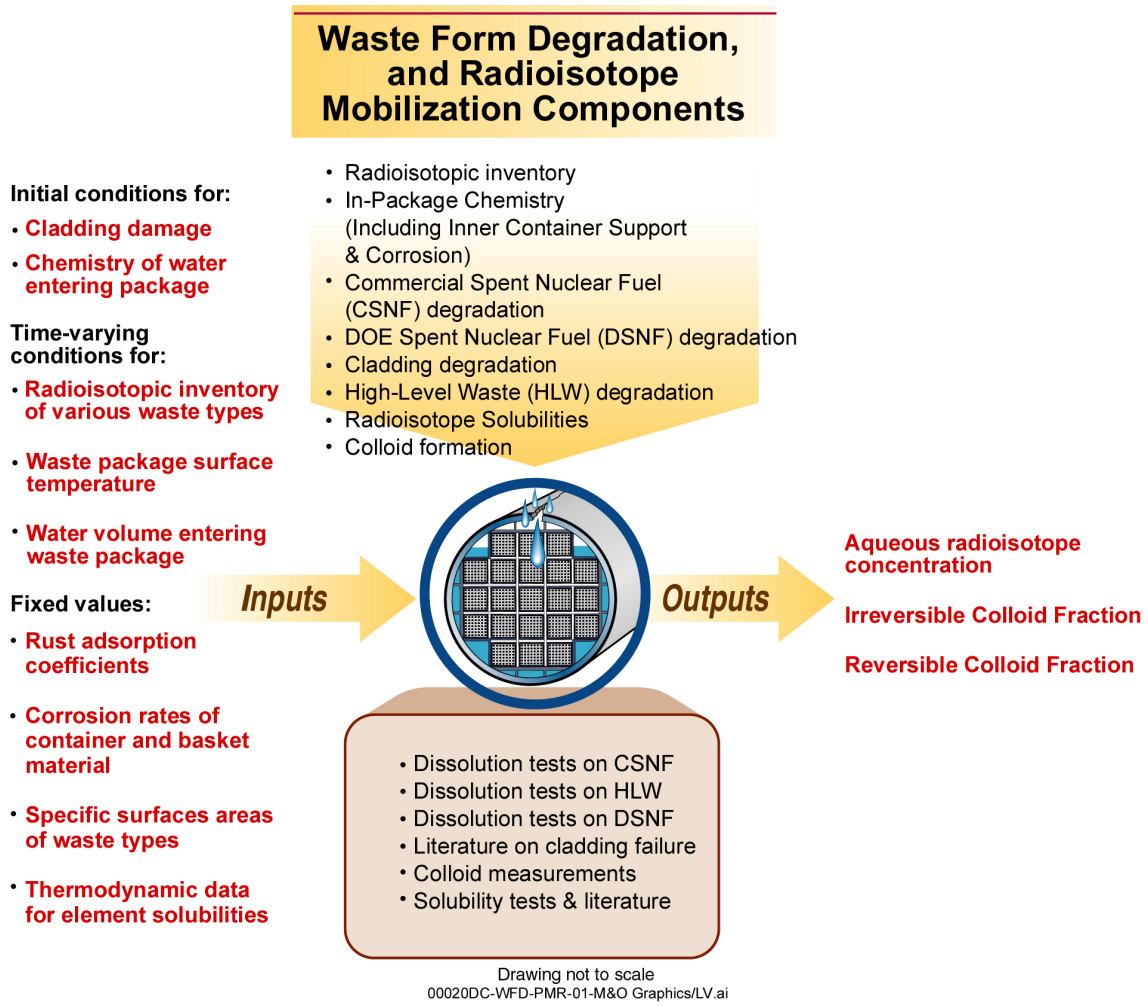


Figure 1.5-1. Summary of Inputs, Outputs, Components, and Assumptions of Waste Form Degradation Model

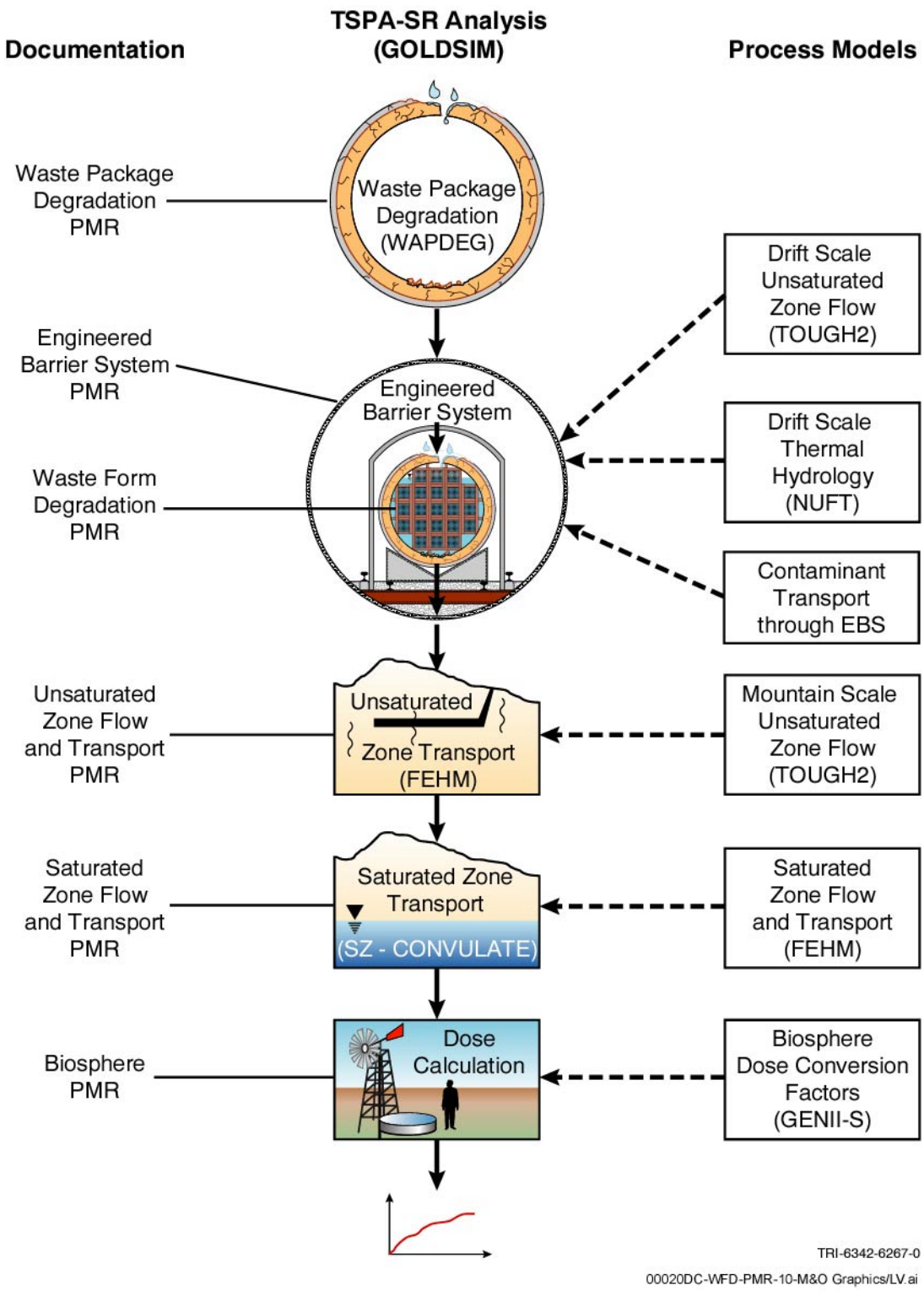


Figure 1.5-2. Major Models within TSPA-SR

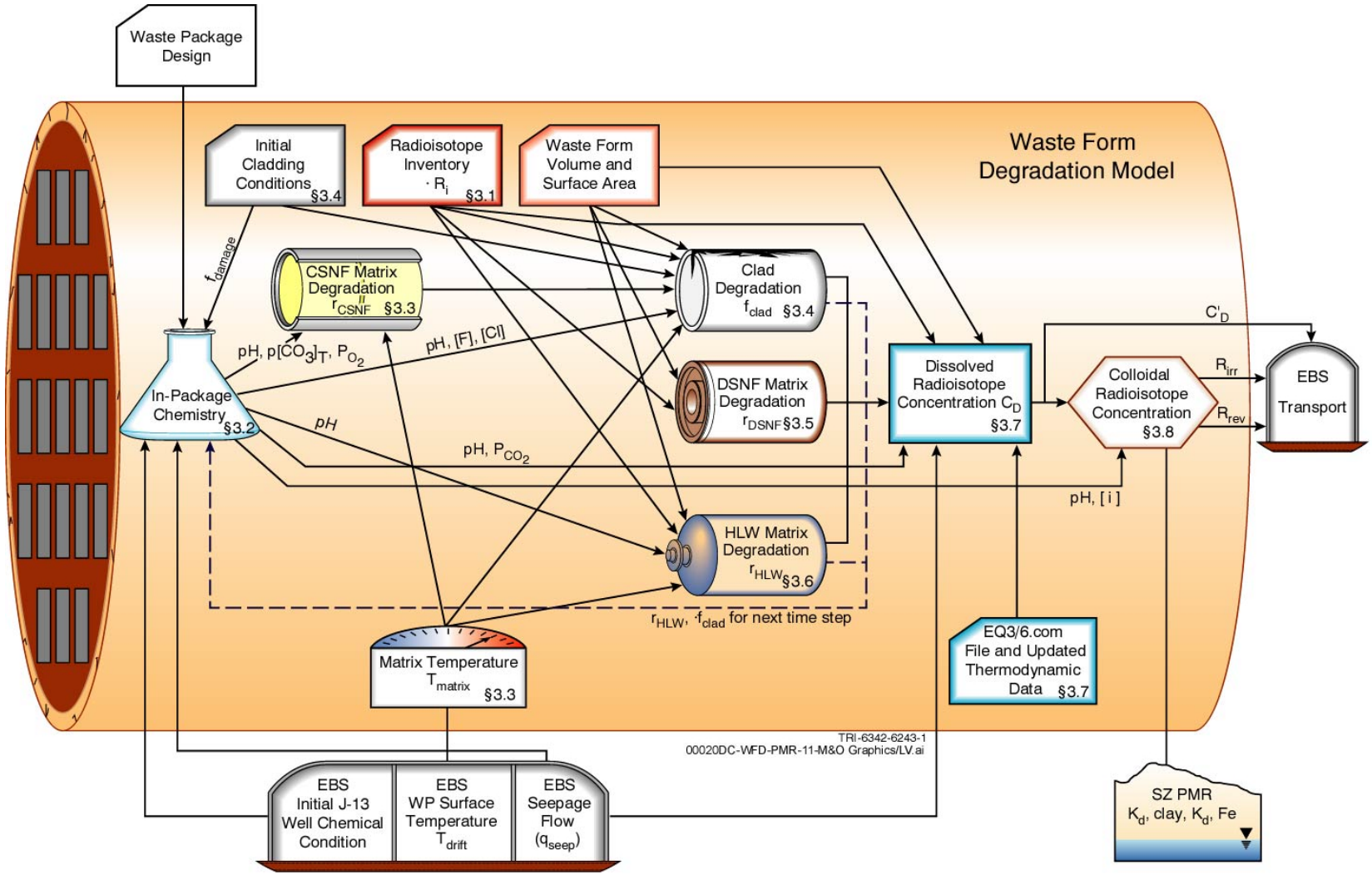


Figure 1.5-3. Linkage of Components in Waste Form Degradation Model

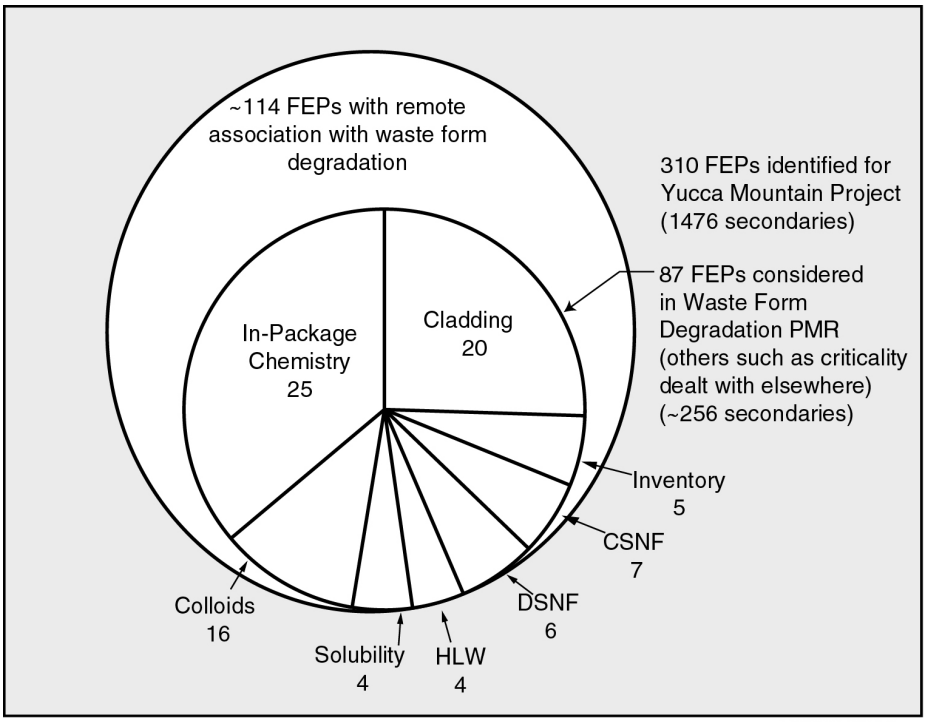


Figure 2.3-1. Waste Form FEPs

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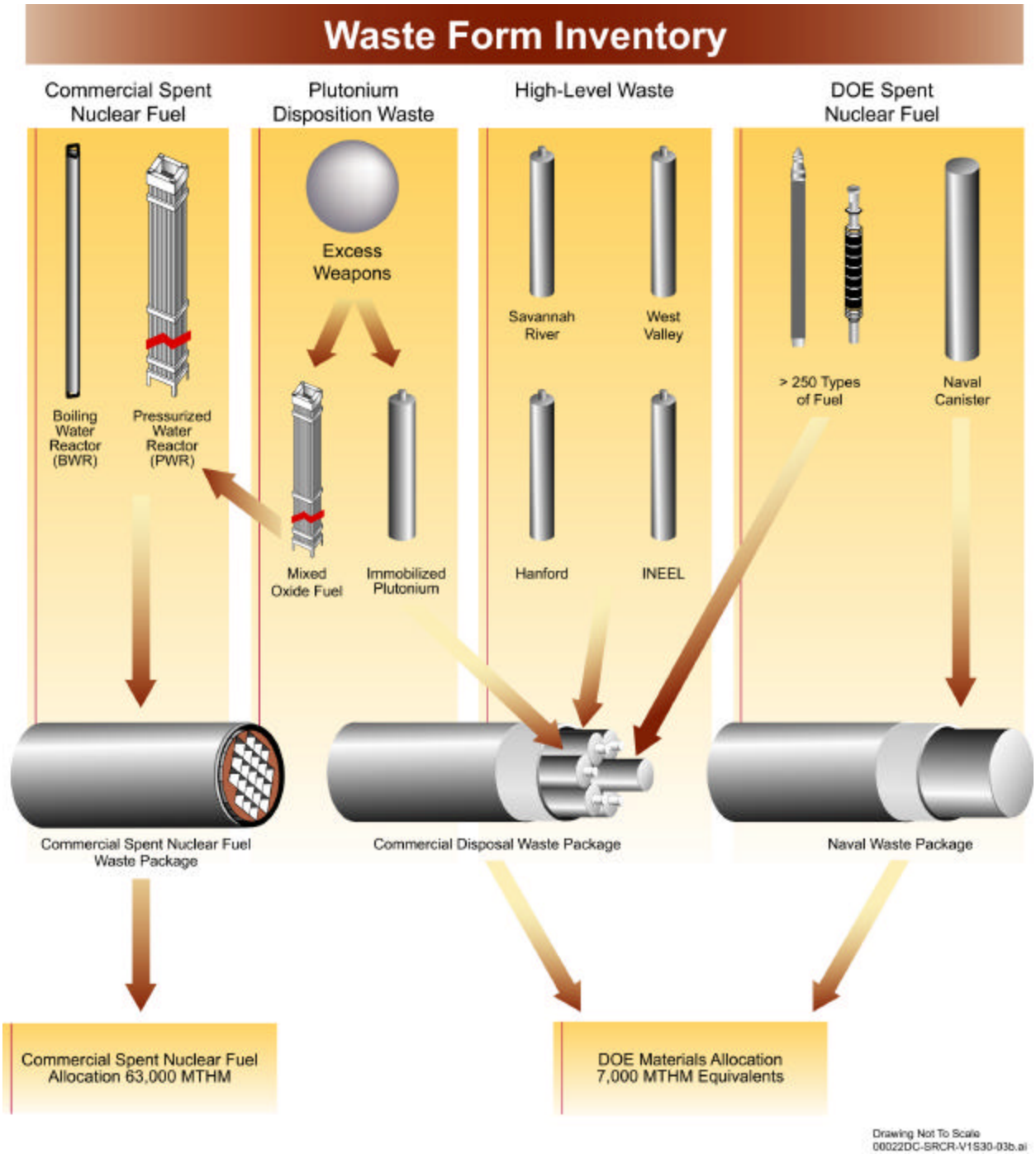


NOTE: The figure depicts the types and quantities of waste forms to be disposed of in the first repository and their representative waste package designs. Until a second repository is in operation, the Nuclear Waste Policy Act limits inventory in the first repository to 70,000 MTHM. Dispositioning excess plutonium from weapons programs into mixed oxide fuel for burnup in reactors and disposal in a repository aids in the fight against nuclear proliferation.

Figure 3.1-1. Waste Types Grouped into Three Representative Waste Packages for Modeling in TSPA-SR

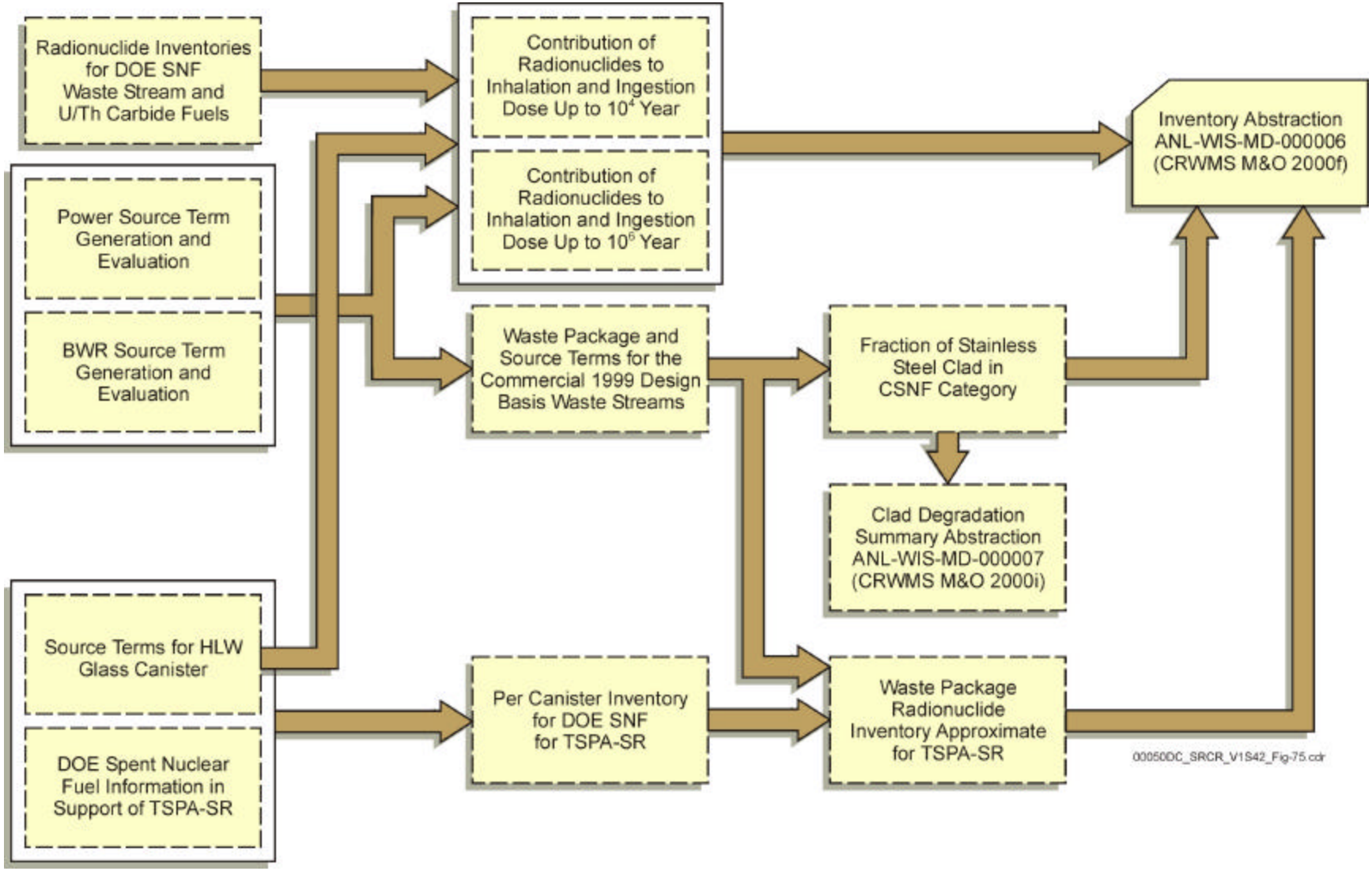


Figure 3.1-2. Flow of Information Supporting the Radionuclide Inventory Component

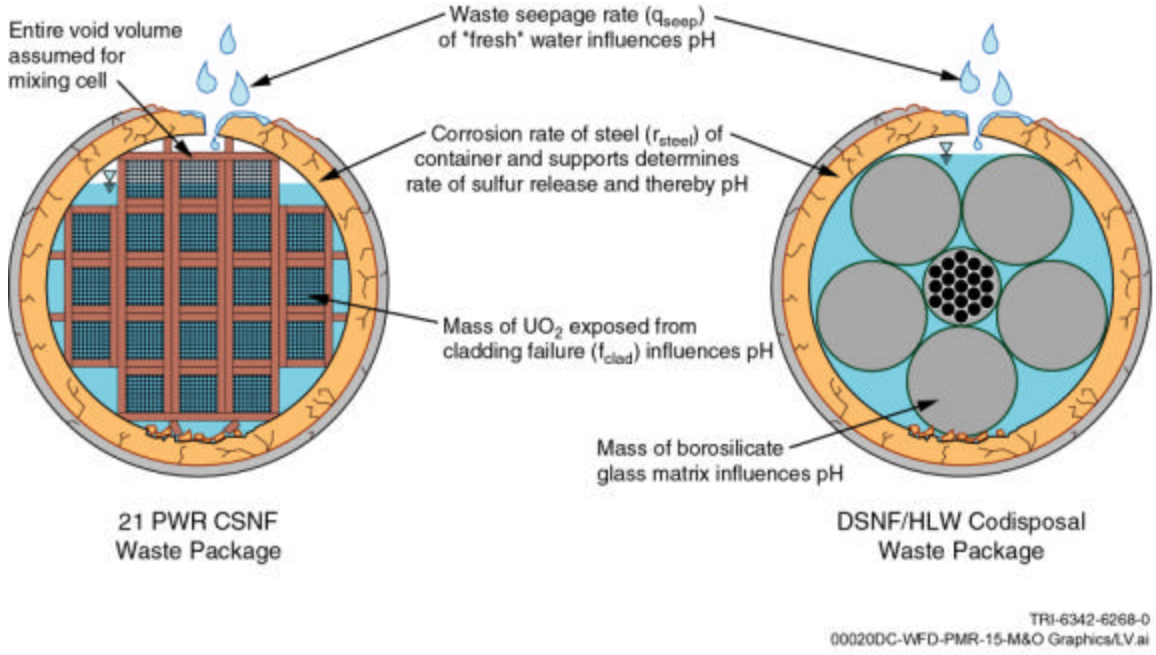
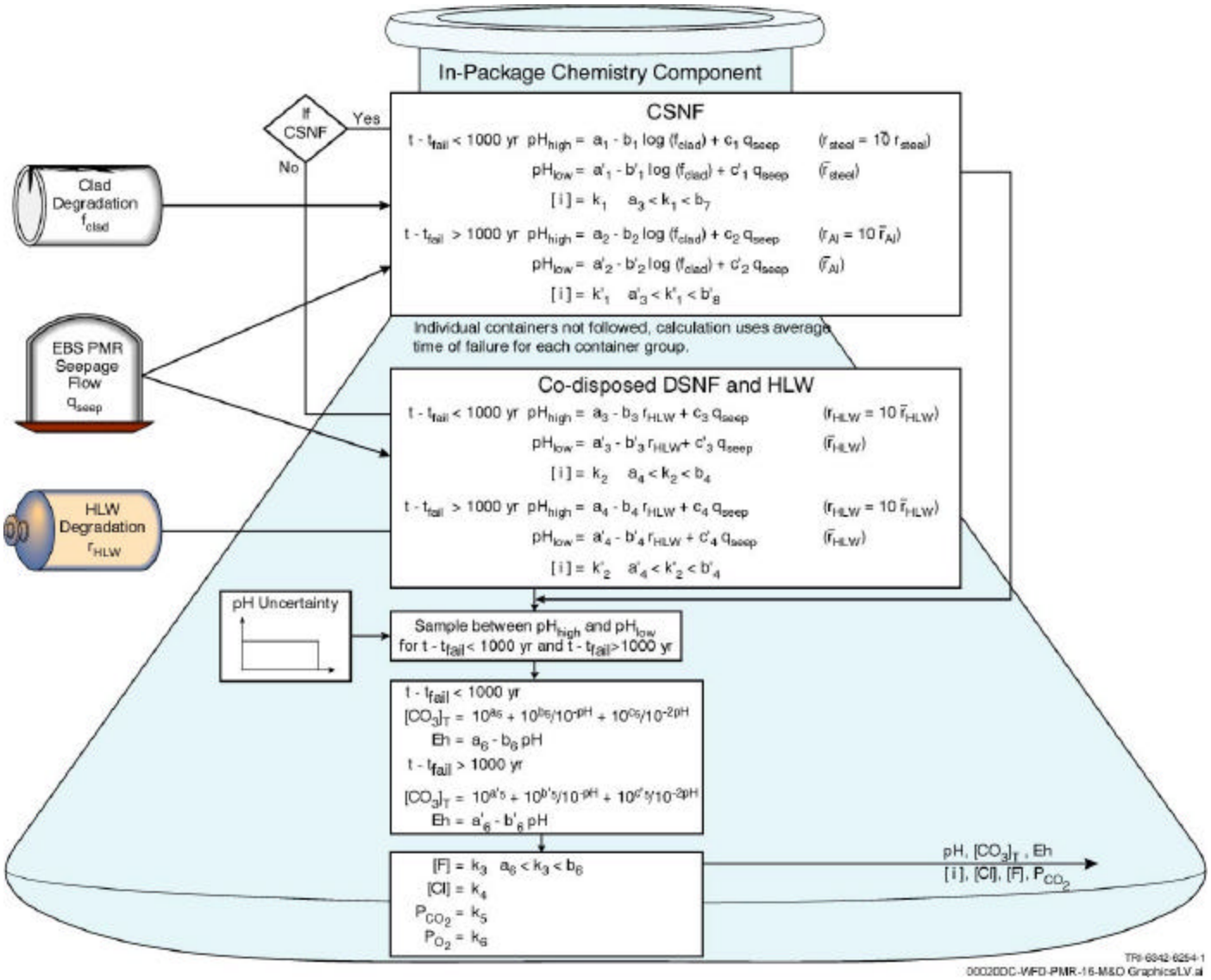
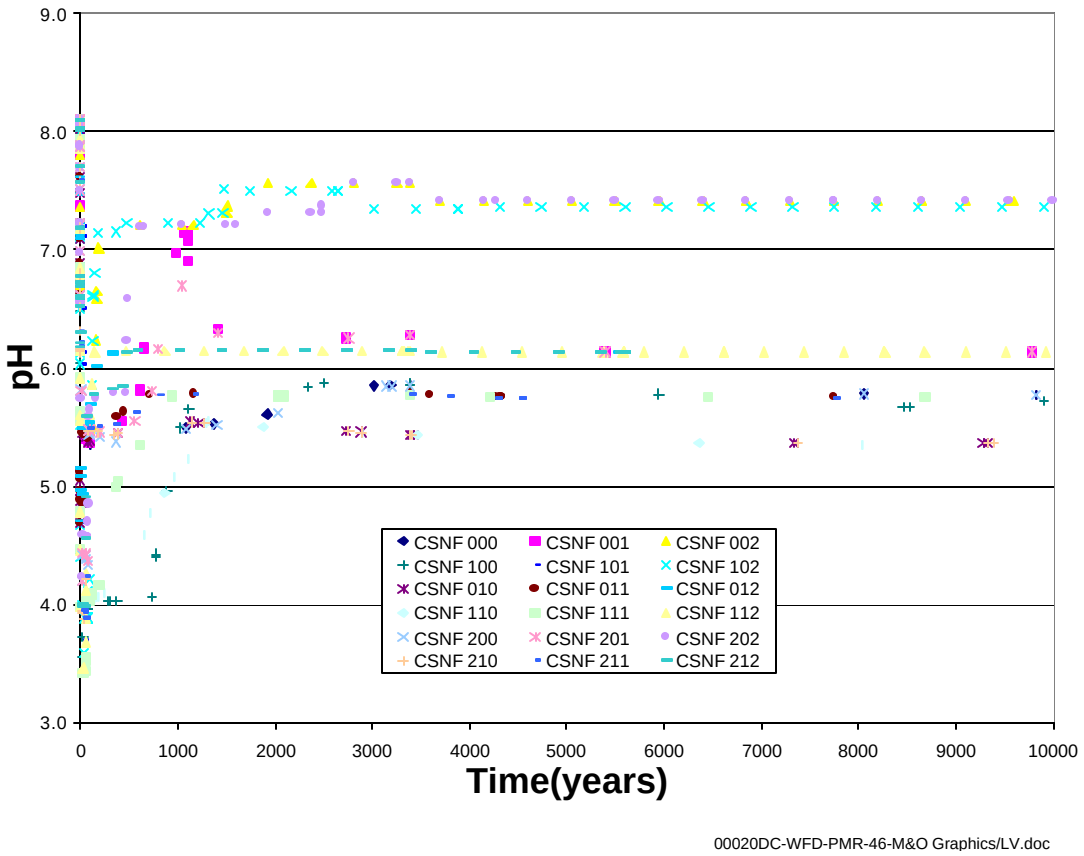


Figure 3.2-1. Conceptual Model of In-Package Chemistry



Source: *In-Package Chemistry Abstraction for TSPA-LA*. ANL-EBS-MD-00037. (CRWMS M&O 2000g)

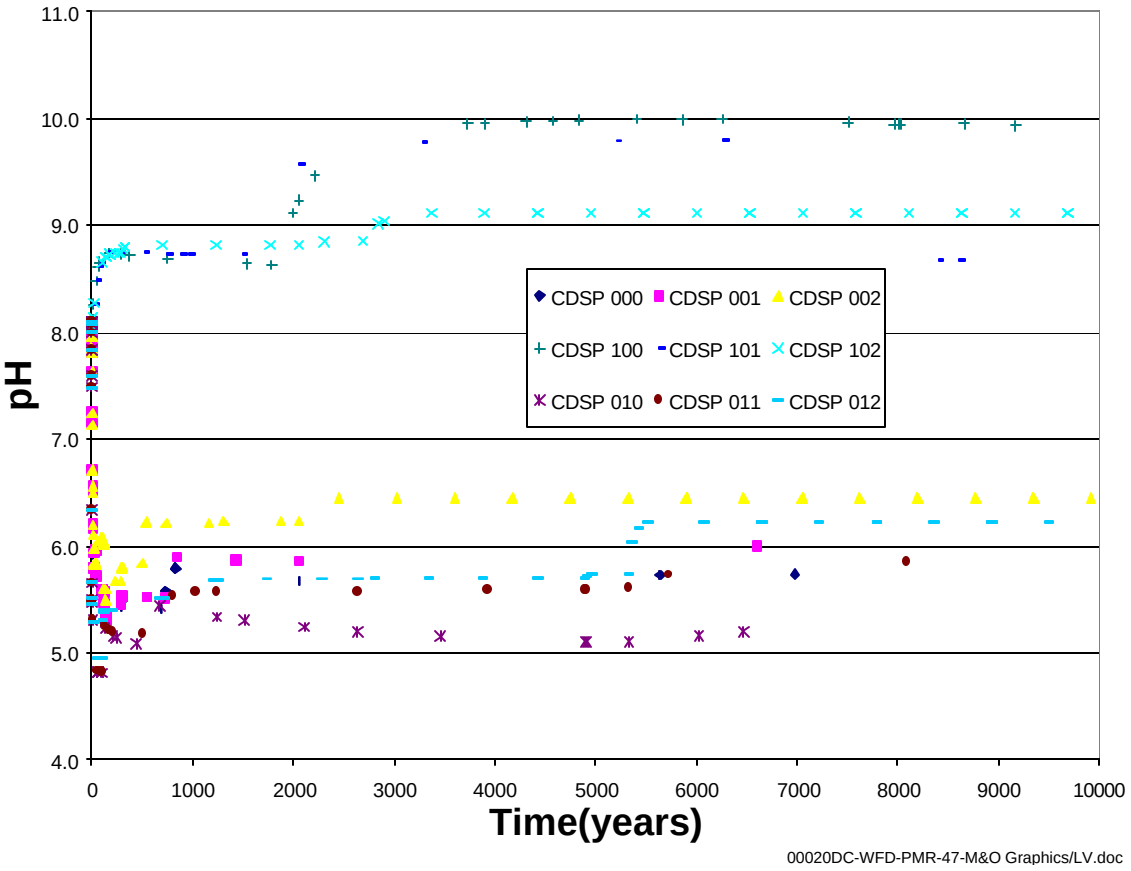
Figure 3.2-2. Abstraction Equations and Linkage of Subcomponents of In-Package Chemistry Component



Source: *In-Package Chemistry Abstraction for TSPA-LA*. ANL-EBS-MD-000037. (CRWMS M&O 2000g, Figure 1)

NOTE: CSNF numbers are code numbers for the CSNF calculations.

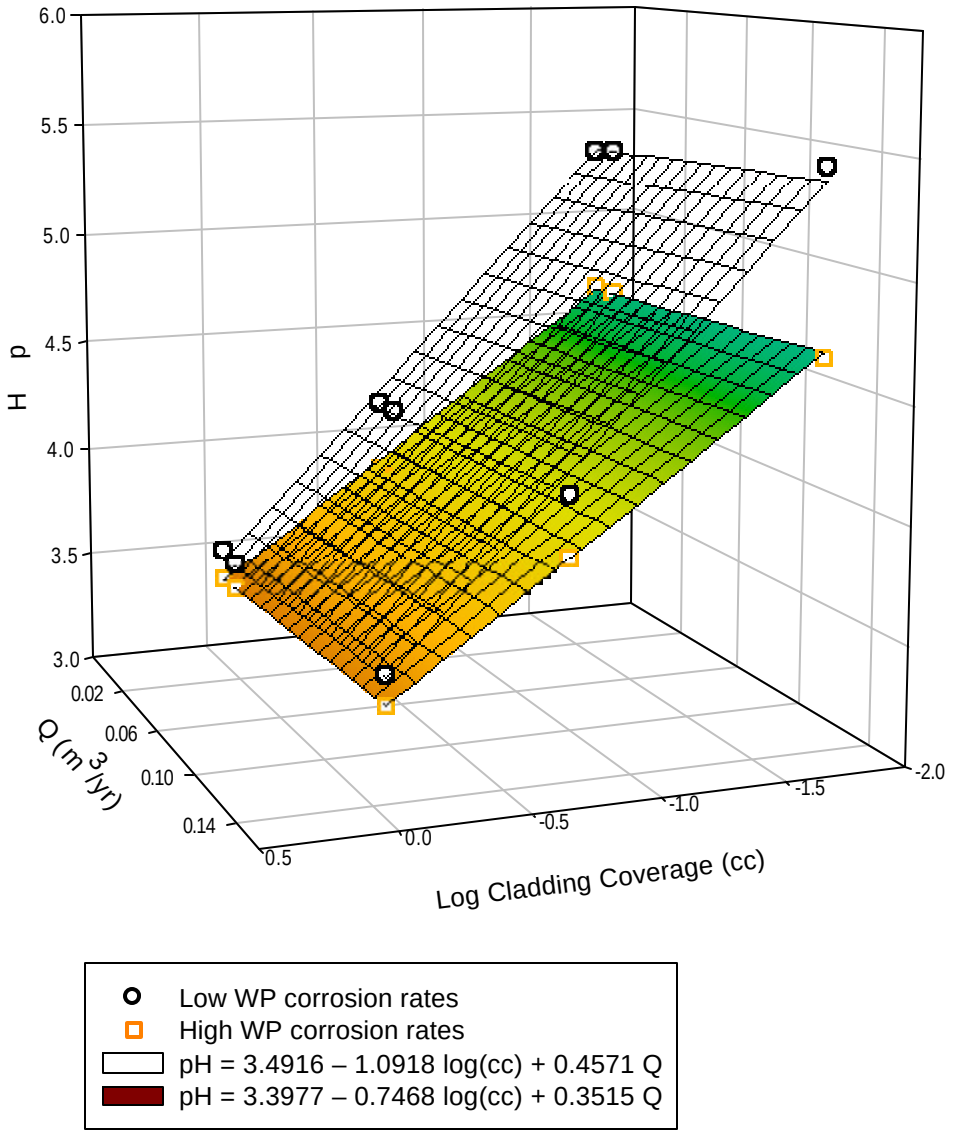
Figure 3.2-3. pH History for CSNF Process Model Calculations



Source: *In-Package Chemistry Abstraction for TSPA-LA*. ANL-EBS-MD-000037. (CRWMS M&O 2000g, Figure 2)

NOTE: CDSP numbers are code numbers for the codisposal waste package calculations.

Figure 3.2-4. pH History for Codisposal Process Model Calculations



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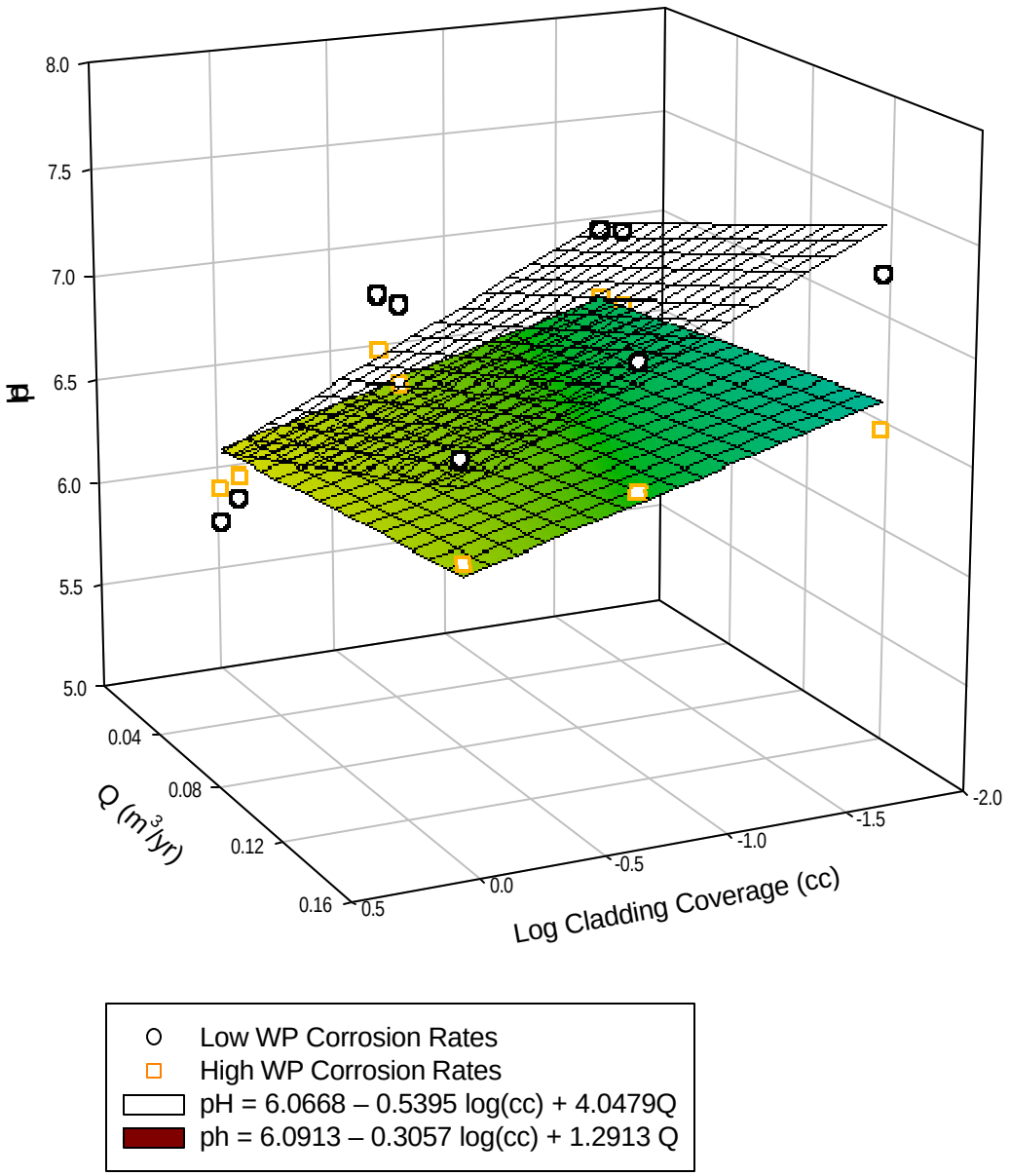
Source: *In-Package Chemistry Abstraction for TSPA-LA*. ANL-EBS-MD-000037. (CRWMS M&O 2000g, Figure 3)

Figure 3.2-5. pH Abstraction for < 1,000 Years, CSNF



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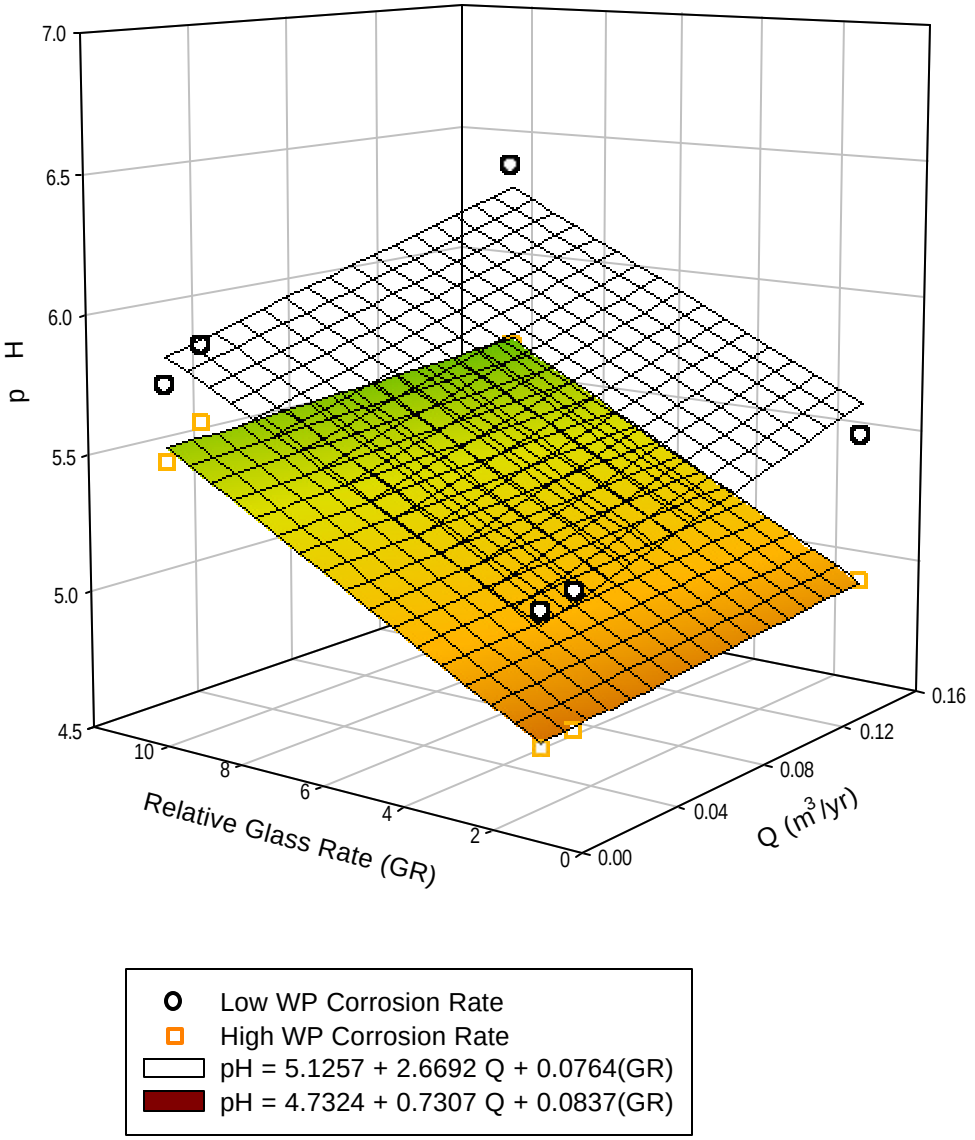
Source: *In-Package Chemistry Abstraction for TSPA-LA*. ANL-EBS-MD-000037. (CRWMS M&O 2000g, Figure 4)

Figure 3.2-6. pH Abstraction for 1,000 to 10,000 Years, CSNF



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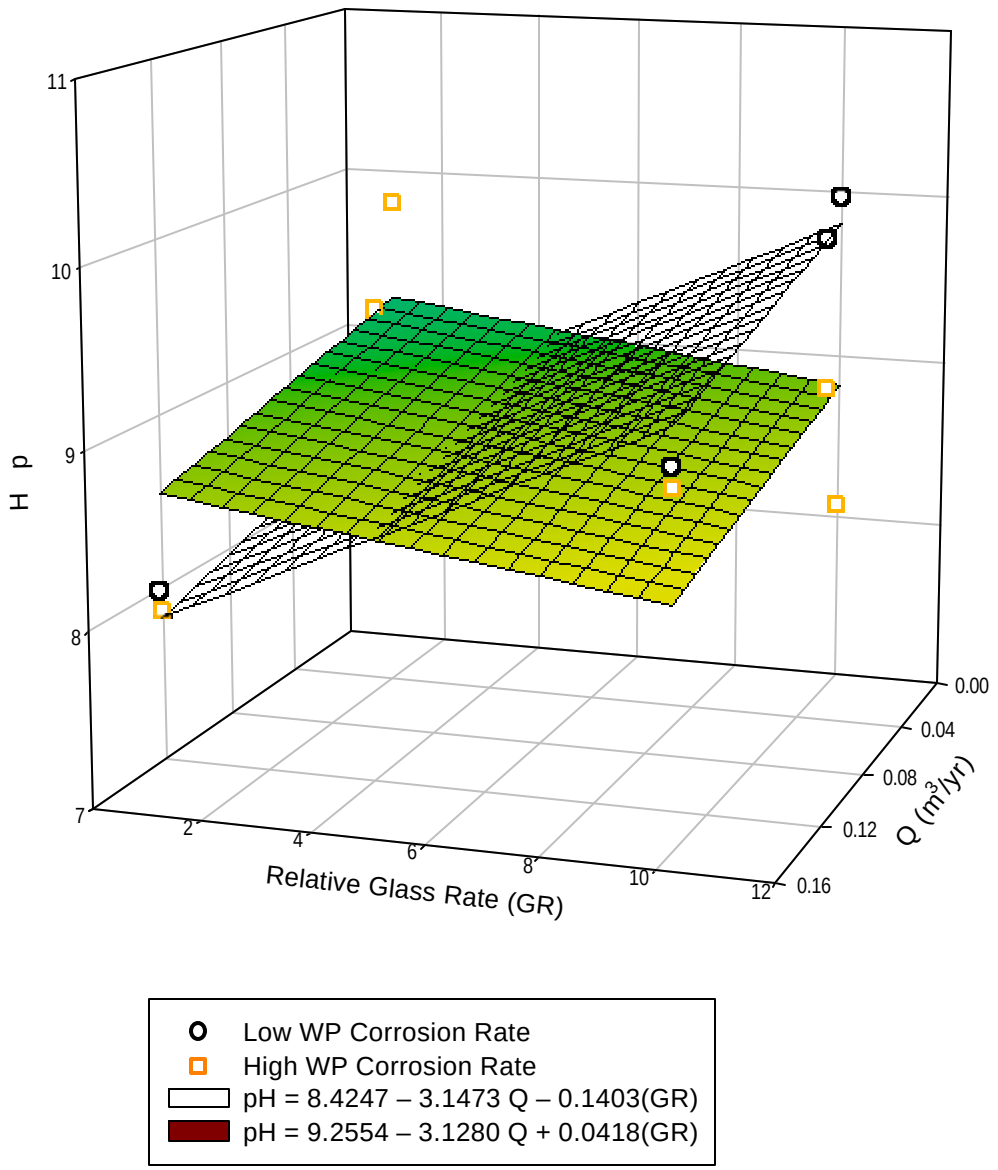
Source: *In-Package Chemistry Abstraction for TSPA-LA*. ANL-EBS-MD-000037. (CRWMS M&O 2000g, Figure 5)

Figure 3.2-7. pH Abstraction for < 1,000 Years, Codisposal Waste Packages



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Source: *In-Package Chemistry Abstraction for TSPA-LA*. ANL-EBS-MD-000037. (CRWMS M&O 2000g, Figure 6)

Figure 3.2-8. pH Abstraction for 1,000 to 100,000 Years, Codisposal Waste Packages

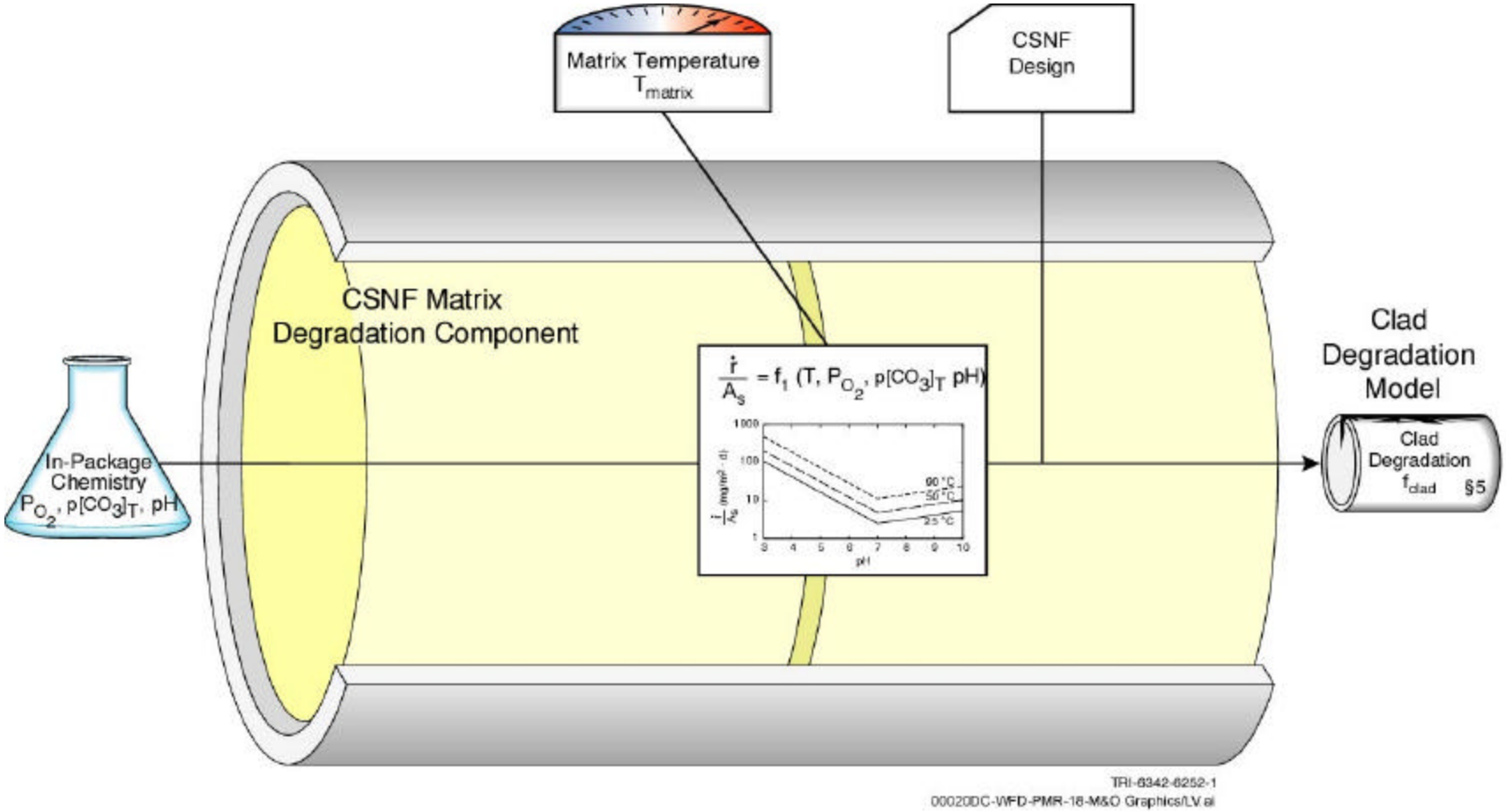
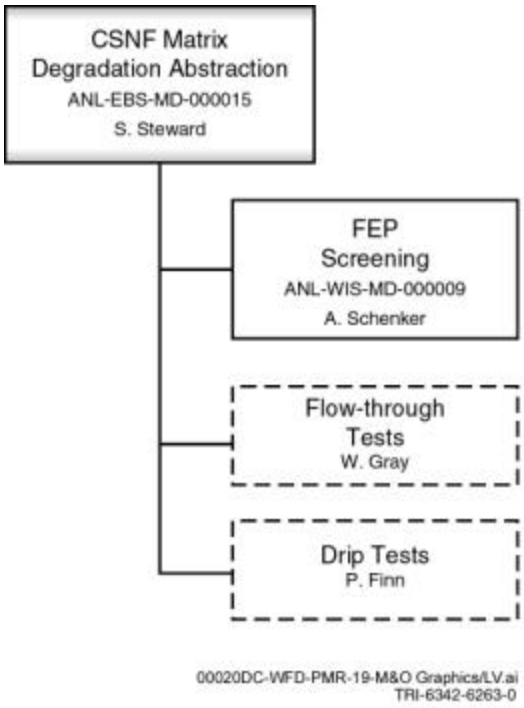
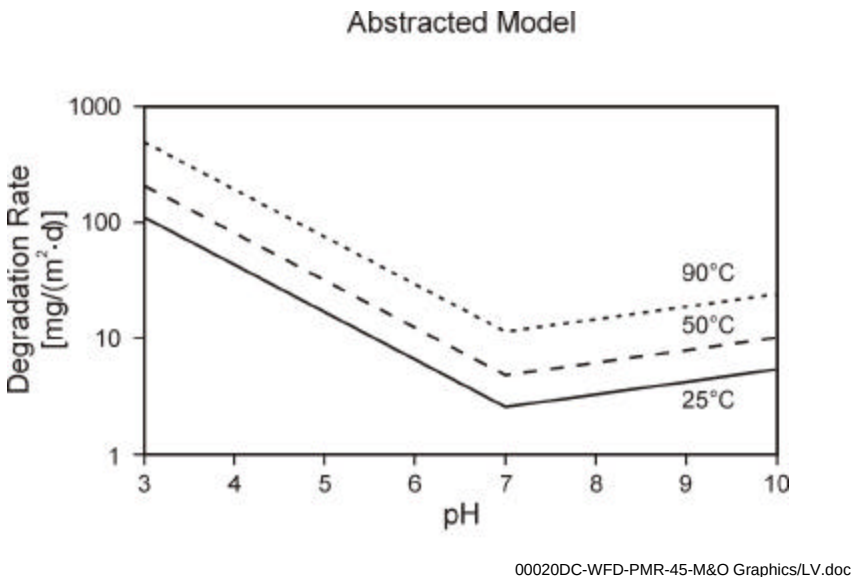


Figure 3.3-1. CSNF Degradation Model



NOTE: Tracking information in CRWMS M&O 2000h.

Figure 3.3-2. Analysis/Model Reports Supporting the Development of the CSNF Degradation Component



Source: *CSNF Waste Form Degradation: Summary Abstraction*. ANL-EBS-MD-000015. (CRWMS M&O 2000h, Figure 2)

Figure 3.3-3. Abstracted Dissolution Model

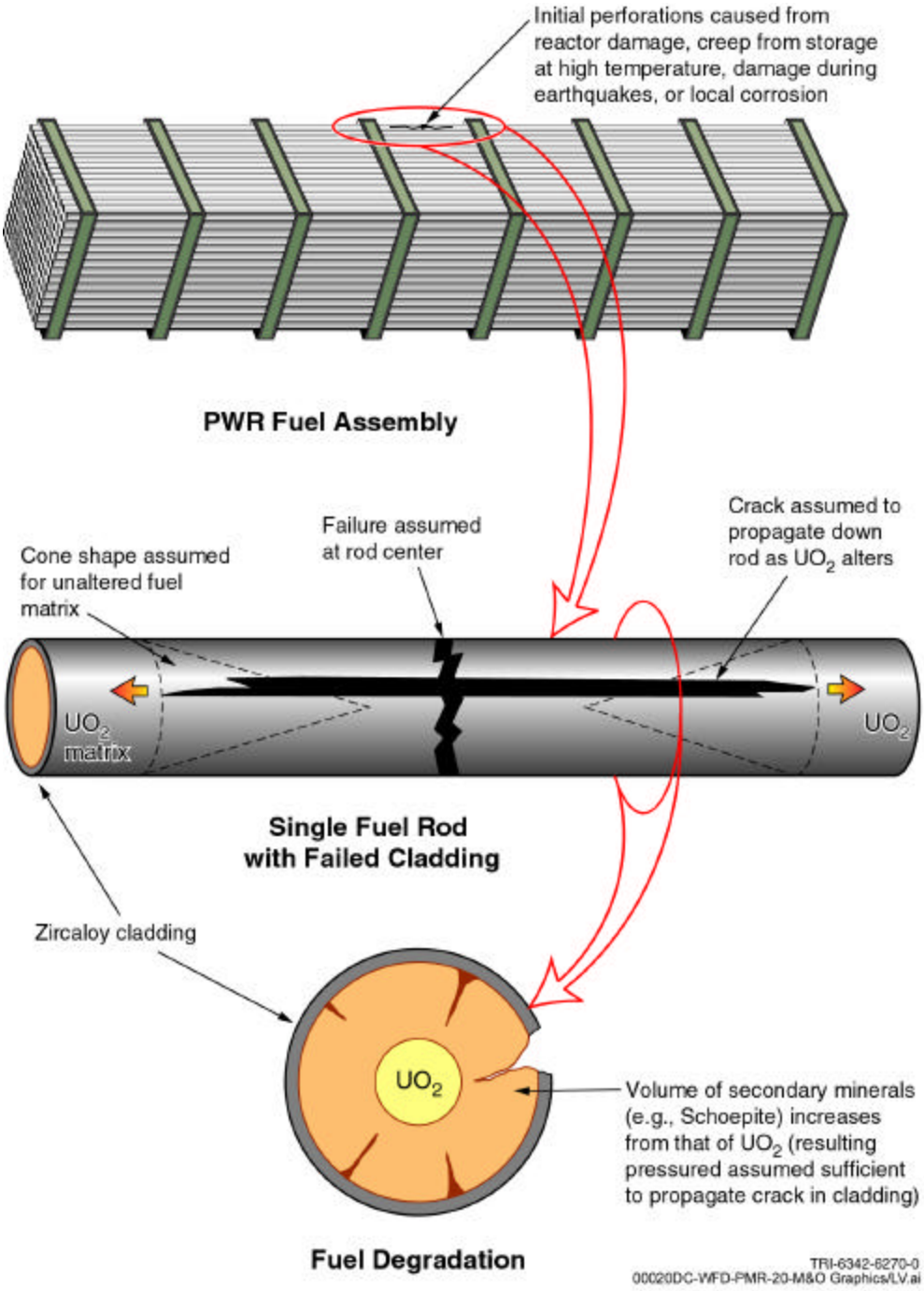


Figure 3.4-1. Conceptual Model of CSNF Cladding Degradation

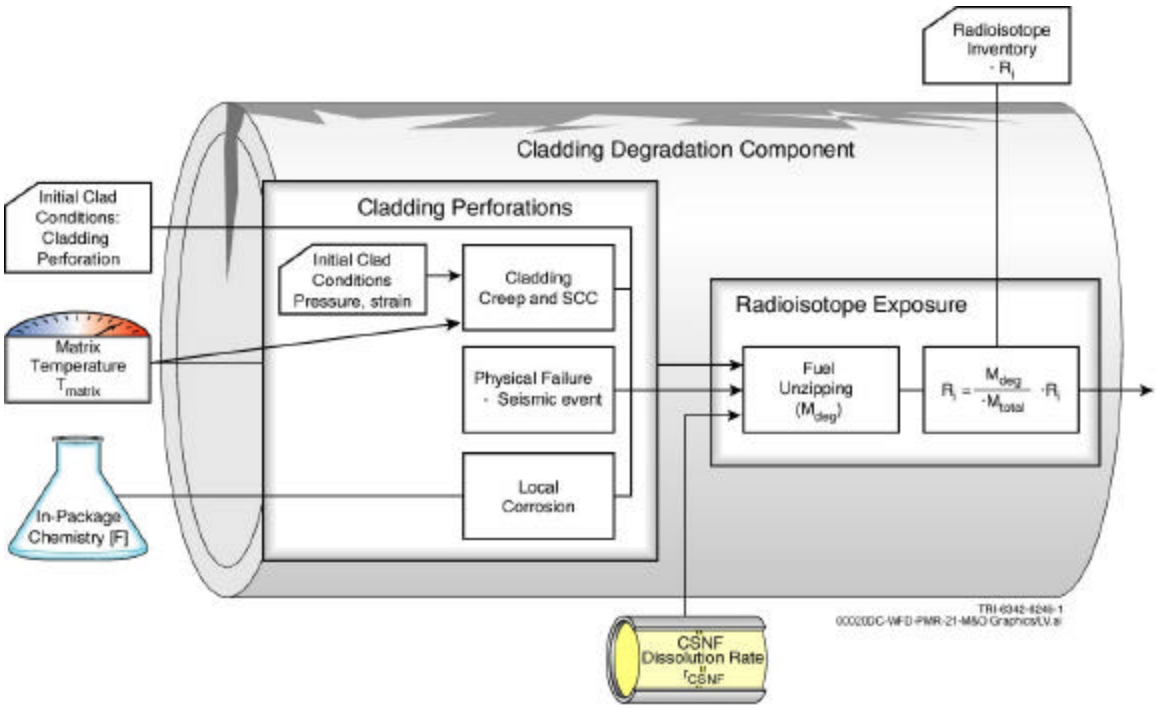
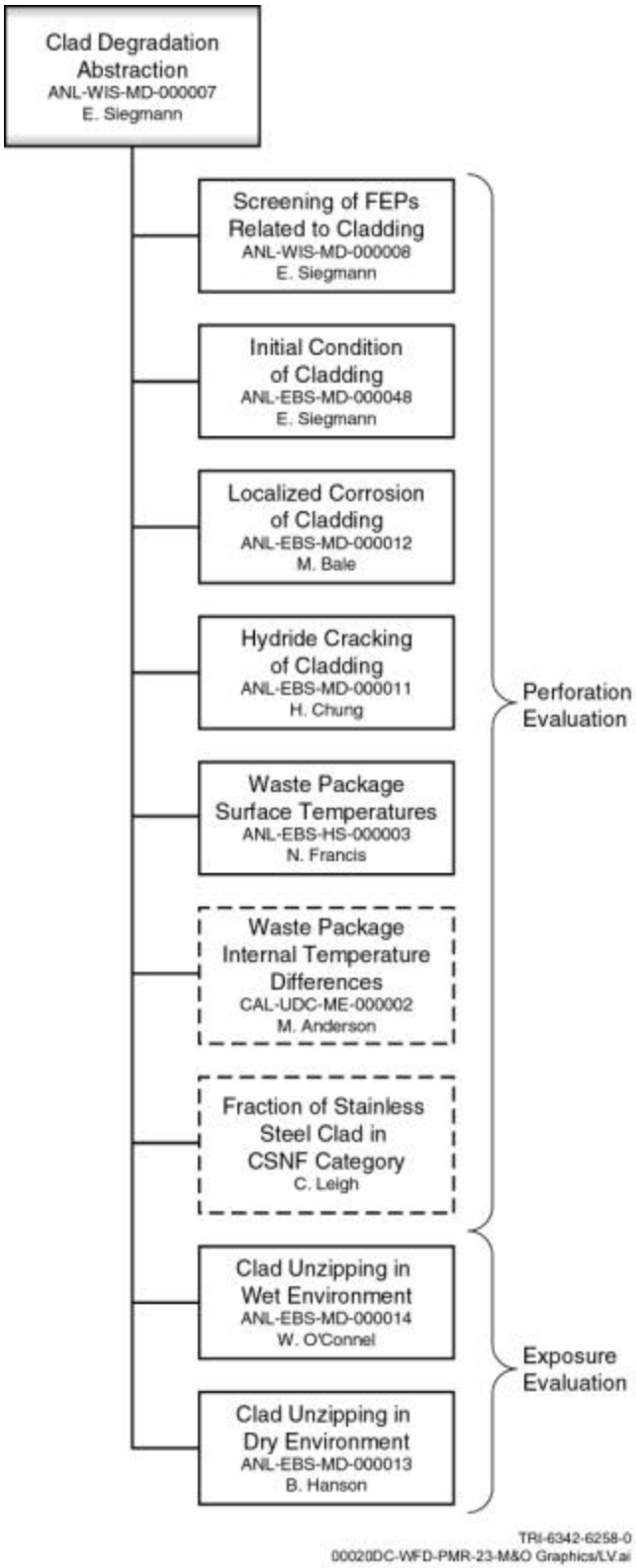
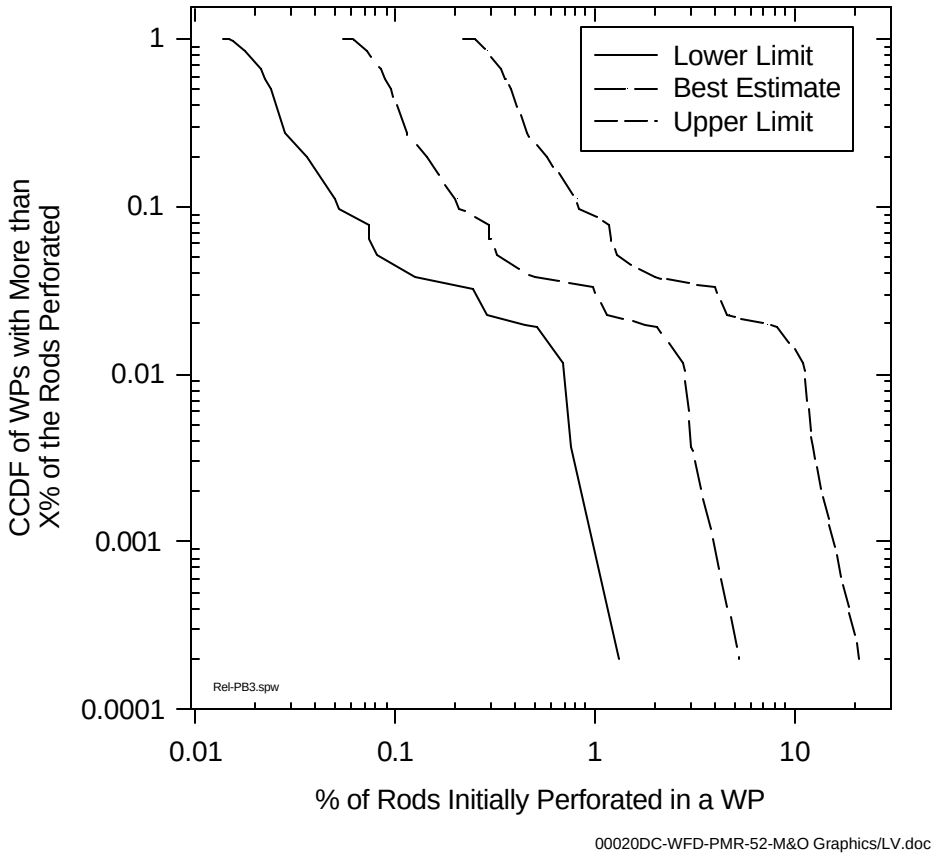


Figure 3.4-2. Implementation of CSNF Cladding Degradation in TSPA-SR



NOTE: Tracking information in CRWMS M&O 2000i

Figure 3.4-3. Calculations and Analysis and Model Reports Supporting the Development of the CSNF Cladding Degradation Component



Source: *Initial Cladding Condition*. ANL-EBS-MD-000048. (CRWMS M&O 2000p, Table 14)

Figure 3.4-4. CCDF for Existing Perforated CSNF

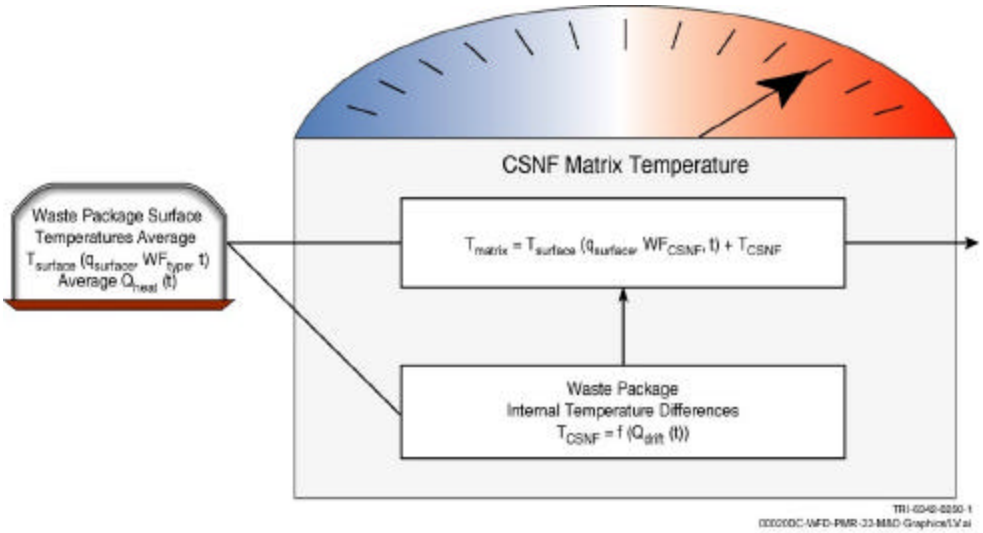
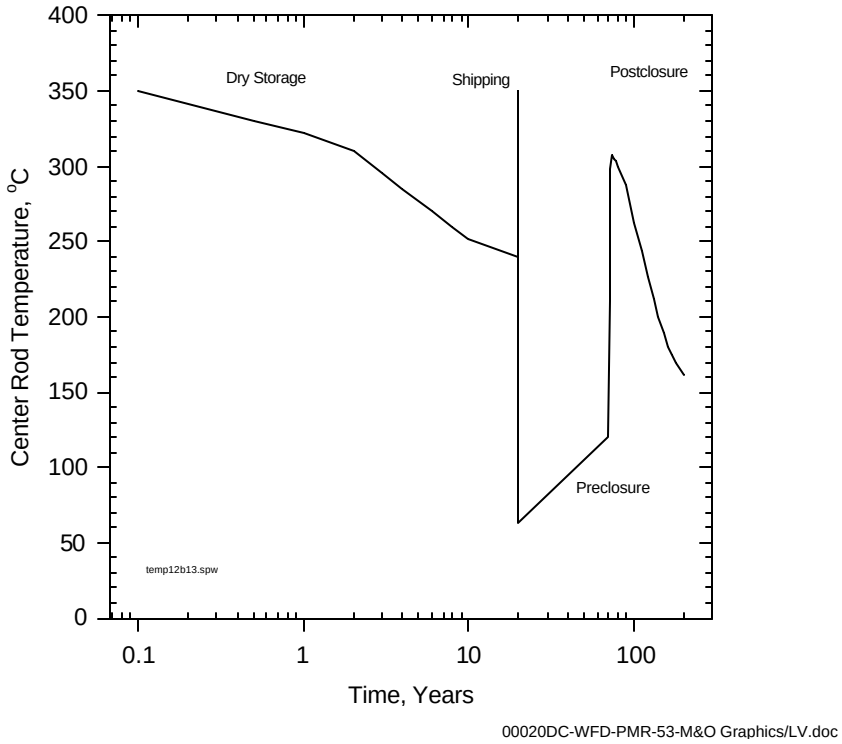
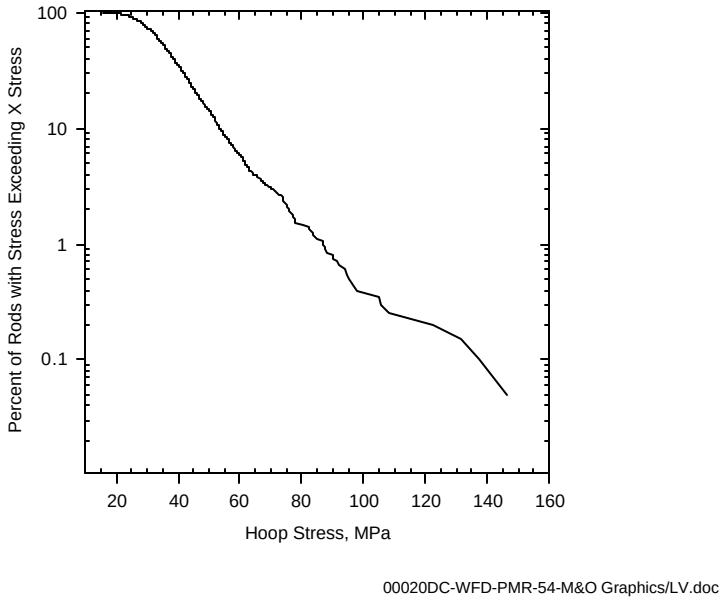


Figure 3.4-5. CSNF Matrix Temperature



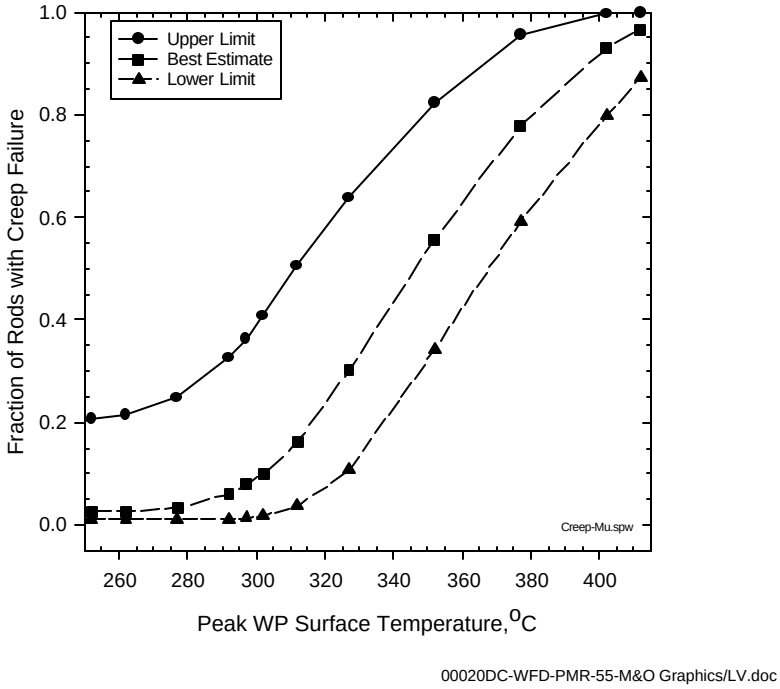
Source: *Clad Degradation–Summary and Abstraction*. ANL-WIS-MD-000007. (CRWMS M&O 2000i, Figure 5)

Figure 3.4-6. Temperature History for Average Center Rod in WP



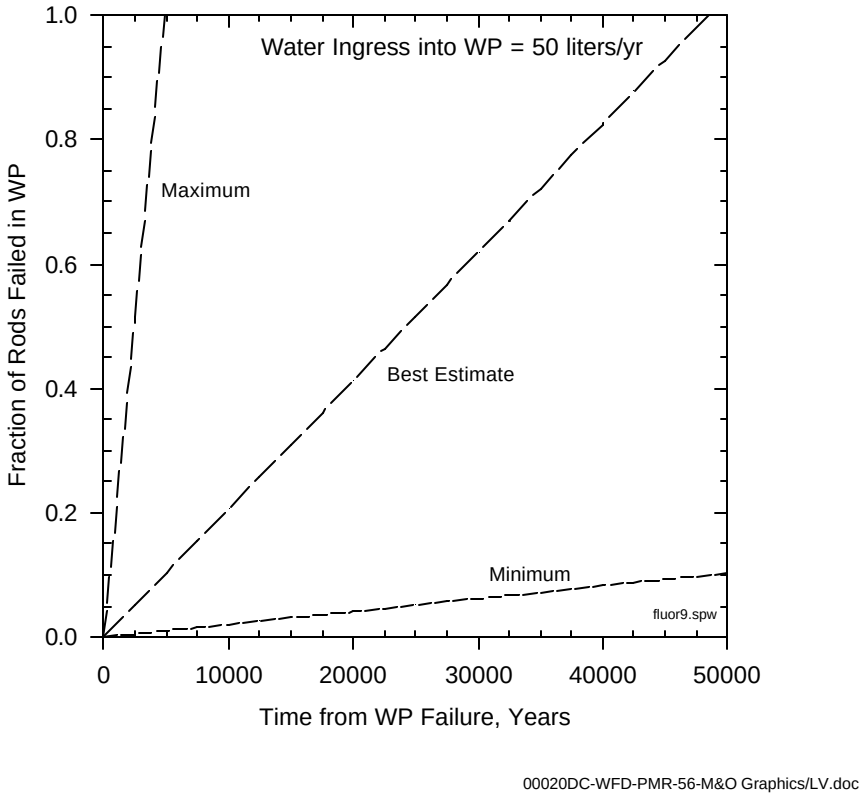
Source: *Clad Degradation–Summary and Abstraction*. ANL-WIS-MD-000007. (CRWMS M&O 2000i, Figure 1)

Figure 3.4-7. CCDF for Rod Stress As Received (room temperature)



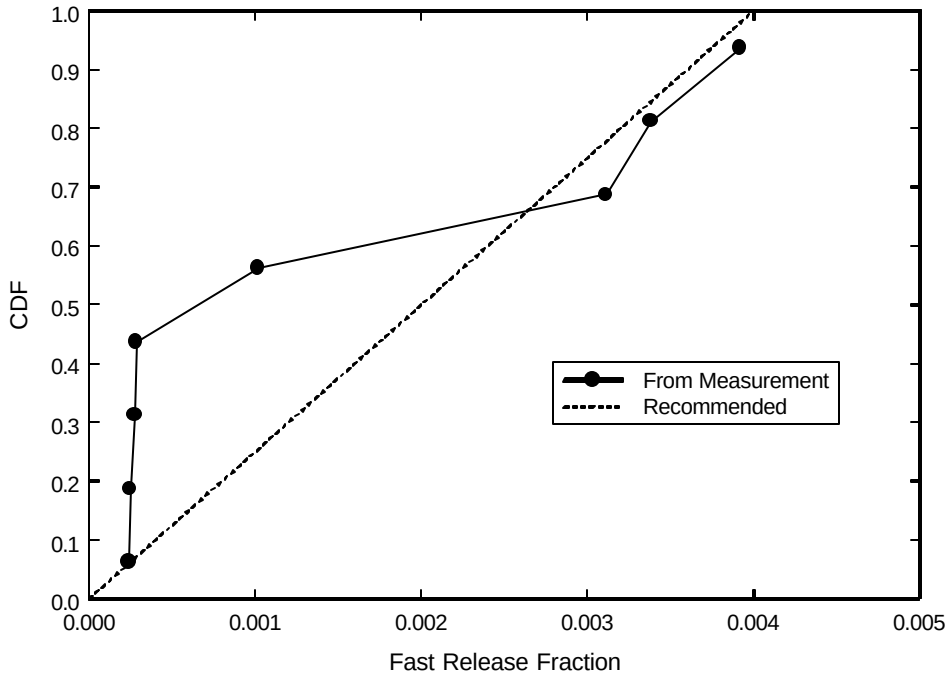
Source: *Clad Degradation–Summary and Abstraction*. ANL-WIS-MD-000007. (CRWMS M&O 2000i, Figure 8)

Figure 3.4-8. Creep and SCC Perforated as a Function of WP Surface Temperature



Source: *Clad Degradation–Summary and Abstraction*. ANL-WIS-MD-000007. (CRWMS M&O 2000i, Figure 9)

Figure 3.4-9. Example of Localized Corrosion with Constant Water Flow into WP



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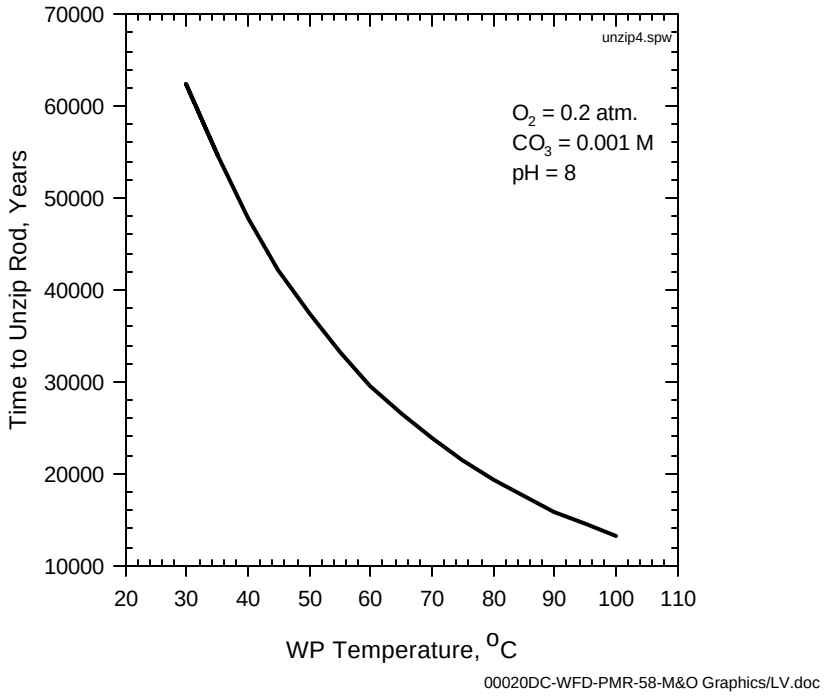
Source: *Clad Degradation–Summary and Abstraction*. ANL-WIS-MD-000007. (CRWMS M&O 2000i, Figure 11)

Figure 3.4-10. CDF for Fast Release Fraction from Fuel Matrix



Source: *Clad Degradation–Summary and Abstraction*. ANL-WIS-MD-000007. (CRWMS M&O 2000i, Figure 13)

Figure 3.4-11. Time to Unzip Rod versus WP Temperature

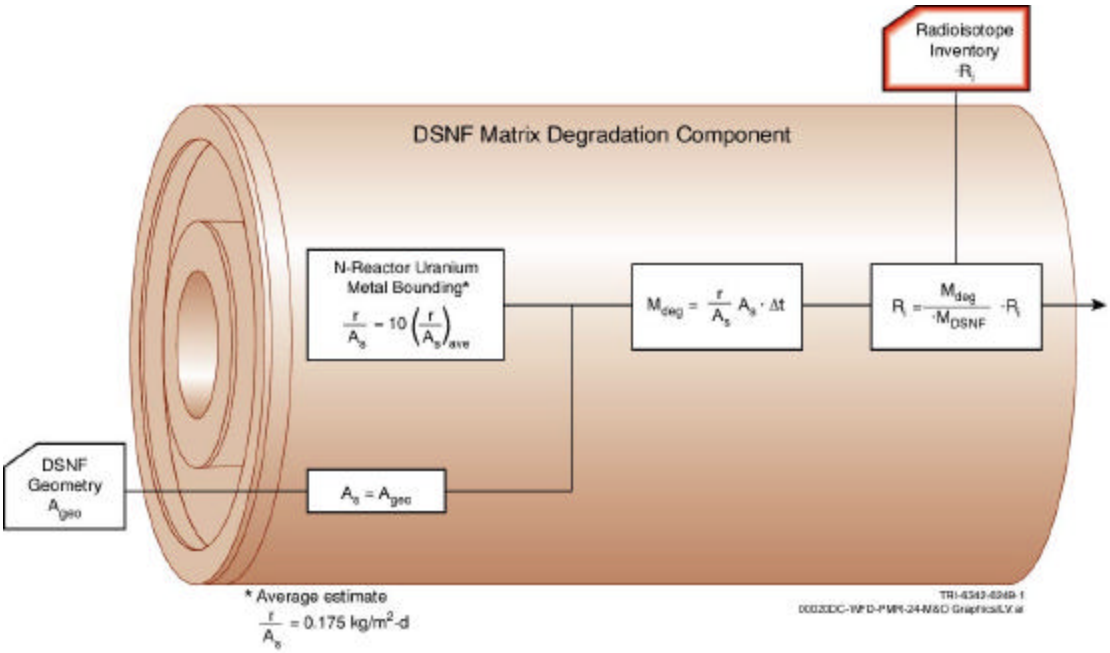


Figure 3.5-1. Implementation of DSNF (exclusive of naval SNF) Degradation in TSPA-SR

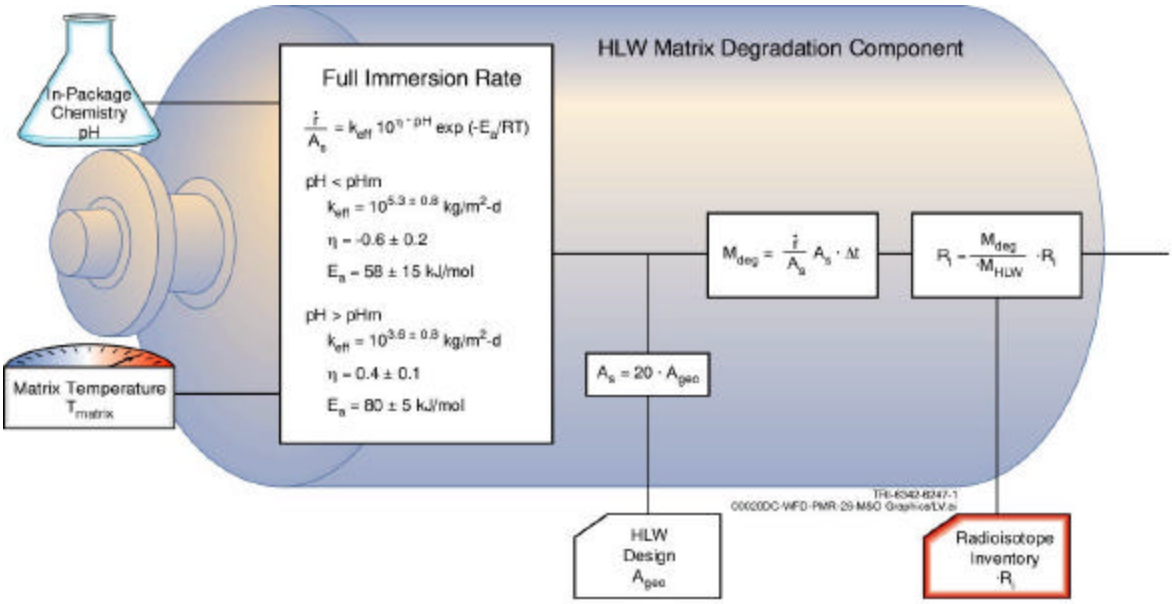


Figure 3.6-1. Bounding Rates in HLW Degradation Component

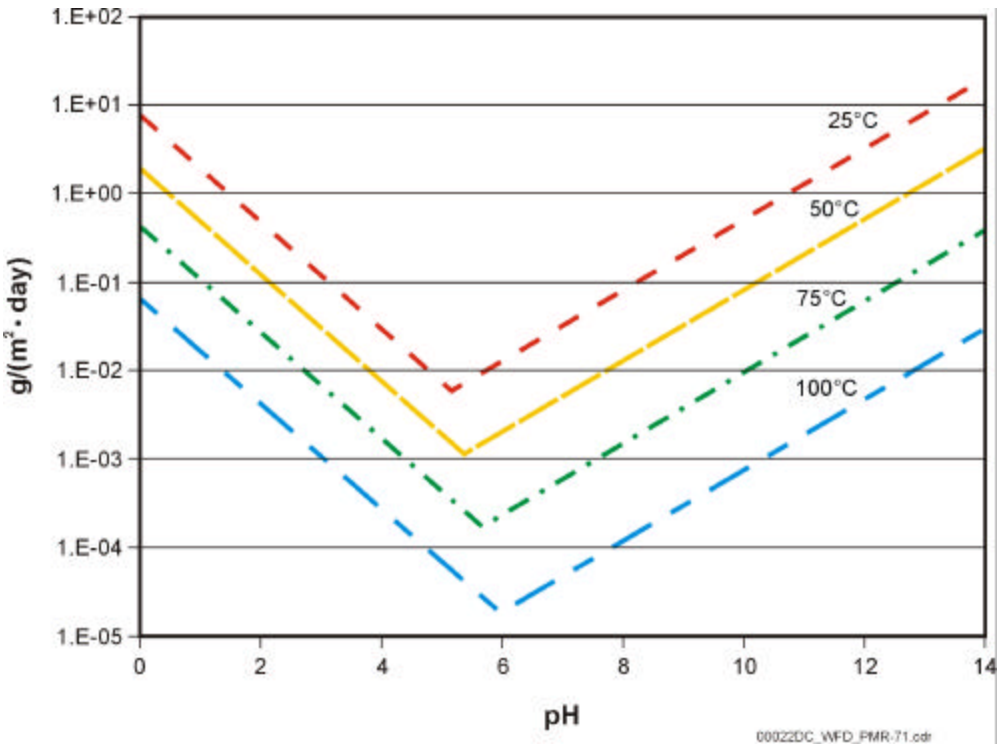


Figure 3.6-2. Abstracted Glass Dissolution Rate Model

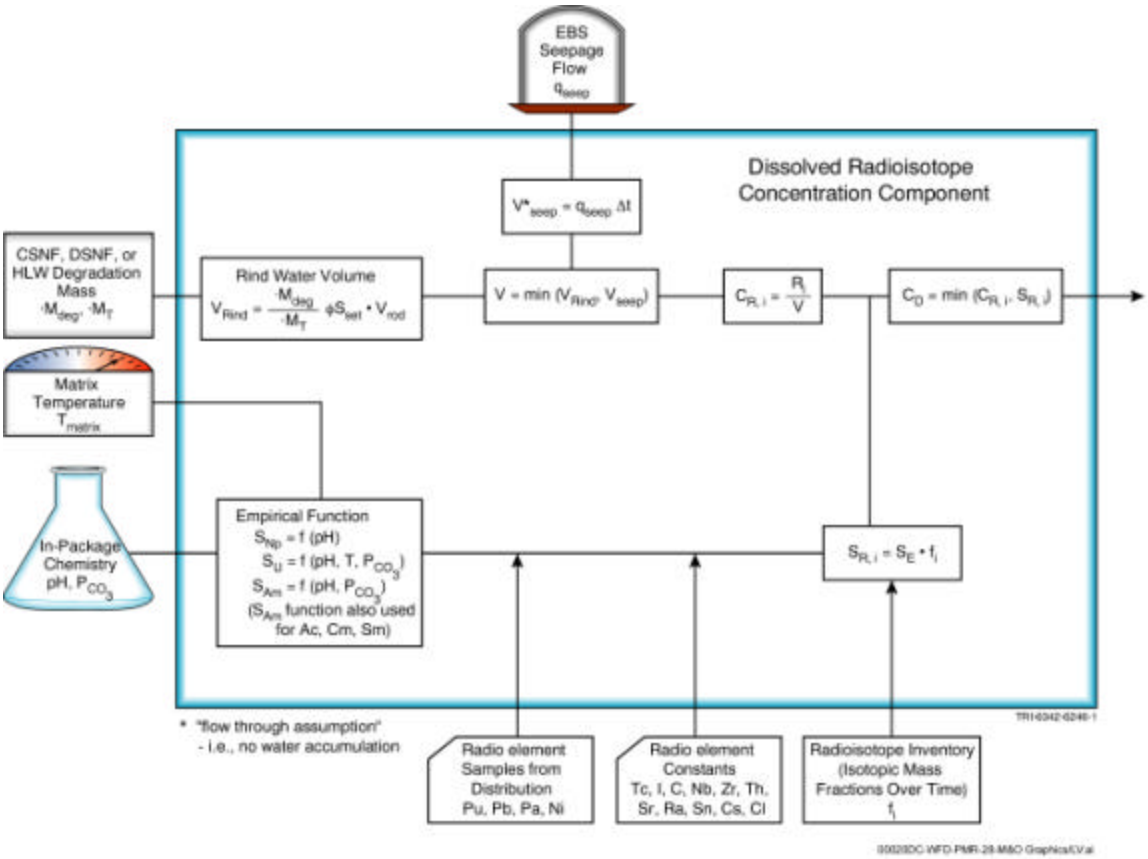
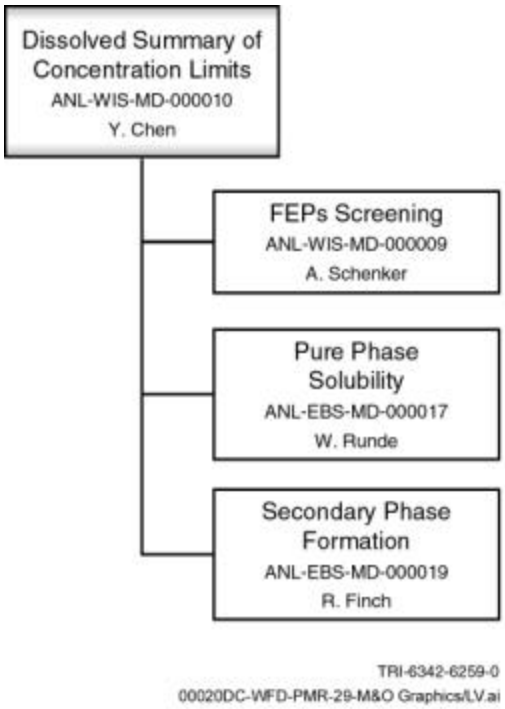


Figure 3.7-1. Linkage of Subcomponents of Solubility Component



NOTE: Tracking information in CRWMS M&O 2000l

Figure 3.7-2. Calculations and Analysis/Model Reports Supporting the Development of the Solubility Component

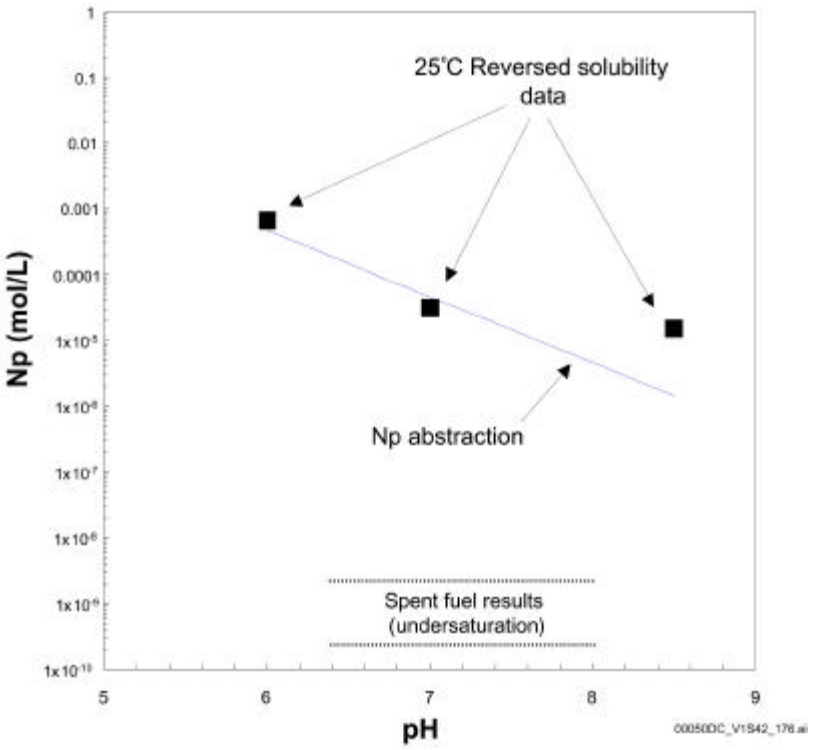
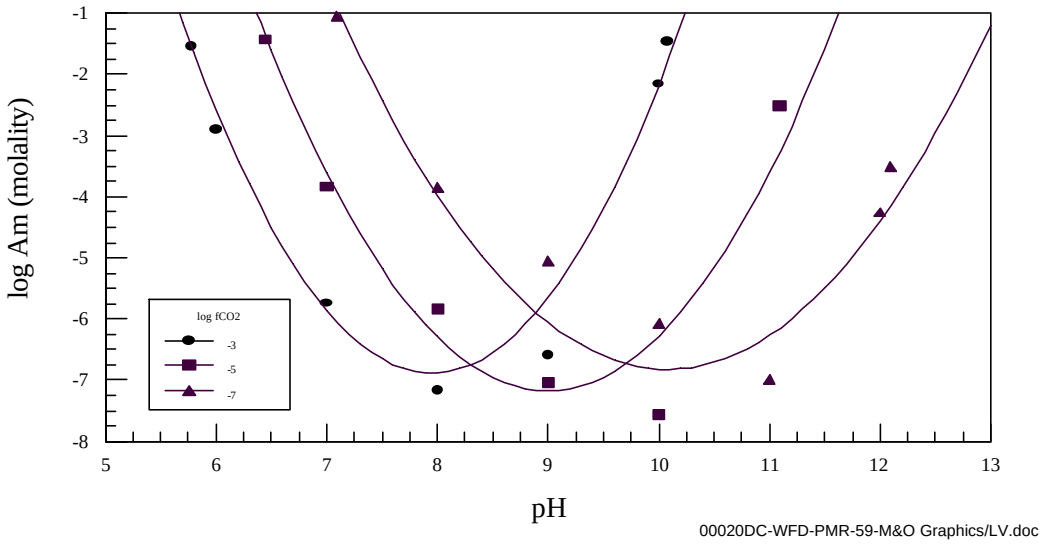


Figure 3.7-3. Comparison of Neptunium Solubility Abstraction to the Data of Efurd et al. (1998)



Source: *Summary of Dissolved Concentration Limits*. ANL-WIS-MD-000010. (CRWMS M&O 2000l, Figure 2)

Figure 3.7-4. Comparison of Fitted Response Surface (solid curves) with Calculated Americium Solubilities

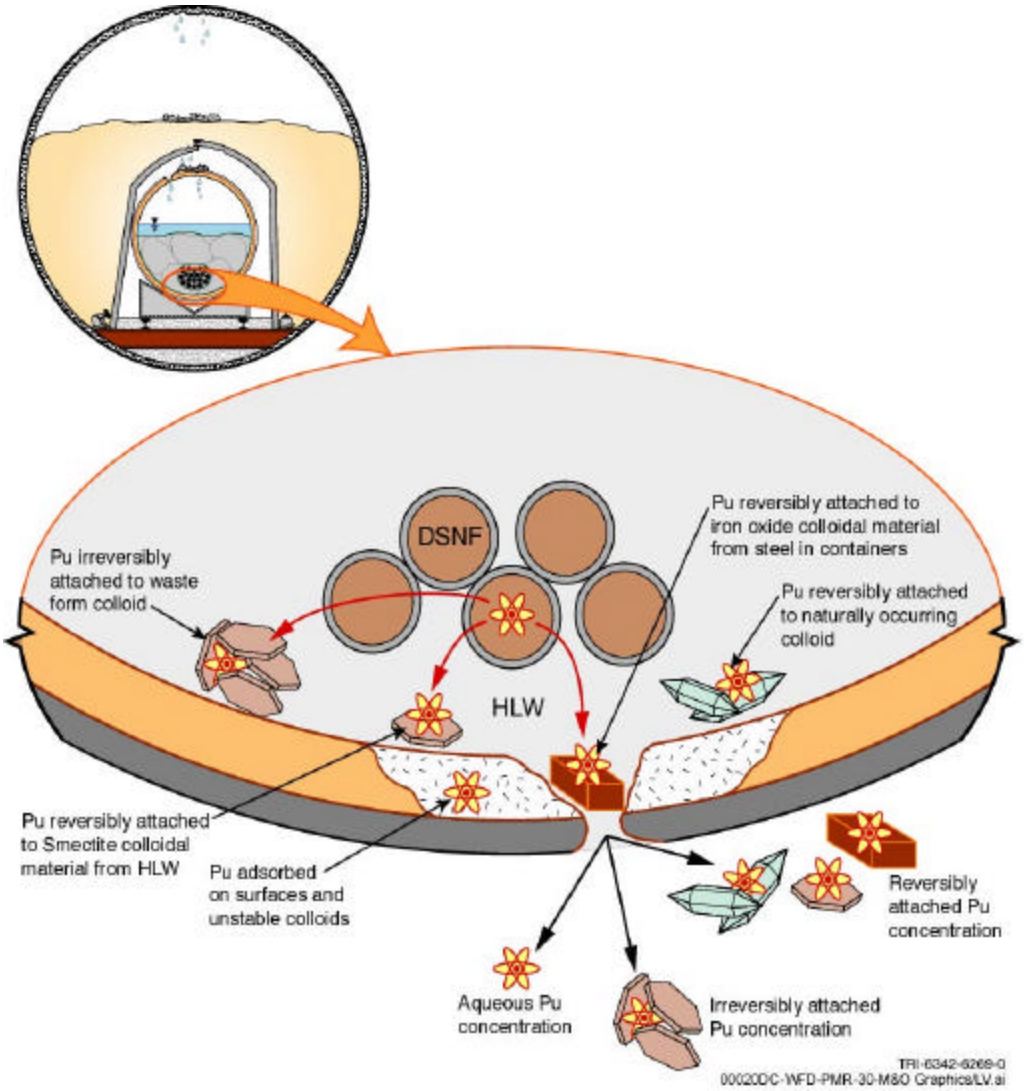


Figure 3.8-1. Conceptual Model of Formation of Reversibly and Irreversibly Attached Radionuclides on Colloids

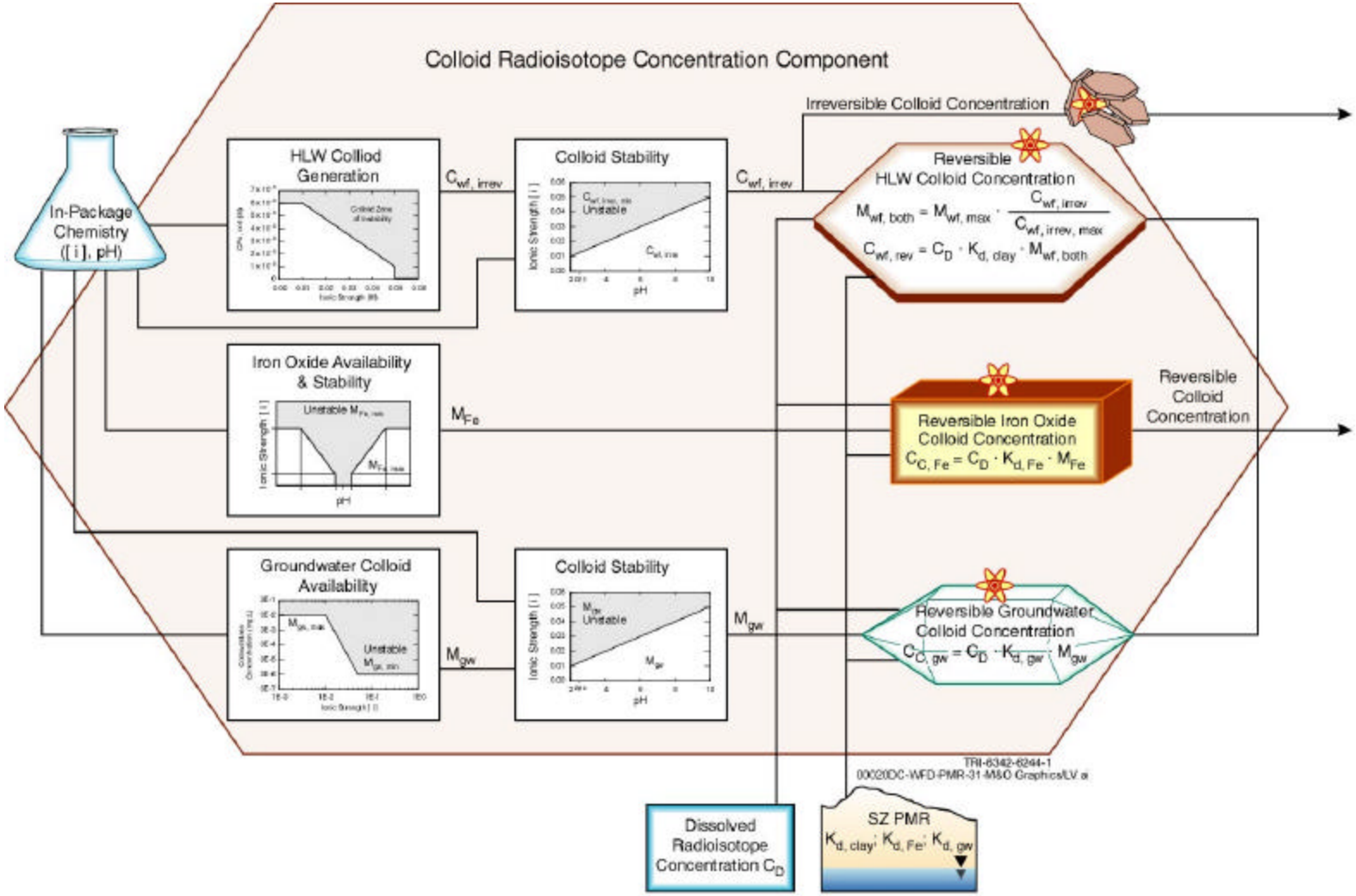
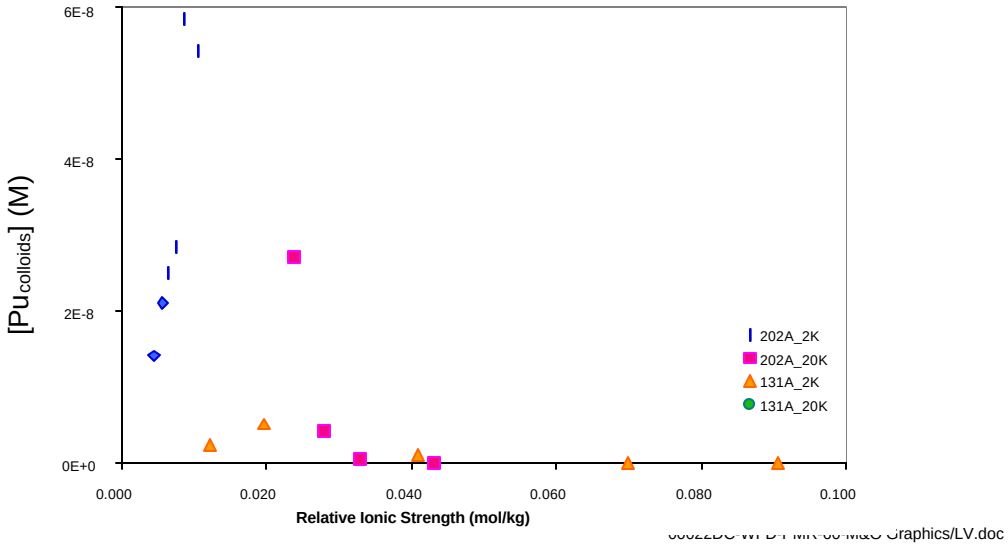
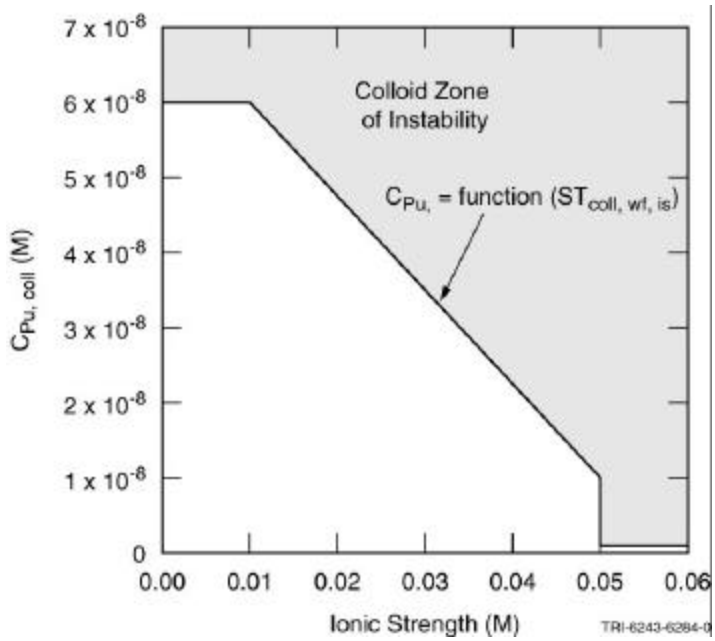


Figure 3.8-2. Linkage of Subcomponents of Colloidal Radionuclide Component



NOTE: Plutonium-bearing colloids as a function of ionic strength for corrosion tests on glass samples SRL 202A and SRL 131A at surface area to volume ratios of 2,000 and 20,000/m (at 90°C). Figure and modified caption from CRWMS M&O (2000m, Figure 7).

Figure 3.8-3. Generation of Waste-Form Colloids; Data



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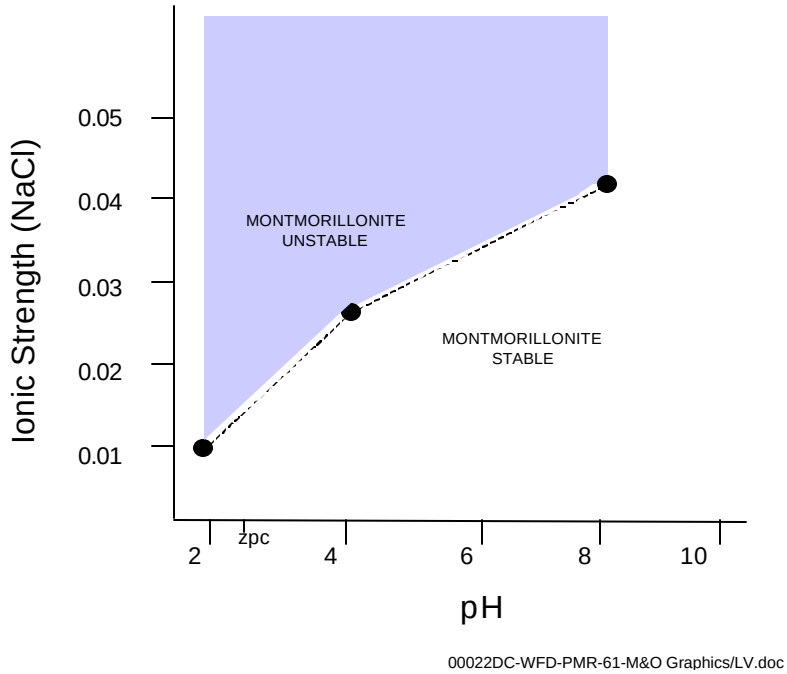
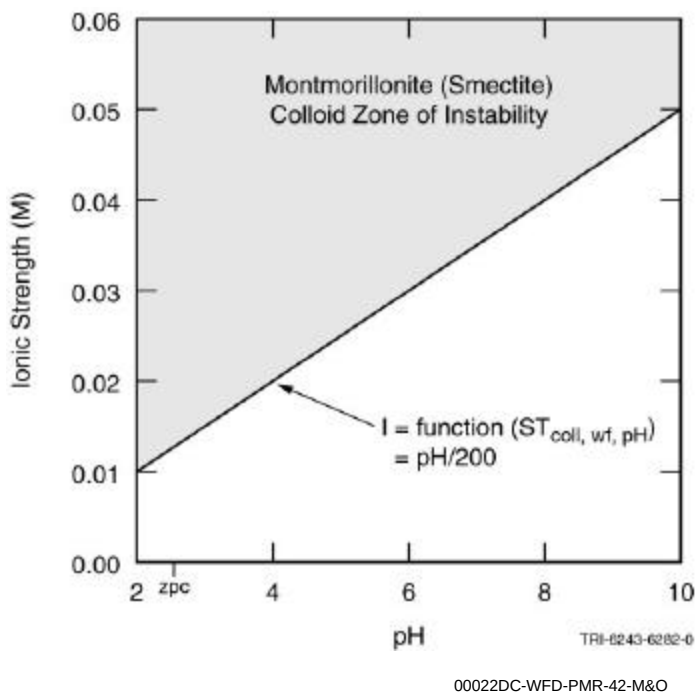
NOTE: Schematic relationship between radionuclide-bearing colloid concentration and ionic strength. The function represents the bound of the HLW degradation experimental data; the maximum value, 6 x 10^{-8} M, is the maximum concentration of colloids observed in the HLW experiments. (Refer to Table 3.8-1 for a description of notation and parameter names and values.)

Source: *Waste Form Colloid-Associated Concentration Limits: Abstraction and Summary*. ANL-WIS-MD-000012. (CRWMS M&O 2000m, Figure 13)

Figure 3.8-4. Generation of Waste-Form Colloids; Abstraction



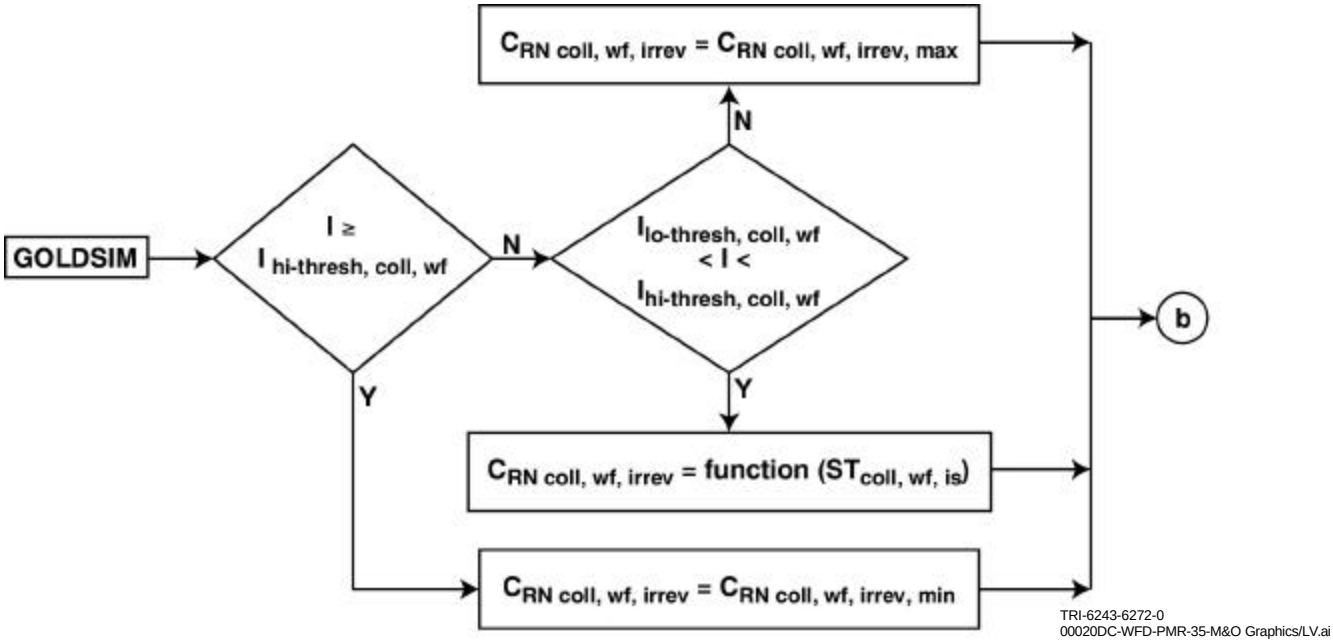
(a) Experimental determination of montmorillonite stability as a function of pH and ionic strength. Data points represent combinations of ionic strength and pH at which montmorillonite stability is significantly decreased. Data from Tombacz et al. (1990).



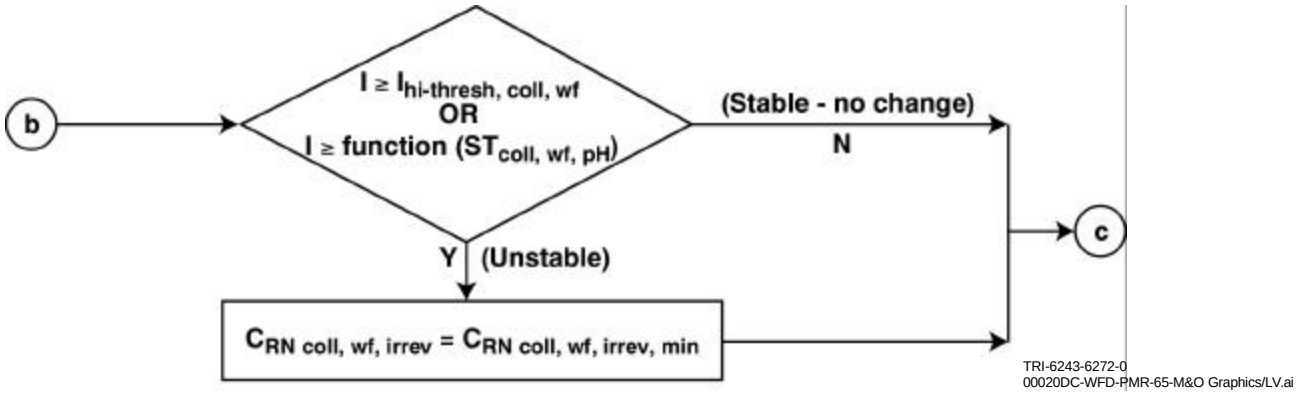
(b) Schematic representation of smectite colloid stability as a function of pH and ionic strength. (Refer to Table 3.8-1 for a description of notation and parameter names and values.)

Source: *Waste Form Colloid-Associated Concentration Limits: Abstraction and Summary*. ANL-WIS-MD-000012. (CRWMS M&O 2000m, Figures 5 and 12)

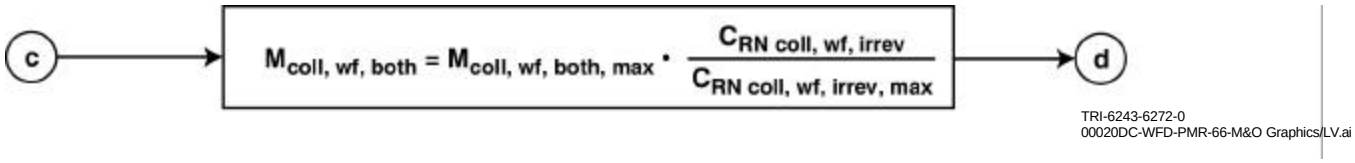
Figure 3.8-5. Stability of Smectite Colloids; Data and Abstraction



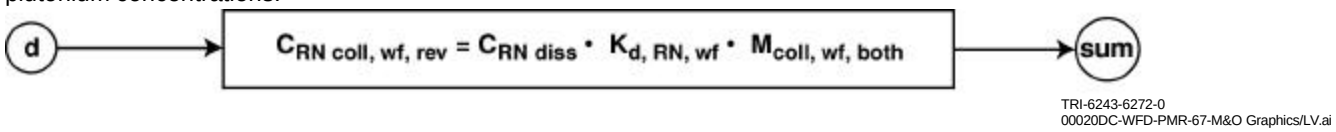
(a) Effect of ionic strength on steady-state concentration of waste-form colloids, based on colloidal plutonium concentrations as shown in Figure 3.8-5.



(b) Effect of ionic strength and pH on stability of waste-form colloids, based on behavior of smectite colloids as shown in Figure 3.8-5.

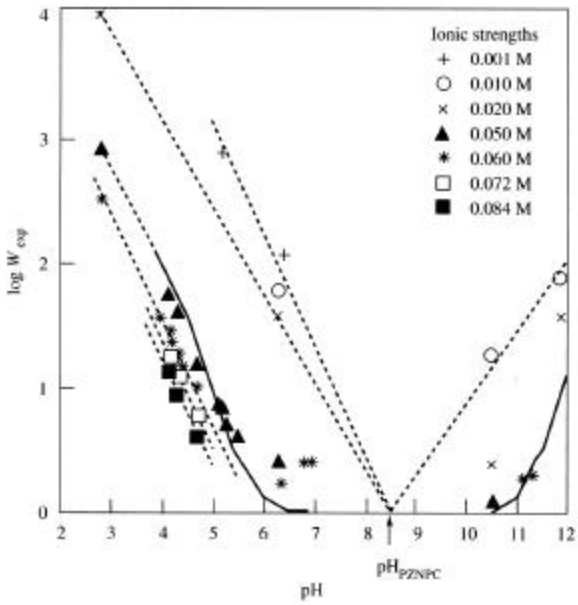


(c) Determination of mass concentration of waste-form colloids, scaled on the basis of maximum and calculated plutonium concentrations.



(d) Calculation of reversibly sorbed radionuclide concentration. (Refer to Table 3.8-1 for a description of notation and parameter names and values.)

Figure 3.8-6. Flow Chart Diagram Depicting Model Implementation for Waste-Form Colloids



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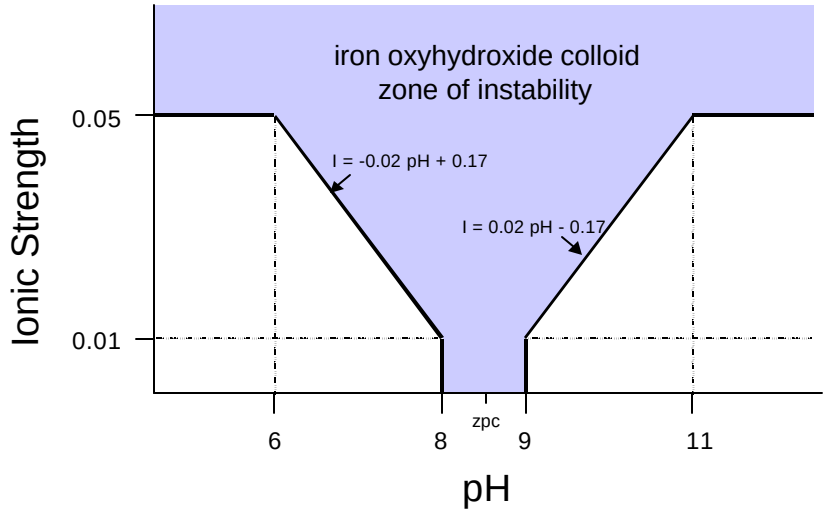
(a) Experimentally derived stability ratio, W_{exp}, of a hematite suspension plotted as a function of pH for differing ionic strengths. The pH of zero proton condition is indicated. Dashed lines are drawn through the experimental points as a guide. Figure and caption from Liang and Morgan (1990, Figure 1). Note that W_{exp} represents the stability of the dispersion, in terms of the rate at which colloidal particles in the dispersion agglomerate. A high value indicates rapid agglomeration; a value of zero represents a stable dispersion. At ionic strengths of about 0.5 m or more, the stability ratio is very low between pH values of about 6 and 11. A relatively low log W_{exp} of approximately 0.2 was selected for abstraction of pH and ionic strength.

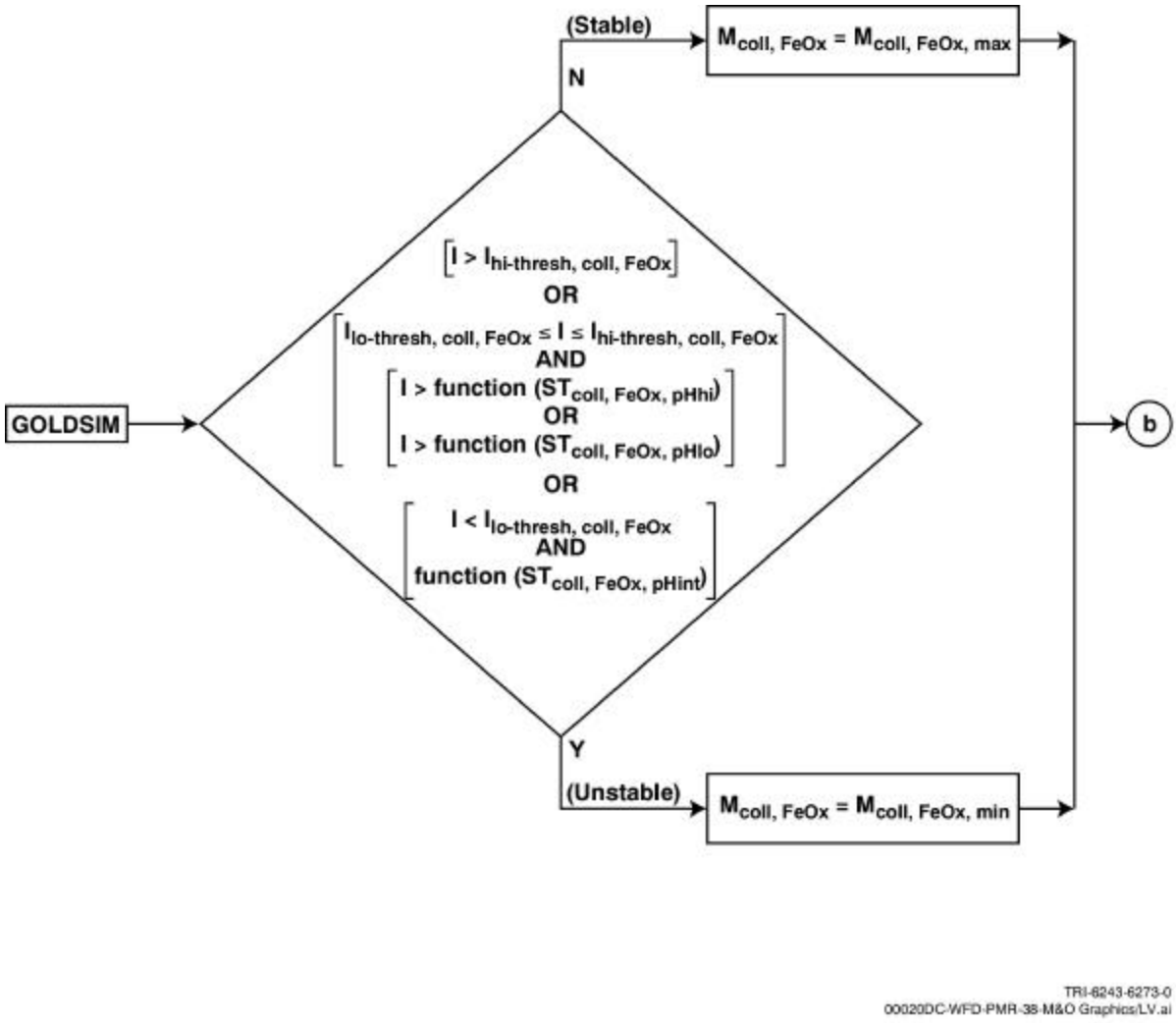


00022DC-WFD-PMR-62-M&O Graphics/LV.doc

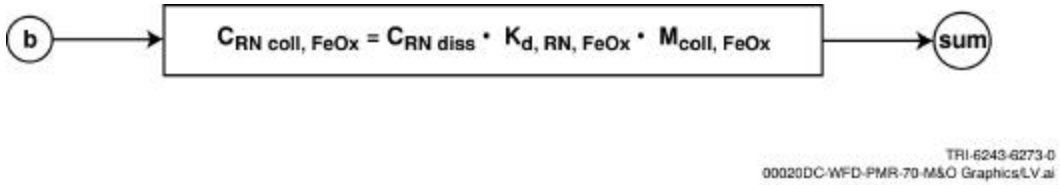
(b) Schematic representation of iron-(hydr)oxide colloid stability as a function of ionic strength and pH. At and near the zero-point-of-charge, colloids are unstable, even at low ionic strengths. At higher ionic strengths the pH range at which colloids are unstable is greater. Above an ionic strength of 0.05 m, colloids are assumed to be unstable at all pH values. (Refer to Table 3.8-1 for a description of notation and parameter names and values.)

Source: *Waste Form Colloid-Associated Concentration Limits: Abstraction and Summary*. ANL-WIS-MD-000012. (CRWMS M&O 2000m, Figures 6 and 11)

Figure 3.8-7. Stability of Corrosion-Product Colloids; Data and Abstraction

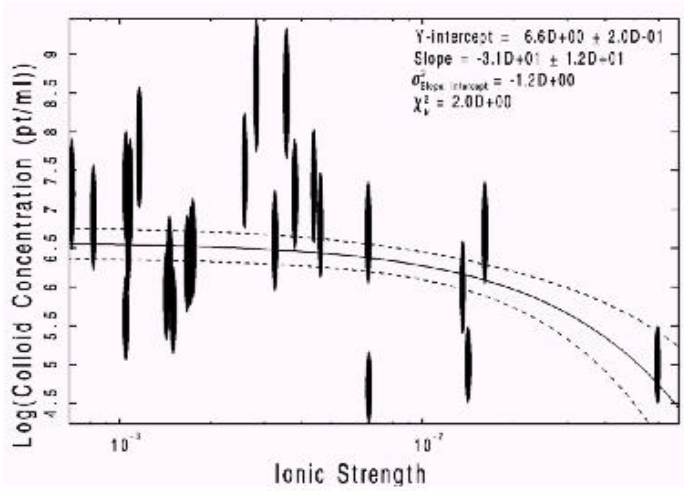


(a) Effect of ionic strength and pH on stability of corrosion-product colloids, based on behavior of iron-(hydr)oxide colloids as shown in Figure 3.8-7.

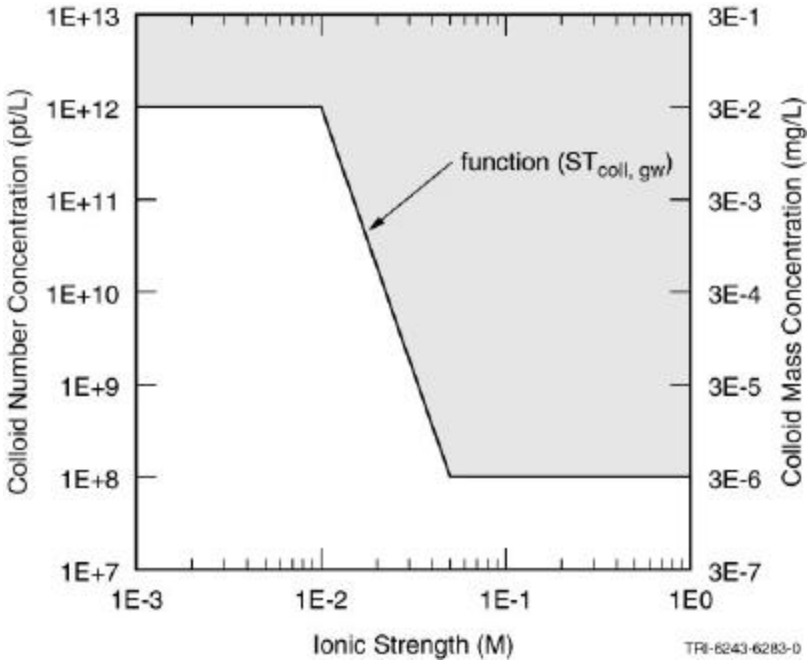


(b) Calculation of reversibly sorbed radionuclide concentration. (Refer to Table 3.8-1 for description of notation and parameter names and values.)

Figure 3.8-8. Flow Chart Diagram Depicting Model Implementation for Corrosion-Product Colloids



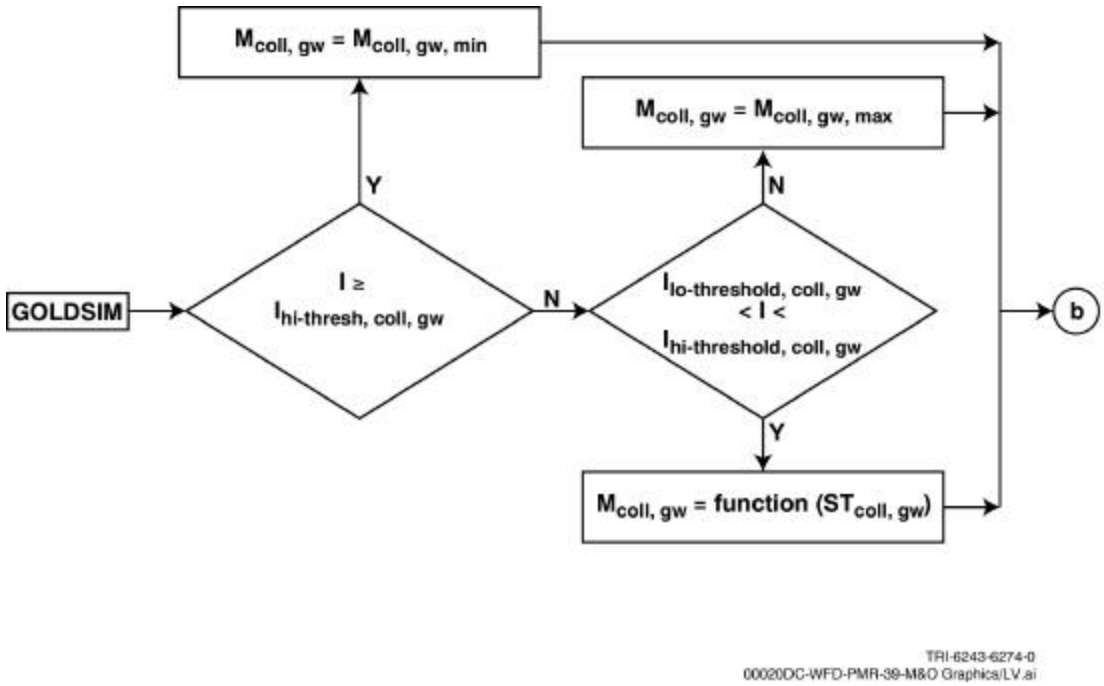
(a) Colloid concentration plotted as a function of ionic strength. Error bars denote uncertainty in concentration values.



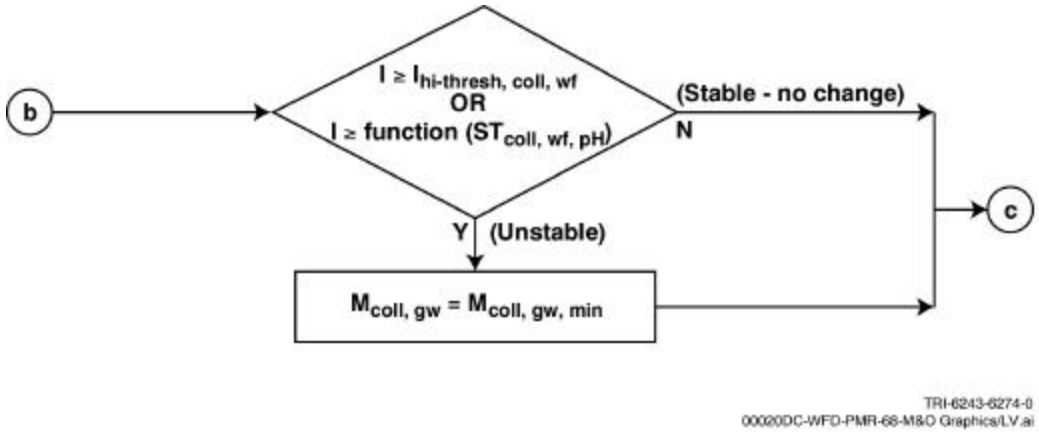
(b) Schematic representation of groundwater colloid mass concentration as a function of ionic strength. (Refer to Table 3.8-1 for a description of notation and parameter names and values.)

Source: *Waste Form Colloid-Associated Concentration Limits: Abstraction and Summary*. ANL-WIS-MD-000012. (CRWMS M&O 2000m, Figure 9 for (a) and Figure 14 for (b))

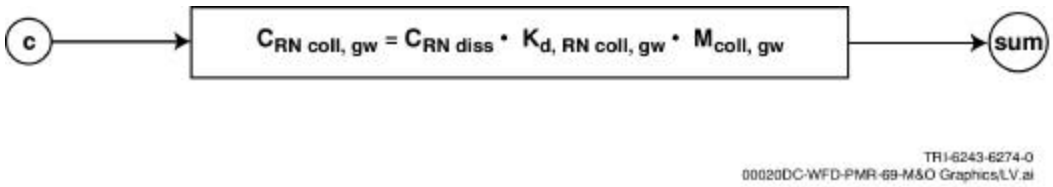
Figure 3.8-9. Concentration of Groundwater Colloids; Data and Abstraction



(a) Effect of ionic strength on the mass of mobile naturally occurring groundwater colloids as shown in Figure 3.8-4.

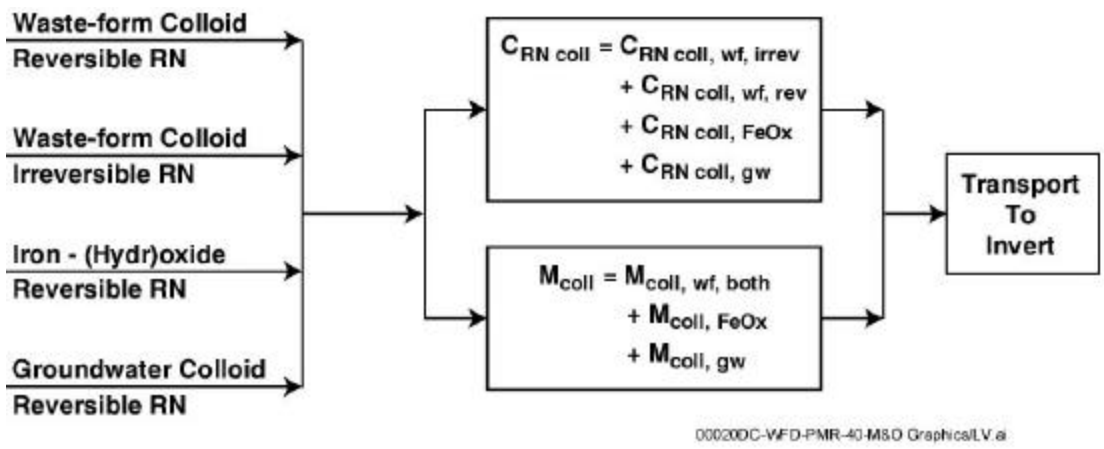


(b) Effect of ionic strength and pH on stability of waste-form colloids, based on behavior of smectite colloids as shown in Figure 3.8-5.



(c) Calculation of reversibly sorbed radionuclide concentration. (Refer to Table 3.8-1 for description of notation and parameter names and values.)

Figure 3.8-10. Flow Chart Diagram Depicting Model Implementation for Groundwater Colloids



NOTE: Summation of the concentration of radionuclides associated with waste-form colloids (both reversibly and irreversibly attached), corrosion-product colloids, and groundwater colloids. Also, summation of the mass of the three types of colloids. (Refer to Table 3.8-1 for description of notation and parameter names and values.)

Figure 3.8-11. Flow Chart Diagram Depicting Model Implementation for all Colloids