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AN ASSESSMENT OF THE IMPORTANT RADIONUCLIDES IN NUCLEAR WASTE

by

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ABSTRACT

The relative importance of the various radionuclides contained in nuclear waste has been assessed by consideration of (1) the quantity of each radionuclide present, (2) the Environmental Protection Agency's release limits for radionuclides, (3) how retardation processes such as solubility and sorption affect radionuclide transport, and (4) the physical and chemical forms of radionuclides in the waste. Three types of waste were reviewed: spent fuel, high-level waste, and defense high-level waste. Conditions specific to the Nevada Nuclear Waste Storage Investigations project potential site at Yucca Mountain were used to describe radionuclide transport. The actinides Am, Pu, Np, and U were identified as the waste elements for which solubility and sorption data were most urgently needed. Other important waste elements were identified as Sr, Cs, C, Ni, Zr, Tc, Th, Ra, and Sn. Under some conditions, radionuclides of three elements (C, Tc, and I) may have high solubility and negligible sorption. The potential for transport of some waste elements (C and I) in the gas phase must also be evaluated for the Yucca Mountain Site.

I. INTRODUCTION

A program to build and license a nuclear waste repository must consider the type of waste to be stored and, in particular, the radionuclide composition of the waste over time after storage begins. This information is needed to assess how well the proposed repository will contain the waste over a long time. Presently, there are several different types of waste that might be stored at potential repository sites being studied by the Office of Civilian Radioactive Waste Management of the Department of Energy. They include spent fuel from boiling water reactors (BWR) or pressurized water reactors (PWR), high-level waste obtained from reprocessing BWR or PWR spent fuel, defense high-level waste, and transuranic (TRU) waste. The different physical forms and radionuclide compositions of these wastes create different problems in site characterization and performance assessment.

The Nevada Nuclear Waste Storage Investigations project is studying a site at Yucca Mountain in southern Nevada as a potential nuclear waste repository. As part of the geochemical site characterization of Yucca Mountain being done at Los Alamos National Laboratory, questions concerning the relative importance of various radionuclides have arisen in the context of studies of how solubility and sorption on local minerals affect radionuclide transport. In assessing the suitability of the site, those radionuclides that are present in large quantities, that must be contained well, or that present special problems should be given attention early in the program.

This report presents an assessment of the relative importance of radionuclides in various types of nuclear waste. Four factors are considered in this assessment:

1. the quantity of various radionuclides present in the waste,
2. the Environmental Protection Agency (EPA) release limits for the radionuclides,
3. how different retardation processes such as solubility and sorption might affect radionuclide transport, and
4. the physical and chemical forms of the radionuclides in the waste.

Because of the uncertainties about waste types, presenting a single list of radionuclides ordered by relative importance is impossible. However, this report highlights radionuclides that may be important regardless of the waste form and others that may be important under certain conditions.

II. REPOSITORY INVENTORY

Before an assessment can be made of the importance of the various radionuclides in nuclear waste, it is necessary to know the radionuclide composition of the waste. For waste from civilian reactors, data from the compilation of Croff and Alexander[1] were used. The two common types of light-water reactors

(PWR and BWR) produce waste (spent fuel and high-level waste) that is sufficiently similar to consider only PWR data in this analysis; conclusions drawn for PWR waste generally apply to BWR waste also. The PWR-waste data are for fuel that was irradiated at 37.5 MW/MTHM to a burnup of 33 000 MWd/MTHM. The unit MTHM is metric ton of heavy metal (uranium plus plutonium) originally charged to the reactor. The radioactivity content of the waste is reported in units of Ci/1000 MTHM. The spent-fuel waste is composed of fuel, cladding, and structural material from the fuel assembly.[1] High-level waste comes from reprocessed fuel from spent-fuel waste. During reprocessing, 99.5% of the uranium and plutonium are assumed to be removed from the waste. Other fission and activation products may also be isolated at this time for separate storage. For the analysis done here, it was assumed that a repository storing high-level waste would also store fission products segregated during reprocessing along with cladding and structural wastes. Although many of these wastes, cladding and structural wastes in particular, are usually not considered to be part of high-level waste, they represent highly radioactive material generated in the fuel cycle that must be isolated from the environment.

Data for defense high-level waste were obtained from calculations by J. R. Fowler and M. D. Boersma of the Waste Solidification Technology Division of Savannah River Laboratory, Aiken, South Carolina. The results[2] give the radionuclide inventory of Savannah River high-level waste from the future Defense Waste Processing Facility. This facility is designed to process alkaline waste sludge that has been stored for 5 years after discharge from a reactor, mixed with cesium concentrate from soluble alkaline waste that has been stored for 15 years, into a borosilicate glass. The radionuclide inventories are reported in terms of the activity or mass of radionuclides per canister of glass (1480 kg). A conversion between the quantity of waste in a canister and the quantity of heavy metal originally charged to the reactor is not available. For the analysis done here, it was assumed that the equivalent loading of defense high-level waste canisters in MTHM/canister could be estimated from the total activity (Ci/canister) of PWR high-level waste and defense high-level waste canisters. PWR high-level waste canisters will contain waste from about 2 MTHM[3] with about 70 000 Ci/canister 100 years after discharge.[1] Defense high-level waste canisters will contain about 16 000 Ci/canister 100 years after discharge. Based on an equivalence of the curie content, the loading of defense high-level waste canisters would be about

0.5 MTHM/canister. This assumption does not affect the relative importance of radionuclides within a given waste type; it does, however, make comparisons between PWR waste and defense waste somewhat uncertain.

TRU wastes, which contain alpha-emitting transuranic radionuclides at levels generally lower than high-level waste, may also be stored in a geologic repository, but they were not specifically considered in this analysis. Presently, the amount of TRU waste that might be stored in a repository and the radionuclide inventory of that waste are uncertain. The radionuclides in TRU waste are generally also present in high-level waste; thus, no new important radionuclides should be added from TRU waste. One important aspect of TRU waste that may influence radionuclide transport is the presence of other materials such as organics.[4] The effect of these other materials on waste-element speciation and solubility must be investigated if a decision is made to store TRU waste in a geologic repository.

Table I is a list of the total inventory in Ci/1000 MTHM for the various types of waste considered here. The inventory is divided into activation products, fission products, and actinides for civilian waste.[1] Throughout an important period (1000 to 10000 years) for the effects of geochemical processes on radionuclide transport, the inventory from PWR spent fuel is about an order of magnitude larger than from PWR high-level waste. Thus, spent fuel presents a more difficult containment problem.

More significant for identifying important radionuclides is the identity of the radionuclides that contribute most to the inventory over time. Tables II, III, and IV list the primary radionuclides and the amounts they contribute to the total inventory of the waste for PWR spent fuel, PWR high-level waste, and defense high-level waste, respectively. The actinides and their decay products are represented by isotopes of Np, U, Pu, Am, Cm, Th, Ra, and Pa. The radionuclides ^{137}Cs, ^{90}Sr, ^{99}Tc, ^{126}Sn, ^{135}Cs, ^{151}Sm, ^{79}Se, and their short-lived daughters are fission products. The radionuclide ^{93}Zr is both a fission product and PWR cladding activation product. Other cladding activation products are ^{63}Ni and ^{59}Ni. The radionuclide ^{14}C comes primarily from activation of ^{14}N, which is present in both PWR fuel and cladding. The ^{14}C content of defense high-level waste is insignificant.[2] The fission product ^{129}I is generally considered important, but it does not appear in Tables II, III, and IV because it does not contribute significantly to the total inventory of the waste; ^{129}I will be discussed

TABLE I

REPOSITORY INVENTORY FOR VARIOUS TYPES OF NUCLEAR WASTE

	Ci/1000 MTHM for Various Decay Times			
	10^2 year	10^3 year	10^4 year	10^5 year
PWR Spent Fuel				
Activation products	3.2×10^5	8.4×10^3	6.4×10^3	2.5×10^3
Actinides	6.8×10^6	1.7×10^6	4.4×10^5	3.9×10^4
Fission products	3.4×10^7	1.9×10^4	1.9×10^4	1.4×10^4
Total	4.1×10^7	1.8×10^6	4.7×10^5	5.6×10^4
PWR High-Level Waste				
Activation products	3.2×10^5	8.4×10^3	6.4×10^3	2.5×10^3
Actinides	3.2×10^5	8.3×10^4	2.0×10^4	2.5×10^3
Fission products	3.4×10^7	1.9×10^4	1.9×10^4	1.4×10^4
Total	3.4×10^7	1.1×10^5	4.5×10^4	1.9×10^4
Defense High-Level Waste				
Total	3.2×10^7	6.0×10^4	3.1×10^4	1.7×10^4

in Sec. VII because of its high solubility and poor sorption behavior. Some radionuclides listed in Tables II, III, and IV have relatively short half-lives and would not be important in a study of transport mechanisms. At 100 years after discharge, ^{137}Cs, ^{90}Sr, and their short-lived daughters dominate the inventory for all types of waste. By 1000 years after discharge, the actinides are the predominant radionuclides. At 10 000 and 100 000 years, activation products, fission products, and actinides are all important, although the actinides are more important for spent fuel than for high-level waste.

III. EPA LIMITS

The total activity of the various radionuclides present is only part of the measure of their importance. In response to the Nuclear Waste Policy Act, the EPA is developing a standard for nuclear waste repositories.[5] In its

TABLE II

PRIMARY RADIONUCLIDES CONTRIBUTING TO
REPOSITORY INVENTORY FOR PWR SPENT FUEL

Radionuclide and Per cent of Total Activity for Various Decay Times							
10^2 year		10^3 year		10^4 year		10^5 year	
^{137}Cs	25%	^{241}Am	51%	^{239}Pu	51%	^{239}Pu	33%
^{137m}Ba	24%[a]	^{240}Pu	27%	^{240}Pu	39%	^{99}Tc	17%
^{90}Sr	17%	^{239}Pu	17%	^{99}Tc	3%	^{59}Ni	4%
^{90}Y	17%[b]	^{243}Am	0.9%	^{243}Am	1%	^{93}Zr	3%
^{241}Am	9%	^{239}Np	0.9%[c]	^{239}Np	1%[c]	^{93m}Nb	3%[d]
^{238}Pu	3%	^{99}Tc	0.7%	^{59}Ni	1%	^{234}U	3%
^{241}Pu	2%	^{59}Ni	0.3%	^{234}U	0.4%	^{242}Pu	3%
^{240}Pu	1%	^{234}U	0.1%	^{93}Zr	0.4%	^{237}Np	2%
^{239}Pu	0.8%	^{93}Zr	0.1%	^{93m}Nb	0.4%[d]	^{233}Pa	2%[e]
^{63}Ni	0.8%	^{93m}Nb	0.1%[d]	^{242}Pu	0.4%	^{226}Ra	2%[f]
^{151}Sm	0.4%	^{242}Pu	0.1%	^{237}Np	0.3%	^{230}Th	2%

[a] Short-lived daughter of ^{137}Cs.
[b] Short-lived daughter of ^{90}Sr.
[c] Short-lived daughter of ^{243}Am.
[d] Short-lived daughter of ^{93}Zr.
[e] Short-lived daughter of ^{237}Np.
[f] Decay products of ^{226}Ra are in secular equilibrium; each decay product also represents 2% of inventory. The ^{226}Ra decay products are generally short lived.

present form, this standard limits the release of radionuclides to the environment for 10 000 years after disposal. Cumulative release limits for radionuclides are prescribed in the standard. Table V lists these release limits as presently set. They cover all radioactive isotopes. All alpha-emitting radionuclides except ^{230}Th and ^{232}Th have a release limit of 100 Ci/1000 MTHM. Most radionuclides that do not emit alpha particles have a release limit of 1000 Ci/1000 MTHM; the exceptions are ^{14}C and ^{129}I, which have lower release limits (100 Ci/1000 MTHM) because of their biological activity, and ^{99}Tc, which has a higher release limit (10 000 Ci/MTHM).

TABLE III

PRIMARY RADIONUCLIDES CONTRIBUTING TO
REPOSITORY INVENTORY FOR PWR HIGH-LEVEL WASTE

Radionuclide and Per cent of Total Activity for Various Decay Times							
10^2 year		10^3 year		10^4 year		10^5 year	
^{137}Cs	30%	^{241}Am	39%	^{99}Tc	28%	^{99}Tc	50%
^{137m}Ba	28%[a]	^{243}Am	14%	^{243}Am	15%	^{59}Ni	12%
^{90}Sr	20%	^{239}Np	14%[c]	^{239}Np	15%[c]	^{93}Zr	10%
^{90}Y	20%[b]	^{99}Tc	12%	^{59}Ni	10%	^{93m}Nb	9%[d]
^{63}Ni	0.9%	^{240}Pu	6%	^{239}Pu	9%	^{239}Pu	3%
^{241}Am	0.5%	^{59}Ni	5%	^{240}Pu	5%	^{126}Sn	2%
^{151}Sm	0.5%	^{239}Pu	2%	^{93}Zr	4%	^{237}Np	2%
^{238}Pu	0.2%	^{93}Zr	2%	^{93m}Nb	4%[d]	^{233}Pa	2%[e]
^{244}Cm	0.1%	^{93m}Nb	2%[d]	^{94}Nb	2%	^{135}Cs	2%
^{243}Am	0.05%	^{14}C	1%	^{126}Sn	2%	^{79}Se	0.7%
^{239}Np	0.05%	^{94}Nb	1%	^{14}C	1%	^{233}U	0.7%

[a]Short-lived daughter of ^{137}Cs.
[b]Short-lived daughter of ^{90}Sr.
[c]Short-lived daughter of ^{243}Am.
[d]Short-lived daughter of ^{93}Zr.
[e]Short-lived daughter of ^{237}Np.

The EPA standard limits the total release of radioactivity to the environment. The contribution of each radionuclide to the total release is calculated from the ratio of the amount released to the EPA limit for that radionuclide. Based on this formulation, a measure of the relative importance of the various radionuclides is the ratio of the radionuclide inventory in Ci/1000 MTHM to the EPA limit. Tables VI, VII, and VIII list radionuclides ordered by this ratio along with values of the ratio for PWR spent fuel, PWR high-level waste, and defense high-level waste, respectively. Radionuclides near the top of the lists have larger values for the inventory/EPA limit ratio. Thus, larger fractions of these radionuclides must be contained by the repository to meet the EPA standard.

TABLE IV

PRIMARY RADIONUCLIDES CONTRIBUTING TO REPOSITORY INVENTORY FOR DEFENSE HIGH-LEVEL WASTE

Radionuclide and Per cent of Total Activity for Various Decay Times							
10^2 year		10^3 year		10^4 year		10^5 year	
^{90}Sr	25%	^{241}Am	31%	^{239}Pu	43%	^{99}Tc	14%
^{90}Y	25%[a]	^{239}Pu	28%	^{240}Pu	13%	^{93}Zr	11%
^{137}Cs	24%	^{240}Pu	16%	^{99}Tc	11%	^{93m}Nb	11%[c]
^{137m}Ba	22%[b]	^{59}Ni	6%	^{59}Ni	11%	^{59}Ni	8%
^{238}Pu	3%	^{99}Tc	6%	^{93}Zr	7%	^{234}U	6%
^{63}Ni	0.7%	^{93}Zr	3%	^{93m}Nb	7%[c]	^{239}Pu	6%
^{151}Sm	0.7%	^{93m}Nb	3%[c]	^{234}U	5%	^{230}Th	4%
^{241}Am	0.2%	^{234}U	2%	^{126}Sn	0.8%	^{226}Ra	4%[d]
^{241}Pu	0.07%	^{238}Pu	1%	^{79}Se	0.5%	^{135}Cs	1%
^{239}Pu	0.05%	^{63}Ni	0.6%	^{135}Cs	0.5%	^{126}Sn	1%
^{240}Pu	0.03%	^{126}Sn	0.4%	^{230}Th	0.4%	^{79}Se	0.4%

[a] Short-lived daughter of ^{90}Sr.

[b] Short-lived daughter of ^{137}Cs.

[c] Short-lived daughter of ^{93}Zr.

[d] Decay products of ^{226}Ra are in secular equilibrium; each decay product also represents 4% of inventory. The ^{226}Ra decay products are generally short lived.

Compared with an ordering of radionuclides based on their inventory (see Tables II, III, and IV), the orderings shown in Tables VI, VII, and VIII increase the importance of actinides because they are usually alpha emitters and thus have lower EPA limits. In particular, ^{99}Tc, which has the highest release limit of any radionuclide, is reduced from being the major contributor to the inventory of PWR high-level waste at 10 000 and 100 000 years (see Table III) to a level below many other radionuclides when ordered on the inventory/EPA limit ratio (see Table VII).

TABLE V

EPA RELEASE LIMITS

Radionuclide	Cumulative Release Limit (Ci/1000 MTHM)
^{241}Am or ^{243}Am	100
^{14}C	100
^{137}Cs or ^{135}Cs	1 000
^{129}I	100
^{237}Np	100
^{238}Pu, ^{239}Pu, ^{240}Pu, or ^{242}Pu	100
^{226}Ra	100
^{90}Sr	1 000
^{99}Tc	10 000
^{230}Th or ^{232}Th	10
^{126}Sn	1 000
^{233}U, ^{234}U, ^{235}U, ^{236}U, or ^{238}U	100
Any other alpha-emitting radionuclide with a half-life greater than 20 years	100
Any other radionuclide with a half-life greater than 20 years that does not emit alpha particles	1 000

IV. EFFECTS OF SOLUBILITY

The previous discussions have not considered any specific repository site; the orderings listed in Tables VI to VIII would be the same for any site storing the type of waste considered. However, solubility, which depends on water chemistry, has site-dependent effects. A simple model has been developed to assess the effects of solubility on element dissolution from solid waste at the proposed Yucca Mountain repository.[6] Results from this model can be used to rank radionuclides in terms of the ratio of the radionuclide dissolution rate (Ci/1000 MTHM year) to the EPA limit (Ci/1000 MTHM). This ratio represents the fraction of the EPA limit of a given radionuclide that is released from the solid waste each year. The ratio is large for radionuclides with large dissolution rates (radionuclides that are not limited by solubility) and with low EPA release limits.

TABLE VI

RADIONUCLIDES ORDERED BY RATIO OF
INVENTORY TO EPA LIMIT FOR
PWR SPENT FUEL

Radionuclide and (Inventory/EPA Limit) for Various Decay Times							
10^2 year		10^3 year		10^4 year		10^5 year	
^{241}Am	3.8×10^4	^{241}Am	9.0×10^3	^{239}Pu	2.4×10^3	^{239}Pu	1.8×10^2
^{238}Pu	1.1×10^4	^{240}Pu	4.8×10^3	^{240}Pu	1.8×10^3	^{230}Th	1.0×10^2
^{137}Cs	1.0×10^4	^{239}Pu	3.1×10^3	^{243}Am	6.7×10^1	^{234}U	1.6×10^1
^{137m}Ba	9.8×10^{3}[a]	^{243}Am	1.6×10^2	^{234}U	1.9×10^1	^{242}Pu	1.5×10^1
^{90}Sr	6.8×10^3	^{234}U	2.0×10^1	^{242}Pu	1.7×10^1	^{237}Np	1.1×10^1
^{90}Y	6.8×10^{3}[b]	^{242}Pu	1.8×10^1	^{230}Th	1.7×10^1	^{226}Ra	1.0×10^{1}[d]
^{240}Pu	5.3×10^3	^{239}Np	1.6×10^{1}[c]	^{237}Np	1.2×10^1	^{233}U	4.1
^{239}Pu	3.1×10^3	^{14}C	1.4×10^1	^{239}Np	6.7[c]	^{236}U	4.0
^{241}Pu	1.0×10^3	^{237}Np	1.0×10^1	^{59}Ni	4.8	^{229}Th	3.7
^{244}Cm	3.3×10^2	^{238}Pu	9.7	^{14}C	4.6	^{238}U	3.2
^{63}Ni	3.1×10^2	^{59}Ni	5.2	^{236}U	3.5	^{59}Ni	2.2
^{243}Am	1.7×10^2	^{238}U	3.2	^{238}U	3.2	^{93}Zr	1.9

[a]Short-lived daughter of ^{137}Cs.

[b]Short-lived daughter of ^{90}Sr.

[c]Short-lived daughter of ^{243}Am.

[d]Decay products of ^{226}Ra are in secular equilibrium. Each alpha emitter in the decay chain has the same inventory/EPA limit ratio; others have an order of magnitude lower ratio. The ^{226}Ra decay products are generally short lived.

The model used in this analysis is based on the assumption that dissolution rates are limited by diffusion of waste elements into water moving past the solid waste or by bulk-waste dissolution, whichever rate is lower.[6] Waste elements with large solubilities or that are present in very small quantities will dissolve at the bulk-waste dissolution rate; elements with low solubilities will dissolve more slowly. The bulk-waste fractional dissolution rate used in these calculations was 1×10^{-4}/yr except for Cs, Sr, C, and I in spent fuel, where 1×10^{-3}/yr was used to account for migration of these

TABLE VII

RADIONUCLIDES ORDERED BY RATIO OF INVENTORY TO EPA LIMIT FOR PWR HIGH-LEVEL WASTE

Radionuclide and (Inventory/EPA Limit) for Various Decay Times

10^2 year		10^3 year		10^4 year		10^5 year	
^{137}Cs	1.0×10^4	^{241}Am	1.8×10^3	^{243}Am	1.6×10^2	^{239}Pu	5.1
^{137m}Ba	9.8×10^{3}[a]	^{243}Am	1.7×10^2	^{239}Pu	4.0×10^1	^{237}Np	3.4
^{90}Sr	6.8×10^3	^{240}Pu	6.3×10^1	^{240}Pu	2.4×10^1	^{230}Th	2.4
^{90}Y	6.8×10^{3}[b]	^{239}Pu	2.1×10^1	^{239}Np	6.7[c]	^{59}Ni	2.2
^{241}Am	2.0×10^3	^{239}Np	1.6×10^{1}[c]	^{59}Ni	4.8	^{93}Zr	1.7
^{238}Pu	5.2×10^2	^{14}C	1.4×10^1	^{14}C	4.6	^{93m}Nb	1.6[d]
^{244}Cm	3.3×10^2	^{59}Ni	5.1	^{237}Np	3.5	^{233}U	1.2
^{63}Ni	3.1×10^2	^{237}Np	3.5	^{93}Zr	1.8	^{229}Th	1.1[f]
^{243}Am	1.7×10^2	^{93}Zr	1.8	^{93m}Nb	1.7[d]	^{99}Tc	9.4×10^{-1}
^{151}Sm	1.7×10^2	^{93m}Nb	1.7[d]	^{99}Tc	1.3	^{126}Sn	3.9×10^{-1}
^{240}Pu	6.8×10^1	^{99}Tc	1.3	^{94}Nb	9.1×10^{-1}	^{234}U	3.6×10^{-1}
^{242}Cm	2.0×10^1	^{94}Nb	1.2	^{126}Sn	7.3×10^{-1}	^{233}Pa	3.4×10^{-1}[e]

[a] Short-lived daughter of ^{137}Cs.
[b] Short-lived daughter of ^{90}Sr.
[c] Short-lived daughter of ^{243}Am.
[d] Short-lived daughter of ^{93}Zr.
[e] Short-lived daughter of ^{237}Np.
[f] Decay products of ^{229}Th are in secular equilibrium. Each alpha emitter in the decay chain has the same inventory/EPA limit ratio; others have an order of magnitude lower ratio. The ^{229}Th decay products are generally short lived.

TABLE VIII

RADIONUCLIDES ORDERED BY RATIO OF INVENTORY TO EPA LIMIT FOR DEFENSE HIGH-LEVEL WASTE

Radionuclide and (Inventory/EPA Limit) for Various Decay Times							
10^2 year		10^3 year		10^4 year		10^5 year	
^{238}Pu	8.7×10^{3}	^{241}Am	1.9×10^{2}	^{239}Pu	1.3×10^{2}	^{230}Th	7.5×10^{1}
^{90}Sr	7.8×10^{3}	^{239}Pu	1.7×10^{2}	^{240}Pu	3.8×10^{1}	^{234}U	1.1×10^{1}
^{90}Y	7.8×10^{3}[a]	^{240}Pu	9.9×10^{1}	^{234}U	1.4×10^{1}	^{239}Pu	9.7
^{137}Cs	7.4×10^{3}	^{234}U	1.5×10^{1}	^{230}Th	1.3×10^{1}	^{226}Ra	7.4[d]
^{137m}Ba	7.0×10^{3}[b]	^{238}Pu	7.1	^{59}Ni	3.3	^{93}Zr	1.9
^{241}Am	7.8×10^{2}	^{59}Ni	3.5	^{93}Zr	2.0	^{93m}Nb	1.9[c]
^{63}Ni	4.3×10^{2}	^{93}Zr	2.0	^{93m}Nb	2.0[c]	^{59}Ni	1.4
^{151}Sm	2.1×10^{2}	^{93m}Nb	2.0[c]	^{226}Ra	9.7×10^{-1}[d]	^{236}U	6.3×10^{-1}
^{239}Pu	1.7×10^{2}	^{230}Th	1.2	^{236}U	6.2×10^{-1}	^{237}Np	2.6×10^{-1}
^{240}Pu	1.1×10^{2}	^{236}U	6.0×10^{-1}	^{99}Tc	3.3×10^{-1}	^{99}Tc	2.4×10^{-1}
^{241}Pu	2.4×10^{1}	^{99}Tc	3.4×10^{-1}	^{237}Np	2.7×10^{-1}	^{238}U	1.5×10^{-1}

[a] Short-lived daughter of ^{90}Sr.
[b] Short-lived daughter of ^{137}Cs.
[c] Short-lived daughter of ^{93}Zr.
[d] Decay products of ^{226}Ra are in secular equilibrium. Each alpha emitter in the decay chain has the same inventory/EPA limit ratio; others have an order of magnitude lower ratio. The ^{226}Ra decay products are generally short lived.

radionuclides to regions of greater access during irradiation. This may be higher than dissolution rates that are ultimately achieved. If lower bulk-waste dissolution rates can be achieved for long times, dissolution rates would be lower than calculated here and solubility would be less important. The model does not account for effects of engineered barriers; in this respect, the model is also very conservative. Estimates of waste-element solubilities and a number of parameters that characterize the solid waste and water flow near the repository are required for the calculation. Table IX lists the solubilities assumed for 16 waste elements in water from Yucca Mountain. These elements were chosen for further analysis because their isotopes are prominent in Tables VI to VIII. Some of the solubilities were calculated,[7] some measured,[8] and others were estimated. The solubilities listed as large are assumed to be large enough that bulk-waste dissolution and not solubility would limit dissolution rates under any conditions.[6] The solubilities of carbon and iodine are listed as large for PWR spent fuel but are lower for PWR high-level waste. This variable solubility reflects an uncertainty about the physical form and release mechanisms of these elements from spent fuel compared with their separation and storage as calcium or barium carbonate and barium iodate for PWR high-level waste.[4] The other parameters characterizing dissolution are the same as described in Ref. 6 for PWR waste, except that the latest estimate of the maximum recharge rate at Yucca Mountain, 1 mm/year, was used.[9] Solid waste sizes for defense high-level waste were taken from Ref. 2.

Tables X, XI, and XII list radionuclides ordered by the ratio of dissolution rate to EPA limit and values of this ratio for PWR spent fuel, PWR high-level waste, and defense high-level waste, respectively. Several short-lived decay products of radionuclides listed in these tables have not been included because they would not exist long enough to provide solubility controls on dissolution (see table footnotes). If the activity of these radionuclides were counted with their parent radionuclides, the dissolution rate/EPA limit ratio of the parents would increase. How much increase would depend on the number of decay products and their EPA limits. As noted above, radionuclides near the top of the list are being released from the solid waste relative to their EPA limits at a faster rate than radionuclides farther down the list. Solubility is not an effective retardation mechanism for elements near the top of the list. In particular, radionuclides of neptunium, carbon, technetium, cesium, strontium, radium, and nickel have generally moved up in Tables X to XII relative to their positions in the tables ordered by inventory/EPA limit ratios (Tables VI to VIII). Radionuclides of thorium, tin, and zirconium have moved down in Tables X to XII; these elements have solubilities low enough to significantly limit their dissolution rates.

TABLE IX

WASTE ELEMENT SOLUBILITIES IN WATER FROM YUCCA MOUNTAIN

Element	Solubility (moles/ℓ) PWR Spent Fuel	PWR High-Level Waste	Defense High-Level Waste
Np	1×10^{-3}	1×10^{-3}	1×10^{-3}
U	4×10^{-3}	4×10^{-3}	4×10^{-3}
Pu	1×10^{-5}	1×10^{-5}	1×10^{-5}
Am	1×10^{-6}	1×10^{-6}	1×10^{-6}
Cm	1×10^{-6}	1×10^{-6}	1×10^{-6}
Th	1×10^{-9}	1×10^{-9}	1×10^{-9}
Ra	3×10^{-7}	3×10^{-7}	3×10^{-7}
Cs	large	large	large
Sr	8×10^{-4}	8×10^{-4}	8×10^{-4}
Tc	large	large	large
C	large	4×10^{-4}	not present
I	large	2×10^{-3}	large
Sn	1×10^{-9}	1×10^{-9}	1×10^{-9}
Ni	1×10^{-2}	1×10^{-2}	1×10^{-2}
Zr	1×10^{-10}	1×10^{-10}	1×10^{-10}
Sm	2×10^{-9}	2×10^{-9}	2×10^{-9}

In addition to the standards imposed by the EPA, the Nuclear Regulatory Commission (NRC) has developed technical criteria for geologic repositories.[10] One criterion limits the release rate of radionuclides from the engineered barrier system to one part in 10^5 per year of the inventory of that radionuclide present at 1000 years following permanent closure of the repository. This release limit does not apply to radionuclides released at less than one part in 10^8 per year of the total inventory at 1000 years (about 1.7×10^{-5} Ci/MTHM year). If dissolution from the solid waste as calculated by the dissolution model described above is assumed to be the only mechanism limiting the release rate of radionuclides from the engineered barrier, the dissolution rates can be used to determine which radionuclides do not meet

TABLE X

RADIONUCLIDES ORDERED BY RATIO OF DISSOLUTION RATE TO EPA LIMIT FOR PWR SPENT FUEL[a]

Radionuclide and Dissolution Rate/EPA Limit $(yr)^{-1}$ for Various Decay Times

10^2 year		10^3 year		10^4 year		10^5 year	
^{137}Cs	1.0×10^{1}	^{14}C	1.4×10^{-2}	^{14}C	4.6×10^{-3}	^{237}Np	1.1×10^{-3}
^{90}Sr	8.7×10^{-1}	^{241}Am	4.5×10^{-3}	^{237}Np	1.2×10^{-3}	^{226}Ra	4.7×10^{-4}
^{244}Cm	3.3×10^{-2}	^{240}Pu	1.1×10^{-3}	^{239}Pu	7.9×10^{-4}	^{239}Pu	4.5×10^{-4}
^{14}C	1.5×10^{-2}	^{237}Np	1.0×10^{-3}	^{240}Pu	6.1×10^{-4}	^{135}Cs	3.4×10^{-4}
^{63}Ni	1.0×10^{-2}	^{239}Pu	6.9×10^{-4}	^{135}Cs	3.4×10^{-4}	^{129}I	3.1×10^{-4}
^{241}Am	5.4×10^{-3}	^{135}Cs	3.5×10^{-4}	^{243}Am	3.4×10^{-4}	^{99}Tc	9.4×10^{-5}
^{238}Pu	2.3×10^{-3}	^{129}I	3.1×10^{-4}	^{129}I	3.2×10^{-4}	^{59}Ni	7.3×10^{-5}
^{242}Cm	2.0×10^{-3}	^{59}Ni	1.7×10^{-4}	^{59}Ni	1.6×10^{-4}	^{242}Pu	3.7×10^{-5}
^{243}Cm	1.3×10^{-3}	^{99}Tc	1.3×10^{-4}	^{226}Ra	1.3×10^{-4}	^{234}U	1.1×10^{-5}
^{240}Pu	1.1×10^{-3}	^{243}Am	7.8×10^{-5}	^{99}Tc	1.3×10^{-4}	^{233}U	2.9×10^{-6}
^{239}Pu	6.7×10^{-4}	^{242}Cm	3.2×10^{-5}	^{234}U	1.4×10^{-5}	^{236}U	2.8×10^{-6}

[a]Short-lived daughters of ^{137}Cs (^{137m}Ba), ^{90}Sr (^{90}Y), ^{243}Am (^{239}Np), and short-lived decay products of ^{226}Ra have not been included.

this NRC technical criterion. Table XIII lists radionuclides that do not meet the NRC release criterion for the three types of waste at 1000, 10 000, and 100 000 years after discharge from the reactor based on this calculation. The radionuclides on this list are generally the same as those that head the lists in Tables X, XI, and XII; that is, they are isotopes of elements whose release is not being limited by solubility. This is a very conservative calculation because it ignores the features of the engineered barrier system except waste-element solubility and solid-waste dissolution. It does highlight those radionuclides that may require low solid-waste dissolution rates or an engineered barrier system to meet this NRC criterion.

TABLE XI

RADIONUCLIDES ORDERED BY RATIO OF DISSOLUTION RATE TO EPA LIMIT FOR PWR HIGH-LEVEL WASTE[a]

Radionuclide and Dissolution Rate/EPA Limit $(yr)^{-1}$ for Various Decay Times							
10^2 year		10^3 year		10^4 year		10^5 year	
^{137}Cs	1.0	^{240}Pu	1.3×10^{-3}	^{239}Pu	6.8×10^{-4}	^{239}Pu	5.1×10^{-4}
^{90}Sr	6.7×10^{-1}	^{241}Am	6.2×10^{-4}	^{240}Pu	4.1×10^{-4}	^{237}Np	3.4×10^{-4}
^{244}Cm	3.3×10^{-2}	^{239}Pu	4.3×10^{-4}	^{237}Np	3.5×10^{-4}	^{233}U	1.2×10^{-4}
^{238}Pu	1.1×10^{-2}	^{237}Np	3.5×10^{-4}	^{243}Am	2.6×10^{-4}	^{99}Tc	9.4×10^{-5}
^{63}Ni	8.1×10^{-3}	^{243}Am	2.3×10^{-4}	^{99}Tc	1.3×10^{-4}	^{59}Ni	5.7×10^{-5}
^{242}Cm	2.0×10^{-3}	^{14}C	2.1×10^{-4}	^{59}Ni	1.2×10^{-4}	^{234}U	3.6×10^{-5}
^{241}Am	1.7×10^{-3}	^{59}Ni	1.3×10^{-4}	^{14}C	7.3×10^{-5}	^{135}Cs	3.3×10^{-5}
^{240}Pu	1.4×10^{-3}	^{99}Tc	1.3×10^{-4}	^{234}U	4.6×10^{-5}	^{129}I	3.1×10^{-5}
^{243}Cm	1.3×10^{-3}	^{234}U	4.7×10^{-5}	^{135}Cs	3.4×10^{-5}	^{226}Ra	2.4×10^{-5}
^{239}Pu	3.5×10^{-4}	^{135}Cs	3.5×10^{-5}	^{129}I	3.1×10^{-5}	^{242}Pu	7.9×10^{-6}
^{237}Np	3.2×10^{-4}	^{242}Cm	3.2×10^{-5}	^{233}U	1.5×10^{-5}	^{236}U	3.2×10^{-6}

[a]Short-lived daughters of ^{137}Cs (^{137m}Ba), ^{90}Sr (^{90}Y), ^{243}Am (^{239}Np), and short-lived decay products of ^{226}Ra have not been included.

V. EFFECTS OF SORPTION

Section IV has outlined how solubility can influence dissolution rates of radionuclides from solid waste. These dissolution rates provide a source term for transport of radionuclides in water passing through the repository and moving toward the environment. Sorption will affect the rate at which radionuclides move with the water. Radionuclides that are strongly sorbed will move more slowly than the average water velocity; radionuclides that are not sorbed may move at about the same velocity as the water. During transport, radionuclides will decay. If the time required to transport a radionuclide from the repository to the environment is much longer than the half-life of the radionuclide, release to the environment will be low. Based on this simplified discussion, a measure of the effect of sorption would be the fraction of a

TABLE XII

RADIONUCLIDES ORDERED BY RATIO OF DISSOLUTION RATE TO EPA LIMIT FOR DEFENSE HIGH-LEVEL WASTE[a]

Radionuclide and Dissolution Rate/EPA Limit $(yr)^{-1}$ for Various Decay Times

10^2 year		10^3 year		10^4 year		10^5 year	
^{90}Sr	7.6×10^{-1}	^{241}Am	1.9×10^{-2}	^{239}Pu	5.7×10^{-3}	^{239}Pu	9.7×10^{-4}
^{137}Cs	7.4×10^{-1}	^{239}Pu	5.4×10^{-3}	^{240}Pu	1.7×10^{-3}	^{234}U	9.5×10^{-4}
^{238}Pu	2.3×10^{-1}	^{240}Pu	3.2×10^{-3}	^{234}U	1.2×10^{-3}	^{226}Ra	7.4×10^{-4}
^{241}Am	3.5×10^{-2}	^{234}U	1.2×10^{-3}	^{59}Ni	3.3×10^{-4}	^{59}Ni	1.4×10^{-4}
^{63}Ni	2.1×10^{-2}	^{59}Ni	3.5×10^{-4}	^{226}Ra	9.7×10^{-5}	^{236}U	5.3×10^{-5}
^{239}Pu	4.6×10^{-3}	^{238}Pu	2.3×10^{-4}	^{236}U	5.2×10^{-5}	^{237}Np	2.6×10^{-5}
^{240}Pu	2.9×10^{-3}	^{236}U	5.1×10^{-5}	^{99}Tc	3.3×10^{-5}	^{99}Tc	2.4×10^{-5}
^{234}U	9.9×10^{-4}	^{99}Tc	3.4×10^{-5}	^{237}Np	2.7×10^{-5}	^{135}Cs	1.5×10^{-5}
^{241}Pu	6.4×10^{-4}	^{63}Ni	3.2×10^{-5}	^{135}Cs	1.5×10^{-5}	^{238}U	1.3×10^{-5}
^{59}Ni	3.6×10^{-4}	^{237}Np	2.7×10^{-5}	^{238}U	1.3×10^{-5}	^{242}Pu	1.3×10^{-5}
^{236}U	5.0×10^{-5}	^{135}Cs	1.5×10^{-5}	^{242}Pu	6.6×10^{-6}	^{233}U	7.9×10^{-6}

[a]Short-lived daughters of ^{137}Cs (^{137m}Ba), ^{90}Sr (^{90}Y), ^{243}Am (^{239}Np), and short-lived decay products of ^{226}Ra have not been included.

radionuclide originally released to the water that is finally released to the environment.

Transport and sorption are very complex processes that depend on details of the hydrology, water chemistry, and mineralogy along transport paths. For this analysis, which is aimed only at assessing the relative importance of various radionuclides for site characterization, a much simpler model was used. It was assumed that water transport from the repository to the environment could be characterized by a travel time (t_w) and that each element could be assigned a retardation factor (R_f) that defines the ratio of water velocity to waste-element transport velocity.[11] From these two parameters, the element travel time from the repository to the environment is ($R_f t_w$). The fraction of that radionuclide left from simple decay (with half-life t_h)

TABLE XIII

RADIONUCLIDES NOT MEETING THE
NRC TECHNICAL CRITERION ON
RELEASE IF ONLY DISSOLUTION LIMITS RELEASE[a]

	Radionuclide Identity at Various Decay Times		
	10^3 year	10^4 year	10^5 year
Spent fuel	^{14}C	^{14}C	^{237}Np
	^{237}Np	^{237}Np	^{59}Ni
	^{59}Ni	^{59}Ni	^{99}Tc
	^{99}Tc	^{99}Tc	^{135}Cs
	^{135}Cs	^{135}Cs	^{226}Ra
	^{129}I	^{129}I	^{129}I
High-level waste	^{14}C	^{237}Np	^{237}Np
	^{237}Np	^{59}Ni	^{59}Ni
	^{59}Ni	^{99}Tc	^{99}Tc
	^{99}Tc	^{135}Cs	^{135}Cs
	^{135}Cs	^{240}Pu	^{239}Pu
	^{240}Pu	^{239}Pu	
	^{239}Pu		
Defense high-level waste	^{59}Ni	^{59}Ni	^{59}Ni
	^{63}Ni	^{99}Tc	^{99}Tc
	^{99}Tc	^{234}U	^{234}U
	^{234}U	^{239}Pu	^{226}Ra
	^{241}Am	^{240}Pu	
	^{239}Pu		
	^{240}Pu		
	^{238}Pu		

[a]Dissolution rates greater than one part in 10^5 per year of the 1000-year inventory of that radionuclide and dissolution rates greater than 1.7×10^{-5} Ci/MTHM year.

after a time (t) is $\exp(-0.693t/t_h)$ (Ref. 12). Thus, for the time required to transport the radionuclide from the repository to the environment, the fraction remaining would be

$$f = \exp(-0.693R_f t_w/t_h) \quad . \qquad (1)$$

The effect of sorption can be combined with the effect of solubility by defining the quantity

$$R_e = (\text{dissolution rate/EPA limit})\exp(-0.693R_f t_w/t_h) \quad , \qquad (2)$$

where the (dissolution rate/EPA limit) ratios have been listed previously in Tables X to XII. The quantity R_e is a measure of the release rate of a radionuclide to the environment relative to its EPA release limit, where the geochemical processes of solubility and sorption are the only retardation mechanisms considered. This release would start at a time ($R_f t_w$) after waste dissolution starts.

Equation (1), which assesses the effects of sorption on radionuclide transport, is only an approximation. Its main deficiency is that it is based on simple radioactive decay. This assumption is generally adequate for activation products, fission products, and many actinides. However, several actinides that are part of long-lived decay chains can be produced along the transport path as precursors decay. A primary example is ^{226}Ra, a decay product of ^{230}Th. These calculations also ignore the effects of dispersion during transport. Although such a simple model is inadequate for actual transport calculations, it is acceptable for the assessment being done here.

Retardation factors for most of the waste elements examined here were chosen from a compilation of sorption data measured on tuffs from Yucca Mountain.[11] Table XIV lists the values used. The measurements have been done on a variety of tuffs and retardation factors can vary significantly with the type of mineral present in the tuff.[11] The retardation factors used in this analysis are generally near the average or median of the measurements. Variations of an order of magnitude or more are possible. Data for carbon (as carbonate), nickel, zirconium, and samarium were estimated. Water travel times from the repository to the environment have not yet been determined for Yucca

TABLE XIV

WASTE-ELEMENT RETARDATION FACTORS

Element	Retardation Factor[a]
Np	50
U	25
Pu	500
Am	1000
Cm	100
Th	500
Ra	1000
Cs	1000
Sr	700
Tc	1
C	1
I	1
Sn	1000
Ni	20
Zr	100
Sm	1000

[a]Retardation factor is the ratio of water velocity to waste-element velocity; see Ref. 11.

Mountain. Preliminary estimates that are consistent with 1 mm/year recharge are in the range of 20 000 years or greater.[9] Water travel times that are greater than 1000 years are acceptable under the NRC's technical criteria.[10] Radionuclide travel time and R_e were calculated for water travel times of 1000 and 20 000 years to assess the effect of this parameter on the identity of the radionuclides released to the environment in significant quantities relative to their EPA release limits.

Tables XV, XVI, and XVII list radionuclides ordered by their travel time and initial value of R_e as defined in Eq. (2) for PWR spent fuel, PWR high-level waste, and defense high-level waste, respectively. Radionuclides that are near the top of the lists have shorter travel times because they are not sorbed well, and solubility is not limiting their concentrations. Because of the exponential term in Eq. (2), radionuclides with short half-lives compared with the water travel time decay before release no matter how well they are sorbed. Radionuclides with longer half-lives decay if their half-lives are short compared with the product $R_f t_w$; thus, their decay depends on retardation by sorption. Other waste elements have long-lived isotopes that are

TABLE XV

RADIONUCLIDES ORDERED BY TRAVEL TIME AND INITIAL RELEASE RATE (R_e) FOR PWR SPENT FUEL

Water Travel Time = 1000 year			Water Travel Time = 20 000 year		
Radionuclide	Radionuclide Travel Time (year)	Initial R_e $(yr)^{-1}$	Radionuclide	Radionuclide Travel Time (year)	Initial R_e $(yr)^{-1}$
^{14}C	1×10^3	1.4×10^{-2}	^{14}C	2×10^4	1.4×10^{-3}
^{129}I	1×10^3	3.2×10^{-4}	^{129}I	2×10^4	3.2×10^{-4}
^{99}Tc	1×10^3	1.3×10^{-4}	^{99}Tc	2×10^4	1.2×10^{-4}
^{59}Ni	2×10^4	1.4×10^{-4}	^{59}Ni	4×10^5	4.3×10^{-6}
^{234}U	2.5×10^4	1.0×10^{-5}	^{234}U	5×10^5	2.7×10^{-6}
^{238}U	2.5×10^4	2.2×10^{-6}	^{238}U	5×10^5	2.2×10^{-6}
^{236}U	2.5×10^4	1.8×10^{-6}	^{236}U	5×10^5	1.8×10^{-6}
^{235}U	2.5×10^4	1.2×10^{-7}	^{235}U	5×10^5	1.2×10^{-7}
^{233}U	2.5×10^4	1.1×10^{-9}	^{233}U	5×10^5	1.3×10^{-10}
^{237}Np	5×10^4	4.1×10^{-4}	^{237}Np	1×10^6	3.0×10^{-4}
^{93}Zr	1×10^5	5.2×10^{-14}	^{93}Zr	2×10^6	2.2×10^{-14}
^{242}Pu	5×10^5	1.5×10^{-6}	^{242}Pu	1×10^7	3.7×10^{-14}
^{230}Th	5×10^5	3.1×10^{-9}	^{135}Cs	2×10^7	3.1×10^{-6}
^{239}Pu	5×10^5	3.8×10^{-10}			
^{135}Cs	1×10^6	2.7×10^{-4}			
^{126}Sn	1×10^6	7.1×10^{-13}			

not strongly sorbed but do have low solubilities; zirconium is one example. The same radionuclides head the lists for all three types of fuel; an exception exists for defense high-level waste where ^{14}C and ^{129}I are not present in significant amounts. The same radionuclides are also present for water travel times of 1000 and 20 000 years; with the longer water travel time, the radionuclide travel time is longer and the values of R_e are smaller.

VI. EFFECTS OF PHYSICAL AND CHEMICAL FORMS OF THE WASTE

As noted earlier, considerable uncertainty still exists about the type of waste to be stored in the proposed repositories. For many waste elements, the

TABLE XVI

RADIONUCLIDES ORDERED BY TRAVEL TIME AND INITIAL RELEASE RATE (R_e) FOR PWR HIGH-LEVEL WASTE

Water Travel Time = 1000 year			Water Travel Time = 20 000 year		
Radionuclide	Radionuclide Travel Time (year)	Initial R_e $(yr)^{-1}$	Radionuclide	Radionuclide Travel Time (year)	Initial R_e $(yr)^{-1}$
^{14}C	1×10^3	2.1×10^{-4}	^{99}Tc	2×10^4	1.2×10^{-4}
^{99}Tc	1×10^3	1.3×10^{-4}	^{129}I	2×10^4	3.2×10^{-5}
^{129}I	1×10^3	3.2×10^{-5}	^{14}C	2×10^4	2.1×10^{-5}
^{59}Ni	2×10^4	1.1×10^{-4}	^{59}Ni	4×10^5	3.3×10^{-6}
^{234}U	2.5×10^4	2.6×10^{-5}	^{234}U	5×10^5	6.7×10^{-6}
^{238}U	2.5×10^4	1.6×10^{-6}	^{238}U	5×10^5	1.6×10^{-6}
^{236}U	2.5×10^4	1.3×10^{-6}	^{236}U	5×10^5	1.3×10^{-6}
^{233}U	2.5×10^4	1.2×10^{-7}	^{235}U	5×10^5	8.6×10^{-8}
^{235}U	2.5×10^4	8.6×10^{-8}	^{233}U	5×10^5	1.6×10^{-8}
^{237}Np	5×10^4	3.1×10^{-4}	^{237}Np	1×10^6	2.3×10^{-4}
^{93}Zr	1×10^5	4.0×10^{-14}	^{93}Zr	2×10^6	1.7×10^{-14}
^{242}Pu	5×10^5	7.6×10^{-7}	^{242}Pu	1×10^7	1.9×10^{-14}
^{230}Th	5×10^5	2.5×10^{-9}	^{135}Cs	2×10^7	3.1×10^{-7}
^{239}Pu	5×10^5	2.0×10^{-10}			
^{135}Cs	1×10^6	2.7×10^{-5}			
^{126}Sn	1×10^6	5.5×10^{-13}			

actual waste form will not strongly influence element dissolution rates or how strongly elements are sorbed along flow paths. However, the physical and chemical form of the waste can control release and transport of some radionuclides. This is particularly true of the repository at Yucca Mountain, which is currently proposed for the unsaturated zone, where vapor as well as aqueous transport are possible. Both ^{14}C (as carbon dioxide) and ^{129}I could be released from spent fuel as gases. The analysis presented here assumed aqueous transport. Thus, for spent fuel, a separate transport path may be important. If both of these radionuclides are separated during reprocessing and stored as

TABLE XVII

RADIONUCLIDES ORDERED BY TRAVEL TIME AND INITIAL RELEASE RATE (R_e) FOR DEFENSE HIGH-LEVEL WASTE

Water Travel Time = 1000 year			Water Travel Time = 20 000 year		
Radionuclide	Radionuclide Travel Time (year)	Initial R_e $(yr)^{-1}$	Radionuclide	Radionuclide Travel Time (year)	Initial R_e $(yr)^{-1}$
^{99}Tc	1×10^3	3.4×10^{-5}	^{99}Tc	2×10^4	3.2×10^{-5}
^{59}Ni	2×10^4	3.0×10^{-4}	^{59}Ni	4×10^5	8.8×10^{-6}
^{234}U	2.5×10^4	9.2×10^{-4}	^{234}U	5×10^5	2.4×10^{-4}
^{236}U	2.5×10^4	5.0×10^{-5}	^{236}U	5×10^5	5.0×10^{-5}
^{238}U	2.5×10^4	1.3×10^{-5}	^{238}U	5×10^5	1.3×10^{-5}
^{235}U	2.5×10^4	2.3×10^{-6}	^{235}U	5×10^5	2.3×10^{-6}
^{233}U	2.5×10^4	2.9×10^{-8}	^{233}U	5×10^5	3.3×10^{-9}
^{237}Np	5×10^4	1.7×10^{-5}	^{237}Np	1×10^6	1.3×10^{-5}
^{93}Zr	1×10^5	2.3×10^{-11}	^{93}Zr	2×10^6	9.7×10^{-12}
^{245}Cm	1×10^5	6.9×10^{-12}	^{247}Cm	2×10^6	1.1×10^{-15}
^{246}Cm	1×10^5	3.2×10^{-15}	^{248}Cm	2×10^6	6.4×10^{-17}
^{247}Cm	1×10^5	1.2×10^{-15}	^{242}Pu	1×10^7	4.0×10^{-14}
^{248}Cm	$1 \times 1C^5$	1.0×10^{-15}	^{135}Cs	2×10^7	1.4×10^{-7}
^{147}Sm	2×10^5	1.1×10^{-13}	^{147}Sm	2×10^7	1.1×10^{-13}
^{242}Pu	5×10^5	1.6×10^{-6}			
^{230}Th	5×10^5	2.1×10^{-8}			
^{239}Pu	5×10^5	2.6×10^{-9}			
^{135}Cs	1×10^6	1.2×10^{-5}			
^{126}Sn	1×10^6	4.8×10^{-12}			

solids, gases probably will not be released. However, ^{14}C as aqueous carbonate could exchange with gaseous carbon dioxide in the unsaturated zone. The entire question of gaseous transport and its relation to aqueous-phase transport remains to be investigated.

A long-lived isotope of nickel, ^{59}Ni, is prominent in the tables presented here. For PWR waste, this isotope comes from subassembly structural material in the form of stainless steel. The solubility of nickel was based on NiO and $NiCO_3$, which give similar solubilities in water from Yucca Mountain. This solubility is relatively high (see Table IX). If the stainless steel passivates during storage, release rates of nickel from PWR waste may be much lower than calculated here based on nickel solubility. For defense high-level waste, nickel is in the borosilicate glass with the other waste elements. The release rates of nickel based on solubility are probably appropriate in this case.

VII. IMPORTANT RADIONUCLIDES

Which radionuclides are most important for a repository at Yucca Mountain? Different answers arise from consideration of different time scales, transport mechanisms, regulations, or waste types. Three aspects of this question are addressed in this report.

1. Which radionuclides are present in waste in large quantities relative to their EPA release limts? These are the radionuclides for which the retardation processes of solubility and sorption must be most effective. Such radionuclides are important because they must be addressed during performance assessment, and thus, must have the solubility and sorption data base for that analysis.
2. Which radionuclides are not retarded by solubility and sorption and are present in large enough quantities to pose release problems? This question can be addressed by performance assessment calculations using estimated solubilities and sorption coefficients. A simplified analysis in this report determined how solubility limits dissolution rates and how sorption limits transport. The results of this analysis highlight radionuclides that may require special consideration because they are not retarded strongly. These radionuclides are important because special design considerations or performance assessments may be required to assure their containment.
3. Which radionuclides might require special consideration because of the physical or chemical form of the waste? These are radionuclides that may exhibit unique source-term or transport behavior. They are also important because they may require special data or assessment techniques for performance-assessment calculations. As solid-waste forms become better defined, other radionuclides may be added to this category.

The radionuclides present in large quantities relative to their EPA release limits are a function of waste type and storage time. Early in the storage period (0 to 500 years), two elements with short-lived isotopes (Sr and Cs) are important (see Tables VI, VII, and VIII). However, the waste package must contain wastes for at least 300 years[10] so that under expected conditions, these short-lived radionuclides would decay before significant release occurred. Solubility and sorption behavior of these elements for assessing the short-lived isotope transport would be important if the waste-package failed early. Solubility and sorption will be important retardation mechanisms over 1000-10 000 years; the same radionuclides are present in large quantities relative to their EPA release limits over that time for the three types of waste considered here (see Tables VI, VII, and VIII). Isotopes of Am, Pu, Th, Np, and U are the most important during that period. Isotopes of C, Ni, Zr, Tc, Ra, and Sn are present in smaller, but still significant, amounts over the same period. Although current estimates of solubilities for many of these elements may be low (see Table IX) or estimates of retardation factors may be high (see Table XIV), data used for transport calculations will have to be supported by an extensive data base.

How solubility influences radionuclide dissolution rates is summarized in Tables X, XI, and XII; how sorption influences radionuclide release rates and travel times is summarized in Tables XV, XVI, and XVII. These analyses were based on estimates of waste element solubilities and sorption coefficients. Three elements with large solubilities and small sorption coefficients that have significant quantities of some isotopes in most types of waste are C, Tc, and I. Other elements with moderately high solubilities and intermediate sorption coefficients are Ni, Np, and U. These elements have isotopes with the shortest travel times and highest release rates in Tables XV, XVI, and XVII. If water travel times from the repository to the environment are as large as expected (20 000 year or more), the proposed Yucca Mountain site should have little problem meeting the EPA release standard.[5] However, water travel times shorter than 10 000 year could result in release of some of the radionuclides with high solubilities and little or no sorption during the period covered by the EPA standard. Some consideration should be given to better containment of these radionuclides.

All considerations of important radionuclides so far have assumed the normal mechanisms of waste dissolution and transport in water passing through the repository. However, some radionuclides may not move in that manner because of the physical or chemical form of the waste. Radionuclides that can be released as gases may have a special transport path through the unsaturated zone at Yucca Mountain. In particular, ^{14}C and ^{129}I release from spent fuel should be considered. These radionuclides will be separated from high-level waste and should not pose the same problem in that case. Also, isotopic exchange between carbonate in the aqueous phase and carbon dioxide in the vapor phase should be examined for its significance. Release of Ni isotopes from PWR wastes may be much less than estimated here based on solubility considerations if the Ni is stored as stainless steel; however, Ni in defense high-level waste would not be constrained by stainless-steel dissolution.

VIII. CONCLUSIONS

Providing a single list of important radionuclides and the work required to assess their release and transport is a difficult task. Uncertainties as to types of waste, transport pathways, and data needed for transport calculations create several problems of similar importance. Based on the assessment done here, the following recommendations are made for solubility and sorption data needs.

1. Provide a solubility and sorption data base for the elements Am, Pu, Th, Np, and U.
2. Provide a solubility and sorption data base for the elements Sr, Cs, C, Ni, Zr, Tc, Ra, and Sn.
3. Study how elements with high solubility and low sorption (C, Tc, and I) could be better contained at Yucca Mountain. Recommend experimental work, design changes, or calculations to assure their containment.
4. Study the potential for gaseous release and transport of radionuclides from various types of wastes, including isotopic exchange between the aqueous and vapor phases. Recommend experimental work, design changes, or calculations to assure containment of gaseous radionuclides if transport is significant in this form.

Many of the data needs mentioned in Items 1. and 2. above are currently being supplied by the experimental program at Los Alamos.[11] Others, such as the solubility of Cs or Tc (under oxidizing conditions), may not be experimentally determined, but may be assumed to have worst-case values (no solubility limits). Decisions concerning the level of experimental effort required to provide a data base for these important waste elements will be made as the site-characterization program develops.

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