739/03

RECORDS ADMINISTRATION

ACFS

DP-MS-81-114

HIGH-LEVEL WASTE FORM PRODUCT PERFORMANCE EVALUATION

by

Thomas A Bernadzikowski, Jeffrey S. Allender,
John A Stone, Donald E. Gordon,
Thomas H. Gould, Jr., and Carlos F. Westberry, III

E. I. du Pont de Nemours & Co.
Savannah River Laboratory
Aiken, South Carolina 29808

A paper proposed for presentation at the
84th Annual Meeting of the American Ceramic Society
Cincinnati, Ohio
May 2-5, 1982

This paper was prepared in connection with work done under Contract No. DE-AC09-76SR00001 with the U.S. Department of Energy. By acceptance of this paper, the publisher and/or recipient acknowledges the U.S. Government's right to retain a nonexclusive, royalty-free license in and to any copyright covering this paper, along with the right to reproduce and to authorize others to reproduce all or part of the copyrighted paper.

DP-MS-81-114

HIGH-LEVEL WASTE FORM PRODUCT PERFORMANCE EVALUATION

by

Thomas A. Bernadzikowski, Jeffrey S. Allender,
John A. Stone, Donald E. Gordon,
Thomas H. Gould, Jr., and Carlos F. Westberry, III

E. I. du Pont de Nemours & Co.
Savannah River Laboratory
Aiken, South Carolina 29808

ABSTRACT

Seven candidate waste forms were evaluated for immobilization and geologic disposal of high-level radioactive wastes. The waste forms were compared on the basis of leach resistance, mechanical stability, and waste loading. All forms performed well at leaching temperatures of 40, 90, and 150°C. Ceramic forms ranked highest, followed by glasses, a metal matrix form, and concrete.

* The information contained in this article was developed during the course of work under Contract No. DE-AC09-76SR00001 with the U.S. Department of Energy.

INTRODUCTION

On August 1, 1981 a comparative screening assessment of seven candidate waste forms (SLIDE 1) as potential media for the immobilization and geologic disposal of high-level nuclear waste (HLW) was completed by the Savannah River Laboratory (SRL). This assessment was conducted in order to select two of the seven waste forms for further development in the National HLW Technology Program which has the goal of developing the technology for immobilizing all U.S. high-level nuclear wastes. Four major inputs formed the basis for the screening assessment: 1) preliminary defense and commercial HLW form evaluations; 2) independent peer review assessments; 3) a processability analysis; and 4) a product performance evaluation. This presentation deals specifically with the product performance evaluation considered in the overall waste form screening process.

PRODUCT PERFORMANCE EVALUATION METHOD

To provide a quantative assessment of product performance on a comparable basis for the seven candidate waste forms, an evaluation of waste loading, mechanical stability, and leaching properties was performed by SRL based on a rating method developed by a Department of Energy (DOE) Interface Working Group (IWG) on HLW Form Selection Factors. 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 (SLIDE 2) in a manner consistent with that specified by the IWG. The properties of radiation stability, thermal stability, and solubility, as specified by the IWG, were not treated in this evaluation because comparative data for the seven forms did not exist. When the rating scheme was used with waste form performance data, a Figure-of-Merit (FOM) rating was generated for each form giving a relative ranking of waste form performance.

FIGURE-OF-MERIT SCORE DETERMINATION

The comparative score for a waste form, for each property in each time period, was determined by converting the performance data to dimensionless values by means of a "value function." The procedure assigned scores of unity to the best-performing waste form for each property. Scores for the other forms for each property were then less than unity. These scores were then multiplied by a weighting factor representing the relative importance of each property. The overall weighting factors for the three waste form properties considered in the evaluation are shown in SLIDE 3.

The FOM scores for each waste form were obtained by summing the weighted scores for all performance properties. The overall procedure is represented in SLIDE 4. Volumetric waste loading was assigned a value function exponent (n_i) of +0.8; leaching and mechanical stability were assigned a value function exponent (n_i) of -0.25.

WASTE FORM PERFORMANCE DATA

Comparative waste form performance data 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 assessment. The types and sources of data considered for waste loading, mechanical stability, and leaching are shown in SLIDE 5.

- 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 composite waste. Waste loading data are given in SLIDE 6.
- Mechanical stability was evaluated as the wt % of fines smaller than 10μm in diameter produced by an impact test at an energy-density of 10 J/cm^3. Mechanical stability data are given in SLIDE 6.

- Leach rates considered in the evaluation were for Cs, Sr, and U derived from MCC-1, 28-day Static Leach Tests at 40 and 90°C, and the MCC-2 Static, High-Temperature Leach Test at 150°C. It is recognized that the 28-day leach rates are conservative values. They were not meant to be representative of leaching in actual repository environments, nor were they meant to be extrapolated to long-term leaching. However, the MCC-1 and MCC-2 tests were judged to be sufficient for screening the relative potential performance of the various waste forms, and were the only procedures sufficiently well developed and standardized to produce the comparable data needed for the evaluation. The matrix of radionuclides, leachants, and temperatures considered in evaluating leaching is shown in SLIDE 7.

The individual developers, the MCC, and a Comparative Leach Testing program at SRL contributed leaching data for the evaluation. No single laboratory contributed an entire data set for all forms. The MCC provided leaching data for four of the forms, SRL tested four of the forms, and each developer tested its own form. Generally, the data from the MCC, SRL, and the developers agreed fairly well. The consensus leach rate data for silicate water considered in the evaluation are shown in SLIDE 8.

Before comparing leaching for the seven waste forms, the data were converted to annual fractional release rates using the waste form densities and surface area/mass ratios for full-size waste forms in canisters. To combine release rates for different elements within one time period, and at the same temperature for a particular form, the data were weighted by the Nuclear Regulatory Commission (NRC) Maximum Permissible Concentrations of the respective radionuclides in drinking water. This treatment of the data was consistent with the procedure specified in the IWG rating method for comparing leach rate data.

APPLICATION OF THE EVALUATION METHOD

When leach rate data for different waste forms were compared, detection limits became an important factor. Before applying the evaluation method to rate the seven waste forms, certain assumptions were made to facilitate comparing leach rate data reported as detection limits. The assumptions used to compare such data are summarized in SLIDE 9. The adjusted set of leaching data for silicate water used in the evaluation is shown in SLIDE 10.

Few performance data were available for the coated particle and metal matrix waste forms. Comparable data were unavailable on mechanical stability for either form, uranium leaching for the metal matrix form, and all leaching for the coated particle form. To address these absences, several assumptions were made so that these forms could be included in the evaluation.

1) The coated particle waste form was given maximum credit for all leaching categories (i.e., it was assigned a score of unity). This treatment was based on preliminary data indicating potentially very low leach rates for this form.

2) When data for the metal matrix 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.

3) The coated particle and metal matrix forms were given maximum credit for mechanical stability. 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 the generation of fines from these forms.

RESULTS

The final ranking of the seven candidate waste forms and the FOM scores resulting from the product performance evaluation are shown in SLIDE 11. 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 two ceramic forms, SYNROC and tailored ceramic, ranked highest because of the high waste loadings and low uranium leach rates reported for them. The coated particle rating was slightly lower because of the lower waste loading, but otherwise indistinguishable.

The glass forms had comparable intermediate ratings, with a slight edge going to borosilicate glass. Both forms demonstrated moderate waste loadings and good Cs and Sr leach rates, which on the average were slightly better than those for the ceramic forms.

However, their uranium leach rates were higher than for the crystalline ceramic forms.

The metal matrix and concrete forms ranked lowest. These forms had the lowest waste loadings of the seven forms. Additionally, FUETAP had the poorest impact resistance and highest overall leach rates. Although the metal matrix form had relatively low leach rates, a moderately high surface area/mass ratio for the form had an effect of increasing the annual fractional release rates.

As can be seen in SLIDE 11, there is a wide margin of difference 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, for several reasons. Relative leach rates for the ceramics versus the glasses were mixed for the three elements (Cs, Sr, and U). Ceramics rated higher because of their low U leach rates, but the glass forms rated slightly better for Cs and Sr. Additional uncertainties include variability in the data reported and the treatment of data reported as detection limits.

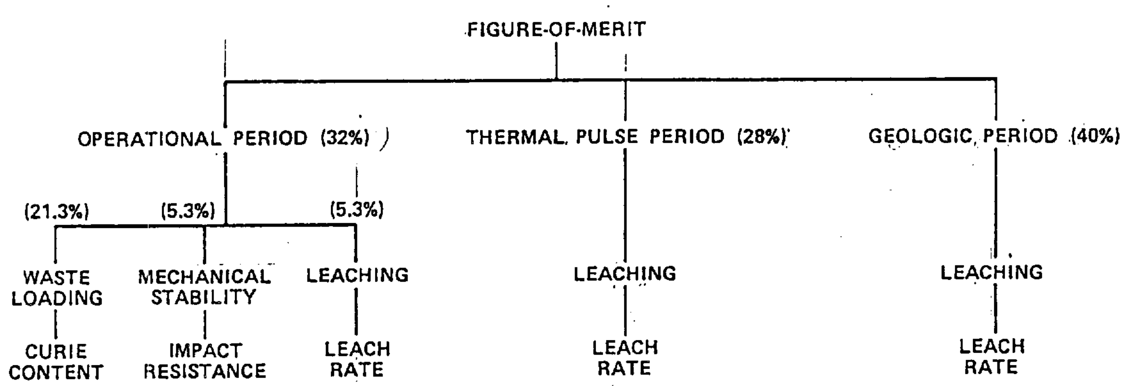
In comparing waste forms within each of the three rating categories, the ratings are too close to permit any meaningful discrimination to be made based on product performance alone.

CONCLUSION

This evaluation was conducted to provide a relative ranking based on potential product performance of seven candidate HLW forms. Of equal importance in evaluating the merits of potential HLW forms is the ability to produce them in a remotely operated and maintained environment. This topic will be discussed in the next presentation by Tom Gould of SRL. Additionally, the worth of various levels of product performance will actually depend on the performance of the entire waste disposal system, not just the waste form. Future evaluations to determine the final waste form or forms for disposal of nuclear wastes should consider the design of other barriers to radionuclide release in a repository, the behavior of the rock surrounding the repository, potential precipitation reactions, dilution of radionuclides in leaching groundwaters, and any time delays before waste elements could be transported to the biosphere. Whether some threshold level of waste form performance would exist, improvement of which would not significantly affect the radiological protection of future human populations, needs to be determined.

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

SLIDE 1. Seven Candidate Waste Forms Evaluated for Geologic Disposal of High-Level Wastes



SLIDE 2. Hierarchy of Waste Form Properties Considered in Product Performance Evaluation

Waste Form Property	% Weighting
Leaching	73
Waste loading	22
Mechanical stability	5

SLIDE 3. Relative Weighting of Waste Form Properties in Product Performance Evaluation

$$FOM = \sum \left(\frac{X_i}{Y_i} \right)^{n_i} W_i$$

where,

X_i = datum for the form being evaluated in the i^{th} category

Y_i = datum for the best form in the i^{th} category

n_i = exponent relating relative worth of any change in the performance measure

W_i = relative weight of the i^{th} performance category

SLIDE 4. Figure-Of-Merit Score Determination

Waste Form Property	Type of Data	Source of Data
Waste Loading	Volumetric Curie Content (Ci/cm^3)	Waste Form Developers
Mechanical Stability	Impact Resistance (wt % fines <10μm)	Argonne National Laboratory
Leaching	Normalized Elemental Leach Rate ($g/m^2 \cdot d$) MCC-1, MCC-2 Leach Tests	Developers MCC SRL

SLIDE 5. Waste Form Performance Data

Waste Form	Waste Loading (Ci/cm^3)	Mechanical Stability (Wt % Fines <10μm)
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*	**
Coated Particles	0.32†	**

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

** Data unavilable.

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

SLIDE 6. Waste Loading and Mechanical Stability Data

Time Period	Elements	Temperature	Leachant	Sources
Operational	Cs, Sr	90°C	Deionized H_2O Silicate H_2O	MCC Developers SRL
Thermal Pulse	Cs, Sr	90, 150°C	Silicate H_2O Brine	MCC Developers SRL
Geologic	U	40, 90°C	Silicate H_2O Brine	MCC Developers SRL

SLIDE 7. Leach Rate Data Matrix MCC-1, MCC-2 Leach Tests

Conditions/	Element	Leach Rate ($g/m^2 \cdot d$) Waste Form BSG	SYN	TC	HSG	FUE	Pb-M	CP
Silicate H_2O								
40°C	U	0.036	<.0185	0.00093	<.02	0.007	-	-
90°C	Cs	0.73	0.38	2.25	0.121	37.	<.04	-
	Sr	<.001	0.089	<.00036	0.0425	0.30	<.01	-
	U	0.31	0.00021	0.0021	0.111	0.02	-	-
150°C	Cs	2.28	0.740	8.14	1.02	-	<.04	-
	Sr	0.006	0.493	<.00036	0.239	-	<.01	-

SLIDE 8. Consensus Leach Rate Data For Silicate Water

1) If several different detection limits were reported for different waste forms,

- all data (real values or detection limits) lower than the highest limit reported were adjusted to that limit.

- real, non-limit values larger than the highest limit were not changed.

2) If detection limits reported at 40°C were higher than actual data or detection limits for the same waste form at 90°C in the same leachant,

- the 90°C data replaced the 40°C limits.

SLIDE 9. Assumptions Used To Compare Leach Rate Data Reported As Detection Limits

Conditions/ Element		Leach Rate ($g/m^2 \cdot d$) Waste Form						
		BSG	SYN	TC	HSG	FUE	Pb-M	CP
Silicate H_2O								
40°C	U	0.036	<0.02	<0.02	<0.02	<0.02	0.036	-
90°C	Cs	0.73	0.38	2.25	0.121	37.	<0.04	-
	Sr	<0.001	0.089	<0.001	0.043	0.30	0.001	-
	U	0.31	<0.0021	<0.0021	0.111	0.02	0.31	-
150°C	Cs	2.28	0.740	8.14	1.02	37.	0.04	-
	Sr	0.006	0.493	<0.00036	0.239	0.30	0.01	-

SLIDE 10. Adjusted Leach Rate Data For Silicate Water

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

SLIDE 11. Final Waste Form Product Performance Ranking and Figure-Of-Merit Scores