Glass Forms for Alpha Waste Management

Michael J. Kupfer
Wallace W. Schulz
Charles W. Hobbick
John E. Mendel

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MASTER

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GLASS FORMS FOR ALPHA WASTE MANAGEMENT

Michael J. Kupfer
Wallace W. Schulz
Charles W. Hobbick

Chemical Technology Laboratory
Research Department
Research and Engineering Division
Atlantic Richfield Hanford Company

John E. Mendel

Nuclear Waste Processing Section
Chemical Technology Department
Battelle Pacific Northwest Laboratories

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ABSTRACT

*We have successfully converted laboratory-
scale amounts of various alpha wastes (e.g.,
plutonium-contaminated soil, incinerator ash,
pyrolysis-process residues, etc.) to immobile
silicate glasses. This paper focuses on prepara-
tion and properties of these glasses. Such
glasses appear to be suitable vehicles for immo-
bilization and densification of residues generated
in volume reduction of both U. S. Energy Research
and Development Administration and commercially
generated plutonium-bearing wastes.*

GLASS FORMS FOR ALPHA WASTE MANAGEMENT

INTRODUCTION

The expanding United States commercial nuclear fuels fabrication industry is generating large volumes of combustible wastes contaminated with varying amounts of plutonium. Such waste comprises a myriad of forms and materials including paper, plastics, rubber gloves, etc. Combustible material generated in operation at U. S. Energy Research and Development Administration (ERDA) sites also contributes heavily to the total volume of such alpha-bearing wastes.

Because plutonium has such a long half-life and its biological hazards are so severe, wastes which contain plutonium must be isolated for man's environment for thousands of years. A prime requisite, therefore, for successful management of both ERDA-generated and commercially derived combustible alpha wastes is to reduce their vast bulk to a volume of material for which reliable storage containers and facilities can be provided. Programs at various ERDA laboratories to find and demonstrate practical ways of achieving the required volume reduction are under way. Thus, scientists and engineers at the Los Alamos Scientific Laboratory are developing technology for incineration of alpha wastes[1] while at the Battelle Pacific Northwest Laboratories (PNL) research relating to pyrolysis of these wastes is in progress.[2] Destruction of the organic portion of combustible alpha waste by reaction with HNO_3-H_2SO_4 solutions (Acid Digestion process) is being studied at Westinghouse Hanford Company's Hanford Engineering Development Laboratory (HEDL).[3] Mass reduction of solid transuranic waste by

molten salt combustion is being evaluated at Atomics
International, Canoga Park, California;[4] and cyclone
incineration is being investigated at the Monsanto Research
Corporation Mound Laboratories.[5] At the US-ERDA Rocky
Flats site presently being operated by Rockwell Interna-
tional, technology for fluidized bed incineration is being
developed.[6] In addition, an alpha waste incineration
process is in routine operation at Atlantic Richfield
Hanford Company (ARHCO)[7] and Rockwell International, Rocky
Flats, Colorado. Details of some of these programs are
described in papers at this symposium.

    The processes and schemes currently being developed to
reduce the volume of the combustible alpha wastes will
likely generate residues from which the bulk of the pluto-
nium can be recovered by aqueous leaching methods. However,
in all cases there will probably still remain various
plutonium-containing solid residues which do not merit
further processing for plutonium recovery. These residues--
powders, ashes, precipitates--will be characterized by their
large surface area and will require densification and incor-
poration into some immobile, nonleachable form (such as
glass) for safe, long-term storage.

    In addition to alpha-bearing residues generated from
volume reduction of combustible wastes, significant quanti-
ties of plutonium-contaminated soil are stored at the US-
ERDA Hanford and Idaho Falls sites. These wastes may also
require conversion to some nonleachable form such as glass.

    There have been no concerted programs in the United
States or abroad dedicated to a systematic study of incor-
poration of alpha-bearing wastes into glass. Exploratory
scouting studies have indicated, however, that silicate
glasses are potentially excellent vehicles for effective

immobilization of plutonium. This paper attempts to sum-
marize some of these exploratory experiments and to suggest
areas where additional research is needed.

REVIEW OF ALPHA-GLASS PREPARATION AND PROPERTIES

Silicate glasses have been made from Hanford alpha-
contaminated soil, ARHCO plutonium-contaminated incinerator
ash, and PNL pyrolysis process residues. Preparation and
properties of these glasses are described in the sections
that follow. Glasses have also been made of HEDL Acid
Digestion process residues and are discussed separately for
convenience. Effects of generation of helium through alpha
decay and stored energy effects are also reviewed.

CHARGE PREPARATION AND MELTING

Experimental conditions for making glasses from Pu-
contaminated soil, incinerator ash, and pyrolysis residue
are summarized in Table I.

Plutonium-Contaminated Soil

A plutonium-contaminated soil sample was obtained from
the 216-Z-9 Trench site at Hanford near the Plutonium
Reclamation Facility (PRF). The sandy, moist sample con-
tained some organic sludge consisting primarily of degraded
PRF solvents. Numerous small pieces of white quartz were
scattered throughout the sample. The chemical composition
of the soil is shown in Table II. The ^{239}Pu concentration
(determined by counting the ^{239}Pu gamma emissions at the
413.6 meV peak) was 680 mg/liter of soil.

Melt charges were prepared by thoroughly mixing the
soil with appropriate amounts of B_2O_3 and Na_2CO_3 which

TABLE I

SUMMARY OF EXPERIMENTAL CONDITIONS
FOR MAKING GLASSES FROM ALPHA-BEARING WASTES

Run	Waste Type	Melt Composition, wt%					Furnace Temperature °C	Melt Time hr
		Waste	Basalt	Frit[a]	B_2O_3	Na_2CO_3		
1	Z-9 Trench soil	20	60	–	10	10	1325	0.5
2	Z-9 Trench soil	80	–	–	10	10	1425	0.2
3	Z-9 Trench soil	70	–	–	10	20	1225	0.5
4	Incinerator ash	2.2	82.8	–	5	10	1300	1.0
5	Pyrolysis residue	20	–	80	–	–	1000 and 1200[b]	3.0
6	Pyrolysis residue	25	–	75	–	–	1000 and 1200[b]	3.0
7	Pyrolysis residue	30	–	70	–	–	1000 and 1200[b]	3.0
8	Pyrolysis residue	35	–	65	–	–	1000 and 1200[b]	3.0
9	Pyrolysis residue	40	–	60	–	–	1000 and 1200[b]	3.0

[a] Zinc borosilicate glass frit composition shown in Table V, p. 8.
[b] Ingredients were initially melted at 1000° C and cooled upon completion of the
tests to determine the leachability of the product specimens; they were remelted
at 1200° C.

TABLE II

COMPOSITION OF Z-9 TRENCH SOIL SAMPLE

Component	wt%
SiO_2	55.4
Al_2O_3	11.2
FeO	6.1
MgO	2.4
CaO	2.5
Na_2O	2.4
K_2O	1.1
TiO_2	1.7
Volatiles*	13.0
^{239}Pu (mg/liter)	680.

*Loss on ignition at 700° C. Volatiles are primarily water and degraded organic solvents from the PRF.

served as fluxes to reduce the temperature necessary to melt the mixture. Although not used during these experiments, anhydrous borax ($Na_2O \cdot 2B_2O_3$) could be used in place of the Na_2CO_3 and B_2O_3.

The first formulation tested (Table I) contained crushed basalt (<35 mesh) in addition to soil as a source of silica. The Hanford basalt (Table III) was obtained from the extensive local deposit. In this formulation the soil was "sandwiched" between layers of a mixture of basalt, B_2O_3, and Na_2CO_3.

TABLE III

COMPOSITION OF HANFORD BASALT

Component	Concentration wt%
SiO_2	52
Al_2O_3	13
FeO	14
CaO	8
Na_2O	3
MgO	4
K_2O	1.5
TiO_2	2.5

The Z-9 Trench soil alone provided the entire source of silica for Runs 2 and 3. Because of the additional sodium added in Run 3, the maximum temperature necessary to achieve a fluid melt was only $\sim$1225° C.

The 700- to 1000-gram charges were contained in Dixon No. 6 graphite-clay crucibles (Dixon Crucible Company). Melting of the charges was performed in a hot cell specially fitted with induction melting equipment. The charges were heated until molten at 1225° to 1425° C, maintained in the molten state from 0.2 to 0.5 hour, and then cooled to 25° C.

Plutonium-Contaminated Incinerator Ash

Plutonium-contaminated ash resulting from incineration of combustible organic material was obtained from the Hanford Plutonium Reclamation Facility. The concentration of ^{239}Pu in the sample was 8 mg per gram. Normally incinerator ash generated at Hanford is sent from the PRF to the Los Alamos Scientific Laboratory where it is contacted with an HNO_3-HF solution for recovery of the plutonium. A small amount of plutonium, typically about 2 mg/g of residue, remains after processing. Both leached and nonleached ash

could conceivably serve as feed to the immobilization process described in this paper.

Only one experiment to immobilize Hanford incinerator ash material was performed. The as-received incinerated ash was double-bagged in plastic. The bagged sample (11 g) was placed in a Dixon No. 6 graphite-clay crucible and was covered by a 500-g mixture of crushed basalt rock, B_2O_3, and Na_2CO_3. The mixture was heated inductively to 1300° C and maintained at this temperature for one hour to promote complete diffusion of the plutonium throughout the glass.

During the heating step black smoke and some flaming resulted from combustion of the plastic bags. The flaming continued for about five minutes before burning out.

Pyrolysis-Gasification Residues

Residues were obtained from the Battelle Pacific Northwest Laboratories laboratory-scale pyrolysis-gasification unit being tested by L. K. Mudge *et al.*[2] The residues resulted from processing of a nonradioactive charge and therefore contained no radioactive component. The as-received ash (chemical composition shown in Table IV) was a grayish-tan, relatively soft material, consisting of particles ranging from about 1 cm in diameter down to fluffy fines (estimated less than 100 mesh). The bulk density was:

	g/cm^3
As-received	0.211
After pulverizing to pass 6 mesh screem	0.273
Tap density of pulverized material	0.409

Charges containing pyrolysis-gasification residues were prepared by mixing the pulverized (<6 mesh) residue with appropriate amounts of a zinc borosilicate glass frit (Table V) that has been studied extensively for fixation of

TABLE IV

CHEMICAL ANALYSIS OF ASH FROM PYROLYSIS-GASIFICATION PROCESS[a]

Ash Constituents	wt%
H_2O[b]	4.6
SiO_2	34.2
TiO_2	16.6
Al_2O_3	17.0
CaO	5.5
$CaCl_2$[c]	3.6
$CaSO_4$[c]	1.6
NaCl[c]	1.8
KCl[c]	1.3

[a]The ash was made by pyrolysis of a feed stock containing 15 wt% rubber tubing, 15 wt% Neoprene, 20 wt% PVC, 10 wt% Tygon, 10 wt% polyethylene, 15 wt% tissue paper, and 15 wt% rags.
[b]Adsorbed water released at 110° C.
[c]Arbitrary assignments of 4.0% Cl and 0.4% S found by analysis.

TABLE V

COMPOSITION OF ZINC BOROSILICATE FRIT

Component	wt%
SiO_2	37
B_2O_3	15.1
Na_2O	5.5
K_2O	5.5
ZnO	28.9
CaO	2.0
MgO	2.0
SrO	2.0
BaO	2.0

commercial high-level waste.[8] The frit/ash ratio was
varied from 80:20 to 60:40. The 6.7-gram charges were
contained in platinum crucibles and were melted at 1000° C
for three hours in a resistance furnace. Upon completion of
tests to determine the leachability of the products
(described later in this report), the product specimens were
remelted at 1200° C. The remelted products were again
leached and the effect of melting temperature (1000° and
1200° C) on leach rates was noted.

PRODUCT QUALITY TESTS

Certain physical and chemical properties of each
product were measured.

1. Appearance and State

Color and porosity were determined by visual
inspection. X-Ray diffraction techniques were
used to differentiate between crystalline and
glassy products.

2. Density

Product densities were calculated from the
volume of water displaced by a weighed sample.

3. Leach Rates in Water

Two techniques were used to evaluate leacha-
bility:

a. Paige-Type Leach Test

Paige-type leach tests were performed with
approximately 20 grams of water-rinsed, 14 to
20 mesh (U. S. Standard Sieve Series) material
which was assumed to consist of 0.11-cm diam-
eter spheres. Pyrex vessels similar to those
described by Paige[9] were used in the leach
tests; the test material was supported on a

stainless steel screen. Airlift circulators
were used to circulate 200 ml of deionized
water over the pieces. Test samples were
leached initially for 24 hours at 25° C and
then, after changing out the leach liquor, for
96 more hours at 25° C. Leach liquor was
thereafter changed weekly. Aliquots of the
leachates were evaporated to dryness on stain-
less steel discs and the concentration of
^{239}Pu was determined using alpha counting
equipment.

The formula

$$\text{Leach rate (g/cm}^2\text{-day) based on Pu} = \frac{\mu Ci \text{ of Pu leached}}{(\mu Ci \text{ of Pu/g of sample)(sample area, cm}^2)(\text{time, days})}$$

was used to calculate leach rates of radioac-
tive products. The term "based on ^{239}Pu" is
explicitly stated when referring to leach rates
calculated by this formula.

b. Soxhlet Extractor Technique

Products were crushed and 1 gram of the
acetone-rinsed and dried 40 to 60 mesh (U. S.
Standard Sieve Series) fraction was leached at
about 99° C using the Soxhlet extractor shown
in Figure 1. Weight loss of the sample was
measured after 24 hours and again after 72
hours.

PROPERTIES OF PRODUCT FORMS

Important features of the results of the experiments
shown in Table I are discussed in the sections that follow.

Product Appearance and State

All of the products made with Z-9 Trench soil were dense
(2.4 to 2.6 g/cm³) glasses containing small inclusions of
white, crystalline quartz located in the top 1- to 2-cm
portion. It is surmised that the melting temperatures were
not sufficiently high to dissolve the small chunks of quartz
typically present in Hanford soil.

REFLUX
CONDENSER

VENT

VAPOR FROM BOILING WATER
ENTERS VENT AND IS CONDENSED
IN THE REFLUX CONDENSER

COILS REHEAT CONDENSATE,
TEMPERATURE IN SAMPLE
CUP IS APPROXIMATELY $99^{0}C$

THERMOWELL

SAMPLE CUP FILLS WITH
CONDENSATE AND DRAINS
THROUGH JACKLEG ON ABOUT
3 MINUTE CYCLE.

1 GRAM OF -45 + 60
MESH GLASS PARTICLES
IS CONTAINED IN 200
MESH STAINLESS
STEEL SCREEN ENVELOPE.

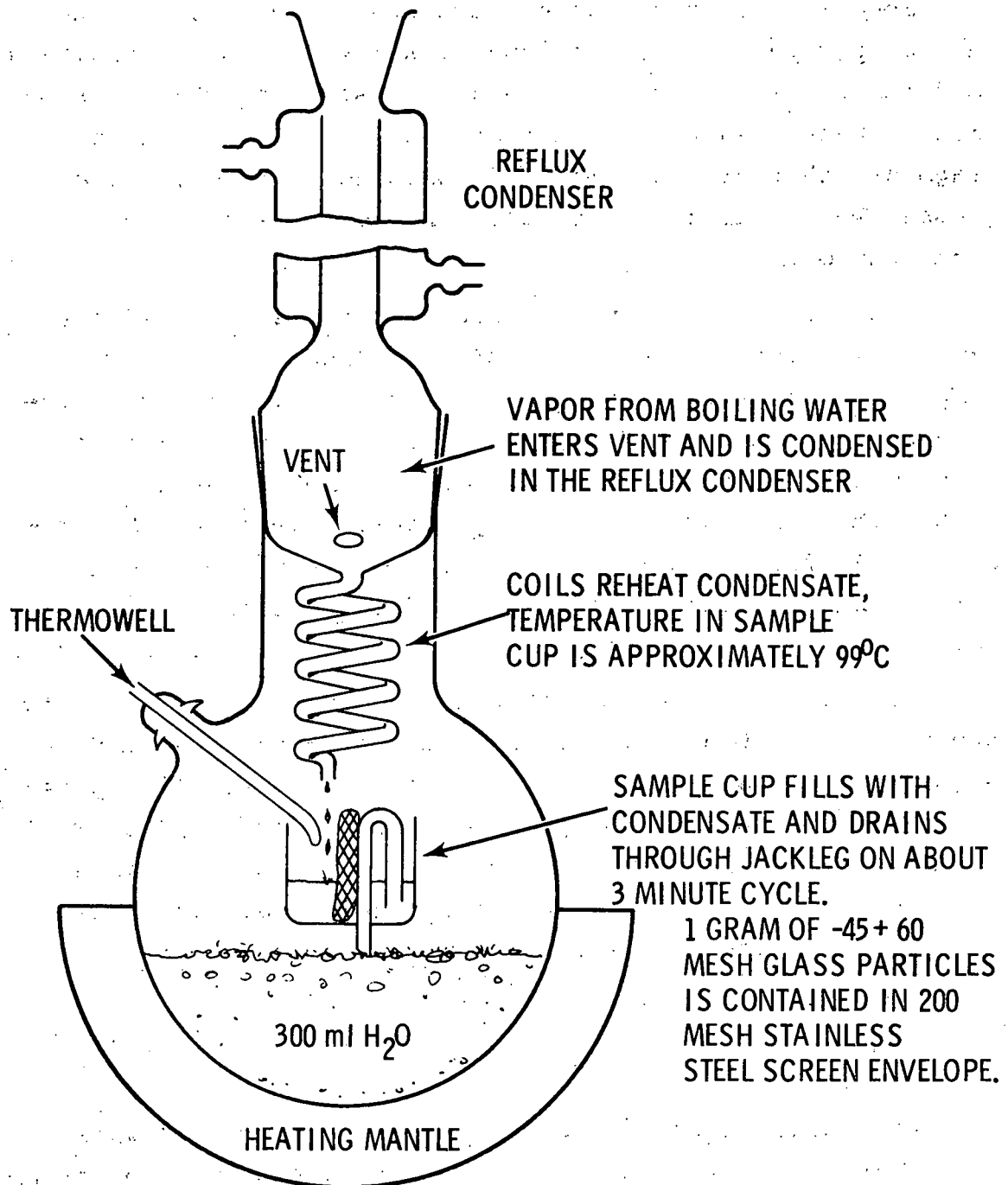300 ml H_2O

HEATING MANTLE

FIGURE 1
SOXHLET EXTRACTOR LEACH TEST

Specimens of the glasses were studied by optical and autoradiographic techniques and by X-ray microprobe analysis to determine microstructural and compositional characteristics. These results showed that, except for the quartz fragments that are disseminated throughout the melt, the glasses were uniform in composition and alpha activity distribution.

The glass made from incinerator ash was black with a micro-thin rust-colored layer on the exposed surface. The composition of this phase has not yet been identified. The product density was 2.44 g/cm^3. Although the ash was not mixed with the other ingredients in the melt charge (see p. 5), the glass product was, by visual appearance, homogeneous. Surface scanning of various pieces of the product with alpha detection instruments indicated that the ^{239}Pu was apparently evenly dispersed throughout the glassy matrix.

The solidified pyrolysis-gasification residue products prepared by heating to 1000° C contained both glass and crystalline phases. [The identity of the crystalline phases was not determined.] The products were dense (3.02 to 3.51 g/cm^3) and nonporous. They ranged from cream-color to a mottled tan as the ash content increased. Remelting the product specimens for three hours at 1200° C resulted in fewer crystalline inclusions.

Leach Behavior

The leach rates of the 216-Z-9 Trench soil products were determined by Method a, p. 9. The resistance to attack by water is very high for these products when the leach rate is based on ^{239}Pu (Figures 2, 3, and 4). The long-term leach behavior is similar to that observed recently at ARHCO for other glass product forms; i.e., the leach rates fall
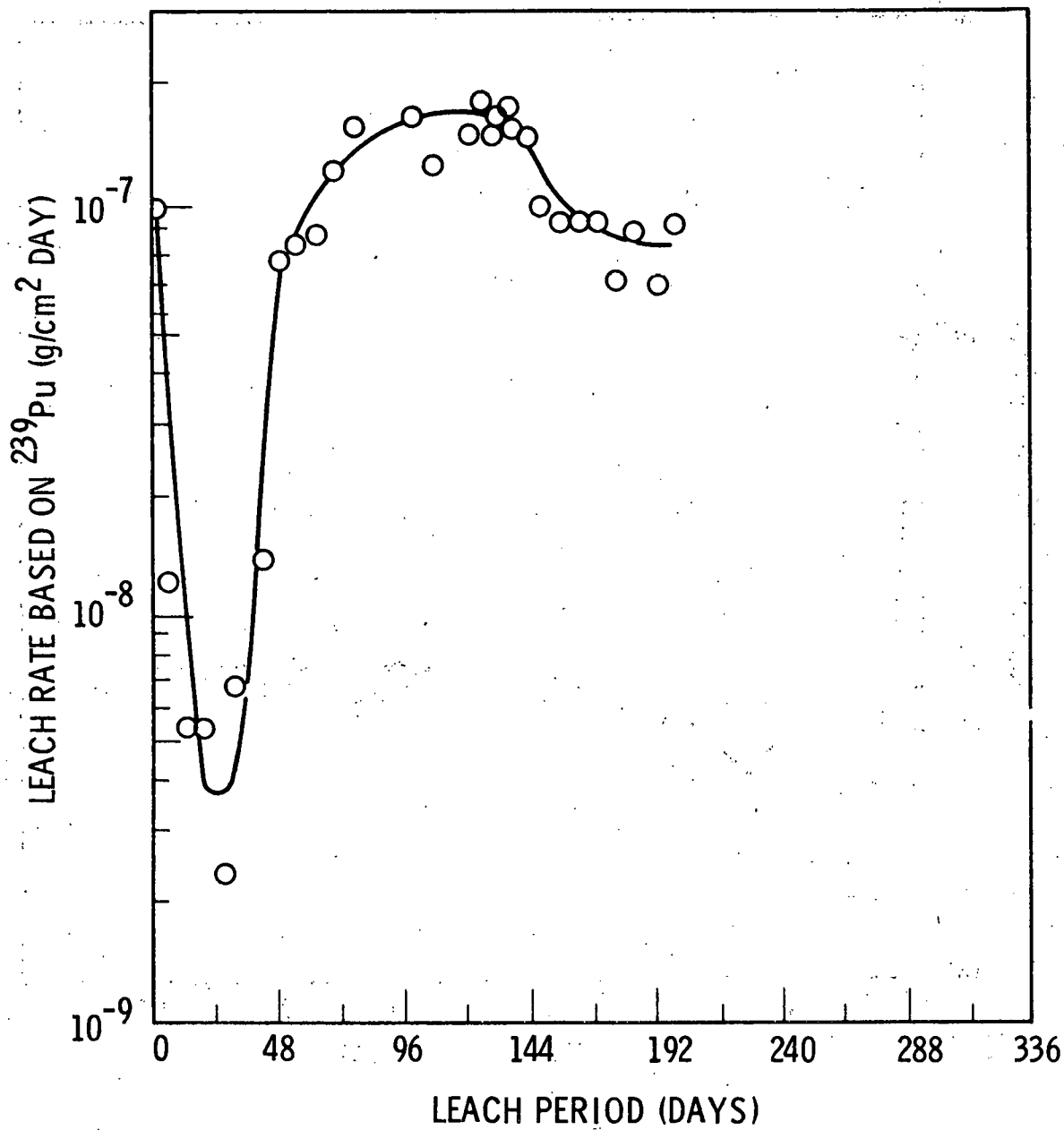
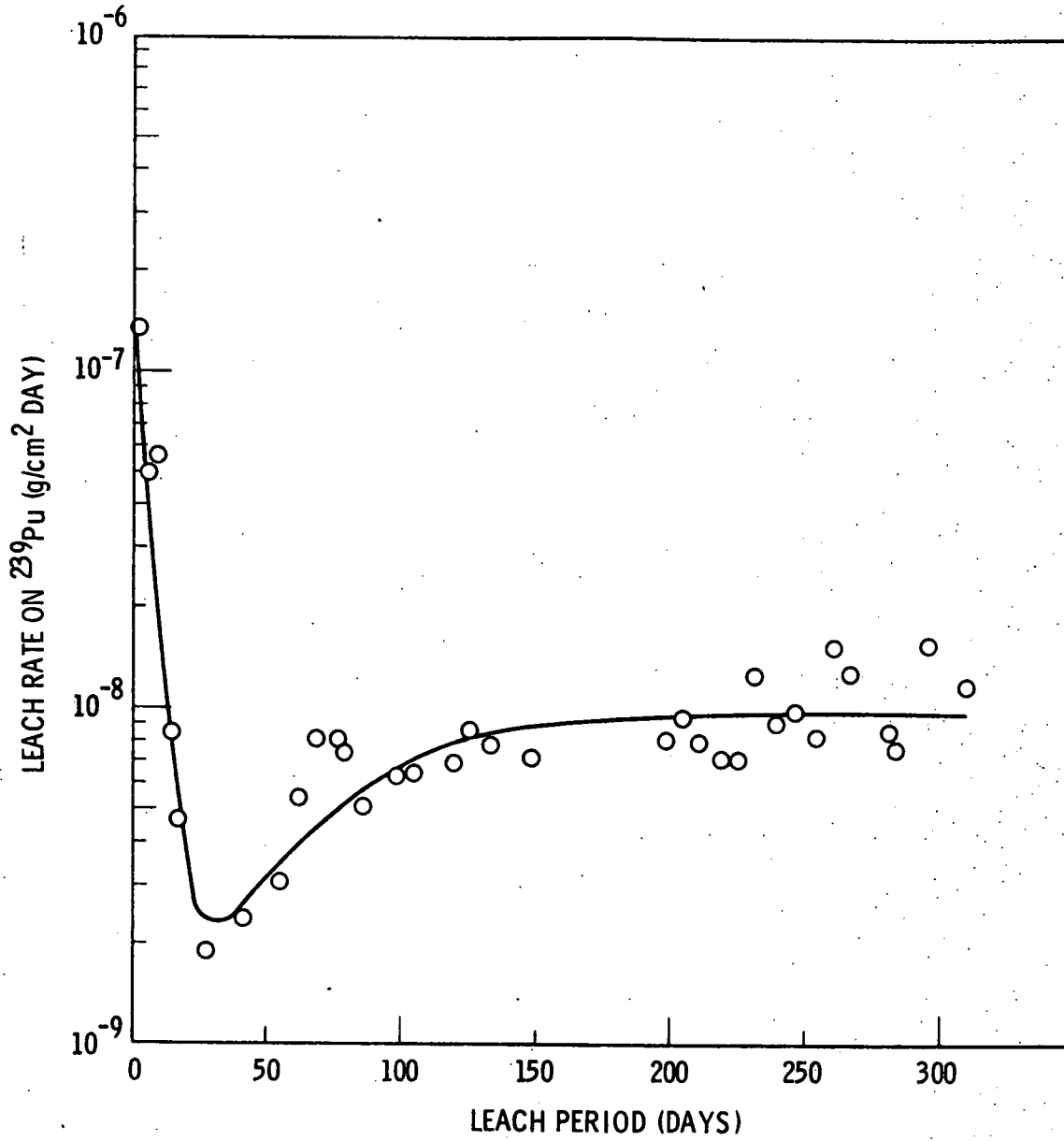FIGURE 2

LEACH DATA FOR 20% Z-9 TRENCH SOIL PRODUCT

FIGURE 3
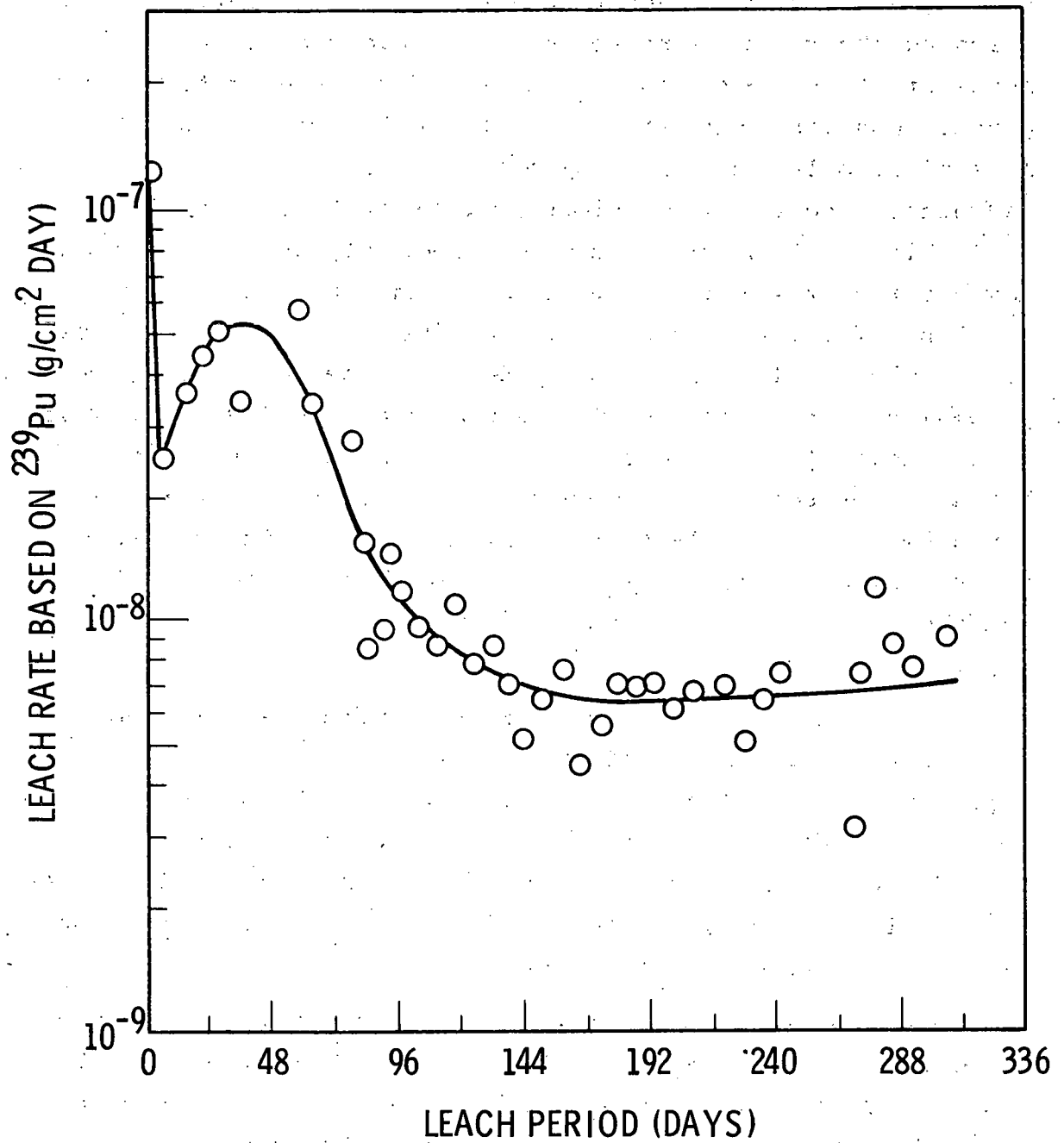
LEACH DATA FOR 70% Z-9 TRENCH SOIL PRODUCT

FIGURE 4

LEACH DATA FOR 80% Z-9 TRENCH SOIL PRODUCT

rapidly at first but then rise again. After several days of leaching the rates decrease again and eventually tend to level off to nearly constant values. The leach rate for the product containing 20 wt% soil (Figure 2) appears to have leveled at only about 10^{-7} g/cm^2 day after 160 days of leaching. The rates for products containing 70 and 80 wt% soil (Figures 3 and 4) have leveled at only about 10^{-8} g/cm^2 day after approximately 150 days of leaching. Apparently the Hanford soil is more effective than basalt as a durable glass former. In all cases, because of the relatively low precision of the radionuclide analyses at the levels present in the leachates, the data points for the leaching curves are somewhat scattered. Perhaps insufficient frequency of replacements of the leachant also contributed to the scattering (J. G. Moore *et al.* have observed depression of the fraction leached as a function of time when leachant is replaced infrequently[10].

Although the leach data are also quite scattered for the product made from incinerator ash (Figure 5) it is apparent that the leach rate (determined by Method a) decreases with time. The rate after 230 days (about 6 x 10^{-9} g/cm^2 day) has not yet reached an equilibrium. The leach data indicate, however, that ^{239}Pu in incinerator ash is very effectively immobilized.

The leaching results for the pyrolysis residue products determined by the Soxhlet Method are not a function of ash content (Table VI). For all five melts the leach rates for products melted at 1000° C were about a factor of 10 higher than previously observed for similar formulations containing synthetic high-level commercial waste.[8] The leach rates were considerably improved, however, for the glasses that were remelted at 1200° C. The weight percent loss after
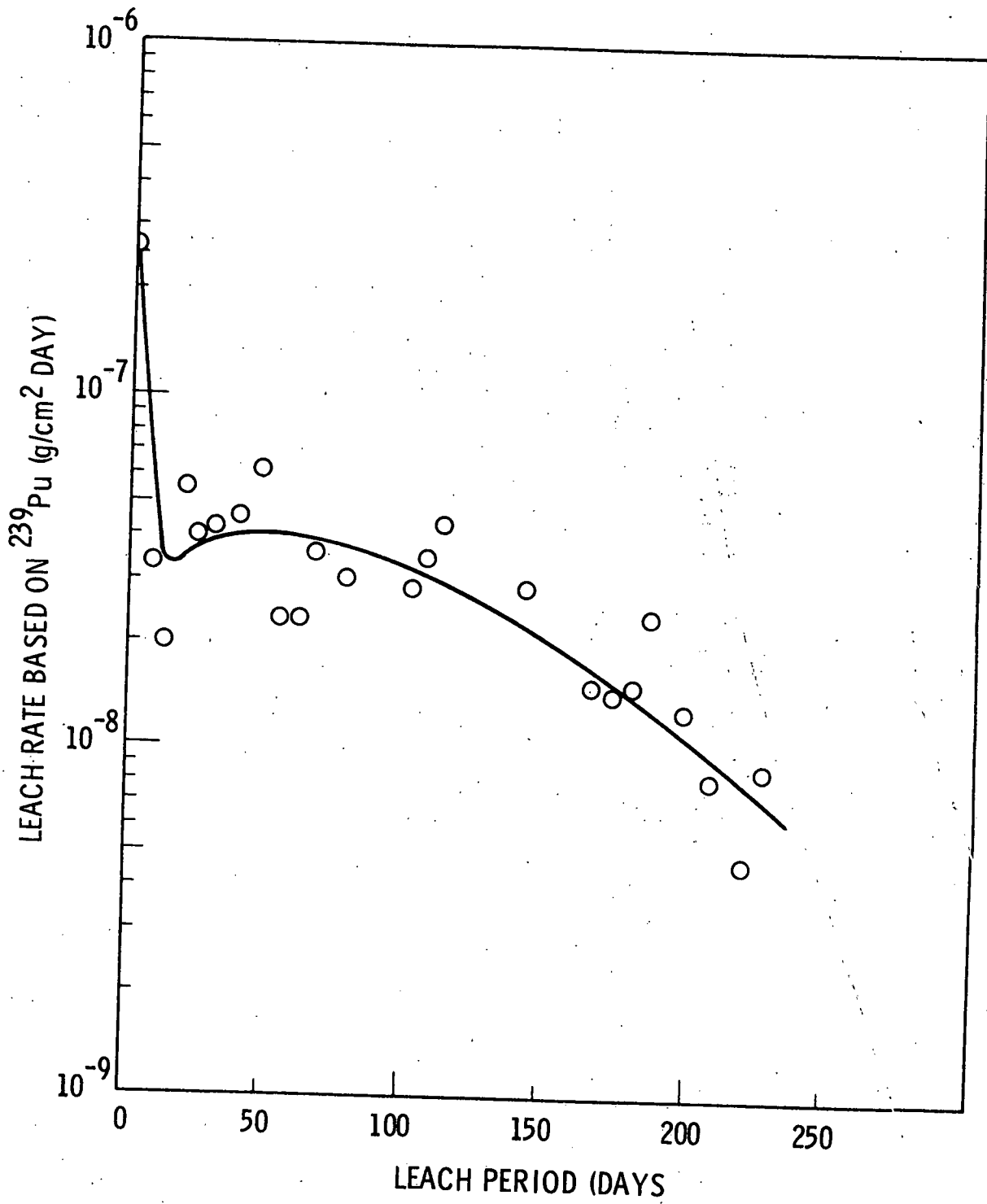
FIGURE 5

LEACH DATA FOR INCINERATOR ASH PRODUCT

TABLE VI

ACCELERATED LEACH TESTING OF PYROLYSIS-GASIFICATION PROCESS ASH FIXED IN GLASS

	Soxhlet Leach Test, wt% Loss							
	After melting @ 1000° C for 3 hr				After Melting @ 1200° C for 3 hr			
Frit/Ash Ratio	1st 24 hr	2nd 24 hr	3rd 24 hr	Overall 72 hr	1st 24 hr	2nd 24 hr	3rd 24 hr	Overall 72 hr
80:20	0.96	2.4	2.0	5.2	0.38	0.15	0.68	1.2
75:25	2.0	2.4	2.7	7.0	0.65	0.42	0.94	2.0
70:30	2.3	2.5	2.3	6.9	0.60	0.51	0.78	1.8
65:35	1.5	2.3	2.3	6.0	0.62	0.31	0.75	1.7
60:40	2.1	1.8	1.8	5.6	0.91	0.55	1.21	2.7

27 hours for all of the formulations tested (after remelting
at 1200° C) ranged from 1.2 to 2.7.

Effects of Alpha Radiation

A major area of concern in alpha-containing glass forms
is the effect of decay of the alpha-emitting isotopes on the
glass structure. Newman and Quimby have shown, for example,
that helium gas formed by alpha particle emission from
actinide nuclides can produce large internal stresses in the
glass.[11]

A series of experiments are being performed at Battelle
Pacific Northwest Laboratories to evaluate the extent of
alpha radiation on solidified high-level wastes in order to
predict the behavior of the wastes after long storage
periods. The results are directly applicable to glasses
used for the immobilization of any alpha-bearing waste with
one major reservation. It cannot be assumed that the distri-
bution of all alpha emitters will occur in a like manner in
glass. By analogy with the behavior of cerium, it is
probable that plutonium, for instance, will not dissolve
completely in low-melting borosilicate glasses, but will be
partially present as discrete crystallites of PuO_2 which
will form loci for concentrated radiation effects. In
contrast, other alpha emitters (curium, for instance) are
believed to be homogeneously distributed in the glass
matrix, yielding isotropic radiation effects. To date the
alpha effects studies at the Battelle Pacific Northwest
Laboratories have utilized only curium; studies with other
actinides in glass are planned and a comparison of the
radiation effects will be made.

A series of test specimens were prepared in the form of
"buttons" approximately 3 cm in diameter and 0.6 cm thick.

The specimens were all of identical nominal composition,
that of a typical zinc borosilicate synthetic commercial
waste glass containing about 20 wt% fission products[8]
(Table VII). The actinides which would normally be present
in a commercial waste-glass were replaced with 1 wt% ^{244}Cm
($t_{1/2}$ = 18.1 years). The radiation rate was accelerated by
the addition of the ^{244}Cm. Thus the alpha dose received by
the test specimens in the first year was approximately equi-
valent to doses that actual glasses would receive in their
first 100 years.

TABLE VII

COMPOSITION OF GLASS USED FOR ACCELERATED ALPHA EFFECTS STUDIES (wt%)

SiO_2	28.21	Ag_2O	0.05
B_2O_3	11.51	CdO	0.06
Na_2O	4.19	TeO_2	0.42
K_2O	4.19	Cs_2O	1.68
ZnO	22.04	BaO	0.91
CaO	1.53	La_2O_3	1.87
MgO	1.53	CeO_2	3.76
SrO	1.53	Pr_6O_{11}	0.37
BaO	1.53	Nd_2O_3	1.34
Rb_2O	0.21	Sm_2O_3	0.23
SrO	0.62	Eu_2O_3	0.08
Y_2O_3	0.02	Gd_2O_3	0.16
ZrO_2	2.88	CmO_2	1.00
MoO_3	3.71	Fe_2O_3	0.88
RuO_2	1.73	Cr_2O_3	0.20
Rh_2O_3	0.28	NiO	0.08
PdO	0.86	PO_4	0.39

Composition of the CmO_2, obtained from
Holifield National Laboratory, was 82.1
wt% $^{244}CmO_2$, 0.8 wt% $^{245}CmO_2$, 4.3 wt%
$^{246}CmO_2$, 0.1 wt% $^{247}CmO_2$, 11.2 wt% $^{240}PuO_2$,
0.1 wt% Fe_2O_3, and 0.2 wt% other impuri-
ties.

The test specimens were prepared in 10-g batches by intimately mixing waste calcine, glass frit, and CmO_2 in the proper ratios to form the glass composition given in Table VII. Melting was done in an electric furnace in an air atmosphere at 1200° C. The melts were stirred several times while being held at temperature for three hours. The test buttons were made by pouring the molten glass on a stainless steel plate and annealing. A few of the test specimens were devitrified by holding at 700° C for seven days.

Stored Energy. Heavy particle interactions with the atoms of a solid can cause dislocations or other effects within the solid which result in metastable energy states. If the solid is subsequently heated, stored energy (sometimes known as Wigner energy) is released equal to the difference between the ground and metastable state of the solid. Calculations have shown that the principal sources of radiation damage giving rise to stored energy are the interactions of the solid with alpha particles and associated heavy particle recoils; thus the [244]Cm-doped specimens serve as excellent media for the estimation of the radiation-induced stored energy which may be induced in radioactive glass.

Stored energy is being measured with a du Pont Model 990 differential scanning calorimeter. The results being obtained on specimens stored at 25° C are shown in Figure 6. The stored energy is approaching a saturation value of less than 25 cal/gram at a cumulative alpha dose of about 10^{18} alphas/gram. The instantaneous release of this amount of energy will not constitute a hazard in any forseeable situation. It is tentatively concluded the concerns about the buildup of stored energy in waste glass may be
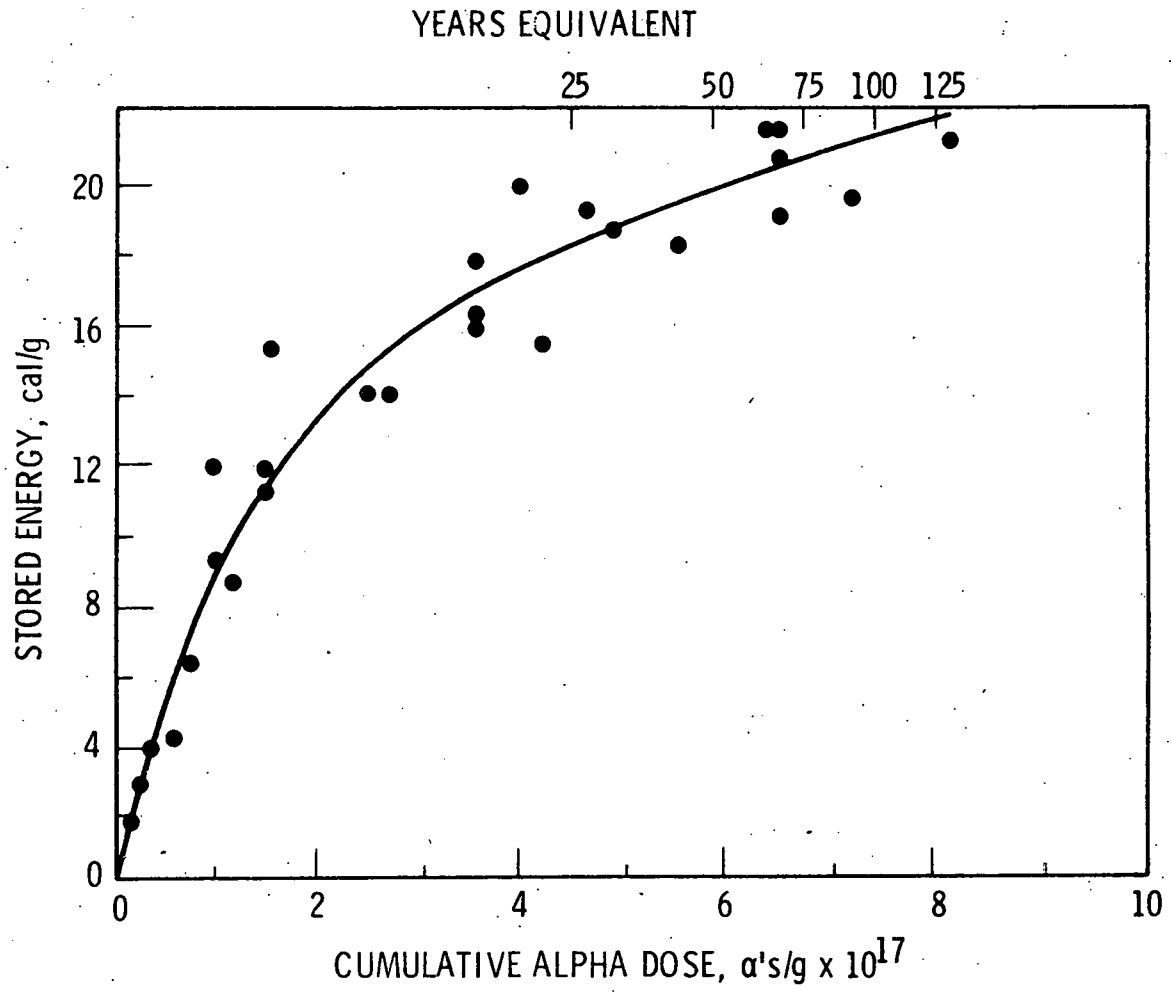
FIGURE 6

STORED ENERGY IN ZINC BOROSILICATE WASTE GLASS STORED AT 25° C

safely discounted.

Other Heavy Particle Radiation Effects. The ^{244}Cm-
doped glass specimens are being used to determine the
effects of heavy particle radiation upon several other
important glass properties, including:

- Leachability.

- Frangibility.

- Helium diffusion.

- Density.

- Microstructure.

The data are not complete enough to discuss in
detail at this time. In general, a cumulative alpha dose of
10^{18} alphas/gram has not produced marked changes in leacha-
bility or frangibility. Perhaps the most significant find-
ing is a definite radiation-induced change in density. If,
as discussed earlier, the alpha-emitting isotopes are
nonuniformly concentrated in the glass, the resulting aniso-
tropic density changes could cause severe internal stresses--
perhaps ultimately causing fracture of the glass.

IMMOBILIZATION OF ACID DIGESTION PROCESS RESIDUES

The residues from the Hanford Engineering Development
Laboratory Acid Digestion process are distinctly different
from the other residues mentioned in this report in that
they may contain large quantities of sulfate. Incorporation
of Acid Digestion process residues into glass was studied
recently at HEDL.[12] The vacuum-filtered residue
(Table VIII) was first heated to 350° C to promote complete
separation of entrained H_2SO_4. Ten- to 20-gram charges
containing 10 to 30 wt% dried residues, 0 to 10 wt% borax,

TABLE VIII

ELEMENTAL ANALYSIS OF VACUUM-FILTERED
ACID DIGESTION RESIDUE

Element	Wt%
Moisture	18
Carbon	0.00
Hydrogen	1.82
Nitrogen	0.00
Sulfur	13.80
Oxygen	26.59
Ash (inorganics)	38.82

Density: 0.77 kg/liter

and 60 to 90 wt% crushed laboratory scrap glass (primarily a
mixture of soda-lime and borosilicate glass) were melted in
platinum crucibles at 1100° C for 4, 5, and 18 hours.
Visual examination of the products indicated that up to 20
wt% dried acid digestion residue could be dissolved in the
glass when heated for 18 hours. Up to 20 wt% residue was
dissolved after five hours when borax was added to the
mixture. Incomplete dissolution was apparent, however, when
30 wt% residue was added. More recent exploratory tests
have indicated that a slightly larger percentage of waste
can be dissolved in the glass when the mixtures are heated
to 1300° C rather than 1100° C.

The above-described exploratory experiments have indi-
cated that high sulfate content residues can indeed be
incorporated into some glasses. The fate of the sulfate
during the heating step must, however, be established.
Other workers[13] have noted, for example, that when exces-
ive quantities of sulfate are included in borosilicate
formulations, a second sulfate phase that is highly leach-
able often segregates during melting. Additionally since
the composition of acid digestion residues can vary con-
siderably, applicability of a glass-forming process to

immobilization of various compositions must be established.

SUMMARY AND CONCLUSIONS

Exploratory tests show that alpha-contaminated soil and some alpha-bearing residues generated from various processes for mass reduction of solid transuranic waste can be successfully incorporated in silicate glasses. Such glasses have excellent physical properties and are suitably resistant to water leaching.

Before silicate glasses can be considered suitable vehicles for immobilization of alpha waste, there is a need to establish concentrated laboratory efforts to define, on a systematic basis, acceptable glass compositions, detailed product properties, and potential problem areas such as effects of helium generation through alpha decay and stored energy effects.

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