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The Analytical Repository Source-Term (AREST) Model: Analysis of Spent Fuel as a Nuclear Waste Form

M. J. Apted
A. M. Liebetrau
D. W. Engel

February 1989

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THE ANALYTICAL REPOSITORY SOURCE-TERM
(AREST) MODEL: ANALYSIS OF SPENT
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SUMMARY

The purpose of this report is to assess the performance of spent fuel as a final waste form. The release of radionuclides from spent nuclear fuel has been simulated for the three repository sites that were nominated for site characterization in accordance with the Nuclear Waste Policy Act of 1982. The simulation is based on waste package designs that were presented in the environmental assessments prepared for each site. Five distinct distributions for containment failure have been considered, and the release for nuclides from the UO_2 matrix, gap (including grain boundary), crud/surface layer, and cladding has been calculated with the Analytic Repository Source-Term (AREST) code.

Separate scenarios involving incongruent and congruent release from the UO_2 matrix have also been examined using the AREST code. Congruent release is defined here as the condition in which the relative mass release rates of a given nuclide and uranium from the UO_2 matrix are equal to their mass ratios in the matrix. Incongruent release refers to release of a given nuclide from the UO_2 matrix controlled by its own solubility-limiting solid phase. Release of nuclides from other sources within the spent fuel (e.g., cladding, fuel/cladding gap) is evaluated separately from either incongruent or congruent matrix release.

Based on current data, congruent UO_2 matrix release is considered to be the anticipated condition at all repository sites evaluated in this report. Releases for individual radionuclides have been evaluated relative to compliance with allowable U.S. Nuclear Regulatory Commission (NRC) fractional release rates. Additional comparison was made to the U.S. Environmental Protection Agency (EPA) 10,000-year cumulative release limits. Although the EPA limits are to be applied at the repository boundary to the accessible environment, it is believed that such a comparison could provide a further meaningful perspective on the performance of spent fuel as a waste form.

For the anticipated condition of congruent matrix release, spent fuel is predicted to meet EPA and NRC release criteria for the distribution of containment failures considered in this report. However, if incongruent matrix

release is assumed, several nuclides are predicted to be released above their NRC limits. The congruent/incongruent nature of UO_2 release is potentially the most significant performance factor that needs to be conclusively established for all disposal conditions. The distribution of containment failures is another performance factor that particularly affects the release rate of readily soluble, inventory-limited nuclides contained in gap and grain boundary sources within spent fuel.

Subsequent to the completion of these calculations, the Nuclear Waste Policy Act (NWPA) was amended in December 1987, so that only the tuff site of the Nevada Nuclear Storage Investigations Project remains as a candidate site. This study and its results are structured solely to evaluate the performance of spent fuel as a waste form. Because of this, it is stressed that these results cannot and should not be used to compare the relative or absolute performance of any geological site for nuclear waste disposal. The purpose of the study was to identify key parameters affecting the performance of spent fuel as a waste form. Given the preliminary nature of the input parameters and assumptions used, it is notable that the key compliance factors for spent fuel were identical for all three geological sites that were used in this evaluation.

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1.0 INTRODUCTION

The U.S. Department of Energy (DOE) is studying the permanent disposal of high-level radioactive waste and spent nuclear fuel in deep geologic repositories. Compliance with the requirements of the U.S. Environmental Protection Agency (EPA) and the U.S. Nuclear Regulatory Commission (NRC) for the safe isolation of high-level waste and spent fuel must be demonstrated. The demonstration of compliance will rely, in part, on an evaluation of the performance and effectiveness of the engineered and natural barriers that provide the containment and isolation of the radioactive materials (Alexander et al. 1985; Apted et al. 1985).

Spent nuclear fuel from civilian reactors is planned to be the dominant waste form for disposal in a U.S. repository. Spent fuel can be considered an assembly of separate materials, including the UO_2 fuel matrix and cladding, and its encasing cladding. In addition to the UO_2 matrix and cladding, different regions within the spent fuel can be identified (Figure 1.1). Spent fuel is inhomogeneous and contains several readily soluble, volatile nuclides (e.g., ^{14}C and ^{129}I) that are not present in the waste forms resulting from the reprocessing of spent fuel (e.g., borosilicate glass). This characteristic has raised certain questions regarding the performance of spent fuel as a waste form (NAS 1983).

Preliminary site-specific calculations by the DOE's Office of Geologic Repository (OGR) projects (DOE 1986a, 1986b, 1986c), however, indicate that radionuclide releases from engineered and natural barrier systems containing spent fuel are expected to comply with NRC and EPA requirements. The DOE requested the Performance Assessment Scientific Support (PASS) program to conduct an additional, independent evaluation of the performance of spent fuel as a waste form. The Analytic Repository Source-Term (AREST) was developed and used by the PASS program to evaluate the performance of spent fuel as a waste form.

The purpose of this report is to provide a quantitative assessment of the performance of spent nuclear fuel as a waste form in geologic repositories. The results of this assessment will be useful for determining the

FIGURE 1.1. Schematic Cross-Section of Spent Fuel with Separate Sources of Radionuclides Identified

relative importance of performance parameters and for deciding what further information is required. This assessment depends upon assumptions about the quantitative release requirements that are applied, the specific characteristics of spent fuel that are assessed, and the geologic repository sites that are considered.

1.1 EVALUATION OF SPENT FUEL AS A WASTE FORM

In May 1986, three sites were nominated for further site characterization in accordance with the Nuclear Waste Policy Act of 1982 (NWP 1983).

These sites are the Yucca Mountain Site, Nevada Research and Development Area, Nevada; the Hanford Site, Reference Repository Location, Washington; and the Deaf Smith County Site, Texas. These sites are geologically diverse and present a wide range of environmental characteristics; accordingly, this report assesses the performance of spent fuel as a waste form at each of these sites. In this report, the repositories will be referred to by the repository project names: the Yucca Mountain Site will be known as the Nevada Nuclear Waste Storage Investigation (NNWSI) Project, the Hanford Site will be known as the Basalt Waste Isolation Project (BWIP), and the Deaf Smith County Site will be known as the Salt Repository Project (SRP). Subsequent to the completion of these calculations, the Nuclear Waste Policy Act (NWPA) was amended in December 1987, so that only the tuff site of the Nevada Nuclear Storage Investigations Project remains as a candidate site.

The evaluations described in this report adopt the waste package designs and repository data presented in the environmental assessment reports (EAs) that were prepared for each site chosen for further characterization (DOE 1986a, 1986b, 1986c). The information in the EAs was selected for use in the AREST evaluation because

- a similar level of detailed analysis is presented for each site
- specific sets of data and designs are presented at a common, fixed time
- current, relevant information on spent fuel and its performance as a waste form is presented.

The authors emphasize that this report presents an evaluation of the performance of spent fuel as a waste form, not an evaluation of any one repository site or waste package design. The use of the spent fuel analysis presented in this report cannot be used to make relative performance comparisons of the three nominated repository sites for the reasons outlined in the following paragraphs.

First, a single standardized performance assessment approach is used in this report for all three repository sites. For the purposes of

evaluation using AREST, this assessment approach is an advantage. Performance characteristics are treated identically at all sites, so that the spent fuel performance itself is evaluated rather than the characteristics of an individual site. Moreover, the mass-transfer theory upon which this assessment is based is applied conservatively and defensibly at each site. The disadvantage to this assessment approach is that it may incorporate certain assumptions that are not equally conservative for all sites, and it may ignore certain favorable conditions that justify a different site-specific approach to waste package performance assessment for licensing purposes.

Second, any meaningful comparison of site performance would necessarily require the assumption of identical waste package designs. Such an approach would contradict the explicit and intended differences in existing waste package designs that are based on different repository environments. Therefore, analyses to compare sites would require the unrealistic and arbitrary use of a single waste package design. Separate waste package designs were used in these analyses instead, which invalidates their application in comparing repository sites.

Third, waste package designs and repository-specific data from the site EAs are not the complete or final designs and data that would be presented in a license application. As noted in Section 3.2 on regulatory requirements, compliance with release limits is a function of both waste form characteristics and waste package design. A finding of potential noncompliance by spent fuel to meet radionuclide release limits does not mean spent fuel is an inadequate waste form. Rather, it may suggest that a revised waste package design should be adopted; that the data used for the assessment may be deficient; or perhaps, that less conservative and more realistic assumptions and processes should be included in a revised assessment. At this early stage of repository design, the purpose of performance assessment analyses such as those reported here is to identify and guide potential design modifications.

Finally, most of the site characterization work has not yet been conducted. The results of the performance assessment calculations reported here depended on site characterization data and, in particular, on the assumed spatial and temporal variation of site parameters. Establishing reasonable

assurance of regulatory compliance may include assessments using a probabilistic approach (51 FR 22288-22301; Sastre, Pescatore, and Sullivan 1986; 40 CFR 191, 1985). The limited sets of data and assumed distributions of that data restrict the degree to which reasonable assurance can be provided before a final preliminary performance assessment. Therefore, the use of these results should be restricted to the purpose for which this evaluation was conducted: assessing the performance of spent fuel as a waste form in geologic repositories.

1.2 ASSESSMENT APPROACH

The performance of spent fuel as a waste form was evaluated using the AREST code (Liebetrau et al. 1987). The AREST code contains three principal models: the waste package containment (WPC) model, the waste package release (WPR) model, and the engineered system release (ESR) model. These models are described more fully by Liebetrau et al. (1987).

The WPR model, based on the mass transfer models of Pigford, Chambré, and coworkers (Chambré et al. 1985; Kim, Chambré, and Pigford 1986) provides the deterministic basis for calculating radionuclide release from individual waste packages containing spent fuel. The WPC model simulates waste package failure times, either from corrosion models or user-defined distributions. These release and containment failure submodels are coupled to the ESR model, which integrates the release from individual waste packages to provide a probabilistically generated assessment of radionuclide release from the total system. Thus, the expected ranges of repository and waste package variables, based on preliminary information and designs for U.S. repository sites, are translated into a probabilistic measure of spent fuel performance. This probabilistic measure of performance can only incorporate the known and quantifiable sources of variation in process models and input variables. For the purposes of this report, spent fuel performance is assessed relative to compliance with the regulatory release rate and cumulative release limits applied to the waste package boundary, as discussed in Section 3.2.

It must be stressed that the AREST code represents a performance assessment tool for preliminary predictions of release rate to guide the overall

program and to focus on technical issues that need resolution. There are still many limitations in the present version and analyses of the AREST code, as discussed in Sections 2.1 and 4.4 of this report. In particular, the use of diffusional mass transfer analysis for release from spent fuel into a tuff host rock is an unexpected, but necessary, assumption that is built into the current version of the AREST code. Later versions of the AREST code are being modified to include a release model based on contact between spent fuel and a drop (or film) of water under unsaturated hydrological conditions. The AREST code and analyses, therefore, will evolve over time, attaining a greater degree of realism and complexity based on an expanding data base.

1.3 REPORT ORGANIZATION

Section 2.0 summarizes the limitations, conclusions, and recommendations of the evaluation of spent fuel performance. Release requirements and the specific characteristics of spent fuel are discussed in Section 3.0. Inputs for spent fuel assessment are described in Section 4.0. Section 5.0 contains a summary of assumptions used in the spent fuel assessment. The spent fuel release rate analyses are discussed in Section 6.0, and the results and discussion for the release rate analyses for the repository sites are presented in Sections 7.0 (BWIP), 8.0 (NNWSI), and 9.0 (SRP). The probabilistic distributions of nuclide release rates at all three repositories are presented in Section 10.0.

2.0 CONCLUSIONS AND RECOMMENDATIONS

This report describes the results of using the AREST performance assessment code to evaluate the performance of spent nuclear fuel as a waste form in geologic repositories. The release of radionuclides from spent fuel has been simulated for three different repository sites and waste package designs. Five distinct distributions for containment failure have been considered, and the release of nuclides from the UO_2 matrix, gap (including grain boundary), crud/surface layer, and cladding has been calculated. Separate scenarios involving incongruent and congruent release from the UO_2 matrix have also been examined.

Calculated releases for individual radionuclides have been evaluated for compliance with allowable release limits (10 CFR 60, 1983; 40 CFR 191, 1985). Compliance with EPA release limits at the boundary between the waste package (engineered barrier system) and the host rock is not required by the EPA or the NRC. It is believed, however, that this comparison against EPA limits imposed at a boundary that is much more restrictive than the accessible environment boundary can add a meaningful perspective on the performance of spent fuel as a waste form.

The limitations of this analysis of spent fuel performance as a waste form, the conclusions drawn, and recommendations for increasing the confidence in the conclusions are summarized in the following sections.

2.1 LIMITATIONS OF SPENT FUEL ANALYSIS

The assumptions adopted for this analysis of spent fuel performance are presented in Section 5.0. The selection of different assumptions would change the details of this analysis; however, because many of these assumptions are well supported by experimental evidence or are demonstrably conservative, it is believed that the general conclusions of this report would remain unchanged.

The most important aspects of any predictive simulation model, such as AREST, are the technical defensibility of its approach and the quality of the data upon which inputs are based. The mass-transfer approach used in the

AREST code is based on the work by Drs. Chambré and Pigford, and colleagues at the University of California, Berkeley (see Chambré et al. 1985). The specifics of the mass-transfer equations used in these analyses are discussed and documented in Section 2.4 and Appendixes D and E of Liebetrau et al. (1987). Limitations of the present code and needed model improvements are discussed in Section 5.0 of Liebetrau et al. (1987). Section 4.0 of this report compares the AREST approach with other, site-specific approaches to waste package performance assessment.

By choice and necessity, the data used for this analysis of spent fuel as a waste form are derived from available information published by the repository projects and are, therefore, preliminary and limited. These data have not been reviewed by the PASS program for their correctness or adequacy.

For example, the solubilities of radionuclides quoted by the SRP (DOE 1986a) are based on "generic" values proposed by the National Academy of Sciences (NAS 1983) in their review of geologic disposal of high-level radioactive wastes. The relevance of the NAS solubility values to a repository in salt is assumed but has not been demonstrated by the SRP. By comparison, the solubilities quoted by the BWIP (DOE 1986b) and NNWSI (DOE 1986c) are based on "thermodynamic" or laboratory studies for their site-specific conditions. Use of the same NAS solubility values, in place of the site-specific values, would lead to steady-state, long-term fractional release rates for waste packages in basalt and tuff that were similar to those calculated for salt. Furthermore, NNWSI has cited several conservatively large solubility values because their approach to release-rate calculation for waste package performance assessment is currently a bulk-flow saturation model (i.e., the release rate is equal to the product of solubility times extremely low annual groundwater flow). These solubility values were conservative, preliminary estimates and it is expected from existing literature that smaller values will be confirmed through later testing. As noted in Section 4.0, this approach is different from the diffusional transport models used in this report.

Another limitation of the data used in this report is the absence of information on the natural variation of site-specific variables. For example, groundwater compositions at all three repository sites undoubtedly

vary laterally and vertically within the host rock. The variations in compositional parameters such pH, carbonate, phosphate, or Eh (redox potential) can, in turn, have pronounced effects on the solubility of radionuclide-bearing solids. The BWIP is the only program that has considered a range of solubilities (DOE 1986b; Yung et al. 1987), and this range is currently attributable more to variation or uncertainty in laboratory measurements than to spatial variation in environmental variables, such as groundwater composition or temperature.

Another limitation, associated with the previous one, concerns the lack of data or models to relate the ranges in environmental parameters to ranges in nuclide-specific parameters, such as solubility or sorption. It may be that the expected range in these environmental parameters has only a small, negligible effect on solubilities, but such supporting data are lacking for most radionuclides. A key exception is uranium, for which a solubility function that expresses the solubility of UO_2 as a function of groundwater composition and temperature over a limited range of conditions has been derived (Garisto and Garisto 1985). This solubility function is crucial, because the expected congruent release of nuclides from the matrix is usually assumed to be controlled by UO_2 solubility (Johnson, Garisto, and Stroes-Gascoyne 1985; Wuertz and Ellinger 1985; Garisto and Garisto 1985; Forsyth and Werme 1985; Pigford and Chambré 1986; LeNeveu 1986; Kim, Chambré, and Pigford 1986). It is notable that results from the AREST code indicate that UO_2 solubility is relatively insensitive to the expected range in groundwater chemistry and temperature calculated for the broad range of initial heat loadings adopted in this report (Liebetrau et al. 1987, Appendix B). This can be attributed to chemical buffering reactions within the localized environment of a waste package that restrict groundwater composition to a relatively limited range, regardless of initial composition or expected temperatures. Variation in reversible sorption coefficients for nuclides because of ranges in environmental variables is not a crucial issue. This is because reversible sorption has a relatively minor effect on waste package radionuclide release rates, particularly if containment times of greater than 300 years are achieved.

An important limitation cited in Section 1.2 of this report is that waste package designs have not been optimized or finalized. For example, the

inclusion of packing materials, changes in metallic container material, and addition of an internal stabilizer within the container could lead to releases significantly different than those calculated in this report. Likewise, the data used in this report are preliminary and often incomplete. The purpose of performance assessment modeling at this stage must be to identify potential problems and guide design modification.

A final limitation to this analysis is the meager amount of available information on spent fuel that is relevant to release rate assessment. The importance of characterizing the separate radionuclide sources within spent fuel has only recently been appreciated and addressed. The variation in properties of spent fuel with different reactor histories is also largely unknown relative to any potential effect on release of nuclides. Performance assessment modeling has an important role to play in guiding and defining the work necessary to establish confidence in spent fuel as a waste form.

2.2 CONCLUSIONS

Results of the separate summaries of spent fuel performance for specific repository sites have been compiled and are tabulated in Table 2.1. When evaluating the meaning of these results, three caveats should be considered:

1. Congruent UO_2 matrix release is the anticipated condition for all repository sites, although additional confirmatory experiments are needed to unequivocally establish this assumption. A review of the available laboratory results supporting congruent dissolution of the UO_2 matrix of spent fuel and the stability of UO_2 in the presence of waste package barrier materials is given in Section 5.0 of this report. The potential for enhanced release arising from precipitation of a low-solubility, uranium-bearing solid is not examined in this report.

Modification of AREST mass transfer equations to model this condition are in progress. Note, however, that precipitation-induced enhancements in release rates by an extreme factor of 40 (Garisto

TABLE 2.1. Summary of Nuclides Released Above Regulatory Limits from Waste Packages Containing Spent Fuel

Assumed Behavior of Containment		BWIP		NNWSI		SRP	
		NRC ^(b,c)	EPA ^(b)	NRC	EPA	NRC	EPA ^(d)
Case 1: (Instantaneous failure at 300 years)	I ^(a)	Cs, Cs*, I, Ra Se, Sr, Tc	Am	C, Cm, Cs, I, Np Ra, Se, Tc	Am, Pu, Pu* C, Np, Cm, Te, Am*	C, Cs, Cs*, I, Se, Sr, Tc	Sum ^(d)
	C ^(a)	Cs*, Se, Sr, Tc	--	C, Cs, I, Se, Tc	--	C, Cs, Cs*, I, Se, Sr, Tc	--
Case 2: (Instantaneous failure at 1000 years)	I	Cs, I, Ra, Se, Tc	Am	C, Cm, Cs, I, Np, Ra, Se, Tc	Am, Pu, Pu* C, Np, Cm, Te, Am*	C, Cs, I, Se, Tc	Sum
	C	Se, Tc	--	C, Cs, I, Se, Tc	--	C, Cs, I, Se, Tc	--
Case 3: (Normal distribution of failures centered at 1000 years)	I	Cs, I, Ra	Am	C, Cm, Cs, I, Np, Ra Se, Tc	Am, Pu, Pu* C, Np, Cm, Tc, Am*	Cs, I	Sum
	C	--	--	--	--	--	--
Case 4: (Normal distribution of failures centered at 5000 years)	I	Cs	Am	C, Cm, Cs, I, Np, Ra, Se, Tc	Am, Pu, Pu* C, Np, Cm, Tc	Cs, I	Sum
	C	--	--	--	--	--	--
Case 5: (Exponential distribution of failures beginning at 300 years)	I	Cs, I	Am	C, Cm, Cs, I, Np Ra, Se, Tc	Am, Pu, Pu* C, Np, Cm, Tc, Am*	Cs, I	Sum
	C	--	--	--	--	--	--

(a) I = incongruent matrix release; C = congruent matrix release.

(b) NRC = fractional release rate limit; EPA = cumulative release limit as applied at boundary to accessible environment.

(c) C = ^{14}C , Cm = ^{245}Cm , Cs = ^{135}Cs , Cs* = ^{137}Cs , I = ^{129}I , Np = ^{237}Np , Ra = ^{226}Ra , Se = ^{79}Se , Sr = ^{90}Sr , Tc = ^{99}Tc , Am = ^{241}Am , Am* = ^{243}Am , Pu = ^{239}Pu , Pu* = ^{240}Pu

(d) Sum = sum of individual release ratios.

and Garisto 1986) would still leave the calculated rates of congruently released nuclides orders of magnitude below NRC and EPA limits for all sites considered.

2. Some distribution of containment failure seems certain for all repository sites (KBS 1983; DOE 1986b; LeNeveu 1986; Yung et al. 1987). The instantaneous failure cases, Cases 1 and 2, are included as a bounding analysis. How closely the actual distribution of containment failures will approach the instantaneous case is unknown. Determining this will require the development of a defensible prediction model for containment time and, perhaps, attaining laboratory and field measurements to support the prediction of containment failure distributions. Resolution of

these important issues, however, lies outside the intended scope of this report. This study assesses the performance of spent fuel as a waste form in meeting release objectives under anticipated conditions (10 CFR 60, 1983; 40 CFR 191, 1985). Substantial release of nuclides prior to 300 years after repository closure would mean failure to meet the NRC containment objective, which is considered an unanticipated event and is not evaluated here.

3. The approach for calculating release with the AREST code taken in this study does not necessarily treat all sites equally. In particular, the waste package site of the NNWSI Project is designed to have a sustained air gap between the unsaturated host rock and the waste package. The conservative assumption of a continuous diffusional pathway between package and tuff, as treated in this report, may be an unanticipated condition requiring a revised set of release criteria (Section 2.1).

From the caveats and the results of Table 2.1, the following general conclusions have been reached regarding the performance of spent fuel as a waste form in a geologic repository:

- The dominant factor in performance of spent fuel as a waste form is whether the UO_2 matrix releases radionuclides incongruently (i.e., release controlled by the solubility of individual radionuclide-bearing solids) or congruently (i.e., release control by the solubility of the UO_2 matrix). Congruent matrix release rates are up to 10 orders of magnitude lower than release rates calculated for incongruent matrix release. This result applies to all the repository sites and waste package designs that have been evaluated with the stated assumptions. Incongruent matrix dissolution would apparently lead to the release of several nuclides above their NRC limits, notably ^{135}C and ^{129}I .
- The cumulative release from spent fuel of all nuclides, assuming congruent matrix release, complied with their EPA limits at all three repository sites for all conditions of containment failure

that were considered. This includes the case of instantaneous failure of all waste packages 300 years after repository closure (Case 1).

- The fractional release rates of all nuclides are below their NRC limits for cases based on congruent UO_2 matrix dissolution and the distributed containment failures considered. This includes failure distributions in which 50% of the waste packages have lost containment 1000 years after repository closure (Cases 3 and 5).
- Based on the previous two points, it is concluded that the EPA cumulative release limits for individual nuclides (intended to be applied kilometers from the waste package) are in many cases more easily met than the NRC fractional release rates intended to be applied at the engineered barrier system boundary for the same nuclides.
- For the incongruent matrix release scenarios, the highest fractional release rates and cumulative releases are attributable to release from the matrix, followed by contributions from the gap and cladding surface layer. Congruent matrix release scenarios lead to large reductions in matrix release rates so that the unchanged release rates from the gap and cladding surface layer constitute the highest fractional release rates and cumulative releases.
- The primary effect of distributed containment failures is to diminish and delay the spike-like maxima in fractional release rates that are observed in Cases 1 and 2 where instantaneous containment failure is assumed. The maximum release rates from the gap and cladding surface layer were significantly below NRC limits for the distributions of containment failure considered in this study. For matrix nuclides, the effect of distribution of container failure is relatively less important than the effect of whether matrix release is congruent or incongruent.
- Short-lived nuclides that are readily soluble, such as ^{137}Cs and ^{90}Sr , can be dismissed from release rate calculations on the basis of the NRC inventory cutoff at 1000 years. These nuclides can,

however, dominate the release rates on a curie basis if a significant fraction of all waste packages lose containment prior to 1000 years after repository closure.

2.3 RECOMMENDATIONS

Three general areas--1) spent fuel characterization and testing, 2) waste package design and data, and 3) programmatic considerations--require work or decisions before this preliminary conclusion of spent fuel as an adequate waste form can be confirmed. Recommendations for further work are briefly described in the following sections. Future developments and enhancements in the AREST code have been recommended in Section 5.0 of Liebetrau et al. (1987).

2.3.1 Spent Fuel Characterization and Testing

The following recommendations apply to spent fuel characterization and testing:

- The congruent release of nuclides from the UO_2 matrix of spent fuel into solutions nearly saturated with respect to UO_2 needs verification.
- The stability of the UO_2 matrix of spent fuel under realistic waste package conditions needs verification.
- The separate sources of nuclides within spent fuel needs to be characterized, the radionuclide content of each source determined, and variability in these distributions established.
- The thermodynamic database used to calculate UO_2 solubility as a function of groundwater composition and temperature needs to be expanded and validated.
- The effect of low-solubility uranium-precipitates on matrix release needs to be analyzed.

2.3.2 Waste Package Designs and Data

The following recommendations apply to waste package designs and data:

- The analysis of spent fuel as a waste form must be updated as revised waste package designs are generated.
- Expected ranges of environmental variables (pH, Eh, groundwater composition, host rock permeabilities, etc.) need to be quantified.

2.3.3 Programmatic Considerations

The following recommendations apply to programmatic considerations:

- Technical bases for containment distributions should be developed according to site- and material-specific constraints.
- The regulatory status and definitive allowable release rates of short-lived nuclides (e.g., ^{90}Sr and ^{137}Cs) under conditions of early (less than 1000 years) containment failure should be clarified. Under these conditions, with certain interpretations, arbitrary normalization of mass transfer rates by the 1000-year inventories of such nuclides as specified by regulations can lead to seemingly unrealistic high fractional release rates above 100% per year.
- The definition of the engineered barrier system boundary for radionuclide release should be clarified; strategies and methodologies for validation of performance assessment models for waste packages should be defined.

3.0 PHYSICAL CHARACTERISTICS AND LEGAL REQUIREMENTS PERTINENT TO THE PERFORMANCE OF SPENT FUEL

The physical characteristics of spent fuel and the federal regulations regarding the acceptable releases of radionuclides are discussed in this section.

3.1 SPENT FUEL CHARACTERISTICS

The assessment of spent fuel as a waste form requires that certain characteristics relevant to performance be identified at the time of disposal. These characteristics, also discussed in Section 4.0, include compositions and dimensions of component materials, radionuclide inventories of components, temperatures, heat generation rates, and cladding integrity. Because several of these spent fuel characteristics vary over time, their values must be specified or calculable for the post-emplacement period of a repository.

Pressurized-water reactor (PWR) fuel has been selected for the assessments in this report because quantities of PWR fuel will dominate those of boiling-water reactor (BWR) fuel in the repository, and PWR and BWR spent fuels display similar radionuclide inventories for similar degrees of burnup (Roddy et al. 1986, Tables 3.6, 3.8, 3.10, and 3.12). This choice is consistent with the requirements of the DOE's Generic Requirements Document (GRD) (DOE 1984), which specifies a characteristic 10-year-PWR fuel with 33,000 MWd/MTU burnup for the purposes of preliminary waste package design analyses.

Variables describing the composition, dimensions, radionuclide inventory, and cladding integrity of the specific PWR spent fuel used for this report are assigned point (single) values. The values for heat generation rate, however, were determined from a distribution that describes the assumed thermal characteristics of the PWR fuel emplaced in the repository. This was done because the heat generation rate determines the time-dependent temperature within the waste package subsystem, and temperature was judged to be an

important parameter affecting both the containment and slow release of radionuclides. The specific values for all of these parameters are presented in Section 4.0.

3.2 RELEASE REQUIREMENTS

Spent fuel performance can be assessed with respect to specific federal regulations regarding acceptable releases of radionuclides from a mined geologic disposal system. These requirements, or performance objectives, include those by the NRC and the EPA. The performance objectives established by these two agencies are applicable to the final safety assessment of spent fuel as a waste form and the licensing of a nuclear waste repository, as discussed in the next section. It is prudent, therefore, to consider the adequacy of spent fuel over the broadest possible set of safety and performance objectives in order to establish its adequacy as a waste form. The NRC and EPA objectives for nuclide release addressed in this evaluation are summarized in the following sections and are portrayed schematically in Figure 3.1. Figure 3.1 also includes the waste package boundary as designated by the DOE's GRD, which interprets the NRC and EPA requirements in its definitions of the mined geologic disposal system, its subsystems, and components. The GRD requirements are internal, baselined DOE requirements, and do not constitute a federal regulation. However, since the GRD governs DOE work, its requirements were also addressed in this assessment of spent fuel performance.

3.2.1 U.S. Nuclear Regulatory Commission 10 CFR Part 60

The NRC has established allowable fractional release rates for nuclides in the period following the 300 to 1000 years during which substantially complete containment is required (10 CFR 60, 1983). Section 60.113(a)(1)(ii)(B) of 10 CFR 60 states that:

"[T]he engineered barrier system shall be designed, assuming anticipated processes and events, so that: . . . (B) The release rate of any radionuclide from the engineered barrier system following the containment period shall not exceed one part in 100,000 per year of the inventory of that radionuclide calculated to be present at 1,000 years following permanent closure, or such

FIGURE 3.1. Schematic Cross Section of Repository Showing Radionuclide Release Boundaries Imposed by NRC, EPA, and DOE

other fraction of the inventory as may be approved or specified by the Commission; provided, that this requirement does not apply to any radionuclide which is released at a rate less than 0.1% of the calculated total release rate limit. The calculated total release rate limit shall be taken to be one part in 100,000 per year of the inventory of radioactive waste, originally emplaced in the underground facility, that remains after 1,000 years of radioactive decay."

There are several important considerations in this NRC performance objective. First, the NRC release rate limit is established for "anticipated processes and events," which in turn has been defined (10 CFR 60, 1983) as meaning those natural processes and events that are reasonably likely to

occur during the period in which the intended performance objective must be achieved. It may be assumed that those processes operating in the repository host rock prior to waste emplacement will continue, provided appropriate allowance is made for the perturbations caused by the mining of the repository and the emplacement of the waste.

Second, the NRC release rate limit is imposed at the "engineered barrier system" (EBS) boundary, not at the waste form surface. The EBS is defined (10 CFR 60, 1983) as the waste packages and the underground facility, including openings and backfilled tunnels (see Figure 3.1). This necessarily means that the assessment of spent fuel performance relative to the fractional release rate objective depends on both spent fuel characteristics and the properties of any additional barriers that may be present in the EBS. Because final EBS designs are not complete at any of the sites chosen for characterization, the assessment of spent fuel as a waste form can only be made on a conditional basis at this time.

Third, 10 CFR 60 requires that the release rate objective must be met with "reasonable assurance." Although this term has not been explicitly defined, the NRC has recently indicated (50 FR 22288-22301) that the level of confidence associated with its concept of "reasonable assurance" is the same as that sought by the EPA term "reasonable expectation" (40 CFR 191, 1985). The EPA advocates reliance on a probabilistic approach in which the consequence of an event (e.g. a measurable fractional or cumulative release) is assessed relative to the probability of occurrence of that event (40 CFR 191, 1985). This approach recognizes and incorporates the qualitative judgments regarding the inherent uncertainty and variation in physical and chemical parameters upon which a quantitative performance assessment must be based. Demonstration of "reasonable assurance" will be based on two principles (51 FR 22288-22301): 1) that performance assessment analysis indicates a high (although unquantified) likelihood of meeting EPA and NRC release requirements, and 2) that the performance assessment is sufficiently conservative that actual performance will be within predicted limits.

Fourth, the allowable NRC fractional release rate for individual nuclides depends on the calculated inventory of that nuclide at 1000 years

after permanent closure of the repository (Bensky and Oliver 1985). For nuclides with 1000-year inventories, $I(1000)_i$, greater than 0.1% of the total 1000-year inventory, $I(1000)_T$, the allowable annual fractional release rate is one part in 100,000 (i.e. 10^{-5}) of $I(1000)_i$. For nuclides with $I(1000)_i$ less than 0.1% (10^{-3}) but greater than 10^{-8} of $I(1000)_T$, the allowable fractional release rate is equal to the ratio of $I(1000)_T/I(1000)_i$, multiplied by 10^{-8} . Finally, nuclides with $I(1000)_i$ below 10^{-8} of $I(1000)_T$ can be exempted from the performance assessment analysis entirely. (a) This dependence of allowable release rate on inventory is shown in Figure 3.2.

Lastly, compliance with the NRC performance objective for the specific nuclide release rates, "or such other fraction of the inventory as may be approved or specified by the Commission . . .," must be demonstrated for final licensing of a nuclear waste repository. It is apparent that, although the basis and formulation of the NRC release rate limit have been criticized (10 CFR 40, 1983), the NRC has expressly retained flexibility in applying and accepting alternate measures of safety assessment.

3.2.2 U.S. Environmental Protection Agency 40 CFR Part 191

The EPA has established "containment requirements" that are quantitative limits on the cumulative release of radionuclides to the accessible environment for the first 10,000 years after repository closure (40 CFR 191, 1985). These limits on cumulative release are expressed in curies per 1000 metric tons of heavy metal (MTHM). The actual permissible limits are a function of the probability of the processes and events used in the performance assessment; that is, the permissible limits increase stepwise with decreasing likelihood of occurrence. The EPA requirement states:

-
- (a) The lower inventory cutoff may not be appropriate for release occurring before 1000 years. For example, short-lived nuclides such as ^{137}Cs and ^{90}Sr fall below the inventory cutoff at 1000 years but represent significant fractions of the total curie content of spent fuel 300 years or earlier after repository closure. When evaluating release prior to 1000 years it is not clear that such nuclides can or should be dismissed under the NRC regulations on the basis of their 1000-year inventory.

FIGURE 3.2. Graphical Representation of NRC's 10 CFR Part 60 Definition of Allowable Fractional Release Rates "R" as a Function of 1000-Year Radionuclide Inventory. "TI" = total inventory of 1.08×10^3 Ci/MTU, "I" = individual nuclide inventory, "R" = expression for allowable fractional release rate. The dashed line defines a region of low-inventory nuclides at 1000 years that are exempt from consideration, based on 10 CFR Part 60.

"... the cumulative releases of radionuclides to the accessible environment for 10,000 years after disposal from all significant processes and events that may affect the disposal system shall:

- (1) Have a likelihood of less than one chance in 10 of exceeding the quantities calculated according to Table 1 (Appendix A); and
- (2) Have a likelihood of less than one chance in 1,000 of exceeding ten times the quantities calculated according to Table 1 (Appendix A)."

The NRC has interpreted EPA's term "all significant processes and events that may affect the disposal system" as the equivalent of "anticipated" and "unanticipated" processes and events in the terminology of (10 CFR 60, 1983). Therefore, the EPA containment requirement would apply for the same "anticipated" conditions as the NRC fractional release rate requirement, as well as for the "unanticipated" conditions for which the NRC fractional release rate requirement does not apply. The accessible environment boundary at which the EPA limits apply are considerably removed from the EBS boundary of the NRC and the waste package (WP) subsystem boundary of the GRD (Figure 3.1).

3.2.3 U.S. Department of Energy Generic Requirements Document

The DOE has developed (DOE 1984) a GRD that establishes a consistent baseline of technical requirements necessary to achieve and demonstrate safe geologic disposal of nuclear waste. The GRD requires that:

"The engineered barrier system performance objective of 10 CFR 60.113 is being applied to the waste package subsystem as a goal for design purposes and a boundary for performance assessment at this time."

The WP subsystem is defined in the GRD as the emplaced waste form, container, any other packing, and absorbent materials immediately surrounding an individual waste container. The WP is also defined in the GRD as a "subsystem" of the EBS. Therefore, the WP boundary must lie entirely within, or contiguous with, the EBS boundary (Figure 3.1). Demonstration of compliance at this inner WP boundary will, therefore, assure compliance at the outer EBS boundary.

There are advantages to such a requirement. The GRD release requirement will be conservative with respect to the release requirements of the NRC. In addition, the location of the WP boundary is clearly defined and confirmed; the location of the EBS boundary, although defined, has been variably interpreted. The key issue for interpretation is whether a portion of the host rock/geologic setting can be incorporated into the EBS.

There are also potential disadvantages to the GRD release requirements. Perhaps the main concern is that analyses performed on the basis of the GRD

requirements may be excessively conservative and, thus, misleading. For example, there can be extremely different transport and hydrologic properties between the underground facilities (i.e. backfilled tunnels) and the host rock. Under such conditions some analyses have shown (Yung et al. 1987) relatively high release rates of nuclides from the WP subsystem, although the release rates from the EBS were well below NRC limits. Overly conservative requirements could lead to spurious revisions to WP designs or negative assessments of waste form performance that are not realistic or defensible, both of which could result in needless expense.

3.2.4 Release Requirements Used for Spent Fuel Assessment

The assessment of spent fuel reported here adopts the release rate requirements of the DOE's GRD (DOE 1984). The requirements are based on the NRC release requirements (10 CFR 60, 1983) for anticipated events and processes, but is conservatively imposed at the waste package subsystem boundary rather than at the boundary of the engineered barrier system. In addition, compliance with the cumulative release requirement of the EPA is also assessed at the waste package subsystem boundary (40 CFR 191, 1985). Only anticipated events and processes are considered. Compliance with EPA release limits at this boundary is not required. It is believed, however, that this scoping comparison against the EPA release limits can establish a meaningful perspective on spent fuel performance as a waste form.

3.3 REPOSITORY SITES

In May 1986, three sites were nominated for further site characterization in accordance with the Nuclear Waste Policy Act of 1982 (NWPA 1983). These sites are the Yucca Mountain Site, Nevada Research and Development Area, Nevada; the Hanford Site, Reference Repository Location, Washington; and the Deaf Smith County Site, Texas. The Nuclear Waste Policy Amendments Act of 1987 directed the DOE to characterize only the Yucca Mountain Site, as previously noted. The purpose of this report is to assess the suitability of spent fuel as a waste form, however, not as a waste form in a specific waste package design or site. Thus, since these three sites are geologically

diverse and present a wide range of environmental characteristics, this report assesses the performance of spent fuel as a waste form by considering each of these three sites.

Specifically, the evaluations conducted in this report adopt the waste package designs and repository data presented in the EAs prepared for each site prior to the passage of the Amendments Act of 1987 (DOE 1986a, 1986b, 1986c). There are several reasons for this selection of data sources. First, EAs present an equivalent amount of data, permitting a similar level of detailed analysis for each site. Second, the data and designs in the EAs date from the same time. The EAs, therefore, provide specific sets of data and designs at a common, fixed time. Third, the EAs contain some of the most current information on spent fuel and its performance as a waste form. However, the EAs contained neither the final designs nor the complete data that would be presented in a license application. As noted in the section on regulatory requirements, compliance with release limits is a function of both waste form characteristics and waste package design. A finding of potential noncompliance by spent fuel to meet radionuclide release limits does not mean spent fuel is an inadequate waste form. Rather, it may suggest that a revised waste package design should be adopted; that the data used for the assessment may be deficient; or, perhaps, that less conservative and more realistic assumptions and processes should be included in a revised assessment. At this early stage of repository design, the purpose of performance assessment analyses such as those reported here is to identify and guide potential design modifications.

It must be stressed that this assessment focuses on the performance of spent fuel, rather than the performance of any one site or waste package design. There are several reasons that prevent the legitimate use of the spent fuel analysis in this report for making relative performance comparisons of the three sites addressed in this report. The first reason for not using the results of this assessment to compare the relative performance of sites is that a single, standardized performance assessment approach is used for all three repository sites. There are advantages to this assessment approach. Spent fuel performance is treated identically at all sites, so that spent fuel performance is evaluated rather than the characteristics of

an individual site. Moreover, the mass transfer theory upon which this assessment is based is applied conservatively and defensibly at each site. A disadvantage is that such a single approach may incorporate certain assumptions that are not equally conservative for all sites, and may even ignore certain favorable conditions that justify a different, site-specific approach to waste package performance assessment for licensing purposes.

A second reason for not using this report to evaluate sites is that site characterization data was in a very preliminary state at the time. (The EAs were written, and only the Yucca Mountain site is to be characterized in the future.) The results of the performance assessment calculations reported here depended on site characterization data and, in particular, on the assumed spatial and temporal variation of site parameters. Establishing reasonable assurance of regulatory compliance will require assessments using a probabilistic approach (51 FR 22288-22301, 1986; Sastre, Pescatore, and Sullivan 1986; 40 CFR 191, 1985). The limited sets of data and assumed distributions of that data restrict the degree to which reasonable assurance can be provided in such a preliminary performance assessment. Because these uncertainties are not explicitly evaluated in this report, it is impossible to confidently compare results among the three repository sites. Therefore, the use of these results should be restricted to the purpose for which this evaluation was conducted; that is, assessing the performance of spent fuel as a waste form in geologic repositories.

4.0 INPUTS FOR SPENT FUEL ASSESSMENT

The required inputs for obtaining radionuclide release rates with the AREST code are described in Sections 2.4 and 3.3 of Liebetrau et al. (1987). The four basic categories for input are:

- containment-failure time distributions
- radionuclide data
- spent fuel data
- repository data.

The repository data category is further divided into three parts corresponding to the basalt, tuff, and salt sites. These data are summarized in the following sections.

4.1 CONTAINMENT-FAILURE TIME DISTRIBUTIONS

A contributing factor in the assessment of spent fuel performance as a waste form is the distribution of waste package containment failure times. This report, however, is primarily concerned with the release performance of spent fuel, rather than with the containment performance of the waste package. Concerns relevant to the performance assessment of containment materials and their ability to meet NRC requirements apply generally to waste packages, whether they contain spent fuel or some other nuclear waste form. Such concerns, while important for overall performance assessment of the engineered barrier system, fall outside the scope of this report on assessment of spent fuel as a waste form.

Consequently, the failure time distributions selected for the spent fuel analysis were not derived from simulations involving the WPC model of the AREST code. Such containment distributions have been developed for certain site-specific conditions and waste package designs (KBS 1983; LeNeveu 1986; Yung et al. 1987). Instead of developing a technical basis for site-specific distributions, theoretical distributions were chosen to parametrically represent a range of extreme or limiting containment cases. Therefore, this approach does not distinguish between containment contributions from the container and the cladding. Compliance with the substantially complete containment performance objective of 10 CFR 60 is assumed to be met; no container

failures during the 300 years after repository closure have been considered. The advantage of this approach is that the analyses focus on the performance of spent fuel as a waste, rather than the separate issue of demonstrating substantially complete containment for any waste form. The disadvantage of this assumption is that containment failure prior to 300 years can present special issues regarding the release of short-lived nuclides (Pigford 1986). Furthermore, the use of a fixed set of containment failure distributions ignores the possibility of significantly different containment performance among potential repository sites and designs. This emphasizes the point made earlier that the results of this report only apply to spent fuel assessment and cannot be used to make a valid comparison of sites.

For this report, releases were evaluated for each of five assumed containment failure time distributions. With one exception, these distributions are conservatively chosen to emphasize failures early in the 10,000 year post-closure period. The selected distributions are:

- Case 1. A point (degenerate) distribution which places all containment failures at 300 years
- Case 2. A point (degenerate) distribution which places all containment failures at 1000 years
- Case 3. A normal (Gaussian) distribution with mean $\mu = 1000$ years and standard deviation $\sigma = 200$
- Case 4. A normal (Gaussian) distribution with mean $\mu = 5000$ years and standard deviation $\sigma = 2000$
- Case 5. An exponential distribution with rate $\lambda = 9.902 \times 10^{-4}$, chosen so that the initial 50% of the failures occur between 300 and 1000 years.

Figures 4.1 to 4.5 show 1) the frequency, or density function (i.e., the rate of waste package failures expressed as a function of time) and 2) the cumulative distribution function (i.e., the proportion of waste packages failed at or before a given time) for each of the failure time distributions listed above.

a)

b)

FIGURE 4.1. Hypothetical Point Distribution Used for Containment Failure (Case 1, Failure = 300 years) a) Failure Distribution, b) Cumulative Failure Distribution

The first two distributions were selected as extreme (limiting) cases in which all the waste packages are assumed to fail simultaneously, in one case at 300 years and in the other at 1000 years. Both cases are unrealistic for the entire engineered barrier system, but they do represent the performance of individual waste packages that fail at 300 and 1000 years, respectively.

a)

b)

FIGURE 4.2. Hypothetical Point Distribution Used for Containment Failure (Case 2, Failure = 1000 years) a) Failure Distribution, b) Cumulative Failure Distribution

As such, these point distribution cases provide useful baseline information with which to evaluate the effects of various containment failure distributions, of congruent versus incongruent matrix dissolution, and the temperature effect on UO_2 solubility. In particular, the first of these two cases is chosen to show the effects of extreme early failure.

a)

b)

FIGURE 4.3. Hypothetical Distribution Used for Containment Failure (Case 3, $\mu = 1000$ years and $\omega = 200$ years) a) Failure Distribution, b) Cumulative Failure Distribution

The normal distributions are selected for their symmetry. The first is centered at $\mu = 1000$ years, with standard deviation $\omega = 200$, and the second is centered at $\mu = 5000$ years, with standard deviation $\omega = 2000$. In the first case, approximately two-thirds of the failures occur between 800 and 1200 years, and approximately 95% of the failures occur between 600 and

a)

b)

FIGURE 4.4. Hypothetical Distribution Used for Containment Failure (Case 4, $\mu = 5000$ years and $\omega = 2000$) a) Failure Distribution, b) Cumulative Failure Distribution

1400 years. In the second case, two-thirds of the failures occur between 3000 and 7000 years, and 95% of the failures occur between 1000 and 9000 years. Simulated failure times less than the 300-year NRC minimum containment time are possible with both assumed normal distributions. These events are discarded when they occur, so for this analysis the distributions are de facto truncated at time $t = 300$ years. The probability of simulating

FIGURE 4.5. Hypothetical Distribution Used for Containment Failure (Case 5, $\lambda = 9.902 \times 10^{-4}$, 50% of failures occur between 300 and 1000 years) a) Failure Distribution, b) Cumulative Failure Distribution

a pre-300-year failure time is 0.0002 (approximately 1 chance in 5000) in the first case and 0.0094 (approximately 1 chance in 100) in the second.

For a normal distribution, the maximum failure rate occurs at its mean and decreases in either direction. The exponential distribution, on the other hand, was selected to examine the effect of high initial failure rate that decreases with time. The rate $\lambda = 9.902 \times 10^{-4}$ was chosen so that 50% of the failures occur between 300 and 1000 years and 50% occur after 1000 years. Except for the distribution which places all containment failure at 300 years, this exponential is the most conservative of the five distributions considered.

4.2 RADIONUCLIDE DATA

Spent fuel contains over 100 radionuclides immediately upon discharge from a nuclear reactor. Screening analyses conducted by all three of the nominated repository sites (DOE 1986a, 1986b, 1986c) have narrowed this list to a much smaller list of key radionuclides. This determination of key radionuclides is achieved by considering data on half-lives, NRC-defined cutoffs based on limited inventory, and conservative estimates based on solubility. Based on these site analyses, the list of key radionuclides to be considered in assessing radionuclide release rates for spent fuel is compiled in Table 4.1. Data on allowable release limits based on NRC (10 CFR 60, 1983) and EPA (40 CFR 191, 1985) regulations are also included in Table 4.1.

Note that ^{137}Cs and ^{90}Sr isotopes are included in Table 4.1. These isotopes can be dismissed from consideration based on NRC's 10 CFR Part 60.113 cutoff with respect to the 1000-year, post-closure curie inventory of spent fuel (Section 3.2.1, Figure 3.2). If release occurs significantly before 1000 years, however, both ^{137}Cs and ^{90}Sr represent appreciable fractions of the total curie inventory of spent fuel. Therefore, the release of ^{90}Sr and ^{137}Cs will be considered in this report for the hypothetical cases involving containment failure occurring prior to 1000 years. The allowable fractional release rates for these nuclides (Table 4.1) are taken from a straight extrapolation from Figure 3.2 based on their 1000-year inventories.

TABLE 4.1. Radionuclide Inventory in Reference PWR Spent Fuel and Allowable Radionuclide Release Limits

Nuclide	1000-Year Inventory, (a) Ci/1000 MTU	NRC Release Limit, (b) parts/yr	EPA Release Limit, (c) Ci/1000 MTU
²⁴⁵ Cm	1.7E02	6.4E-05	100
²⁴³ Am	1.3E04	1.0E-05	100
²⁴¹ Am	3.5E05	1.0E-05	100
²⁴⁰ Pu	4.1E05	1.0E-05	100
²³⁹ Pu	2.8E05	1.0E-05	100
²³⁸ U	3.2E02	3.4E-05	100
²³⁷ Np	5.8E02	1.9E-05	100
²²⁶ Ra	1.5E00	7.2E-03	100
¹³⁷ Cs	2.2E-03	(4.9E00) (d)	(d)
¹³⁵ Cs	2.7E02	4.0E-05	1,000
¹²⁹ I	3.3E01	3.3E-04	100
¹²⁶ Sn	4.8E02	2.3E-05	1,000
⁹⁹ Tc	1.3E04	1.0E-05	10,000
⁹⁰ Sr	6.5E-04	(1.7E01) (d)	(d)
⁷⁹ Se	4.0E02	2.7E-05	1,000
¹⁴ C	6.9E02	1.6E-05	100

(a) DOE 1986c, total 1000-yr inventory equal to 1.08×10^6 Ci/1000 MTU.

(b) DOE 1986b, 1986c.

(c) 40 CFR 191, 1985.

(d) ¹³⁷Cs and ⁹⁰Sr fall below NRC's cutoff for release rate calculations, based on a 1000-year inventory. The parenthetical values are extrapolated from NRC regulations (10 CFR 60, 1983) to provide comparative values in the cases of early (less than 1000 years) containment failure.

It must be recognized that alternate interpretation regarding the allowable release rates of short-lived nuclides is possible. Pigford (1986), for example, has calculated an allowable release rate for ¹³⁷Cs based on inventory at emplacement rather than 1000 years after emplacement. Yet another approach would be to define a time-dependent allowable release rate based on the inventory at the time containment is lost. The current assumption used in this report of a straight line extrapolation to calculate allowable release rates (see Figure 3.2) seems the most consistent with NRC's 10 CFR 60, but this remains a key issue for programmatic resolution.

4.3 SPENT FUEL DATA

Pressurized water reactor fuel has been selected for the assessments in this report. This selection is consistent with the DOE requirements in the GRD, which specifies a characteristic 10-year-old, PWR fuel with 33,000 MWd/MTU burnup for the purposes of preliminary waste package design analyses. The waste loading of this PWR fuel is 0.461 MTU/assembly (DOE 1986a).

The assessment of spent fuel as a waste form requires that certain relevant spent fuel characteristics be identified at the time of disposal. These include:

- identification of component materials
- compositions of component materials
- dimensions of components
- radionuclide inventories of components
- temperatures and heat generation rates
- cladding integrity (optional).

Studies of the spent fuel (Barner 1985) and the interaction of spent fuel with groundwater solutions (Johnson, Garisto, and Stroes-Gascoyne 1985; Wilson and Oversby 1985; Wilson 1985) indicate that release of radionuclides from spent fuel can be divided into four distinct components or radionuclide sources: 1) the matrix of UO_2 grains, 2) the cladding, 3) the gap between the fuel and cladding, and 4) the boundaries between the UO_2 grains. These sources can be distinguished on the basis of physical form, radionuclide inventory, and processes controlling release, as reviewed in Appendix D in Liebetrau et al. (1987). A schematic cross section of spent fuel showing these separate sources is presented in Figure 1.1.

Because radionuclide inventories and heat generation rates vary with time, their values must be specified or calculated for the post-emplacement period of a repository. Inventories for reference PWR fuel at 1000 years after permanent closure, $I(1000)$, are identified in Table 4.1 for the key radionuclides (DOE 1986c). Table 4.2 presents the percentages of these nuclides in the different sources within spent fuel. These inventories correspond to spent fuel 1060 years after reactor discharge, assuming a 60-year

TABLE 4.2. Distribution of Key Radionuclides Used for Reference
PWR Spent Fuel (Percentage)

<u>Nuclide</u>	<u>UO₂ Matrix</u>	<u>Gap (+ Grain Boundary) (a)</u>	<u>Cladding</u>	<u>Crud</u>
²³⁸ U	100	--	--	--
²⁴¹ Am	100	--	--	--
²⁴³ Am	100	--	--	--
¹⁴ C (b)	35.0	1.00	63.0	1.0
²⁴⁵ Cm	100	--	--	--
¹³⁵ Cs	98.0	2.00	--	--
¹³⁷ Cs	98.0	2.00	--	--
⁹⁰ Sr	98.0	2.00	--	--
¹²⁹ I	98.0	2.00	--	--
²³⁷ Np	100	--	--	--
²³⁹ Pu	100	--	--	--
²⁴⁰ Pu	100	--	--	--
²²⁶ Ra	100	--	--	--
⁷⁹ Se	98.0	2.00	--	--
⁹⁹ Tc	98.0	2.00	--	--
¹²⁶ Sn	100	--	--	--

(a) The 2% of total volatile fission products in the combined gap and grain boundary source is based on fission gas measurements of moderate burnup PWR fuel (Baker et al. 1987; Barner 1985) and experimental data on release in closed system tests (Oversby 1987).

(b) Based on data obtained by Van Konynenburg et al. 1986.

preclosure time period for repository construction and waste emplacement. The allowable NRC fractional release rates are based on mass transfer rates of nuclides that are normalized with respect to these 1000-year inventories (see Liebetrau et al. 1987, Appendix E). The 1000-year inventory, I(1000), and the half-life of a radionuclide can be used to calculate its inventory at other times as needed.

Radionuclide inventories in spent fuel rods vary because of radioactive decay, as well as differences in age and burnup (Roddy et al. 1986). Inventory variations because of spent fuel type, age, and burnup have not been modeled in the AREST code. It is expected that fuels with lower burnup than the 33,000 MWd/MTU used as a reference in this report should perform better than the calculated values of this report; higher burnup fuel is not significantly different in composition (inventories higher by a factor of 3 or less; Roddy et al. 1986). The effect of burnup will be investigated in later studies.

The heat generation rate, which varies with time, is used in the AREST code to calculate the waste package temperature, as described in Appendix B of Liebetrau et al. (1987). The time dependence of temperature is judged to be of primary importance because other processes (corrosion, diffusion) and variables (solubility, groundwater composition) may be temperature dependent.

Appendix B of Liebetrau et al. (1987) also describes how the heat generation rate distribution for disposed spent fuel used in this report is developed. The WASTES II computer code (Shay and Buxbaum 1986) is used to analyze the repository receipt scenario described in the DOE Mission Plan for the Office of Civilian Radioactive Waste Management (OCRWM) Program (DOE 1985). The heat generation rate distribution is developed for both PWR and BWR fuel, but PWR will constitute 65% of the fuel in the repository. As noted previously, PWR and BWR fuel with the same burnup and age have similar radionuclide inventories (Roddy et al. 1986) and can, therefore, be expected to have similar heat generation rates.

The dimensions of the reference PWR spent fuel rod used in this report are: 370.8 cm (146 in.) in length, 1.12 cm (0.44 in.) in diameter, and $364 \text{ cm}^3/\text{rod}$ ($22.2 \text{ in.}^3/\text{rod}$) in volume (Roddy et al. 1986). Because theoretical distributions of containment failure are used in this report, the containment performance of the cladding and the container are not distinguished. The WPC model of the AREST code does, however, permit the option of defining a percentage for prefabricated cladding or container materials (see Section 2.3 in Liebetrau et al. 1987).

4.4 SITE DATA

The site-specific data used in the AREST code for analysis of spent fuel performance as a waste form are reviewed in the following sections. The majority of these data come from the EA reports (DOE 1986a, 1986b, 1986c) for the SRP, BWIP, and NNWSI. In Section 6.4.2 of each EA, details are given concerning the performance assessment approaches being adopted by the projects to demonstrate compliance with the NRC fractional release requirement. In some instances, the assumptions used in this analysis of spent fuel are inconsistent with assumptions made in the EAs or other project documents. These differences in assumptions are discussed in the following sections. Many of these differences arise because the purpose of the EAs (analysis of preliminary waste package design) and the purpose of this report (assessment of spent fuel as a waste form) are different.

4.4.1 Basalt Waste Isolation Project

The release rate analyses performed by the BWIP are obtained from finite element mass-transfer codes for performance assessment, CHAIN-T and REPREL (DOE 1986b; Yung et al. 1987; Sagar, Eslinger, and Baca 1984). The BWIP has also developed analytical models for calculating release (Oliver 1985; Bensky and Oliver 1985). Both BWIP codes and the AREST code rely chiefly on solubility-limited, diffusion-controlled release of radionuclides in a saturated medium. For highly soluble radionuclides, the inventory-limited mass transfer model of AREST (Liebetrau et al. 1987, Section 2.4 and Appendix E) is slightly different than the solubility-limited model adopted by the BWIP. The AREST code also can specifically model the potential effects of congruent dissolution of the UO_2 matrix, while the BWIP codes currently treat the dissolution of UO_2 as incongruent (i.e. separate and independent radionuclide solubilities). On the other hand, the finite-element basis of the BWIP codes is more flexible for incorporating many concentric barriers or layers with different physical and chemical properties.

Table 4.3 contains the information on the waste package design and dimensions presented in the BWIP EA (DOE 1986b). This table also shows the dimensions of the spherical waste package geometry, used in AREST, that are

TABLE 4.3. Design Parameters for BWIP Release Calculations (DOE 1986b)

Diffusion coefficient in steam (D_s) = $5.0000\text{E-}01 \text{ cm}^2/\text{sec}$
Diffusion coefficient in water (D_f) = $1.0000\text{E-}05 \text{ cm}^2/\text{sec}$
Packing thickness (b) = $1.9300\text{E+}01 \text{ cm}$
Spherical waste form radius (R_0) = $5.9353\text{E+}01 \text{ cm}$
Radius of waste package (R_1) = $7.8653\text{E+}01 \text{ cm}$
Packing porosity (ϵ_1) = 0.3
Rock porosity (ϵ_2) = 0.001
Gap width (a) = $6.4000\text{E+}00 \text{ cm}$
Gap surface area (S_v) = $4.4269\text{E+}04 \text{ cm}^2$
Gap volume (V_v) = $2.8323\text{E+}05 \text{ cm}^3$
PWR Assemblies/Waste Package = 4
Waste Loading = $1.8456\text{E+}00 \text{ MTU/Waste Package}$
Bulk density of packing (ρ_1) = $1.8000\text{E+}00 \text{ gm/cm}^3$
Bulk density of host rock (ρ_2) = $3.0000\text{E+}00 \text{ gm/cm}^3$
Saturation (ϕ) = 100%

obtained from the reference BWIP dimensions. The failed container barrier is not included in the analysis for this report because of current limitations in the AREST code.

The GR-4 groundwater of the BWIP is used as the reference groundwater for the spent fuel analysis; its composition is tabulated in Liebetrau et al. (1987, Appendix C). Reference BWIP EA values of expected radionuclide solubilities and sorption coefficients for packing and basalt are provided in Table 4.4 for key radionuclides. The BWIP temperature versus time (T-t) profile for a reference spent fuel (Yung et al. 1987), as well as the distribution of temperature-versus-time profiles calculated in AREST, are plotted in Liebetrau et al. (1987, Appendix B) for the range of heat generation rates for spent fuel expected at a first repository. Note that the distribution of T-t profiles assumes that each BWIP waste container will contain the same number of spent fuel assemblies regardless of their heat generation rate. The BWIP, or any repository site, could opt to reduce the number of spent fuel assemblies per package to lower the upper limit in these calculated T-t profiles.

TABLE 4.4. Nuclide Parameters for BWIP Release Calculations (DOE 1986b)

Nuclide	Half-life, yr	Inventory mg/package	Solubility, ^(a) gm/m ³	K _D (P), ^(b) mL/gm	K _D (R), ^(b) mL/gm	Specific Activity, Ci/gm	K ₁ ^(c)	K ₂ ^(c)
²³⁸ U	4.51E+09	1.77E+09	2.38E-05	2.00E+01	2.00E+01	3.33E-07	1.21E+02	6.00E+04
²⁴¹ Am	4.58E+02	1.99E+05	2.04E-03	1.70E+02	1.70E+02	3.24E+00	1.02E+03	5.10E+05
²⁴³ Am	7.95E+03	1.30E+05	3.77E-04	1.70E+02	1.70E+02	1.85E-01	1.02E+03	5.10E+05
¹⁴ C	5.73E+03	2.86E+02	5.60E-05	0.00E+00	0.00E+00	4.45E+00	1.00E+00	1.00E+00
²⁴⁵ Cm	9.30E+03	2.00E+03	2.45E-03	2.50E+01	2.50E+01	1.57E-01	1.51E+02	7.50E+04
¹³⁵ Cs	3.00E+06	5.65E+05	1.35E+01	2.50E+02	2.50E+02	8.82E-04	1.50E+03	7.50E+05
¹³⁷ Cs	3.00E+01	4.67E-08	1.35E+01	2.50E+02	2.50E+02	8.70E+01	1.50E+03	7.50E+05
⁹⁰ Sr	2.90E+01	8.76E-09	9.00E-03	1.00E+02	1.00E+02	1.37E+02	6.01E+02	3.00E+05
¹²⁹ I	1.59E+07	3.50E+05	1.29E+03	0.00E+00	0.00E+00	1.74E-04	1.00E+00	1.00E+00
²³⁷ Np	2.14E+06	1.54E+06	2.37E-05	1.00E+01	1.00E+01	7.05E-04	6.10E+01	3.00E+04
²³⁹ Pu	2.44E+04	8.43E+06	1.43E-04	2.10E+01	2.10E+01	6.13E-02	1.27E+02	6.30E+04
²⁴⁰ Pu	6.58E+03	3.35E+06	7.20E-05	2.10E+01	2.10E+01	2.26E-01	1.27E+02	6.30E+04
²²⁶ Ra	1.60E+03	2.80E+00	2.26E-03	5.00E+01	5.00E+01	9.88E-01	3.01E+02	1.50E+05
⁷⁹ Se	6.50E+04	1.06E+04	7.90E-04	4.00E+00	4.00E+00	6.97E-02	2.50E+01	1.20E+04
⁹⁹ Tc	2.15E+05	1.41E+06	1.98E-03	1.50E+01	1.50E+01	1.70E-02	9.10E+01	4.50E+04
¹²⁶ Sn	1.00E+05	3.12E+04	3.78E-06	5.00E+00	5.00E+00	2.84E-02	3.10E+01	1.50E+04
Zr	0.00E+00	2.12E+09	4.60E-05	0.00E+00	0.00E+00	4.45E+00	1.00E+00	1.00E+00

(a) For elements with more than one long-lived isotope, an "isotopic solubility," is equal to the elemental solubility times the mass fraction of that particular isotope at 1000 years after emplacement. Note, because ¹³⁷Cs is extremely short-lived compared to ¹³⁵Cs, the elemental solubility was used for evaluation of ¹³⁷Cs released for early (less than 1000 years) times of containment failure.

(b) P = pecking (or equivalent); R = Host Rock; K_D = sorption coefficient.

(c) K₁ = Packing Retardation Coefficient; K₂ = Host Rock Retardation Coefficient.

4.4.2 Nevada Nuclear Waste Storage Investigation

Three models are identified in the EA of the NNWSI (DOE 1986c) for evaluating the release of radionuclides from a spent fuel waste package. The first of these is a bulk-flow saturation model, in which the entire volume of groundwater flowing through the repository is assumed to reach solubility limits with respect to each waste element (Kerrisk 1985). This model must assume that the air gap between the waste package and the tuff is maintained to assure no diffusional pathways. Such pathways would dominate mass transport at the low groundwater flow rates at the tuff site. As noted by the NNWSI (DOE 1986c), this approach is needlessly unrealistic.

The second model advocated in the NNWSI EA is a diffusion-limited model in which the water present at the waste form surface is assumed to be

saturated with radionuclides and radionuclide release is controlled by diffusional transport out of the waste package (Kerrisk 1985). This model is more physically realistic than the bulk-flow saturation model, yet provides a technically defensible approach to estimating radionuclide release rates. However, the pseudo-empirical boundary-layer approximation of Kerrisk (1985) is only for steady state. As shown in this report and work by Chambré et al. (1985), early transient release rates can be much higher than steady-state values. Also the NNWSI notes (DOE 1986c) that highly soluble radionuclides may not be limited by solubility. This does not mean, however, that the release of such nuclides cannot be analyzed by a mass transfer approach; Liebetrau et al. (1987, Section 2.4 and Appendix E) discuss a mass-transfer model that was developed by Kim, Chambré, and Pigford (1986) and was used in AREST for highly soluble ("inventory-limited") nuclides.

The third model proposed by the NNWSI as the most realistic for release calculations is also a bulk-flow model. In this model, flow is coupled with the kinetics of waste form dissolution to predict release rates (Oversby and McCright 1984). The "flow" is interpreted to be the slow dripping of groundwater across a waste form under unsaturated hydrologic conditions. Geometric analysis of the limited perforation of container and cladding, as well as exposed surface area of waste form, are included in the model.

Although this model is claimed by the NNWSI to be both realistic and conservative, these assumptions have been questioned (Pigford and Chambré 1986) because the model predicts no waste package release at zero flow. Their assessment of this model is that it requires that there is no possibility of pathways for molecular diffusion of dissolved species being established between the waste form and the surrounding, unsaturated tuff host rock. If such pathways were to be formed, a nonzero release rate of radionuclides would occur even at zero flow because of diffusion-controlled transport (i.e. NNWSI's second model, cited in a previous paragraph). Under such conditions, this diffusion-controlled release would be far more important than the convection-controlled transport at the low flow rates predicted for the tuff site.

The WPR submodel of the AREST code is based on mass transfer models assuming diffusion-controlled transport. This approach relies on the existence of a connective pathway in the region between the waste container/waste form and the tuff host rock. Establishment of diffusional pathways is not an expected condition for a waste package located in unsaturated tuff; however, it is beyond the scope of this report to demonstrate convincingly that such pathways cannot and will not be established. Furthermore, the diffusion-limited, mass transfer approach adopted in this report is similar to the second model advocated by the NNWSI, and AREST has the added advantage of calculating transient release rates, as well as the steady-state release rates.

Table 4.5 contains the information on the waste package design and dimensions presented in the NNWSI EA (DOE 1986c). For the purposes of the spent fuel analyses conducted for this report, it is assumed that the void region between the NNWSI waste container and tuff host rock is filled with a matrix of crushed tuff with a 50% porosity. This table also shows the dimensions of the spherical waste package geometry, used in AREST, that was

TABLE 4.5. Design Parameters for NNWSI Release Calculations (DOE 1986c)

Diffusion coefficient in steam (D_s) = 5.0000E-01 cm ² /sec
Diffusion coefficient in water (D_f) = 1.0000E-05 cm ² /sec
Packing thickness (b) = 3.0000E+00 cm
Spherical waste form radius (R_0) = 8.5895E+01 cm
Radius of waste package (R_1) = 8.8895E+01 cm
Packing porosity (ϵ_1) = 0.5
Rock porosity (ϵ_2) = 0.1
Gap width (a) = 1.0300E+01 cm
Gap surface area (S_v) = 9.2715E+04 cm ²
Gap volume (V_v) = 9.5701E+05 cm ³
PWR Assemblies/Waste Package = 7
Waste Loading = 3.2298E+00 MTU/Waste Package
Bulk density of packing (ρ_1) = 1.2900E+00 gm/cm ³
Bulk density of host rock (ρ_2) = 2.3300E+00 gm/cm ³
Saturation (ϕ) = 60%

obtained from the dimension presented. As in the BWIP case, the failed container barrier is not included in the analysis because of current limitations in the AREST code.

The J-13 groundwater of the NNWSI is used as the reference groundwater for the spent fuel analyses; its composition is tabulated in Liebetrau et al. (1987, Appendix C). Reference NNWSI EA values of expected radionuclide solubilities and sorption coefficients on tuff are provided in Table 4.6 for key radionuclides. The NNWSI temperature versus time (T-t) profile for a reference spent fuel, as well as the distribution of temperature versus time profiles calculated in AREST, are shown in Liebetrau et al. (1987, Appendix B) for the range of heat generation rates for spent fuel expected at a first repository. As with the BWIP case, the upper T-t profiles of NNWSI waste

TABLE 4.6. Nuclide Parameters for NNWSI Release Calculations (DOE 1986c)

Nuclide	Half-life, yr	Inventory mg/pack	Solubility, (a) gm/m ³	K _D (P), (b) mL/gm	K _D (R), (b) mL/gm	Specific Activity, Ci/gm	K ₁ (c)	K ₂ (c)
²³⁸ U	4.51E+09	3.10E+09	5.00E+01	1.80E+00	1.80E+00	3.33E-07	5.64E+00	4.29E+01
²⁴¹ Am	4.58E+02	3.49E+05	1.45E-03	1.20E+03	1.20E+03	3.24E+00	3.10E+03	2.80E+04
²⁴³ Am	7.95E+03	2.27E+05	9.72E-04	1.20E+03	1.20E+03	1.85E-01	3.10E+03	2.80E+04
¹⁴ C	5.73E+03	5.01E+02	1.40E+00	0.00E+00	0.00E+00	4.45E+00	1.00E+00	1.00E+00
²⁴⁵ Cm	9.30E+03	3.50E+03	2.45E-03	1.20E+03	1.20E+03	1.57E-01	3.10E+03	2.80E+04
¹³⁵ Cs	3.00E+06	9.89E+05	8.10E+05	2.90E+02	2.90E+02	8.82E-04	7.49E+02	6.76E+03
¹³⁷ Cs	3.00E+01	8.17E-08	8.10E+05	2.90E+02	2.90E+02	8.70E+01	7.49E+02	6.76E+03
⁹⁰ Sr	2.90E+01	1.53E-08	9.40E-04	5.30E+01	5.30E+01	1.37E+02	1.38E+02	1.24E+03
¹²⁹ I	1.59E+07	6.13E+05	7.74E+05	0.00E+00	0.00E+00	1.74E-04	1.00E+00	1.00E+00
²³⁷ Np	2.14E+06	2.69E+06	7.11E+02	7.00E+00	7.00E+00	7.05E-04	1.91E+01	1.64E+02
²³⁹ Pu	2.44E+04	1.48E+07	3.10E-01	6.40E+01	6.40E+01	6.13E-02	1.66E+02	1.49E+03
²⁴⁰ Pu	6.58E+03	5.86E+06	1.20E-01	6.40E+01	6.40E+01	2.26E-01	1.66E+02	1.49E+03
²²⁶ Ra	1.60E+03	4.90E+00	2.26E-02	1.50E+03	1.50E+03	9.88E-01	3.87E+03	3.50E+04
⁷⁹ Se	6.50E+04	1.86E+04	5.50E+05	7.00E+00	7.00E+00	6.97E-02	1.91E+01	1.64E+02
⁹⁹ Tc	2.15E+05	2.47E+06	9.90E+05	3.00E-01	3.00E-01	1.70E-02	1.77E+00	7.99E+00
¹²⁶ Sn	1.00E+05	5.46E+04	1.26E-04	1.00E+02	1.00E+02	2.84E-02	2.59E+02	2.33E+03
Zr	0.00E+00	3.71E+09	4.60E-05	0.00E+00	0.00E+00	4.45E+00	1.00E+00	1.00E+00

(a) For elements with more than one long-lived isotope, an "isotopic solubility," is equal to the elemental solubility times the mass fraction of that particular isotope at 1000 years after emplacement. Note, because ¹³⁷Cs is extremely short-lived compared to ¹³⁵Cs, the elemental solubility was used for evaluation of ¹³⁷Cs released for early (less than 1000 years) times of containment failure.

(b) P = packing (or equivalent); R = Host Rock; K_D = sorption coefficient.

(c) K₁ = Packing Retardation Coefficient; K₂ = Host Rock Retardation Coefficient.

packages calculated here are upper limits. These upper limits are based on waste container loadings of spent fuel with above-average heat generation rates. A reduction in the number of spent fuel assemblies per package could significantly lower the upper limit in these calculated T-t profiles.

4.4.3 Salt Repository Project

The EA of the SRP (DOE 1986a) estimates radionuclide release rates from the waste package by a bulk-saturation model. The time-dependent volume of brine inclusions that migrate to the waste package is calculated and this quantity of accumulated brine is assumed to become saturated with respect to all nuclides. The physical basis for the brine-migration model used in such assessments has been questioned (Lieberman et al. 1985; Pigford and Chambré 1985). Furthermore, sources of water other than inclusions, such as water-bearing sedimentary layers in the salt, may provide water to the waste package environment in excess of that calculated by brine-migration models.

This report adopts Pigford and Chambré's (1985) recommendations for predicting radionuclide release from a waste package surrounded by salt. A continuous brine phase along the grain boundaries of salt and waste package materials is assumed. Thus, the waste package and the immediately adjacent salt are saturated media through which nuclides can migrate by diffusion. Convective transport by pressure-induced brine flow is expected to be negligible. This assumption is based on the rapid readjustment of any stress differentials by the means of salt creep in the hot salt immediately adjacent to a waste package (Ratigan 1984). It is conservatively assumed that upon failure, the waste container is immediately filled with brine and that the brine attains saturation-limited (or an inventory-limited) concentration at the same time.

Table 4.7 contains information on the waste package design and dimensions presented in the SRP EA (DOE 1986a). The SRP's EA waste package design for emplacement includes a thin (2.75 cm) packing of crushed salt between the waste container and the host salt formation. It is expected that rapid salt creep will act to consolidate this salt packing material soon after emplacement. This will result in physical properties (porosity, density, heat conductivity) that are indistinguishable from the bulk, undisturbed host salt

TABLE 4.7. Design Parameters for SRP Release Calculations (DOE 1986a)

Diffusion coefficient in steam (D_s)	= 5.0000E-01 cm ² /sec
Diffusion coefficient in water (D_f)	= 1.0000E-05 cm ² /sec
Packing thickness (b)	= 1.2000E+01 cm
Spherical waste form radius (R_o)	= 7.7112E+01 cm
Radius of waste package (R_1)	= 8.9112E+01 cm
Packing porosity (ϵ_1)	= 0.5
Rock porosity (ϵ_2)	= 0.0001
Gap width (a)	= 4.2900E+00 cm
Gap surface area (S_v)	= 7.4723E+04 cm ²
Gap volume (V_v)	= 3.2070E+05 cm ³
PWR Assemblies/Waste Package	= 10
Waste Loading	= 4.6140E+00 MTU/Waste Package
Bulk density of packing (ρ_1)	= 2.1400E+00 gm/cm ³
Bulk density of host rock (ρ_2)	= 2.2000E+00 gm/cm ³
Saturation (ϕ)	= 100%

formation. For this reason and because of the extreme thinness of this consolidated salt packing layer, the relatively thick (12 cm) container material is explicitly incorporated into the AREST calculations as the prime barrier material between the waste form and the host rock. The transport across the thin consolidated salt packing is neglected and the waste package boundary is effectively defined as the outer surface of the waste container. Table 4.7 also shows the dimensions of the spherical waste package geometry, used in AREST, obtained by transforming the given dimensions.

The PBB-1 brine developed by the SRP is used as the reference groundwater for these analyses; its composition is tabulated in Liebetrau et al. (1987, Appendix C). Reference SRP EA values of expected radionuclide solubilities are provided in Table 4.8 for key radionuclides. Because the modeling approach advocated by SRP in its EA (DOE 1986a) does not require reference sorption coefficients on salt and iron corrosion products, these data are not available for use in the calculations of this report. Sorption coefficients for salt are conservatively assumed to be zero for all radionuclides. The sorption coefficients used for the corroded container

TABLE 4.8. Nuclide Parameters for SRP Release Calculations (DOE 1986a)

Nuclide	Half-life, yr	Inventory mg/peck	Solubility, ^(a) gm/m ³	K _D (P), ^(b) mL/gm	K _D (R), ^(b) mL/gm	Specific Activity, Ci/gm	K ₁ ^(c)	K ₂ ^(c)
²³⁸ U	4.51E+09	4.43E+09	1.00E-03	1.00E+02	0.00E+00	3.33E-07	4.29E+02	1.00E+00
²⁴¹ Am	4.58E+02	4.98E+05	8.45E-05	5.00E+02	0.00E+00	3.24E+00	2.14E+03	1.00E+00
²⁴³ Am	7.95E+03	3.24E+05	1.55E-05	5.00E+02	0.00E+00	1.85E-01	2.14E+03	1.00E+00
¹⁴ C	5.73E+03	7.15E+02	6.00E-02	0.00E+00	0.00E+00	4.45E+00	1.00E+00	1.00E+00
²⁴⁵ Cm	9.30E+03	5.00E+03	1.00E-03	5.00E+02	0.00E+00	1.57E-01	2.14E+03	1.00E+00
¹³⁵ Cs	3.00E+06	1.41E+06	6.00E+05	1.00E+02	0.00E+00	8.82E-04	4.29E+02	1.00E+00
¹³⁷ Cs	3.00E+01	1.17E-07	6.00E+05	1.00E+02	0.00E+00	8.70E+01	4.29E+02	1.00E+00
⁹⁰ Sr	2.90E+01	2.19E-07	8.00E-01	1.00E+02	0.00E+00	1.37E+01	4.29E+02	1.00E+00
¹²⁹ I	1.59E+07	8.75E+05	6.00E+05	1.00E-01	0.00E+00	1.74E-04	1.43E+00	1.00E+00
²³⁷ Np	2.14E+06	3.84E+06	1.00E-03	1.00E+02	0.00E+00	7.05E-04	4.29E+02	1.00E+00
²³⁹ Pu	2.44E+04	2.11E+07	6.00E-04	5.00E+02	0.00E+00	6.13E-02	2.14E+03	1.00E+00
²⁴⁰ Pu	6.58E+03	8.37E+06	3.00E-04	5.00E+02	0.00E+00	2.26E-01	2.14E+03	1.00E+00
²²⁶ Ra	1.60E+03	7.01E+00	4.20E-04	1.00E+02	0.00E+00	9.88E-01	4.29E+02	1.00E+00
⁷⁹ Se	6.50E+04	2.65E+04	1.00E-03	5.00E-01	0.00E+00	6.97E-02	3.14E+00	1.00E+00
⁹⁹ Tc	2.15E+05	3.53E+06	1.00E-03	2.50E+01	0.00E+00	1.70E-02	1.08E+02	1.00E+00
¹²⁶ Sn	1.00E+05	7.80E+04	1.00E-04	5.00E+00	0.00E+00	2.84E-02	2.24E+01	1.00E+00
Zr	0.00E+00	5.30E+09	4.60E-05	0.00E+00	0.00E+00	4.45E+00	1.00E+00	1.00E+00

- (a) For elements with more than one long-lived isotope, an "isotopic solubility," is equal to the elemental solubility times the mass fraction of that particular isotope at 1000 years after emplacement. Note, because ¹³⁷Cs is extremely short-lived compared to ¹³⁵Cs, the elemental solubility was used for evaluation of ¹³⁷Cs released for early (less than 1000 years) times of containment failure.
- (b) P = packing (or equivalent); R = Host Rock; K_D = sorption coefficient.
- (c) K₁ = Packing Retardation Coefficient; K₂ = Host Rock Retardation Coefficient.

material were adapted from data on "average sediments" (McKinley and Hadermann 1984). Experimental studies are being conducted by the PASS program to obtain data to access this process.

The SRP temperature-time (T-t) profile for a reference spent fuel, as well as the distribution of temperature versus time profiles calculated in AREST, are given in Liebetrau et al. (1987, Appendix B) for the range in heat generation rates of spent fuel expected at a first repository. The agreement between the AREST and SRP T-t profiles for the reference heat-loading case was not as good as for the BWIP and NNWSI cases, as noted in Liebetrau et al. (1987, Appendix B). As in the BWIP and NNWSI cases, the upper T-t profiles for the SRP waste packages calculated here are upper limits. Reduction in the number of spent fuel assemblies per package could significantly lower the upper limit in these calculated T-t profiles.

5.0 SUMMARY OF ASSUMPTIONS USED IN THE SPENT FUEL ASSESSMENT REPORT

Numerous assumptions have been made for this analysis of spent fuel performance as a waste form in a geologic repository. These assumptions are summarized here.

5.1 PROGRAMMATIC ASSUMPTIONS

- Waste package designs from the site-specific EA reports, although not necessarily final or optimal, are used for this spent fuel analysis (DOE 1986a, 1986b, 1986c).
- Repository data presented by the individual repository projects in the EA reports represent "anticipated processes and events" (DOE 1986a, 1986b, 1986c).
- A distribution of spent fuel heat generation rates, and hence waste package temperatures, is developed from the 1985 DOE Mission Plan (DOE 1985) scenario for spent fuel receipts at a first repository.
- Pressurized-water reactor fuel, having a 33,000 MWd/MTU-burnup, is adopted as the reference for nonthermal spent fuel characteristics.
- Compliance with the NRC performance objective for containment is met by assuming release will not occur before the 300-year period following repository closure. This does not imply that NRC prohibits any release before 300 years, but the issue of meeting substantially complete containment is outside the intended scope of this study.
- The NRC, EPA, and DOE release rate performance objectives are conservatively evaluated at the boundary of the waste package. This is especially conservative with respect to the EPA release objective, which is actually applied at distances up to 5 km from the waste package.
- Nuclides with short half-lives that can be eliminated from consideration on the basis of the NRC 1000-year inventory-cutoff are reconsidered if release begins significantly before 1000 years.

5.2 MODELING ASSUMPTIONS

- The performance of individual waste packages is independent of the performance of adjoining waste packages. Early containment failures would release radionuclides into the host rock. This assumption conservatively neglects this effect on the mass transfer rates from waste packages that subsequently lose containment.
- The waste package and spent fuel eventually come into contact with a continuous hydrologic system with a fixed degree of water saturation. This assumption may significantly "penalize" repository sites where the anticipated (but as yet undemonstrated) condition is a sustained air gap or void between the waste package and host rock.
- Under anticipated conditions, transport within the waste package and in the immediately adjacent host rock is by diffusion (NAS 1983; Lieberman et al. 1985).
- Three types of containment failure distributions are assumed: point (simultaneous failure of all packages), symmetric, and asymmetric.
- The UO_2 matrix dissolves congruently at all repository sites. Experimental evidence of congruent release of nuclides contained in the UO_2 matrix of spent fuel under oxidizing and reducing conditions is well established and documented by U.S., Canadian and Swedish researchers (Johnson et al. 1982; Johnson et al. 1984; Wilson et al. 1985; Oversby and Wilson 1985; Johnson, Garisto, and Stroes-Gascoyne 1985; Wilson 1985; Forsyth and Werme 1985). The assumption of congruent release has been adopted for most performance assessment modeling studies of spent fuel (LeNeveu 1986; Wuertz and Ellinger 1985; Johnson, Garisto, and Stroes-Gascoyne 1985; Garisto and Garisto 1985; Pigford and Chambré 1986; Kim, Chambré, and Pigford 1986). It is theoretically possible that some radionuclides may form sufficiently low-solubility precipitates that the net release from UO_2 is apparently incongruent, despite

actual congruent dissolution and release from the UO_2 matrix. Liebetrau et al. (1987, Appendix E) discuss the conditions necessary for this eventuality.

As an upper bounding condition, however, this report also calculates nuclide release rates conservatively assuming incongruent (i.e. nuclides controlled by separate solubility-limiting solids) release. It must be kept in mind that incongruent release of nuclides from the UO_2 matrix is treated here as an unanticipated condition for all repository sites based on testing and modeling results obtained to date.

- The UO_2 matrix of spent fuel is stable in proximity to metallic container and waste package materials.

This is a key assumption because the waste package release rates of nuclides included in a congruently dissolving UO_2 matrix can be directly related to the release rate of uranium (see Liebetrau et al. 1987, Appendix E), unless UO_2 is thermodynamically unstable and a new, lower-solubility uranium-bearing solid were to precipitate within the waste package (Johnson, Garisto, and Stroes-Gascoyne 1985). Auto-oxidation of UO_2 by alpha radiolysis has been suggested as one mechanism for making UO_2 metastable (Neretnieks 1984). Subsequent laboratory testing of spent fuel in the presence of other waste package materials, such as metallic container materials, has indicated that alpha radiolysis-induced oxidation of UO_2 is not observed, possibly because of buffering by waste package materials, including the UO_2 itself (Johnson et al. 1982; Forsyth and Werme 1985).

It has recently been argued that even under conditions at which UO_2 is stable, the irradiated UO_2 matrix of spent fuel must be considered as a metastable solid solution of UO_2 , UO_3 , other actinide oxides, fission products, and structural defects (Murphy and Smith 1987). Murphy and Smith developed a model for irreversible dissolution and precipitation of such solutions for ideal binary systems. The principle assumptions of their model are that the forward (dissolution) reaction is

stoichiometric and zero order, that the reverse (precipitation) reaction stoichiometry corresponds to the solid solution phase that is most stable with respect to the aqueous solution, and that the surface composition of the phase is homogeneous but variable in time. Calculations with this model show enrichments or depletions in non-uranium components leading to new equilibrium concentrations in a reconstructed UO_2 surface. Although these calculations were made for closed systems (i.e., no mass transport) conditions, it may have important implications for the release of trace components incorporated into the UO_2 matrix under conditions approaching, or at, UO_2 saturation. This model would lead to time-invariant concentrations of trace components at the UO_2 surface, although these values would be controlled to the relative solubility products for these trace components compared to pure UO_2 , rather than the initial inventory of the trace nuclides. Confirmation of these models must rely on direct testing of UO_2 from spent fuel. Unfortunately, experimental studies of UO_2 under expected repository conditions are difficult because of low solubilities and relatively high detection levels for uranium and other components. It is proposed that studies of UO_2 from spent fuel in mixed-flow reactors (e.g., Rimstidt and Dove 1986) using aqueous solution presaturated with UO_2 could be used to discriminate among these models.

Notwithstanding these models for release under conditions of UO_2 stability, the unanticipated condition of a uranium-bearing precipitate in the waste package would lead to an enhanced release rate of uranium (hence, included nuclides) from the waste form, but not necessarily from the waste package. Garisto and Garisto (1986) have calculated enhancements in waste form release of uranium by up to a factor of 40 under extreme conditions (i.e. precipitation 1 cm from the UO_2 matrix of a uranium-bearing solid with a solubility 1000 times lower) for a simple one-dimensional mass transfer model. Further work on the potential effect of UO_2 matrix restructuring and/or uranium precipitation on the assessment of spent fuel is

needed. Appropriate modifications to existing mass transfer equations within the AREST code (see Liebetrau et al. 1987, Appendix E) are in progress.

- The grain boundary of spent fuel is identical in composition to the gap source, and the two sources are treated as the "gap" release. A conservatively high value of 2% of the entire inventory of fission products is assumed to reside in this combined gap and grain boundary source (Baker et al. 1987; Barner 1985).
- The surface concentrations are set by solubility relationships immediately upon containment failure; no credit is taken for the time to reach surface saturation.
- No credit is taken for potential attenuation of radionuclide release arising from containment failure over a limited surface area.
- No credit is taken for the effect of pathway tortuosity on effective diffusion coefficients.
- No credit is taken for fuel cladding as a containment barrier.

6.0 DISCUSSION OF SPENT FUEL RELEASE RATE ANALYSES

The graphical results of the AREST analysis of spent fuel as a waste form are presented in Sections 7.0 (BWIP), 8.0 (NNWSI), and 9.0 (SRP). These divisions are based upon the repository location and the type of host rock--basalt, tuff, and salt--in which the fuel-bearing waste package is located. Each of these sections are divided into five cases based upon the assumed containment failure distributions:

- Case 1. All containment failures are placed at 300 years after repository closure.
- Case 2. All containment failures are placed at 1000 years after repository closure.
- Case 3. The containment failures have a normal distribution with a mean value of 1000 years and a standard deviation of 200 years, truncated at 300 years.
- Case 4. The containment failures have a normal distribution with a mean value of 5000 years and a standard deviation of 2000 years, truncated at 300 years.
- Case 5. The containment failures have an exponential distribution with a rate of 9.902×10^{-4} , chosen so that 50% of the failures occur between 300 and 1000 years after repository closure.

The repository average release from the UO_2 matrix, gap/grain boundary, and cladding sources are calculated for each failure distribution. Separate scenarios for congruent and incongruent release of nuclides from the UO_2 matrix (see Liebetrau et al. 1987, Appendix E) are also assessed. Radionuclide release is evaluated with respect to both NRC fractional release rates limits and EPA cumulative release limits (Section 3.2). In addition, the distribution of release rates for ^{237}Np (matrix nuclide) and ^{129}I (gap and

matrix nuclide) are evaluated for Case 3 of each host rock. These distributions are calculated at 1000 and 5000 years after repository closure, and provide an indication of the range in fractional release rates within the engineered barriers system of a repository.

7.0 SPENT FUEL RELEASE RATE ANALYSIS FOR THE BASALT WASTE ISOLATION PROJECT

The graphical results and discussion of the AREST analysis for BWIP are presented in this section. The results and discussion are presented for each of the five container failure distribution cases.

7.1 CASE 1

Figures 7.1 through 7.7 present the AREST analyses of waste package releases for the case where containment for all waste packages is instantaneously lost 300 years after permanent repository closure. This is the most conservative (and perhaps most unrealistic) case considered in this report.

For incongruent release from the UO_2 matrix (Figures 7.1 to 7.2), all nuclides are below their allowable NRC fractional release rates except ^{90}Sr , ^{137}Cs , ^{129}I , ^{135}Cs , and ^{226}Ra . The latter three are released at such high rates that they are rapidly depleted from the matrix. The ingrowth of ^{226}Ra is not considered for incongruent release.

In the congruent release scenario, the surface concentration (hence, release rate) of a contained nuclide is controlled by either its own solubility-limiting solid or the solubility of the UO_2 matrix (see Liebetrau et al. 1987, Appendix E), whichever is lower. Congruent releases from the UO_2 matrix (Figures 7.3 to 7.4) of all of the nuclides contained in the matrix are well below their NRC fractional release rates limits. Note that the release rates of all nuclides are lower for the congruent case than for the incongruent case, in some cases by a factor of almost 10 orders of magnitude.

The fractional release rate of gap (including grain boundary) nuclides is shown in Figure 7.5. These nuclides display the typical early "spike" in the release rate that arises from instantaneous dissolution of nuclides that are not solubility-controlled. Of these nuclides, only ^{79}Se and ^{99}Tc are released at a rate above their allowable NRC limits, and this occurs only for a relatively short (less than 200 years) duration. The long-term release

rates for all of these inventory-limited nuclides are at least a factor of 10 below their NRC limits, and continue to decrease with time.

Release of ^{14}C is assumed to occur from both the cladding itself and the crud/surface layer covering of the cladding (Figure 7.6). The release from the crud is inventory-limited, and displays the same characteristic spike as early release rate for ^{14}C . Releases of ^{14}C from both crud and cladding are, however, below the NRC limit for this nuclide.

Strontium-90 and ^{137}Cs are short-lived nuclides, with half-lives of approximately 30 years. Because of this, the inventory in curies of each of these nuclides 1000 years after repository closure falls below the cutoff for release rate consideration established by the NRC (10 CFR 60, 1983). For this case, with containment failure times significantly before 1000 years, both of these isotopes can represent a significant fraction of the total curie content of spent fuel. Figure 7.7 shows the combined release of these two nuclides from both UO_2 matrix (congruent dissolution) and gap sources. The initial fractional releases are high, largely because these releases are normalized by their 1000-year inventories. That is, at 300 years the total inventory of ^{90}Sr and ^{137}Cs is approximately 7 to 8 orders of magnitude greater than the 1,000 year inventories of these nuclides. An annual release rate of one part in 100,000 of the 300-year ^{90}Sr inventory corresponds to the release of approximately 10 times the entire 1000-year ^{90}Sr inventory. As Figure 7.7 shows, however, these artificially high release rates occur for a relatively short period of time; because of the rapid radioactive decay of these nuclides, their release rate decreases rapidly to values well below NRC limits.

Figures 7.8 through 7.13 show the cumulative release of radionuclides from all sources within spent fuel, normalized to their individual, allowable curie limits for 10,000 years (40 CFR 191, 1985). For the scenario of incongruent release from UO_2 , ^{241}Am exceeds its EPA limit approximately 700 years after containment failure (Figure 7.8), while ^{129}I , ^{135}Cs and ^{245}Cm are released at values greater than 10% of their EPA cumulative limit (Figure 7.9). All other nuclides are released well below their EPA limit (Figures 7.10 and 7.11). For the case of congruent release from UO_2

(Figure 7.12 and 7.13), ^{14}C , ^{99}Tc , ^{79}Se , ^{129}I , and ^{135}Cs dominate total cumulative release, although all nuclides are well below their EPA limits and the sum is about 3% of the allowable cumulative release.

7.2 CASE 2

Figures 7.14 to 7.25 present release rate calculations for the case in which all waste packages lose containment at 1000 years after permanent repository closure. This case is slightly less conservative but equally unrealistic to the previous one. The nuclides considered in this analysis have half-lives that are greater than 1000 years. For the long-lived nuclides, the difference between 300 and 1000 years of containment has a negligible effect on release rates. Therefore, it is expected that the release behavior for the case of total containment failure at 1000 years should closely match that described for total failure at 300 years (Section 7.1).

For the UO_2 matrix, Figures 7.14 and 7.15 show incongruent release and Figures 7.16 and 7.17 show congruent release. For the incongruent scenario, only ^{126}Ra , ^{129}I , and ^{135}Cs have release rates that exceed their NRC limits. In the congruent release scenario, no release rates exceed their NRC limits. The results shown in Figures 7.14 to 7.17 closely match those shown in Figures 7.1 to 7.4. The congruent release rates are all slightly lower for the 1000-year case; this is attributable to the lower temperature, and hence lower UO_2 matrix solubility, at 1000 years after repository closure than at 300 years.

The release rates of gap radionuclides, assuming total containment failure at 1000 years (Figure 7.18), are also nearly identical to those for the 300-year containment case (Figure 7.5). Only the gap releases of ^{79}Se and ^{99}Tc briefly exceed their NRC limits.

The situation that applies for the cladding is the same as for the gap. The release rate of ^{14}C is essentially identical for total containment failure at 1000 years (Figure 7.19) and at 300 years (Figure 7.6). In both cases, the calculated release rates for ^{14}C are below the NRC allowable limit.

Because of their short half-lives, ^{90}Sr and ^{137}Cs fall below the curie inventory cutoff at 1000 years established by the NRC (10 CFR 60, 1983). Therefore, both of these nuclides are dismissed from this analysis.

Figures 7.20 through 7.25 show the cumulative release of nuclides from all sources for the incongruent and congruent scenarios, respectively. For both scenarios, the cumulative releases are essentially identical to those for the 300-year containment case (Figures 7.8 through 7.13). Only ^{241}Am for the incongruent matrix release case is released above its EPA cumulative release limit.

7.3 CASE 3

Figures 7.26 to 7.38 present the nuclide release results for the case of a normal containment failure distribution, centered at 1000 years after repository closure. The distribution is truncated so that no containment failures occur prior to 300 years after repository closure.

The release from the UO_2 matrix is shown for both incongruent (Figures 7.26 and 7.27) and congruent (Figures 7.28 and 7.29) scenarios. Comparison of these figures with Figures 7.14 to 7.17 (Case 2) confirms that the distribution of containment failures over time attenuates release in two ways: 1) it diminishes the maximum value of the early "spike" in nuclide release rates, and 2) it displaces this maximum to later times. The steady-state release rates of nuclides are not, however, significantly affected by this distribution of containment failures. Except for ^{226}Ra , ^{129}I , and ^{135}Cs of the incongruent release scenario, all release rates are below their NRC limits. This result is identical to that for Cases 1 and 2, as discussed previously.

Gap nuclides are all released at rates below 10^{-5} parts per year for this case (Figure 7.30). In this case, the distribution of containment failures reduces and delays the maximum fractional release rate for gap nuclides in exactly the same manner as for matrix nuclides. The release rates of ^{14}C from cladding and crud show the same effect (Figure 7.31); the release rate of ^{14}C from the crud is well below the NRC limit for this case.

Figure 7.32 presents the release of ^{90}Sr and ^{137}Cs from gap and UO_2 matrix (congruent release) sources. Approximately 50% of containment failures occur before 1000 years; this results in gap fractional release rates greater than 10^{-5} parts per year for these two nuclides during the first several hundred years of release, but are lower than their extrapolated allowable release rates (see Table 4.1). Results in this case are lower than those for Case 1.

For Case 3, the cumulative release of nuclides from all sources are shown for both the incongruent (Figures 7.33 to 7.36) and congruent release (Figures 7.37 and 7.38). As for Cases 1 and 2, ^{241}Am from incongruent matrix release was the only nuclide in violation of its EPA limit. Relative to the instantaneous failure situations treated in Cases 1 and 2, the normal distribution of containment failures centered at 1000 years does not result in a significant reduction of the cumulative releases.

7.4 CASE 4

In this case, the effect of a normal distribution of containment failures centered at 5000 years on the radionuclide release from spent fuel is evaluated. This distribution has a standard deviation of 2000 years and is truncated to exclude failures prior to 300 years. This distribution is similar to the containment failure distribution used in the environmental assessment for BWIP (DOE 1986b). The plotted results described below show irregularities that are artifacts of the small number of early waste packages failures.

The fractional release rates are presented for incongruent release from the matrix (Figures 7.39 and 7.40) and congruent release from the matrix (Figures 7.41 and 7.42). Release rates from the gap (Figure 7.43) and cladding (Figure 7.44) are also shown. In addition, the combined fractional release rates of ^{90}Sr and ^{137}Cs from gap and matrix sources are shown in Figure 7.45. Only the incongruent release of ^{129}I , ^{135}Cs and, possibly, ^{226}Ra from the matrix exceed their NRC limits. All other nuclides, including ^{90}Sr and ^{137}Cs , are well below NRC limits.

The cumulative releases of nuclides from all sources for both incongruent and congruent matrix release are shown in Figures 7.46 through 7.51, respectively. As for the previous Cases 1 through 3, only ^{241}Am in the incongruent matrix scenario is released above its EPA cumulative limit. The normal distribution of containment times centered at 5000 years does reduce the cumulative release of many nuclides relative to Cases 1 through 3.

7.5 CASE 5

The release behavior of nuclides for an asymmetric distribution of containment failures is the last case to be considered. Specifically, containment failure is initiated 300 years after emplacement at a high rate that decreases exponentially over time in such a way that 50% of the failures occur before 1000 years.

Figures 7.52 and 7.53 show rates for incongruent matrix release and Figures 7.54 and 7.55 show rates for congruent release. The only nuclides released above their NRC limits are ^{226}Ra , ^{129}I and ^{135}Cs for the incongruent scenario. The dip in ^{129}I (Figure 7.53) at about 7000 years is attributable to absence of new containment failures and the relatively rapid transport of ^{129}I through the waste package. Figures 7.56 and 7.57 present nuclide release rates for gap and cladding release, respectively. Release rates for all nuclides are below their NRC limits. Figure 7.58 indicates that ^{90}Sr and ^{137}Cs releases from gap sources will exceed a release of 10^{-5} but are lower than their extrapolated allowable release rate.

Figures 7.59 through 7.64 show the cumulative nuclide releases from all sources for the scenarios of incongruent and congruent matrix release, respectively. Both of these cases show that all nuclides except ^{241}Am (incongruent matrix scenario) are released well below their 10,000-year allowable EPA limits. These releases are closely similar to those shown for Cases 1 through 3.

7.6 BASALT WASTE ISOLATION PROJECT CASE SUMMARY

If nuclides are released congruently from the UO_2 matrix, then the highest fractional release rates result from the gap (^{14}C , ^{79}Se , ^{99}Tc , ^{129}I ,

^{135}Cs and crud (^{14}C). It should be stressed, however, that even for the highly conservative and unrealistic assumption of total containment failure at 1000 years, the release rates of radionuclides from the gap and crud fall below allowable NRC limits; any distribution of failures further reduces releases.

If the matrix release is incongruent, then the releases of ^{226}Ra , ^{129}I , and ^{135}Cs from the matrix may also exceed their NRC limits for certain distributions of containment failure. In addition, the short-lived nuclides ^{90}Sr and ^{137}Cs can be released at fractional release rates considerably above 10^{-5} parts per year if there are a significant number of containment failures prior to 1000 years.

With respect to cumulative release, it was found for all cases evaluated that all nuclides except ^{241}Am (incongruent matrix scenario) were released at rates less than their allowable EPA limits. For the anticipated condition of congruent UO_2 matrix release, the sum of all nuclides released was only 3% of the allowable EPA limit. This is true even for the most conservative case of total containment failure at 300 years.

FIGURE 7.1. Release Rate of Actinides for Incongruent Matrix Release (BWIP Case 1)

FIGURE 7.2. Release Rate of Fission Products and ^{226}Ra for Incongruent Matrix Release (BWIP Case 1)

FIGURE 7.3. Release Rate of Actinides for Congruent Matrix Release (BWIP Case 1)

FIGURE 7.4. Release Rate of Fission Products and ^{226}Ra for Congruent Matrix Release (BWIP Case 1)

FIGURE 7.5. Release Rate of Fission Products for Gap Release (BWIP Case 1)

FIGURE 7.6. Release Rate of ^{14}C for Cladding Release (BWIP Case 1). "Crud" refers to the release from the surface layer on cladding; "Zr" refers to the release from the zircaloy cladding.

FIGURE 7.7. Release Rate of ⁹⁰Sr and ¹³⁷Cs for Gap Release (BWIP Case 1).

FIGURE 7.8. Cumulative Release, Multiplied by a Factor of 0.1, of Nuclides for Incongruent Release (BWIP Case 1)

FIGURE 7.9. Cumulative Release, Multiplied by a Factor of 0.1, of Nuclides for Incongruent Release (BWIP Case 1)

FIGURE 7.10. Cumulative Release, Multiplied by a Factor of 10, of Nuclides for Incongruent Release (BWIP Case 1)

FIGURE 7.11. Cumulative Release, Multiplied by a Factor of 100, of Nuclides for Incongruent Release (BWIP Case 1)

FIGURE 7.12. Cumulative Release, Multiplied by a Factor of 10, of Nuclides for Congruent Release (BWIP Case 1)

FIGURE 7.13. Cumulative Release, Multiplied by a Factor of 100, of Nuclides for Congruent Release (BWIP Case 1)

FIGURE 7.14. Release Rate of Actinides for Incongruent Matrix Release (BWIP Case 2)

FIGURE 7.15. Release Rate of Fission Products and ^{226}Ra for Incongruent Matrix Release (BWIP Case 2)

FIGURE 7.16. Release Rate of Actinides for Congruent Matrix Release (BWIP Case 2)

FIGURE 7.17. Release Rate of Fission Products and ^{226}Ra for Congruent Matrix Release (BWIP Case 2)

FIGURE 7.18. Release Rate of Fission Products for Gap Release (BWIP Case 2)

FIGURE 7.19. Release Rate of ^{14}C for Cladding Release (BWIP Case 2). "Crud" refers to the release from the surface layer or cladding; "Zr" refers to the release from the zircaloy cladding.

FIGURE 7.20. Cumulative Release, Multiplied by a Factor of 0.1, of Nuclides for Incongruent Release (BWIP Case 2)

FIGURE 7.21. Cumulative Release, Multiplied by a Factor of 1.0, of Nuclides for Incongruent Release (BWIP Case 2)

FIGURE 7.22. Cumulative Release, Multiplied by a Factor of 10, of Nuclides for Incongruent Release (BWIP Case 2)

FIGURE 7.23. Cumulative Release, Multiplied by a Factor of 100, of Nuclides for Incongruent Release (BWIP Case 2)

FIGURE 7.24. Cumulative Release, Multiplied by a Factor of 10, of Nuclides for Congruent Release (BWIP Case 2)

FIGURE 7.25. Cumulative Release, Multiplied by a Factor of 100, of Nuclides for Congruent Release (BWIP Case 2)

FIGURE 7.26. Release Rate of Actinides for Incongruent Matrix Release (BWIP Case 3)

FIGURE 7.27. Release Rate of Fission Products and ^{226}Ra for Incongruent Matrix Release (BWIP Case 3)

FIGURE 7.28. Release Rate of Actinides for Congruent Matrix Release (BWIP Case 3)

FIGURE 7.29. Release Rate of Fission Products and ^{226}Ra for Congruent Matrix Release (BWIP Case 3)

FIGURE 7.30. Release Rate of Fission Products for Gap Release (BWIP Case 3)

FIGURE 7.31. Release Rate of ^{14}C for Cladding Release (BWIP Case 3). "Crud" refers to the release from the surface layer on cladding; "Zr" refers to the release from the zircaloy cladding.

FIGURE 7.32. Release Rate of ^{90}Sr and ^{137}Cs for Gap Release (BWIP Case 3)

FIGURE 7.33. Cumulative Release, Multiplied by a Factor of 0.1, of Nuclides for Incongruent Release (BWIP Case 3)

FIGURE 7.34. Cumulative Release, Multiplied by a Factor of 1.0, of Nuclides for Incongruent Release (BWIP Case 3)

FIGURE 7.35. Cumulative Release, Multiplied by a Factor of 10, of Nuclides for Incongruent Release (BWIP Case 3)

FIGURE 7.36. Cumulative Release, Multiplied by a Factor of 100, of Nuclides for Incongruent Release (BWIP Case 3)

FIGURE 7.37. Cumulative Release, Multiplied by a Factor of 10, of Nuclides for Congruent Release (BWIP Case 3)

FIGURE 7.38. Cumulative Release, Multiplied by a Factor of 100, of Nuclides for Congruent Release (BWIP Case 3)

FIGURE 7.39. Release Rate of Actinides for Incongruent Matrix Release (BWIP Case 4)

FIGURE 7.40. Release Rate of Fission Products and ^{226}Ra for Incongruent Matrix Release (BWIP Case 4)

FIGURE 7.41. Release Rate of Actinides for Congruent Matrix Release (BWIP Case 4)

FIGURE 7.42. Release Rate of Fission Products and ²²⁶Ra Nuclides for Congruent Matrix Release (BWIP Case 4)

FIGURE 7.43. Release Rate of Fission Products for Gap Release (BWIP Case 4)

FIGURE 7.44. Release Rate of ^{14}C for Cladding Release (BWIP Case 4). "Crud" refers to the release from the surface layer on cladding; "Zr" refers to the release from the zircaloy cladding.

FIGURE 7.45. Release Rate of ^{90}Sr and ^{137}Cs for Gap Release (BWIP Case 4)

FIGURE 7.46. Cumulative Release, Multiplied by a Factor of 0.1, of Nuclides for Incongruent Release (BWIP Case 4)

FIGURE 7.47. Cumulative Release, Multiplied by a Factor of 1.0, of Nuclides for Incongruent Release (BWIP Case 4)

FIGURE 7.48. Cumulative Release, Multiplied by a Factor of 10, of Nuclides for Incongruent Release (BWIP Case 4)

FIGURE 7.49. Cumulative Release, Multiplied by a Factor of 100, of Nuclides for Incongruent Release (BWIP Case 4)

FIGURE 7.50. Cumulative Release, Multiplied by a Factor of 10, of Nuclides for Congruent Release (BWIP Case 4)

FIGURE 7.51. Cumulative Release, Multiplied by a Factor of 100, of Nuclides for Congruent Release (BWIP Case 4)

FIGURE 7.52. Release Rate of Actinides for Incongruent Matrix Release (BWIP Case 5)

FIGURE 7.53. Release Rate of Fission Products and ^{226}Ra for Incongruent Matrix Release (BWIP Case 5)

FIGURE 7.54. Release Rate of Actinides for Congruent Matrix Release (BWIP Case 5)

FIGURE 7.55. Release Rate of Fission Products and ^{226}Ra for Congruent Matrix Release (BWIP Case 5)

FIGURE 7.56. Release Rate of Fission Products for Gap Release (BWIP Case 5)

FIGURE 7.57. Release Rate of ^{14}C for Cladding Release (BWIP Case 5). "Crud" refers to the release from the surface layer on cladding; "Zr" refers to the release from the zircaloy cladding.

FIGURE 7.58. Release Rate of ^{90}Sr and ^{137}Cs for Gap Release (BWIP Case 5)

FIGURE 7.59. Cumulative Release, Multiplied by a Factor of 0.1, of Nuclides for Incongruent Release (BWIP Case 5)

FIGURE 7.60. Cumulative Release, Multiplied by a Factor of 1.0, of Nuclides for Incongruent Release (BWIP Case 5)

FIGURE 7.61. Cumulative Release, Multiplied by a Factor of 10, of Nuclides for Incongruent Release (BWIP Case 5)

FIGURE 7.62. Cumulative Release, Multiplied by a Factor of 100, of Nuclides for Incongruent Release (BWIP Case 5)

FIGURE 7.63. Cumulative Release, Multiplied by a Factor of 10, of Nuclides for Congruent Release (BWIP Case 5)

FIGURE 7.64. Cumulative Release, Multiplied by a Factor of 100, of Nuclides for Congruent Release (BWIP Case 5)

FIGURE 2.52. Cumulative Release, Multiplied by a Factor of 10, of Nucleoside for Conjugate Release (Case 2)

FIGURE 2.53. Cumulative Release, Multiplied by a Factor of 100, of Nucleoside for Conjugate Release (Case 3)

8.0 SPENT FUEL RELEASE RATE ANALYSIS FOR THE NEVADA NUCLEAR WASTE STORAGE INVESTIGATIONS

The graphical results and the discussion of the AREST analysis for NNWSI are presented in this section. The results and discussion are presented for each of the five container failure distribution cases.

8.1 CASE 1

Figures 8.1 to 8.12 present the AREST analyses of waste package releases for the conservative case in which containment failure occurs instantaneously at 300 years after permanent repository closure. Note that steam conditions are expected to occur for approximately the first 1500 years after repository closure. Because of this, many sources, such as the matrix and gap, do not release radionuclides until water conditions are reestablished (i.e., waste package temperatures fall below 95°C). The release behavior of short-lived nuclides (i.e., ^{90}Sr and ^{137}Cs) are not treated for any of the tuff cases because these nuclides have radioactively decayed to negligible amounts by the time they can be released from their sources.

Figures 8.1 to 8.2 show release rates of radionuclides from the UO_2 matrix under incongruent control. For the incongruent case, release rates for many nuclides are greater than their NRC limits. This arises because of the extremely high solubilities assumed by the NNWSI project (Kerrisk 1985) and the relatively short pathway between the waste form and the waste package boundary in their waste package design (DOE 1986c).

Figures 8.3 and 8.4 show release rates of radionuclides from the UO_2 matrix under congruent control. Release rates calculated for congruent release of nuclides from the matrix, however, indicate that all matrix nuclides are less than their allowable NRC limits. A critical factor for congruent release is the solubility of UO_2 . For these analyses, it was assumed that the local geochemical conditions at the waste form surface are such that UO_2 is the stable uranium phase. This assumes, therefore, that these conditions, particularly Eh, are controlled by local solids within the waste package (UO_2 , cladding, container) rather than by the repository

environment surrounding the waste package. If appropriate reaction and transport rates are known (Zavoshy, Chambré, and Pigford 1985; Liebetrau et al. 1987, Appendix D), this supposition can be (but has not been) tested.

The spike-like maxima for release rates from the gap (Figure 8.5) are close to, or slightly above, the allowable NRC release rates for ^{135}Cs , ^{129}I , ^{79}Se , ^{99}Tc , and ^{14}C . This is attributable to the instantaneous nature of the assumed containment failure and the relatively short transport pathway to the waste package boundary. Note, however, that the long-term release rates for all of these nuclides are considerably less (up to four orders of magnitude) than their allowable NRC limits.

Figure 8.6 shows ^{14}C release from the cladding and the crud layer on the cladding. The crud release of ^{14}C begins at the 300-year time of failure as gaseous diffusion of CO_2 . When steam conditions are superseded by water conditions (an average time of approximately 1500 years after repository closure), the release from the crud continues as aqueous diffusion of carbonate. The initial ^{14}C release from the crud is above the NRC limit because of the instantaneous nature of the containment failure, the relatively short transport pathway to the boundary of the waste package, and the extremely rapid rate of gaseous diffusion compared to aqueous diffusion. The long-term release of ^{14}C is well below its NRC limit. The undulating nature of ^{14}C release rate from the crud source results from the combination of releases that are occurring simultaneously from waste packages under steam and water conditions.

Cumulative release of nuclides from all sources have been calculated for scenarios assuming incongruent matrix release (Figures 8.7 to 8.10) or congruent matrix release (Figures 8.11 to 8.12). For this highly conservative containment case including incongruent matrix release (Figures 8.7 to 8.10), the following nuclides are released above their EPA limits: ^{241}Am , ^{240}Pu , ^{239}Pu , ^{14}C , ^{237}Np , ^{245}Cm , ^{99}Tc , and ^{243}Am . Consideration of the anticipated congruent UO_2 matrix release results in a tremendous decrease in cumulative releases (Figures 8.11 and 8.12) such that all nuclides and their sum are below allowable EPA limits.

8.2 CASE 2

The release of nuclides from spent fuel is shown in Figures 8.13 to 8.24 for the case of instantaneous containment failure 1000 years after repository closure. Because of the persistence of steam conditions up to about 1500 years after repository closure and the relatively long half-lives (more than 1000 years) of nuclides considered, the results of Case 2 are identical to those of Case 1 for matrix and gap release rates. This includes the results of the cumulate EPA release limits for incongruent (Figures 8.19 through 8.22) and congruent (Figures 8.23 and 8.24) matrix release.

The release rate of ^{14}C from the crud or surface layer of cladding (Figure 8.18) briefly exceeds its NRC limit, but then rapidly decreases to a value several orders of magnitude below this limit.

8.3 CASE 3

The effect of a normal distribution of containment failures centered at 1000 years on radionuclide release from spent fuel is calculated for this case of nuclides from all sources for the incongruent and congruent matrix release (Figures 8.25 to 8.36). The distribution is assumed to have a standard deviation of 200 years and failures prior to 300 years are not permitted. The highly variable fractional release rates shown at early times for several nuclides are attributable to the small number of waste packages that have failed and the relatively short transport pathway within the NNWSI waste package.

Incongruent release of nuclides from the UO_2 matrix (Figures 8.25 and 8.26) results in the same nuclides being released above their NRC limits for this case as for Cases 1 and 2. The fractional release rates of nuclides for the congruent matrix scenario (Figures 8.27 and 8.28) are far below their allowable NRC limits. A normal distribution of containment failures produces a significant effect for the release of nuclides from the gap (Figure 8.29) and cladding (Figure 8.30) sources of spent fuel. Comparing Figure 8.29 with Figures 8.5 and 8.17, for example, shows that the distributed failures attenuates the maximum release rates for all nuclides and delays this maximum to later times. The early release rate maximum of gap nuclides (Figure 8.29)

are near or slightly above their allowable NRC limits; the releases of ^{14}C from cladding and crud (Figure 8.30) are below the NRC limit for this nuclide.

Figures 8.31 through 8.36 display the cumulative releases of nuclides from spent fuel for the scenarios for incongruent and congruent matrix release, respectively. The effect of this normal distribution of containment failures on cumulative release is not appreciable compared to the instantaneous failure conditions of Cases 1 and 2.

8.4 CASE 4

Case 4 assumes a normal distribution containment failure as with Case 3, but the distribution is centered at 5000 years with a standard deviation of 2000 years. Containment failures prior to 300 years are not allowed. The calculated releases for this case (Figures 8.37 to 8.49) often display significant variation around an average trend that results from the small number of containment failures at early times and the short transport pathway for nuclides to the waste package boundary.

For the scenario of incongruent UO_2 matrix release (Figures 8.37 and 8.38), the same nuclides are released at fractional release rates greater than their NRC limits as for Cases 1 to 3. The NRC release rate limits are met by all nuclides for the congruent matrix release scenario (Figures 8.39 and 8.40). The release rates of all nuclides in the gap (Figure 8.41) and cladding (Figure 8.42) sources comply with their NRC limits.

Cumulative releases (Figures 8.43 through 8.49) show only slight changes between this distribution of containment failures and those for Cases 1 to 3. The cumulative releases for all nuclides are below their EPA limits for the anticipated congruent matrix release scenario. Note, however, that cumulative releases for the congruent scenario are higher for Case 4 than for Case 3. This seemingly contradictory result is because of the calculated higher UO_2 solubility as temperature decreases below 95°C for the groundwater equilibrated with tuff. In particular, slightly higher phosphate concentrations calculated for groundwater at 70°C than at 95°C leads to higher overall calculated UO_2 solubility. This in turn, for congruent release, leads to

higher overall releases for containers failing at later times. This slight, artificial effect arises because changing solubility with time (temperature) is not accounted for by AREST.

8.5 CASE 5

This case examines the effect of an exponential distribution of containment failures on the release of nuclides from spent fuel. Containment failure is assumed to start at 300 years after repository closure and to exponentially decrease, so that 1000 years after repository closure 50% of all waste packages have lost containment. The "spikes" in release of some nuclides arise because of their relatively rapid release from a limited number of individual packages that lose containment at long times after emplacement.

The results of this case (Figures 8.50 to 8.62) are essentially the same as for Cases 1 to 4. For incongruent matrix release, several nuclides are calculated to be released at fractional release rates in excess of their NRC limits (Figures 8.50 and 8.51); ^{241}Am , ^{240}Pu , ^{239}Pu , ^{14}C , ^{237}Np , ^{245}Cm , ^{99}Tc , and ^{243}Am display cumulative releases greater than their EPA limits (Figures 8.56 to 8.59). Congruent UO_2 matrix release (Figures 8.52, 8.53, and 8.60 to 8.62) leads to releases of all matrix nuclides that are well below their NRC and EPA limits. The release rates of gap nuclides (Figure 8.54) and ^{14}C from the crud (Figure 8.55) display early maxima that approach their NRC limits. The long-term release rates of these nuclides, however, are all several orders of magnitude below their NRC fractional release rate limits. None of the gap or cladding nuclides are found to have cumulative releases greater than their EPA limits.

8.6 NEVADA NUCLEAR WASTE STORAGE INVESTIGATIONS CASE SUMMARY

Incongruent release of nuclides from the UO_2 matrix, based on cited NNWSI references (Kerrisk 1985; DOE 1986c), can lead to fractional release rates of ^{237}Np , ^{245}Cm , ^{79}Se , ^{135}Cs , ^{129}I , ^{99}Tc , ^{226}Ra , and ^{14}C that exceed allowable NRC limits for certain assumed distributions of containment failure. Congruent release of nuclides from the UO_2 matrix, which has been

observed for spent fuel tests conducted by NNWSI (Wilson and Oversby 1985), leads to fractional release rates for all of these matrix nuclides that are at least four orders of magnitude below their NRC limits. Analyses of cumulative release from the UO_2 matrix indicate that, while many nuclides are released above their EPA cumulative limit for the incongruent matrix scenario, no nuclides are released in excess of their EPA limits for congruent matrix release and the containment distributions assumed here.

For nuclides contained in the gap and cladding sources of spent fuel, the fractional release rates comply with their NRC limits for the cases with distributed containment failures (Cases 3 to 5); only for unrealistic, instantaneous containment failures (Cases 1 and 2) are fractional release rates in excess of their NRC limits. The fractional release rates of nuclides for the NNWSI waste package design are particularly sensitive to distributed containment failures because of the relatively short (approximately 3 cm) pathway between the waste form and the waste package boundary. It must be stressed that even for the instantaneous failure cases, the cumulative releases of all matrix, gap and cladding nuclides under assumed anticipated conditions complied with EPA limits that are normally imposed far from the waste package.

FIGURE 8.1. Release Rate of Actinides for Incongruent Matrix Release (NNWSI Case 1)

FIGURE 8.2. Release Rate of Fission Products and ^{226}Ra for Incongruent Matrix Release (NNWSI Case 1)

FIGURE 8.3. Release Rate of Actinides for Congruent Matrix Release (NNWSI Case 1)

FIGURE 8.4. Release Rate of Fission Products and ^{226}Ra for Congruent Matrix Release (NNWSI Case 1)

FIGURE 8.5. Release Rate of Fission Products for Gap Release (NNWSI Case 1)

FIGURE 8.6. Release Rate of ^{14}C for Cladding Release (NNWSI Case 1). "Crud" refers to the release from the surface layer on cladding; "Zr" refers to the release from the zircaloy cladding.

FIGURE 8.7. Cumulative Release, Multiplied by a Factor of 0.001, of Nuclides for Incongruent Release (NNWSI Case 1)

FIGURE 8.8. Cumulative Release, Multiplied by a Factor of 0.01, of Nuclides for Incongruent Release (NNWSI Case 1)

FIGURE 8.9. Cumulative Release, Multiplied by a Factor of 0.1, of Nuclides for Incongruent Release (NNWSI Case 1)

FIGURE 8.10. Cumulative Release, Multiplied by a Factor of 1.0, of Nuclides for Incongruent Release (NNWSI Case 1)

FIGURE 8.11. Cumulative Release, Multiplied by a Factor of 10, of Nuclides for Congruent Release (NNWSI Case 1)

FIGURE 8.12. Cumulative Release, Multiplied by a Factor of 100, of Nuclides for Congruent Release (NNWSI Case 1)

FIGURE 8.13. Release Rate of Actinides for Incongruent Matrix Release (NNWSI Case 2)

FIGURE 8.14. Release Rate of Fission Products and ^{226}Ra for Incongruent Matrix Release (NNWSI Case 2)

FIGURE 8.15. Release Rate of Actinides for Incongruent Matrix Release (NNWSI Case 2)

FIGURE 8.16. Release Rate of Fission Products and ^{226}Ra for Congruent Matrix Release (NNWSI Case 2)

FIGURE 8.17. Release Rate of Fission Products for Gap Release (NNWSI Case 2)

FIGURE 8.18. Release Rate of ^{14}C for Cladding Release (NNWSI Case 2). "Crud" refers to the release from the surface layer on cladding; "Zr" refers to the release from the zircaloy cladding.

FIGURE 8.19. Cumulative Release, Multiplied by a Factor of 0.001, of Nuclides for Incongruent Release (NNWSI Case 2)

FIGURE 8.20. Cumulative Release, Multiplied by a Factor of 0.01, of Nuclides for Incongruent Release (NNWSI Case 2)

FIGURE 8.21. Cumulative Release, Multiplied by a Factor of 0.1, of Nuclides for Incongruent Release (NNWSI Case 2)

FIGURE 8.22. Cumulative Release, Multiplied by a Factor of 1.0, of Nuclides for Incongruent Release (NNWSI Case 2)

FIGURE 8.23. Cumulative Release, Multiplied by a Factor of 10, of Nuclides for Congruent Release (NNWSI Case 2)

FIGURE 8.24. Cumulative Release, Multiplied by a Factor of 100, of Nuclides for Congruent Release (NNWSI Case 2)

FIGURE 8.25. Release Rate of Actinides for Incongruent Matrix Release (NNWSI Case 3)

FIGURE 8.26. Release Rate of Fission Products and ^{226}Ra for Incongruent Matrix Release (NNWSI Case 3)

FIGURE 8.27. Release Rate of Actinides for Congruent Matrix Release (NNWSI Case 3)

FIGURE 8.28. Release Rate of Fission Products and ^{226}Ra for Congruent Matrix Release (NNWSI Case 3)

FIGURE 8.29. Release Rate of Fission Products for Gap Release (NNWSI Case 3)

FIGURE 8.30. Release Rate of ^{14}C for Cladding Release (NNWSI Case 3). "Crud" refers to the release from the surface layer on cladding; "Zr" refers to the release from the zircaloy cladding.

FIGURE 8.31. Cumulative Release, Multiplied by a Factor of 0.001, of Nuclides for Incongruent Release (NNWSI Case 3)

FIGURE 8.32. Cumulative Release, Multiplied by a Factor of 0.01, of Nuclides for Incongruent Release (NNWSI Case 3)

FIGURE 8.33. Cumulative Release, Multiplied by a Factor of 0.1, of Nuclides for Incongruent Release (NNWSI Case 3)

FIGURE 8.34. Cumulative Release, Multiplied by a Factor of 1.0, of Nuclides for Incongruent Release (NNWSI Case 3)

FIGURE 8.35. Cumulative Release, Multiplied by a Factor of 10, of Nuclides for Congruent Release (NNWSI Case 3)

FIGURE 8.36. Cumulative Release, Multiplied by a Factor of 100, of Nuclides for Congruent Release (NNWSI Case 3)

FIGURE 8.37. Release Rate of Actinides for Incongruent Matrix Release (NNWSI Case 4)

FIGURE 8.38. Release Rate of Fission Products and ^{226}Ra for Incongruent Matrix Release (NNWSI Case 4)

FIGURE 8.39. Release Rate of Actinides for Congruent Matrix Release (NNWSI Case 4)

FIGURE 8.40. Release Rate of Fission Products and ^{226}Ra for Congruent Matrix Release (NNWSI Case 4)

FIGURE 8.41. Release Rate of Fission Products for Gap Release (NNWSI Case 4)

FIGURE 8.42. Release Rate of ^{14}C for Cladding Release (NNWSI Case 4). "Crud" refers to the release from the surface layer on cladding; "Zr" refers to the release from the zircaloy cladding.

FIGURE 8.43. Cumulative Release, Multiplied by a Factor of 0.001, of Nuclides for Incongruent Release (NNWSI Case 4)

FIGURE 8.44. Cumulative Release, Multiplied by a Factor of 0.01, of Nuclides for Incongruent Release (NNWSI Case 4)

FIGURE 8.45. Cumulative Release, Multiplied by a Factor of 0.1, of Nuclides for Incongruent Release (NNWSI Case 4)

FIGURE 8.46. Cumulative Release, Multiplied by a Factor of 1.0, of Nuclides for Incongruent Release (NNWSI Case 4)

FIGURE 8.47. Cumulative Release, Multiplied by a Factor of 1.0, of Nuclides for Congruent Release (NNWSI Case 4)

FIGURE 8.48. Cumulative Release, Multiplied by a Factor of 10, of Nuclides for Congruent Release (NNWSI Case 4)

FIGURE 8.49. Cumulative Release, Multiplied by a Factor of 100, of Nuclides for Congruent Release (NNWSI Case 4)

FIGURE 8.50. Release Rate of Actinides for Incongruent Matrix Release (NNWSI Case 5)

FIGURE 8.51. Release Rate of Fission Products and ^{226}Ra for Incongruent Matrix Release (NNWSI Case 5)

FIGURE 8.52. Release Rate of Actinides for Congruent Matrix Release (NNWSI Case 5)

FIGURE 8.53. Release Rate of Fission Products and ^{226}Ra for Congruent Matrix Release (NNWSI Case 5)

FIGURE 8.54. Release Rate of Fission Products for Gap Release (NNWSI Case 5)

FIGURE 8.55. Release Rate of ^{14}C for Cladding Release (NNWSI Case 5). "Crud" refers to the release from the surface layer on cladding; "Zr" refers to the release from the zircaloy cladding.

FIGURE 8.56. Cumulative Release, Multiplied by a Factor of 0.001, of Nuclides for Incongruent Release (NNWSI Case 5)

FIGURE 8.57. Cumulative Release, Multiplied by a Factor of 0.01, of Nuclides for Incongruent Release (NNWSI Case 5)

FIGURE 8.58. Cumulative Release, Multiplied by a Factor of 0.1, of Nuclides for Incongruent Release (NNWSI Case 5)

FIGURE 8.59. Cumulative Release, Multiplied by a Factor of 1.0, of Nuclides for Incongruent Release (NNWSI Case 5)

FIGURE 8.60. Cumulative Release, Multiplied by a Factor of 1.0, of Nuclides for Congruent Release (NNWSI Case 5)

FIGURE 8.61. Cumulative Release, Multiplied by a Factor of 10, of Nuclides for Congruent Release (NNWSI Case 5)

FIGURE 8.62. Cumulative Release, Multiplied by a Factor of 100, of Nuclides for Congruent Release (NNWSI Case 5)

FIGURE 8.51 Cumulative Release, Multiplied by a Factor of 10, of Nucleoside for Congruent Release (HMSI Case 5)

FIGURE 8.52 Cumulative Release, Multiplied by a Factor of 100, of Nucleoside for Congruent Release (HMSI Case 5)

9.0 SPENT FUEL RATE ANALYSIS FOR THE SALT REPOSITORY PROJECT

The graphical results and the discussion of the AREST analysis for SRP are presented in this section. The results and discussion are presented for each of the five failure distribution cases.

9.1 CASE 1

Figures 9.1 to 9.13 present the AREST analyses of waste package releases for the case in which total containment failure occurs instantaneously at 300 years after permanent repository closure. This is the most conservative (and perhaps most unrealistic) case considered in this report.

Figures 9.1 and 9.2 show the fractional release rates of nuclides for the scenario of incongruent release from the UO_2 matrix. The ^{135}Cs and ^{129}I nuclides are released at rates in excess of their NRC limits. Because of their limited inventory in 1000-year old spent fuel, both ^{14}C and ^{226}Ra have allowable fractional release rates greater than 10^{-5} parts per year. For congruent release from the UO_2 matrix (Figures 9.3 and 9.4), all nuclides are released at rates far below their NRC limits. The fractional release rates of ^{135}Cs and ^{129}I , for example, are more than 13 orders of magnitude lower if their release is controlled by the solubility of the UO_2 matrix (congruent release) rather than by their own solubility-limiting solid (incongruent release).

The rates of nuclide release from the gap and cladding are shown in Figures 9.5 and 9.6, respectively. Release rates for all of the gap and cladding nuclides (^{14}C , ^{79}Se , ^{99}Tc , ^{129}I , ^{135}Cs) display initial maxima, or spikes, that approach, or even slightly exceed, the allowable NRC limits.

Figure 9.7 presents the release rates of the short-lived nuclides ^{90}Sr and ^{137}Cs from the gap and matrix (congruent release). These nuclides are released at extremely high fractional release rates when total containment failure at 300 years is assumed.

Cumulative releases of all nuclides from all sources are presented in Figures 9.8 through 9.13 for the incongruent and congruent matrix release scenarios. In both scenarios, the ratios of cumulative releases to their EPA

limits are well below the required value of 1 for all nuclides. For the incongruent release scenario, ^{14}C , ^{129}I , and ^{135}Cs are the dominant contributors, as might be expected from Figure 9.1. Note that while the ratio of cumulative release to the EPA limit is less than 1.0 for all nuclides, the "sum" is greater than 1.0. This would correspond to noncompliance with the overall EPA cumulative release criterion. For the case including congruent matrix release, releases of nuclides from the crud and gap (^{14}C , ^{99}Tc , ^{79}Se , ^{129}I) yield the largest fractions of their EPA limits. Note that the sum of the EPA release ratios is about 2% of the allowable limit for the congruent matrix release scenario.

9.2 CASE 2

Figures 9.14 to 9.25 show the radionuclide release behavior for the case in which all of the waste packages lose containment 1000 years after repository closure. Because all of the nuclides considered, except ^{90}Sr and ^{137}Cs , have half-lives that are longer than 1000 years, the results for this Case 2 are similar to those for Case 1. Note that nuclide fractional release rates for congruent matrix release are lower for Case 2 than Case 1 (compare Figures 9.3 and 9.4 with Figures 9.16 and 9.17). These release rates are attributable to the lower waste package temperatures, and therefore, lower UO_2 solubilities, at 1000 years after repository closure than at 300 years. The fractional release rates for ^{90}Sr and ^{137}Cs are not calculated in this case because they both fall below the 1000-year, curie-inventory cutoffs established by the NRC (10 CFR 60, 1983).

Cumulative releases of all nuclides for incongruent (Figures 9.20 to 9.23) and congruent (Figures 9.24 and 9.25) are essentially identical to Case 1. The dominant contributors for the case including incongruent matrix release are ^{14}C , ^{135}Cs , and ^{129}I from the matrix; for the congruent matrix release scenario, release of ^{14}C from the gap dominates.

9.3 CASE 3

Figures 9.26 to 9.38 present the nuclide release results for the case of a normal containment failure distribution centered at 1000 years after

repository closure. The distribution is truncated so that no containment failures occur prior to 300 years after repository closure.

For incongruent release from the UO_2 matrix (Figures 9.26 and 9.27), ^{129}I and ^{135}Cs releases are again the only nuclides that are released clearly above their allowable NRC fractional release rate limits. When congruent release of nuclides from the UO_2 matrix is evaluated (Figures 9.28 and 9.29), all of the nuclides are released at rates many orders of magnitude below their NRC limits.

The releases of nuclides from the gap (Figure 9.30) and cladding (Figure 9.31) sources are also well below allowable NRC limits. As noted in previous cases for BWIP and NNWSI, the effect of containment failure distributed over time is to suppress the spike-like maxima in release rates that are calculated for the instantaneous failure cases (Cases 1 and 2). Not only are the maximum release rate values decreased, but the time at which the maximum occurs is also delayed. The release rates of ^{90}Sr and ^{137}Cs from the gap and matrix sources is also calculated (Figure 9.32). The peak releases from both sources are near but below 10^{-5} parts per year, and well below their extrapolated allowable release limit (Table 4.1).

The cumulative releases of all nuclides from all sources for scenarios of incongruent (Figures 9.33 to 9.36) and congruent (Figures 9.37 and 9.38) matrix releases are evaluated. The releases for the congruent scenario are well below both the allowable EPA limits. The effect of normally distributed containment failures centered at 5000 years is to slightly decrease cumulative releases relative to Cases 1 and 2.

9.4 CASE 4

In this case, the effect of a normal distribution of containment failures (centered at 5000 years) on the release of radionuclides from spent fuel is considered. The distribution is assumed to have a standard deviation of 2000 years and is truncated to exclude failures prior to 300 years. Because of the limited number of early containment failures observed, the fractional release rates tend to display noticeable fluctuations initially.

Figures 9.39 to 9.51 give the results of release calculations for this case. Only for the scenario of incongruent matrix release (Figure 9.39) are any nuclides (^{129}I and ^{135}Cs) released at rates above their allowable NRC limits. Based on their inventories at 1000 years, both ^{90}Sr and ^{137}Cs (Figure 9.45) are also released at rates below their NRC limits. The ^{14}C , ^{129}I , and ^{135}Cs nuclides are also the dominant contributors to the cumulative release for the case including incongruent matrix release (Figures 9.46 to 9.49). The normally distributed containment failures centered at 5000 years effectively prevents the "Sum" of ratios of individual nuclides from exceeding 1.0. All nuclides are released well below their EPA cumulative release limits for the congruent UO_2 matrix release scenario (Figures 9.50 and 9.51).

9.5 CASE 5

Case 5 evaluates the effect of an asymmetric distribution of containment failure on radionuclide release. Specifically, containment failure is initiated 300 years after emplacement at a high rate that decreases exponentially over time in such a way that 50% of the failures occur before 1000 years.

Radionuclide release rates have been calculated for a number of sources, including incongruent release from the UO_2 matrix (Figures 9.52 and 9.53), congruent release from the matrix (Figures 9.54 and 9.55), the gap (Figure 9.56), and the cladding (Figure 9.57). In addition, the release rate of ^{90}Sr and ^{137}Cs from gap and matrix sources has been evaluated (Figure 9.58). Only the fractional release rates for ^{135}Cs and ^{129}I for the incongruent matrix release scenario are greater than their NRC limits. The dip in ^{129}I release (Figure 9.53) at about 7000 years is attributable to an absence of new container failures and the relatively rapid transport of ^{129}I through the waste package. These nuclides are eliminated from consideration if the UO_2 matrix congruent release scenario applies. Both ^{90}Sr and ^{137}Cs are released at rates greater than 10^{-5} parts per year, but the rates are still below their extrapolated allowable NRC limits (see Table 4.1).

For individual nuclides, the ratios of cumulative releases from all sources to their EPA limits (Figures 9.59 to 9.62 and 9.63 and 9.64) are below the value of 1, as required by the EPA. The "Sum" of the individual

release ratios is, however, slightly greater than 1 for the incongruent matrix release scenarios. As for Cases 1 to 4, ^{14}C , ^{129}I , and ^{135}Cs are the dominant contributors to the EPA cumulative release "Sum." For the congruent matrix scenario, the "Sum" is about 2% of the allowable EPA limit.

9.6 SALT REPOSITORY REPORT CASE SUMMARY

Iodine-129 and ^{135}Cs are released at rates in excess of their allowable NRC limits if the UO_2 matrix undergoes incongruent release of nuclides. The distribution of containment failure over time attenuates the excess, but did not eliminate it. If the release of nuclides from the UO_2 matrix is congruent, however, there are significant decreases (of up to 13 orders of magnitude relative to the incongruent scenario) in the fractional release rates of these two nuclides, as well as in the release rates of other nuclides contained in the UO_2 matrix.

The release rates of nuclides from the gap and cladding are well below their NRC limits for all cases in which the containment failures are distributed over time (Cases 3 through 5). Release rates from these sources for instantaneous containment failures (Cases 1 and 2) approach and may slightly exceed NRC limits; however, these cases are considered to be extremely conservative and unrealistic.

In all cases examined, the ratios of cumulative release to the EPA limit for individual nuclides were below the required value of 1. For the scenarios including incongruent matrix release, cumulative releases are dominated by ^{14}C , ^{129}I , and ^{135}Cs ; for congruent matrix release scenarios, the cumulative release is dominated by ^{14}C . The sum of the ratios of the individual nuclides is less than 1 for all radionuclides for congruent matrix release, and only slightly greater than 1 for incongruent matrix release.

FIGURE 9.1. Release Rate of Actinides for Incongruent Matrix Release (SRP Case 1)

FIGURE 9.2. Release Rate of Fission Products and ^{226}Ra for Incongruent Matrix Release (SRP Case 1)

FIGURE 9.3. Release Rate of Actinides for Congruent Matrix Release (SRP Case 1)

FIGURE 9.4. Release Rate of Fission Products and ^{226}Ra for Congruent Matrix Release (SRP Case 1)

FIGURE 9.5. Release Rate of Fission Products for Gap Release (SRP Case 1)

FIGURE 9.6. Release Rate of ^{14}C for Cladding Release (SRP Case 1). "Crud" refers to the release from the surface layer on cladding; "Zr" refers to the release from the zircaloy cladding.

FIGURE 9.7. Release Rate of ^{90}Sr and ^{137}Cs for Gap Release (SRP Case 1)

FIGURE 9.8. Cumulative Release, Multiplied by a Factor of 0.1, of Nuclides for Incongruent Release (SRP Case 1)

FIGURE 9.9. Cumulative Release, Multiplied by a Factor of 1.0, of Nuclides for Incongruent Release (SRP Case 1)

FIGURE 9.10. Cumulative Release, Multiplied by a Factor of 100, of Nuclides for Incongruent Release (SRP Case 1)

FIGURE 9.11. Cumulative Release, Multiplied by a Factor of 1000, of Nuclides for Incongruent Release (SRP Case 1)

FIGURE 9.12. Cumulative Release, Multiplied by a Factor of 10, of Nuclides for Congruent Release (SRP Case 1)

FIGURE 9.13. Cumulative Release, Multiplied by a Factor of 100, of Nuclides for Congruent Release (SRP Case 1)

FIGURE 9.14. Release Rate of Actinides for Incongruent Matrix Release (SRP Case 2)

FIGURE 9.15. Release Rate of Fission Products and ^{226}Ra for Incongruent Matrix Release (SRP Case 2)

FIGURE 9.16. Release Rate of Actinides for Congruent Matrix Release (SRP Case 2)

FIGURE 9.17. Release Rate of Fission Products and ^{226}Ra for Congruent Matrix Release (SRP Case 2)

FIGURE 9.18. Release Rate of Fission Products for Gap Release (SRP Case 2)

FIGURE 9.19. Release Rate of ^{14}C for Cladding Release (SRP Case 2). "Crud" refers to the release from the surface layer on cladding; "Zr" refers to the release from the zircaloy cladding.

FIGURE 9.20. Cumulative Release, Multiplied by a Factor of 0.1, of Nuclides for Incongruent Release (SRP Case 2)

FIGURE 9.21. Cumulative Release, Multiplied by a Factor of 1.0, of Nuclides for Incongruent Release (SRP Case 2)

FIGURE 9.22. Cumulative Release, Multiplied by a Factor of 100, of Nuclides for Incongruent Release (SRP Case 2)

FIGURE 9.23. Cumulative Release, Multiplied by a Factor of 1000 of Nuclides for Incongruent Release (SRP Case 2)

FIGURE 9.24. Cumulative Release, Multiplied by a Factor of 10, of Nuclides for Congruent Release (SRP Case 2)

FIGURE 9.25. Cumulative Release, Multiplied by a Factor of 100, of Nuclides for Congruent Release (SRP Case 2)

FIGURE 9.26. Release Rate of Actinides for Incongruent Matrix Release (SRP Case 3)

FIGURE 9.27. Release Rate of Fission Products and ^{226}Ra for Incongruent Matrix Release (SRP Case 3)

FIGURE 9.28. Release Rate of Actinides for Congruent Matrix Release (SRP Case 3)

FIGURE 9.29. Release Rate of Fission Products and ^{226}Ra for Congruent Matrix Release (SRP Case 3)

FIGURE 9.30. Release Rate of Fission Products for Gap Release (SRP Case 3)

FIGURE 9.31. Release Rate of ^{14}C for Cladding Release (SRP Case 3). "Crud" refers to the release from the surface layer on cladding; "Zr" refers to the release from the zircaloy cladding.

FIGURE 9.32. Release Rate of ^{90}Sr and ^{137}Cs for Gap Release (SRP Case 3)

FIGURE 9.33. Cumulative Release, Multiplied by a Factor of 0.1, of Nuclides for Incongruent Release (SRP Case 3)

FIGURE 9.34. Cumulative Release, Multiplied by a Factor of 1.0, of Nuclides for Incongruent Release (SRP Case 3)

FIGURE 9.35. Cumulative Release, Multiplied by a Factor of 100, of Nuclides for Incongruent Release (SRP Case 3)

FIGURE 9.36. Cumulative Release, Multiplied by a Factor of 1000, of Nuclides for Incongruent Release (SRP Case 3)

FIGURE 9.37. Cumulative Release, Multiplied by a Factor of 10, of Nuclides for Congruent Release (SRP Case 3)

FIGURE 9.38. Cumulative Release, Multiplied by a Factor of 100, of Nuclides for Congruent Release (SRP Case 3)

FIGURE 9.39. Release Rate of Actinides for Incongruent Matrix Release (SRP Case 4)

FIGURE 9.40. Release Rate of Fission Products and ^{226}Ra for Incongruent Matrix Release (SRP Case 4)

FIGURE 9.41. Release Rate of Actinides for Congruent Matrix Release (SRP Case 4)

FIGURE 9.42. Release Rate of Fission Products and ²²⁶Ra for Congruent Matrix Release (SRP Case 4)

FIGURE 9.43. Release Rate of Fission Products for Gap Release (SRP Case 4)

FIGURE 9.44. Release Rate of ^{14}C for Cladding Release (SRP Case 4). "Crud" refers to the release from the surface layer on cladding; "Zr" refers to the release from the zircaloy cladding.

FIGURE 9.45. Release Rate of ^{90}Sr and ^{137}Cs for Gap Release (SRP Case 4)

FIGURE 9.46. Cumulative Release, Multiplied by a Factor of 0.1, of Nuclides for Incongruent Release (SRP Case 4)

FIGURE 9.47. Cumulative Release, Multiplied by a Factor of 1.0, of Nuclides for Incongruent Release (SRP Case 4)

FIGURE 9.48. Cumulative Release, Multiplied by a Factor of 100, of Nuclides for Incongruent Release (SRP Case 4)

FIGURE 9.49. Cumulative Release, Multiplied by a Factor of 1000, of Nuclides for Incongruent Release (SRP Case 4)

FIGURE 9.50. Cumulative Release, Multiplied by a Factor of 10, of Nuclides for Congruent Release (SRP Case 4)

FIGURE 9.51. Cumulative Release, Multiplied by a Factor of 100, of Nuclides for Congruent Release (SRP Case 4)

FIGURE 9.52. Release Rate of Actinides for Incongruent Matrix Release (SRP Case 5)

FIGURE 9.53. Release Rate of Fission Products and ^{226}Ra for Incongruent Matrix Release (SRP Case 5)

FIGURE 9.54. Release Rate of Actinides for Congruent Matrix Release (SRP Case 5)

FIGURE 9.55. Release Rate of Fission Products and ^{226}Ra for Congruent Matrix Release (SRP Case 5)

FIGURE 9.56. Release Rate of Fission Products for Gap Release (SRP Case 5)

FIGURE 9.57. Release Rate of ^{14}C for Cladding Release (SRP Case 5). "Crud" refers to the release from the surface layer on cladding; "Zr" refers to the release from the zircaloy cladding.

FIGURE 9.58. Release Rate of ^{90}Sr and ^{137}Cs for Gap Release (SRP Case 5)

FIGURE 9.59. Cumulative Release, Multiplied by a Factor of 0.1, of Nuclides for Incongruent Release (SRP Case 5)

FIGURE 9.60. Cumulative Release, Multiplied by a Factor of 1.0, of Nuclides for Incongruent Release (SRP Case 5)

FIGURE 9.61. Cumulative Release, Multiplied by a Factor of 100, of Nuclides for Incongruent Release (SRP Case 5)

FIGURE 9.62. Cumulative Release, Multiplied by a Factor of 1000, of Nuclides for Incongruent Release (SRP Case 5)

FIGURE 9.63. Cumulative Release, Multiplied by a Factor of 10, of Nuclides for Congruent Release (SRP Case 5)

FIGURE 9.64. Cumulative Release, Multiplied by a Factor of 100, of Nuclides for Congruent Release (SRP Case 5)

FIGURE 2.53 Cumulative Release, Multiplied by a Factor of 10, of Material for Component Release (50% Case 2)

FIGURE 2.54 Cumulative Release, Multiplied by a Factor of 100, of Material for Component Release (50% Case 2)

10.0 PROBABILISTIC DISTRIBUTION OF NUCLIDE RELEASE RATES AT ALL THREE REPOSITORIES

The calculated variation in release rates of ^{237}Np and ^{129}I for Case 2 (normal distribution of containment failures centered at 1000 years) at all three repositories is plotted in Figures 10.1 to 10.10 for times 1000 and 5000 years after permanent repository closure. Congruent release from the UO_2 matrix is assumed. Note that no plots are provided from the tuff case at 1000 years because aqueous conditions have not yet been reestablished at this time.

The relative range in release rates for both nuclides is less at 5000 years than at 1000 years. This is attributable to the fact that at 5000 years most waste packages have lost containment and have closely approached a steady-state release rate. For release at 1000 years, the rate of containment failure is the highest and most of the failed containers display highly variable, transient release rates.

The variability in the release rates of the two nuclides can also be evaluated. The variation in the release rate of ^{237}Np arises from both the distribution of containment failure times and the range in waste package temperatures. This range in temperatures creates a range in groundwater within the waste package, both of which contribute to the range in UO_2 solubility (hence, congruent matrix release rates). By comparison, the gap is the dominant source for ^{129}I release. As such, the variation in ^{129}I release rates results solely from the distribution containment failures and is independent of temperature.

FIGURE 10.1. Variation in ^{237}Np Release Rate at 1000 Years for BWIP Case 3

FIGURE 10.2. Variation in ^{237}Np Release Rate at 5000 Years for BWIP Case 3

FIGURE 10.3. Variation in ^{129}I Release Rate at 1000 Years for BWIP Case 3

FIGURE 10.4. Variation in ^{129}I Release Rate at 5000 Years for BWIP Case 3

FIGURE 10.5. Variation in ^{237}Np Release Rate at 1000 Years for SRP Case 3

FIGURE 10.6. Variation in ^{237}Np Release Rate at 5000 Years for SRP Case 3

FIGURE 10.7. Variation in ^{129}I Release Rate at 1000 Years for SRP Case 3

FIGURE 10.8. Variation in ^{129}I Release Rate at 5000 Years for SRP Case 3

FIGURE 10.9. Variation in ^{237}Np Release Rate at 5000 Years for NNSWI Case 3

FIGURE 10.10. Variation in ^{129}I Release Rate at 5000 Years for NNWSI Case 3

REFERENCES

- Alexander, D. H., M. J. Apled, A. M. Liebetrau, A. E. Van Luik, R. E. Williford, and P. G. Doctor. 1985. "Conceptual Model for Deriving the Repository Source Term." In Scientific Basis for Nuclear Waste Management VIII, eds. C. M. Jantzen, J. A. Stone, and R. C. Ewing, pp. 439-450. Materials Research Society, Pittsburgh, Pennsylvania.
- Apled, M. J., D. H. Alexander, A. M. Liebetrau, A. E. Van Luik, R. E. Williford, and P. G. Doctor. 1985. "A Conceptual Model for Repository Source-Term Evaluation." In Proceedings of the Workshop on the Source Term Realistic Repository Conditions, eds. T. Hunter and A. Muller, pp. 125-136. SAND85-0380, Sandia National Laboratory, Albuquerque, New Mexico.
- Baker, D. A., W. J. Bailey, C. E. Beyer, F. C. Bold, and J. J. Tawil. 1987. Assessment of the Use of Extended Burnup Fuel in Light Water Power Reactors. PNL-6258, Pacific Northwest Laboratory, Richland, Washington.
- Barner, J. O. 1985. Characterization of LWR Spent Fuel MCC-Approved Testing Material - ATM-101. PNL-5109, Rev. 1, Pacific Northwest Laboratory, Richland, Washington.
- Bensky, M. S., and D. L. Oliver. 1985. "Transient Diffusional Release from Waste Packages in a Repository in Basalt." In Scientific Basis for Nuclear Waste Management IX, ed. L. O. Werme, pp. 779-787. Materials Research Society, Pittsburgh, Pennsylvania.
- Chambré, P. L., T. H. Pigford, W. W.-L. Lee, J. Ahn, S. Kajiwarra, C. L. Kim, H. Kimura, H. Lung, W. J. Williams, and S. Zavoshy. 1985. Mass Transfer and Transport in a Geologic Environment. LBL-19430, Lawrence Livermore Berkeley Laboratory, Berkeley, California.
- Disposal of Nuclear Radioactive Waste in Geologic Repositories. Title 10 CFR, Part 60 (1983). (Cited in text as 10 CFR 60, 1983.)
- Environmental Radiation Protection Standards for Management and Disposal of Spent Nuclear Fuel, High-Level and Transuranic Radioactive Wastes. Title 40 CFR Part 191 (1985). (Cited in text as 40 CFR 191, 1985.)
- Forsyth, R. S., and L. O. Werme. 1985. "The Corrosion of Spent UO₂ Fuel in Synthetic Groundwater." In Scientific Basis for Nuclear Waste Management IX, ed. L. O. Werme, pp. 327-336. Materials Research Society, Pittsburgh, Pennsylvania.
- Garisto, F., and N. Garisto. 1985. "A UO₂ Solubility Function for the Assessment of Used Nuclear Fuel Disposal." Nucl. Sci. Eng. 90:103-110.
- Garisto, N. C., and F. Garisto. 1986. "The Effect of Precipitation on the Long-Term Release of Radionuclides from Used Fuel." Ann. Nucl. Energy 13(11):591-596.

Johnson, L. H., D. W. Shoesmith, G. E. Lunansky, M. G. Bailey, and P. R. Tremaine. 1982. "Mechanisms of Leaching and Dissolution of UO_2 Fuel." Nucl. Technol. 56(2):238-253.

Johnson, L. H., S. Stroes-Gascoyne, D. W. Shoesmith, M. G. Bailey, and D. M. Sellinger. 1984. "Leaching and Radiolysis Studies on UO_2 Fuel." In Proceedings of the third spent fuel workshop, ed. L. Werme. SKBF/KBS, Technical Report 83-76, SKBF/KBS, Stockholm, Sweden.

Johnson, L. H., N. C. Garisto and S. Stroes-Gascoyne. 1985. "Used-Fuel Dissolution Studies in Canada." In Waste Management '85: Proceedings of the 1985 Symposium on Waste Management, ed. R. G. Post, Vol. 1, pp. 479-481. University of Arizona, Tucson, Arizona.

Karnbranslesakerhet (KBS). 1983. Final Storage of Spent Nuclear Fuel - KBS-3. Karnbranslesakerhet Report, SKBF/KBS, Stockholm, Sweden.

Kerrisk, J. F. 1985. "Solubility Limits on Radionuclide Dissolution." In Scientific Basis for Nuclear Waste Management VIII, eds. C. M. Jantzen, J. A. Stone, R. C. Ewing, pp. 237-244. Materials Research Society, Pittsburgh, Pennsylvania.

Kim, C., P. Chambré, and T. Pigford. 1986. Mass-Transfer-Limited Release of a Soluble Waste Species. LBL-20899, Lawrence Berkeley Laboratory, Berkeley, California.

LeNeveu, D. M. 1986. Vault Submodel for the Second Interim Assessment of the Canadian Concept for Nuclear Fuel Waste Disposal: Post-Closure Phase. AECL-8383, Atomic Energy of Canada, Ltd., Whiteshell Nuclear Research Establishment, Pinawa, Manitoba.

Lieberman, J. A., S. N. Davis, D. R. F. Harleman, R. L. Keeney, D. C. Kocher, D. Langmuir, R. B. Lyon, W. W. Owens, T. H. Pigford, and W. W.-L. Lee. 1985. Performance Assessment National Review Group. RFW-CRWM-85-01, Roy F. Weston, Washington, D.C.

Liebetrau, A. M., M. J. Apted, D. W. Engel, M. K. Altenhofen, C. R. Reid, D. M. Strachan, R. L. Erikson, and K. I. Johnson. 1987. The Analytical Repository Source-Term (AREST) Model: Description and Documentation. PNL-6346, Pacific Northwest Laboratory, Richland, Washington.

McKinley, I. G. and J. Hadermann. 1984. Radionuclide Sorption Database for Swiss Safety Assessment. EIR-Bericht Nr. 550, Swiss Federal Institute for Reactor Research, Wurenlingen, Switzerland.

Murphy, W. M., and R. W. Smith. 1987. Irreversible Dissolution of Solid Solutions: A Kinetic and Stoichiometric Model. WHC-SA-0090, Westinghouse Hanford Company, Richland, Washington.

National Academy of Sciences (NAS). 1983. A Study of the Isolation System for Geologic Disposal of Radioactive Wastes, National Academy Press, Washington, D.C.

Neretnieks, I. 1984. "The Impact of Alpha-Radiolysis on the Release of Radionuclides from Spent Fuel in a Geologic Repository." In Scientific Basis for Nuclear Waste Management VII, ed. G. L. McVay, pp. 1009-1022. Elsevier, Amsterdam, Netherlands.

Nuclear Waste Policy Act of 1982 (NWPA). 1983. Public Law 97-425, U.S. Code. Title 42, Sec. 10101-10226.

Oliver, D. L. 1985. Parametric Analysis of Release of Radionuclides from a Waste Package in Basalt. SD-BW1-TI281, Rockwell Hanford Operations, Richland, Washington.

Oversby, V. M. 1987. "Spent Fuel as a Waste Form: Data Needs to Allow Long-Term Performance Assessment Under Repository Disposal Conditions." In Scientific Basis for Nuclear Waste Management, Vol. X, ed. J. K. Bates and W. B. Seefeldt, pp. 87-101. Materials Research Society, Pittsburgh, Pennsylvania.

Oversby, V. M., and R. D. McCright. 1984. Laboratory Experiments Designed to Provide Limits on the Radionuclide Source Term for the NNWSI Project. UCRL-91257, Lawrence Livermore National Laboratory, Livermore, California.

Oversby, V., and C. N. Wilson. 1985. "Derivation of a Waste Package Source Term for NNWSI." In Scientific Basis for Nuclear Waste Management IX, ed. L. O. Werme, pp. 337-347. Materials Research Society, Pittsburgh, Pennsylvania.

Pigford, T. H. 1986. Can ^{137}Cs be Dismissed Under the NRC Release-Rate Criterion for Geologic Repositories? UCB-NE-4088, University of California, Berkeley, California.

Pigford, T. H. and P. L. Chambré. 1985. Mass Transfer in a Salt Repository. LBL-19918, Lawrence Berkeley Laboratory, Berkeley, California.

Pigford, T. H. and P. L. Chambré. 1986. "Reliable Predictions of Waste Performance in a Geologic Repository." In High-Level Nuclear Waste Disposal, ed. H. C. Burkholder, pp. 163-186. Battelle Press, Columbus, Ohio.

Ratigan, J. L. 1984. "A Finite Element Formulation for Brine Transport in Rock Salt." International Journal for Numerical and Analytical Methods in Geomechanics 8:225.

Rimstidt, J. D., and P. M. Dove. 1986. "Mineral/Solution Reaction Rates in a Mixed Flow Reactor: Wollastonite Hydrolysis." Geochim. Cosmochim. Acta 50:2509-2516.

Roddy, J. W., H. C. Clairborne, R. C. Ashline, P. J. Johnson, and B. T. Rhyne. 1986. Physical and Decay Characteristics of Commercial LWR Spent Fuel. ORNL/TM-9591/V1&R1, Oak Ridge National Laboratory, Oak Ridge, Tennessee.

Sagar, B., P. W. Eslinger and R. G. Baca. 1984. Probabilistic Modeling of Radionuclide Release at the Waste Packages Engineered Boundary of a Repository in Basalts. Draft RHO-BW-ST-68P, Rockwell Hanford Operations, Richland, Washington.

Sastre, C., C. Pescatore and T. Sullivan. 1986. Waste Package Reliability. NUREG/CR-4509, U.S. Nuclear Regulatory Commission, Washington, D.C.

Shay, M. R. and M. E. Buxbaum. 1986. WASTES II: Waste System Transportation and Economic Simulation - Version II. Users Guide. PNL- 5714, Pacific Northwest Laboratory, Richland, Washington.

U.S. Department of Energy (DOE). 1984. Generic Requirements for a Mined Geologic Disposal System. DOE/OGR/B-2, Office of Civilian Radioactive Waste Management, Washington, D.C.

U.S. Department of Energy (DOE). 1985. Mission Plan for the Civilian Radioactive Waste Management Program. DOE/RW-0005, Volume 1, Office of Civilian Radioactive Waste Management, Washington, D.C.

U.S. Department of Energy (DOE). 1986a. Environmental Assessment: Deaf Smith County, Texas. DOE/RW-0069, Vol. 2 of 3, Office of Civilian Radioactive Waste Management, Washington, D.C.

U.S. Department of Energy (DOE). 1986b. Environmental Assessment: Reference Repository Location, Hanford Site, Washington. DOE/RW-0070, Vol. 2, Office of Civilian Radioactive Waste Management, Washington, D.C.

U.S. Department of Energy (DOE). 1986c. Environmental Assessment: Yucca Mountain Site, Nevada Research and Development Area, Nevada. DOE/RW-0073, Vol. 2 of 3, Office of Civilian Radioactive Waste Management, Washington, D.C.

"Disposal of High-Level Radioactive Wastes in Geologic Repositories; Conforming Amendments," 51 Federal Register 22288-22301. (Cited in text as 51 FR 22288-22301.)

Van Konyneburg, R., C. Smith, H. Culham, and C. Otto, Jr. 1986. "Behavior of Carbon-14 in Waste Packages for Spent Fuel in a Repository in Tuff." In Scientific Basis for Nuclear Waste Management VIII, eds. C. Jantzen, J. Stone, and R. Ewing, pp. 405-412. Elsevier, Amsterdam, Netherlands.

Wilson, C. N. 1985. Results from NNWSI Series 1 Spent Fuel Leach Tests, HEDL-TME 84-30, Hanford Engineering Development Laboratory, Richland, Washington.

Wilson, C., and V. Oversby. 1985. "Radionuclide Release from PWR Fuels in A Reference Tuff Repository Groundwater." In Waste Management 85: Proceedings of the 1985 Symposium on Waste Management, p. 497. University of Arizona, Tucson, Arizona.

Wilson, C. N., R. E. Einziger, R. E. Woodley, and V. M. Oversby. 1985. LWR Spent Fuel Characteristics Relevant to Performance as a Waste Form in a Potential Tuff Repository. UCRL-92891, Lawrence Livermore National Laboratory, Livermore, California.

Wuertz, R., and M. Ellinger. 1985. "Source Term for the Activity Release from a Repository for Spent Fuel." In Scientific Basis for Nuclear Waste Management IX, ed. L. O. Werme, pp. 392-400. Materials Research Society, Pittsburgh, Pennsylvania.

Yung, S. C., C. F. McLane, R. P. Anantatmula, R. T. Toyooka, and W. K. Terry. 1987. Waste Package Preliminary Reliability Analysis Report. SO-BWI-TI-287, Basalt Waste Isolation Project, Rockwell Hanford Operations, Richland, Washington.

Zavoshy, S. J., P. L. Chambré and T. H. Pigford. 1985. "Mass Transfer in a Geologic Environment." In Scientific Basis for Nuclear Waste Management VIII, eds. C. M. Jantzen, J. A. Stone and R. C. Ewing, pp. 311-322. Materials Research Society, Pittsburgh, Pennsylvania.

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