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RELEASES FROM EXOTIC WASTE PACKAGES FROM PARTITIONING AND TRANSMUTATION

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ABSTRACT

Partitioning the actinides in spent nuclear fuel and transmuting them in actinide-burning liquid-metal reactors has been proposed as a potential method of reducing the public risks from geologic disposal of nuclear waste. To quantify the benefits for waste disposal of actinide burning, we calculate the release rates of key radionuclides from waste packages resulting from actinide burning, and compare them with release rates from LWR spent fuel destined for disposal at the potential repository at Yucca Mountain. The wet-drip water-contact mode has been used. Analytic methods and parameter values are very similar to those used for assessing Yucca Mountain as a potential repository. Once released, the transport characteristics of radionuclides will be largely determined by site geology. For the most important nuclides such as I-129 and Tc-99, which are undiminished by actinide-burning reactors, it is not surprising that actinide burning offers little reduction in releases. For important actinides such as Np-237 and Pu isotopes, which are reduced in inventory, the releases are *not* reduced because the release rates are proportional to solubility, rather than inventory.

1. Introduction

Partitioning the actinides in light-water reactor (LWR) spent fuel and transmuting them in actinide-burning liquid-metal reactors (ALMR) has been put forth as a potential method of reducing the public risks from geologic disposal of nuclear waste. However, the real benefits of such partitioning and transmutation for waste disposal have not been analysed. Efforts to quantify these benefits are now underway. This paper provides the following

a. Elucidation of an equal energy produced basis of comparison.

b. Characteristics and inventories of exotic waste packages from aqueous and pyro-reprocessing schemes,

c. Release rates of selected radionuclides that are likely to travel to the accessible environment from the potential repository at Yucca Mountain.

2. Need for Evaluating the Benefits of Partitioning and Transmutation

The slow pace of technological progress as well as seemingly overwhelming public opposition to geologic disposal of spent nuclear fuel has brought forth the concepts of partitioning and transmutation to reduce the risks to the public of waste disposal. Spent nuclear fuel can be reprocessed, and the waste can be *partitioned* or separated into elemental fractions which can then be *transmuted* into stable or short-lived isotopes by bombardment with neutrons. Partitioning involves chemical processes and can be done in a reprocessing facility. Transmutation can be accomplished in accelerators or reactors. Actinide burning is the concept of using the transuranics in LWR spent fuel in a liquid-metal fast reactor to generate electricity as well as perform transmutation.

While the technology for partitioning and transmutation was developed in the 1970's and 1980's, the waste disposal community has always regarded the benefits from partitioning and transmutation to be marginal, compared to the magnitude of the undertaking.[1] However, recent difficulties at Yucca Mountain[2] have given new impetus to partitioning and transmutation.

The main claimed benefits of partitioning and transmutation are

- partitioning and transmutation reduce health risk to future generations.
- partitioning and transmutation reduce the heat placed in the repository.
- partitioning and transmutation ease the licensing of a repository.
- partitioning and transmutation make the repository more acceptable to the public.

Only the first two claims can be evaluated quantiatively. In this paper we give the inventories of major nuclides in the repository from various schemes, and calculate the release of the nuclides from waste packages. The inventories and release rates are used by total systems analysts.

3. An Equal Energy Production Comparison

In this Section we describe the basis of comparing the reference case of spent-fuel disposal at Yucca Mountain with two variants of partitioning and transmutation.

The schemes being compared are shown in Figure 1.

Scheme 1 is disposal of light-water reactor spent-fuel.

In Scheme 2, the geologic repository receives waste from the reprocessing of LWR and the reprocessing of ALMR fuel. In order to provide initial fuel, reloads and makeup for actinide-burning liquid-metal reactors, light-water reactor (LWR) spent-fuel is reprocessed, by either conventional aqueous reprocessing technology, the PUREX process,[3] or pyrochemical reprocessing technology under development.[4] We shall take the 63000 MTIHM of LWR spent fuel destined for the first repository and reprocess for use in the General Electric PRISM reactor,[5] the reference U. S. Department of Energy advanced liquid-metal reactor. We assume

- Nine modules of PRISM produce 1395 MWe;
- The reactors have 40 years of economic life;
- The ALMR's have a capacity factor of 0.8 and conversion ratio of 0.76.

In Scheme 2a, the LWR spent fuel is reprocessed with pyrochemical processes, and the ALMR fuel recycled using pyrochemical processes. We shall designated waste streams in the pyro-processing of LWR fuel as A1-x, and waste streams in the pyro-processing of ALMR fuel as A3-x.[6]

In Scheme 2b, the LWR spent fuel is reprocessed with aqueous processes, and the ALMR fuel processed with pyrochemical processes. We shall designated waste streams in the aqueous processing of LWR fuel as B1-x, and waste streams in the pyro-processing of ALMR fuel as A3-x.[6]

For the following calculations, we use 33,000 MWd/ton burnup fuel from pressurized water reactors as the reference case. With each Mg U or MTIHM of LWR spent fuel giving 9.72 kg of transuranics, 3878 MTIHM of LWR spent fuel are needed to support one 1395-MWe ALMR, and the 63,000 MTIHM would support about 16 in all. In the course of their economic lives, these 16 ALMR's would produce 9.1×10^5 MWe-a of energy. Thus a repository serving Scheme 2a or 2b would contain the waste of 9.1×10^5 MWe-a of energy plus the waste from the reprocessing of 63,000 MTIHM of LWR spent fuel. For a fair or equal energy produced comparison, we now add to Scheme 1 the equivalent LWR spent fuel that would have resulted from the generation of 9.1×10^5 MWe-a of energy using LWR's, or a total of

$$63000 + 9.1 \times 10^5 \times \frac{28 \text{ MTIHM SF}}{1000\text{MWe} - \text{a}} = 88400 \text{ MTIHM}$$

Therefore the repository serving Scheme 1 should contain 88400 MTIHM for an equal-energy produced comparison.

4. Waste Characteristics and Inventories

Figure 2 shows the waste streams from the three Schemes. The waste characteristics and inventories were originally developed by Thompson and Taylor,[6] revised by Wilems and Danna[7] and we revised them further. We use the inventories given by Thompson and Taylor.[6] We adopted the simplified waste packaging suggested by Wilems and Danna[7] and their per package thermal limit of 2.5 Kw/package.

These are the major modifications we made.

- In Scheme 2b we considered low-recovery (99.9%) aqueous processing. The inventories we use are from the high-recovery (99.999%) tables by Thompson and Taylor and scaled back to 99.9%. For pyro-processing, we use 99.9% recovery.

- Where I-129 is considered a gas, we convert it to AgI, a low-solubility compound that is a more leach-resistant waste form.

- We put the fuel hardware from ALMR (A3-2) into the electro-refining metal waste (A3-5), which has a copper matrix, forming A3-2,5.

- Gaseous nuclides and short-lived wastes such as A1-5 can be allowed to decay. If disposed in any repository, these species will not affect dose to humans except in human intrusion scenarios.

Table 1 shows the waste packages from pyro-processing of LWR spent fuel. Table 2 shows the waste packages from aqueous processing of LWR spent fuel. Table 3 shows the waste packages from pyro-processing of ALMR fuel. In each case, the dimensions, materials, heat output, matrix and number of packages are shown.

In this study, we track 33 radionuclides. They have been chosen because of their significance in waste disposal. Such species have one or more of the following characteristics

- Long half life
- Large inventory
- High toxicity
- High heat generation
- Low sorption.

Radioactive inventories of waste packages have been calculated for 10, 100, 300, 1000, 5000 and 10,000 years after emplacement.[8]

5. Calculation of Release Rates

We assume that waste from LWR and ALMR cycles will be placed in the potential repository at Yucca Mountain.

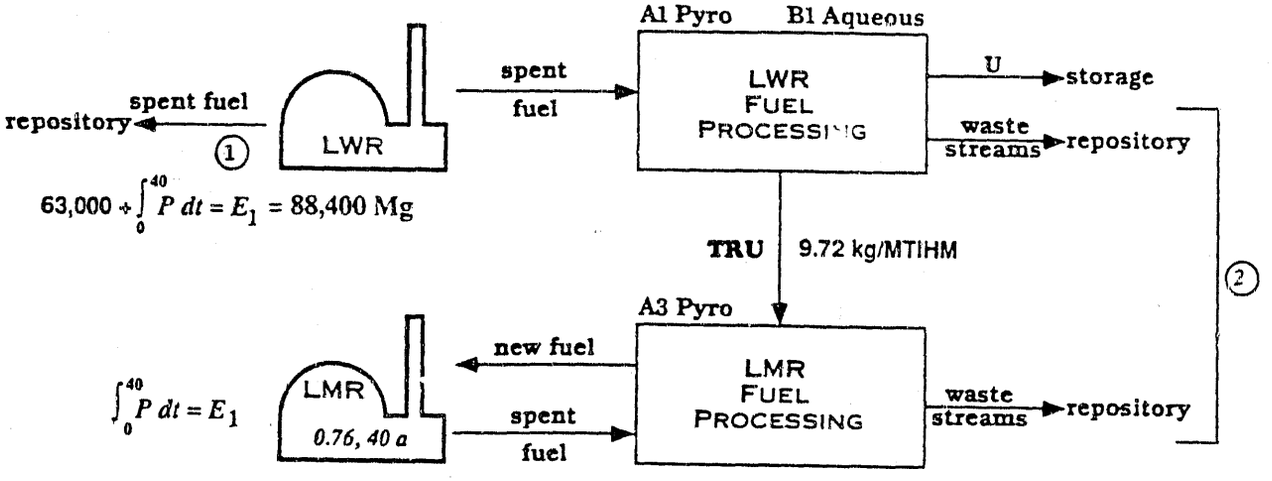

Figure 1. Equal Energy Production Comparison

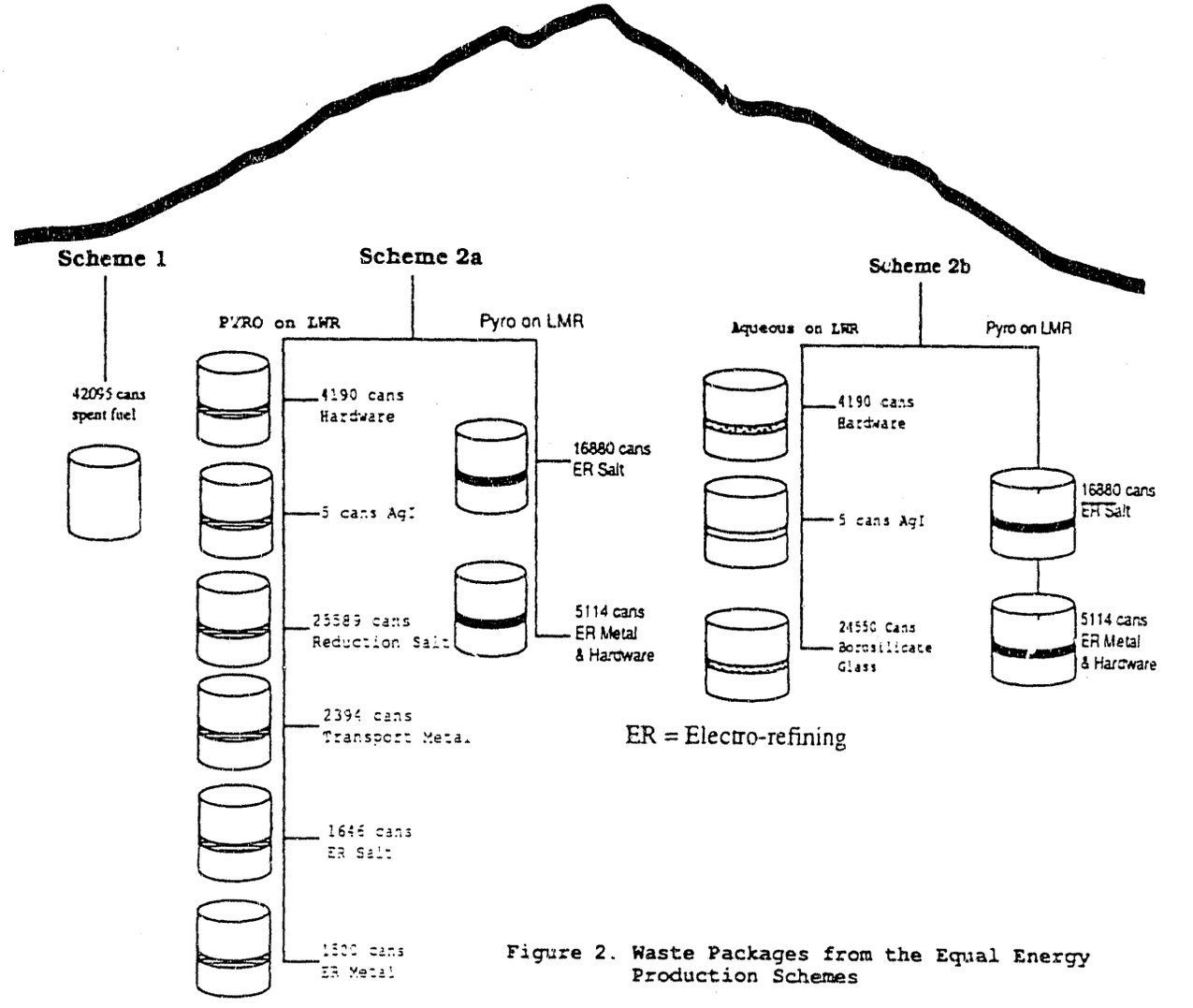

Figure 2. Waste Packages from the Equal Energy Production Schemes

Table 1. Waste packages from pyro-processing of LWR spent fuel

Pyro-processing of LWR SF	A1-1,2	A1-3	A1-4	A1-6	A1-7	A1-8
Waste Stream	Hardware	Gases	Reduction Salt	Transport Metal	Electro-refining Salt	Electro-refining Metal
Container Type	6	6	5	6	5	5
Inside Container Dia (m)	0.59	0.59	0.4	0.59	0.4	0.4
Inside Container Ht (m)	5.0	5.0	5.0	5.0	5.0	5.0
Inside X-Sectional Area (m^2)	0.273	0.273	0.125	0.273	0.125	0.125
Container Material	SS	SS	SS	SS	SS	SS
Outside Container Dia (m)	0.66	0.66	0.47	0.66	0.47	0.47
Outside Container Ht (m)	5.22	5.22	5.22	5.22	5.22	5.22
Outside X-Sectional Area (m^2)	0.342	0.342	0.173	0.342	0.173	0.173
Waste Volume (m^3)	1.16	1.16	0.53	1.16	0.53	0.53
Void Volume (m^3)	0.624	0.624	0.37	0.624	0.37	0.37
MATRIX	None	AgI	Zeolite	Copper	Zeolite	Copper
KW/pkg at 10 years	0.57	0.00126	2.08	0.19	0.35	1.45
Number of Containers	4190	5	25589	2394	1646	1500

Table 2. Waste packages from aqueous processing of LWR spent fuel

Waste Stream	B1-2	B1-3	B1-4
	Hardware	Gas	Glass
Inside Container Diameter (m)	0.59	0.59	0.4
Inside Container Height (m)	5.0	5.0	5.0
Inside X-Sectional Area (m^2)	0.273	0.273	0.125
Container Material	SS	SS	SS
Outside Container Dia. (m)	0.66	0.66	0.47
Outside Container Ht. (m)	5.22	5.22	5.22
Outside X-Sectional Area (m^2)	0.342	0.342	0.173
Waste Volume (m^3)	1.16	1.16	0.53
Void Volume (m^3)	0.624	0.624	0.37
MATRIX	None	AgI	Glass
Kilowatts/Package at 10 years	0.57	0.00126	2.50
Number of Containers	4191	5	24550

Table 3. Waste packages from pyro-processing of ALMR fuel

Waste Stream	A3-4	A3-2,5
	Electro-refining Salt	Hardware & Electro-refining Metal
Inside Container Dia. (m)	0.59	0.59
Inside Container Ht. (m)	5.0	5.0
Inside X-Sectional Area (m^2)	0.273	0.273
Outside Can Dia. (m)	0.66	0.66
Outside Can Ht. (m)	5.22	5.22
Outside X-Sectional Area (m^2)	0.342	0.342
Container Material	SS	SS
Waste Volume (m^3)	1.16	1.16
Void Volume (m^3)	0.624	0.624
MATRIX	Zeolite	Copper
Kilowatts/Package at 10 years	0.70	1.84
Number of Containers	16880	5114

Current design calls for vertical emplacement of waste containers, and for the containers to be surrounded by an air gap. Although the waste package is generally not seen as the primary barrier for nuclear waste isolation, it must in fact meet specific regulatory requirements. In 10 CFR 60.113(a)(1)(ii)(B), the U. S. Nuclear Regulatory Commission requires that the release rate of any radionuclide from the engineered barrier system following the containment period shall not exceed one part in 100,000 per year of the inventory of that radionuclide calculated to be present at 1,000 years following permanent closure. For low-inventory radionuclides, those that constitute less than 0.1 percent of the calculated total curie inventory at 1,000 years, the allowable annual release is a constant value, equal to 10^{-8} of the total curie inventory in the repository at 1,000 years. The release rate is input to total system performance calculations. Therefore it is necessary to calculate release rates for waste packages at Yucca Mountain.

We calculate release rates for the selected radionuclides using analytic solutions in Sadeghi *et al.*[9] for the wet-drip bathtub water-contact mode. For the radionuclides, we consider the release of three types of species: solubility-limited species, species released congruent with solid-solid alteration of the waste matrix, and readily soluble species. In each case we give the release rates of the species as a function of time.

5.1 The Wet-Drip Water-Contact Mode

Here we refer to the dripping of water from overhead rock onto waste packages. This dripping may happen because of episodic fracture flow or a change in rock permeability may divert water into fractures that intersect the borehole. Drips are assumed to penetrate cracks in a failed container and to dissolve radionuclides as the radionuclide solution slowly rises in the container and finally overflows through other cracks and penetrations. Overflow of contaminated water is assumed to occur only near the top of the container. The contaminated water drips to the rock below. Water within the container is always well mixed from diffusion and thermal convection. We refer to this as the "wet-drip bathtub water-contact mode." We showed in Sadeghi *et al.*[10] that the release rates from the wet-drip bathtub water-contact mode are not very different from the wet-drip flow-through or the moist-continuous water-contact modes.

For details of calculations of release rates from LWR spent fuel (Scheme 1), see Sadeghi *et al.*[10]

5.2 Parameters Adopted for Calculating Release Rates

Hydrogeologic Conditions

The far-field averaged flux at the emplacement horizon is taken to be 0.5 mm/a, which appears to be an upper bound for expected conditions.[11] For the wet-drip water-contact mode we assumed that water contact begins at 1000 years after emplacement.

Release Mechanisms

For the exotic waste matrix encountered in pyrochemical processing, Table 4 summarizes the release mechanisms for actinides and fission products.

Table 4. Release Mechanisms

Matrix	None	AgI	Zeolite	Copper	Glass
Actinides	Solubility-limited	NA	NA	Solubility-limited	Solubility-limited
Fission Products	Instant	Solubility-limited	Instant	Alteration-controlled	Alteration-controlled

Table 5. Solubility Data

Solubility (g/m^3)		
	LWR SF & Copper	Glass
Np	3.0×10^{-4}	9.4×10^{-2}
Pu	9.5×10^{-4}	3.8×10^{-8}
U	0.3	6×10^{-2}
Am	3.8×10^{-5}	1.5×10^{-3}
Source	Ref. 12	Ref. 13

Solubility

For calculating the release rates of the solubility-limited species, the elemental solubility is needed. For solubilities of U, Np, Pu and Am dissolving from hardware and copper matrix, we use the concentrations of these elements measured in hot-cell leaching experiments of decladded LWR spent fuel,[12] shown in Table 5.

Solubilities of U, Np, Pu and Am dissolving from borosilicate glass have been calculated using the geochemical code EQ3/6 to simulate hot-cell leaching experiments of Wilson, also shown in Table 5.[13]

See Sadeghi *et al.*[10] for a discussion of the uncertainties in these solubilities.

For AgI, the solubility was obtained from the commonly known solubility product constant.[14]

Matrix Alteration Rates

For LWR spent fuel, we use an UO_2 alteration rate of 10^{-3} per year from Wilson's leaching experiments.[15]

For borosilicate glass, there is a slow corrosion reaction which releases fission products and actinides. From the

experimental dissolution rate of lithium from borosilicate glass,[16] the rate of reaction of the SiO_2 glass matrix with water is 5.2 g/m^2-a. For a container with 1660 kg glass and assuming that the total reaction surface area, due to internal cracks, is 25 times the geometrical surface area (0.27 m^2),[17,13] the reaction rate would become 36 g/a. This results in a fractional alteration rate of 2×10^{-5}/a.

Several of the new waste containers have copper matrix. Elemental copper is not stable in the oxidizing environment at Yucca Mountain. To estimate copper corrosion rate, we used data from a 16-year corrosion damage study of copper alloy in aqueous environments in tropical countries, conducted by the U. S. Naval Research Laboratory.[18] In these tests, samples exposed to intermittent immersion in Pacific Ocean water and complete immersion in soft-water lake water resulted in the same corrosion rate. Over 16 years the average weight loss was 5 g/m^2-a. We use this corrosion rate, in the form of a fractional alteration rate per year, for copper-matrix waste containers.

5.3 Calculated Release Rates

Release rates have been calculated for the 33 species tracked in this study. However, in this paper we shall present only selected calculated release rates.

In a parallel study, Hirschfelder *et al.*[19] showed that only a few nuclides will reach the water table and have the potential to reach the accessible environment.

Figure 3 shows the release of Cs-135 from single containers, in Ci/a, from all reprocessing wastes, as well as from LWR spent fuel.[10] The release rates of Cs-135 from reprocessed packages are generally lower than for LWR spent fuel, but the fractional release rates of several reprocessed packages are above the USNRC limit of 5×10^{-5} for Cs-135.

Figure 4 shows the release rates of plutonium species from single containers, in Ci/a, from all reprocessing wastes. The release rate of Pu is partitioned into the three longest-lived isotopes, and that partition is shown for only one waste stream, A1-8. The release rate of only Pu-242, the longest-lived isotope, is shown from the other waste packages. Because Pu is solubility limited, all release rates are low.

We now calculate the aggregate release from entire repositories, represented by the schemes in Figure 1. An equal amount of nuclides released from either scheme should result in the same dose at the point of discharge. Once radionuclides are released from waste, the buffering capacity of the rock controls the chemical form of the species, and its transport properties.

We multiply the release rates of key radionuclides from the individual waste packages by the number of waste packages, and compare the overall release rate of that species from the two schemes, LWR spent fuel (SF) versus reprocessing, for I-129, Tc-99, Np-237 and Pu isotopes. Thus the following figures are repository-wide comparisons. Figure 5 shows the release rates of I-129 from LWR spent fuel and reprocessed wastes. The peak release rate of I-129 from reprocessed wastes is approximately the same as that from LWR spent fuel, but starts earlier. For reprocessing wastes from both Schemes 2a and 2b, the releases are dominated by instant release from the zeolite waste in A3-4. The solubility-limited release from AgI from gaseous I-129 does not appear until about 80,000 years, in the form of a tail.

Figure 6 shows that for Tc-99 the peak release rate from LWR SF is higher by about a factor of 10 than the peak release rate from reprocessed wastes. However, the releases from reprocessed wastes start earlier and stay at a near constant level for a much longer time. Release from the LWR spent fuel waste container begins much later because it has a larger void volume, but the peak release rate of Tc-99 from LWR SF is higher because the alteration rate of LWR spent fuel is about two-orders of magnitude faster than the copper-matrix waste containers resulting from reprocessing.

For solubility-limited Np-237, Figure 7 shows that the release rate from LWR spent fuel is between that of Scheme 2a and Scheme 2b. Within the uncertainty of our parameter values, we can say that the release rate of Np-237 from LWR spent fuel and reprocessed wastes are equal.

Figure 8 shows the composite release rates of the plutonium isotopes from LWR spent fuel and reprocessing wastes. The combined release from LWR spent fuel is usually higher, but within a factor of 10. Within the accuracy of the parameter values, these release rates can be considered equal.

The release rates in Figures 5 through 8 assumes that all waste packages begin water contact at 1000 years, and no credit has been taken for any metallic container or the time-distributed nature of package failure.

6. Conclusions

This paper provides some of the basis for evaluating the benefits for waste disposal of partitioning and transmutation. Inventories of exotic waste packages are given. Release rates, for the wet-drip water-contact mode relevant to Yucca Mountain, have been calculated. For key radionuclides that are likely to reach the accessible environment, the release rates from reprocessed waste packages are shown to be approximately the same as the release rate from LWR spent fuel.

Several caveats are in order about the results presented here. While we use the same methodology for calculating release rates as for the potential repository at Yucca Mountain, in calculating release rates for Yucca Mountain we use well established solubilities. In this study we *assumed* that solubilities for LWR spent fuel can be used for pyro-processed hardware and copper-matrix packages, a step that has to be justified by experiments.

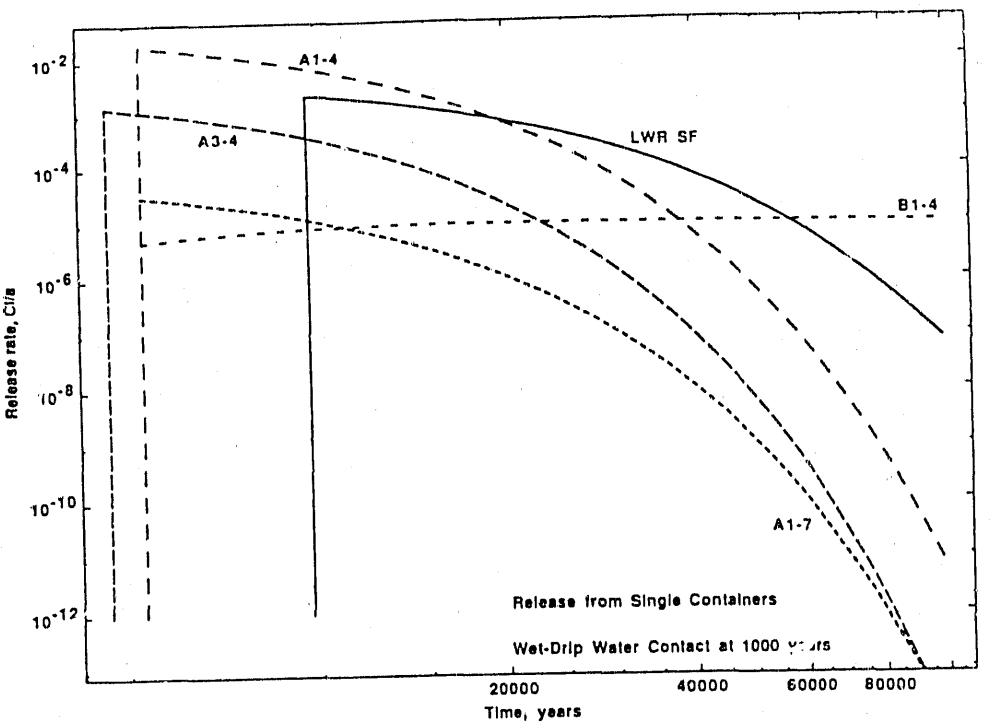

Figure 3. Release rates of Cs-135 from LWR spent fuel and reprocessed wastes

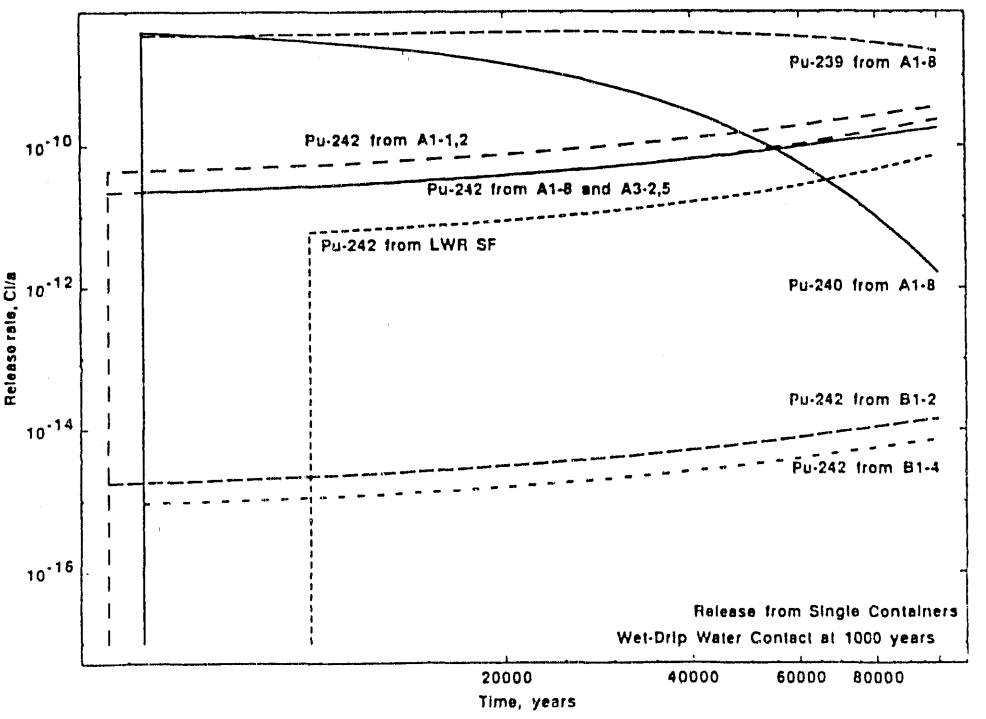

Figure 4. Release rates of plutonium from LWR spent fuel and reprocessed wastes

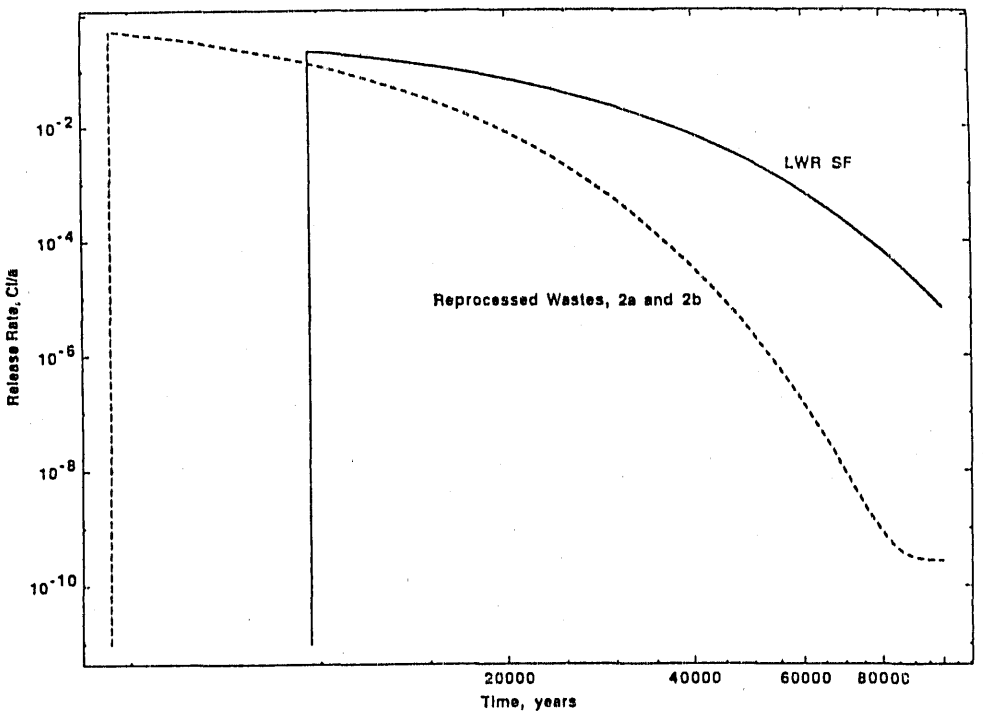

Figure 5. Release rates of I-129 from Schemes 1, 2a and 2b

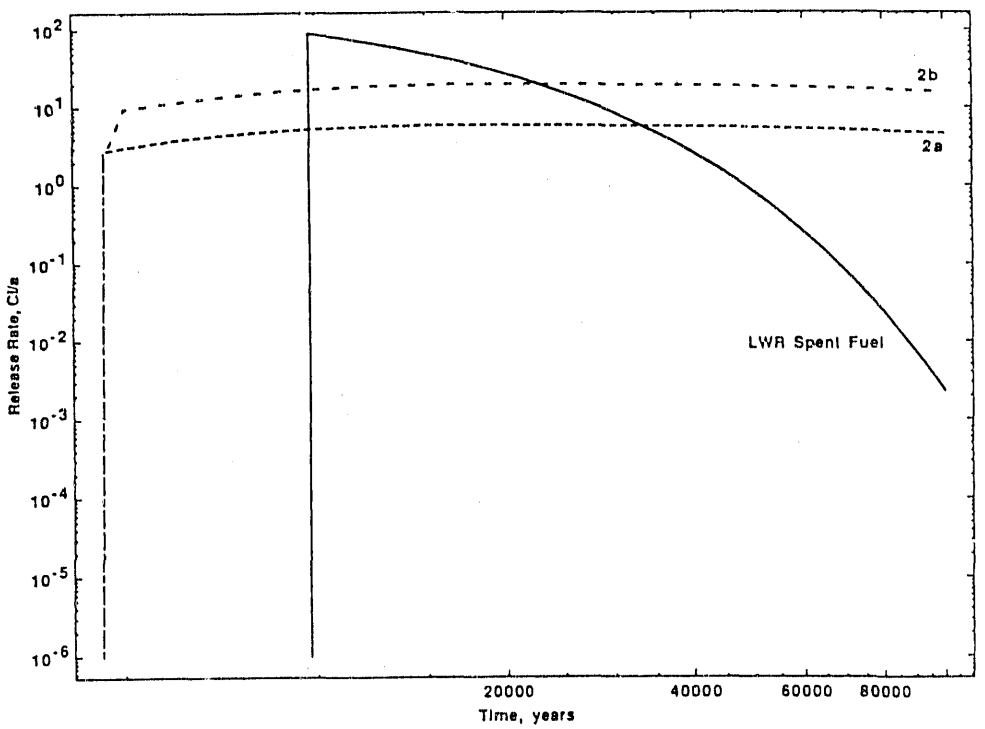

Figure 6. Release rates of Tc-99 from Schemes 1, 2a and 2b

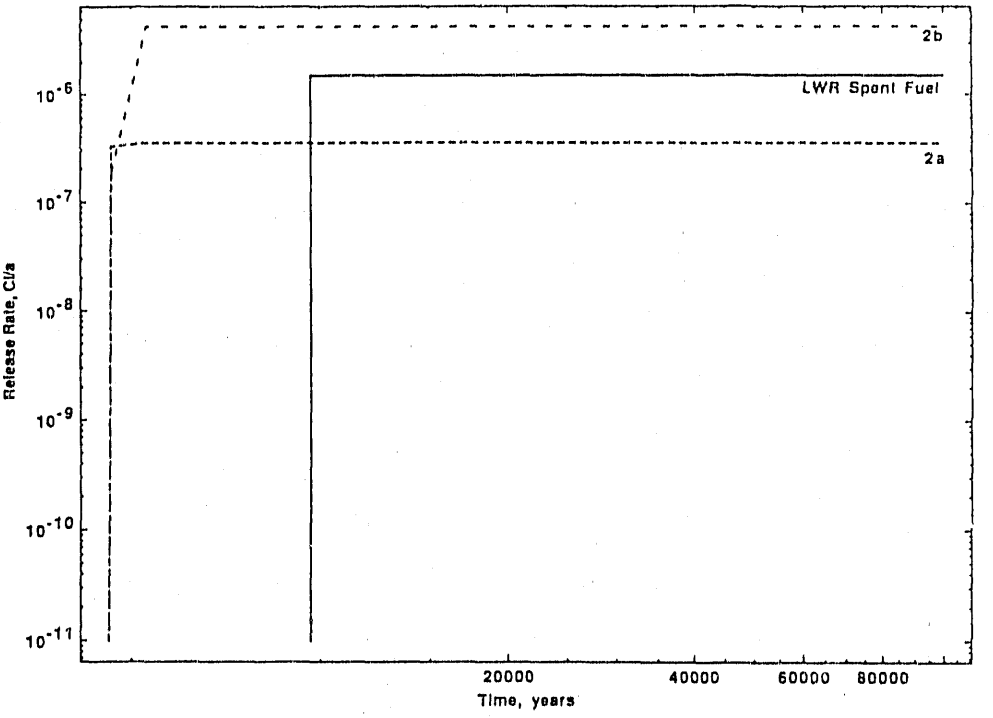

Figure 7. Release rates of Np-237 from Schemes 1, 2a and 2b

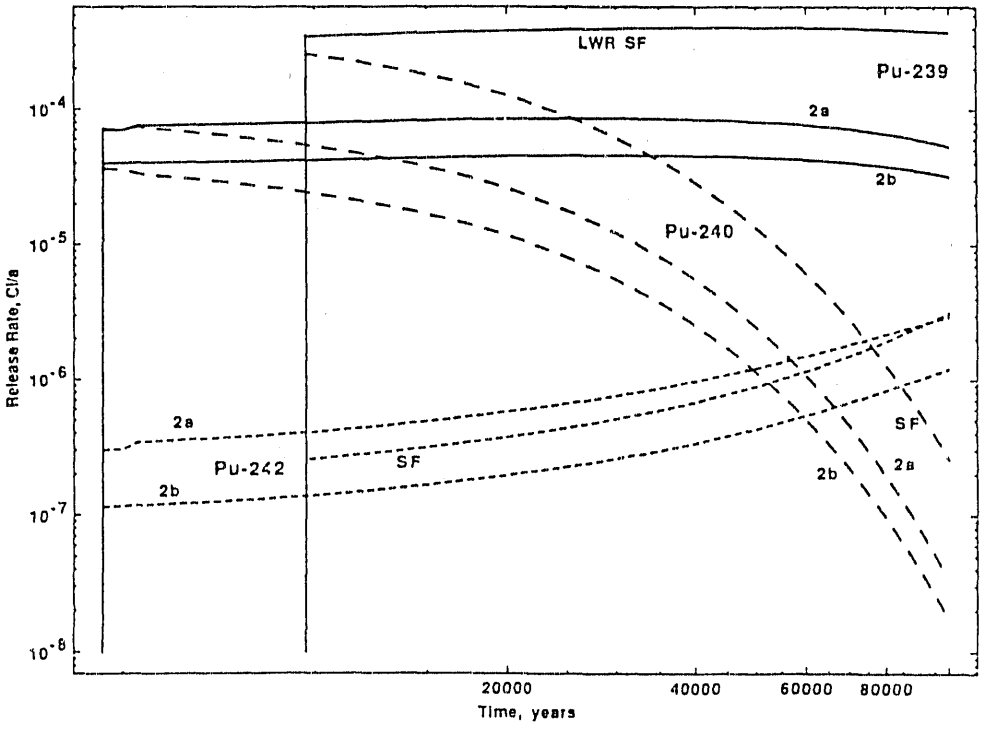

Figure 8. Release rates of Pu isotopes from Schemes 1, 2a and 2b

We also assumed, quite arbitarily, that water contact begins at 1000 years. For spent fuel disposal at Yucca Mountain, extensive thermal studies showed that re-condensation can begin at about that time. For the exotic wastes from partitioning and transmutation, we do not know whether this is true.

Given the validity of these assumptions, actinide-burning appears to offer marginal benefit for waste disposal, in terms of radionuclide releases from a geologic repository. Our conclusion collaborates similar studies in other countries.[20]

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