SPENT FUEL AS A WASTE FORM: ANALYSIS WITH AREST PERFORMANCE ASSESSMENT CODE

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ABSTRACT

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 with the AREST code. Separate scenarios involving incongruent and congruent release from the UO_2 matrix have also been examined. Based on current data, congruent release from the UO_2 matrix is considered the anticipated condition for all three repository sites. Releases for individual radionuclides have been evaluated relative to compliance with allowable NRC fractional release rates and EPA 10,000-year cumulative release limits.

For the expected congruent matrix release scenario, spent fuel is predicted to meet all EPA and NRC release criteria for the distributed containment failures considered in this evaluation. However, for the incongruent matrix release scenario, releases of several nuclides are found to be above their NRC limits. Additional studies, especially to determine whether release of nuclides from UO_2 is congruent or not and define anticipated, site-specific distributions in containment failure, are needed before these general to conclusions can be further substantiated.

INTRODUCTION

Spent nuclear fuel from civilian reactors is planned to be the dominant waste form for disposal in a U.S. geologic repository. Spent fuel is actually a combination of separate radionuclide sources, including the UO_2 fuel matrix and its encasing cladding. This inhomogeneity and presence of certain key nuclides not found in reprocessed waste forms (e.g., I-129, C-14) has raised doubts regarding the performance of spent fuel as a waste form (1). However, preliminary, site-specific calculations by the Department of Energy's (DOE) Office of Geologic Repository (OGR) Projects (2-4) indicate that radionuclide releases from engineered and natural barrier systems containing spent fuel could comply with requirements of the Nuclear Regulatory Commission (NRC) and the Environmental Protection Agency (EPA). The Performance Assessment Scientific Support (PASS) program was requested by the DOE to make an additional, independent evaluation of the performance of spent fuel as waste form. The results of that study are reported here.

APPROACH

The performance of spent fuel as a waste form is evaluated using the AREST code (see Liebetrau et al. this volume). In the AREST code, the deterministic bases for the calculation of radionuclide releases from individual waste packages are the mass transfer models of Pigford, Chambre' and coworkers (5-8). Solubility-limited (5) and inventory-limited (7) mass transfer models are used within the AREST code for calculating radionuclide release rates.

The inventory-limited, mass transfer model of the AREST code assumes complete dissolution of readily soluble nuclides at the time of containment failure. The solubility-limited, mass transfer model adopts a boundary condition in which the surface concentration at the waste form surface is set equal to the solubility of the waste form matrix.

The solubility-limited, mass transfer model in the AREST code can be subdivided, in turn, into "congruent" and "incongruent" scenarios. Congruent release requires that the mass transfer rates of individual nuclides included in a waste matrix, normalized to their mass inventories, are equal to the normalized mass transfer rate of the matrix component. The congruent release of included nuclides will be proportional to the solubility-limited release rate of the matrix component (e.g., UO_2) times the mass ratio of the included nuclide and the matrix component. This assumes that no subsequent precipitation of a phase containing the matrix component. For the incongruent scenario, the release of individual nuclides are controlled by the solubility of separate solids rather than by the matrix solubility.

Radionuclide releases from separate waste packages are coupled by the AREST code with simulated waste package failure times, either from

corrosion models or user-defined distributions, 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, are translated into a probabilistic assessment of spent fuel performance.

For the purposes of this assessment of spent fuel, the release-rate performance objectives of the DOE's (9) Generic Requirements Document (GRD) have been selected. These objectives are based on the NRC release limits (10) for anticipated events and processes, but are conservatively imposed at the waste package subsystem (WPS) boundary rather than at the boundary of the engineered barrier system (EBS). In addition, compliance with the cumulative release requirement of the EPA is also assessed at the waste package subsystem boundary (11), assuming anticipated events and processes. It is stressed that compliance with EPA release limits at the WPS/EBS boundary is not required. It is believed, however, that this comparison against the EPA release limits can add a meaningful prospective on the performance of spent fuel as a waste form.

INPUTS FOR SPENT FUEL ASSESSMENT

The inputs to the AREST code that are required to compute radionuclide release rates are described elsewhere (see Liebetrau et al. this volume). The four basic categories of input information are:

- repository/waste package data
- containment-failure time distributions
- spent fuel data.
- radionuclide data

Specific input values used in this assessment of spent fuel as a waste form are discussed in the following sections.

Repository Data

The evaluations conducted in this report adopt the waste package designs and repository data presented in the Environmental Assessments (EA's) prepared for basalt, tuff, and salt sites that have been nominated for further site characterization in the U.S. (2-4). This information is summarized in Tables I-III. Data on solubilities and sorption coefficients cited in the EA's have not been critically evaluated. For the tuff site, an unconsolidated region of tuff particles is assumed to fill the air gap surrounding the waste container in order to permit a diffusional pathway between the waste form and the host rock. For the salt site, the failed container is treated as a barrier of porous corrosion products between the waste form and the consolidated salt host rock.

There are several advantages to the selection of the EA's for repository data. First, the EA's present an equivalent amount of data, permitting a similar level of detailed analysis for each site. Second, the data and designs in the EA's date from the same time. This is desirable because design revisions and the collection of new data are ongoing activities at all three sites. The EA's, therefore, provide specific sets of data and designs at a common, fixed time. Third, the EA's contain some of the most current information on spent fuel and its performance as a waste form.

It must be stressed, however, 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 nominated repository sites.

First, the current waste package designs and repository-specific data in the EA's are not the final, complete information that would be presented in a license application. 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 necessarily 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 are deficient, or, perhaps, that less conservative and more realistic assumptions should be used in a revised assessment.

TABLE I

Design Parameters for Basalt Release Calculations (2)

Diffusion coefficient in steam (D_s)	5.0000E-01 cm^2/sec
Diffusion coefficient in water (D_f)	1.0000E-05 cm^2/sec
Outer edge of packing radius (r_1)	7.8653E+01 cm
Packing thickness (b)	1.9300E+01 cm
Spherical waste form radius (r_1-b)	5.9353E+01 cm
Radius of waste (R_0)	7.8653E+01 cm
Packing porosity (ε_1)	0.3
Rock porosity (ε_2)	0.001
Gap width (a)	6.4000E+00 cm
Gap surface area (S_v)	4.4269E+04 cm^2
Gap volume (V_v)	2.8323E+05 cm^3
PWR Assemblies/Waste Package	4
Waste Loading	1.8456E+00 MTU/Waste Package
Bulk density of packing (ρ_1)	1.8000E+00 gm/cm^3
Bulk density of host rock (ρ_2)	3.0000E+00 gm/cm^3
Saturation (ϕ)	100%

Second, 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, to evaluate spent-fuel performance rather than the characteristics of an individual site. Moreover, the mass transfer theory upon which this assessment is based has been judged to be a technically defensible approach (1, 8). A disadvantage of adopting a single approach is that certain assumptions may be incorporated that are not equally conservative for all sites. A single approach may even ignore certain favorable conditions that justify a different, site-specific approach to waste package performance assessment for licensing purposes.

Finally, much of the site characterization work is yet to be 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 compliance with regulatory requirements will require a probabilistic approach (11,12). The limited sets of data and assumed distributions of that data restrict the degree to which reasonable assurance can be provided by any performance assessment at this time.

TABLE II

Design Parameters for Tuff Release Calculations (2)

Diffusion coefficient in steam (D_s)	5.0000E-01 cm^2/sec
Diffusion coefficient in water (D_f)	1.0000E-05 cm^2/sec
Outer edge of packing radius (r_1)	8.8895E+01 cm
Packing thickness (b)	3.0000E+00 cm
Spherical waste form radius (r_1-b)	8.5895E+01 cm
Radius of waste (R_o)	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%

TABLE III

Design Parameters for Salt Release Calculations (2)

Diffusion coefficient in steam (D_s)	5.0000E-01 cm^2/sec
Diffusion coefficient in water (D_f)	1.0000E-05 cm^2/sec
Outer edge of packing radius (r_1)	8.9112E+01 cm
Packing thickness (b)	1.2000E+01 cm
Spherical waste form radius (r_1-b)	7.7112E+01 cm
Radius of waste (R_o)	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%

For the cited reasons, the use of the results presented here should be restricted to the purpose for which this evaluation was conducted; that is, to assess the performance of spent fuel as a waste form in geologic repositories.

Containment-Failure Time Distribution

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. Consequently, the failure-time distributions selected for the spent fuel analysis were not derived from corrosion simulation models within the AREST code. Instead, five hypothetical distributions were chosen to represent a range of containment cases, with a conservative emphasize on failures early in the 10,000 year post-closure period. This approach does not distinguish between containment contributions from the container and cladding. Additional site-specific work is required to better define what failure distributions will occur under expected repository conditions. The selected failure distributions, based on time after repository closure, are as follows:

- A point (degenerate) distribution that places all containment failures at 300 years,
- A point (degenerate) distribution that places all containment failures at 1,000 years,
- A normal (Gaussian) distribution with a mean containment failure at 1,000 years and a standard deviation (s) = 200,
- A normal (Gaussian) distribution with a mean containment failure at 5,000 years and a standard deviation (s) = 2,000,
- An exponential distribution with rate $\lambda = 9.902 \times 10^{-4}$, chosen so that the initial 50 percent of the failures occur between 300 and 1,000 years.

Previous performance assessments of waste package systems have derived failure distributions that are similar to, or are far more favorable than these (2, 13, 14).

Spent Fuel Data

Pressurized water reactor (PWR) fuel has been selected for the assessments in this analysis because quantities of PWR fuel will dominate those of boiling water reactor (BWR) fuel in the repository, and because PWR and BWR spent fuels display similar radionuclide inventories for similar degrees of burnup (15). This choice is also consistent with the requirements of the GRD (9), 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 dimensions of the reference PWR spent fuel rod used in this report are a length of 370.8 cm (146 in.), a diameter of 1.12 cm (0.44 in.), and a volume of 364 cm^3/rod (15). Variables

describing the composition, dimensions, and radionuclide inventory 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 (see Liebetrau et al. this volume). This was done because the heat generation rate determines the time-dependent temperature within the waste package subsystem, and temperature is an important parameter affecting groundwater chemistry, both of which affect the UO_2 matrix solubility.

"Spent fuel" is divided into four separate sources of radionuclides for this study; the UO_2 matrix, the gap/grain boundary, the cladding, and the surface layer on the cladding. This is done because of the different reactivity of these components with water and because of their different radionuclide inventories. Release from the matrix and cladding are calculated using solubility-limited, mass transfer models (5). The release of nuclides from the gap/grain boundary and surface layer are calculated using an inventory-limited, mass transfer model (7).

Radionuclide Data

Screening analyses conducted for all three of the nominated repository sites (2-4) have been used to determine a list of key radionuclides, based on consideration of data on half-lives, NRC and EPA release regulations (10, 11), and conservative estimates of radionuclide solubilities. Based on these site analyses, the list of key radionuclides considered in this assessment of radionuclide release rates for spent fuel is complied in Table IV. Data on allowable release limits based on NRC (10) and EPA (11) regulations are also included in Table IV.

The allowable NRC fractional release rates are calculated from mass transfer rates of nuclides that are normalized relative to their 1,000 year inventories. Inventories for reference PWR fuel at 1,000 years after permanent closure are identified in Table IV for the key radionuclides. These inventories corresponds to spent fuel 1,060 years after reactor discharge, assuming a 60-year pre-closure time period for repository construction and waste emplacement. Inventory variations due to spent fuel age and burnup have not been modeled because these effects are expected to be relatively unimportant. The 1,000 year inventory, and the half-life of a radionuclide can be used to calculate its inventory at other times as needed.

Table V presents the percentages of these nuclides in the different sources within spent fuel. The exact distribution of nuclides, particularly in the gap, will be a function of rod fabrication and power history. Because the majority of nuclear fuel that could be disposed in a geologic repository has not yet been fabricated, a single set of values for radionuclide distribution within spent fuel based on available data (3, 16-18) was assumed for this study.

Note that Cs-137 and Sr-90 isotopes are included in Table IV. These isotopes can be dismissed from consideration, based on NRC's 10 CFR *60.113 cutoff* with respect to the 1,000-year, post-closure curie inventory of spent fuel (10). If release occurs significantly before 1,000 years,

TABLE IV

Nuclide Inventories in Reference PWR Spent Fuel and Allowable Release Limits (2, 3, 10, 11).

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
^{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 1986b.
(b) DOE 1986a, 1986b.
(c) Yuan et al. 1986.
(d) ^{137}Cs and ^{90}Sr fall below NRC's cutoff for release rate calculations, based on 1,000 year inventory. The parenthetical values are extrapolated from NRC regulations (NRC 1983) to provide comparative values in the cases of early (<1,000 years) containment failure.

Table V

Assumed Distribution (by Percentages) of Key Nuclides for Reference PWR Spent Fuel.

Nuclide	UO_2 Matrix	Gap (+ Grain Boundary)	Cladding	Crud
^{238}U	100	--	--	--
^{241}Am	100	--	--	--
^{243}Am	100	--	--	--
^{14}C	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	--	--	--

however, both Cs-137 and Sr-90 represent large fractions of the total curie inventory of spent fuel. Therefore the releases of Sr-90 and Cs-137 are considered in this study for those cases in which containment failure occurs prior to 1,000 years. The allowable fractional release rates for these nuclides are calculated from a linear extrapolation of NRC's 10 CFR 60.113 cutoff based on their 1,000-year inventories. It must be recognized that alternate, more restrictive interpretations regarding the allowable release rates of short-lived nuclides are possible (19) and this remains a key issue for programmatic resolution.

Summary of Modelling Assumptions

In addition to these data and design constraints, other assumptions have been made for this analysis of spent-fuel performance as a waste form in a geologic repository. These assumptions are summarized here.

- Compliance with the NRC performance objective for containment (10) is assumed (i.e., release will not occur before the 300 year period following repository closure).
- The performance of individual waste packages is independent of the performance of adjoining 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 saturation. This assumption may significantly "penalize" repository sites where the anticipated condition is a sustained air gap or void between waste package and host rock.
- Under anticipated conditions, transport within the waste package and in the immediately adjacent host rock is by aqueous-phase diffusion (1, 8, 20).
- The grain boundary of spent fuel is identical in composition to the gap source, and the two sources are treated as the "gap" release. Two percent of the inventory of fission products is assumed to reside in the combined gap and grain boundary source.
- The surface concentrations are set by solubilities that occur immediately upon containment failure; no credit is taken for the time to reach saturation.
- No credit is taken for potential attenuation of radionuclide release that arise from container or cladding failure over a limited surface area.
- No credit is taken for the effect of pathway tortuosity on effective diffusion coefficients.
- The UO_2 matrix dissolves congruently at all repository sites.
- The UO_2 matrix of spent fuel is stable in proximity to metallic container and waste package materials.

These last two assumptions are perhaps the most important. Evidence of congruent release of nuclides contained in the UO_2 matrix of spent fuel under oxidizing and reducing conditions is well documented by U.S., Canadian and Swedish researchers (18, 21-26). The assumption of congruent release has been adopted as an anticipated condition in many assessments of spent fuel performance (7, 8, 13, 26-28). As an upper bounding condition, however, radionuclide release rates are calculated conservatively assuming incongruent (i.e., nuclides controlled by separate solubility-limiting solids) release. Based on current testing and modeling results, however, incongruent release of nuclides from the UO_2 matrix is judged to be an unanticipated condition for all repository sites.

The last assumption is especially important 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 (8, 13, 26-28), if UO_2 is stable. It is possible, however, that UO_2 may be thermodynamically unstable and a new, lower-solubility, uranium-bearing solid may precipitate within the waste package. Auto-oxidation of UO_2 by alpha radiolysis has been suggested as one mechanism leading to UO_2 instability (29). Subsequent laboratory testing of spent fuel in the presence of other waste package materials has indicated that alpha radiolysis-induced UO_2 instability is suppressed or buffered by waste package materials, including the UO_2 itself (21, 25).

Nonetheless, the precipitation of a new uranium-bearing solid within the waste package would lead to an enhanced release rate of uranium and, included nuclides from the waste form, although not necessarily from the waste package. Garisto and Garisto (30) have calculated for a one-dimensional mass transfer model enhancements in release of uranium from UO_2 by up to a factor of 40 under the extreme condition of precipitation close to the UO_2 matrix of a uranium-bearing solid with a much lower solubility. Further analyses and data on the potential effect of precipitation on the assessment of spent fuel are being conducted with the AREST code. It should be noted that an enhancement in matrix release rates by a factor of 40 would not change the calculated results from regulatory compliance to non-compliance.

RESULTS

Representative graphical results of the AREST analysis of spent fuel, based on data from the basalt site (2), are presented in the Figs. 1 to 4. Similar results and graphs have been obtained for the tuff (3) and salt (4) sites. The plotted "fractional release rates" are based on NRC regulations (9) and are obtained by dividing the nuclide mass transfer rate, calculated from mass transfer models (5-8), by the mass inventory of that nuclide within a waste package 1,000 years after repository closure.

Figure 1 presents the UO_2 matrix release of selected nuclides for the case of instantaneous failure of all waste packages 300 years after repository closure (Case 1). Note that the fractional release rates of nuclides for the congruent release scenario are significantly lower (up to 10 orders of magnitude) than those for the incongruent release scenario. This decrease is attributable to both the low solubility of the UO_2

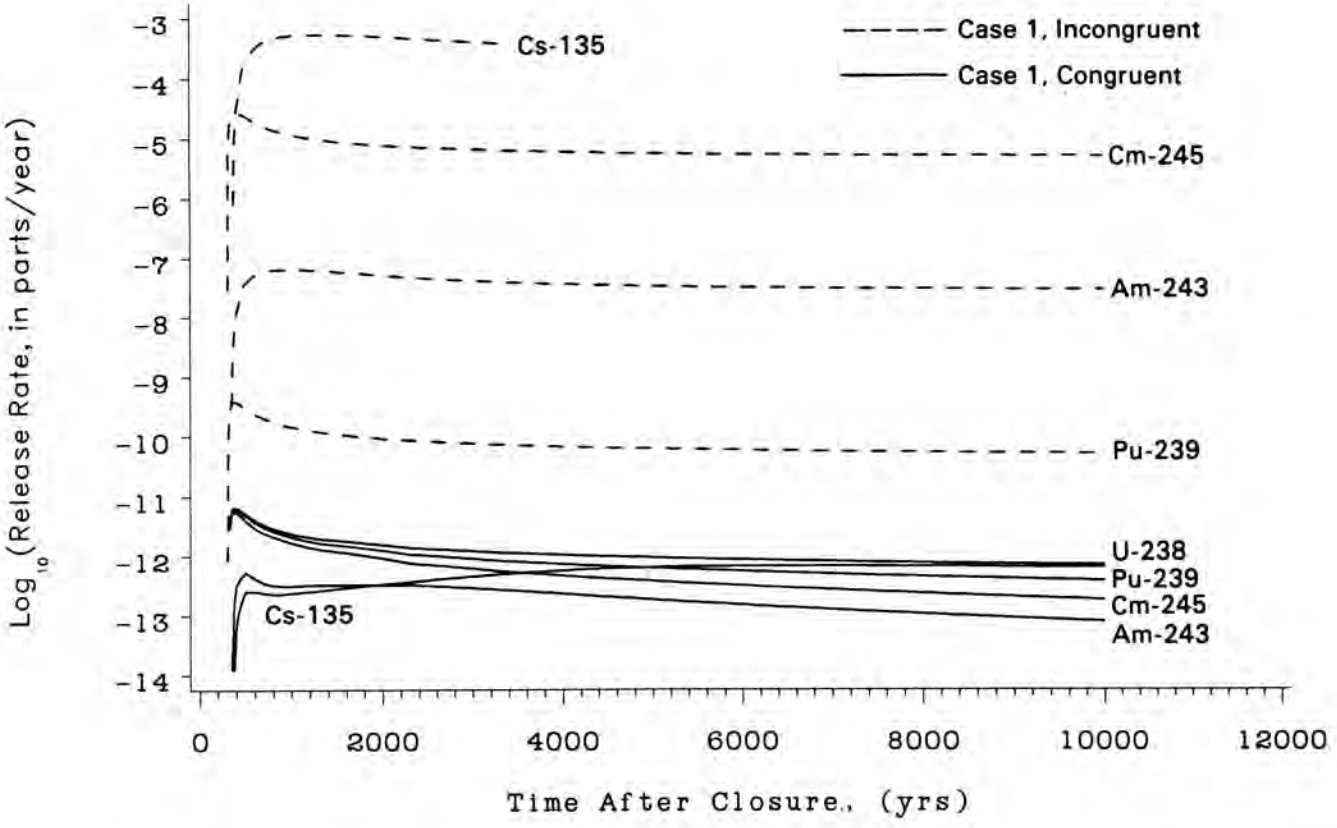

Fig. 1. Incongruent and Congruent Release Rates of Selected Nuclides from UO_2 Matrix for Basalt, Case 1.

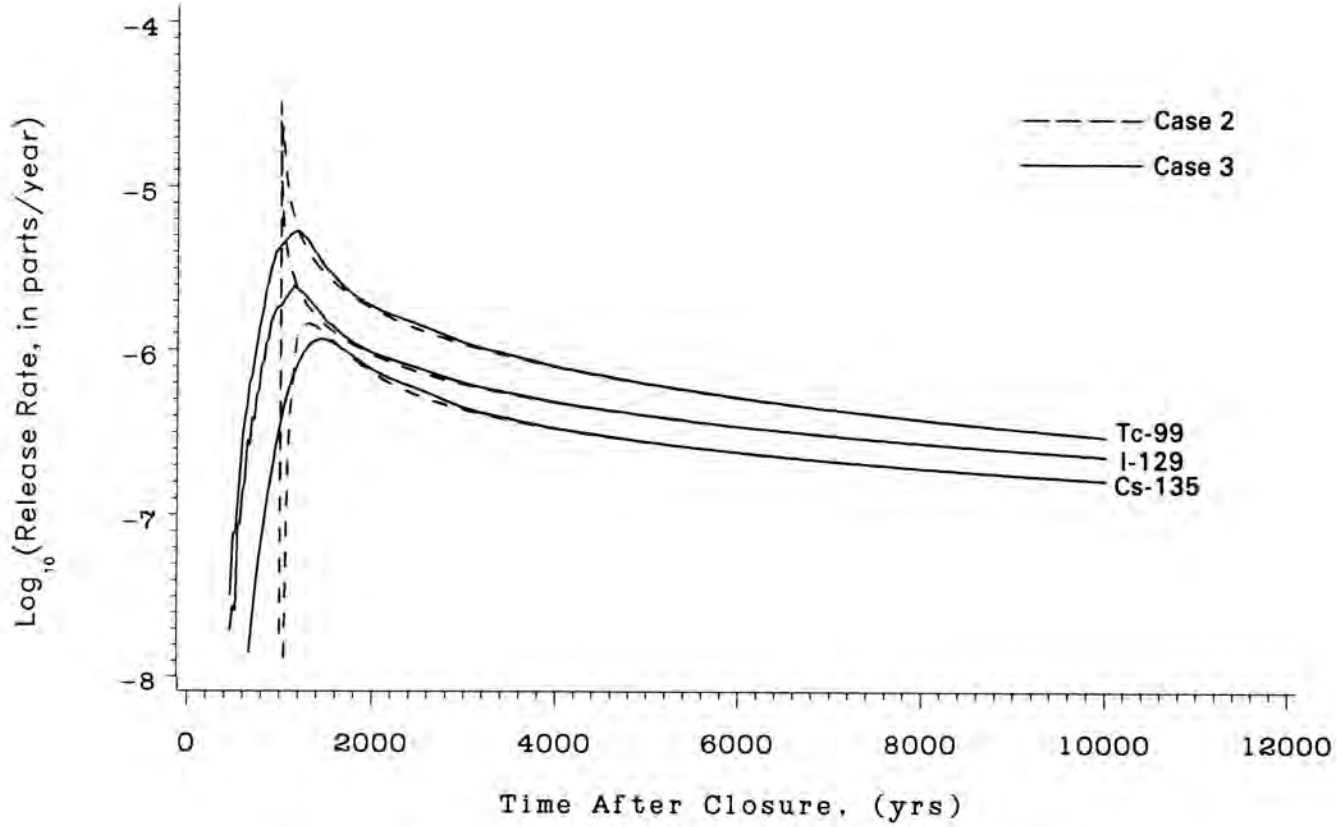

Fig. 2. Release Rates of Selected Gap Nuclides for Basalt, Cases 1 and 3.

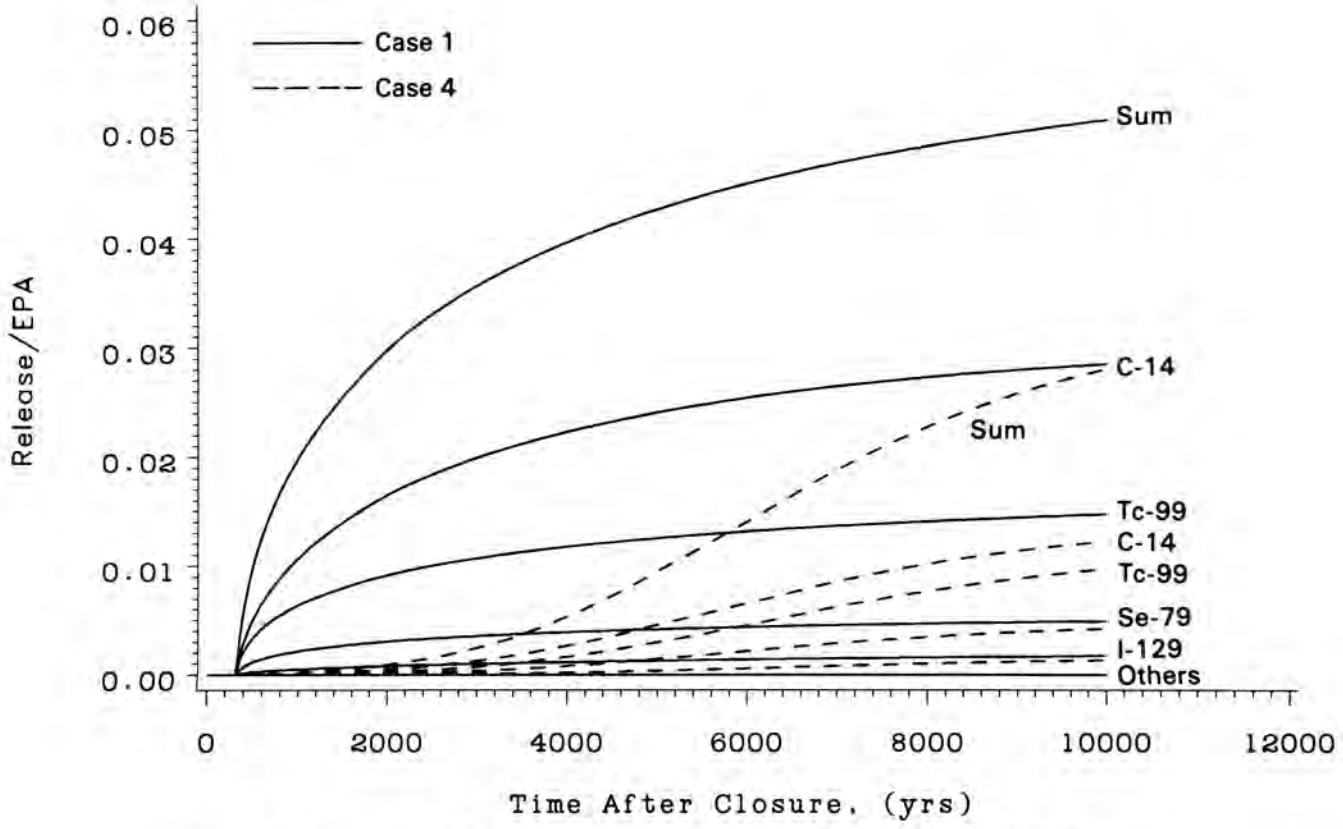

Fig. 3. Cumulative Release Rates, Normalized to EPA Limits, of Nuclides from All Sources for Basalt, Cases 1 and 4.

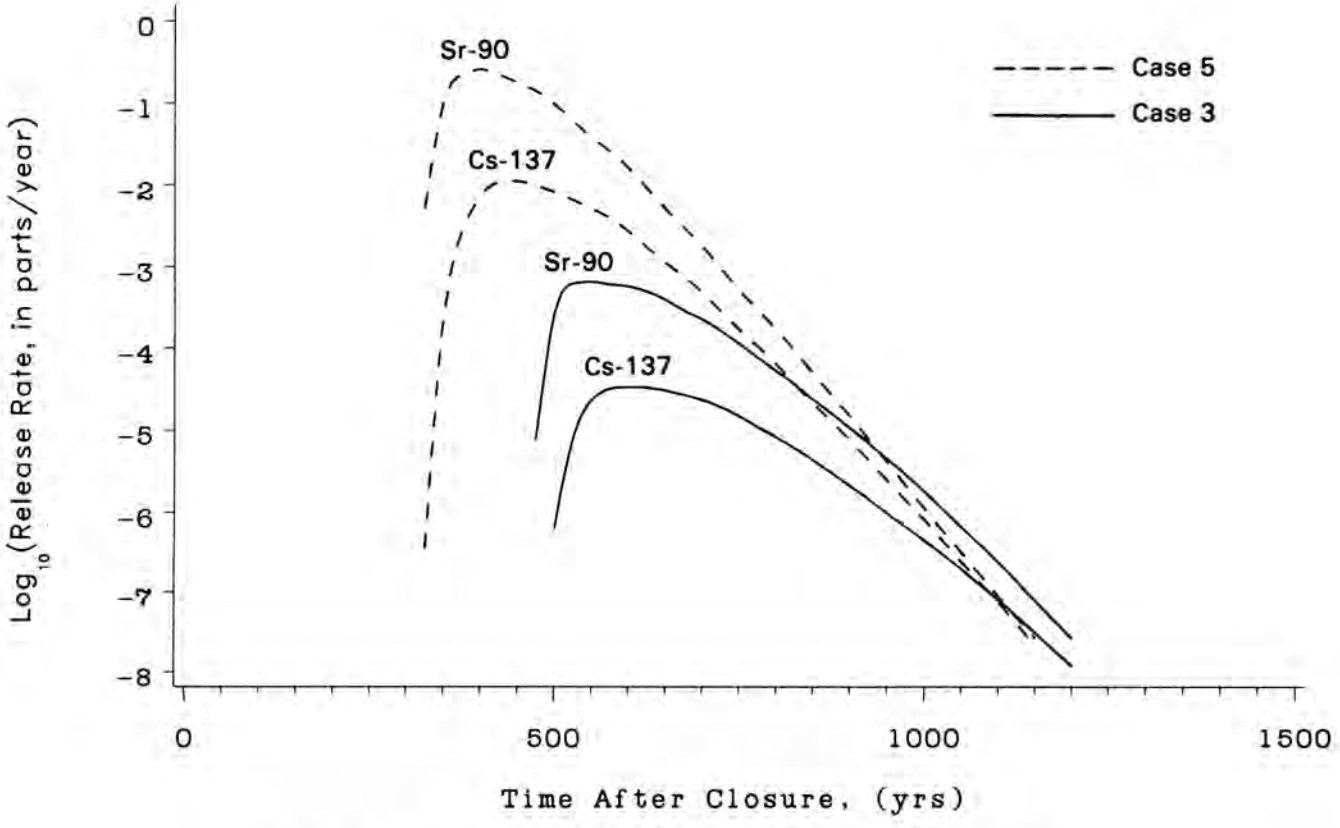

Fig. 4. Release Rates of Sr-90 and Cs-137 for Basalt, Cases 3 and 5.

matrix and the low mass ratio of those nuclides contained in the UO_2 matrix.

Figure 2 compares the release of inventory-limited gap nuclides for the cases of instantaneous containment failure of all waste packages 1,000 years after repository closure (Case 2) and a normal distribution of containment failures centered at 1,000 years, with a standard deviation of 200 years (Case 3). The spike-like maxima in release for the instantaneous failure case are greatly attenuated by the assumption that failures are distributed over time.

Figure 3 shows the cumulative releases of nuclides, normalized to their EPA limits (11), from all sources within spent fuel, assuming congruent matrix release. The releases shown are for the cases of total containment failure at 300 years after repository closure (Case 1) and normally distributed containment failures centered at 5000 years (Case 4), respectively. The difference in compliance with EPA limits between the most unfavorable and most favorable cases considered in this study is surprisingly slight. For example, the release of C-14 at the boundary of the waste package reaches approximately 2.5% of its cumulative release limit to the biosphere after 10,000 years for the instantaneous failure case. For the normally distributed failure case, C-14 release reaches a maximum of about 1% of its cumulative release limit after the same 10,000 years.

Finally, Fig. 4 presents the predicted gap release of the short-lived nuclides Sr-90 and Cs-137 for normally distributed containment failures centered at 1,000 years (Case 3) and exponentially distributed failures (Case 5) in which one-half of the failures occur between 300 and 1,000 years after repository closure (see earlier section on containment failure). Releases of Sr-90 and Cs-137 are higher in the latter case because of the early, higher rate of failure. Note that although these fractional releases are high, they are below the extrapolated NRC limits for these nuclides used in this study (Table IV). Alternate interpretations (14) of allowable releases for such short-lived nuclides could, however, show that these nuclides were above intended NRC limits.

SUMMARY

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. Releases of individual radionuclides have been evaluated relative to compliance with allowable NRC fractional release rates and EPA 10,000-year cumulative release limits (10,11).

Results of the separate summaries of spent fuel performance for specific repository sites have been compiled. Based on these results, 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., control by the solubility of the UO_2 matrix). For the congruent matrix release scenario (the anticipated condition based on current data) rates are lower, in some cases by factors of 10 orders of magnitude, than release rates calculated for the incongruent matrix release scenario. This result applies to all the repository sites and waste package designs that have been evaluated. It should also be noted, however, that incongruent matrix dissolution apparently leads to the release of several nuclides, notably Cs-135 and I-129, above their NRC limits.

- For congruent matrix release, the cumulative release of all nuclides from spent fuel complied with their EPA limits at all three repository sites for all scenarios of containment failure that were considered. This includes the case of instantaneous failure of all waste packages 300 years after repository closure.

- For congruent matrix release, the fractional release rates of all nuclides from spent fuel are below their NRC limits for cases based on the distributed containment failures considered. This includes failure distributions in which 50 percent of the waste packages have lost containment 1,000 years after repository closure.

- Based on these previous two points, the EPA cumulative release limits for individual nuclides are generally less restrictive than the NRC fractional release rates 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, gap and cladding surface layer. For the congruent matrix release scenarios, the gap and cladding surface layer contributed 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 when instantaneous containment failure is assumed. For gap and cladding surface-layer nuclides, distributed container failures can significantly reduce the maximum release rates. For matrix nuclides, the effect of distributed container failures is relatively less important than whether matrix release is congruent or incongruent.

- Short-lived nuclides that are readily soluble, such as Cs-137 and Sr-90, can be dismissed from release rate calculations on the basis of the NRC inventory cutoff. These nuclides can, however, dominate the release from spent fuel if a significant fraction of all waste packages lose containment prior to 1,000 years after repository closure.

- Additional data on site characterization, waste package design, and spent fuel characterization are needed to substantiate these general conclusions.

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