DL 1000-0307-92-195

DP-1553 ✓

RECORDS ADMINISTRATION
R0139141

VITRIFICATION OF TRANSURANIC AND BETA-GAMMA CONTAMINATED SOLID WASTES

M. D. DUKES

TIS FILE
RECORD COPY

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

PREPARED FOR THE U. S. DEPARTMENT OF ENERGY UNDER CONTRACT DE-AC09-76SR00001

DISCLAIMER

This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Department of Energy, nor any of their employees, make any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, mark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

Printed in the United States of America
Available from
National Technical Information Service
U. S. Department of Commerce
5285 Port Royal Road
Springfield, Virginia 22161

Price: Printed Copy A02; Microfiche A01

DP-1553 726260
Distribution Category: UC-70

VITRIFICATION OF TRANSURANIC AND BETA-GAMMA CONTAMINATED SOLID WASTES

by

M. D. DUKES

Approved by

J. K. Okeson, Research Manager
Chemical Technology Division

Publication Date: June 1980

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

PREPARED FOR THE U. S. DEPARTMENT OF ENERGY UNDER CONTRACT DE-AC09-76SR00001

ABSTRACT

Vitrification of solid transuranic contaminated (TRU) wastes alone and with high-level liquid wastes (HLLW) was studied. Homogeneous glasses containing 20 to 30 wt % ash were made by using glass frits previously developed at the Savannah River Plant and Pacific Northwest Laboratories. If the ash is vitrified along with the HLLW, 1.0 wt % ash can be added to the waste forms without affecting their quality. This loading of ash is well above the loading required by the relative amounts of HLLW and TRU ash that will be processed at the Savannah River Plant. Vitrification of TRU-contaminated electropolishing sludges and high efficiency particular air filter materials along with HLLW would require an increase in the quantity of glass to be produced. However, if these TRU-contaminated solids were vitrified with the HLLW, the addition of low-level beta-gamma contaminated ash would require no further increase in glass production.

CONTENTS

VITRIFICATION OF TRANSURANIC AND BETA-GAMMA CONTAMINATED SOLID WASTES

INTRODUCTION

Transuranic (TRU) contaminated combustible wastes generated at the Savannah River Plant (SRP) and from processing fuels from power reactors will be incinerated to reduce the volume and the weight of the waste. The ash produced must be immobilized, and glass may be the preferred form for long-term storage. In vitrification studies of these wastes, ash and high-level liquid waste (HLLW) were assumed to be vitrified either together or separately, but in the same facility. Therefore, the candidate glass compositions for SRP HLLW and for power reactor HLLW were used.

Solid wastes other than TRU ash will also be generated at SRP. These wastes include high efficiency particulate air (HEPA) filter media, sludges from the electropolishing and chemical cleaning of noncombustible wastes, and beta-gamma contaminated ash. Because these wastes must also be immobilized, the possibility of vitrifying them with the HLLW and TRU ash in the proposed Defense Waste Processing Facility (DWPF) at SRP was studied. Tests to establish the feasibility of vitrifying these waste mixtures are discussed in this report.

VITRIFICATION OF TRU CONTAMINATED INCINERATOR ASH

Ash

TRU contaminated incinerator ash must be immobilized for future storage, perhaps in a Federal repository. A possible method of immobilization is vitrification into borosilicate glass. Borosilicate glass is a candidate for the immobilization of HLLW from SRP reactors and from commercial fuel reprocessing. Because a large amount of work has been done on glass compositions for the vitrification of HLLW, candidate glass-making mixtures for HLLW were used in these waste vitrification studies. A glass-making mixture developed at Pacific Northwest Laboratories (PNL) for commercial HLLW, Frit 77-62 (Table 1),[1] and glass-making mixtures developed at Savannah River Laboratory for SRP HLLW, Frits 21, 22, and 411 (Table 1) [2] were used in these studies.

The ash will be produced from the incineration of combustible wastes contaminated with transuranics during normal operations. These wastes include bags and shoe covers, rubber gloves, tubing, and paper products.

TABLE 1

Frit Compositions, wt %

Component	*SRL Frits* 21	22	411	*PNL Frit* 77-62
SiO_2	52.5	52.5	58.3	52.2
B_2O_3	10.0	10.0	11.1	19.4
Na_2O	18.5	15.2	12.5	7.5
Li_2O	4.0	7.3	12.5	-
K_2O	-	-	-	6.0
CaO	5.0	5.0	5.6	3.0
TiO_2	10.0	10.0	-	4.5
ZnO	-	-	-	7.5

Nonradioactive waste mixtures have been incinerated in a pilot-scale operation.[3] Samples of the ash produced were analyzed, and a representative ash was prepared from reagent oxides (Table 2). The elements found in the ash are commonly used as property modifiers in plastics and rubbers. Details of the ash simulation, the glass-making process, and the glass-testing procedures are discussed in the appendix.

Table 2

Simulated Ash Composition

Component	*wt %*
CaO	24
TiO_2	18
SiO_2	16
Al_2O_3	15
Fe_2O_3	11
ZnO	6
MgO	6
BaO	1
C	3

To determine the largest quantity of ash that could be vitrified with Frit 77-62 to give a homogeneous glass, mixtures containing 35, 30, 25 and 20 wt % simulated ash were tested. Only the mixture of 20 wt % simulated ash and 80 wt % Frit 77-62 yielded a homogeneous glass. Mixtures containing 35 and 30 wt % ash left undissolved particles in the crucible bottoms, and fines were left with 25 wt % ash. The Soxhlet leachability of the glass containing 20 wt % ash was 1.2 x 10^{-4} g/(cm^2)(day) (Table 3).

Tests of Frit 21 with a series of simulated ash-frit mixtures showed that up to 25 wt % ash would give a homogeneous glass. With greater than 25 wt % simulated ash, a uniform melt was not obtained after three hours at 1150°C. The leachability of the glass containing 25 wt % simulated ash, 1.1 x 10^{-4} g/(cm^2)(day), is approximately equal to those of other ash-loaded glasses (Table 3).

Frit 411, a TiO_2-free frit was also tested. TiO_2, used as a whitener in plastics, is a major ash component. While small amounts of TiO_2 increase glass durability, large amounts increase the tendency for devitrification. With Frit 411, 30 wt % ash was vitrified to give a homogeneous green-brown glass. The Soxhlet leachability of this glass, 1.1 x 10^{-4} g/(cm^2)(day), is approximately equal to those of other ash-containing glasses (Table 3) as well as glasses containing HLLW.

TABLE 3

Soxhlet Leachability of Glasses Containing Simulated Ash

Glass Former *Frit*	*Ash Content, wt %*	*Leachability,** (x 10^{-4} $g/(cm^2)(day)$)
77-62	20	1.2
21	25	1.1
411	30	1.1

*In distilled water at 100°C.

Ash and Power Reactor High-Level Liquid Waste

Combustible TRU contaminated waste generated from power reactor fuel processing must be immobilized. TRU ash and HLLW could be incorporated into the same waste form. Therefore, the effects of small quantities of ash on HLLW waste glass were studied. In proposed plans for the vitrification of power reactor HLLW, the glass production rate is approximately two tons/day.

The ash production from the incineration of TRU combustibles would be approximately six pounds/day. The addition of six pounds of ash to the waste stream in a two-ton/day vitrification facility would yield a glass product containing only 0.15 wt % ash.

Calcined HLLW was simulated with nonradioactive reagent oxides (Table 4) for this study. The glass product would contain 33.3 wt % calcined HLLW and 66 wt % PNL Frit 77-62. To test the effect of adding ash to the waste, glasses containing 33.3 wt % simulated calcined HLLW with 5.0, 2.5, and 1.0 wt % ash were prepared with the remainder being Frit 77-62. Of these glasses, only the one containing 1.0 wt % ash was completely homogeneous. The glass with 2.5 wt % ash contained gray fines, and the glass with 5.0 wt % contained large insoluble particles. Thus, only 1.0% ash can be loaded with HLLW glasses if the 33.3 wt % loading of HLLW is maintained. This, however, is well above the 0.15 wt % loading necessary to handle the expected ash generation.

Samples of glass both with and without ash were leached in Soxhlet extractors. The leachabilities of both glasses are approximately 1.1×10^{-4} g/(cm^2)(day) (Table 5). Also, no difference in homogeneity was noted between the two glasses.

To determine if ash would promote excessive devitrification in HLLW glass, glasses with and without ash were heated for two weeks at 600°C. After heating, the glasses had an opaque brown appearance. Microscopic analysis revealed crystals scattered over the opaque surface with no apparent difference for glass with and without ash. The Soxhlet leach rates based on the weight loss of the glasses were similar: 7×10^{-5} g/(cm^2)(day) for glass with ash, 9×10^{-5} g/(cm^2)(day) for glass without ash (Table 5). The leach rates based on the sodium content of the leachant were identical: 2.6×10^{-4} g/(cm^2)(day). Thus, the addition of 1.0 wt % ash has no adverse effect on the Soxhlet leachability of HLLW glass.

Ash and SRP High-Level Liquid Waste

Incineration of stored TRU contaminated combustible materials from SRP operations is an alternative to storage of the wastes on pads. Ash will be generated at a maximum rate of 5200 pounds/year. In the DWPF, proposed for startup in the year 1988, HLLW will be vitrified with a glass production of 1.6×10^6 pounds/yr. Because TRU contaminated ash will require immobilization, one processing option is to add the ash to the HLLW calcine. Tests showed that this option would not affect glass quality.

TABLE 4

Simulated Power Reactor High-Level Waste Calcine

Inert Compounds	*wt %*
Na_2O	1.7
Fe_2O	3.0
Cr_2O_3	0.5
NiO	0.2
Gd_2O_3	16.6
MnO_2	12.7
Na_2SO_4	1.4
Fission Products	*wt %*
SeO	0.1
Rb_2O	0.6
SrO	1.7
Y_2O_3	1.0
ZrO_2	8.1
MoO_3	8.5
Tc_2O_7	2.2 (Mo)*
RuO_2	4.7 (Fe)
Rh_2O_3	0.8 (Co)
PdO	2.5 (Ni)
Ag_2O	0.1
CdO	0.2
SnO_2	0.1
Sb_2O_3	0.3
TeO_2	1.1
Cs_2O	4.7
BaO	2.6
La_2O_3	2.4
CeO_2	5.3
Pr_6O_{11}	2.4
Nd_2O_3	7.6
Pm_2O_3	0.2
Sm_2O_3	1.6
Eu_2O_3	0.3
Gd_2O_3	0.2
Actinides	*wt %*
U_3O_8	3.7
NpO_2	0.0
PuO_2	0.0
Am_2O_3	0.3 (RE)**
Cm_2O_3	0.1 (RE)

*Where used, chemical substitutes are shown in parentheses.

**RE = a commercial rare earth mixture commonly used by PNL in vitrification studies.[1] Its content (wt %) is 0.2 Y_2O_3, 24.0 La_2O_3, 48.0 CeO_2, 5.0 Pr_6O_{11}, 17.0 Nd_2O_3, 3.0 Sm_2O_3, 0.8 Eu_2O_3, and 2.0 Gd_2O_3.

TABLE 5

Soxhlet Leachability of Frit 77-62 Glasses Containing 33.3 wt % Power Reactor Calcine and Simulated Ash

Ash Content, wt %	*Heat Treatment*	*Leachability**
None	None	1.1 (2.7)**
1.0	None	1.2 (3.1)
None	600°C for 2 Weeks	0.9 (2.6)
1.0	600°C for 2 Weeks	0.7 (2.6)

*Distilled water at 100°C. Values should be multiplied by 10^{-4} to give units of $g/(cm^2)(day)$.

**Value in parentheses are sodium leachabilities based on neutron activation analyses of leach water.

Tests to determine the maximum loading of the TRU ash in HLLW glass included preparing glasses containing 25 wt % simulated HLLW calcine and 10, 5.0, 2.5, or 1.0 wt % ash with the remainder of the composition made up of Frit 21 (Table 1). These tests showed that glasses with >5.0 wt % ash contained substantial quantities of insoluble materials. The glass with 2.5 wt % ash was homogeneous, but the crucible bottom contained a small quantity of gray fines. The 1.0 wt % ash glass was the only one that was completely homogeneous.

The addition of 1.0 wt % ash to the SRP HLLW calcine for vitrification would be approximately three times the amount needed to handle the volume of ash expected from an incinerator.

To ascertain that a 1.0 wt % ash content would not change SRP glass properties, glasses containing a range of simulated waste compositions were prepared and tested. All ash-containing glasses consisted of 25 wt % simulated SRP HLLW calcine, 1.0 wt % simulated ash, and 74 wt % frit. The properties of these glasses were compared with those containing 25 wt % simulated SRP HLLW calcine and 75 wt % frit. The simulated wastes used (Table 6) were composite sludge, representing a weighted average of waste in tank storage at SRP, and high-aluminum and high-iron sludges, which represent two composition extremes found in SRP waste tanks. Frits 21, 22, and 411 (Table 1) were used in these tests.

TABLE 6

Simulated SRP Sludge Compositions, wt %

Component	*Composite Sludge*	*High Al Sludge*	*High Fe Sludge*
Fe_2O_3	31.6	6.0	61.4
Al_2O_3	46.4	86.3	5.6
MnO_2	10.3	4.9	4.1
U_3O_8	6.1	1.5	14.2
CaO	3.3	0.4	4.2
NiO	2.3	0.9	10.5

Glasses containing 1.0 wt % ash were examined microscopically to determine homogeneity. These glasses were homogeneous as were those containing no ash. Samples of glass both with and without ash were leached in Soxhlet extractors with distilled water. Comparison of leach data showed that with all frits used, the leach rates were not changed notably by the addition of 1.0 wt % ash. The leachabilities for all glasses tested were 1-2 x 10^{-4} g/(cm^2)(day) (Table 7).

TABLE 7

Leachabilities of Glasses Containing Simulated SRP Sludge and Ash

	*Leachability**		
	Frit 21	*Frit 22*	*Frit 411*
25% Composite Sludge	1.1 (1.5)**	1.4 (2.0)	1.5 (1.8)
25% Composite Sludge + 1% Ash	1.3 (2.0)	1.3 (3.0)	1.4 (2.3)
25% High Al Sludge	1.4 (1.7)	1.7 (2.0)	1.7 (1.9)
25% High Al Sludge + 1% Ash	1.5 (1.7)	1.5 (1.6)	1.7 (1.9)
25% High Fe Sludge	1.4 (3.3)	1.8 (3.3)	1.6 (3.1)
25% High Fe Sludge + 1% Ash	1.2 (2.9)	1.5 (3.7)	1.7 (3.3)

*Distilled water at 100°C. Values should be multiplied by 10^{-4} to give units of g/(cm^2)(day)

**Values in parentheses are sodium leachabilities measured by neutron activation analyses.

The effect of ash on glass devitrification was also studied. Glasses containing 1.0 wt % ash and glasses without ash were tested. After two weeks of heating at 600°C, all glasses had an opaque brown appearance. Microscopic analyses revealed crystals on a rough surface for all glasses, with no apparent difference for glasses with and without ash. Soxhlet leachabilities for these glasses were 5-25 x 10^{-4} g/(cm^2)(day) (Table 8), with no systematic increase in the leachability of the ash-containing glasses.

The viscosities of melts with and without ash were measured. Glass containing 25 wt % composite sludge and 75 wt % Frit 21 had a viscosity of 38 poise at 1150°C. An identical melt, except for the addition of 1.0 wt % ash,had a viscosity of 40 poise. Thus, the viscosity of the melt is almost unchanged by this small amount of ash.

TABLE 8

Leachabilities of Glasses Containing Simulated SRP Sludge and Ash (Glass Heated at 600°C for Two Weeks)

	*Leachability**	
	Frit 21	*Frit 411*
25% Composite Sludge	5.7 (12)**	6.1 (4.7)
25% Composite Sludge + 1% Ash	11 (21)	6.5 (17.6)
25% High Al Sludge	11 (8.9)	8.2 (2.4)
25% High Al Sludge + 1% Ash	15 (13)	8.3 (3.6)
25% High Fe Sludge	26 (52)	23 (68)
25% High Fe Sludge + 1% Ash	26 (49)	25 (61)

*Distilled water at 100°C. Values should be multiplied by 10^{-4} to give units of g/(cm^2)(day).

**Values in parentheses are sodium leachabilities measured by neutron analyses.

VITRIFICATION OF SOLID WASTES WITH SRP HIGH-LEVEL LIQUID WASTE

Transuranic Contaminated Solid Wastes

HEPA filter media and electropolishing sludges may also require long-term storage. Vitrifying these materials with SRL HLLW, however, would almost double the amount of glass to be made in the DWPF and is not recommended.

Based on a possible ratio of TRU solid waste to HLLW oxides (1:3, assuming TRU solid waste would be processed during the first 5 years of DWPF operations), testing for optimum waste loading into glass was conducted. The simulated TRU solid waste mixture used is listed in Table 9.

TABLE 9

Simulated TRU and Beta-Gamma Contaminated Solid Wastes

Component	*TRU Waste Only, wt %*	*Combined TRU and Beta-Gamma Waste, wt %*
Simulated Ash (Table 1)	4	32
HEPA Filter Media	16	11
Simulated Electropolish Sludge*	80	57

*The simulated electropolish sludge contained 73.3 wt % Fe_2O_3, 17.2 wt % NiO, and 9.4 wt % Cr_2O_3. This is approximately the composition which would result from the dissolution of 304 stainless steel, followed by precipitation and oxidation.

Initial tests showed that less of the combined HLLW-TRU waste could be vitrified when high-iron sludge (Table 6) made up the HLLW calcine. This is due to the formation of Fe-spinel crystals, a known problem in SRP glasses with high-iron contents.[2] Therefore, for conservatism, experiments to determine maximum loading were done with a 1:3 mixture of simulated TRU solids and high-iron sludge.

Glasses were made with 25, 22, and 19 wt % of the 1:3 waste mixture along with Frit 21 or 411. The glasses containing 19 wt % waste were homogeneous but the others contained undissolved particles and crystal clusters on the glass surface. The Soxhlet leachabilities for the homogeneous glasses were approximately equal to those for typical SRP waste glasses, 1-2 x 10^{-2} g/(cm^2)(day) (Table 10).

TABLE 10

Soxhlet Leachabilities of Glasses Containing High Fe Sludge and TRU or Beta Gamma Waste

wt %	*Glass Composition*	*Leachability**	
19	High Fe Sludge + TRU Waste in Frit 21	1.9	(3.6)**
19	High Fe Sludge + TRU Waste in Frit 411	1.6	(2.9)
21	High Fe Sludge + TRU, β-γ Waste in Frit 21	1.7	(2.7)
21	High Fe Sludge + TRU, β-γ Waste in Frit 411	1.9	(2.7)

*Distilled water at 100°C. Values should be multiplied by 10^{-4} to give units of $g/(cm^2)(day)$.

**Values in parentheses are sodium leachabilities determined by neutron activation analyses.

Glass will contain 25 to 27.5 wt % oxide sludges if only SRP HLLW is processed in the DWPF. Glass containing 19 wt % of the waste mixture described in the previous paragraph contains only 14 wt % simulated HLLW. Thus, the addition of the TRU solid wastes into the DWPF waste stream would require the production of 75 to 95% more glass to dispose of the same quantities of HLLW that would be vitrified if no TRU solid wastes were added. This increased glass production would be necessary as long as the TRU solid waste was added to the DWPF waste stream. At the addition rates assumed here, it could take as long as 5 years for the expected quantities of TRU solid wastes to be processed.

Beta-Gamma Contaminated Solid Wastes and Transuranic Contaminated Solid Wastes

The possibility of vitrifying ash from incineration of beta-gamma wastes in the DWPF with TRU solid waste was studied. The simulated waste mixture used in these tests was 67.5 wt % simulated HLLW calcine and 32.5 wt % simulated TRU and beta-gamma contaminated solids (Table 9). Again for conservatism, high-iron sludge was chosen for the HLLW calcine because it gave the lowest waste solubility in the glass. Tests showed that the maximum loading obtainable with the mixture of high-iron sludge and simulated TRU and beta-gamma contaminated solids was 21 wt %.

CONCLUSION

- As much as 1.0 wt % ash can be vitrified along with calcined HLLW with no adverse effect on glass quality or processing characteristics.
- Homogeneous glasses containing 20 to 30% ash and no calcined HLLW can be prepared.
- Glass containing 19 wt % waste can be prepared with the HLLW and TRU waste (including high efficiency particulate air filter materials and electropolishing sludges) mixture used in this study. Vitrifying this mixture would increase the required glass production by 75 to 95% until all TRU waste is processed.
- Adding beta-gamma contaminated ash to glass containing combined TRU and HLLW would require no further increase in glass production.

APPENDIX

EXPERIMENTAL PROCEDURE

Simulated ash was used in these vitrification studies. The composition of the simulated ash was based on data from spark source mass spectral analyses of six actual ash samples. An average composition was prepared by using reagent grade oxides and carbon (Table 2). Use of simulated ash for the vitrification studies gave sufficient quantities of ash for the necessary experiment. This ash also had a constant composition, as would a mixture of large amounts of different ashes. At the beginning of this work, the melting and pouring process and the glass products made with simulated ash and frit were compared with those of actual-ash glasses (nonradioactive). The actual ash was obtained from pilot-scale incinerator operations.[3] The properties of the glasses were similar.

Glass was prepared in 50-g batches in crucibles of approximately 100-mL volume. Glasses prepared with the premelted SRP frits (Frit 21, 22 and 411) were heated for three hours at 1150°C, poured into carbon crucibles, annealed at 500°C for one hour, and then allowed to cool slowly (three to four hours). When PNL Frit 77-62 (Table 1) was used to prepare glass, the mixtures were heated for three hours at 1050°C and then allowed to cool slowly to room temperature (five to six hours). To prepare glass with Frit 77-62, ash was combined with batch chemicals rather than premelted frit. When actual ash (non-radioactive) was used to make glass, it was ground and mixed thoroughly with frits. Otherwise, the dissolving rate of the ash was very poor. Glasses were checked for homogeneity with a microscope at 220X.

Glass samples were leached with a Soxhlet extractor[4] (Figure 1). The Soxhlet leach test is used frequently as a relative measure to compare the durability of glass samples.[5-7] This test allows continuously distilled, ~100°C water to contact the glass. Crushed glass is used in the Soxhlet test to expose a larger glass surface area. The -40 +60 mesh glass particles used in this work had a surface area of 70 cm^2/g as determined by the Brunauer-Emmet-Teller method.[8] Leach rates were calculated from the weight loss during leaching and checked by sodium content of leachant as determined by neutron activation analyses.

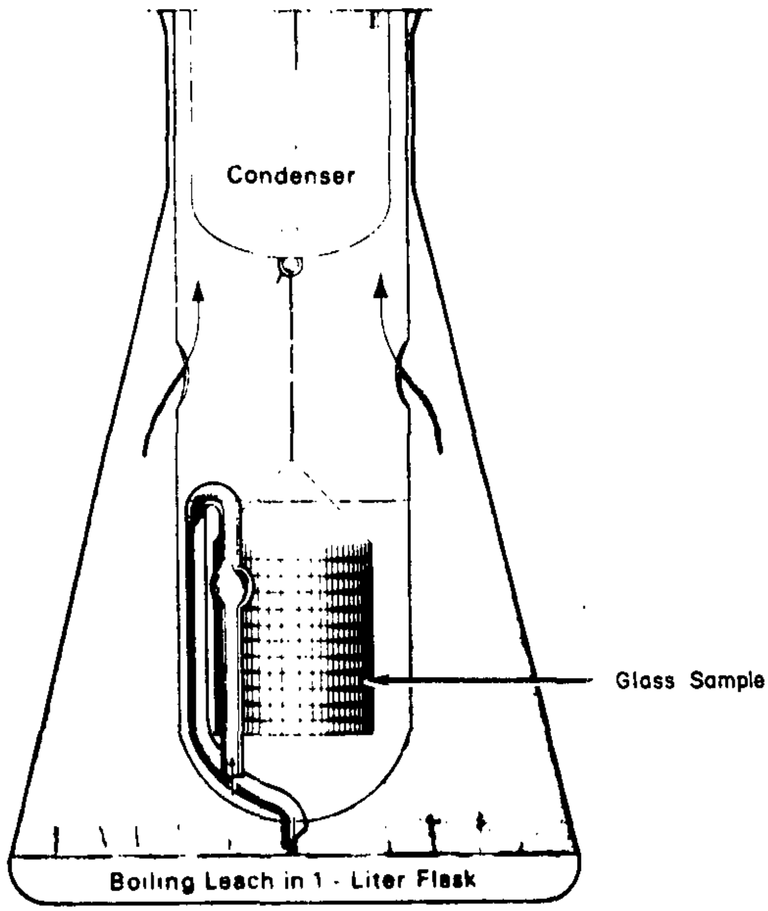

FIGURE 1. Soxhlet Extractor

REFERENCES

1. J. E. Mendel, et al. *Annual Report on the Characterization of High Level Waste Glasses*. USDOE Report BNWL-2252, Battelle-Pacific Northwest Laboratories, Richland, WA (1977).

2. M. J. Plodinec. *Development of Glass Compositions for Immobilization of SRP Waste*. USDOE Report DP-1517, E. I. du Pont de Nemours & Co., Savannah River Laboratory, Aiken, SC (1979).

3. H. E. Hootman, D. J. Trapp, and J. H. Warren. "Operation of a Pilot Solid Waste Incinerator." *Am. Nucl. Soc. Trans., 32*, 391-2 (1979).

4. A. M. Platt. *Quarterly Progress Report, Research and Development Activities, Waste Fixation Program, December 1972-March 1973*. USAEC Report BNWL-1741, pp 6-7, Battelle-Pacific Northwest Laboratories, Richland, WA (1973).

5. J. A. Kelley. *Evaluation of Glass as a Matrix for Solidification of Savannah River Plant Waste, Nonradioactive and Tracer Studies*. USERDA Report DP-1382, E. I. du Pont de Nemours & Co., Savannah River Laboratory, Aiken, SC (1975).

6. W. A. Ross, et al. *Annual Report on the Characterization of High-Level Waste Glasses*. USDOE Report PNL-2625, Battelle-Pacific Northwest Laboratories, Richland, WA (1978).

7. D. F. Luthy and W. H. Bond. *Volume Reduction System for Solid and Liquid TRU Waste from the Nuclear Fuel Cycle: January-March, 1977*. USDOE Report MLM-2436, Monsanto Research Corporation, Mound Laboratory, Miamisburg, OH (1977).

8. S. Brunauer. "The Absorption of Gases and Vapors." *Physical Adsorption* Vol. I, Princeton University Press, Princeton, NJ (1943).