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An Assessment of Radiation Effects in Defense Transuranic- Waste Forms

F. P. Roberts

July 1981

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Prepared for the U.S. Department of Energy
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Pacific Northwest Laboratory
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AN ASSESSMENT OF RADIATION EFFECTS
IN DEFENSE TRANSURANIC-WASTE FORMS
2

F. P. Roberts

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1.0 SUMMARY

The actinide concentrations of the defense transuranic (TRU) wastes were analyzed and the potential effects of the radiation on the properties of the wastes after conversion to immobile forms were assessed. The study focused on the contact-handled^(a) retrievably-stored wastes.

The major components of the current inventory are defense plutonium-contaminated wastes containing various amounts of ^{241}Am . The wastes stored at Idaho National Engineering Laboratory (INEL) are typical of the wastes in this category. There is also a substantial amount of wastes contaminated with plutonium enriched in ^{238}Pu arising from the Department of Energy's (DOE) isotopic heat-source programs. Most of these wastes are stored at the Los Alamos National Laboratory (LANL) and the Savannah River Plant (SRP) sites.

Four reference wastes were selected representing a credible range of actinide activities and were used for estimating radiation doses to the final waste forms. The four are based on:

- INEL first stage sludge,
- a composite of all wastes at INEL,
- a composite of all wastes at LANL including both defense and heat-source plutonium wastes,
- a composite of all heat-source plutonium wastes at SRP free from defense plutonium.

From integrated alpha and beta-gamma doses over a 10^5 y storage period, it is concluded that:

- Accumulated beta-gamma doses will be 10^6 to 10^8 rad over a 10^5 y storage period. These doses will not significantly change physical properties of the waste form.

(a) Contact-handled TRU wastes contain only small amounts of penetrating radiation and can be handled without the special techniques required for remote-handled wastes.

- The alpha decay doses are $3\text{--}30 \times 10^{16}$ decays/cm³ (up to 3×10^{10} rad ionizing radiation from alpha particles) accumulated over a 10^5 y storage period. At these dose levels changes in microstructure and phase stability are expected to be negligible.
- Radiolytic gas generation can be substantial in waste forms containing water or organic materials. A standard test that employs doping with short-lived actinides to measure this effect is recommended.
- The effect of leachant radiolysis on the leachabilities of TRU waste forms is not sufficiently understood to rule out the need for testing with actinide-doped specimens. Further studies need to be undertaken to determine the impact of this phenomenon at the dose rates expected for TRU waste forms.

2.0 INTRODUCTION

This report is a preliminary assessment of the expected effects of self-irradiation on the durability of the solid waste forms proposed for immobilizing transuranic (TRU) wastes before their ultimate disposal. The findings are intended to provide information to be used in determining the need for testing the effects of radiation on the proposed waste forms.

This assessment was performed by the Material Characterization Center at the Pacific Northwest Laboratory. It was sponsored by DOE's office of Nuclear Waste Management as part of the TRU Waste Management Program under the coordination of the TRU Waste Systems Office of Rockwell International, Rocky Flats Plant (RFP).

Conversion of the TRU wastes to stable immobile forms before final disposal in geologic repositories is an option being considered for the permanent isolation of TRU wastes (DOE 1980b). This option involves incineration of combustible wastes, decontamination of noncombustible wastes, or other volume-reduction steps followed by fixation of the radioactive residues in a solid form. Such processing of TRU wastes reduces the volume, reduces dispersibility, and reduces waste-cannister pressurization and formation of potentially explosive gas mixtures. In addition, the solidified waste forms may serve as barriers to the re-entry of the radionuclides contained in the waste into man's environment. To act as a barrier, the waste form must be stable over the very long isolation periods required of the wastes.

TRU wastes consist of materials that are contaminated with long-lived alpha-emitting radionuclides, specifically ^{233}U and others with atomic numbers greater than 92. Generally, wastes contaminated with actinide levels less than 10 nCi/g are classified as low-level wastes. High-level wastes from reprocessing operations are not considered to be TRU wastes even though they contain substantial actinide concentrations.

The effects of radiation on high-level waste forms have been studied extensively both in the United States and in Europe during the last 10 years. The subject has recently been reviewed in detail by Weber and Roberts (1981). On the other hand, the radiation stability of TRU waste forms has received less

attention. The radiolysis behavior of TRU wastes, particularly radiolytic gas generation, has been studied. Much of this work has focused on the radiolytic decomposition of TRU combustible wastes, but work has also been done on some TRU waste forms, notably that of Bibler (1978, 1979) on concrete and Kosiewicz (1980) on bitumen.

3.0 DEFENSE TRANSURANIC WASTES

Transuranic wastes consist of materials contaminated with actinide elements. The principal sources of these materials are the processes associated with the defense program. In defense wastes, plutonium is the major actinide element present. Its isotopic composition is characterized by a very low ^{238}Pu content. Heat-source plutonium waste, by contrast, consists mostly of ^{238}Pu and represents the second major source of TRU wastes. The isotopic compositions of these two kinds of plutonium used in this study are given in Table 1.

TABLE 1. Plutonium Isotopic Compositions Found in TRU Wastes(a)

Nuclide	Half-Life, y	Weight Fraction	
		Defense Pu	Heat Source Pu
^{238}Pu	87.4	0.0003	0.802
^{239}Pu	24,390	0.939	0.159
^{240}Pu	6,540	0.057	0.0302
^{241}Pu	14.4	0.005	0.0064
^{242}Pu	3.76×10^5	0.0003	0.0013

(a) From Shefelbine (1978).

Americium-241 is always present in plutonium-contaminated wastes from the beta decay of ^{241}Pu :

The amount of americium in defense wastes is highly variable because of the differences in decay times and because many of the processes remove americium from the plutonium and, thus, enrich the wastes in americium.

The national inventory of defense TRU wastes exists in a variety of degrees of retrievability. Before 1970, wastes containers were emplaced in trenches or pits and covered with soil. Extra efforts were not made to package them for ease of future retrieval. Subsequently, it was required that all

TRU wastes with >10 nCi/g of long-lived actinides be stored in packages that could be retrieved intact and free from external surface contamination for a period of 20 years.

In addition to buried and retrievably stored wastes that can be handled without extraordinary radiation protection, there is a relatively small volume of TRU wastes containing β , γ emitting radionuclides having surface dose rates >200 mrem/h. These are referred to as remotely handled wastes. These may have surface dose rates averaging 100 rem/hr and some containers may be potentially as high as 200,000 rem/h (Shefelbine 1978). The national inventory of defense TRU wastes as of December 31, 1980 is summarized in DOE (1980a). Summaries of TRU waste inventories held at INEL, LANL, and SRP are given in Appendix A.

3.1 RADIATION CHARACTERISTICS OF DEFENSE TRU WASTES

3.1.1 Alpha Decay Doses

From the TRU waste inventories described in Appendix A, actinide compositions of four TRU waste categories were defined to estimate doses to waste forms. These four cases represent typical concentrations of plutonium and americium in final waste forms after processing and immobilization. In addition, a limiting case representing the maximum expected activity is postulated based on a proposed acceptance criterion for the Waste Isolation Pilot Plant (Irby 1980). The proposed criterion limits the total fissile content of any 0.21 m^3 (55 gal) container to 0.2 kg. The assumptions for each of the four cases are described in the following paragraphs.

Case I is a waste form prepared from the first stage sludge stored at INEL. The product is a monolith with a density of 3 g/cm^3 . The waste loading is 50 wt% (0.5 g sludge/g product) giving a final composition of 1.2×10^{-3} wt% defense plutonium and 4.53×10^{-4} wt ^{241}Am . These values assume an average concentration of 2.37×10^{-3} wt% Pu and 9.05×10^{-4} wt% Am in the sludge. This assumption is consistent with actual measured values of current production RFP sludges reported by Bryan (1981) that have a range of 4×10^{-4} to 39×10^{-4} wt% Pu and 1×10^{-4} to 32×10^{-4} wt% Am.

Case II represents a composite of all retrievable stored wastes at INEL. Again, assuming a monolithic product having a density of 3 g/cm^3 and a waste loading of 50 wt%, the waste form will have a final composition of:

- 7.40×10^{-4} wt% defense Pu
- 3.93×10^{-6} wt% heat source Pu
- 4.02×10^{-5} wt% Am

Case III is a composite representing all the TRU waste stored at LANL. Again, assuming a monolithic product with a density of 3 g/cm^3 and a waste loading of 50 wt%, the waste form will have a final composition of:

- 1.15×10^{-3} wt% defense Pu
- 2.80×10^{-4} wt% heat source Pu
- 1.35×10^{-4} wt% Am

Case IV is a composite of the heat-source plutonium wastes stored at SRP. The weights of the waste containers are not available, but the gross volume and the amounts of plutonium for the containers are known. Using these values and an estimated bulk density of the waste of 0.55 g/cm^3 , a waste form was assumed having a density of 3.0 g/cm^3 and an actinide concentration of 1.9×10^{-3} wt% heat-source plutonium.

Case V is a "worst case" upper limit for actinide concentration based on the WIPP acceptance criterion discussed above. The hypothesized waste form is concrete containing 0.2 kg of defense plutonium in a 0.21 m^3 canister filled to 90% capacity. The density of the concrete is 1.8 g/cm^3 and the actinide concentration is 0.059 wt% of defense plutonium. This case is highly atypical and it is unlikely that many canisters will have activities this high.

The cumulative alpha-decay dose (in alpha decays/ cm^3 of waste) calculated for each of the five cases for times up to 10^5 y after immobilization are presented in Figure 1. The maximum dose at 10^5 y ranged from 3×10^{16} up to 2.5×10^{17} alpha decays/ cm^3 for the typical cases. The "worst" case, Case V, had a value of 2.5×10^{18} alpha decays/ cm^3 .

FIGURE 1. Alpha Decay Doses in Immobilized TRU Waste Forms

Cases III and IV had relatively high doses at times up to ~300 y because of the decay of the short-lived ^{238}Pu ($t_{1/2} = 87.4$ y) that dominates heat-source plutonium wastes.

Much of the TRU waste is not directly convertible to an immobile solid. Some of it is combustible solids that may be incinerated. Another large fraction is contaminated equipment such as hoods, tanks and machinery that may be decontaminated and disposed as non-TRU waste. However, plutonium concentrations in the ash and decontamination residues are not likely to be more than 2 to 3 times greater than in the unprocessed wastes.

3.1.2 Beta, Gamma Doses

The principal radioactivity in contact-handled TRU wastes is, of course, alpha decay. However, the actinide nuclides do have beta-emitting daughters in their decay chains as shown in Tables 2 to 5. The decay of these daughters, as well as the gamma activity associated with some of the alpha active nuclides (notably ^{241}Am), results in a small but potentially significant β , γ dose. The calculated α , and β , γ doses expressed in rads are shown in Figure 2 for Cases I to IV. The ratio, alpha dose/ $(\beta$, $\gamma)$ dose, is highly dependent on the isotopic composition of the plutonium and the amount of ^{241}Am . Wastes containing large amounts of ^{238}Pu as in Case IV (Figure 2d) have large ratios ($>10^4$ except for very long times). Wastes containing high concentrations of ^{241}Am or ^{241}Pu have smaller ratios, e.g., Case I (Figure 2a).

3.3 TRU WASTE FORMS UNDER CONSIDERATION

A large number of waste forms have been considered for immobilization of radioactive wastes. Crisler (1980) identified and assessed some of the most prominent ones. The processes and waste forms are listed in Table 6. Other forms will undoubtedly be proposed and developed in the future.

TABLE 2. ^{238}Pu Decay Chain

Nuclide	Half-life	Principal Decay Mode	Decay Energy MeV/decay
^{238}Pu	87.4 y	α	5.61
^{234}U	2.44×10^5 y	α	4.86
^{230}Th	7.7×10^4 y	α	4.76
^{226}Ra	1600 y	α	4.76
^{222}Rn	3.82 d	α	5.49
^{218}Po	3.05 m	α (0.02% β)	6.10
^{214}Pb	26.8 m	β	0.411
^{214}Bi	19.8 m	β (0.04% α)	2.34
^{214}Po	1.64×10^{-4} s	α	7.61
^{210}Pb	22.34 y	β ($10^{-5}\%$ α)	6.98×10^{-3}
^{210}Bi	5.01 d	β	0.443
^{206}Po	138.4 d	α	5.30
^{206}Pb	Stable	--	--

TABLE 3. ^{239}Pu Decay Chain

Nuclide	Half-life	Principal Decay Mode	Decay Energy MeV/decay
^{239}Pu	2.439×10^4 y	α	5.24
^{235}U	7.04×10^8 y	α	4.69
^{231}Th	25.5 h	β	0.133
^{231}Pa	3.25×10^4 y	α	5.13
^{227}Ac	21.8 y	β (1% α)	0.086
^{227}Th	18.7 d	α	5.82
^{223}Ra	11.4 d	α	5.86
^{219}Rn	3.96 s	α	6.82
^{215}Po	1.8×10^{-3} s	α	7.39
^{211}Pb	36.1 m	β	0.566
^{211}Bi	2.1 m	α	6.57
^{207}Tl	4.8 m	β	0.509
^{207}Pb	Stable	--	--

TABLE 4. ^{240}Pu Decay Chain

Nuclide	Half-life	Principal Decay Mode	Decay Energy MeV/decay
^{240}Pu	6540 y	α	5.27
^{236}U	2.342×10^7	α	4.76
^{232}Th	1.40×10^{10} y	α	4.09
^{228}Rd	6.7 y	β	0.013
^{228}Ac	6.13 h	β	0.900
^{228}Th	1.91 y	α	5.53
^{224}Ra	3.64 d	α	5.78
^{220}Rn	53 s	α	6.41
^{216}Po	0.15s	α	6.92
^{212}Pb	10.6 h	β	0.24
^{212}Bi	61 m	35% α 65% β	2.17 0.77
^{208}Tl	3.1 m	β	3.93
^{208}Pb	Stable	--	--

TABLE 5. ^{241}Pu Decay Chain

Nuclide	Half-life	Principal Decay Mode	Decay Energy MeV/decay
^{241}Pu	14.4	β	7.0×10^{-3}
^{241}Am	433 y	α (60 Kev γ in 36% of decays)	5.64
^{237}Np	2.14×10^6 y	α	4.97
^{233}Pa	27 d	β	0.228
^{233}U	1.58×10^5 y	α	4.91
^{229}Th	7.34×10^3 y	α	5.12
^{225}Ra	14.8 d	β	0.111
^{225}Ac	10.0 d	α	5.80
^{221}Fr	4.8 m	α	6.28
^{217}At	3.2×10^{-4} s	α	7.08
^{213}Bi	46 m	β	1.04
^{213}Po	4×10^{-9} s	α	8.40
^{209}Pb	3.3 h	β	0.194
^{209}Bi	Stable	--	--

Figure 2a

Figure 2b

Figure 2c

Figure 2d

FIGURE 2. Integrated Alpha and Beta-Gamma Doses for Immobilized TRU Waste Forms

TABLE 6. Listing of Generic Process Types and Waste Forms for TRU Waste Immobilization^(a)

<u>Process</u>	<u>Waste Forms</u>
Hydrocarbon encapsulant	Bitumen
Cementitious encapsulant	FUETAP ^(b) CPWF ^(c)
Vitrification	Borosilicate glass High-Silicon glass
Fusion-cast ceramics	Basalt Glass Ceramics
Crystalline or mineral ceramics	"SYNROC B"
Cermets	Cermet

(a) Adapted from Crisler (1980).

(b) An acronym for Formed Under Elevated Temperature and Pressure

(c) Cold pressed waste forms.

4.0 EXPECTED RADIATION DAMAGE IN IMMOBILIZED TRU WASTES

4.1 GENERAL CONCERNS

The concerns about the radiation stability of the immobilized waste forms are both short-term and long-term. The short-term concern is the period between the conversion of the waste to its ultimate form and the final closure of the repository. During this period, the waste is packaged, transported to and emplaced in the repository. The waste packages will be under surveillance during this period and may be required to be retrievable for up to 50 years after the end of the repository operational period. One of the major potential effects of self-irradiation is radiolytic gas production with attendant risks from container pressurization and the build-up of flammable gas mixtures.

The long-term concerns are related to the capability of the waste form to retard the movement of the radionuclides back to the biosphere. This period is indefinite, but, in the case of TRU wastes, may be $>10^5$ y. During this time, irradiation-induced changes that affect the chemical durability of the waste form are of concern. These may include chemical changes leading to the formation of soluble phases or structural changes that increase surface area, e.g., cracking, pulverization, etc. In addition, radiolysis of ground waters intruding the system can accelerate leach rates.

4.2 EFFECTS OF RADIATION ON WASTE FORM PROPERTIES

As was shown in Figure 2, alpha decay is the dominant source of radiation in contact-handled TRU wastes. In alpha decay, two projectiles are formed--alpha particles and recoil nuclei. The alpha particles have energies mostly in the range of 5-6 MeV. A few are produced with energies as high as 8.4 MeV (see Tables 3 through 6). Typical recoil nuclei energies are near 100 keV. The ranges of the projectiles are relatively short and thus exhibit high linear-energy transfer.

Interaction of alpha particles with matter is primarily through ionization processes, but a significant number of atomic displacements are formed by elastic collisions. The recoil nuclei dissipate nearly all their energy by elastic collisions producing atomic displacements. Much of the radiation damage may be caused by atomic displacements, but ionization effects such as covalent bond

rupture, valence change and radiolysis may be significant particularly in waste forms containing water or organic material. Alpha decay may produce a number of measurable changes in waste solids including:

- density changes
- structural and microstructural changes
- leach rate changes
- mechanical property changes
- stored energy build-up
- helium and radiolytic gas generation.

The potential effects of these radiation-induced changes on the properties of TRU waste forms are discussed in the following sections.

4.2.1 Density Changes

Density changes can produce internal stresses in waste forms, particularly forms having more than one phase. If expansions occur, there is potential for inducing stresses in the canisters. These concerns have led to experimental studies on the effect of irradiation on waste form densities. Much of the available data is based on actinide-doped materials and is, therefore, applicable to TRU wastes. These data have been recently reviewed (Weber and Roberts 1981).

The density change as a function of dose tends to follow exponential behavior as described by the following equation:

$$\Delta\rho/\rho = a [1-\exp(-bn)] \quad (1)$$

where ρ is the density of the waste form and n is the dose. The constants a and b are material-dependent parameters and can be derived by regression analysis of experimental data.

The behavior of a relatively large number of waste glasses indicates the maximum density change is less than 1.5% for vitreous waste forms (Roberts, Turcotte and Weber 1981; Malow, Marples and Sombret 1980; Bibler and Kelley 1978; Hall et al. 1976). Less extensive data is available on ceramic waste forms, but the behavior of supercalcine-SPC-2 (Rusin, Gray and Wald 1979), celsian glass ceramic (Malow, Marples and Sombret 1980) and SYNROC-B (Reeve,

Levine and Woolfrey 1981) have been studied. Actinide doping was used for the tests of supercalcine and the glass ceramic; fast neutron irradiation was used in the study of SYNROC-B and the neutron doses were converted to equivalent alpha decays/cm³ in the review by Weber and Roberts (1981).

The data from these studies were obtained at the relatively large doses expected for high-level wastes, but by using Equation 1 it is possible to extrapolate the density changes back to the dose-levels expected for TRU wastes. Absolute density changes were calculated for four of the generic TRU waste forms; SYNROC B, supercalcine, borosilicate glass (using data for PNL-72-68a glass) and celsian glass ceramic. The values are shown in Figure 3 both as a function of alpha-decay dose and equivalent age of the immobilized waste forms using as the reference wastes Cases I-IV (See Section 3.1). Although this approach has some uncertainty because of the large extrapolation, density changes are likely to be <0.5%.

4.2.2 Structural and Microstructural Changes

Structural or microstructural changes caused by radiation are manifested in the macro-properties of the solids such as density, leach rate and mechanical strength. Only in a relatively few studies have crystallinity changes and devitrification behavior after irradiation been reported.

In one notable experimental study, Weber et al. (1979) observed amorphization of two crystalline phases in a ²⁴⁴Cm-doped high-level waste glass partially devitrified by thermal treatment. One phase with an apatite structure of Ca₃Gd₇(SiO₄)₅(PO₄)O₂ became amorphous at a dose of 2×10^{18} alpha decays/cm³. A second major phase, gadolinium titanate, which was originally reported by Weber et al. (1979) to be radiation stable, became x-ray amorphous at a much higher dose ($>6 \times 10^{18}$ alpha decays/cm³) (Weber and Roberts 1981). (In that same study, observations showed no enhanced devitrification of any of the glasses as the result of alpha decay.)

Similar crystalline phase behavior has also been reported in supercalcine-SPC-2 (Rusin, Gray and Wald 1979) and celsian glass ceramic (Weber and Roberts 1981). In both these materials, the transformation of the crystalline phases to x-ray amorphous phases became perceptible only at dose levels greater than 10^{18} alpha decays/cm³.

FIGURE 3. Radiation-Induced Density Changes in TRU Waste Forms

There is no experimental evidence presently available that suggests amorphization of crystalline phases will take place at the dose levels expected in TRU waste forms.

4.2.3 Leach Behavior

The leachability of radioactive waste forms can be affected by radiation damage to the solid itself and by the formation of radiolytic degradation products at the waste form surfaces and in the solution with which it is in contact.

None of the experimental studies reported to date on glass waste forms show evidence of significant acceleration of leaching due to radiation damage. No studies on ceramic forms other than concrete have been reported to date (Weber and Roberts 1981). In a comparative study, Malow, Marples and Sombret (1980) measured leach rates on eight European waste glasses doped with 2.5 wt% ^{238}Pu . Only one glass formulation exhibited a leach-rate increase by more than a factor of two. The leach rate increased from $23 \text{ g/m}^{-2} \cdot \text{d}^{-1}$ to $69 \text{ g/m}^{-2} \cdot \text{d}^{-1}$ after irradiation to 3.3×10^{18} alpha decays/cm³ at 50°C.

Neither beta nor gamma irradiation have been found to produce permanent changes in borosilicate glasses that increase leachability. Wastes containing actual fission products showed no increase in leach rates with β , γ doses up to 1.2×10^{11} rads in one study (Jacquet-Francillon, Bonniand and Sombret 1978) and 4.5×10^{11} rads in another study (Mendel 1977). Also external gamma irradiation up to 10^{11} rad had no effect on the leach rates of borosilicate glasses (Johnson and Marples 1979) nor on concrete at a dose of 10^{10} rad (Stone 1979). These doses are at least 100 fold greater than the integrated β , γ doses expected in TRU wastes at 10^5 y (See Figure 2) and therefore it is not anticipated that internal β , γ irradiation will change leach rates.

Although radiation damage to the waste forms does not seem to play a significant role in leachabilities, radiolysis of the leachant may have an effect. In a study by McVay and Pederson (1981), both borosilicate glass and supercal-cine showed increased leach rates in a gamma field of 2.4×10^6 R/hr. Nitric acid, as well as other radiolysis products, was formed and presumably contributed to the increased leachability (McVay, Weber and Pederson 1981). The tests were carried out at much higher dose rates (as much as 10^4) than expected at

the surfaces of TRU waste packages that are in the range of ~200 mR/hr (Shefelbine 1978). Additional studies may be needed to assess this effect at the lower dose rates encountered with TRU waste forms.

4.2.4 Mechanical Properties

The strength of TRU waste forms is a potential concern from the standpoint of the effect of mechanical shock on dispersability (formation of fine particles) and increased leachability resulting from increased surface areas. Impacts producing fine particles and cracking within the waste canister are most likely to occur during the transportation of wastes to the repository. Since transportation is early in the life of the waste form, the radiation doses will be extremely small and radiation-induced changes in the mechanical properties are highly implausible and may be assumed to be negligible during this phase.

4.2.5 Stored Energy Buildup

The thermal annealing of radiation damage in glass or ceramic waste forms results in the release of energy as heat, a phenomenon known as stored-energy release. Stored-energy behaviors of a relatively large number of potential waste forms have been surveyed recently (Weber and Roberts 1981). The heat energy released at saturation damage ranges from 83 to 125 J/g for borosilicate waste glasses developed in the U.S.A. (Roberts, Jenks and Bopp 1976; Roberts, Turcotte and Weber 1981). Scheffler and Riege (1977) observed values as high as 347 J/g in German developed waste glasses. Measurements have also been made on supercalcine (Rusin, Gray and Wald 1979) and celsian glass ceramic (Malow, Marples and Sombret 1980). The stored-energy releases are so small and occur gradually over such wide temperature range that any hazard is considered negligible.

4.2.6 Helium and Radiolytic Gas Formation

The pressurizing of TRU waste canisters and the potential release of flammable or explosive gas mixtures need to be considered, particularly during the operational phase of the repository. The formation of helium from alpha decay and the radiolytic generation of by-products from the decomposition of water or organic materials in the waste form are two possible sources of gas that might threaten the integrity of a TRU waste package.

Pressurization of waste containers from helium buildup is relatively minor, even for the worst case postulated in Section 3.1. In this case, the container is a 0.023 m^3 canister holding TRU waste with 0.2 kg of weapons plutonium with a void space of 0.21 m^3 . The total helium at the end of 10^5 y is 0.8 moles. The pressure increase at 45°C is only 100 kPa. (This increase assumes all the helium diffuses into the void space.)

Pressurization resulting from the generation of radiolytic gases is potentially more severe than from helium buildup. Concretes and bitumen are two waste forms that are sensitive to radiolysis.

Alpha radiolysis of bitumen produces significant amounts of hydrogen. Kosiewicz (1980) reported that the initial value of $G_{\text{gas}}^{(a)}$ at low radiation doses lies in the range of 0.8 to 1.7 gas molecules/100 eV energy deposition. The value is dose dependent and approaches a value in the range of 0.5 to 0.7 at higher doses. In this same study, hydrogen was found to be the major gas evolved amounting to 97 mol%.

Concretes contain substantial amounts of water that can undergo radiolysis to form hydrogen and oxygen. This behavior has been examined in detail at SRL (Bibler 1978, 1979). In tests performed with ^{244}Cm -doped concretes, the G_{gas} value was independent of dose at least up to the maximum pressure of the tests (1380 kPa). The G_{gas} values varied with the concrete composition over a range of 0.03 to 0.61. The hydrogen evolution could be substantially reduced by adding nitrate or nitrite salts to the mix.

(a) G_{gas} is defined as the total number of gas molecules formed per 100 eV of energy deposited in the waste matrix.

Hydrogen and oxygen are also produced by gamma radiolysis of concretes. Unlike alpha radiolysis, a steady state pressure is attained at about 350 kPa (Bibler 1978). In gamma irradiation experiments carried out at Karlsruhe, Mockel and Koster (1980) also found hydrogen and oxygen as radiolytically generated gas in concrete containing simulated low and intermediate-level wastes. Specimens of pure cement, however, produced only hydrogen.

Canister-pressurization rates were calculated for the TRU waste compositions postulated in Section 3.1.1. The assumptions used were for a 0.21 m^3 canister with a void space of 0.021 m^3 . The waste form was concrete having a density of 1.8 g/cm^3 . A G_{gas} value of 0.3 was used. The calculated pressure increases are shown in Figure 4. Calculated pressure increases at 100 y ranged from 250 to 23,000 kPa. The high value was for the SRP heat source waste Case IV. At 10^5 y , the pressure increases for all compositions were in the range of 10^4 – 10^5 kPa .

FIGURE 4. Calculated Canister Pressurization from Radiolytic Gas Generation in Concrete TRU Waste Forms

5.0 CONCLUSIONS

In view of the relatively low radiation doses expected for immobilized TRU waste forms and in light of existing information on radiation effects in solids, it can be concluded that radiation impacts on the long-term stabilities of forms not containing water or organic compounds will be minor. Radiation-induced structural changes that may potentially reduce chemical durability or modify desirable mechanical properties of proposed waste forms are small at doses of 10^{17} alpha decays/cm³, and the times required to accumulate these levels are very long as was shown in Figure 1. Because of the decay characteristics of defense plutonium the activity levels in typical TRU wastes are well below 100 nCi/g after 10^5 y. (This is not true for the special case of heat-source plutonium because of the in-growth of ²³⁴U and its daughters.) Testing for structural changes using the accelerated method proposed by the Materials Characterization Center for defense and commercial high-level waste forms (Roberts, Turcotte, and Weber 1981) does not appear to be warranted unless actinide concentrations much greater than projected in this report are to be incorporated in the immobilized wastes.

The radiolytic-generation of oxygen and hydrogen from alpha decay may pose potentially serious problems for waste forms containing significant amounts of water such as concretes and organic matrix waste forms such as bitumen both from the standpoint of canister pressurization and the development of flammable or explosive gas mixtures. Transuranic waste forms that are subject to the gas generation problem need to be tested. A standardized test should be designed to measure the G_{gas} value under the range of conditions anticipated during the operational phase of the waste repository. The test variables should include pressure, temperature and dose rate.

Although radiation-induced structural changes in the waste forms may not significantly alter leachabilities, there is a potential for enhanced leaching caused by radiolysis of the leachant. The ionization doses to the leachant will be quite low because of the low β , γ activities in the wastes and the self-absorption of the alpha particles by the waste form. The effects at such low doses and dose rates should be small but need to be investigated further. In any case, leach testing of TRU waste forms should be performed with test

specimens having plutonium and americium concentrations at levels expected for the actual waste forms in order to include the radiolysis effects in the test.

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APPENDIX A

TRANSURANIC WASTE INVENTORIES

This appendix presents the relevant information used to derive the reference wastes used in this report. The reference wastes are based on actual wastes stored at INEL, LANL and SRP. The wastes at these sites were thought to represent an adequate range of compositions for the purposes of the study. The inventory of TRU wastes at Hanford is the second largest at all the DOE storage sites (DOE 1979). Its composition is believed to be similar to the INEL inventory and was not included in deriving the reference wastes.

A.1 INEL INVENTORY

Wastes stored at INEL are generated predominately at RFP but smaller amounts come from Mound Laboratory, Bettis Atomic Power Laboratory, Argonne National Laboratory, and other sites. As of December 1978, the INEL site contained $3 \times 10^4 \text{ m}^3$ of waste weighing $1.66 \times 10^7 \text{ kg}$. The actinide content consisted of 229 kg plutonium and 13.2 kg americium. The bulk of the plutonium is defense grade (Shefelbine 1978).

There are 82 categories of waste at the INEL TRU storage area. The actinide concentration varies widely among the categories. Table A.1 lists the gross weight, the plutonium concentration and the fraction of the total plutonium inventory in the waste category ranked by plutonium content. The top 14 categories contain 85% of the total plutonium. The highest plutonium concentration is 0.18 wt% in the ash heels. The average concentrations of the entire inventory is $1.4 \times 10^{-3} \text{ wt\%}$ plutonium and $8 \times 10^{-5} \text{ wt\% Am.}^{(a)}$

Greater than 90% of the total americium inventory is in the first stage sludge, which has a concentration of $9 \times 10^{-4} \text{ wt\% Am.}^{(a)}$

(a) These percentages are based on known gross weights that include container and container liner weights. Actual plutonium and americium concentrations may be 1.25 times the value stated, assuming a weight of 35 kg per 210L drum and polyethylene liner and 300 kg for the fiberglass reinforced plywood boxes used for RFP wastes.

TABLE A.1 Composition of TRU Waste at INEL Site(a)

Rank	Description	Gross Weight, kg	Pu, Wt (b)	% of Total Pu in Inventory
1	First stage sludge	1.35×10^6	2.17×10^{-3}	12.8
2	Metal scrap	3.91×10^6	0.739×10^{-3}	12.6
3	Composition unknown	1.84×10^6	1.36×10^{-3}	10.9
4	Ash heels	9.74×10^3	182×10^{-3}	7.8
5	Cement insulation and filter media	7.09×10^4	24.9×10^{-3}	7.7
6	Metal, leached	8.62×10^4	17.1×10^{-3}	6.5
7	Leaded rubber gloves and aprons	8.89×10^4	16.5×10^{-3}	6.4
8	Raschig rings	1.35×10^5	9.19×10^{-3}	5.41
9	Graphite	1.00×10^5	7.40×10^{-3}	3.2
10	Dry paper and rags	2.13×10^6	0.311×10^{-3}	2.90
11	Insulation and filter media	8.89×10^4	7.31×10^{-3}	2.8
12	CWS filters	4.02×10^5	1.38×10^{-3}	2.4
13	Leached and cemented resin	2.48×10^4	20.0×10^{-3}	2.2
14	Tantalum	2.48×10^4	13.2×10^{-3}	1.7
	All remaining categories	7.40×10^6	2.06×10^{-3}	14.7

(a) Data estimated from a more detailed listing of items stored between September 1971 through December 1978 obtained from the TRU waste program office.

(b) Not corrected for container weight.

A.2 DESCRIPTION OF LANL INVENTORY

There was 1600 m^3 of TRU waste in retrievable storage at LANL as of January 1, 1978.^(a) The wastes had a gross weight of 1.24×10^6 kg and a total actinide content of 44.2 kg including 18.4 kg uranium (Kosiewicz 1979); Table A.2 lists the principle TRU radionuclides present (a number of other

(a) The waste volume has increased substantially since then because of the addition of large amounts of decontamination and decommissioning wastes.

TABLE A.2. Amounts of Transuranic Nuclides in LANL TRU Wastes

<u>Nuclide</u>	<u>Total Weight, kg</u>
^{237}Np	3
^{238}Pu	4,832
^{239}Pu	18,735
^{241}Am	2,195

nuclides are present in insignificant amounts). The waste categories, gross weights, weight percent actinide composition and fraction of the total actinides in the inventory, as analyzed by Kosiewicz et al. (1979), are listed in Table A.3. The ranking is by fraction of the total actinides in the inventory.

The major category, containing 40% of the total actinides, is the leached process residues. This waste, which is an appropriate candidate for immobilization, contains little heat-source plutonium or excess americium but the defense plutonium concentration is nearly 0.1 wt%. The average actinide concentrations for the entire inventory are:

defense plutonium	- 0.0023 wt%
heat-source plutonium	- 0.00056 wt%
americium-241	- 0.00027 wt%

A.3 DESCRIPTION OF SRP INVENTORY

The retrievably stored TRU wastes at SRP have three major sources; SRP, Mound Facility and LANL. The waste from SRP operations are mainly plutonium-contaminated materials generated in the plutonium conversion and fabrication facilities at the plant site. The actinide content is almost entirely defense plutonium and americium. Other SRP-generated wastes arise from special production campaigns, heat-source fabrication and research and development at the Savannah River Laboratory. These activities have resulted in wastes contaminated with heat-source plutonium (>80% ^{238}Pu), ^{244}Cm , ^{252}Cf and ^{233}U (DOE 1979).

TABLE A.3 Composition of TRU Wastes at LANL (a)

Ranking	Description	Gross Weight, kg	(b) Actinide Content, wt%			% of Total Actinides in Inventory
			(c) Pu _W	(c) Pu _H	²⁴¹ Am	
1	Leached process residues	18,540	0.090	--	--	40.8
2	Unclassified noncombustibles	122,410	0.014	0.091	4×10^{-5}	15.2
3	Cement paste	577,800	0.00014	3×10^{-7}	0.0004	11.5
4	Metal crucibles scrap and dies	14,700	0.0191	0.0023	--	7.2
5	Mixed combustibles and non-combustibles	49,400	0.0053	0.032	--	5.9
6	Other combustibles	26,350	0.010	0.0001	--	5.0
7	Mixed cellulose	27,450	0.010	0.0030	--	4.0
8	Mixed paper, plastic, rubber, etc.	64,500	0.0047	0.0093	--	3.5
9	Other scrap metals	34,100	0.0043	0.0082	--	3.1
10	Equipment	166,600	0.00035	0.000035	--	2.1
	All others	278,200	6.6×10^{-5}	0.00076	--	1.7

(a) adapted from Kosiewicz et al. (1979)

(b) The actinide wt% is based on the estimated net weight of the waste category, i.e., the gross weight less the container weight.

(c) Pu_W is defense plutonium and Pu_H is heat-source plutonium.

The wastes from Mound Facility and LANL are predominantly material contaminated with heat-source plutonium.

In addition, there is a substantial quantity of contaminated soil and debris resulting from the cleanup of two military airplane crash sites. The contamination in this case is entirely defense plutonium.

As of December 31, 1977, the entire inventory of retrievably-stored TRU wastes at SRP had a volume of 1700 m³ and contained 287,000 Ci of all actinides (DOE 1979). The projected inventory for 1990 (DOE 1979) is 3400 m³ containing 375,000 Ci of all actinides. A breakdown of the weights, volumes and activities of the individual actinides in the 1990 projection is shown in Table A.4.

The 20.5 kg heat-source plutonium represents a special category of TRU wastes because of its relatively high specific activity. Fifteen kg of the heat source plutonium originated at Mound Facility. Only about 10% of this is in the form of combustible wastes. The rest is metal, filters, glassware, resin, slag, oxide ash and equipment including large items such as hoods, machinery and tanks. Fifty-three percent of the heat-source plutonium originating at LASL and stored at SRP is in the form of combustible waste. The remainder consists of crucibles, discarded equipment, glass, graphite residues and sweepings.

TABLE A.4. Projected Retrievably-Stored SRP TRU Waste Inventory Through the Year 1990.

<u>Radionuclide</u>	<u>Weight of Nuclide, kg</u>	<u>Activity, Ci</u>	<u>Volume of Waste, m³</u>
²³⁷ Np	4.80	3.4	42
²³⁸ Pu	20.5	354,000	1,240
²³⁹ Pu	69.5	4,240	1,560
²⁴⁴ Cm	0.2	17,100	400
²⁵² Cf	--	0.3	200

(a) From DOE 1979.

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