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ABSTRACT

Seven candidate waste forms, developed under the direction of the Department of Energy's National High-Level Waste (HLW) Technology Program, were evaluated as potential media for the immobilization and geologic disposal of high-level nuclear wastes. The evaluation, completed on August 1, 1981, combined preliminary waste form evaluations conducted at Department of Energy (DOE) defense waste-sites and at independent laboratories, peer review assessments, a product performance evaluation, and a processability analysis. Based on the combined results of these four inputs, two of the seven forms, borosilicate glass and a titanate-based ceramic, SYNROC, were selected as the reference and alternative forms, respectively, for continued development and evaluation in the National HLW Program. The borosilicate glass and ceramic forms were further compared during FY-1982 on the basis of

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risk assessments, cost comparisons, properties comparisons, and conformance with proposed regulatory and repository criteria. Both the glass and ceramic forms are viable candidates for use at DOE defense HLW sites; they are also candidates for immobilization of commercial reprocessing wastes. This paper describes the waste form screening process, discusses each of the four major inputs considered in the selection of the two forms in 1981, and presents a brief summary of the comparisons of the two forms during 1982 and the selection process to determine the final form for immobilizing defense HLW at the Savannah River Plant (SRP).

INTRODUCTION

Background

As specified by Federal law (DOE/NE-0008, 1980), DOE has responsibility for the isolation of U.S. nuclear wastes from the human environment. Consequently, DOE is conducting comprehensive programs for the long-term management of these wastes. The overall objective of these programs is to provide the required confinement of radioactive wastes 1) with minimum reliance on future human surveillance, and 2) in a way that ensures a high degree of isolation from the human environment during the period of potential radiation hazard.

The DOE long-term waste management efforts are organized by nuclear waste category, specifically high-level waste (HLW), low-level waste (LLW), transuranic waste (TRU), and airborne waste. Specific DOE sites were designated as "Lead Offices" to coordinate the development of disposal technology for each waste type. Responsibility for coordinating the long-term management program for HLW which has been generated as a byproduct of national defense activities, or which may be generated from reprocessing of commercial spent power reactor fuels, has resided with DOE's Savannah River Operations Office (DOE-SR) (DOE/SR-WM-79-3, 1981). Technical support and guidance to DOE-SR has been provided by the Savannah River Laboratory (SRL), operated by E. I. du Pont de Nemours and Company.

Program Strategy

The HLW Technology Program was established in FY-1979 to develop the technology for immobilizing HLW into solid waste forms which would provide highly efficient barriers against radionuclide release to the environment. The waste form would be the innermost component in a waste package which was being developed in the National Waste Terminal Storage (NWTS) Program for ultimate disposal in a geologic repository. The waste form development effort in the HLW Technology Program has progressed in a parallel and co-ordinated manner with the NWTS Program.

The strategy for HLW immobilization involves 1) the development of technology to support the choice of an isolation system for each DOE defense HLW site and for any commercial HLW site, 2) development of candidate waste forms and production processes, and 3) support of R&D activities required to construct and operate immobilization facilities for defense HLW.

In response to recommendations by the Interagency Review Group (IRG) on Nuclear Waste Management (TID-29442, 1979) and the National Academy of Sciences (NUREG/CR-0895, 1979) a variety of waste forms were defined and developed to permit flexibility in the immobilization and terminal storage systems. Milestones for the national program tasks related to waste form development activities were established as follows:

1. The development and characterization of all viable candidate forms during FY-1979 to FY-1981.
2. The selection of two waste forms at the end of FY-1981 for further development.
3. Selection of one waste form before the end of FY-1983 for the Defense Waste Processing Facility (DWPF), to be located at the Savannah River Plant (SRP), as the first defense-waste immobilization facility in the United States.

On August 1, 1981, a screening comparison of seven candidate waste forms was completed from which two forms were selected for further development and evaluation in FY-1982. Four major inputs served as the bases for the selection of the two forms:

1. Preliminary evaluations conducted to determine the preferred forms for immobilization and geologic disposal of DOE defense and commercial HLW.
2. A series of annual independent reviews, by the Alternative Waste Form Peer Review Panel, of the waste forms being developed in the National HLW Technology Program.
3. A quantitative evaluation of waste form product performance based on comparative characterization data available on or before August 1, 1981.
4. A quantitative evaluation of the processes required to produce the seven candidate forms.

During FY-1982 a final selection process was initiated to choose one waste form for the DWPF. Factors considered in the final comparison of the two forms, in addition to those presented in this paper, were further product performance comparisons, comparative risk assessments, cost analyses, and assessments of how each form conforms with proposed regulatory and repository criteria. The selections of waste forms for immobilization of HLW at other DOE defense sites (Hanford and Idaho), as well as for potential commercial HLW, will be made sequentially.

This paper describes the FY-1981 waste form screening process and selection of the final two waste forms, including descriptions of the seven candidate forms and discussions of the four major decision inputs. Also, a brief summary is presented on the comparison of these two forms and the selection process during FY-1982 to determine the waste form for the DWPF.

CANDIDATE HIGH-LEVEL WASTE FORMS

Since the inception of the national program in May 1979, seventeen candidate waste forms have been developed and characterized by fourteen participating contractors as potential media for the geologic disposal of high-level nuclear wastes (Table 1). The broad-based research efforts have been conducted at the waste processing sites, national laboratories, industrial laboratories, and universities. Research and development for ten of the forms was terminated at various stages of the program based on preliminary reviews which raised technical concerns about the viability of these forms for geologic disposal of wastes. Following continued development and characterization, the seven remaining forms (Table 2) were evaluated to select, at the end of FY-1981, two forms for further development and evaluation in support of the DWPF project and in support of the overall program goal of immobilizing all HLW in the United States.

TABLE I

CANDIDATE WASTE FORMS CONSIDERED FOR GEOLOGIC DISPOSAL OF HIGH-LEVEL WASTE

Waste Form	Developer/Contractor
Borosilicate Glass	Savannah River Laboratory Pacific Northwest Laboratory
High-Silica Glass	Catholic University of America NPD Nuclear Systems, Inc.
Phosphate Glass	Pacific Northwest Laboratory Brookhaven National Laboratory
Clay Ceramic	Rockwell Hanford Operations Pacific Northwest Laboratory
Glass Ceramic	Idaho Chemical Processing Plant
Tailored Ceramic	Rockwell International Pennsylvania State University
SYNROC	Lawrence Livermore National Laboratory Argonne National Laboratory North Carolina State University
Titanate Ion Exchanger	Sandia National Laboratory
Stabilized Calcine	Idaho Chemical Processing Plant
Pelletized Calcine	Idaho Chemical Processing Plant
Normal Concrete	Savannah River Laboratory Oak Ridge National Laboratory Pennsylvania State University
Hot-Pressed Concrete	Pennsylvania State University
FUETAP Concrete	Oak Ridge National Laboratory
Matrix Forms	Pacific Northwest Laboratory Argonne National Laboratory
Coated Sol-Gel Particles	Oak Ridge National Laboratory
Cermet	Oak Ridge National Laboratory
Disc-Pelletized Coated Particles	Pacific Northwest Laboratory Battelle Columbus Laboratory

TABLE 2
SEVEN CANDIDATE WASTE FORMS
EVALUATED FOR GEOLOGIC DISPOSAL OF HIGH-LEVEL WASTES

Waste Form	Developer/Contractor
Borosilicate Glass	Savannah River Laboratory Pacific Northwest Laboratory
SYNROC	Lawrence Livermore National Laboratory Argonne National Laboratory North Carolina State University
Tailored Ceramic	Rockwell International Pennsylvania State University
High-Silica Glass	Catholic University of America NPD Nuclear Systems, Inc.
FUETAP Concrete	Oak Ridge National Laboratory
Coated Sol-Gel Particles	Oak Ridge National Laboratory
Glass Marbles in a Lead Matrix	Pacific Northwest Laboratory

The seven final forms are described below. Because performance data were obtained from forms containing simulated SRP waste, the product and process descriptions apply specifically to immobilizing SRP HLW. Other waste types may affect the specific waste form compositions and possibly processing as well. In the production processes for all of the waste forms except the crystalline ceramics, aluminum is removed from the waste prior to solidification. Aluminum, a desirable component for the ceramics, is not removed from the waste when producing these forms. Because of this difference, the waste loadings in the descriptions of the crystalline ceramic forms would be somewhat lower on a comparable basis with the other forms.

Borosilicate Glass

Borosilicate glass (Plodinec et al., 1982) is the best developed of all the candidate waste forms for high-integrity radionuclide containment. Typically, a borosilicate waste glass consists of about 20 to 35 wt % waste oxides (with Al removal), 40 to 50 wt % silica, 5 to 10 wt % boron oxide and 10 to 15 wt % alkali oxides, plus other additives. The reference process for the DWPF involves feeding a slurry waste stream and glass additives to a continuous joule-heated glass melter, from which the waste glass is poured into 2 ft diameter by 10 ft high canisters approximately 80% full. Melter temperatures for the above compositions range from 1050 to 1150°C.

High-Silica Glass

High-silica natural glasses (obsidians and tektites) have persisted for long periods in both terrestrial and lunar environments. However, these glasses are formed at temperatures of 1600 to 1800°C — high enough to vaporize ruthenium and cesium radionuclides present in HLW. The key to the development of this form has been the lower-temperature Porous Glass Matrix process (Macedo et al., 1979), which provides a means for fabricating high-silica waste forms at 1200°C by sintering an intimate mixture of porous glass frit and waste calcine. The form is produced by a rising level, in-can sintering process in which porous glass powder (10 to 300 μm pores) is blended with waste calcine and charged under vacuum into an inductively heated canister. Pores in the glass grains collapse at 600 to 900°C to trap volatiles, followed by sintering of the frit and waste particles at about 1100°C. Typically, the high-silica waste glass contains from 50 to 60 wt % silica at a waste loading of approximately 20 to 30 wt % waste oxides (with Al removal).

SYNROC

SYNROC (Ringwood et al., 1981) is a densely consolidated, titanate-based, polyphase crystalline ceramic form in which radionuclides are incorporated as dilute solid solutions in the crystal lattices of perovskite, zirconolite, and either hollandite, nepheline, or pollucite. "SYNROC-D" (Campbell et al., 1982), a variation of this waste form developed for the immobilization of SRP high-level wastes, is formed from tailored additions of TiO_2, SiO_2, CaO, and perhaps other oxides to the waste sludge. In the reference SYNROC process, an intimately blended mixture of waste and additives is calcined, tamped into a canister, preheated under vacuum, and then hot isostatically pressed (HIPped) at 1100 to 1200°C and 25,000 psi, to synthesize the mineral phases. Waste loadings of 50 to 70 wt % (as oxides, without Al removal) have been achieved. Potentially, the form also could be produced via hot pressing or sintering.

Tailored Ceramic

Tailored ceramic (Harker et al., 1981) is a densely consolidated, alumina-based, polyphase crystalline ceramic material developed for SRP waste. It is formed by tailored additions of Al_2O_3, SiO_2, rare earths, and perhaps other oxides to the waste sludge. The waste components are incorporated into stable crystalline phases such as uraninite, magnetoplumbite, spinel, corundum, nepheline, and perovskite. The reference process for production of the tailored ceramic waste form is essentially identical to the SYNROC-D process. Waste loadings of 50 to 90 wt % (as oxides, without Al removal) have been achieved, depending on waste composition.

FUETAP Concrete

Concretes formed under elevated temperatures and pressures (FUETAP concretes) (Moore et al., 1981) are prepared from common Portland cements, fly ash, sand, clays, and waste calcines. The production process for this form involves dry blending the cement additives with the waste calcines, followed by wet blending with the addition of water. This mixture is poured into 2 ft diameter by 10 ft canisters and cured under mild autoclave conditions (100°C, 1 atm steam). Subsequently, the concrete is dewatered under vacuum at 250°C for 24 hours. Typically, the FUETAP concrete waste form has a 20 wt % waste loading (with Al removal).

Coated Sol-Gel Particles

Coated sol-gel particles (Angelini et al., 1981) are 750-micron zirconia-based ceramic spheres enclosed in three separate coatings. The inner coating is low-density silicon carbide; the middle coating is high-density silicon carbide; and the outer coating is high-density pyrolytic carbon. In the sol-gel process, matrix elements are added to a solution of waste sludge dissolved in nitric acid, followed by urea addition to neutralize excess acid. A chilled solution (about 0°C) of HMTA (hexamethylenetetraamine) is added to the urea mixture and fed through a vibrating nozzle into an immiscible liquid at 55°C to form the sol-gel spheres. After aging, rinsing, and washing, the spheres are transferred to fluidized beds for application of the coating layers by chemical vapor deposition (CVD). The coated spheres would then be packaged together with an inorganic binder (probably an aluminate or silicate-aluminate compound) into a canister, to obtain the final waste form. A canister would contain approximately 10^8 spheres. Single-uncoated particle waste loadings from 70 to 90 wt % (with Al removal) have been achieved. The net canister waste loading, considering coating volumes and canister void spaces, is about half the single-particle waste loading.

Glass Marbles in a Lead Matrix

Glass marbles in a lead matrix (Jardine and Steindler, 1978) provide an alternative to the production of large canisters of glass. Molten glass similar to the borosilicate glass composition, with approximately 28 wt % waste calcine, is cast into marbles 1.3 cm in diameter. The marbles are batch-loaded into a canister and the void space is filled with low-melting lead-tin alloy shot. The canister is then zone heated in an induction furnace to melt the shot and provide a continuous metal matrix which solidifies from the bottom upward. Only about 60% of the volume of a 2 ft diameter by 10 ft canister would be occupied by the glass marbles, giving a significantly lower net canister waste loading than for a canister of glass.

PRELIMINARY WASTE FORM EVALUATIONS

Introduction

Independent evaluations were conducted at each of the DOE defense HLW sites, Savannah River (near Aiken, South Carolina), Hanford (near Richland, Washington), and the Idaho Chemical Processing Plant (near Idaho Falls, Idaho), to determine the preferred solid forms for immobilization of the existing HLW at each location. The individual evaluations rated from eight to nineteen candidate waste forms according to process complexity and potential product performance. All of the evaluations weighted product and process equally. Similar assessments were sponsored by the Pacific Northwest Laboratory (PNL) for nuclear wastes from potential reprocessing of commercial power reactor spent fuel. The first of two studies investigated product durability, and the second evaluated potential production processes.

Defense Waste Form Evaluations

Savannah River. A study (Stone et al., 1979) conducted by SRL evaluated eleven waste forms (borosilicate glass as the reference form, plus ten alternatives) that had been proposed for solidification of SRP HLW. The goal of this preliminary evaluation was to select a limited number of forms for more detailed studies. At that time, most of the waste forms lacked detailed experimental data for the factors upon which they were evaluated. Information on each form was obtained from published articles and reports, supplemented by discussions with the proponents of the forms and technical judgments made to develop the waste form comparison. The major conclusions of this study were:

- Borosilicate glass had the highest overall score of all the forms considered.
- Six waste forms potentially have better product properties than borosilicate glass.
- Coated waste forms had the highest product scores but were poorest in process characteristics.
- Concrete waste forms had the poorest product scores, but were rated highly on process characteristics.

Hanford. At the Hanford site, a preliminary evaluation (Schulz et al., 1980) of solid waste forms for immobilization of Hanford high-level defense wastes was conducted using published articles, reports, and conference proceedings. Additional data were obtained from detailed discussions with scientists and engineers involved in developing the various forms. Nineteen waste forms (including the eleven forms considered at Savannah River) were evaluated and compared to determine their applicability for immobilization of Hanford salt cake, residual liquid, sludges, and mixtures thereof. The objective was to identify those waste forms and processes meriting further research and development to qualify them for consideration in the final selection of waste forms for disposal of Hanford HLW.

Two general conclusions were made from the final waste form selections and rankings.

- Borosilicate glass is a viable waste form for fixation of all types of Hanford HLW.
- No one waste form was judged distinctly superior to any other. Waste forms typically exhibited either high product and low process scores, or low product and high process scores. Borosilicate glass, however, had good rating values for both product and process properties.

Idaho Chemical Processing Plant (ICPP). An evaluation was performed by an independent review panel (Post, 1981) chartered to provide Exxon Nuclear Idaho Company with objective recommendations on the most promising waste forms for final disposal of calcined ICPP wastes. Four categories of waste forms were evaluated by the panel: glasses, glass ceramics, tailored ceramics, and composites. Composites consisted of one of the previous forms or calcine, in granular size, in a matrix material such as concrete or metal.

The panel concluded that although stabilized calcine in a concrete matrix had a slightly higher rating, borosilicate glass was the best choice because of its acceptable product performance, its well developed process, and its potential for expeditious implementation. The panel also concluded that the matrix form could prove acceptable. However, they felt that development was not sufficiently advanced to provide adequate confidence in the concept.

Commercial Waste Form Evaluations

Two studies were sponsored by PNL to assess potential commercial HLW forms and processes. The first study (Wald et al., 1980) assessed the potential product durability of candidate commercial waste forms based on volatility, mechanical strength, and leaching tests. Additionally, bulk property, phase composition, and microstructural examinations were conducted. The second study (E. R. Johnson Associates, Inc., 1980) assessed various processes in the context of the feasibility of establishing practical production-scale facilities in a highly radioactive environment requiring remote operation and maintenance.

Glass, glass ceramic, and Supercalcine ranked highest based on product durability, while glass, glass ceramic, and concrete processes were rated as the top three production processes.

Conclusions

The preliminary evaluations involved a significant amount of technical judgment and were partially based on qualitative or nonstandardized quantitative data. Although continuing research and development has refined and changed the forms and processes, these early evaluations were valuable screening studies. They identified unacceptable waste forms and directed development efforts towards the most promising alternatives for waste immobilization. While significant revisions in the waste forms and processes have been made, these changes have not been so drastic as to negate the results and conclusions of the preliminary studies. The most recent evaluations of product performance and waste form processability for the seven candidate waste forms were based on current comparative data, as discussed in the following sections of this paper.

The results of the preliminary waste form evaluations exhibited several important and consistent trends.

- In each evaluation by the DOE defense-sites, borosilicate glass was selected as a viable and preferred form for immobilization of HLW. Similarly, glass was rated highest in both commercial waste form evaluations.
- Generally, waste forms had either low product ratings and high process ratings, or high product ratings and low process ratings. Glass forms always had high process ratings and good product ratings.
- The second most preferred form was a ceramic, usually a crystalline ceramic. These forms exhibited product characteristics potentially superior to glass forms, but required comparatively complex processes.
- Concrete forms generally had good process ratings (although lower than those of glass), but demonstrated some of the poorest product performance characteristics.

ALTERNATIVE WASTE FORM PEER REVIEW PANEL EVALUATIONS

Introduction

The second major input to the waste form evaluation and selection process was a series of annual independent assessments of the waste forms being developed in the National HLW Technology Program. These reviews were conducted by the Alternative Waste Form Peer Review Panel chaired by Dr. L. L. Hench of the University of Florida. The panel is composed of eight scientists and engineers representing independent non-DOE laboratories from industry, government, and universities; and the disciplines of materials science, ceramics, glass, metallurgy, and geology. Its charter has been to review and evaluate the relative scientific merits and engineering practicality of candidate waste forms for geologic disposal of high-level radioactive wastes. The panel has convened annually since August 1979. To facilitate each assessment the most current data, reports, and other information for each waste form were provided by the waste form developers for distribution to the panel members. For the second and third evaluations by the panel, program reviews were conducted at which the waste form developers presented their most recent results from ongoing research and development efforts.

Evaluation Numbers 1 and 2

The first two reviews (DOE/TIC 10228, 1979 and DOE/TIC 11219, 1980) of candidate waste forms were conducted in August 1979 and May 1980. The first review evaluated the relative merits and potential of eleven candidate forms; the second considered fifteen forms. More waste forms were considered in the second review because the general category of "multibarrier forms" was expanded to include five specific composite forms. The 1980 review also assessed four approaches to presolidification processing. The waste form rankings presented in the reports of the 1979 and 1980 reviews were accompanied by discussions of the relative strengths and weaknesses of the candidate forms and recommendations for future program direction. These reviews and recommendations were instrumental in selecting the seven candidate waste forms considered in the National Program. These seven remaining waste forms were assessed in the panel's third review in May 1981.

Evaluation Number 3

The third review (DOE/TIC 11472, 1981) by the Peer Review Panel in May 1981, considered the remaining seven forms under study by DOE: borosilicate glass, high-silica glass, SYNROC, tailored ceramic, coated sol-gel particles, FUETAP concrete, and glass marbles in a lead matrix. A thermal-spray-coated marble form was also evaluated by the panel, although this form was not considered in the final evaluation. The thermal spray concept was investigated briefly by PNL as an alternative glass or ceramic marble coating process.

Prior to panel deliberations, developers presented the most recent waste form information to the panel. Progress reports describing details of waste form development, characterization, testing procedures, and data for a comparison of physical properties, chemical durability, radiation stability, and thermal stability were provided to the panel several weeks before the review. Reports summarizing the most recent data obtained from ongoing research and development efforts were provided to the panel at the time of the presentations. These uniform reporting procedures facilitated the comparison of waste form performance parameters. The 1981 review provided a consensus ranking of the candidate forms, shown in Table 3, based on their relative merits as determined by the panel. This ranking, the discussions of the forms' advantages and disadvantages, and the recommendations for future research and development were considered in selection of the final two candidate waste forms.

TABLE 3
RELATIVE RANKING OF CANDIDATE WASTE FORMS BY
ALTERNATIVE WASTE FORM PEER REVIEW PANEL - MAY 1981

1) Borosilicate Glass
2) SYNROC
3) High-Silica Glass
4) Tailored Ceramic
5) Coated Sol-Gel Particles
6) FUETAP Concrete
7) Glass Marbles in a Lead Matrix
8) Thermal Spray Coatings

The panel's conclusions for the two top rated forms are summarized below.

- Borosilicate glass was ranked as the preferred form for geologic disposal of HLW. This conclusion was based on factors such as a relatively simple processing operation, evidence of process insensitivity to waste stream variability, routine operation of a full scale remote facility in France, good leach resistance, relative insensitivity to radiation, and impact resistance equivalent to the other candidate forms.
- The panel felt that SYNROC, the second ranked form, was the best characterized and understood of the forms other than borosilicate glass. They concluded that radiation effects should be minimal, based on data that demonstrate the radiation insensitivity of natural analogs of the synthetic mineral phases present in SYNROC. Also, potential process flexibility had been demonstrated (i.e., hot pressing, sintering, and hot isostatic pressing).

WASTE FORM PRODUCT PERFORMANCE EVALUATION

Introduction

To provide a quantitative assessment of product performance on a comparable basis for the seven candidate waste forms, an evaluation of several key waste form properties was performed by SRL based on a rating method developed by a DOE Interface Working Group (IWG) on HLW Form Selection Factors. The IWG was comprised of representatives from DOE waste management programs on HLW, isolation, transportation, and transuranic waste. It was established to provide a broad spectrum of expert opinion from which a consensus could be reached on the relative importance of waste form properties, and to develop a method for translating performance data into waste form ratings. The rating method used to compare the seven forms was modeled after the IWG method, with some modifications to account for unavailable characterization data. A detailed explanation of the method developed by the IWG is described elsewhere (DOE/TIC 11612, 1982).

Product Performance Evaluation Method

The IWG identified five categories of waste form properties to be considered in judging the relative merits of candidate waste forms based on product performance. These were leaching (leach rates and solubilities), thermal stability, radiation stability, waste loading, and mechanical stability. These properties were quantified, weighted by relative importance, and organized into a numerical rating scheme representing the three major time periods in the waste disposal system:

1. The Operational Period, which includes interim storage, transportation, and handling.
2. The Thermal Pulse Period, during which short-lived fission products would still generate significant decay heat. This period was nominally assumed to last about 1000 years after emplacement of the waste in a repository.
3. The Geologic Period, after the short-lived radionuclides have decayed.

Radiation stability, thermal stability, and solubility were not included in this evaluation because of an insufficient comparative data base. Therefore, a hierarchy was used which contained the waste form properties of waste loading, mechanical stability, and leaching (Figure 1). The rating scheme used a linear, additive scoring model with constant criteria weights

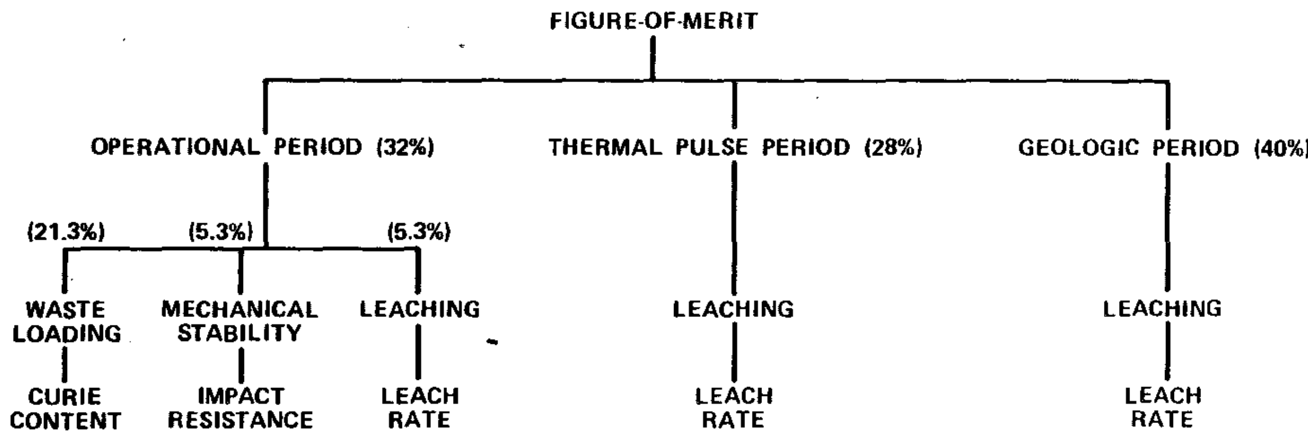

Fig. 1 Figure-of-Merit Hierarchy Used in the High-Level Waste Form Product Performance Evaluation

and nonlinear value functions. When this scheme was used with the waste form performance data, a "Figure-of-Merit" (FOM) rating was generated for each form. These ratings were then compared, giving a relative ranking of waste form performance.

Waste Form Characterization Data

Characterization data for waste loading, mechanical stability, and leaching were provided by the waste form developers, Argonne National Laboratory (ANL), the Materials Characterization Center (MCC), and SRL. Only data available on or before August 1, 1981, the final date for completion of the waste form comparison, were considered in the product performance evaluation. The significance of these data and the methods by which they were obtained are described below.

Waste Loading. Waste loading affects the volume and number of waste forms that must be produced. Lower waste loadings would lead to increased risks in handling and storing a larger number of waste forms, more shipments to the repository, and a larger number of packages to be emplaced in the repository. Waste loading was specified in units of Ci/cm^3 in the final product, as calculated from the wt % loading in waste forms containing simulated SRP waste. Waste loading data for the seven candidate waste forms are shown in Table 4.

Mechanical Stability. Mechanical stability is related to the risk of public exposure from respirable fines generated by an accidental impact. The data evaluated were wt % of fines smaller than 10 micrometers in diameter produced by an impact test at a constant energy-density of 10 $joules/cm^3$ (MCC-10 type test) (Mecham et al., 1981). Mechanical stability data are shown in Table 4.

TABLE 4
WASTE LOADING AND MECHANICAL STABILITY DATA
FOR THE SEVEN CANDIDATE WASTE FORMS

Waste Form	Waste Loading, Ci/cm^3	Mechanical Stability, wt % fines <10 μm
Borosilicate Glass	0.36	0.14
SYNROC	0.99	0.16
Tailored Ceramic	0.81	0.06
High-silica Glass	0.45	0.29
FUETAP Concrete	0.22	0.40
Glass Marbles in a Lead Matrix	0.19[a]	b
Coated Sol-Gel Particles (SYNROC)	0.32[c]	b

a. Indicates bulk waste loading with 62% of canister volume as lead.

b. Data unavailable.

c. Indicates bulk waste loading with 38% canister void space.

Leaching. The ability of a waste form to resist chemical dissolution in groundwaters, as determined by the inverse of its leach rate, is an important indicator of waste form performance. Leach rates were derived from MCC-1, 28 day Static Leach Tests (DOE/TIC 11400, 1981) at 40 and 90°C, and the MCC-2 Static, High Temperature Leach Test (DOE/TIC 11400, 1981) at 150°C. The MCC-1 and MCC-2 tests were judged to be sufficient for screening the relative potential performance of the various waste forms, and they were the only procedures sufficiently well developed to produce the comparable standard data needed for the evaluation.

For evaluating leach rates, the elements, leachants, and temperatures considered were: Cs and Sr, in deionized water, at 90°C; Cs and Sr, in silicate water and brine, at 90 and 150°C; and uranium in silicate water and brine, at 40 and 90°C. The leaching temperatures were chosen to bracket anticipated repository conditions. Because only simulated waste was incorporated into the waste forms for testing, uranium was used to simulate the leaching of all actinides. To compare leaching for the seven candidate waste forms, the leach rates were converted to annual fractional release rates, using the waste form densities and surface area/mass ratios for full size waste forms in canisters.

Several laboratories including the waste form developers, the MCC, and SRL contributed leaching data. Table 5 shows the consensus leach rate data set determined from all of the data submitted.

Unavailable Performance Data

Few performance data were available for the coated particle and metal matrix waste forms. To address the data absences, several assumptions were made so that these forms could be included in the evaluation.

TABLE 5

COMPARATIVE LEACH RATE DATA FOR THE SEVEN CANDIDATE WASTE FORMS

Conditions/		Leach Rate, $g/m^2 \cdot d$						
		Waste Form[a]						
Elements		BSG	SYN	TC	HSG	FUE	Pb-M	CP
Deionized H_2O								
90°C	Cs	1.12	0.75	4.50	0.028	48.0	0.13	-
	Sr	<0.001	0.33	0.0011	0.157	0.27	<0.01	-
Silicate H_2O								
40°C	U	0.036	<0.0185	0.00093	<0.02	0.007	-	-
	Cs	0.73	0.38	2.25	0.121	37.0	<0.04	-
	Sr	<0.001	0.089	<0.00036	0.0425	0.30	<0.01	-
	U	0.31	0.00021	0.0021	0.111	0.02	-	-
150°C	Cs	2.28	0.740	8.14	1.02	-	<0.04	-
	Sr	0.006	0.493	<0.00036	0.239	-	<0.01	-
Brine Solution								
40°C	U	<0.71	<0.0185	<0.0011	0.038	0.06	-	-
90°C	Cs	0.35	<0.10	5.46	<0.20	53.0	-	-
	Sr	<0.001	<0.20	<0.00036	0.546	23.0	<0.10	-
	U	0.011	0.0005	<0.0018	0.0028	0.06	-	-
150°C	Cs	2.28	1.96	5.64	0.654	-	<3.8	-
	Sr	<1.33	1.53	<0.039	1.43	-	0.15	-

a. Waste Form Abbreviations: BSG: borosilicate glass; SYN: SYNROC-D; TC: tailored ceramic; HSG: high-silica glass; FUE: FUETAP concrete; Pb-M: glass marbles in a lead matrix; CP: coated sol-gel particles.

Normalized fractional release rates for coated particles were set equal to the best rates measured for any other waste form. This treatment was based on preliminary data presented at a program review in 1981 indicating potentially very low leach rates for the coated particle form. If data for the glass marble form were unavailable, borosilicate glass leach rates were used, assuming that glass marbles in a lead matrix would have leach rates no greater than those of a glass monolith. For mechanical stability, both forms were assigned performance measures equal to the best of the other forms. It was assumed that in an impact accident, the presence of the lead matrix or the void spaces between coated particles would provide buffering against any significant generation of fines from these forms.

Results and Conclusions

The final ranking of the seven candidate waste forms and the FOM scores resulting from the product performance evaluation are shown in Table 6. The seven forms can be grouped into three categories: 1) the two ceramic forms and coated particles rated highest; 2) the borosilicate glass and high-silica glass forms had intermediate ratings; and 3) the metal matrix form and FUETAP concrete had the lowest ratings.

The product performance ratings are only an approximate indication of the waste forms' relative performance. As shown in Table 6, there is a wide margin between the highest and lowest rated waste forms, which allows a reasonably confident distinction to be made between them. Discrimination is less clear between waste forms in the high and intermediate rating categories (viz. ceramics versus borosilicate glass), for several reasons. Relative leach rates for ceramic versus glass forms are mixed for the three

TABLE 6
FINAL WASTE FORM PRODUCT PERFORMANCE RANKING AND FIGURE-OF-MERIT SCORES

Waste Form	Figure-of-Merit Score
SYNROC	95
Tailored Ceramic	93
Coated Sol-Gel Particles	87
Borosilicate Glass	67
High-Silica Glass	64
Glass Marbles in a Lead Matrix	40
FUETAP Concrete	39

elements evaluated (U, Cs, and Sr). Ceramic forms rated highest because of their low uranium leach rates, but the glass forms rated slightly better for Cs and Sr. Additional uncertainties include variability in the data, the variability in detection limits reported for leaching, and uncertainties in applying short-term leach test results to repository conditions for geologic time periods. As an example of the latter, borosilicate glass tends to build a protective oxide layer, which reduces its leach rate for long (>28 day) leaching times. Possibly this phenomenon could reduce the observed difference in uranium leachabilities between the ceramic and glass forms; alternatively, similar phenomena may lower long-term leachability for the ceramics.

In comparing waste forms within each of the three rating categories, the ratings were too close to permit any meaningful discrimination to be made based on product performance alone.

WASTE FORM PROCESSABILITY ANALYSIS

Introduction

To provide a quantitative comparison of the processes required for producing the seven candidate waste forms, a processability analysis was conducted by the Du Pont Engineering Department for SRL. The purpose of the study was to evaluate the relative feasibility of processing the candidate forms in a shielded and remotely operated DWPF. This assessment was considered equal in importance to the product performance evaluation in the screening of the seven candidate forms. Results of the processability analysis and the product performance evaluation were combined to obtain a single rating of the seven forms.

TABLE 7
CANDIDATE WASTE FORM PROCESSES EVALUATED

Waste Form	Process
Borosilicate glass	Slurry Fed Glass Melter
High-Silica Glass	Rising-Level In-Can Sintering
SYNROC	Hot Isostatic Pressing
Tailored Ceramic	Hot Isostatic Pressing
FUETAP Concrete	Curing Under Elevated Temperatures and Pressures; Dewatering
Glass Marbles in a Lead Matrix	Slurry Fed Glass Melter; Marble Machine; Lead Matrix
Coated Sol-Gel Particles	Sol-Gel Sphere Formation with CVD Coating of PyC and SiC

Six waste form processes were evaluated (Table 7). The processability analysis involved the definition of individual waste form processes by the developers; development of flowsheets, equipment concepts and conceptual designs by the Engineering Department in collaboration with the waste form developers and SRL; and the quantitative rating of the processes by the Engineering Department. Process factors considered in the analysis included complexity/reliability, resource requirements, safety, and quality control. Full details of the evaluation are documented in a report published by SRL (Dunson et al., 1982).

Ground Rules

Several ground rules were set by SRL as bases for the processability analysis. The principal ground rules were:

1. Processes should be for the immobilization of all SRP HLW, including both sludge and cesium concentrate from the supernate.
2. Conceptual designs should include only Stage 1 of the proposed DWPF.*
3. Conceptual designs should incorporate Du Pont operating and maintenance philosophy for nuclear material processing facilities (e.g., completely remote operation and maintenance and no explosive gas mixtures allowed).
4. All process steps must have defined, but not necessarily developed equipment concepts.

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* Present plans for the DWPF call for the construction of two facilities built in stages to reduce the initial capital investment. The Stage 1 facility is the waste solidification facility which will initially immobilize the sludge component of SRP wastes. In the Stage 2 facility, the radionuclides (primarily cesium) in the dissolved or supernate portion of the waste will be concentrated and then transferred to the Stage 1 facility for solidification with the sludge.

Rating Method

To assess the relative merits of the waste form processes, twenty-one processability criteria in four major categories were defined and weighted according to their overall importance. Design data from process flowsheets, equipment definitions, and facility layouts were used to rate each process against these criteria. The four major criteria categories, complexity/reliability, resource requirements, safety, and quality control were weighted 40, 25, 20, and 15 percent, respectively. The processability criteria and their relative weights are shown in Table 8. A Figure-of-Merit (FOM) rating method, similar to and compatible with the product performance evaluation method, was used to rate each process for all 21 criteria.

Results and Conclusions

The processability scores for the major criteria categories are shown in Table 9. The final ranking of the processes and their associated FOM scores are shown in Table 10. A process having the best score for each criterion would have achieved a FOM score of 100. The borosilicate glass process was rated as the best (simplest) process, followed closely by FUETAP concrete. These two processes can be categorized as relatively simple. Glass marbles in a lead matrix and high-silica glass can be categorized as moderately complex; crystalline ceramics (SYNROC and tailored ceramic) as complex; and coated sol-gel particles as very complex. These results compare favorably with two other analyses described earlier (Stone et al., 1979, and E. R. Johnson Associates, Inc., 1980), which also assessed the relative merits of candidate waste form production processes.

TABLE 8
WASTE FORM PROCESSABILITY ANALYSIS CRITERIA

Factor	Relative Weight
A. Complexity/Reliability (40%)	
1. Critical control parameters	10
2. Process cell requirements	8
3. Process steps	6
4. Equipment pieces at high temperature (>350°C) or high pressure (>150 psi)	4
5. Unusual service facilities	4
6. Recycle loops	2
7. Equipment pieces in covered cells	2
8. Chemical additions through walls	2
9. Dry radioactive materials transfer steps	2
	40
B. Resource Requirements (25%)	
1. Capital cost	7.5
2. Development items	7.5
3. Raw materials	2.5
4. Decontamination and disposal	3.75
5. Lag storage	3.75
	25
C. Personnel Safety (20%)	
1. Radioactive high maintenance equipment pieces	10
2. Process steps at high temperature or pressure	5
3. Other hazardous materials	5
	20
D. Quality Control and Assurance (15%)	
1. Product tolerance to process variations	7.5
2. Dry particle processing steps	3
3. Steps to prepare and test sample	3
4. QC sampling points	1.5
	15

TABLE 9
PROCESSABILITY ANALYSIS FIGURE-OF-MERIT SCORES FOR THE MAJOR PROCESSABILITY CRITERIA CATEGORIES

Major Processability Category	Borosilicate Glass	Glass Marbles in Lead Matrix	High-Silica Glass	FUETAP Concrete	Crystalline Ceramics	Coated Particles
Complexity/ Reliability	38	26	23	31	17	11
Resource Requirements	21	13	12	14	14	8
Safety	15	12	12	20	7	6
Quality Control/ Assurance	9	7	4	11	4	6

TABLE 10
PROCESSABILITY ANALYSIS FINAL FIGURE-OF-MERIT RATINGS

Process	Figure-of-Merit
Borosilicate Glass	83
FUETAP Concrete	77
Glass Marbles in a Lead Matrix	58
High-Silica Glass	51
Crystalline Ceramics	42
Coated Sol-Gel Particles	32

COMBINED PRODUCT PERFORMANCE AND PROCESSABILITY RATINGS

The processability analysis rating method was structured to be compatible with the rating procedure used in the product performance evaluation. This permitted combining the results from the two evaluations to give an overall comparative assessment of the seven waste forms based on both product and process considerations. A single combined FOM rating for each waste form was obtained by calculating the geometric mean of the two individual scores. The individual and combined FOM ratings are shown in Table 11.

The combined scores were calculated as geometric means rather than simple arithmetic means to provide a realistic representation of overall merit. Geometric means more effectively take into account poor product and/or process ratings and penalize any wide disparity between individual product and process scores. Thus, the fact that a waste form which ranked highly for product performance was extremely difficult to produce, or that a highly ranked process would produce a relatively less durable form, would be reflected in the overall FOM. As shown in Table 11, forms such as the crystalline ceramics and coated particles, which have the highest performance ratings, are lowered in the overall rating because of the complexity of their production processes. Similarly, FUETAP concrete, while requiring a relatively simple process, is ranked low because of its poor product performance rating. The high process rating for borosilicate glass and the intermediate product performance score resulted in its overall top ranked position.

TABLE 11
FINAL PRODUCT, PROCESS, AND COMBINED FIGURE-OF-MERIT SCORES FOR THE SEVEN CANDIDATE WASTE FORMS

Waste Form	Product	Process	Combined[a]
Borosilicate Glass	67	83	75
SYNROC	95	42	63
Tailored Ceramic	93	42	62
High-Silica Glass	64	51	57
FUETAP Concrete	39	77	55
Coated Sol-Gel Particles	87	32	53
Glass Marbles in a Lead Matrix	40	58	48

a. Geometric mean.

WASTE FORM SCREENING EVALUATION RESULTS

The results of each of the four major inputs discussed above served as the bases for the August 1981 comparison and screening of the seven candidate waste forms being developed in the National HLW Technology Program. These inputs were: 1) the preliminary waste form evaluations for defense and commercial HLW, 2) the Alternative Waste Form Peer Review Panel reviews and recommendations, 3) the product performance evaluation, and 4) the processability analysis. Based on the results of these considerations, two of the seven forms were selected for further development and evaluation.

First, borosilicate glass was selected for continued development as the reference form for the DWPF. The bases for this selection are as follows:

1. The process for fabricating the borosilicate glass waste form is the simplest and least expensive of all those considered.
2. Borosilicate glass performance properties rated well relative to the other forms.
3. Borosilicate glass was rated as the preferred form for HLW immobilization by the Alternative Waste Form Peer Review Panel.
4. Borosilicate glass was consistently selected as the preferred form by the DOE defense-sites, and it was rated highest in the commercial waste form evaluations.

Second, the crystalline ceramic forms, although ranking rather low in processing, were chosen as the best alternatives to borosilicate glass. The bases for this selection are as follows.

1. The crystalline ceramic forms, SYNROC and tailored ceramic, ranked highest in the product performance evaluation, and have some characteristics potentially superior to glass.
2. The ceramics have generally better high-temperature leaching characteristics than borosilicate glasses.
3. A number of mineral analogs of the crystalline ceramics have proven extremely durable in nature.
4. The SYNROC form, rated second by the Alternative Waste Form Peer Review Panel, was judged to be the best characterized and understood of the forms other than borosilicate glass.
5. Ceramic waste forms consistently ranked high in each of the DOE defense-site evaluations.

Technical distinction between the two ceramic forms, SYNROC and tailored ceramic, was not possible. Therefore, it was decided to pursue an optimized ceramic form utilizing the expertise of both primary developers, Lawrence Livermore National Laboratory and Rockwell International. SYNROC, the more developed and characterized form, was selected as the base ceramic for this Ceramic Waste Form Development Program.

Both the glass and ceramic waste forms are considered candidates for use at the DOE defense sites, and are potential candidates for immobilizing commercial reprocessing wastes should reprocessing of U.S. power reactor fuels be resumed. Both forms are thus considered to be applicable to the attainment of the overall goal of the National HLW Technology Program, which is to develop the technology for immobilizing all U.S. defense and commercial HLW.

SAVANNAH RIVER PLANT FINAL WASTE FORM SELECTION

The focal point of national waste form development efforts in FY-1982 was the technology needs for the DWPF at Savannah River, scheduled as the first defense HLW immobilization plant in the United States. The Environmental Impact Statement (EIS) (DOE/EIS-0082, 1982) for the planned DWPF, which was issued in final form in February 1982, promised further National Environmental Policy Act (NEPA) documentation on the specific waste form selection. As background for the (NEPA) documentation, various technical assessments were made comparing the borosilicate glass and ceramic waste forms to support a final DWPF waste form selection.

The comparison during 1982 of the glass and ceramic forms for immobilization of SRP HLW was based on several considerations. These included a production and interim storage risk assessment (Huang and Wright, 1982), a transportation risk assessment (Moyer, 1982), a repository risk assessment (Cheung et al., 1982), a comparative cost analysis (McDonell, 1982), a comprehensive waste form properties comparison (Stone et al., 1982), and assessments of the two forms' abilities to comply with regulatory and repository criteria (Gordon et al., 1982). These assessments were used as input for the preparation of an Environmental Analysis. Based on the Environmental Analysis, DOE determined that the appropriate NEPA documentation for the final waste form selection was an Environmental Assessment (EA). The purpose of the EA (DOE/EA-0179, 1982), issued in July, 1982, was to analyze the impact of the selection of the DWPF waste form for immobilization of SRP HLW. The EA concluded there was no significant environmental impact in choosing borosilicate glass as the preferred DWPF

HLW form, and in September, 1982, borosilicate glass was selected as the waste form to immobilize SRP HLW.

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