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Comparative Assessment of TRU Waste Forms and Processes

Volume I: Waste Form and Process Evaluations

September 1982

Prepared for the U.S. Department of Energy
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Pacific Northwest Laboratory
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COMPARATIVE ASSESSMENT OF TRU WASTE FORMS AND PROCESSES

VOLUME I: WASTE FORM AND PROCESS EVALUATIONS

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SUMMARY

This study provides an assessment of seven waste forms and eight processes for immobilizing transuranic (TRU) wastes. Developing the assessment involved 1) the preparation and characterization of TRU-containing waste forms, and 2) the preparation of preconceptual process designs and their evaluation in terms of risks and costs. The waste forms considered in this study are:

- cast cement
- cold-pressed cement
- FUETAP ("formed under elevated temperature and pressure") cement
- borosilicate glass
- aluminosilicate glass
- basalt glass-ceramic
- cold-pressed and sintered silicate ceramic.

The waste-immobilization processes considered are:

- in-can glass melting
- joule-heated glass melting
- glass marble forming
- cement casting
- cement cold-pressing
- FUETAP cement processing
- ceramic cold-pressing and sintering
- basalt glass-ceramic processing.

Quantitative criteria by which to judge the acceptability of a waste form have not been generally established. However, qualitative criteria have been established by the Waste Isolation Pilot Plant for defense transuranic wastes, and draft criteria for commercial wastes have been published by the Nuclear Regulatory Commission. Therefore, the comparisons of waste forms in this report are generally not made with respect to quantitative criteria, but are made, instead, with respect to the relative behavior of each of the waste forms considered in the study. The waste forms compared were prepared from the same waste composition. Properties considered in the study included gas generation, chemical durability, mechanical strength, thermal stability, and radiation stability.

The ceramic products demonstrated the best thermal, chemical, and mechanical properties, except for plutonium release during leaching. The two glass products and the ceramic products had similar properties. The cement products generally had poorer properties than the other forms, except for plutonium release during leaching. Calculations of the fraction of plutonium released from full-scale products under static leaching conditions indicated that the waste forms met the proposed NRC release rate limit of 1 part in 10^5 per year in most test conditions.

From the viewpoint of processing costs, the cast-cement process had the lowest cost, followed closely by the cold-pressed and FUETAP cement processes. Joule-heated glass melting had the lower cost of the glass processes. In-can melting in a high-quality canister had the

highest cost, and cold-pressed and sintered ceramic the second highest. Labor costs and the costs of canisters used for in-can melting were identified as major cost differentiating items among the processes. The major contributor to costs of disposing of TRU wastes in a defense waste repository is waste processing costs. Repository costs could become the dominant cost for disposing of TRU wastes in a commercial repository. Based on the assumptions and evaluations in this study, it is recommended that cast and FUETAP cement and borosilicate glass waste-form systems be considered for further development and application in transuranic-waste immobilization.

Additionally, it is recommended that 1) further development of cast cement be directed to reducing water content, which would eliminate concerns about radiolysis and free water and improve thermal stability; 2) a full-scale leach test be conducted to verify calculations made from laboratory-scale leach tests; and 3) future process development emphasize methods that can operate with limited labor requirements and avoid expensive containers.

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INTRODUCTION

Transuranic (TRU) wastes are generated from defense activities related to production and processing of plutonium for weapons. TRU wastes will also be generated in the commercial nuclear power fuel cycle. Current policy requires that these wastes be isolated from the biosphere for long periods of time. The Waste Isolation Pilot Plant (WIPP) is planned as a demonstration and research and development facility for defense TRU waste disposal. Requirements for the waste to be shipped to the WIPP have been published (Irby 1980). Battelle Memorial Institute's Office of Waste Isolation (OWI) under U.S. Department of Energy (DOE) funding is developing plans for commercial waste repositories to handle both TRU and high-level wastes (HLW). The Nuclear Regulatory Commission (NRC) has also published draft criteria (10 CFR 60) for the disposal of TRU and high-level wastes.

Provided in this volume is a summary of a comprehensive comparative assessment of seven TRU waste forms and eight waste processes performed at Pacific Northwest Laboratory (PNL). The assessment was based on cost and risk considerations. Additional details of the assessment are contained in companion Volume II. Volume II contains additional information on the fabrication, test methods, and characterization data for the waste forms considered; and detailed process descriptions and additional cost data.

This comparative assessment study began in 1979 with a request from the Transuranic Waste Systems Office (TWSO) of the U.S. Department of Energy to provide comparative data on promising systems for immobilizing TRU wastes. The first phase of the study defined the following:

- basic criteria to be used for the selection of waste-processing systems
- criteria by which waste-processing systems would be compared
- the waste composition to be used for fabrication of the waste forms
- the size and location of the reference facility to process the waste.

The second phase of the study included:

- laboratory-scale fabrication and characterization of the waste forms
- preparation of process descriptions
- estimation of system costs
- evaluation of risks associated with the process.

The third phase of the study is the assembly of the data and the comparison of the systems, which is the purpose of this report.

PREVIOUS STUDIES

A wide variety of comparative studies, assessments, and compilations of data have been published during the past few years. Many of these studies have been directed toward HLW (ERDA 1976; E. R. Johnson 1980; Hench 1979; Mendel et al. 1981; Ross, Rusin and McElroy 1979; Ross et al. 1979; NRC 1979; Rusin 1980; Schulz et al. 1980; Stone, Goforth, and Smith 1979; Wald et al. 1980). One study was directed toward TRU wastes and processes and consisted of a review of nine potential waste forms and an assessment of their flexibility to allow incorporation of the wide variety of TRU wastes currently generated (Crisler 1980). Another study to assess the ability of various TRU waste forms to meet the proposed Nuclear Regulatory Commission (NRC) 10 CFR 60 criteria for commercial-waste disposal has been published by Brookhaven National Laboratory (Bida and MacKenzie 1982).

STUDY EMPHASIS

This study considers both risk and cost factors. These evaluation criteria were organized into a hierarchy so that relationships could be established and relative importance quantified. The hierarchy that was established during the first phases of the program is shown in Figure 1. In the risk side of the hierarchy, both long-term and short-term risks were considered. For the long term, the primary factors are the leaching and transport of radionuclides. Gas generation was also considered as an important characteristic for some TRU wastes in the WIPP report (Irby 1980). Study members also felt that the ability to provide quality assurance to the whole package could help quantify and reduce the long-term risk.

Short-term risks arise in processing and handling, interim storage, transportation of the waste, and placement in the repository. A major factor during processing is the occupational exposure to radiation during both routine and maintenance operations. Processing operations will expose workers to normal industrial hazards such as fire, explosions, and

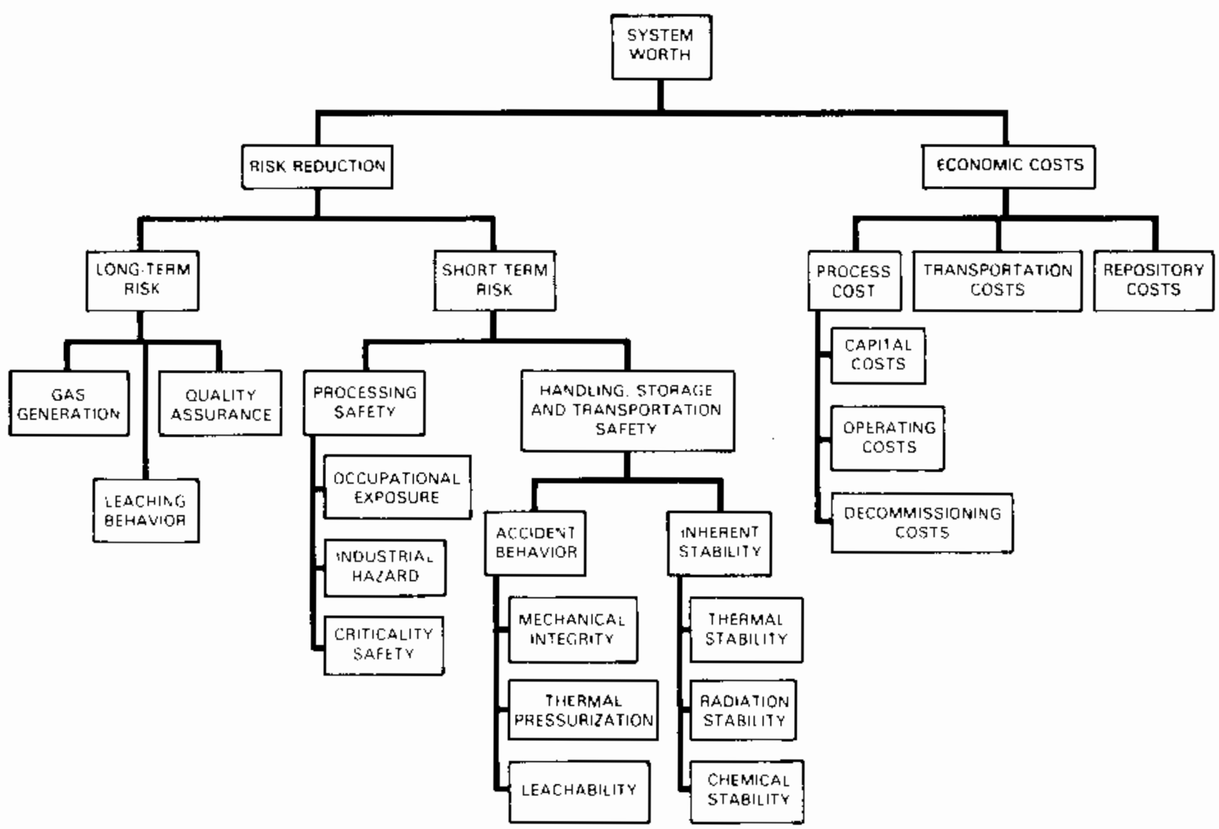

FIGURE 1. Hierarchy of TRU Waste Immobilization Systems Objectives

chemical vapors and dusts. During the handling of transuranic elements, the potential of nuclear criticality should also be considered. The post-processing considerations are inherent stability and dispersibility of the wastes and the behavior of waste systems during accidents. Other factors such as population exposure to radiation during transportation were considered, but were excluded from this study since they are controlled by engineering design in other parts of the waste management system.

Many of the risk factors for the different waste forms can be compared by using waste-form property data. It is generally recognized that data generated with different test methods, with different waste compositions, or at different laboratories are not directly comparable. Thus, a major factor in this study has been the selection of a single reference waste and the standardization of the fabrication and characterization of selected waste forms in order to provide comparable data.

The total costs of the eight systems have been estimated. The major variables in processing costs were determined by engineering estimates of processes conceptualized for the same facility. These estimates provided details of facility costs, manpower and operating costs, and decommissioning costs. The transportation and disposal costs, which are not highly immobilization-system dependent, were taken from other studies.

The final objective of this study was to bring together the waste immobilization system information in such a manner that it can be used by those selecting waste immobilization systems in the future. Each specific application of a waste immobilization system will have a different importance rating for each of the system characteristics. Therefore, no attempt has been made in this report to arrive at final ranking of the immobilization processes.

SELECTION OF REFERENCE WASTE AND WASTE-FORM CRITERIA AND ASSUMPTIONS

The variety of TRU wastes and waste-form options required selection of a reference waste and a few waste forms to reduce the number of variables in the study. This section discusses factors considered in developing a reference waste and the criteria used in finally selecting the waste forms for comparison and evaluation.

SELECTION OF A REFERENCE WASTE

The chemical and physical diversity of TRU wastes is greater than in HLW, owing to the many different processes and facilities that produce TRU waste materials. Table 1 provides some inventory data for defense TRU wastes stored at the Idaho National Engineering Laboratory (INEL) during a seven-year period (Bryan 1981). The WIPP criteria specifically state that sludges and fine powders must be immobilized for shipment to WIPP, thereby reducing handling risks in case of container failure. Because of this specific requirement for immobilization, a sludge and incinerator ash were selected for the study. The reference waste used in this comparative assessment is a mixture of three parts sludge and one part incinerator ash.

The chemical composition of incinerator ash can vary considerably since it is primarily dependent upon the mixture of combustible wastes charged to the incinerator. It is also dependent, to a lesser extent, upon the incinerator design and operating conditions. The ash is very heterogeneous owing to poor mixing and the occasional presence of noncombustible materials in the incinerator feed. If plutonium concentrations are high enough to warrant recovery (e.g. >0.5 wt%), the ash may be leached in concentrated nitric acid containing a small amount of hydrofluoric acid. This treatment will alter the chemical composition of the residual ash. The ash typically consists of oxides of iron, aluminum, silicon, and titanium with lesser amounts of alkali and alkaline earth oxides, sulfates and chlorides. A few weight percent of carbonaceous residue generally remains from incomplete oxidation. The composition of the ash (Table 2) used in this study was derived from the flowsheets for the new Rocky Flats Plant (RFP) Waste Treatment Facility (Buildings 371/374).

TABLE 1. Volumes and Plutonium Contents for Important Waste Forms Received for Storage at INEL from September 1971 through December 1978 (from Bryan 1981)

Waste Form	Volume		Plutonium Content
	m^3	% of Total	Wt% of Total
Metal Scrap	9248	30	13
Paper and Rags--Dry	3023	10	3
Second-Stage Sludge	1562	5	1
Paper and Rags--Moist	1554	5	1
Filters--CWS	1491	5	2
First-Stage Sludge	1262	4	13
Raschig Rings--Unleached	317	1	5
Incinerator Ash-Leached	21	0.1	8

TABLE 2. Chemical Compositions for Typical Process Sludge and Incinerator Ash

Compound	Composition, wt%		
	Process Sludge	Incinerator Ash	Waste Blend
Al_2O_3	3	20	7
CaO	24	8	20
Cr_2O_3	--	2	0.5
Fe_2O_3	10	8	9
K_2O	1	1	1
MgO	3	4	3
Na_2O	16	2	13
NiO	--	1	0.3
SiO_2	29	35	31
TiO_2	--	10	3
ZnO	--	2	0.5
C	--	3	0.8
Cl	0.5	0.5	0.5
H_2O	14	3	11
	100	100	100

Process sludges are generated during decontamination of aqueous wastes that contain unrecoverable concentrations of plutonium, americium and other radionuclides. Decontamination is accomplished by a precipitation/flocculation/filtration process in which a hydrated ferric oxide flocculant scavenges plutonium and americium from the aqueous stream. The precipitated sludge, which is collected on a precoated vacuum drum filter and dried to 10 to 15% water, consists of a variety of metallic compounds and diatomaceous earth filter aid. The principal sludge components and their concentrations are indicated in Table 2. Fluoride, sulfate and phosphate anions may also be present significant concentrations.

Data from the RFP indicate that the ratio of sludge to ash production would be about 3:1. The waste blend in Table 2 represents the waste composition that was used in preparation of the comparative waste-form materials. Plutonium-containing ash was obtained from Rocky Flats and was used in the fabrication of most samples. Details of sample fabrication are in Volume II.

WASTE-FORM SYSTEM SELECTION CRITERIA AND ASSUMPTIONS FOR THE COMPARATIVE STUDY

About 40 waste forms and processes were initially identified for possible inclusion in the study (Platt and Powell 1980). To reduce this group to a practical number for detailed review, a preliminary screening was performed. For the screening, minimum design objectives were adopted to partition the candidate systems into "probably acceptable" and "probably nonacceptable" categories. The screening was based on the waste-form performance criteria under development within the WIPP project and at NRC and on discussions of these developments with RFP-TWSO. Subsequently, the set of products and processes was further reduced based on degree of performance in various areas.

The design objectives used for preliminary screening fell mostly into the six categories of performance listed in Table 3. Each of these are briefly discussed below.

Dispersible Fines. Sludges, ashes and other particulate solids (contained in 55 gal drums, for instance) are generally considered undesirable for transportation or handling in a repository due to the possibility of nuclide respiration following accidental container breach. Further, if small-sized waste forms or particles were loosely packed in a container that was breached in a transportation accident, retrieval of all the contents might become difficult. Consequently, a prime processing objective is consolidation of these solid wastes to minimize the consequences of postulated accidents.

Gas Generation. Gases generated during degradation of waste forms may be of concern during interim storage and after emplacement in the repository. Degradation mechanisms include radiolysis of organics or contained water, thermal and bacterial degradation of organics, and corrosion of metals. The two performance specifications for gas generation noted in Table 3 were explicit in the draft WIPP acceptance criteria (December 1979) and appeared to eliminate organic-based waste forms such as bitumen or urea formaldehyde.

Waste Loading. Although the minimum waste loading is somewhat arbitrary, the choice was based on 1) the notion that economics will be very important in selection of a system for these wastes, and 2) our judgment of what loadings are achievable in most of the waste forms under consideration.

Production Capacity. We assumed the TRU waste-generating facility was the 371/374 complex at the Rocky Flats Plant. This assumption was necessary in order to develop detailed information on the waste characteristics and the immobilization process and facility designs. The capacity indicated is thus specific to RFP.

TABLE 3. Design Objectives Used in Preliminary Screening of Immobilization Alternatives

Immobilization Objective	Minimum Acceptable Performance Level
Minimize Dispersible Fines	As-formed, no smaller than 5 mm diameter sphere.
	<1 wt% respirable (10 μm) fines generated in 150-ft-lb impact test.
Minimize Gas Generation	<0.5 moles/ft^3/yr and <800 moles/ft^3 total.
Maximize Waste Loading	Minimum design loading 20 wt%; occasional loading of <10 wt% acceptable (e.g., to accommodate waste variability).
Maximize Production Capacity	Design feed rate should be 145,000 kg sludge plus 46,000 kg ash per year.
Minimize Implementation Time	Technology should be available for detailed design of demonstration plant by 1986.
Maximize Chemical Durability	Maximum corrosion rate of matrix elements (e.g., Si) of 10 g/m^2·d in deionized water at 25°C.

Availability. The year 1986 for the design of the demonstration plant is fixed by the WIPP operation schedule in place at the time this study was being conducted. It was anticipated that initial waste receipts at WIPP would be unprocessed wastes in retrievable storage (e.g., metals and combustibles), and that processing of newly generated sludge and ash would not start until the repository acceptance criteria were firmly adopted.

Chemical Durability. The WIPP acceptance criteria do not include a durability criterion. This is based on the Final Environmental Impact Statement analyses for WIPP, which demonstrated that long-term risk is not sensitive to leach rate. On the other hand, the NRC philosophy evolving at the time was based on the concept of multiple independent barriers, each barrier sufficient to meet Environmental Protection Agency standards. Hence, some waste-form release specification is likely for a licensed repository. Therefore, we judged that, given these wastes are to be processed, it would be prudent to achieve some minimum chemical durability. We chose a specification that is relatively easy to comply with, recognizing that solubility limits (rather than waste-form degradation kinetics) would likely constrain plutonium and americium releases and that waste packaging may also be available to reduce release rates.

With these criteria, the 40 waste forms and process combinations were reduced to seven waste forms and eight waste processes as noted in Table 4. These waste forms and processes are discussed below and in the remainder of this report.

- Cast Hydraulic Cement. Cement has long been used in waste disposal (Lokken 1978) and its processing is one of the simplest. Cast hydraulic cement has been extensively evaluated for HLW and has met the criteria for inclusion in our study.
- Pressed Hydraulic Cement. One of the major problems encountered with cast cement has been the residual water incorporated in the pores of the cement. The Mound Facility has developed the pressed-cement system to reduce the amount of water present in the cement (Lewis and Herbert 1981). Since this is a process currently under development, it was included in our study.
- FUETAP Cement. Cement formed under elevated temperature and pressure has been under development at ORNL for a number of years (Dole et al. 1982). The FUETAP

TABLE 4. Waste Forms and Processes Selected for the Comparative Study

Process	Waste-Form Product
In-Can Glass Melting	Borosilicate glass monolith
Joule-Heated Glass Melting	Borosilicate/aluminosilicate glass monolith
Glass Marble	Borosilicate/aluminosilicate glass marble
Basalt Glass Ceramic	Basalt glass-ceramic monolith
Cast Cement	Cast cement monolith
Pressed Cement	Pressed cement pellet
FUETAP Cement	FUETAP cement monolith
Cold-Pressed Sintered Ceramic	Pressed ceramic pellet

cement formulation can be tailored to specific wastes including TRU wastes. The process includes steps to accelerate curing and to dewater the cement.

- Borosilicate Glass. Glass was considered an improved product compared to cement. Glasses for incorporation of TRU wastes, and in particular for incinerator ash, have been under development for several years. Borosilicate glass is also considered the reference waste form for HLW.
- Aluminosilicate Glass. Aluminosilicate glasses are considered by some to have a higher chemical durability than borosilicate glasses, but they require higher processing temperatures (1350°C versus 1050°C for borosilicate glass). The study provided an opportunity to compare the two glasses directly.
- Basalt Glass Ceramics. Early tests have indicated that crystallized basalt glass has improved chemical durability over the parent glass from which the ceramic is formed. Improved durability appears as if it may be important in meeting the NRC requirements for a waste product. A high-iron basalt glass-ceramic has also been developed at EG&G in Idaho Falls (Flinn et al. 1979) for immobilization of their stored TRU wastes.
- Cold-Pressed and Sintered Silicate-Ceramic. This waste form was developed at LANL for immobilization of their TRU wastes. It offers the potential of conventional ceramic processing to produce a nearly crystalline material. It was included as a feasible ceramic waste form.

WASTE-FORM EVALUATION DATA

This section provides a summary of the characteristics of the waste forms and a comparison of their properties. The data included in this section address only waste-form factors identified in Figure 1. Additional basic waste-form data, such as density, microstructure and homogeneity, are contained in Volume II. The Materials Characterization Center (MCC) tests were utilized where available and are designated by MCC test number. However, such tests were in draft form at the time of testing.

GAS GENERATION

The generation of large volumes of gas from radiolytic, thermal, corrosive, or bacterial degradation is considered potentially detrimental for WIPP. The generation of radiolytic gas and the generation of gas from thermal events are considered in later sections. Corrosion is considered to be an important mechanism for gas generation for metals, but our wastes do not contain any significant quantity of metals, other than the canister. Therefore, evaluation of gas generation by corrosion was not necessary. Bacterial degradation requires an organic content above the residual carbon content of the ash waste. High-temperature processing will further reduce the carbon content for most of the waste forms. The cement forms have less than 0.4 wt% carbon or 0.008 g/cm^3, which is significantly below the WIPP limit of 0.096 g/cm^3 (Irby 1980). All waste forms, therefore, compare very favorably.

CHEMICAL DURABILITY

The static leach test (MCC-1) developed by the Materials Characterization Center was used for this comparative study. This test generates data to compare the behavior of waste forms as a function of temperature, leachant composition and time. Each waste form was leached in deionized water and silicate and brine solutions at temperatures of 40 and 90°C for times up to six months. A more detailed discussion of the procedure is available in Volume II. The data for each waste form are tabulated in that volume and are summarized in this section.

Some of the results of the leach tests are shown in Figures 2 and 3. Figure 2 shows final pHs and normalized mass losses based on Pu, Si, and Ca for the 90°C tests in deionized water as a function of time. Figure 3, a series of bar graphs, shows the effect of temperature and leachant in the 28-day leach tests. Because of the similarities in performance, it is convenient to group the cements, glasses, and ceramics for discussion.

The cast-hydraulic cement, the cold-pressed hydraulic cement and the FUETAP cement may be characterized as having the lowest plutonium releases of the seven waste forms. As Figure 2 shows, this behavior can be attributed to the high pH of the leachate which is quickly attained. Plutonium releases are low and constant with time; the releases of Ca, Na, and Al are much higher than for the glasses or the ceramics. Releases of Si from the cements lie between those of the glasses and the ceramics. Silicon solubility may be depressed by the higher aluminum concentration (Iler 1979). Figure 3 shows that temperature plays a minor

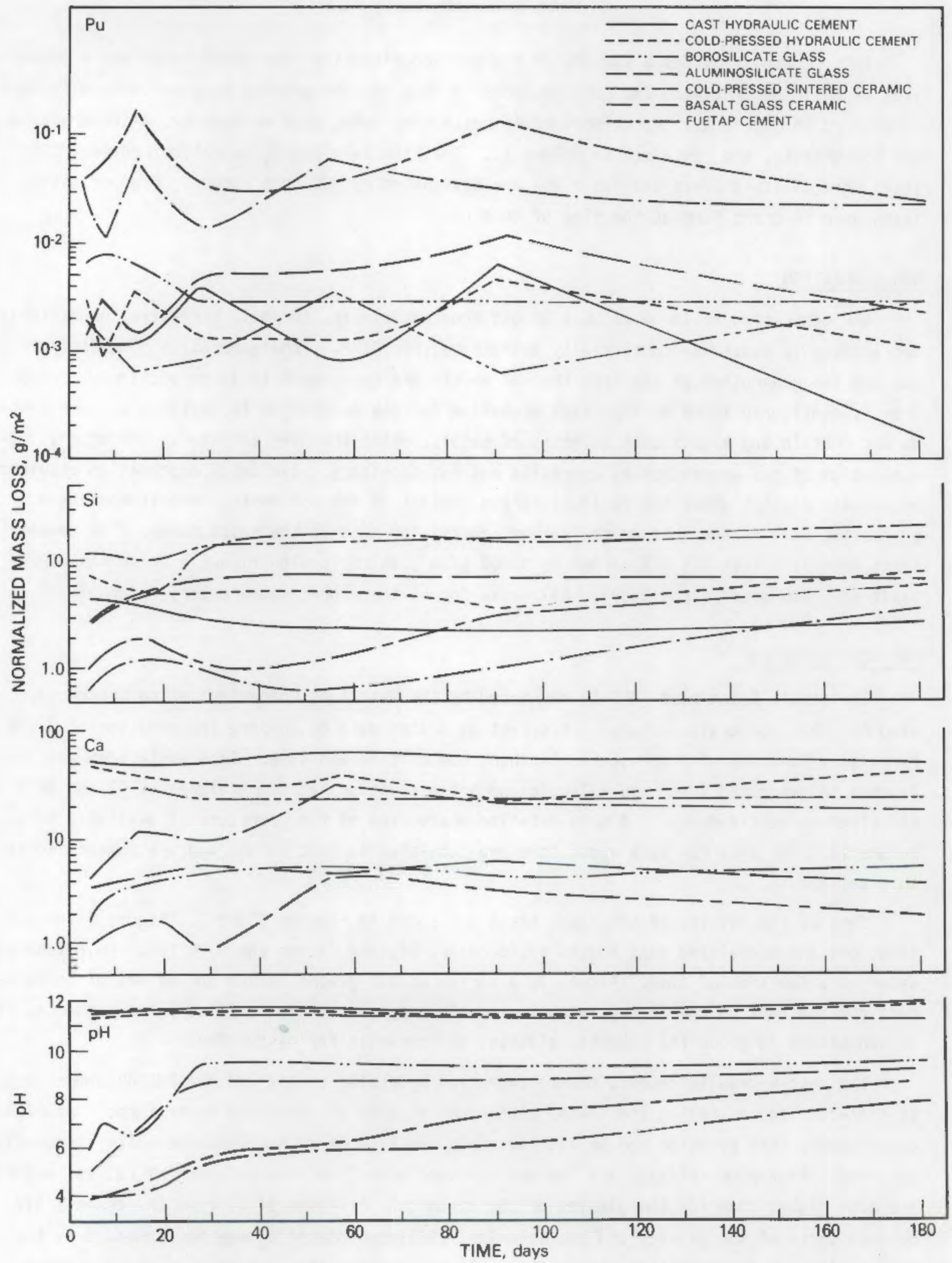

FIGURE 2. Leach Results for TRU Waste Forms Leached in Deionized Water at 90°C

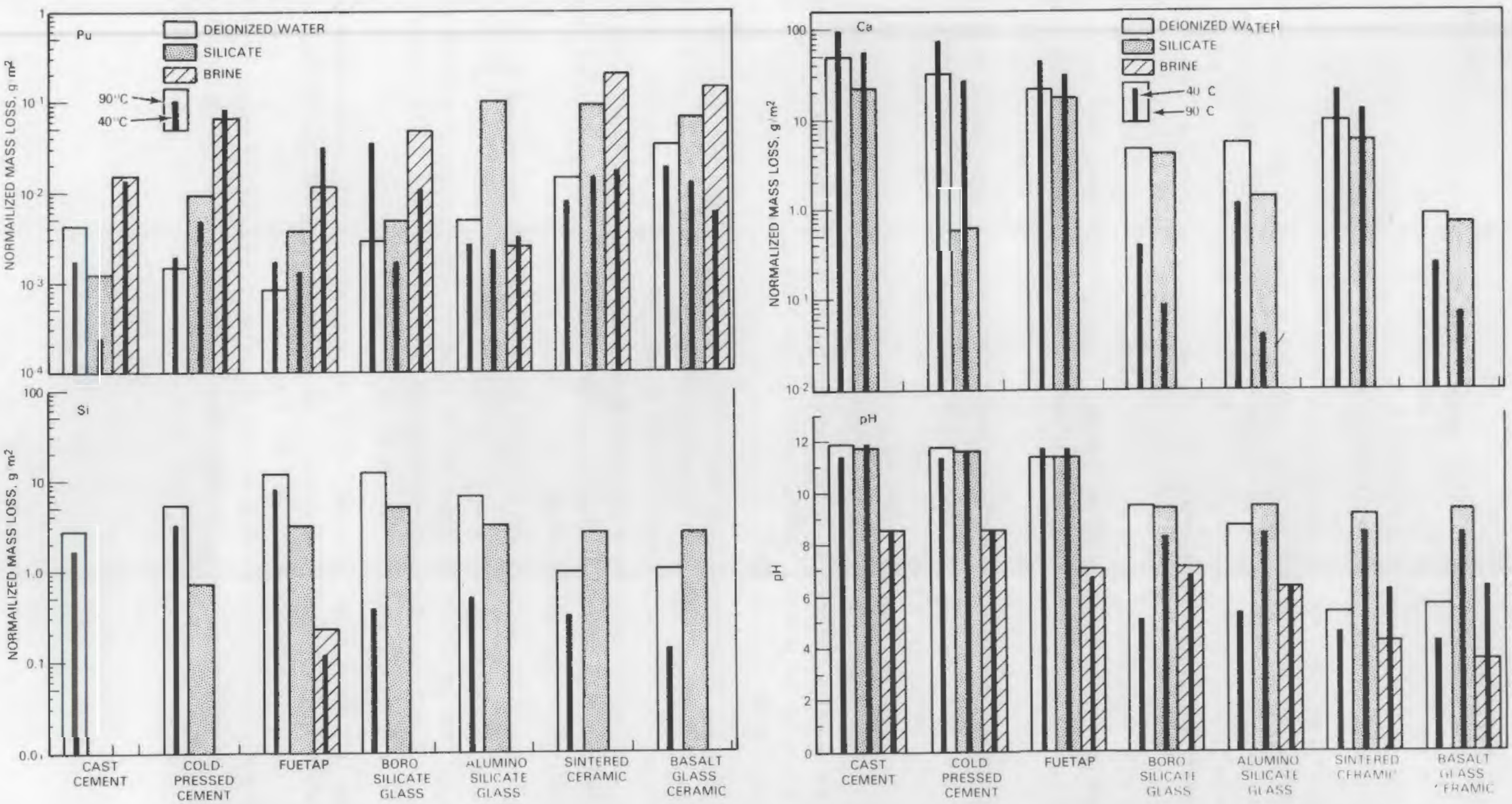

FIGURE 3. Leach Results Showing Effects of Temperature and Leachant Composition (28-day MCC-1 Static Leach Test)

role in the releases from the cements. There appears to be no significant difference in the behavior of the cements relative to the glasses and ceramics as far as their behavior in each of the leachants. Only information on Pu behavior is available in the brine because the high salt content precludes measuring the small concentrations of most leached species.

The borosilicate glass and the aluminosilicate glass leach behaviors generally fall between those of the cements and the ceramics. The glasses cause a change in leachate pH although the change is not as large or as rapid as that for the cements. Normalized mass losses based on Si are highest for the glasses. Mass losses based on Pu, Ca, Al and Na are between those for the cements and ceramics. The glasses do show a temperature dependence, with normalized mass losses higher after 28 days at 90°C compared with similar tests of the same time at 40°C. One anomaly in the results of the leach tests is evident in Figure 3. The normalized mass losses based on Pu are highest in the silicate leachant for the aluminosilicate glass. For the other six waste forms, the highest Pu releases occur in the brine leachant.

The basalt glass-ceramic and the cold-pressed sintered ceramic appear to be the most durable of the waste forms. Normalized mass losses based on Si, Na, and Al are lowest for these ceramics. However, Pu releases are highest for the ceramics. This corresponds with only a small change in the leachate pH with time. The basalt glass-ceramic had the lowest release of Ca, while releases of Ca from the sintered ceramic lie between those for the cements and the glasses. Interestingly, the sintered ceramic was the only waste form with measurable release of phosphorous. Similar to the glasses, the ceramics have higher normalized mass losses at 90°C than at 40°C.

To summarize the differences in the leachability of the seven TRU waste forms studied, the cements have the lowest releases of Pu but are otherwise less durable than the glasses or the ceramics. The leachate pH appears to play an important role in the releases from the waste forms. Even so, the differences among the waste forms are not more than two orders of magnitude. As demonstrated in the following, all seven waste forms may exceed proposed performance objectives for the waste package.

Recently proposed NRC criteria would require that for TRU waste the maximum annual release of radionuclides be not more than 1 x 10^{-5} of the maximum inventory of that radionuclide (Federal Register 1981). By comparing the volume of water necessary to remove one 10^{-5} fraction from each waste form with measured ground-water flow rates, it is possible to compare the seven waste forms with respect to the proposed NRC criteria.

In making this analysis the following assumptions are made: 1) plutonium concentrations in the leachate are controlled by solubility; 2) the plutonium concentrations measured in the 182-day, 90°C leachates represent the solubility limit for Pu and that waste form; and 3) the wastes are disposed of in 55 gal drums at typical densities. The amount of water required to remove a 10^{-5} fraction per year of Pu from a drum is then calculated from the formula:

$$\text{Annual flow rate} = \frac{(10^{-5}/\text{yr})(\text{Pu concentration in waste form})(\text{mass of waste form in drums})}{(\text{Pu concentration in 182-day leachate})}$$

The results are shown in Figure 4.

The potential ground-water flow rates through a repository are still being investigated. However, there appears to be some agreement that the maximum rates under "expected conditions" would be less than 10^3 liters/yr per drum. Assuming this flow rate, all the waste forms except the sintered ceramic in the silicate or brine leachants would meet the proposed NRC criteria. If lower ground-water flow rates through the repository are verified, then any of the seven waste forms would be acceptable.

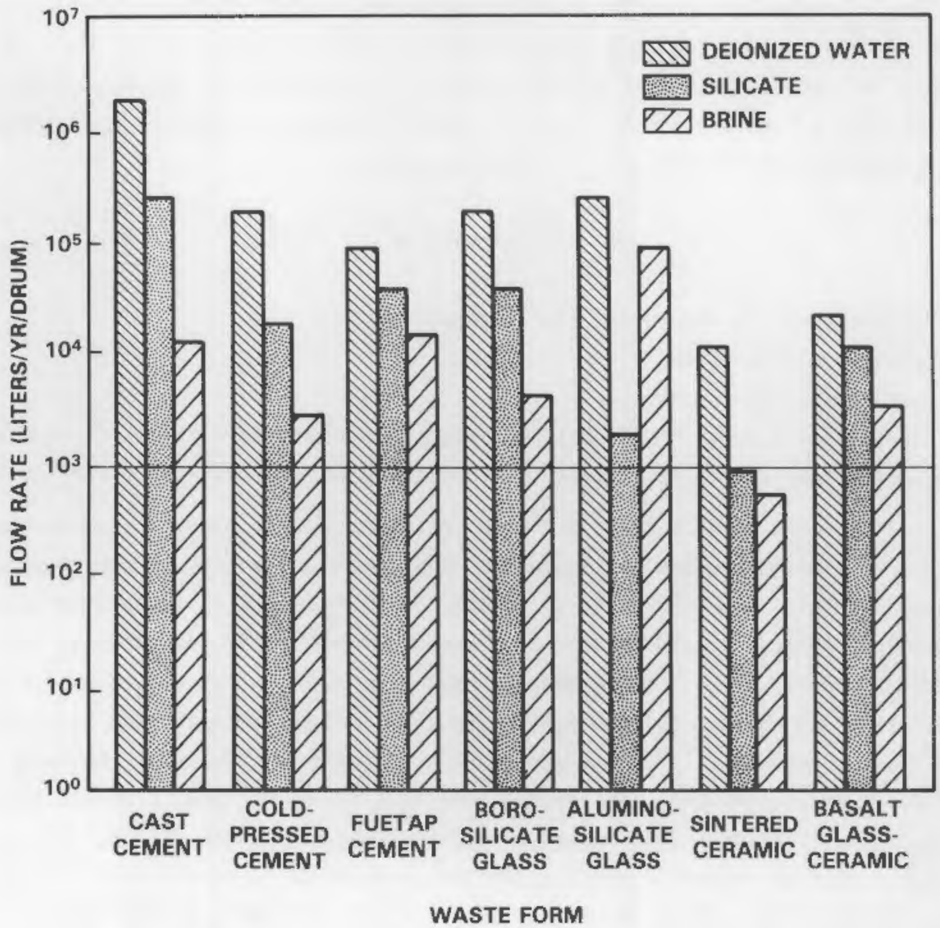

FIGURE 4. Minimum Flow Rates for 10^{-5}/Year Plutonium Release

MECHANICAL INTEGRITY

The ability of a waste form to resist fracturing during handling and possible accident situations is an important parameter that can affect long- and short-term safety assessments. In the short term, a waste form must be able to withstand routine handling following preparation and before its transportation to a repository. These activities include loading the waste form into canisters or drums and transporting the waste containers within the solidification facility. Additionally, the waste forms must be capable of withstanding transportation to and placement in a repository.

The mechanical integrity of the seven waste forms in this study was comparatively assessed using the results of a tensile test (MCC-11) designed for strength measurements of brittle solids. In the test, a right circular cylinder of waste form is loaded diametrically between two platens, one moving at a constant speed relative to the other. The stress is applied along the vertical diameter of the sample until the fracture strength of the material is reached, at which time the sample fractures along its vertical plane. The fracture strength (splitting tensile strength) is calculated by:

$$T = 2P/\pi dt, \text{ MPa}$$

where P = applied force at initiation of fracture, N

d = specimen diameter, mm

t = specimen thickness, mm.

Each sample used in the tests had a nominal diameter of 12 mm and a thickness of 6 mm and was fabricated in the same manner as samples in other tests except that they contained no Pu.

The results of the tensile tests are shown in Figure 5. As expected, the waste forms made from cement yielded the lowest strengths. The inherent porosity of cement products creates materials that, when subjected to tensile stresses, fracture at applied loads that are dependent on the actual load-bearing area within the material. As porosity is increased, the load-bearing area, or the total number of hydration bonds, is decreased, which results in lower apparent strengths. This phenomenon is evidenced in comparing the strengths of cast cement and pressed cement. The volume percent of open porosity of cast cement was 23% compared to 13% for pressed cement. The strength of this FUETAP is about one-half the strength of other cement waste forms. This may be a result of the high waste loading in the FUETAP and the lower number of hydration bonds. The two glass waste forms exhibited average tensile strengths of 35 to 40 MPa, while the crystalline ceramic and basalt glass ceramic had strengths of 48 and 60 MPa, respectively.

THERMAL PRESSURIZATION

Thermal pressurization is of concern when temperatures exceed ambient conditions during handling, storage, or transportation of the waste form. The most obvious scenario is that of a fire.

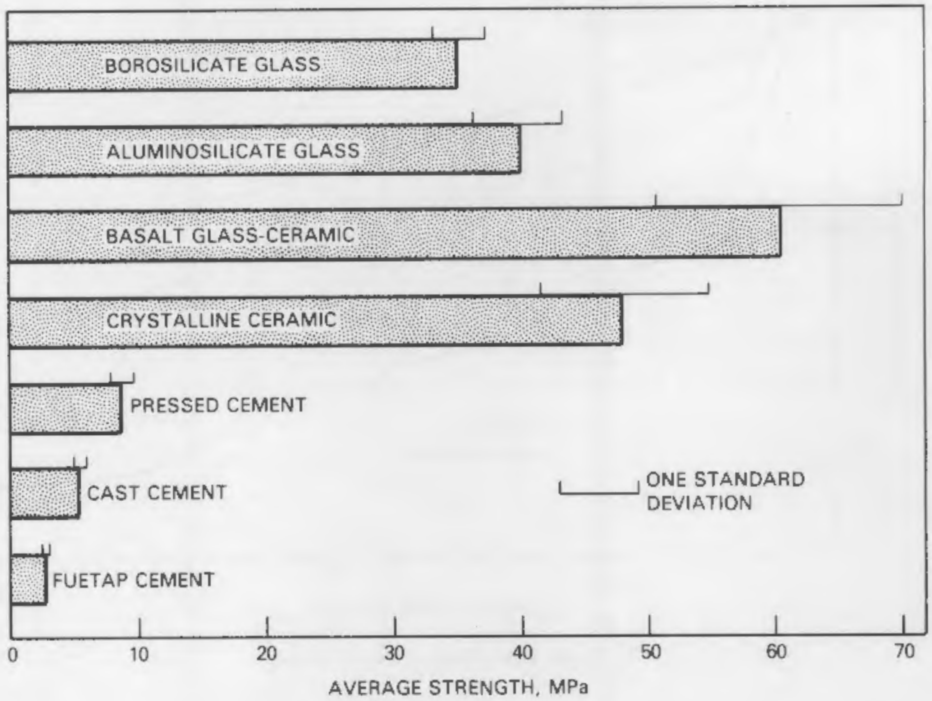

FIGURE 5. Average Tensile Strength of TRU Comparative Studies Waste Forms

Thermogravimetric analysis (TGA) was used to determine the potential for thermal pressurization. Weight loss was measured on samples weighing 20 to 40 mg which were ground to -40 +60 mesh. The samples were heated at 10°C/min from 25 to 800°C. The samples used for the TGA were prepared identically to those used for the leach testing, except that they contained no plutonium.

No detectable weight losses were observed with samples of borosilicate glass, aluminosilicate glass, basalt glass-ceramic or cold-pressed sintered ceramic. Thermal pressurization is not expected to be a problem with these waste forms.

As expected, the three cements showed substantial weight losses as seen in Figure 6. The loss for these materials began immediately upon heating and was not complete at 800°C. The cast cement showed distinct transitions at 190 to 210°C, 400 to 430°C and 520 to 630°C. These transitions were not evident with the cold-pressed or FUETAP cements. The weight loss in cement waste forms was shown by Stone (1977) to be predominantly due to evolution of H_2O, although some CO_2 is formed from the thermal decomposition of $CaCO_3$ at temperatures above 500°C. The volume of water generated as the temperature rises are sufficient to pressurize a 55 gal drum to the water saturation pressure. These pressures are high enough to rupture the 55 gal drums at temperatures greater than about 150°C. Such temperatures would require a fire or other external heat source and would, therefore, have a low probability of occurring.

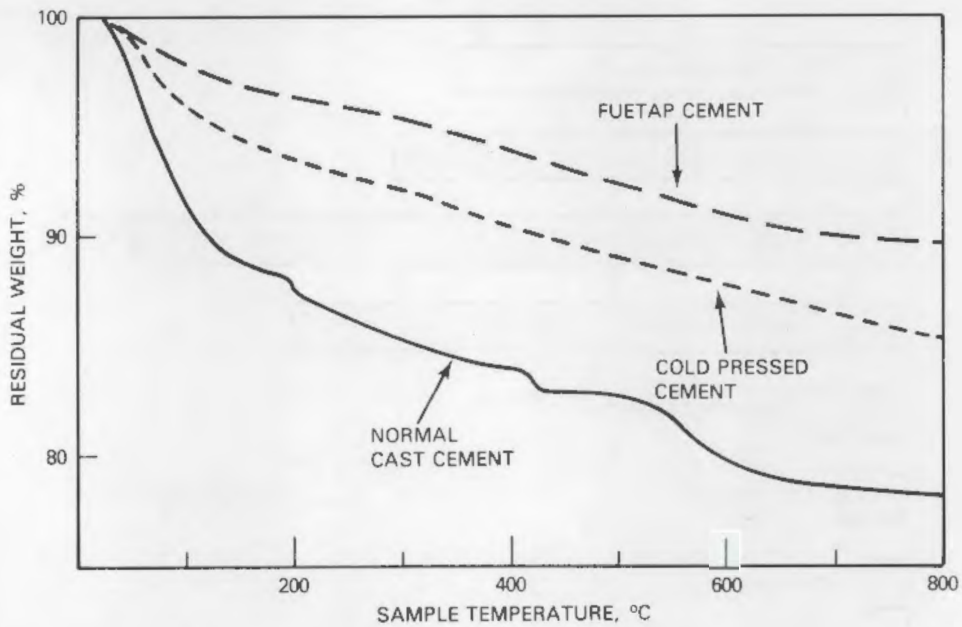

FIGURE 6. TGA Data for Cement Waste Forms

THERMAL STABILITY

Differential thermal analyses (DTA) were performed on the seven waste forms as a measure of their thermal stability. The measurements were made with 15- to 25-mg samples of the simulated waste forms ground to a particle size of -40 +60 mesh. The simulated waste forms were prepared identically to those used for leach testing except that the samples contained no plutonium. The DTA curves for the vitreous and ceramic waste forms are shown in Figure 7 and for the cements in Figure 8.

The DTA curves for the aluminosilicate glass, basalt glass-ceramic and cold-pressed sintered ceramic are completely featureless which, in conjunction with the TGA results (see the previous section), is evidence of thermal stability in the region of 25 to 600°C. The borosilicate glass displays a relatively small endotherm in the region of the glass transition temperature (530°C), which is characteristic of borosilicate waste glasses (Roberts, Jenks and Bopp 1976). This transition appears to be reversible because an identical DTA curve is obtained if the sample is cooled slowly and then reheated. This endotherm is not accompanied by a weight loss. Glasses can be expected to devitrify at temperatures above 600°C if sufficient time elapses.

The DTA curves of the cold-pressed cement and cast cement waste forms both show endothermic peaks at 135 and 505°C. The cast cement also undergoes an exothermic reaction over the temperature range of 370 to 600°C. The FUETAP cement shows two endothermic peaks--one at 180°C and one at 570°C. The peak at 570°C appears to be a reversible phase transition.

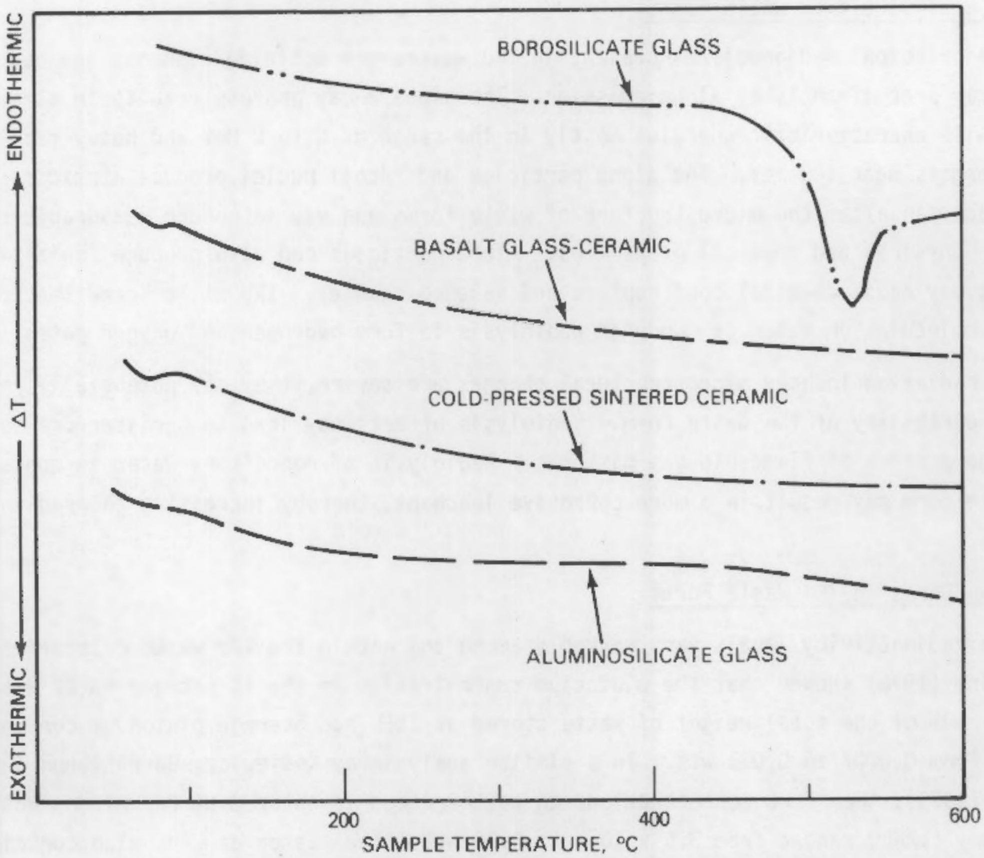

FIGURE 7. DTA Curves for Vitreous and Ceramic TRU Waste Forms

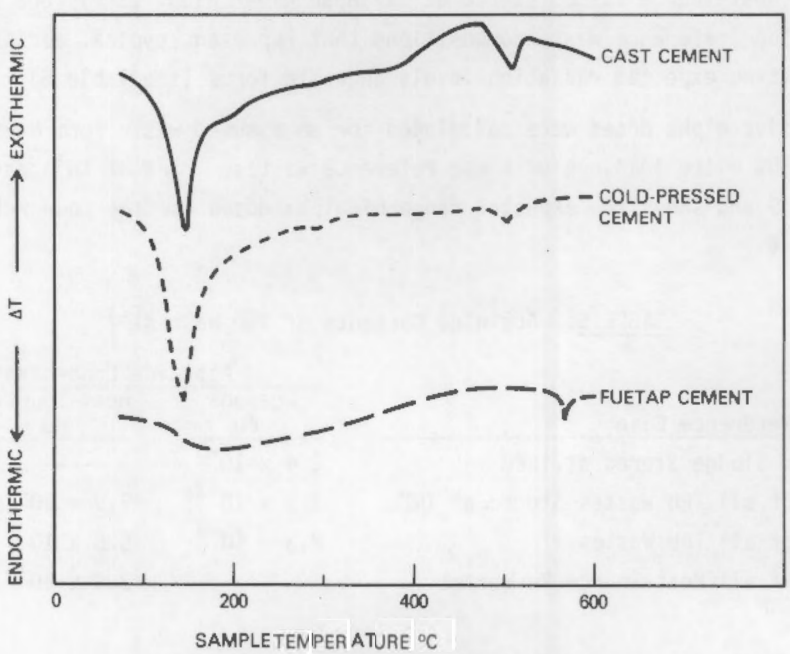

FIGURE 8. DTA Data for Cement Waste Forms

RADIATION STABILITY OF WASTE FORMS

The principal radionuclides present in TRU wastes are actinide elements and daughters that decay predominantly by alpha emission. The alpha decay process results in alpha particles with characteristic energies mostly in the range of 6 to 8 MeV and heavy recoil nuclei with energies near 100 keV. The alpha particles and recoil nuclei produce atomic displacements that can alter the microstructure of waste forms and may introduce measurable changes in their physical and chemical properties. Alpha particles can also produce ionization damage that may cause chemical bond rupture and valence changes. TRU waste forms that contain organic molecules or water can undergo radiolysis to form hydrogen and oxygen gases.

If radiation-induced microstructural changes are severe, they may potentially reduce the overall durability of the waste form. Radiolysis effects may lead to canister pressurization or the generation of flammable gas mixtures. Radiolysis of repository water in contact with the waste form may result in a more corrosive leachant, thereby increasing the radionuclide releases.

Radiation Doses in TRU Waste Forms

The radioactivity levels vary markedly among and within the TRU waste categories. Shefelbine (1978) showed that the plutonium concentration in the 10 categories of wastes containing >80% of the total weight of waste stored at INEL had average plutonium concentrations ranging from 0.0007 to 0.021 wt%. In a similar analysis by Kosiewicz, Barraclough and Zerwekh (1979), the ^{239}Pu concentrations in waste categories stored at Los Alamos National Laboratory (LANL) ranged from 3.5 x 10^{-6} to 0.09 wt%. The wastes at LANL also contain significant amounts of ^{238}Pu, which has a high specific activity and is used as a heat source. Based on these analyses (Shefelbine 1978; Kosewicz, Barraclough and Zerwekh 1979) and a description of the heat-source wastes stored at Savannah River Plant (SRP) (DOE 1979), Roberts (1981) derived four reference waste compositions that represent typical actinide concentrations for estimating expected radiation levels in waste forms (see Table 5).

The cumulative alpha doses were calculated for an assumed waste form having a density of 3 g/cm^3 and 50% waste loadings of these reference wastes. Part of this data is reproduced in Figure 9 and shows the expected range of alpha doses for the four reference TRU wastes in Table 5.

TABLE 5. Actinide Contents of TRU Wastes[(a)]

Reference Case	Actinide Concentration, wt% Weapons Pu	Heat-Source Pu	^{241}Am
I. First-Stage Sludge Stored at INEL	2.4 x 10^{-3}	---	9.0 x 10^{-4}
II. Composite of all TRU Wastes Stored at INEL	1.5 x 10^{-3}	7.9 x 10^{-6}	8.0 x 10^{-5}
III. Composite of all TRU Wastes	2.3 x 10^{-3}	5.6 x 10^{-4}	2.7 x 10^{-4}
IV. Composite of all Heat-Source Pu Wastes Stored at SRP	---	3.8 x 10^{-3}	---

(a) Adapted from Roberts (1981).

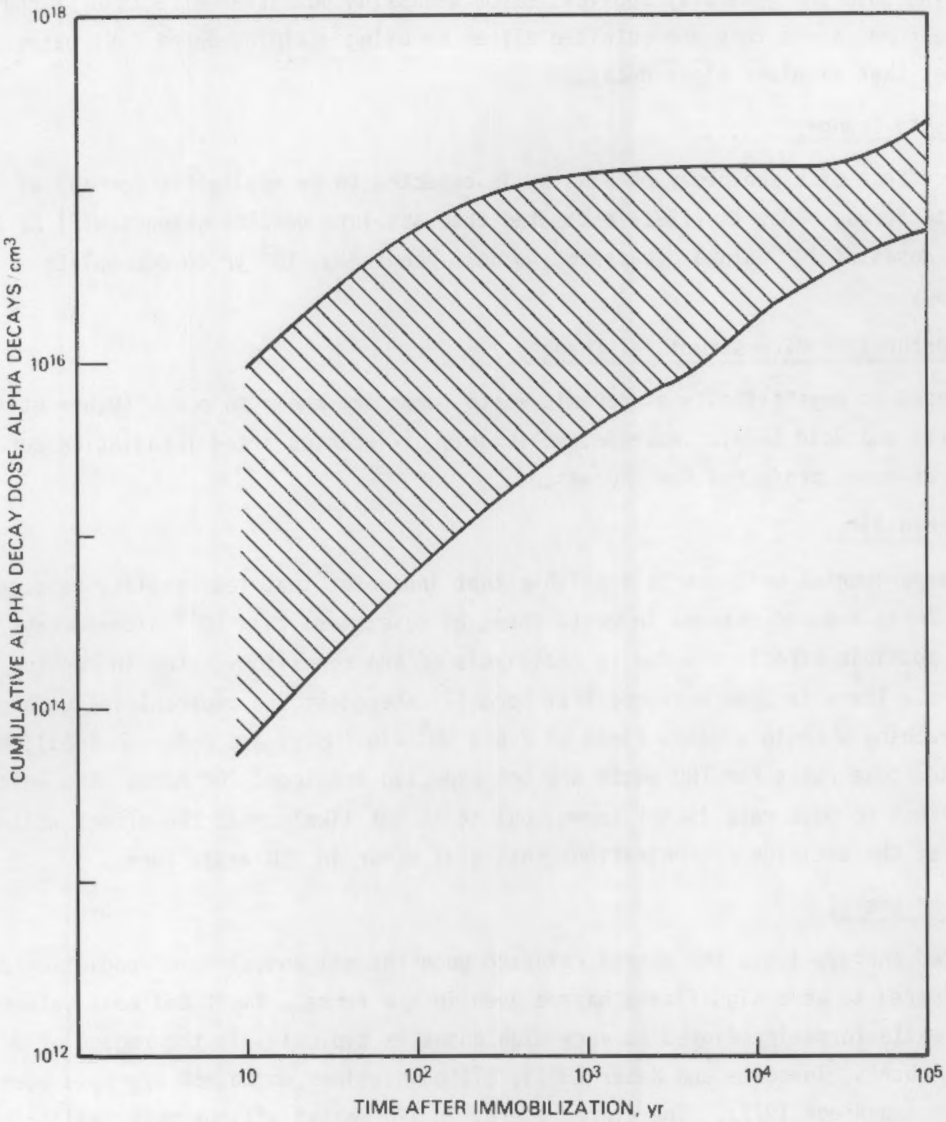

FIGURE 9. Expected Range of Alpha Decay Doses in TRU Waste Forms

Potential Effects of Radiation

Alpha decay may potentially produce measurable changes in the properties, which include:

- density
- structure and microstructure
- leachability
- stored energy
- mechanical properties
- radiogenic helium and radiolytic gas.

The status of research on radiation-induced property change has been recently reviewed (Weber and Roberts 1981). Much of the experimental work to date has focused on HLW waste forms that have radioactivity levels substantially higher than projected for TRU waste forms. The data are generally applicable for assessing potential effects of alpha decay on TRU waste forms since they are obtained either by using actinide-doped test materials or techniques that simulate alpha decay.

Density Changes

The effect of alpha decay on density is expected to be negligible for all of the reference waste forms. Roberts (1981) estimated that absolute density changes will be less than 0.002 at doses of 10^{17} alpha decays/cm^3, a dose requiring >10^3 yr to accumulate (Figure 9).

Structural or Microstructural Changes

Changes in crystallinity of ceramic waste forms are known to occur (Weber et al. 1979; Rusin, Gray and Wald 1979). However, measurable differences after irradiation were not observed at doses projected for TRU wastes.

Leachability

No experimental evidence is available that indicates that leachability is accelerated by alpha-decay-induced charges in waste forms at doses less than 10^{18} alpha decays/cm^3. However, possible effects are due to radiolysis of the repository water in contact with the waste form. There is some evidence that borosilicate glass and supercalcine undergo accelerated leaching when in a gamma field of 2.6 x 10^4 R/h (McVay and Pederson 1981); however, the surface dose rates for TRU waste are not expected to exceed 200 mR/h. The relationship of the effect to dose rate is not known, but it is not likely that the effect will be significant at the actinide concentrations that will occur in TRU waste forms.

Stored Energy

Stored energy, i.e., the energy released upon thermal annealing of radiation damage, is not considered to be a significant hazard even in HLW forms. The total heat released in vitreous waste forms irradiated to very high doses is typically in the region of 80 to 125 J/g (Roberts, Turcotte and Weber 1981), although values up to 350 J/g have been observed (Scheffler and Riege 1977). The stored energy in TRU wastes will be much smaller because of the relatively low doses.

Mechanical Properties

Changes in mechanical properties resulting from irradiation are of concern principally during preparation and transportation of the waste form to and emplacement in the repository. Preparation and transportation will occur early in the life of the waste canisters when the radiation doses are extremely small. Therefore, effects on the mechanical properties can be ignored.

Gas Generation

The formation of helium from alpha decay and the radiolytic generation of gas from the decomposition water are the two sources of radiogenic gas in TRU waste forms considered here.

Helium formation is minor, and the potential for canister pressurization is small. For example, the pressure increase calculated for a 55 gal drum having 10% void volume and containing 200 g weapon plutonium is only 100 kPa after 10^5 yr storage (Roberts 1981). Typical canisters will contain much less plutonium, and the pressures will be correspondingly smaller.

Radiolytic gas generation will be insignificant for the borosilicate glass, aluminosilicate glass, basalt glass-ceramic and the cold-pressed sintered ceramic forms because of their very low water contents. The three cement waste forms contain sufficient concentrations of water that can undergo radiolysis to form hydrogen and oxygen. The radiolytic gas yields are dramatically reduced by removing free water. The $G_{(H_2)}$[a] for cold-pressed cement containing SRP simulated wastes ranged from 0.003 to 0.009 as compared with values ranging from 0.28 to 0.65 for normal cast cement (Molecke 1979). The radiolytic gas generated in FUETAP cement is expected to be negligible. Studies at ORNL predict a $G_{(gas)}$ value of 0.005 (Dole et al. 1981). Experiments with ^{244}Cm doped simulated waste showed only slight pressure buildup at alpha decay doses up to doses that are equivalent to $>10^5$ yr exposure for a concrete containing 20 wt% actual defense waste (Moore et al. 1981).

To compare the potential canister pressurizations from radiolytic gas formation in the cast and cold-pressed cement waste forms, pressure increases as a function of time (or alpha decay dose) were estimated and are presented in Figure 10. The estimates are based on the reference canisters described in Table 6. The upper and lower ranges of pressure increases shown in the figure represent the ranges of alpha decay doses expected for TRU wastes given in Figure 9. We believe the estimates are conservatively high. The gas generation rates were assumed to be linear with respect to alpha decay dose over very long periods and independent of pressure. Also, no credit was taken for the open porosity of the cement waste forms, which will increase the void volume substantially.

Assessment of Difference

The cumulative radiation doses over very long periods of time ($>10^5$ yr) are not expected to produce significant chemical or physical property changes in the seven TRU waste forms considered in this report. The only effect resulting from radiation that may affect the comparative analysis is radiolytic gas generation. This phenonenon is important only for the cast and cold-pressed cement waste forms and if sealed, gas-tight canisters are required. The pressurization of waste canisters from radiolytic gas generation is dependent on the magnitude of $G_{(gas)}$, the amount of waste, the fractional void volume of the canister, and the temperature of the repository. These combined factors result in a 100-fold lower pressure increase in the canister of cold-pressed cement as compared to a canister of cast cement. While the calculated pressures in the canisters of cast cement are clearly unacceptable, the maximum 100 kPa pressure in the canister of cold-pressed cement may be tolerable, particularly when considering the retrieval periods of 10 to 100 yr being considered for geologic disposal.

(a) $G_{(gas)}$ is the number of molecules of gas evolved per 100 eV of radiation energy absorbed by the matrix.

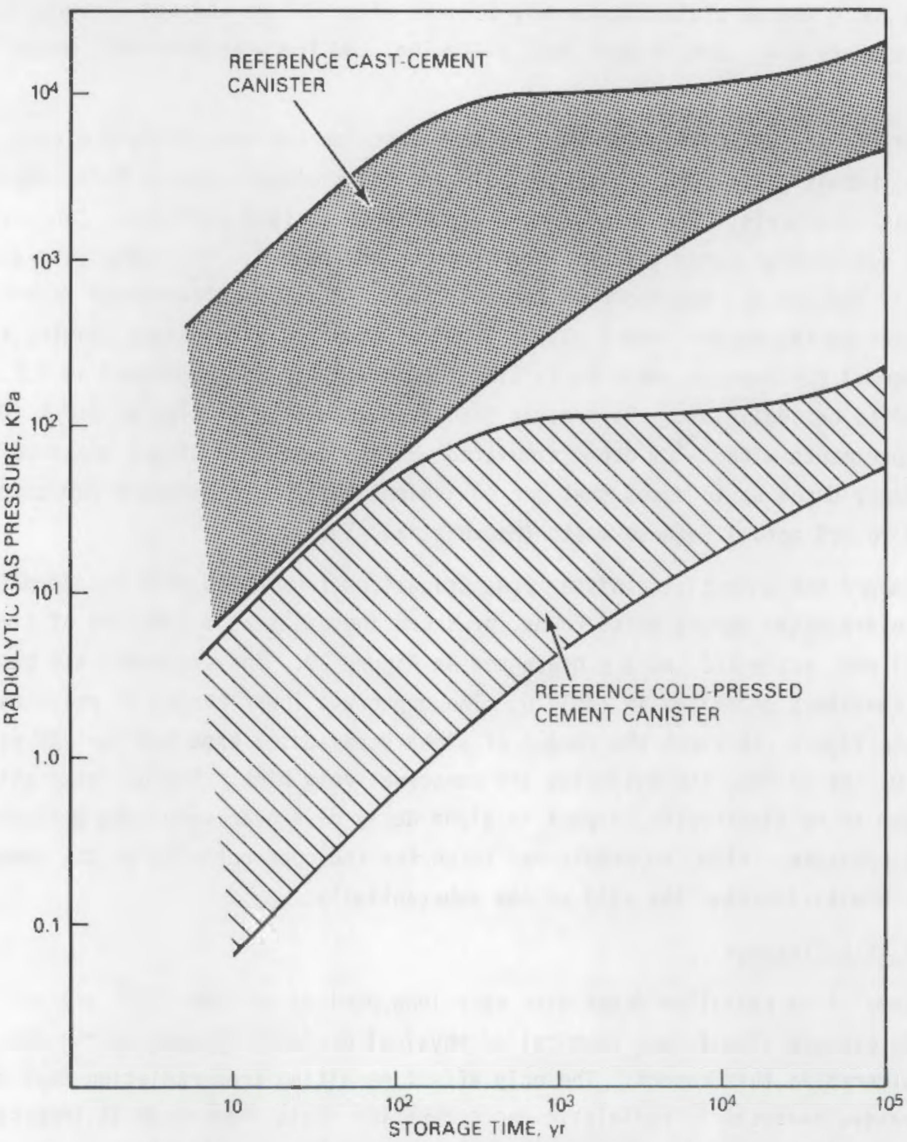

FIGURE 10. Calculated Canister Pressurization from Radiolytic Gas Formation in Cement TRU Waste Forms

TABLE 6. Characteristics for Calculation of Radiolytic Gas Pressurization of Cement Waste Forms[a]

	Cast Cement	Cold-Pressed Cement
Canister Volume	210 L	210 L
Void Volume	42 L	84 L
Weight of Cement	320 kg	154 kg
Waste Loading	25%	35%
G(gas)	0.30	0.01
Temperature	45°C	45°C

(a) Based on flowsheets presented by Timmerman (1980).

COMPARATIVE ASSESSMENT OF WASTE FORM PROPERTIES

The waste form data in the preceding sections have been utilized to provide an overall property comparison summarized here. Ratings of "1," "2," and "3" have been applied to the behavior of each waste form as compared to the behavior of the other six waste forms. For many of the properties, significant differences were not apparent from our data, and therefore, near-equivalent "top performance" is given the best rating. The rankings given in Table 7 indicate that cast cement has the poorest overall rating. Pressed cement and FUETAP cement are slightly improved over cast cement. The two glasses receive considerably higher ratings--the aluminosilicate has slightly better thermal stability although the borosilicate generally has lower plutonium release. The two ceramic forms received the highest overall ranking; however, the only noted improvements are in mechanical strength and thermal stability. It would seem likely that any of the waste forms may be an acceptable product for either the WIPP or NRC requirements.

In selecting a waste form for a specific application, it will be very important to determine what requirements are needed for that application. A review of property data, and particularly leaching data, should be made for that application and an assessment made with that perspective in mind since performance beyond the needed requirements is of limited value.

TABLE 7. Comparison of Waste Form Characterization Data

Waste Form	Gas Generation	Plutonium Leach Behavior	Matrix Leach Behavior	Mechanical Behavior	Thermal Pressurization	Thermal Stability	Radiation Stability
Cast Cement	1	1	3	3	3	3	3
Cold-Pressed Cement	1	1	3	3	3	3	2
FUETAP Cement	1	1	3	3	3	3	2
Borosilicate Glass	1	1	2	2	1	2	1
Aluminosilicate Glass	1	1	2	2	1	1	1
Sintered Silicate Ceramic	1	2	1	1	1	1	1
Basalt Glass-Ceramic	1	2	1	1	1	1	1

KEY: 1--Waste form property values of most worth (i.e., lowest leach rate or highest strength).
2--Waste form property values of intermediate worth.
3--Waste form property values of least worth (i.e., highest leach rate or lowest strength).

PROCESS EVALUATIONS

This section describes the evaluation of the eight processes that can be used to produce the seven characterized waste-form products. The processes are assessed primarily on the basis of economics and safety. A detailed description of a conceptual design and facility layout for each of these processes is provided in Volume II.

ECONOMIC EVALUATION

The economics of TRU waste disposal have been examined to understand the effects that each component of a waste immobilization system has on the cost. Specific cost components estimated include processing costs, transportation costs and repository or disposal costs.

Processing Costs

Processing costs were based on a new facility constructed at the Rocky Flats Plant in Colorado. Each process facility was considered to handle 145,000 kg/yr of process sludge and 46,000 kg/yr of incinerated low-specific activity (LSA) and high-specific activity (HSA) ash. Each product must meet the disposal requirements of the WIPP disposal site. For additional details on other process assumptions and bases, refer to Volume II.

Processing costs were determined by Kaiser Engineers under subcontract to this program. The cost category breakdown was as follows:

- capital costs
- operating costs
- maintenance costs
- decommissioning costs
- total processing facility costs.

Processing cost estimates for the eight processes are summarized in Table 8. Capital costs for all processes are very similar since the facility and construction costs are the major items, and equipment costs only have a minor influence. As Table 8 shows, the three cement processes are much lower in cost. This cost differential is primarily the result of the substantially lower operating and maintenance costs. The cement processes operate on an 8 h/day-5 day/wk schedule, whereas the glass and ceramic processes require continuous 24 h/day-7 day/wk operation. This difference of work schedules requires more operating personnel for the continuous processes, which increases their operating costs by a factor of approximately three. One shift/day operation is a key advantage for the cement processes or any other process that could operate on this schedule.

The very high operating cost for the in-can glass melting process results from costs of material, fabrication, and quality assurance associated with the waste canister, which total approximately $10,000 per canister. Investigations were made to find out if a more economical canister could be produced. The results of the investigation indicated that a canister could be produced for about $1,000. The costs of both canisters are given here to show the effect of these costs on total operating and processing costs.

TABLE 8. Cost Estimates for a Processing Facility for Seven Waste Immobilization Processes (all costs in thousands of 1981 dollars)

Process	Capital Cost	30-Yr Operating Cost	30-Yr Maintenance Cost	Decommissioning Cost	Total 30-Yr Processing Cost
In-Can Glass Melting					
$10,000 canister	$33,000	$164,000	$13,000	$2,000	$212,000
$1,000 canister	33,000	81,000	13,000	2,000	129,000
Joule-Heated Glass Melting	30,000	69,000	18,000	2,000	119,000
Glass Marble	33,000	69,000	20,000	2,000	124,000
Basalt Glass-Ceramic	30,000	77,000	18,000	2,000	127,000
Cast Cement	29,000	20,000	10,000	2,000	61,000
Pressed Cement	32,000	20,000	12,000	2,000	66,000
FUETAP Cement	34,000	22,000	12,000	2,000	70,000
Cold-Pressed Sintered Ceramic	34,000	63,000	13,000	2,000	112,000

Maintenance costs include general facility and equipment maintenance along with equipment replacement costs. The processes that require a joule-heated melter have higher maintenance costs because the replacement of the melter is anticipated to occur every three years. The decommissioning costs are estimated to be the same for all the processes since the processing facilities are essentially similar in size; the costs are based on the decommissioning cost/ft^2 (Nuclear Energy Services 1980).

Transportation Costs

The transportation costs are based on the cost of a shipment from the processing facility to the repository, and on the number of shipments required to meet the processing rates. The TRUPACT container (Eakes et al. 1980) is the reference shipping container. The number of waste-form containers that can be shipped in each TRUPACT is determined either by space or weight limitations. All processes, except the in-can glass melting process, use a 6-pack, 55 gal drum configuration as shown in Figure 11. The in-can glass melting process uses a 4-pack arrangement as illustrated and described in Volume II. Truck TRUPACT for all processes, except the joule-heated glass melting and the basalt glass-ceramic, are space-limited and can allow thirty-six 55 gal drums or eight in-can melting canisters in their 6- and 4-pack arrangements, respectively. The joule-heated glass melting and basalt glass-ceramic products are sufficiently dense that only twenty-four 55 gal drums or four 6-packs are permitted in a truck TRUPACT to remain below the payload limit of 12,700 kg (28,000 lb). Using the described limitations for each process, the number of truck TRUPACT shipments required per year were calculated and are supplied in Table 9. From the TRUPACT/yr number, a cost of $1,600/shipment (Andrews et al. 1982), based on published 1980 transportation tariffs and container use rates between the RFP site and the WIPP site, was used to estimate the transportation cost per year and the total cost over the expected 30-yr life of the processing facility. These transportation costs are also provided in Table 9.

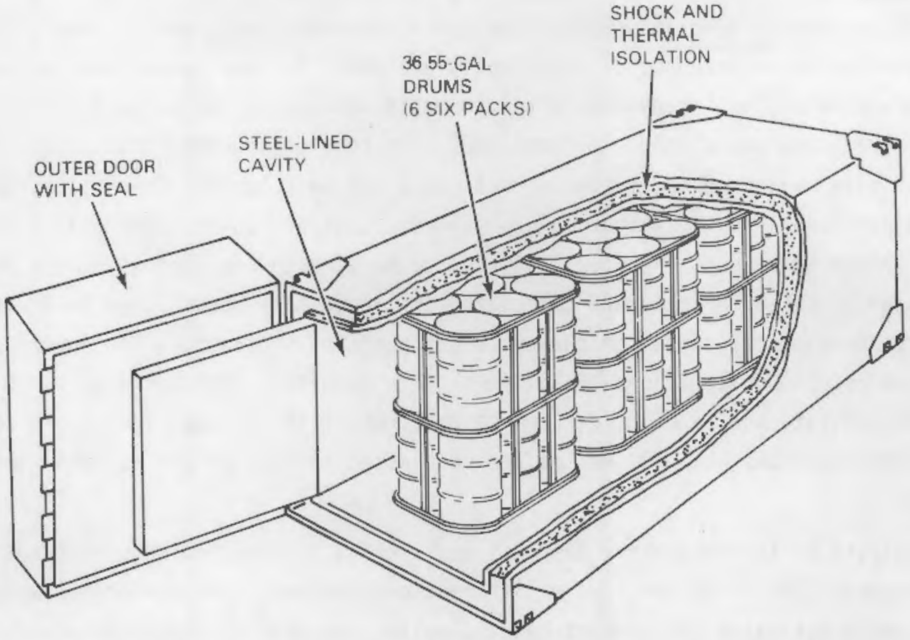

FIGURE 11. Truck TRUPACT Container Showing Configuration with 6-Packs

TABLE 9. Cost Estimate of Transportation of TRU Waste Forms (all costs in thousands of 1981 dollars)

Process	Containers[a]/Yr	Truck TRUPACT/Yr	Cost/Yr	Total 30 Yr Transportation Cost
In-Can Glass Melting	308	39	$ 62	$1,870
Joule-Heated Glass Melting	795	34	54	1,630
Glass Marble	1,325	37	59	1,770
Basalt Glass-Ceramic	795	34	54	1,630
Cast Cement	2,330	65	104	3,120
Pressed Cement	2,775	78	125	3,750
FUETAP Cement	1,530	43	69	2,070
Cold-Pressed Sintered Ceramic	2,800	80	128	3,840

(a) All processes use 55 gal drums, except in-can glass melting, which uses a 0.7 m dia x 1.5 m tall canister.

Estimated transportation costs indicate a slight advantage to the lower volume generation processes. However, the cost of transporting the waste to the repository are small when compared to the processing costs.

Repository Costs

The costs of disposal of immobilized TRU wastes based on the RFP generation rates have been considered for two disposal scenarios: wastes disposed in the defense-related WIPP repository and the same volume of wastes placed in a commercial TRU repository.

The costs for disposal of TRU wastes in a defense facility such as WIPP have been reported to be rather insensitive to volume since the major costs are for the surface facility construction and maintenance. Also, costs are minor for mining out storage rooms. A recent evaluation of the incentives to reduce waste volume has indicated a savings of $640/m^3$ for reducing waste volume (Brown 1982). In considering total costs for the near term, Brown also evaluated the probable costs on a per unit basis. This value, $3300/m^3$, is much higher than $640/m^3$, which implies that the cost per unit volume will increase if the total volume decreases. For our analysis, we have taken the largest-volume waste form (cold-pressed sintered ceramic) and calculated the repository costs based on $3300/m^3$. We have then calculated the costs for the other processes by subtracting the potential costs savings from reduced waste volume at $640/m^3$. This results in the costs given in Table 10. These costs indicate that a major difference does not exist in repository costs for the various process options when TRU wastes are considered in this manner for the WIPP defense repository.

An analysis of the repository disposal costs using information generated for commercial waste management (DOE 1979) provides a different perspective. The commercial waste repository costs were estimated for contact-handled wastes received at repositories of four different formations--salt, granite, shale, and basalt. These costs assume that the facility, acceptance rate, and all other cost-influencing facts are the same as those in DOE (1979). The costs per 55 gal drum container were $900 and $1800 (1978 dollars) for the salt and basalt repositories, respectively. Determining the costs on a unit basis and escalating these costs with increases of 9.88%, 9.1%, and 10.7% for 1979, 1980, and 1981 provides a unit cost range of $5650/m^3$ to $11,300/m^3$. When applying this range to the volume of immobilized wastes generated by each process considered in this study, we arrive at the cost estimates in Table 11. Based on this table, the disposal costs for a commercial TRU waste repository become a major portion of the total cost for the higher-volume generating processes, namely the cast and pressed cement and sintered ceramic processes.

Table 11 also illustrates other volume-related cost trends dependent on waste loading and void space. For similar density waste forms, the waste-loading difference between cast

TABLE 10. Estimate of Disposal Costs for TRU Waste in a Defense Repository (all costs in thousands of 1981 dollars)

Process	Annual Volume, m^3	Cost/Yr	Total 30-Yr Cost
In-Can Glass Melting	187	$1,720	$51,500
Joule-Heated Glass Melting	166	1,700	51,000
Glass Marble	276	1,770	53,100
Basalt Glass-Ceramic	166	1,700	51,000
Cast Cement	485	1,910	57,200
Pressed Cement	578	1,970	59,100
FUETAP Cement	319	1,800	54,000
Cold-Pressed Sintered Ceramic	600	1,980	59,400

TABLE 11. Estimate of Disposal Cost Range for TRU Waste in a Commercial Repository (all costs in thousands of 1981 dollars)

Process	Annual Volume, m^3	Cost/Yr		Total 30-Yr Cost	
		Low	High	Low	High
In-Can Glass Melting	187	1,055	2,110	31,600	63,200
Joule-Heated Glass Melting	166	935	1,870	28,100	56,100
Glass Marble	276	1,560	3,120	46,800	93,500
Basalt Glass-Ceramic	166	935	1,870	28,100	56,100
Cast Cement	485	2,740	5,480	82,200	164,000
Pressed Cement	578	3,270	6,530	98,000	196,000
FUETAP Cement	319	1,800	3,600	54,000	108,000
Cold-Pressed Sintered Ceramic	600	3,390	6,780	102,000	203,000

and FUETAP cement (26 and 40 wt%, respectively) caused the increased immobilized-waste volume and corresponding increase in disposal costs. Comparing the pressed cement with either of these monolithic cement waste forms shows the increased disposal costs of pelletization because of more void volume and increased containerized volume.

Total Costs

The total cost of TRU waste immobilization and disposal is the summation of processing, transportation, and repository costs. The individual total costs for disposal of TRU wastes in a defense repository are provided in Table 12 for the eight processes considered in this study. As the table illustrates, the cast cement process, because of its low processing cost, is the most economical of the eight processes. The pressed and FUETAP cement processes are slightly more expensive than the cast cement process. The other processes have very similar costs, except in-can glass melting when a $10,000 canister is used.

For TRU wastes placed in a commercial repository, the total costs are shown in Table 13. The cement processes (specifically the FUETAP cement process) have the lowest total cost with the glass processes becoming competitive in cost. It should be noted that the FUETAP cement system is economically better than the cast cement because of the waste-loading difference between the two--40 wt% for FUETAP and 26 wt% for cast. These waste loadings have not been optimized for this study but are the as-formulated waste loading used to produce and evaluate the various waste forms. Obviously if a 40 wt% waste-loading cast cement product was made, its lower processing costs would allow it to perform even better in this economic evaluation. However, with the results presented, the reader must refer to the product evaluation sections to compare the effects waste loading and processing sequences have on waste form characteristics. The figures presented in Table 13 do, however, illustrate the need for good overall evaluation of costs since higher repository costs can overcome lower process costs and make lower volume generating systems cost competitive. It is also important to note again that the difference in total cost for the various processes is not very large.

TABLE 12. Total Cost Estimate for Immobilization of TRU Wastes and Disposal in a Defense Repository (all costs in thousands of 1981 dollars)

Process	30 Yr Costs Processing	Transportation	Repository	Total	Total Annual Cost
In-Can Glass Melting					
$10,000 canister	$212,000	$1,870	$51,500	$265,000	$8,900
$1,000 canister	129,000	1,870	51,500	182,000	6,100
Joule-Heated Glass Melting	119,000	1,630	51,000	172,000	5,700
Glass Marble	124,000	1,770	53,100	179,000	6,000
Basalt Glass-Ceramic	127,000	1,630	51,000	180,000	6,000
Cast Cement	61,000	3,120	57,200	121,000	4,000
Pressed Cement	66,000	3,750	59,100	129,000	4,300
FUETAP Cement	70,000	2,070	54,000	126,000	4,200
Cold-Pressed Sintered Ceramic	112,000	3,840	59,400	175,000	5,800

TABLE 13. Total Cost Estimate for Immobilization of TRU Wastes and Disposal in a Commercial Repository (all costs in thousands of 1981 dollars)

Process	30 Yr Costs					
	Processing	Transportation	Repository Low	Repository High	Total Low	Total High
In-Can Glass Melting						
$10,000 canister	$212,000	$1,870	$31,600	$63,200	$245,000	$277,000
$1,000 canister	129,000	1,870	31,600	63,200	162,000	194,000
Joule-Heated Glass Melting	119,000	1,630	28,100	56,100	149,000	177,000
Glass Marble	124,000	1,770	46,800	93,500	173,000	219,000
Basalt Glass-Ceramic	127,000	1,630	28,100	56,100	157,000	185,000
Cast Cement	61,000	3,120	82,200	164,000	146,000	228,000
Pressed Cement	66,000	3,750	98,000	196,000	168,000	266,000
FUETAP Cement	70,000	2,070	54,000	108,000	126,000	180,000
Cold-Pressed Sintered Ceramic	112,000	3,840	102,000	203,000	218,000	319,000

ECONOMIC ASSESSMENT

The major cost influencing factors for the immobilization systems are indicated by the cost breakdown provided. Processing costs are reduced via a simpler process that performs effectively in a shorter operating time. The cement processes accomplish this and therefore are approximately a factor of two less costly than the other processes. Transportation and repository costs are lower for immobilization processes that generate lower volume waste forms. However, transportation costs are not major compared to the other costs involved. Repository costs were calculated based on the cost per volume basis of two referenced scenarios: one for the defense-related WIPP repository and the other for commercial repositories. Each of these illustrate the different impact the repository costs can have on total cost of a specific immobilization system.

Total costs for the immobilization and disposal of TRU wastes in a defense-based WIPP repository indicate the cement immobilization systems have a 25 to 30% cost reduction over the glass and ceramic systems. The cast cement is the least costly of the cement systems because it is a simpler process than the pressed or FUETAP cement processes.

Total costs for immobilized TRU wastes in a commercial repository present a similar but slightly different view from the above TRU waste total costs relating to a defense repository. These costs indicate that lower volume waste form products can provide up to a factor of four reduction in repository costs. Using the lower salt repository costs as a guideline, the total costs of the FUETAP system proved to be the lowest, followed closely by the cast cement and joule-heated glass-melting systems. This shows the advantage of a high waste loading in reducing the immobilized volume generated and thereby the disposal costs. It also illustrates the cost incentives of reduced volume generation systems in that the glass systems become competitive or even economically superior (for a basalt repository) to the cements. It also illustrates the need to review all the costs that go into producing and disposing of an immobilized TRU waste form.

OCCUPATIONAL EXPOSURE

The occupational radiation exposure, or dose estimate, is a function of "hands-on" operations. These operations include routine maintenance, equipment replacement, and normal process operations. Routine maintenance operations include lubricating bearings and seals, dislodging material blockages, cleaning material spills, changing filters, changing worn or damaged parts, etc. Equipment replacement items are shredders, pneumatic and/or hydraulic systems, dryers, scrubbers, pumps, blenders, feeders, rotary airlocks, heating elements, melters, mixers, presses, conveyors, etc. Processing operations entail ash unloading, scrap cleanup, load-out of recycle containers, filter coating, weld inspection, drum assay and weighing, drum lidding and load-out, etc. These operations vary in need and frequency from process to process; therefore, a rating of the processes as to their occupational exposure risk has been derived by estimating the number of these operations and their frequency for each process. The estimate of these operations (20,000 to 25,000/yr) for the conceptualized processes is provided in Figure 12.

Figure 12 shows that the in-can glass melting process offers the minimum work doses. Analysis of the occupational exposure also indicates that the amount of exposure-related operations is a function of the number of containers processed and the overall complexity of the process. These two factors, more than any other, lead to the relative results of the table. The FUETAP cement process illustrates the effects reduced container handling has on occupational exposure over the cast cement due to waste loading differences between the two.

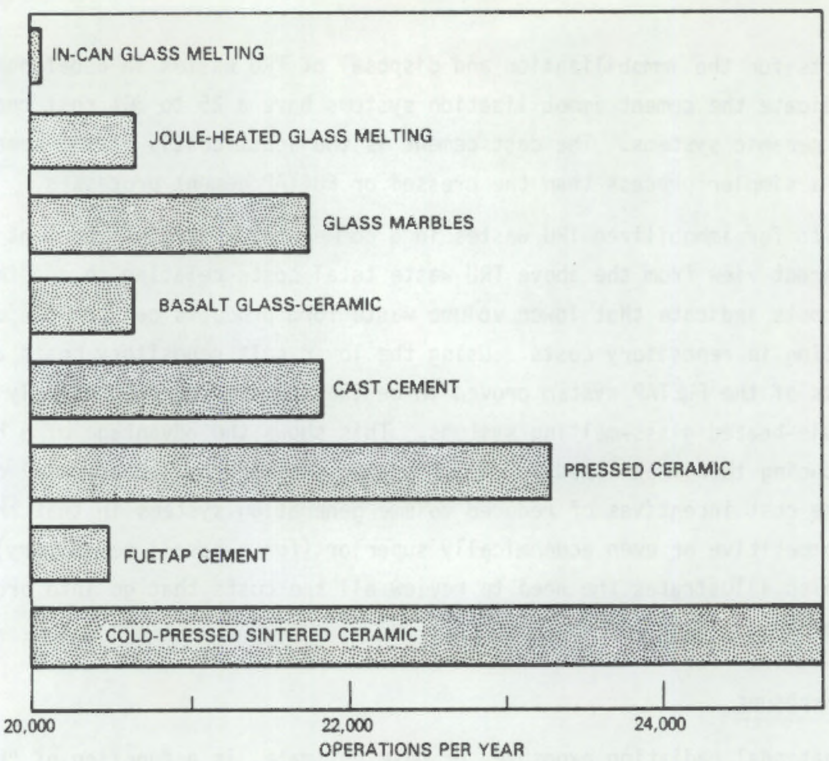

FIGURE 12. Occupational Exposure Operations Estimated per Year for Each Process

INDUSTRIAL HAZARDS

The industrial hazards considered for the processes were fire, explosion, pressurization, high temperature, hazardous off gas, and electrical- and mechanical-related injury potentials. The hazards relate to the potential for causing injury to a worker.

Fire and minor explosions would not be of major concern since they would be contained in the process canyon and would not likely cause any personnel injuries or a release of hazardous material outside the canyon. However, any fire or explosion outside the process canyon, or on a large scale, would have the same effect on personnel safety as at any other industrial complex. Pressurization is an unlikely processing concern since no violent chemical reactions occur and all processes have adequate off-gas vacuum to prevent any minor pressurization effects that may occur because of processing. The containerized cement products have shown drum pressurization resulting from radiolysis of water. This radiolytic gas generation is unlikely to occur during the brief interim storage of these drums at the solidification facility, but must be considered for longer-term interim storage if they are not sent directly to a repository or if vented cans are not used.

Processing temperature effects can take their toll in the form of burns to operating or maintenance personnel. The higher temperature processes, namely the glass and ceramic processes, do have the potential for causing skin burns if maintenance operations are performed around the operating equipment while at high temperatures or during container-handling for loadout purposes. Loadout handling would be a daily operation where personnel would be exposed to higher-temperature (>200°C) containers of glass or sintered-ceramic material. Normal operating precautions, such as insulated gloves and protective clothing, should provide adequate protection against these potential burn injuries. The cast and pressed cement processes would not have any effects related to temperature since the maximum temperature generated by heat of hydration is 60°C. Normal operating precautions as described above for the glasses would be required for FUETAP cement.

The potential for personnel injury because of hazardous off gas is very low because the processing area is semi-remote, which would limit exposure to off gas if an accident occurred. Also, the building is designed to sweep air from areas of least contamination to areas of high contamination. This air sweep would confine any hazardous off gas to the processing area. The major hazardous components of off gas would include NO_x, SO_x and CO. No volatile radionuclides exist in these TRU wastes; however, entrained radionuclides could enter the off-gas system as particulates. Only the high-temperature processes (glass and ceramic) will generate hazardous off gas.

Mechanical- and electrical-related injuries will also be minimized because of the remote nature of the processing. Such injuries may occur during maintenance operations. Some of the equipment related to these types of injuries are:

1. LSA ash-bag shredder
2. vacuum drum filter
3. paddle dryer
4. solids blender
5. in-can furnace
6. joule-heated glass melter
7. drum roller conveyors
8. marble machine
9. batch cement mixer
10. hydraulic presses
11. screw conveyors
12. belt conveyors
13. roller conveyors
14. sintering kiln
15. pneumatic or hydraulic cylinders or rams
16. autoclave

The scope of this study does not allow an extensive safety analysis of these pieces of equipment and their potential for mechanical- and electrical-related injuries. Naturally, if the above equipment was not operated or maintained in a safe manner, significant personnel injuries could result.

A process that operates at ambient temperature and has a minimum number of moving parts or mechanical operations would be best from the perspective of industrial hazards. The cast-cement process is the simplest mechanically, operates at a low temperature and is considered to have the least industrial hazard. The FUETAP cement process ranks next due to its use of the low-pressure and low-temperature autoclaves. The simpler high-temperature process follows next, with the mechanically complex processes rated below them. A review of the processes provides the following ranking in terms of least to greatest potential hazard:

1. cast cement
2. FUETAP cement
3. in-can melting
4. joule-heated glass melting
5. basalt glass-ceramic
6. pressed cement
7. glass marble
8. cold-pressed sintered ceramic

CRITICALITY SAFETY

The plutonium concentration normally present in the TRU waste blend is ~0.025 wt% or 0.5 to 0.75 g Pu/L. At these concentrations, the infinite multiplication factor (K_∞) is less than unity, and any quantity of the blend will remain subcritical. However, if plutonium migration and/or nonhomogeneous plutonium waste feeding occurs, it is conceivable that higher plutonium concentrations might result. Two computer calculations were made to simulate the glass and waste mixture and the cement and waste mixture. These calculations assumed a homogeneous waste composition with a varying plutonium density. Spherical geometry and full-water reflection were modeled to optimize conditions for minimum critical mass. The results are plotted in Figure 13 to indicate the mass and concentration of Pu where a criticality may occur for the TRU waste in glass and in cement. Note that below plutonium concentrations of ~500 g Pu/L in glass, infinite quantities of material remain subcritical. For the cement and waste mix, a relative minimum occurs at ~20 g Pu/L and ~40 kg Pu because of the water present in the cement/waste mixture. Both these concentrations and amounts of plutonium are unrealistic for the TRU wastes considered in this study.

The above computer analysis assumed that the plutonium was nonhomogeneous. Should other elements also preferentially migrate, the minimum critical mass would be reduced. The worst conceivable case would be to have all the plutonium mix with all the water in the cement.

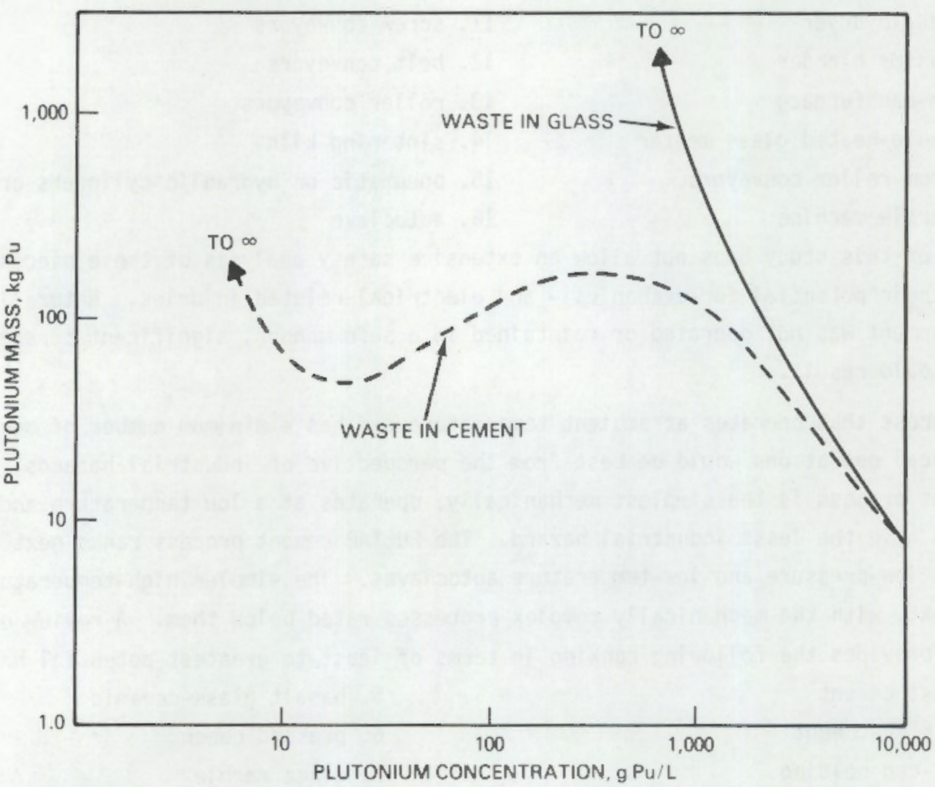

FIGURE 13. Plutonium Critical Mass for Glass and Cement TRU Waste Form

A minimum mass for this case would be ~640 g Pu at 30 g Pu/L. This is a very unlikely situation, however, as all variables would have to be optimally configured.

Even though this analysis shows criticality as an unlikely event, because of the high concentrations and large amount of Pu needed, the immobilization processes should be designed to be critically safe and should be monitored at potential holdup points. Equipment to be monitored for criticality prevention is identified below:

- surge hoppers
- all containers
- joule-heated glass melter
- batch cement mixer
- solids blender.

With the design and monitoring considerations, all the conceptual processes are considered to be critically safe; therefore, all processes are ranked the same with respect to criticality safety.

QUALITY ASSURANCE

The assurance of quality in the waste product is a matter of how easily inspection and testing of the product can be performed to determine its quality. This concept could be easily applied through sampling of the product stream. However, this product stream varies from process to process and so does the ease of obtaining a sample. In the order of ease of sampling, the eight processes fall into three categories of waste product: 1) small marble or pellet; 2) monolith waste form material produced outside the container; and 3) monolith form produced within the container. Of these categories, the glass marble, cement pellet, and ceramic pellet are the easiest to sample. The cast and FUETAP cement waste products, joule-heated glass monolith and basalt glass-ceramic monolith all have a pour stream that can be sampled but which presents difficulties in obtaining a representative sample. The in-can melting glass-monolith process is the most difficult to sample.

Along with the ease of sampling, another consideration in quality assurance is how representative the sample is of the product. This consideration can be answered in terms of the homogeneity of production and the stage of the sample in the production process. Homogeneity will vary slightly from process to process since some processes inherently provide a better mix because of superior blending equipment in the processing scheme. The solids blender used in all the processes (except the cast and FUETAP cement processes) is the initial operation in the development of a homogeneous product. The cast and FUETAP cement processes use the batch mixer to obtain homogeneity. The FUETAP process, however, requires an additional processing step in the autoclave to obtain a quick cure and drying of this cement waste form. This autoclave processing on a smaller sample may not simulate the same effects as experienced on the larger 55 gal size product. A basalt glass-ceramic product requires a heat treatment to simulate the controlled cooldown after the sample is taken; therefore, the grab sample from the melter may not be truly representative of the final form. Large glass castings may also undergo some devitrification during the cooldown cycle, which would not be disclosed from the sample of the glass stream from the melter. Considering the ease of obtaining a representative sample, the various processes and products would be ranked as follows:

1. glass marble
2. pressed cement pellet
3. pressed ceramic pellet
4. cast cement monolith
5. joule-heated glass monolith
6. FUETAP cement monolith
7. basalt glass-ceramic monolith
8. in-can melting glass monolith

The glass marble is believed to be the most homogeneous of the small sample products. The other products, grouped previously, are ranked by product temperature, which affects the complexity of sampling, and by post-sampling processing activities (e.g., FUETAP cement and basalt glass-ceramic), which affect the assurance that the sample is representative of the larger product. A summary of the sampling ease, homogeneity, and sampling temperature factors is shown in Table 14.

ASSESSMENT OF PROCESSES

The total assessment of the processes considers costs, occupational exposure, industrial hazard, criticality safety and quality assurance. Each process is ranked in Table 15 as "1," "2," or "3," as was done earlier for the waste forms in Table 7. From this subjective approach, the cast cement process ranks the highest and is followed closely by the FUETAP cement and joule-heated glass melting processes. The others rank much further behind. Therefore, we recommend that a cast-cement process be the first choice for immobilizing TRU ashes and sludge, and that the FUETAP cement and glass melting processes be kept available as an option if product criteria become stricter and require a waste form that has the properties of a dewatered cement or a glass waste form.

TABLE 14. Quality Control Factors

Product	Sampling Ease	Homogeneity	Sampling Temperature, °C
Glass Marble	1	1	400
Pressed Cement Pellet	1	2	<50
Pressed Ceramic Pellet	1	2	200
Cast Cement	2	2	<50
Joule-Heated Glass Monolith	2	1	1000
FUETAP Cement Monolith	2	2	<50
Basalt Glass-Ceramic Monolith	2	2	1200
In-Can Melting Glass Monolith	3	2	1050

TABLE 15. Ranking of Processes

Process	Overall Cost	Occupational Exposure	Industrial Hazard	Criticality Safety	QA
In-Can Glass Melting	3	1	2	1	3
Joule-Heated Glass Melting	2	2	2	1	2
Glass Marble	3	2	3	1	1
Basalt Glass-Ceramic	2	2	2	1	3
Cast Cement	1	2	1	1	2
Pressed Cement	2	3	2	1	1
FUETAP Cement	1	2	2	1	3
Cold-Pressed Sintered Ceramic	3	3	3	1	2

COMPARISON OF TRU-WASTE IMMOBILIZATION SYSTEMS

This study provides quantitative data for an engineering analysis of TRU waste immobilization systems. Trade-offs in terms of cost, complexity, and product performance are required where a particular immobilization system is selected. The major advantages and disadvantages of each immobilization system are summarized in Table 17 and discussed briefly below:

- Cast Cement. The cast cement process has the lowest process cost because of its inherent simplicity. Cast cement also has the lowest plutonium releases combined with low external surface area, which means cast cement should have the lowest plutonium fractional release rate. The major disadvantages of this process are its higher final waste volume and the residual water content of the waste product.

  Higher volumes do not appear to be a significant cost factor for the defense-related WIPP repository where disposal costs are reported to be lower and insensitive to volume. However, when costs are considered to increase linearly with increasing volume and at the level anticipated for commercial repositories, then disposal costs can exceed the estimated processing savings.

  The high water content of the waste product will allow radiolysis and possibly subsequent pressurization of the drum. Cast cement may also have free water on occasion, and the pore water may be desorbable. These concerns of water content may be resolvable by dewatering of the cement before final sealing of the drum. While the release rates of plutonium are the lowest with this process, the release rates of Na, Ca, and Al are the highest and may indicate longer-term durability problems.

- Pressed Cement. Pressed cements share many of the characteristics of cast cements. The pressed cement process has the advantage of lower water content and overcomes concerns about radiolysis, free water, and desorbable water. Also, the process has low processing costs and can be considered as an alternative to dewatering cast cement. In addition, pellet formation provides an easy opportunity to sample the product, which enhances the ease of quality assurance. However, pellet formation has two negative features. First, the volume of the drummed product increases because of voids in the packed pellets. This feature, although of minor significance for defense wastes, would be very significant for commercial wastes. Second, pellet formation also increases the surface area available for leaching and may allow a high release fraction. It also increases the likelihood of the generation of fine particulates during handling and transportation.

- FUETAP Cement. FUETAP cement also possesses the advantage of lower water content and closely approximates the effects of the pressed cement. The chemical durability is similar to the other cements. As with the other cements, the FUETAP has a relatively low processing cost but the highest among the cement group. The

TABLE 16. Comparison of TRU Immobilization Systems

System	Major Advantages	Major Disadvantages	Comments
Cast Cement	• Lowest process cost • Lowest Pu release	• Higher volume • High water content with radiolysis, free water, and desorbable water concerns • High matrix leachability • Lower mechanical strength	• Dewatering could overcome most product concerns
Pressed Cement	• Quality assurance ease • Overcomes most water concerns with cast cement • Low process cost	• Highest canister volume and repository disposal cost • High matrix leachability	
FUETAP Cement	• Low process cost • Overcomes water concerns of cast cement	• Lowest mechanical strength • QA difficult • High matrix leachability	• Larger-scale quick curing and dewatering method for cements
In-Can Melting • Borosilicate Glass	• Simple process • Low-volume waste	• Highest total cost • QA difficult	• Development of less expensive canister and 8-h/day operations would reduce costs
Joule-Heated Melter • Borosilicate Glass • Aluminosilicate Glass	• Low-volume waste • Simple canister	• High labor cost for continuous operations	• Commercial repository costs can make system lower and competitive in cost
Joule-Heated Melter • Glass marbles	• QA easier	• Higher process cost and complexity • Higher surface area for leaching	
Basalt Glass-Ceramic	• High thermal stability • Highest mechanical strength • Low matrix leachability	• High Pu release • QA difficult • High process cost	
Cold-Pressed Sintered Ceramic	• Good thermal stability • High mechanical strength • QA ease	• High volume • Highest Pu release in brine solutions	

FUETAP cement has the highest waste loading (~40 wt%) of the cement products tested, which causes it to generate the lowest volumes of the cements. This is not to say that the cast or pressed cement cannot achieve this waste loading. This higher waste loading does apparently cause a reduction by a factor of two in the mechanical strength when compared with the cast product.

The FUETAP method does appear viable as a larger-scale (55 gal drum size) processing option in reducing the cure time and lowering the water content of cement waste forms. These advantages may outweigh other negative factors associated with quality assurance and additional processing costs.

- In-Can Melting--Borosilicate Glass. In-can melting has potentially the highest cost because of the requirements of high-temperature/high-strength metallic canisters. The development of a lower-cost container would make the process cost competitive with other processes. The process does offer process simplicity and has some potential for set-up and operation on an 8 h/day shift basis, which could further lower process costs. Product formation within a thick metallic canister makes quality assurance based on product sampling difficult. However, volume of the glass product is low and offers incentives for processing commercial wastes on the basis of the lower volume and generally good product properties.

- Joule-Heated Melting--Borosilicate or Aluminosilicate Glass. With this process, the melting of waste glass in a separate melter allows increased flexibility of process temperature and, therefore, in the composition of the glass. The process also allows a low-cost container to be used and permits the container to be more completely filled, which provides the lowest volume product. However, continuous operation requirements of the process make for high labor costs, and the need to regularly replace the melters increases maintenance costs.

  As noted for in-can melting, joule-heated-melter glass products generally have good product properties. While it was anticipated that an aluminosilicate glass would have improved properties compared to borosilicate glass, the observed differences between these two waste forms are not large. Compared to borosilicate glass, the aluminosilicate waste product does have improved thermal stability above 400°C and potentially some minor improvement in strength. However, the plutonium release fractions of the two glasses are comparable. In terms of leaching, borosilicate glass has better behavior in silicate waters, and aluminosilicate glass has better behavior in brine.

- Joule-Heated-Melting--Glass Marbles. The forming of glass into marbles allows for easier quality control inspection of the product, but like pelletized cement this waste form increases the volume, the surface area, processing and total costs compared to monolithic products. The glass-marble system only would have a significant advantage if detailed product quality control were required.

- Basalt Glass-Ceramic. The thermal heat treatment of a specific glass composition to form a basalt glass-ceramic increases the thermal stability of the final product. Such treatment also forms a mechanically strengthened product and reduces the matrix leachability. Plutonium release, however, may be increased. Sampling of the final product may be difficult, and the product can be expected to vary with radius because thermal gradients during heat treatment will affect the crystallization behavior. The extra thermal treatments and processing increase process costs.

- Cold-Pressed Sintered Ceramic. The sintered-ceramic product has high thermal stability and mechanical strength. Like glass marbles and pressed cement, ceramic pellets would offer ease of quality assurance but have higher volume and higher surface area. High surface area, in conjunction with high measured plutonium leach rates, leads to the highest calculated plutonium release fractions.

The TRU immobilization system preferred for a particular application is dependent on the repository medium and cost assignments. For defense wastes in salt, we believe that a cast cement, FUETAP cement, or pressed cement process would be an appropriate selection (in that order). For commercial wastes, it would appear that a borosilicate or aluminosilicate glass or FUETAP or cast cement would be most appropriate, depending on whether the medium were hard rock or salt. These glass forms would have lower total costs in the higher-cost hard-rock (basalt or granite) repositories, in addition to improved properties, compared to the cement waste forms. The cement waste forms, however, again show a slight economic advantage in the lower-cost salt repositories. This illustrates the need to review the waste form criteria, volume generation differences, and total system economics before selecting a TRU immobilization system.

REFERENCES

Andrews, W. B., B. M. Cole, R. L. Engel, and J. M. Olylear. 1982. Defense Waste Transportation Cost and Logistics Studies. PNL-3721, Pacific Northwest Laboratory, Richland, Washington.

Bida, G., and D. R. MacKinzie. 1982. Nuclear Waste Management Technical Support in the Development of Nuclear Waste Form Criteria for the NRC. Task 2. Alternative TRU Technologies, Vol. II. NUREG-CR-2333/BNL-NUREG-51458, Brookhaven National Laboratory, Upton, New York.

Brown, C. M. 1982. Economic Evaluation of Volume Reduction for Defense Waste. RFP-3245, Rockwell International Energy Systems Group, Rocky Flats Plant, Golden, Colorado.

Bryan, G. H. 1981. Characterization of Transuranium-Contaminated Solid Wastes Residues. PNL-3776, Pacific Northwest Laboratory, Richland, Washington.

Crisler, L. R. 1980. A Preliminary Assessment of Nine Waste-Form Products/Processes for Immobilizing Transuranic Wastes. RFP-3131, Rockwell International Energy Systems Group, Rocky Flats Plant, Golden, Colorado.

Department of Energy (DOE). 1979. Technology for Commercial Radioactive Waste Management, Volume 5. DOE/ET-0028, Washington, D.C.

Dole, L. R., et al. 1982. "Cementiteous Radioactive Waste Hosts Formed Under Elevated Temperatures and Pressures (FUETAP Concretes)." In Scientific Basis for Nuclear Waste Management, Vol. 6. SiV. TOPP ed. North Holland, New York.

Eakes, R. G., et al. 1980. "TRU Waste Transportation Package Development." In Sixth International Symposium--Packaging and Transportation of Radioactive Materials, pp. 114-117. Berlin (West), November 10-14, 1980, Federal Republic of Germany.

Energy Research and Development Administration (EROA). 1976. Alternatives for Managing Wastes from Reactors and Post-Fission Operations in the LWR Fuel Cycle. ERDA-76-43, Washington, D.C.

Federal Register, July 1981, Proposed Rules Vol. 36, No. 130.

Flinn, J. E., P. V. Kelsey, W. C. Seymour, and R. L. Tallman. 1979. Summary of FY-1979 Materials Support Studies for SPI-Migration and Immobilization. EG&G-FM-5041, EG&G, Inc., Idaho Falls, Idaho.

Hench, L. L., chairman. 1979. The Evaluation and Review of Alternate Waste Forms for Immobilization of High-Level Radioactive Wastes. Alternative Waste Form Peer Review Panel, DOE/TIC-10228, Oak Ridge, Tennessee.

Iler, R. K. 1979. The Chemistry of Silica. John Wiley and Sons, New York.

Irby, H. H. , editor. 1980. Report of the Steering Committee on TRU Waste Acceptance Criteria for the Waste Isolation Pilot Plant. WIPP-DOE-069, Westinghouse Electric Corp.

Johnson, E. R., and Associates. 1980. Preliminary Evaluation of Alternative Waste Form Solidification Processes, Volume II: Evaluation of the Processes. PNL-3477, Pacific Northwest Laboratory, Richland, Washington.

Kosiewicz, S. T., B. L. Barraclough and A. Zerwekh. 1979. Studies of Waste Storage Conditions Expected in the Waste Isolation Plant (WIPP). LA-7931-Pr, Los Alamos Scientific Laboratory, Los Alamos, New Mexico.

Lewis, E. L. and Robert F. Herbert, Jr. 1981. Pelletized Waste Form Demonstration Program: October 1980-March 1981. MLM-2827, Mound Facility, Miamisburg, Ohio.

Lokken, R. O. 1978. A Review of Radioactive Waste Immobilization in Concrete. PNL-2654, Pacific Northwest Laboratory, Richland, Washington.

McVay, G. L., and L. R. Pederson. 1981. "Effect of Gamma Radiation on Glass Leaching." In J. Am. Cer. Soc. 64:34.

Mendel, J. E., et al. 1981. A State-of-the-Art Review of Materials Properties of Nuclear Waste Forms. PNL-3802, Pacific Northwest Laboratory, Richland, Washington.

Molecke, M. A. 1979. Gas Generation from Transuranic Waste Degradation: Data Summary and Interpretation. SAND 79-1245, Sandia Laboratories, Albuquerque, New Mexico.

Moore, J. G., E. Newman, L. R. Dole, and S. Katy. 1981. Alpha Radiolysis of FUETAP Concrete. CONF-81103, ANS Winter Meeting 1981, San Francisco, California.

Nuclear Energy Services, Inc. (NES). 1980. Study of Postimmobilization Disposition. Prepared for U.S. Department of Energy for WNYNSC, NES.

Nuclear Regulatory Commission (NRC). 1979. Solidification of High-Level Radioactive Wastes. NUREG/CR-0895, National Academy of Sciences, Washington, D.C.

Platt, A. M., and J. A. Powell, editors. 1980. Nuclear Waste Management Quarterly Progress Report, October Through December 1979. PNL-3000-4, Pacific Northwest Laboratory, Richland, Washington.

Roberts, F. P. 1981. An Assessment of Radiation Effects in Defense Transuranic Waste Forms. PNL-3913, Pacific Northwest Laboratory, Richland, Washington.

Roberts, F. P., G. H. Jenks and C. D. Bopp. 1976. Radiation Effects in Solidified High-Level Wastes--Part I, Stored Energy. BNWL-1944, Pacific Northwest Laboratory, Richland, Washington.

Roberts, F. P., R. P. Turcotte and W. J. Weber. 1981. Materials Characterization Center Workshop on the Irradiation Effects in Nuclear Waste Form, Summary Report. PNL-3588, Pacific Northwest Laboratory, Richland, Washington.

Ross, W. A., J. M. Rusin, and J. L. McElroy. 1979. "Process for Production of Alternative Waste Forms." In Proceedings of Waste Management 1979. R. G. Post, editor, University of Arizona, Tucson, Arizona.

Ross, W. A., R. P. Turcotte, J. E. Mendel and J. M. Rusin. 1979. "A Comparison of Glass and Crystalline Waste Materials." In Proceedings Ceramics in Nuclear Waste Management, CONF-790420, April 1979.

Rusin, J. M. 1980. A Review of High-Level Waste Form Properties. PNL-3035, Pacific Northwest Laboratory, Richland, Washington.

Rusin, J. M., W. J. Gray and J. W. Wald. 1979. Multibarrier Waste Forms, Part II: Characterization and Evaluation. PNL-2668-2, Pacific Northwest Laboratory, Richland, Washington.

Scheffler, K., and U. Riege. 1977. Investigations on the Long-Term Radiation Stability of Borosilicate Glasses Against Alpha Emitters. KFK 2422, Kernforschungszentrum, Karlsruhe, Germany.

Schulz, W. W., et al. 1980. Preliminary Evaluation of Alternative Forms for Immobilization of Hanford High-Level Defense Wastes. RHO-ST-32, Rockwell Hanford, Richland, Washington.

Shefelbine, H. C. 1978. Preliminary Evaluation of the Characteristics of Defense Transuranic Wastes. SAND-78-1850, Sandia Laboratories, Albuquerque, New Mexico.

Stone, J. A. 1977. Evaluation of Concrete as a Matrix for Solidification of Savannah River Plant Waste. DP-1448, Savannah River Laboratory, Aiken, South Carolina.

Stone, J. A., S. T. Goforth, Jr., and P. K. Smith. 1979. Preliminary Evaluation of Alternative Forms for Immobilization of Savannah River Plant High-Level Wastes. DP-1545, E. I. du Pont de Nemours & Company, Savannah River Laboratory, Aiken, South Carolina.

Timmerman, C. L. 1980. Description of Processes for Immobilization of Selected Transuranic Wastes. PNL-3479, Pacific Northwest Laboratory, Richland, Washington.

Wald, J. W., R. O. Lokken, J. W. Shade, and J. M. Rusin. 1980. Comparative Waste Forms Study. PNL-3516, Pacific Northwest Laboratory, Richland, Washington.

Weber, W. J., and F. P. Roberts. 1981. "Radiation Effects." In State-of-the-Art Review of Materials Properties of Nuclear Waste Forms. J. E. Mendel, ed., PNL-3802, Pacific Northwest Laboratory, Richland, Washington.

Weber, W. J., R. O. Turcotte, L. R. Bunnell, F. O. Roberts, and J. H. Westsik, Jr. 1979. "Radiation Effects in Vitreous and Densified Simulated Waste Glass." In Ceramics in Nuclear Waste Management, T. D. Chickalla and J. E. Mendel, eds., pp. 294-299, CONF-790420, National Technical Information Service, Springfield, Virginia.

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