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SUMMARY ASSESSMENT OF THE RADIONUCLIDES RELEVANT TO RESEARCH ON THE GEOLOGIC DISPOSAL OF HIGH-LEVEL RADIOACTIVE WASTES

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BACKGROUND

The potential release of radionuclides to the accessible environment is the key issue of concern in the design and licensing of a geologic repository. The high-level radioactive wastes (HLW) expected to be emplaced in the proposed Yucca Mountain repository comprise a large number of radionuclides. For example, of the 103 known elements, 81 are found in spent fuel; of these, 57 have radioactive isotopes in spent fuel (Manaktala, 1993). Understanding radionuclide chemistry and transport is fundamental to appraising the potential migration of radionuclides and the long-term performance of geologic repositories. Unfortunately, the migration behavior and chemistry of radionuclides in the geologic system can be complex because they are governed by a variety of geochemical processes such as: (i) dissolution/precipitation, (ii) hydrolysis, (iii) complexation with inorganics and organics, (iv) redox reactions, (v) colloid formation, (vi) coprecipitation, and (vii) sorption reactions, etc. (Kim, 1993). As a result, many technical uncertainties exist that are related to the potential release of radionuclides. The Nuclear Regulatory Commission (NRC) staff intends to make full use of the Department of Energy (DOE) site characterization data to develop its independent understanding of basic physical processes that occur or may occur at the proposed HLW geologic repository at Yucca Mountain, Nevada; however, it needs to conduct its own investigations[1]. In view of limited resources and the expected complex chemistry and transport behavior of HLW radionuclides, it is important to identify those radionuclides on which to focus research efforts. This summary assessment seeks to address the questions: What are the criteria for selecting HLW radionuclides that should be studied in NRC-sponsored research projects? How can these be used to screen (at least in a preliminary sense) relevant radionuclides for study?

REGULATORY BASIS

The regulatory requirements set forth in Title 10 Part 60 of the Code of Federal Regulations (10 CFR Part 60) require the geologic setting of a HLW repository to exhibit an appropriate combination of geologic and geochemical conditions which are sufficient to provide reasonable assurance that releases of radioactive materials to the accessible environment following permanent closure of the repository conform to the generally applicable environmental standards for radioactivity established by the Environmental Protection Agency (EPA) and set forth in 40 CFR Part 191 (Table 1). The requirements for containment of HLW in disposal systems for 10,000 yr set forth in 40 CFR Part 191.13 are based on the likelihood of less than 1 chance in 10 of exceeding the quantities listed in Table 1 and on the likelihood of less than 1 chance in 1,000 of exceeding ten times those same quantities. (40 CFR 191 has been remanded but, until new regulations are issued, these standards will continue to be used for performance assessment analyses).

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1 Birchard, G.F. and J.D. Randall, eds. (in draft) NRC High-Level Radioactive Waste Research Program Plan. NUREG-1406.

Table 1. Release limits for containment requirements (Cumulative releases to the accessible environment for 10,000 yr after disposal) taken from 40 CFR Part 191, Appendix A

Radionuclide	Release limit per 1,000 metric tons of heavy metal or other unit of waste, curies
Americium-241 or -243	100
Carbon-14	100
Cesium-135 or -137	1,000
Iodine-129	100
Neptunium-237	100
Plutonium-238, -239, -240, or -242	100
Radium-226	100
Strontium-90	1,000
Technetium-99	10,000
Thorium-230 or -232	10
Tin-126	1,000
Uranium-233, -234, -235, -236, -238	100
Any other alpha-emitting radionuclide with a half-life greater than 20 yr	100
Any other radionuclide with a half-life greater than 20 yr that does not emit alpha particles	1,000

In addition to the overall system performance requirements, which are based on the EPA release limits, a subsystem performance measure for the engineered barrier system (EBS) is also set forth in 10 CFR 60 [specifically, Sections 113(a)(1)(i)(B) and 113(a)(1)(ii)(B)], which requires that the release rate of any radionuclide from the EBS following the containment period of 300-1,000 yr shall not exceed one part in 100,000 per yr of the inventory of that radionuclide calculated to be present at 1,000 yr following permanent closure. Radionuclides released at rates less than 0.1 percent of the calculated total radionuclide release limit are exempt from this requirement. The calculated total release limit is taken to be one part in 100,000 per year of the total inventory of radioactive waste at 1,000 yr. Consideration of these differing requirements can lead to differing lists of important radionuclides (Oversby, 1987).

Other dose based limits also apply. The EPA (40 CFR 191.15) requires that exposure to any member of the public in the accessible environment, within the first 1000 years following permanent closure of a geologic repository, not exceed 25 mrem to the whole body or 75 mrem to any critical organ in one year.

Protection requirements for groundwater are also established (40 CFR 191.16). Radionuclides that have a high potential for biological uptake and dose transfer can be considered important under these dose based performance criteria. The relative importance of radionuclides has also been evaluated based on the exposure limits outlined in 10 CFR Part 20 (Barney and Wood, 1980).

CRITERIA FOR SELECTING IMPORTANT RADIONUCLIDES

There have been numerous investigations seeking to identify radionuclides associated with HLW that are most important for research related to geologic disposal of HLW (e.g., Barney and Wood, 1980; Kerrisk, 1985; Oversby, 1987; Barnard, 1993). However, because different answers may result from consideration of different waste types, time scales, geologic and geochemical environments, and retardation and transport mechanisms, and because of the differences in the nature of the uncertainties associated with study of each radionuclide, particularly isotopes of the actinides, no single definitive list of "important" radionuclides can be provided. It is possible, however, to develop a general list of criteria that may be used to develop a list of radionuclides to be studied. Aspects of radionuclides that might be used as criteria include (in no particular order):

- half-life of the nuclide
- mode of decay
- potential leach rate of the nuclide (dissolution/solubility)
- regulatory release limit
- chemical/radiological toxicity of the nuclide
- mass and/or activity inventory of the nuclide in HLW
- propensity to form colloids in groundwater
- expected valence state, oxidation/reduction potential
- speciation/complexation in expected repository environment
- whether or not the nuclide(s) is(are) representative of the entire HLW inventory
- potential retardation of the nuclide during transport
- influence of the nuclide on the surrounding environment (competitive sorption, radiolysis)
- identified as contributing to release in performance assessment calculations
- potential use as an analogue for other nuclides of interest.

Though the experimental database is growing, many of the aspects in the preceding list, such as potential speciation, colloid formation, and retardation, that would be helpful in identifying important radionuclides are not well known. An additional criterion then might be those nuclides for which there is little data regarding chemical and/or transport behavior (Kerrisk, 1985). Other aspects in the list are heavily dependent on the waste form (e.g., leach rate and radiolysis effects) or may be closely interrelated (e.g., speciation and valence state). Nevertheless, of the factors listed above, half-lives of radionuclides are well-known, inventories of radionuclides in HLW can be determined with good confidence, and regulatory guidelines for release and personnel exposure have been established. The various criteria can be grouped to provide a rationale for selection of nuclides for research. Five broad categories can be delineated: (i) ratio of activity inventory to regulatory limits, (ii) potential for migration and transport, (iii) radionuclide thermodynamic uncertainties, (iv) repository environmental uncertainties, and (v) importance based on TSPA (Total System Performance Assessment) calculations. Each of these is discussed, in turn, in the following sections.

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Ratio of Activity Inventory to Regulatory Limit

In the U.S., the proposed repository at Yucca Mountain is currently expected to contain 70,000 MTU (metric tons of uranium) of nuclear waste: about 54 percent spent nuclear fuel from pressurized-water reactors (PWRs), 35 percent spent fuel from boiling-water reactors (BWRs), and 11 percent glassified HLW (DOE, 1988). Considering that about 90 percent of the expected HLW comprises spent nuclear fuel, a good estimate of HLW radionuclide activity inventory can be determined using values derived for typical spent fuel. (Radionuclide activity inventory present in spent fuel is a function of burnup, decay, and reactor type). A list of radionuclide activity inventories for typical PWR and BWR spent fuel was tabulated by Wilson (1991) based on the Nuclear Waste Terminal Storage Program database (Roddy et al., 1986). Both inventories are for spent fuel 10 yr after discharge from the reactor and only list isotopes with half-lives greater than 20 yr. The PWR inventory is for fuel with a burnup of 33,000 MWd/MTHM (megawatt-days per metric ton of heavy metal) and the BWR inventory is for fuel with a burnup of 27,500 MWd/MTHM. These burnups are considered typical of the spent fuel to be disposed of in a waste repository. The two inventories are similar enough so that for preliminary evaluation of important radionuclides, it should be adequate to use one of the two inventories tabulated in Wilson (1991) or a combination of them. Because of radioactive decay and associated in-growth of daughter products, this inventory will change with time.

A list of 25 radionuclides with the highest activity inventory in spent fuel 10 yr after discharge is presented in Table 2, based on Wilson's (1991) data for PWR spent fuel. The radionuclides are listed in descending order of total activity. The actinides are represented by isotopes of Np, U, Pu, Am, and Cm. The radionuclides ^{135}Cs, ^{137}Cs, ^{90}Sr, ^{99}Tc, ^{121m}Sn, ^{126}Sn, ^{151}Sm, ^{79}Se are fission products. The radionuclide ^{93}Zr is both a fission product and PWR cladding activation product. Other cladding activation products are ^{59}Ni, ^{60}Ni, and ^{94}Nb. ^{14}C comes primarily from activation of ^{14}N, which is present in both PWR fuel and cladding.

As mentioned previously, the radionuclide activity inventory in HLW will vary with time due to radioactive decay. A graphical illustration of this time dependence is given in Figure 1, which plots the activity inventory of radioactive elements in PWR spent fuel versus time. Radionuclide inventories over times of regulatory concern are tabulated in Wilson (1991), Oversby (1987) and Kerrisk (1985). Relatively short-lived radionuclides, such as ^{137}Cs and ^{90}Sr, dominate the activity inventory early (first 200 yr after discharge), but decrease sharply after that due to radioactive decay. By +1,000 yr, the actinides are the predominant radionuclides. At +10,000 yr, activation products, fission products, and actinides are all major contributors to the total inventory (Kerrisk, 1985).

The EPA standards limit 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. Compared to an inventory-based criterion, a measure that better accommodates the EPA requirements is the ratio of the radionuclide activity inventory to the EPA limit. Table 3 lists the radionuclides in PWR spent fuel along with values of inventory/EPA-limit ratio for several times. Radionuclides near the top of the list have larger values of inventory/EPA-limit ratio; thus, larger fractions of these radionuclides must be kept from reaching the accessible environment to meet the EPA standard (Kerrisk, 1985). Based solely on this criterion, nuclides whose inventory to regulatory limit ratio is significantly less than one could be excluded from the list of important nuclides.

Table 2. Radionuclides with highest activity inventory (10 yr after discharge) in PWR spent fuel with a burnup of 33,000 MWd/MTHM (from Wilson, 1991).

Nuclides	Half-life (yr)	Molar activity (Ci/mol)	Inventory in spent fuel (Ci/MTHM)	Location in spent fuel
Cs-137	3.000E+01	1.19E+04	8.21E+04	matrix, gap
Sr-90	2.912E+01	1.23E+04	5.72E+04	matrix, gap
Pu-238	8.774E+01	4.07E+03	2.33E+03	matrix
Am-241	4.322E+02	8.27E+02	1.69E+03	matrix
Ni-63	9.200E+01	3.89E+06	6.52E+02	structural
Pu-240	6.537E+03	5.47E+01	5.27E+02	matrix
Sm-151	8.999E+01	3.97E+03	3.31E+02	matrix
Pu-239	2.406E+04	1.49E+01	3.13E+02	matrix
Am-243	7.380E+03	4.84E+01	1.71E+01	matrix
Cm-243	2.850E+01	1.25E+04	1.66E+01	matrix
Tc-99	2.130E+05	1.68E+00	1.31E+01	matrix, gap
Am-242m	1.520E+02	2.35E+03	6.93E+00	matrix
Ni-59	8.000E+04	4.47E+00	5.15E+00	structural
Zr-93	1.530E+06	2.34E−01	1.93E+00	matrix, cladding
Pu-242	3.869E+05	9.24E−01	1.72E+00	matrix
C-14	5.729E+03	6.24E+01	1.55E+00	matrix, gap, cladding, structural
Nb-94	2.030E+04	1.76E+01	1.28E+00	structural
U-234	2.445E+05	1.46E+00	1.19E+00	matrix
Sn-126	1.000E+05	3.58E+00	7.76E−01	matrix, gap
Sn-121m	4.997E+01	7.15E+03	6.83E−01	structural
Se-79	6.496E+04	5.50E+00	4.09E−01	matrix, gap
Cs-135	2.300E+06	1.55E−01	3.45E−01	matrix, gap
U-238	4.468E+09	8.00E−05	3.17E−01	matrix
Np-237	2.140E+06	1.67E−01	3.15E−01	matrix
U-236	2.341E+07	1.53E−02	2.56E−01	matrix

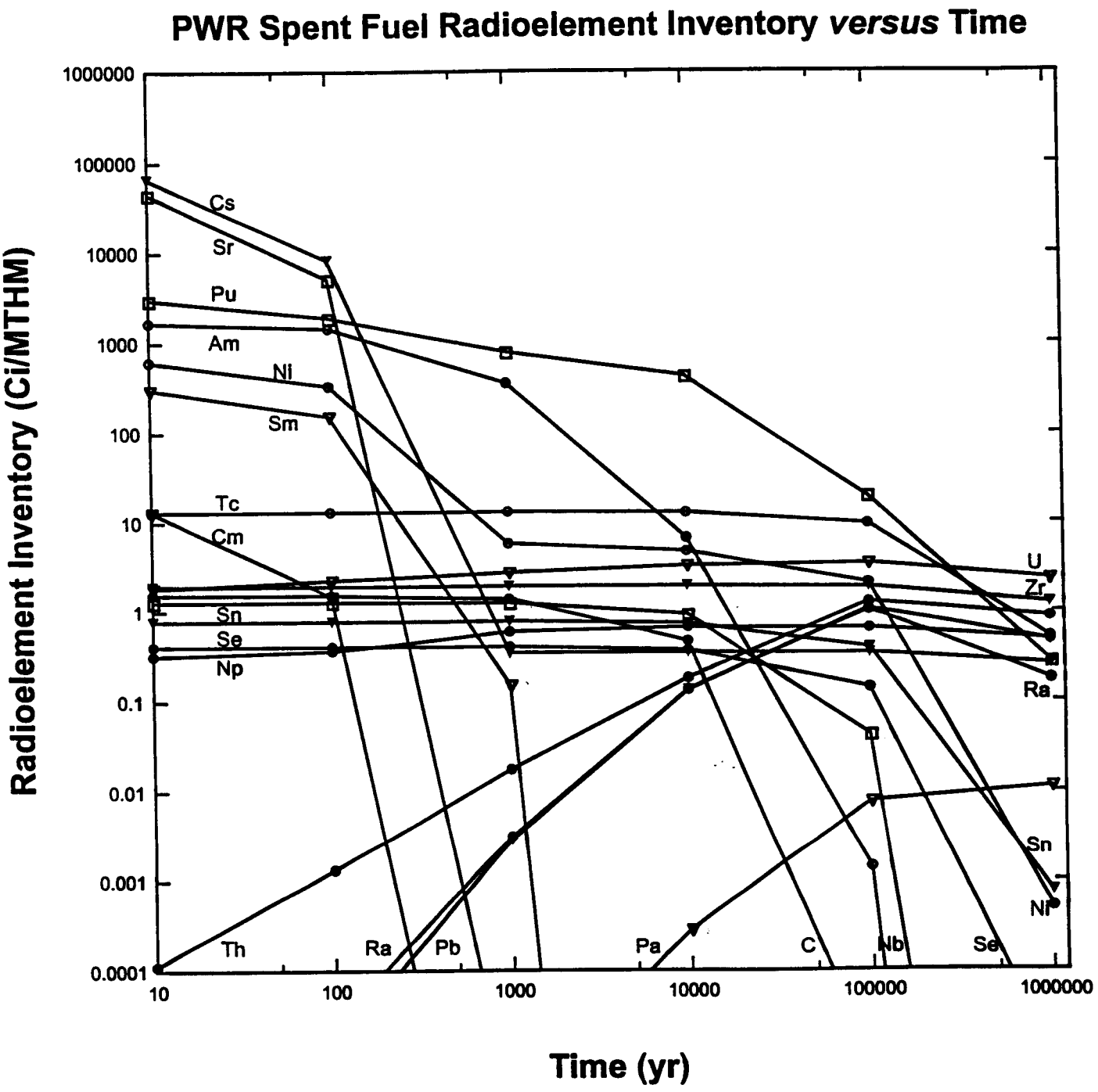

Figure 1. Inventories of various radioactive elements in PWR spent fuel as a function of time. Initial values of radionuclide inventories of PWR spent fuel (10 yr after discharge) were taken from Wilson (1991). For each element, only radionuclides with half-lives greater than 20 yr were included in calculating the total inventory.

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Table 3. Radionuclides ranked according to the ratio of their activity inventory in PWR spent fuel (Table 3) to their EPA release limit (Table 1). Values are listed for various time intervals.

+ 100 years		+ 1,000 years		+ 10,000 years	
Nuclide	Ratio of Inventory to EPA-limit	Nuclide	Ratio of Inventory to EPA-limit	Nuclide	Ratio of Inventory to EPA-limit
Am-241	1.44E+04	Pu-240	4.74E+03	Pu-239	2.38E+03
Pu-238	1.06E+04	Am-241	3.40E+03	Pu-240	1.83E+03
Cs-137	8.25E+03	Pu-239	3.04E+03	Am-243	6.68E+01
Pu-240	5.21E+03	Am-243	1.56E+02	U-234	1.99E+01
Sr-90	5.07E+03	U-234	2.03E+01	Th-230	1.71E+01
Pu-239	3.12E+03	Pu-242	1.72E+01	Pu-242	1.69E+01
Ni-63	3.26E+02	C-14	1.37E+01	U-236	9.15E+00
Am-243	1.69E+02	Pu-238	1.01E+01	Np-237	6.54E+00
Sm-151	1.53E+02	Np-237	5.88E+00	Ni-59	4.70E+00
Pu-242	1.72E+01	Ni-59	5.10E+00	C-14	4.60E+00
U-234	1.65E+01	U-236	3.57E+00	U-238	3.17E+00
C-14	1.53E+01	U-238	3.17E+00	Zr-93	1.92E+00
Cm-243	1.46E+01	Zr-93	1.93E+00	Ra-226	1.34E+00
Ni-59	5.15E+00	Th-230	1.75E+00	Tc-99	1.27E+00
Np-237	3.66E+00	Tc-99	1.31E+00	Nb-94	9.10E−01
U-238	3.17E+00	Nb-94	1.24E+00	Sn-126	7.24E−01
U-236	2.67E+00	Sn-126	7.71E−01	Se-79	3.70E−01
Zr-93	1.93E+00	Ni-63	6.41E−01	Cs-135	3.44E−01
Tc-99	1.31E+00	Se-79	4.05E−01	U-233	2.70E−01
Nb-94	1.28E+00	Cs-135	3.45E−01	Pb-210	1.32E−01
Sn-126	7.76E−01	Sm-151	1.50E−01	Th-229	9.40E−02
Se-79	4.09E−01	Ra-226	3.10E−02	U-235	2.72E−02
Cs-135	3.45E−01	U-233	2.14E−02	Pa-231	2.81E−03
Th-230	1.33E−01	U-235	3.00E−03	Am-241	1.83E−03
U-233	1.51E−03	Pb-210	2.90E−03	Th-232	3.20E−05

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Compared with an ordering of radionuclides based on their activity inventory alone, the ordering shown in Table 3 increases the relative importance of actinides because they have lower EPA limits. In the case of ^{99}Tc, which has the highest release limit of any radionuclide, ordering based on inventory/EPA-limit ratio lowers its ranking from the top five radionuclides to below the top thirteen at +1,000 and +10,000 yr. The values in Table 3 indicate that radioisotopes of the actinides are the most important radionuclides in the inventory at ~1,000 to ~10,000 yr after discharge, based on this criterion.

Potential for Migration and Transport

It is obvious that, for the radionuclides to be major contributors to releases over 10,000 yr, they must be mobilized from the waste package and transported to the accessible environment. Three types of release mechanisms are possible: (i) aqueous transport, (ii) gaseous transport, and (iii) direct surface release (e.g., by human intrusion or igneous activity). For direct surface release, the ratio of radionuclide activity inventory in the HLW to EPA release-limit at the particular time of release determines the relative importance of the different radionuclides. For instance, at time periods less than 300 yr after repository closure, contributions from radionuclides with shorter half-lives (e.g., fission products such as ^{90}Sr and ^{137}Cs) must be considered. For gaseous transport, ^{14}C is the radionuclide of most concern because of its potential transport as CO_2 gas in a hydrologically unsaturated and geochemically oxidizing environment like Yucca Mountain. However, the thermodynamic properties of $CO_2(g)$, aqueous carbonate species, and carbonate minerals are relatively well-known so that modeling of $^{14}CO_2$ transport is currently tractable (e.g., Codell and Murphy, 1992).

It is generally acknowledged that the principal release of radionuclides from the repository to the accessible environment will likely occur through transport of radionuclides by groundwater, most likely as dissolved species and possibly as colloids or particulates. For the period 1,000 to 10,000 yr after permanent closure, the geologic system is expected to provide the major barrier to migration of radionuclides to the accessible environment, and geochemical processes involving dissolution, precipitation, and sorption will be important to the retardation of radionuclide migration. Because the actinides dominate the inventory to regulatory limit ratio of spent fuel during this time period, it is clear that information on the solubility and sorption behavior of Pu, Am, U, and Np is critical to understanding their migration behavior, as well as the migration of their decay products (e.g., ^{230}Th, ^{226}Ra, and ^{210}Pb). This conclusion is consistent with the recommendations made to DOE by Kerrisk (1985). More recently, Barnard (1993) reviewed the aspects of radionuclide source terms used for DOE's TSPA analyses and concluded that Kerrisk's observation regarding the importance of Am, Pu, Np, and U remains valid.

Preliminary data suggest that Pu and Am solids have low solubilities in Yucca Mountain groundwaters (Nitsche et al., 1993; Kerrisk, 1985) and high sorption coefficients, both of which favor retardation of Pu and Am migration. In the case of Th, its estimated sorption coefficient is similar to those of Pu and Am, whereas its estimated solubility is a few orders of magnitude lower than those of Pu and Am (Kerrisk, 1985); thus retardation of Th transport is favored to a greater degree relative to Pu and Am. On the other hand, U and Np have higher solubilities in Yucca Mountain groundwaters (Ogard and Kerrisk, 1984; Nitsche et al., 1991; Nitsche et al., 1993) and lower sorption coefficients (Meijer, 1990), which make them more susceptible to transport by groundwaters to the accessible environment and more likely to exceed the EPA release limits. Other radionuclides also have low sorption coefficients; ^{79}Se, ^{99}Tc, and ^{129}I have all been shown to be potential major contributors to release because of their high transport potential (Simmons, 1992). Though U has lower sorption coefficients than Pu and Am, its higher solubility compared to Pu and Am and its much higher mass-inventory compared to all the other

radioelements may cause sorption of these other radionuclides to be less effective due to competition for sorption sites. The effect of competitive sorption on radionuclide migration is not known because very limited data are available.

It is important to note that spent fuel is composed of >95 percent UO_2 (Johnson and Shoesmith, 1988). The great majority of radionuclides are contained within the grains of the UO_2 fuel pellets, even though a small percentage (typically 0.1 to 5 percent), notably Cs and I, migrates to the fuel/cladding gap and the grain boundaries in the fuel (Gray et al., 1992). Table 2, for example, gives the location in the spent fuel of the radionuclides with the highest activity inventory. The actinides, in particular, are expected to be in solid solution with the UO_2 fuel. Thus the major factor controlling the long-term release of soluble radionuclides is the oxidative dissolution of the UO_2 fuel matrix (Forsyth and Werme, 1992; Shoesmith and Sunder, 1992). This fact may have significant relevance to meeting the gradual release limits set forth in 10 CFR Part 60.

Radionuclide Thermodynamic Uncertainties

Performance assessment calculations regarding release of radionuclides to the accessible environment require a knowledge of the key parameters that affect radionuclide solubility and sorption behavior and also require radionuclide dissolution and transport models that properly account for the effects of these parameters. It is evident from published data that sorption processes are influenced by several factors, including physical and chemical properties of the rock and the groundwater; thus, predictions of radionuclide transport using empirically-derived distribution coefficients (Kds) are frequently unjustified (Turner, 1991). On the other hand, incorporation of sophisticated geochemical models in transport codes require a well-constrained thermodynamic dataset on aqueous speciation and solid phase stabilities for the radionuclide of interest. Based on the values listed in Table 3 and a consideration of potential for migration and transport, it is clear that reliable predictions of transport are most needed for Pu, Am, U, and Np. Among these actinide elements, U has the most extensively evaluated set of thermodynamic parameters (e.g., Grenthe et al., 1992), although various laboratories are currently generating new data on the other actinides (e.g., Nitsche et al., 1993). The detailed aqueous chemistry of U is different from that of the other actinides; however, the latter exhibit similarly strong tendencies towards hydrolysis and polynuclear ion formation as well as aqueous complex formation (Cotton and Wilkinson, 1980; Allard, 1982). Thus, U is a good analogue for understanding the transport and retardation behavior of the other actinides and for evaluating different approaches for quantitative prediction of actinide migration in complex geochemical systems (Chapman et al., 1984). Studies involving U solubility, sorption and transport can be used to develop experimental and modeling approaches, which can then be applied to Pu, Am, and Np.

Repository Environmental Uncertainties

The geochemical environment of the proposed HLW repository will directly affect the source term and transport behavior of radionuclides. The fact that U is the predominant radioelement on a mass-basis throughout the period of regulatory concern has important implications to near-field and far-field geochemical processes. For example, interaction between the groundwater and the UO_2 spent fuel will shift substantially the chemistry of the groundwater, such as pH and oxidation state (Apted, 1989). This, in turn, can change the solubility limits of radionuclides and affect the stability of radiocolloids. Furthermore, precipitation of various primary and secondary uranyl solid phases, e.g., uranophane, is likely to occur during the course of groundwater/UO_2-fuel interaction. This is evidenced by results of

laboratory-scale dissolution experiments on unirradiated UO_2 (e.g., Wronkiewicz et al., 1992) and spent fuel (Wilson, 1990; Wilson and Bruton, 1989), as well as by the occurrence of secondary U minerals associated with altered UO_2 ore deposits (Pearcy et al., 1993). Incorporation of various radionuclides, particularly those ranked high in Table 3, with these precipitates is possible, which will contribute to the retardation of their migration to the accessible environment (Bruno and Sandino, 1988). On the other hand, it is feasible that these uranyl solid phases, which may precipitate some distance away from the waste package, may serve as a secondary, but important, source of U and coprecipitated radionuclides (Tripathi et al., 1989) at some distance which may be closer to the accessible environment.

Importance Based on TSPA Calculations

The release limits given in Table 1 are the quantitative bases for evaluating the overall performance of a geologic repository (Barnard et al., 1992; EPRI, 1992; Wescott et al., 1994). However, these Total System Performance Assessment (TSPA) calculations are based on preliminary hypotheses and conceptual models and their results may change as knowledge advances. Thus, existing TSPA results by themselves are not sufficient for ranking the radionuclides for research. However, TSPA calculations do provide feedback to investigators regarding radionuclides and repository conditions most likely to play a critical role in repository performance.

Using available geochemical data and assumptions based on empirical relationships, TSPA results have identified several nuclides which have high release potential. ^{237}Np and ^{79}Se, both of which have relatively high solubilities and HLW inventories, were major contributors to radionuclide release in the Electric Power Research Institute's TSPA (EPRI, 1992). ^{99}Tc and ^{129}I dominated aqueous releases in the TSPA done by Sandia National Laboratories (Barnard et al., 1992); those nuclides are highly soluble and are considered nonsorbing. Significant contributions to release in the Sandia TSPA were also made by ^{79}Se, ^{234}U, and ^{237}Np, which are weakly sorbing radionuclides. Although these TSPA results are preliminary and are dependent on the particular data and models used, it is interesting to note that Pu and Am, which are ranked highest based on inventory/EPA-limit ratio (Table 3), are not among the radionuclides with highest release to the accessible environment. Thus, factors in addition to the inventory/EPA-limit ratio need to be considered in evaluating the importance of radionuclides. Barnard (1993) presents an excellent review of the factors that should be applied to the selection of radionuclides to provide a more sensitive source term for TSPA. Among the critical factors listed are how well the radionuclide represents the HLW inventory, dose conversion factors, estimated solubility distributions, and "indicator" distribution coefficients (Kds). Barnard's recommended listing includes nearly all of the nuclides listed as important by previous studies (Kerrisk, 1985; Oversby, 1987)

RECOMMENDATIONS

Determination of the radionuclides on which research supporting a proposed high-level waste repository should be focused depends on a number of factors. These factors include the mission of the research project, the system conceptual model, system performance requirements, and data uncertainties. In general, the mission of NRC-sponsored research is to develop an independent understanding of the basic physical processes surrounding the geologic disposal of HLW. This mission is served not only by utilizing data on radionuclides that is better constrained (e.g., U sorption experiments) but also by developing data in areas where much uncertainty exists (e.g., Np and Pu sorption). Some criteria that may be used to identify important radionuclides have been outlined in this document. The specific goals of an individual research project can be combined with these criteria to identify radionuclides relevant to that project.

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Based on the CNWRA's Sorption Project goals of identifying sorption processes that are most important in the Yucca Mountain geologic system and the physical and chemical parameters that control these processes, and based on the identification of important nuclides from the criteria outlined in this paper, additional experiments on actinides other than U are warranted. The relationship between aqueous speciation and sorption behavior, which has been observed for U (e.g., Pabalan et al., 1993), needs to be investigated for Pu, Am, and Np. This will entail conducting experiments over a wide range of pH, actinide concentration, and CO_2 partial pressure. A critical evaluation of thermodynamic data for aqueous species of Pu, Am, and Np will also be needed. The importance of sorbent properties (e.g., crystal structure, pH of zero charge, site density, etc.) needs to be investigated by using a variety of mineral types as the sorbent phase. In addition, competition among different radionuclides for sorption sites and its effect on the retardation of radionuclide migration is something that has received little attention and deserves intensive study. The potential importance of U mineral precipitates as a secondary source of radionuclides should also be considered, particularly at temperatures above 25 °C. Thus, experimental studies, e.g., uranophane solubility and solid solution experiments, designed to derive those properties are warranted. In addition, the modeling study of release rates from spent fuel should eventually be expanded to include Pu, Am, Np and other radionuclides. CNWRA investigators are in the process of acquiring Pu and Np spikes and designing and setting-up experiments to study the sorption behavior of Pu and Np. Continued CNWRA participation in programs focused on radionuclide sorption/transport processes is also recommended.

REFERENCES

Allard, B. 1982. Solubilities of actinides in neutral or basic solutions. *Actinides in Perspective.* N.M. Edelstein, ed. Elansford, NY: Pergamon Press: 553–580.

Apted, M.J. 1989. Engineered barrier system of the near field: The "Little Brother" of performance assessment. *Proceedings of the Symposium on Safety Assessment of Radioactive Waste Repositories*. Organization for Economic Cooperation and Development. Paris, France: Nuclear Energy Agency: 471–480.

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

Barnard, R.W. 1993. *Review of Radionuclide Source Terms used for Performance-Assessment Analyses.* SAND92-2431. Albuquerque, NM: Sandia National Laboratories.

Barney, G.S., and B.J. Wood. 1980. *Identification of Key Radionuclides in a Nuclear Waste Repository in Basalt.* RHO-BWI-ST-9. Richland, WA: Rockwell International, Basalt Waste Isolation Project.

Bruno, J., and A. Sandino. 1988. The thermodynamics and kinetics of coprecipitation and its effect on radionuclide solubility. *Radiochimica Acta* 44/45: 17–21.

Chapman, N.A., I.G. McKinley, and J.A.T. Smellie. 1984. *The Potential of Natural Analogues in Assessing Systems for Deep Disposal of High-Level Radioactive Waste.* EIR-Report No. 545: Swiss Federal Institute for Reactor Research.

Codell, R.B., and W.M. Murphy. 1992. Geochemical model for ^{14}C transport in unsaturated rock. *Proceedings of the Third International Conference on High-Level Radioactive Waste Management* 2. La Grange Park, IL: American Nuclear Society: 1,959–1,965.

Cotton, F.A., and G. Wilkinson. 1980. *Advanced Inorganic Chemistry*. New York, NY: John Wiley & Sons.

Electric Power Research Institute. 1992. *Demonstration of a Risk-Based Approach to High-Level Waste Repository Evaluation Phase 2*. EPRI TR-100384: Palo Alto, CA.

Forsyth, R.S., and L.O. Werme. 1992. Spent fuel corrosion and dissolution. *Journal of Nuclear Materials* 190: 3–19.

Gray, W.J., H.R. Leider, and S.A. Steward. 1992. Parametric study of LWR spent fuel dissolution kinetics. *Journal of Nuclear Materials* 190: 46–52.

Grenthe, I., J. Fuger, R. Konings, R. Lemire, A. Muller, C. Nguyen-Trung, and H. Wanner. 1992. *Chemical Thermodynamics of Uranium*. Amsterdam: North-Holland.

Johnson, L.H., and D.W. Shoesmith. 1988. Spent Fuel. *Radioactive Waste Forms for the Future*.W. Lutze and R.C. Ewing, eds. Amsterdam: Elsevier. 635–698.

Kerrisk, J.F. 1985. *An Assessment of the Important Radionuclides in Nuclear Waste*. LA-10414-MS. Los Alamos, NM: Los Alamos National Laboratory.

Kim, J.I. 1993. The chemical behavior of transuranium elements and barrier functions in natural aquifer systems. *Scientific Basis for Nuclear Waste Management XVI*. C.G. Interrante and R.T. Pabalan, eds. Pittsburgh, PA: Materials Research Society: MRS Symposium Proceedings 294. 1–12.

Manaktala, H.K. 1993. *Characteristics of Spent Nuclear Fuel and Cladding Relevant to High-Level Waste Source Term*. CNWRA 93-006. San Antonio, TX: Center for Nuclear Waste Regulatory Analyses.

Meijer, A. 1990. *Yucca Mountain Project Far-Field Sorption Studies and Data Needs*. LA-11671-MS. Los Alamos, NM: Los Alamos National Laboratory.

Nitsche, H., R.C. Gatti, E.M. Standifer, S.C. Lee, A. Muller, T. Prussin, R.S. Deinhammer, H. Maurer, K. Becraft, S. Leung, and S.A. Carpenter. 1991. *Measured Solubilities and Speciations of Neptunium, Plutonium, and Americium in a Typical Groundwater (J-13) from the Yucca Mountain Region*. LBL-30958. Berkeley, CA: Lawrence Berkeley Laboratory.

Nitsche, H., K. Roberts, T. Prussin, D. Keeney, S.A. Carpenter, K. Becraft, and R.C. Gatti. 1993. Radionuclide Solubility and Speciation Studies for the Yucca Mountain Site Characterization Project. *High-Level Radioactive Waste Management, Proceedings of the Fourth Annual International Conference, Las Vegas, NV, April 26-30, 1993*. 1,490–1,495.

Ogard, A.E., and J.F. Kerrisk. 1984. *Groundwater Chemistry Along Flow Paths Between a Proposed Repository Site and the Accessible Environment*. LA-10188-MS. Los Alamos, NM: Los Alamos National Laboratory.

Oversby, V.M. 1987. Important Radionuclides in High-Level Nuclear Waste Disposal: Determination Using a Comparison of the U.S. EPA and NRC Regulations. *Nuclear and Chemical Waste Management* 7: 149–161.

Pabalan, R.T., J.D. Prikryl, P.M. Muller, and T.B. Dietrich. 1993. Experimental study of uranium(6+) sorption on the zeolite mineral clinoptilolite. *Scientific Basis for Nuclear Waste Management XVI*. C.G. Interrante and R.T. Pabalan, eds. Pittsburgh, PA: Materials Research Society: MRS Symposium Proceedings 294: 777-782.

Pearcy, E.C., J.D. Prikryl, W.M. Murphy, and B.W. Leslie. 1993. *Uranium Mineralogy of the Nopal I Natural Analog Site, Chihuahua, Mexico*. CNWRA 93-012. San Antonio, TX: Center for Nuclear Waste Regulatory Analyses.

Roddy, J.W., H.C. Claiborne, 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, TN: Oak Ridge National Laboratory.

Simmons, A.M. 1993. Significance of Geochemical Characterization to Performance at Yucca Mountain, Nevada. *High-Level Radioactive Waste Management, Proceedings of the Fourth Annual International Conference, Las Vegas, NV, April 26-30, 1993*. 1,482–1,489.

Shoesmith, D.W., and S. Sunder. 1992. The prediction of nuclear fuel (UO_2) dissolution rates under waste disposal conditions. *Journal of Nuclear Materials* 190: 20–35.

Tripathi, V.S., G.T. Yeh, and G.K. Jacobs. 1989. *Simulation of Groundwater-Transport-Dynamics of Chemically Reactive Radionuclides*. Letter Report LR-287-84. Oak Ridge, TN: Oak Ridge National Laboratory.

Turner, D.R. 1991. *Sorption Modeling for High-Level Waste Performance Assessment: A Literature Review*. CNWRA 91-011. San Antonio, TX: Center for Nuclear Waste Regulatory Analyses.

Turner, D.R. 1993. *Mechanistic Approaches to Radionuclide Sorption Modeling*. CNWRA 93-019. San Antonio, TX: Center for Nuclear Waste Regulatory Analyses.

U.S. Department of Energy. 1988. *Site Characterization Plan*. (Site Hydrologeologic System). DOE/RW-0199. Washington, DC: Department of Energy.

U.S. Environmental Protection Agency. 1989. *Environmental Radiation Protection Standards for Management and Disposal of Spent Nuclear Fuel, High-Level and Transuranic Radioactive Wastes*. Title 40, Protection of Environment, Part 191 (40 CFR Part 191). Washington, DC: U.S. Government Printing Office.

U.S. Nuclear Regulatory Commission. 1992. *Disposal of High-Level Radioactive Wastes in Geologic Repositories*. Title 10, Energy, Part 60 (10 CFR Part 60). Washington, DC: U.S. Government Printing Office.

U.S. Nuclear Regulatory Commission. 1992. *Standards for Protection Against Radiation*. Title 10, Energy, Part 20 (10 CFR Part 20). Washington, DC: U.S. Government Printing Office.

Wescott, R.G., M.P. Lee, N.A. Eisenberg, and T.J. McCartin, eds. 1994. *Phase 2 Demonstration of the NRC's Capability to Conduct a Performance Assessment for a High-Level Waste Repository*. NUREG-1464. Washington, DC: U.S. Nuclear Regulatory Commission.

Wilson, C.N., and C.J. Bruton. 1989. *Studies on Spent Fuel Dissolution Behavior Under Yucca Mountain Repository Conditions*. UCRL-100223. Livermore, CA: Lawrence Livermore National Laboratory.

Wilson, C.N. 1990. *Results from NNWSI Series 2 Bare Fuel Dissolution Tests*. PNL-7169/UC-802. Richland, WA: Pacific Northwest Laboratory.

Wilson, M.L. 1991. *A Simplified Radionuclide Source Term for Total-System Performance Assessment*. SAND91-0155. Albuquerque, NM: Sandia National Laboratories.

Wronkiewicz, D.J., J.K. Bates, T.J. Gerding, E. Veleckis, and B.S. Tani. 1992. Uranium release and secondary phase formation during unsaturated testing of UO_2 at 90 °C. *Journal of Nuclear Materials* 190: 107–127.