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Quarterly Progress Report
Research and Development Activities
Waste Fixation Program
April through June 1976

J. L. McElroy

August 1977

Prepared for the Energy Research and Development Administration under Contract EY-76-C-06-1830

Battelle
Pacific Northwest Laboratories

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QUARTERLY PROGRESS REPORT
RESEARCH AND DEVELOPMENT ACTIVITIES
WASTE FIXATION PROGRAM
APRIL THROUGH JUNE 1976

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BATTELLE
Pacific Northwest Laboratories
Richland, Washington 99352

PREVIOUS REPORTS

This progress report is the fifteenth in a series that presents research and development activities in radioactive waste fixation. Previous progress reports were BNWL-1699, 1741, 1761, 1788, 1809, 1826, 1841, 1871, 1893, 1908, 1932, 1949, 1994 and 2070.

SUMMARY

Research and development activities of the Waste Fixation Program for April through June 1976 are described in this report. The objective of this program is to develop processes to convert high-level liquid waste (HLLW) from the light water reactor (LWR) fuel cycle to solid forms that are demonstrated to be physically, chemically, and radiolytically stable and inert.

Several significant results were obtained during this reporting period. These are listed below and detailed in the text of this report.

- Tests of the spray calciner, fed with simulated Nuclear Fuel Services (NFS) wastes, were successful.
- The joule-heated ceramic melter has operated continuously for more than 5 months in 1976 and for more than 16 months total. Since the last report over 2500 lb of simulated high-level waste glass have been produced while feeding dry powder calcine and slurried liquid waste.
- A report on water leaching at 25°C of UO_2 fuel with an average burnup of 54,500 MWD/MTU is being issued.
- Fluidized bed waste calcine particles were monocoated with Al_2O_3 or duplex-coated with PyC-Al_2O_3 in a fluidized bed; larger calcine pellets were formed with a disk pelletizer and coated with Pyrex frit.

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INTRODUCTION

The objective of the Commercial High-Level Waste Fixation Program is to provide the technological bases which will enable industry to adopt and implement, at the earliest possible time, fully appropriate techniques for the conversion of high-level radioactive waste to stable, nondispersible solid forms such as silicate glasses. The program is designed to be a means through which industry and government can cooperate in an effective and optimum approach to this common problem. These objectives are being accomplished by a comprehensive program which includes: development and characterization of glass formulations; equipment and process development; and design, construction and demonstration of full-scale and plant-duplicate process equipment in cooperation with industry.

The research and development activities in radioactive waste fixation for the reporting period of April through June 1976 are described in this report.

SECTION 1 - COMMERCIAL WASTE FIXATION

The purpose of this task is to develop processes and equipment for converting liquid high-level radioactive waste to a stable, relatively nondispersible form for storage and, ultimately, disposal. This objective is generally being accomplished by development of a two-step approach (calcination or concentration) followed by melting to form a silicate glass.

IN-CAN MELTING (H. T. Blair)

The purpose of this study is to develop and demonstrate on a full scale the vitrification of calcined nuclear waste and glass-forming frit using the storage can as the melting crucible.

Now that the melting capacity of the in-can melter (ICM) has been demonstrated to be sufficient to support the reference 5 MTU/day fuel reprocessing plant, we are studying the off-gas from the process.

ICM run No. 17 (ICM-17) was made with the melter coupled to the fluidized-bed calciner to demonstrate and evaluate the performance of this HLLW vitrification system when the fluidized-bed calciner is operated with a continuous inert bed. The simulated HLLW fed to the fluidized-bed calciner contained a full level of cesium and 10% of the expected ruthenium content of HLLW. The run demonstrated the compatibility of the two pieces of equipment. The can of melt was water quenched from 760°C and the effects of this quench on the can and product are being evaluated. The off-gas analyses, which are reported under "Fluidized Bed Calcination" in this report (p. 4), were encouraging. The counterflow of batch and off-gas in the coupling section and the relatively cool, small surface of the melt in the ICM are believed responsible for the low level of effluents from the process.

ICM run No. 18 (ICM-18) was made with a blend of spray-dried calcine containing cesium plus 10% of the expected ruthenium contents, and frit containing 1.5% powdered silicon fed from a screw feeder to an 8-in. diameter can. The melter off-gas was exhausted through the spray calciner filters. The can has not been opened for product evaluation and the results of the off-gas analyses are not yet available.

Following each of the last four ICM developmental runs (ICM-15, 16, 17 and 18) accumulations of agglomerated batch have been found in the coupling section, which is a 4-in. diameter pipe sloping at an angle of 60° from horizontal. Dried deposits of calcine were also found after run ICM-16 in the cone and frit feed tube. It is believed that steam from the calciner and melter condensed in these cool areas and moistened the batch, which then stuck to the surfaces of the equipment. During the Waste Solidification Engineers Prototype (WSEP) program, hot air was blown into the frit feed tube to prevent this condensation; however, this increases the volume of air that must be treated by the off-gas system. Heating the cone, frit feed tube, and coupling section to >100°C may be necessary to prevent these accumulations.

<u>DEVELOPMENTAL SPRAY CALCINER</u> (L. S. Romero)

The objective of this work is to provide a reliable system for drying radioactive liquid waste prior to vitrification.

Run DSS-28 was made using simulated NFS startup and shutdown waste (PW-8e-2) composed mostly of sodium nitrate. The spray calciner ran very smoothly at 700°C. A glossy brown scale deposited on the upper half of the chamber, while the lower half showed indications of molten material trickling down to the cone area. However, the calcine produced was a dry buff-colored material. Work is being continued to eliminate melting of the sodium nitrate on the spray chamber wall by using additives to the feed.

Run DSS-29 was made using simulated NFS waste (PW-8a-2) spiked with ruthenium. The purpose of the run was to obtain data on the amount of ruthenium lost either by volatilization or entrainment of fine calcine particles through the filters. The spray chamber temperatures and the frequency of the blowback cycle were increased slightly higher than normal. Operation during the run was very smooth and indications are that approximately 4% of the total ruthenium was lost during calcination. A full-level ruthenium run is scheduled for the near future.

JOULE-HEATED CERAMIC MELTER (C. C. Chapman)

The objective of this work is to develop a reliable ceramic melter which can be used for converting radioactive waste to glass.

Since restart in January, the engineering-scale melter has been maintained at temperature for more than 5 continuous months, which brings the total operating experience with this unit to 16 months. Since last reporting, over 2500 lb of simulated high-level waste (HLW) glass have been produced while feeding dry powder calcine or the slurried liquid waste. During the calcine-fed operation, the unit was operated at a capacity in excess of 100 lb/hr.

A simple technique for boosting the liquid throughput for this type of melter was tested. In a short experiment using this boosting technique, a liquid waste throughput in excess of 45 ℓ/hr was demonstrated. This approach for increasing liquid process capacity promises to provide the capability of processing dilute liquid waste solutions.

FLUIDIZED-BED CALCINATION (W. J. Bjorklund and F. E. Haun)

Fluidized-bed calcination studies provide data for process and equipment development in the conversion of simulated high-level commercial nuclear waste to a solidified form. The calcine product can then be fed directly or indirectly to a melting system for vitrification.

Calciner operation continued on an intermittent basis this period, the most significant operation being a 13.5-hr run while coupled to the ICM system (ICM-17). While operating in the continuous inert bed mode and feeding a special silica-free frit to the melter, 200 kg of glass were produced. A full level of cesium and 10% ruthenium were included in the PW-7a type feed. Decontamination factors for both ruthenium and cesium were greater than 10^4 across the calciner/melter coupled system. Other tests, including a conical distributor plate and new fuel and feed nozzle designs, are ongoing.

ICM INSTRUMENT SUPPORT (R. D. Dierks)

The information obtained in this study is intended to aid in developing design specifications for instrument monitoring systems for HLW vitrification equipment.

Improvements in the vacuum compensation portion of the canister weighing system have increased the resolution of the indicated canister weight to about ±0.25 kg, and the accuracy of the indicated weight changes to within 0.3% of measured weight changes.

A substantial error is introduced in the canister weighing system due to the vacuum fluctuations that normally occur in the ICM system. This error is caused by the vacuum in the calciner that supports part of the canister weight, and thus is dependent on the cross sectional area of the bellows connecting the canister to the spray calciner discharge chute. For the development unit, this error amounts to 0.49 kg/in. of water pressure change. An electromechanical system to compensate for these fluctuations was developed and proved to be quite effective. However, the mechanical retransmitting slide wire lagged in its response to fluctuations and introduced some noise into the weigh system readout signal. This system has now been modified to directly introduce into the output of the transducer load indicator a portion of the output from an electronic d/p cell measuring the pressure in the calciner, thus eliminating the retransmitting slide wire with its noise and maintenance problems.

During the last ICM experiment (ICM-18), the indicated weight fluctuations were reduced to about ±0.25 kg during the course of the experiment, except during two short periods when the calciner barrel vibrator was operated. Two disturbances were noted during these periods; one resulting from the vibration of the load cell, and the other, a transient spike induced in the signal whenever the vibrator solenoid was opened or closed. At the conclusion of the experiment, the indicated canister weight increase matched the measured weight increase of 69.3 kg within 0.2 kg. The gross weight of the filled canister was 148.0 kg.

SECTION 2 - WASTE FORM CHARACTERIZATION

The purpose of waste form characterization is to measure the properties of candidate solidified products (solidified waste and canister) as functions of composition, processing parameters, and storage conditions. The measurements are used: 1) to assure operability of the manufacturing processes, and 2) to provide data for safety analyses of high-level waste management. The ultimate goal is to characterize the physical and chemical properties of the waste forms so thoroughly that when they are placed in retrievable storage, and later in ultimate disposal, full confidence can exist that their behavior is known and that any changes or any interactions with their environment which may occur are wholly predictable.

MELT DEVELOPMENT (W. A. Ross)

The objective of this work is to develop melt compositions suitable for incorporating typical HLW in durable glasses at low processing temperatures.

The 76-76(1) composition has been tested for sensitivity to devitrification by holding samples for 1 week in a thermal gradient. The tests indicated that the glass behavior during devitrification is acceptable. The limits on PW-8a calcine content were determined to be 22 to >43 wt% based on viscosity and leachability respectively. Leach tests have also indicated that the glass has a low leach rate over a wide range of pH. Differences in sample form are affecting the leach rate values; therefore, further tests are desirable to clarify these differences.

Development of a non-zinc glass for use with silicon-containing glass batches has continued. Reasonable 25°C leach rates have been obtained, but further reductions in Soxhlet leachability are desired.

COMMERCIAL RADIOACTIVE WASTE COMPOSITIONS USED IN THE WASTE FIXATION PROGRAM (W. A. Ross)

There is a continuing need to assure that the waste compositions being studied are relevant to projected commercial reprocessing plant practice.

Two new waste compositions shown in Table 1 have been defined which represent nonstandard wastes that may occur during operations at NFS. The PW-8d will result when evaporator bottoms are not available to the waste stream. The PW-8e will occur during startup and shutdown operations and, therefore, contains a high sodium content and low fission product loading. The -1 compositions are used in most laboratory melts and the -2 compositions are used in engineering-scale tests.

THERMAL EFFECTS ON STORED GLASS (J. H. Westsik, Jr.)

The purpose of this work is to determine property changes of HLW glass as functions of storage time and temperature.

Results of leach testing of 72-68 glass according to a modified IAEA leach test procedure are reported this quarter. The as-formed glass showed leach rates of the order of 10^{-7} g/cm^2-day while the worst case samples stored at 700°C for 2 months had leach rates only a factor of ten higher. Figures 1 and 2 show these data normalized to strontium and cesium behavior respectively.

PHASE BEHAVIOR (R. P. Turcotte and J. W. Wald)

This task is directed toward general understanding of the glass-making and devitrification processes, primarily through microstructural examination (including microprobe analysis) and x-ray diffraction methods.

This quarter, kinetic studies of glass 72-68 included examination of 1000 and 1200°C melts and the calcine-free (73-1) frit. In addition, qualitative analyses of devitrification in new glass compositions 76-68 and 76-76 were completed. The kinetic study emphasized ingrowth of the major devitrification product, Zn_2SiO_4, with the objective of establishing some general principles concerning the importance of other phases present and/or sample preparation. The rate of ingrowth is in this order:

$$\text{frit 73-1} < 1200°\text{C melt 72-68} < 1000°\text{C melt 72-68}$$

TABLE 1. Waste Compositions Used in Waste Fixation Program - Supplement No. 4[a]

		Kilograms per MTU					
		PW-8d as defined	PW-8d-1	PW-8d-2	PW-8e as defined	PW-8e-1	PW-8e-2
Inerts	Na_2O	---	---	---	14.077	14.077	14.077
	Fe_2O_3	27.225	27.225	29.008	0.302	0.338	0.338
	Cr_2O_3	0.086	0.086	0.086	0.086	0.086	0.086
	NiO	0.028	0.933	0.933	0.028	0.046	0.046
	P_2O_5	1.342	1.342	1.342	1.342	1.342	1.342
Fission Products	Rb_2O	0.354	0.354	0.178(K)	0.004	0.004	0.002(K)
	SrO	1.059	1.059	1.059	0.011	0.011	0.011
	Y_2O_3	0.598	0.027(RE)[b,c]	0.043(RE)	0.006	--- (RE)	--- (RE)
	ZrO_2	4.944	4.944	4.944	0.049	0.049	0.049
	MoO_3	5.176	6.375	6.375	0.052	0.064	0.064
	Tc_2O_7	1.291	--- (Mo)	--- (Mo)	0.013	--- (Mo)	--- (Mo)
	RuO_2	2.972	2.972	--- (Fe)	0.030	--- (Fe)	--- (Fe)
	Rh_2O_3	0.480	0.304(Co)	0.304(Co)	0.005	0.003(Co)	0.003(Co)
	PdO	1.483	--- (Ni)	--- (Ni)	0.015	--- (Ni)	--- (Ni)
	Ag_2O	0.088	0.088	0.088	0.001	0.001	0.001
	CdO	0.097	0.097	0.097	0.001	0.001	0.001
	TeO_2	0.725	0.725	0.725	0.007	0.007	0.007
	Cs_2O	2.880	2.880	0.963(K)	0.029	0.028	0.009(K)
	BaO	1.567	1.567	1.567	0.016	0.016	0.016
	La_2O_3	1.480	3.213(RE)	5.114(RE)	0.015	0.032(RE)	0.051(RE)
	CeO_2	3.323	6.426(RE)	10.228(RE)	0.033	0.064(RE)	0.102(RE)
	Pr_6O_{11}	1.482	0.669(RE)	1.065(RE)	0.015	0.007(RE)	0.011(RE)
	Nd_2O_3	4.522	2.276(RE)	3.622(RE)	0.045	0.023(RE)	0.036(RE)
	Pm_2O_3	0.123	--- (RE)	--- (RE)	0.001	--- (RE)	--- (RE)
	Sm_2O_3	0.924	0.402(RE)	0.639(RE)	0.009	0.004(RE)	0.006(RE)
	Eu_2O_3	0.200	0.107(RE)	0.170(RE)	0.002	0.001(RE)	0.002(RE)
	Gd_2O_3	0.137	0.268(RE)	0.426(RE)	0.001	0.002(RE)	0.004(RE)
Actinides	U_3O_8	11.689	12.683	--- (RE)	11.689	11.781	--- (RE)
	NpO_2	0.865	--- (U)	--- (RE)	0.009	--- (U)	--- (RE)
	PuO_2	0.085	--- (U)	--- (RE)	0.085	--- (U)	--- (RE)
	Am_2O_3	0.181	--- (RE)	--- (RE)	0.002	--- (RE)	--- (RE)
	Cm_2O_3	0.040	--- (RE)	--- (RE)	---	--- (RE)	--- (RE)
Total	--	77.4	77.0	68.9	28.0	28.0	16.3

a. See also p. 28, Ref 2, p. 23, Ref 3, p. 34, Ref 4, and Ref 5.
b. Where used, chemical substitutes are shown in parentheses. Values listed represent actual amount present. If no value shown, then weight included with substitute.
c. RE = a commercial rare earth mixture nominally containing wt% 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

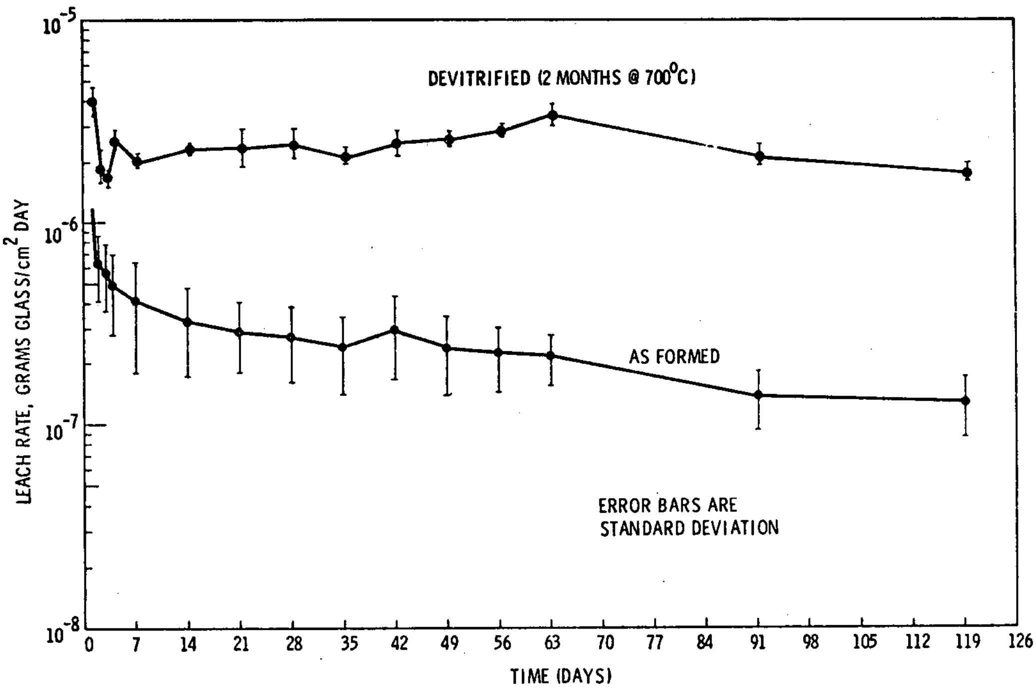

FIGURE 1. Leachability of HLW Glass, Normalized to Cesium Behavior

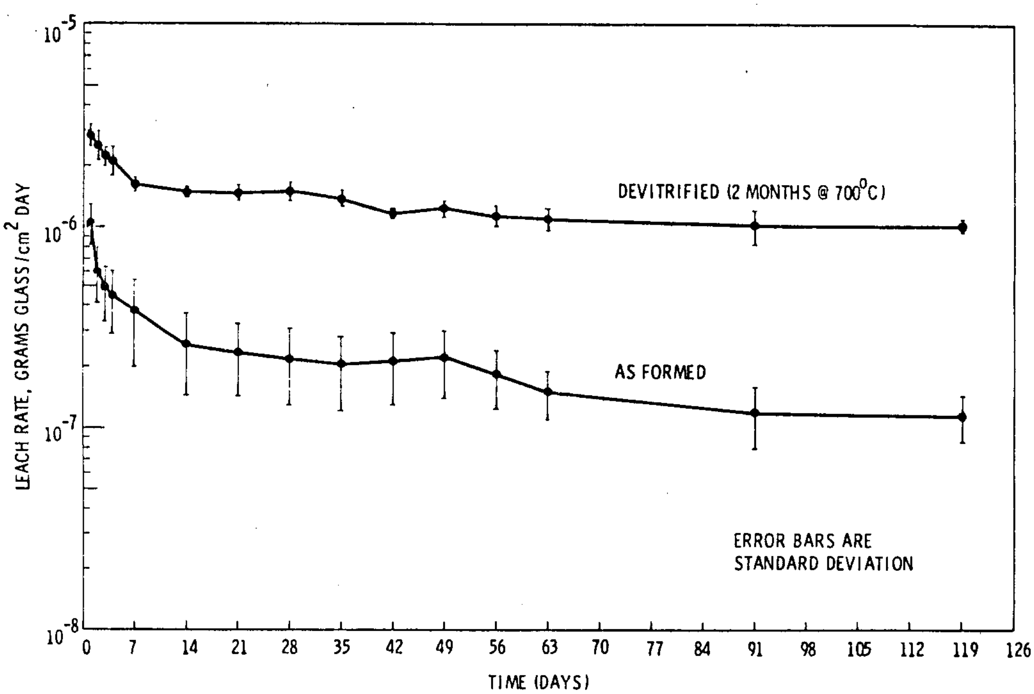

FIGURE 2. Leachability of HLW Glass, Normalized to Strontium Behavior

This is also the order of increasing second phase concentrations (i.e., Pd, RuO_2, CeO_2), which serve as nucleation sites for crystal growth. Isothermal ingrowth curves for frit 73-1 yield an activation energy for crystal growth of 39 kcal/mole which is compatible with a diffusion-controlled mechanism. For glass 72-68 a much higher temperature dependence was obtained with E = ∿78 kcal/mole.

The new glass compositions based on calcine PW-8a-1 have been examined after devitrification by the gradient furnace technique. Maximum crystalline concentrations after 1 week anneals occurred in the range 700 to 800°C and the major phase ingrowing in both 76-68 and 76-76 is a fluorite structure (mainly UO_2). The homogeneity of both glasses in their vitreous and fully devitrified state is substantially better than the high zinc glass 72-68. Elements showing some phase separation (via x-ray mapping on the microprobe) are listed in Table 2.

TABLE 2. Elements Showing Phase Separation, Seven Day Anneals, 1050°C Melt

76-68 400°C	76-68 700°C	76-76 325°C	76-76 700°C
Ru[a]	Ru[a]	Ru[a]	Ru[a]
Ce[b]	Fe[c]	U[b]	Fe[c]
	Ni[c]	Ni[c]	Ni[c]
	U[b,d]		U[b,d]
	Ce[b,d]		Ce[b,d]
	La[d]		La[d]
	Mo		Mo

(a) Both Ru and RuO_2 in 76-68, mainly Ru metal in 76-76.
(b) Fluorite type MO_2, a_0 = 5.45Å, high uranium.
(c) Spinel phases M_3O_4, at least two compositions.
(d) Dendritic crystals/ hexagonal crystals(?)

WASTE VAPORIZATION STUDIES (W. J. Gray and S. A. Gallagher[a])

The purpose of this study is to investigate the vaporization behavior of fission product-containing wastes during processing, shipment, and storage. Accident conditions involving high temperature and breach of the container during shipment and storage are of particular interest.

Some of the earlier[6] vaporization work on PW-4b calcine has been redone to verify the analytical results obtained on the vapor which was collected. Figure 3 shows some of the recent results, which are somewhat different from earlier results. A different calcine batch was used, but the primary difference is believed due to improved analytical procedures. A document describing the results of all vaporization studies to date will be issued shortly.

Table 3 lists weight losses of several materials at 1200°C in dry air and includes previous data on 72-68 glass for comparison. All materials, except the glass, were hot-pressed pellets. Interestingly, the disilicate ($CsAlSiO_4$) lost less than the pollucite ($CsAlSi_2O_6$), and adding sodium and potassium to the pollucite had no effect. It is also interesting to note that the small addition of pyrex to the supercalcine increased the weight loss only slightly.

RADIOLYTIC GAS GENERATION (G. L. Tingey and W. D. Felix)

Significant pressures of gas will be generated in nuclear waste materials containing residual moisture or nitrate compounds. The gases are released either by thermal decomposition or radiolytically from the ionizing radiation emanating from the radioactive waste materials.

We have measured the rate of release of gas from a simulated nuclear waste calcine (PW-7-2) containing up to 7 wt% volatile material. The radiolysis was conducted in a ^{60}Co gamma flux and the rate of gas generation was measured over a 23-g sample of the waste material. The rate of gas

(a) NORCUS student from Pennsylvania State University

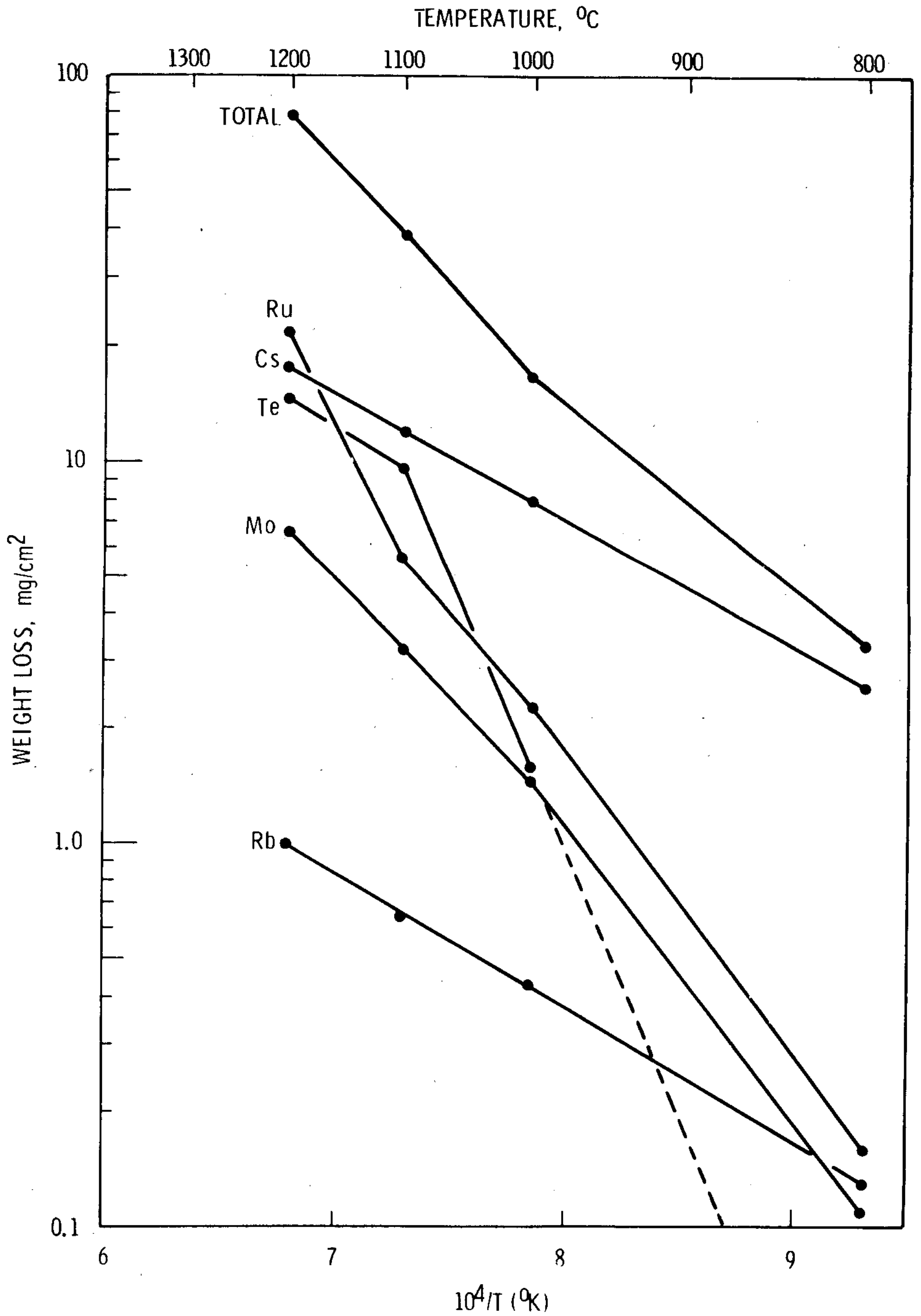

FIGURE 3. Weight Loss from PW-4b Calcine After 4 hr in Dry Air

TABLE 3. Weight Loss of Waste Materials After 12 hr at 1200°C

Material	Weight-Loss, %
$CsAlSiO_4$	0.24
$CsAlSi_2O_6$ (Pollucite)	0.43
$(Cs_{0.74}Na_{0.13}K_{0.13})AlSi_2O_6$	0.43
Supercalcine[a]	0.86
Supercalcine[a] + 10% Pyrex	1.11
72-68 Glass	8.25

(a) Formulation 75-2 developed by Dr. Greg McCarthy at Pennsylvania State University

evolution was determined to be a linear function of dose rate in the range from 5×10^5 to 2×10^7 R/hr. The gas generated was analyzed and shown to be essentially pure oxygen. This finding is consistent with published data[7] on the decomposition of inorganic nitrate compounds following the net chemical reaction:

$$NO_3^- \longrightarrow NO_2^- + 1/2O_2$$

in competition with the reverse process:

$$NO_2^- + 1/2O_2 \longrightarrow NO_3^-.$$

The rate of oxygen generation as a function of time (see Figure 4) decreases as the pressure increases, but does not appear to saturate until several atmospheres pressure is achieved. At a pressure of 1000 torr, oxygen is being generated at a rate of about 0.23 cm^3 of gas (STP)/g of waste-hr in a flux of 2×10^7 R/hr. If one assumes a flux of 5×10^6 R/hr for a waste calcine and the 8.5 torr/hr rate were to continue, the pressure in a

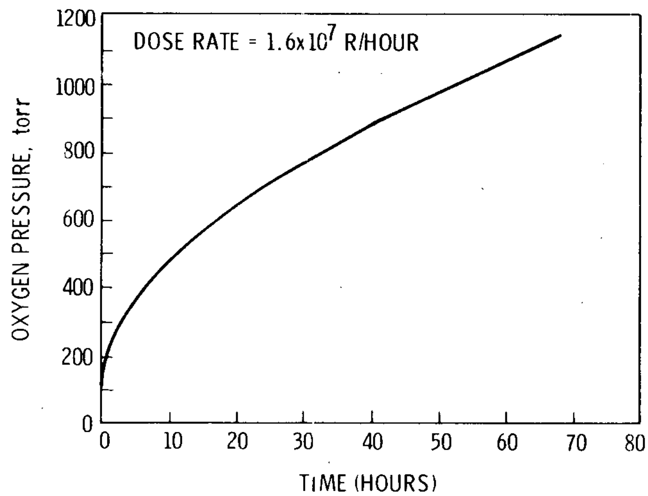

FIGURE 4. Radiolysis of PW-7a Simulated Nuclear Waste

canister with 0.5 cm^3 free volume/g of waste calculates to be about 45 atmospheres after 1 yr. This pressure is the maximum expected because the pressure generation rates appear to continue to decrease as the oxygen pressure increases. Determination of the actual pressure achieved will depend on geometry, flux, and character of the waste material.

FULL-LEVEL RADIOACTIVE TESTS (Y. B. Katayama)

Fully radioactive waste glass specimens are being prepared on a laboratory scale. The off-gas from melting the radioactive glass is being analyzed. Characteristics of fully radioactive glass specimens will be compared with nonradioactive glasses of similar chemical composition as a function of storage conditions.

Four 100-g batches of glass were made during the quarter. The macroporosity of the glass was found to be dependent on the heat-up or out-gassing rate. Slowly heated frit/calcine mixtures resulted in glasses with low macroporosity and low weight losses. The ^{106}Ru losses were 0.05 wt% of the ^{106}Ru in the feed, and the ^{137}Cs losses were 0.36 wt% of the ^{137}Cs in the feed.

FULL-LEVEL RADIOACTIVE GLASS CHARACTERIZATION (D. J. Bradley and C. E. Bigelow)

A gamma spectroscopy unit is being designed to analyze high-level glass and low-level leach samples being generated in the HLW glass characterization program.

LEACHING OF IRRADIATED LWR FUEL PELLETS (Y. B. Katayama)

Irradiated fuel fragments are being leach tested and the results correlated with similar information about common solidified HLW forms.

Leach tests were conducted on enriched UO_2 fuel fragments, with 5.81 wt% ^{235}U, that had been removed from LWR fuel rods irradiated to an average burnup of 54,500 MWD/MTU. The test results are correlated with similar information about common solidified HLW forms in a report entitled "Leaching of Irradiated LWR Fuel Pellets in Deionized and Typical Ground Water," BNWL-2057.

The relative leachability of the elements was found to decrease in the order of Cs > Sb > Sr + Y > Pu > Cm.

Two release mechanisms were observed for each radioisotope over the time interval studied. The short-term mechanism was linear for ^{244}Cm and logarithmic for the other radioisotopes. This short-term mechanism ranged in duration from 5 to 31 days. In every case, the long-term mechanism was logarithmic and could be fitted to the following expression:

$$\text{Fraction released} = Bt^m.$$

The value for m was found to be 0.07 for Hanford ground water, 0.31 for deionized water, and 0.06 to 0.35 for building distilled water. The deionized water was a reference leachant. The Hanford ground water represented typical arid ground water where the ionic species retarded the effective diffusional release of the radioisotope to the leachant. The building distilled water was a comparison leachant to the WSEP leach tests and resulted in intermediate leach rates.

SECTION 3 - ALTERNATIVE WASTE FIXATION PROCESSES

The goal of this task is to develop alternative waste fixation processes and evaluate their waste form products to assure that viable backup processes to the mainline processes will be available if needed. The alternative processes and products are to be compared for cost and safety to the current reference process and product, silicate glass castings in large metal canisters. The alternative processes are being developed on the laboratory scale. In the concept currently emphasized the waste is formed into small granules or pellets which are coated with nonradioactive inert materials to provide containment and leach resistance. The coated waste shapes are then incorporated into a metal matrix which provides impact resistance and increased thermal conductivity.

COATING DEVELOPMENT (M. F. Browning)[a]

The purpose of this study is to develop coating materials and processes that can be applied to waste particles to serve as an impermeable barrier to the release of radioactivity. Current efforts are being directed toward duplex, PyC/Al_2O_3, coatings on 1- to 10-mm pellets.

The feasibility of coating 5- to 5,000-μm calcined radioactive waste "stand-in" particles by a chemical vapor deposition process using a fluidized-bed technique has been demonstrated. Alumina coatings have been applied to the larger particles by a modified chemical vapor deposition/fluidized-bed system which utilized mechanical vibration to augment the particle agitation normally obtained in a fluidized bed. Several approaches to obtain improved coatings on the large particles have been under investigation including:

- a modification of the vibration system to increase particle movement
- a redesign of the reactor to improve particle circulation

(a) Battelle Columbus Laboratories.

- an increase in flow and improved preheat of the fluidizing gas to more closely simulate a conventional fluidized bed particle circulation pattern.

As a result of the changes listed above, the following Al_2O_3-coated particle lots were prepared according to specifications given in Table 4. These particles, which exhibited promising oxidation protection, were shipped to PNL for their evaluation. These lots were evaluated by heating at 750°C in air. Weight changes were monitored during the test period, but the interim changes were so slight that only the total change over the entire test period is given in Table 5. Photomicrographs are shown in Figures 5, 6, 7.

TABLE 4. Al_2O_3-Coated Particle Lots Prepared at Battelle Columbus

Lot No.	Type	Dia, mm	Coating Temp., °C	Coating Thickness, µm	Figure Number
31917-96-44	PW-4a, 50% 75-75 Frit	1	825	33	5
31917-98-45	Supercalcine	1	1025	38	6
32556-4-46A	Supercalcine	2.5	1025	68	7

TABLE 5. Weight Changes Observed on Heating Al_2O_3-Coated Particles

Specimen	Time, hr	Weight, g		Change, %
		Initial	Change	
31917-96-44	194	1.8618	-0.0010	-0.05
31917-98-45	134	2.0132	+0.0004	+0.02
32556-4-46A	203	3.9537	+0.0055	+0.14
PW-4a[a]	43	1.5614	+0.0128	+0.82
Supercalcine[b]	134	1.4986	+0.0290	+1.94

(a) Substrate for Lot No. 31917-96-44
(b) Substrate for Lot No. 31917-98-45

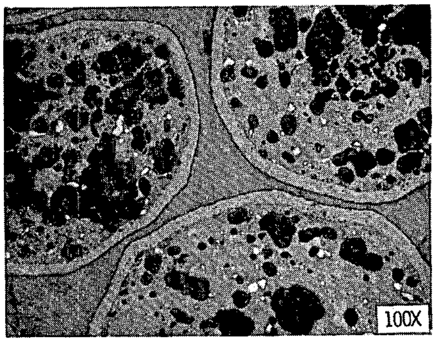

FIGURE 5. Nominal 1-mm Calcined Waste (PW-4a, 50% 75-75 Frit) Coated with ∿33 μm of Al_2O_3 at 825°C (No. 31917-96-44)

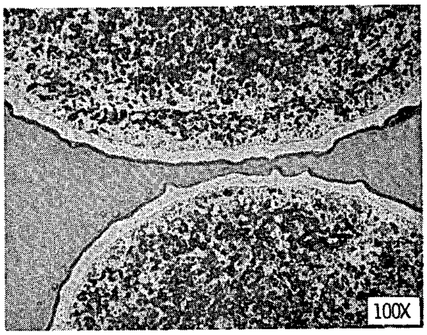

FIGURE 6. Nominal 1-mm Supercalcine Waste Coated with 38 μm of Al_2O_3 at 1025°C (No. 31917-98-45)

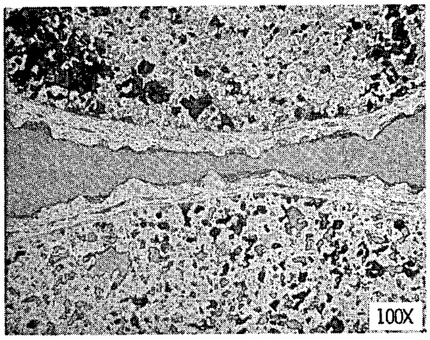

FIGURE 7. Nominal 2.5-mm Supercalcine Waste Coated with 68 μm of Al_2O_3 at 1025°C (No. 32556-4-46A)

Additional investigations were aimed at the development of a duplex coating consisting of a carbon inner coating to take advantage of the superior leach properties of this material, and an Al_2O_3 overcoat to protect the carbon from oxidation in the event of an accidental fire.

Pyrolytic carbon (PyC) coatings were applied to the nominal 1-mm supercalcine particles at 950 to 1050°C by the thermal decomposition of C_3H_8. Deposition was initiated at the lower temperature and completed at the higher temperature to improve the deposition rate. The coatings appeared continuous, were of good integrity, and were noted to have infiltrated the porosity of the supercalcine material to a considerable extent. A total PyC coating thickness of ≈26 μm was applied.

The PyC-coated material was then overcoated with Al_2O_3 at 1025°C by the hydrolysis of $AlCl_3$ in a manner similar to that used in preparing lot No. 31917-98-45. An Al_2O_3 coating thickness of nominally 46 μm (range 16 to 73 μm) was obtained. The coating thickness range is much greater than typically obtained in a chemical vapor deposition/fluidized-bed process, and the cause has yet to be determined. The Al_2O_3 coatings were of relatively high density, but a significant portion were cracked. It appears that the cracking is related to the relative coating thickness of the PyC and Al_2O_3 coatings with the thicker PyC and thinner Al_2O_3 coatings predominant in those particles having cracked coatings. A mismatch in the thermal expansion of PyC ($\sim 6 \times 10^{-6} C^{-1}$) and Al_2O_3 ($\sim 8 \times 10^{-6} C^{-1}$) is thought to be responsible for this cracking. On smaller particles, this difference has not been sufficient to crack the Al_2O_3 coatings on cool-down from deposition temperature, but on these larger particles it appears to have presented a problem. However, the fact that only a portion of the Al_2O_3 coatings cracked would seem to indicate a borderline situation that could possibly be overcome by reducing the PyC coating thickness and overcoating the particle with a stronger more uniform (>30 μm) Al_2O_3 coating.

An investigation has begun to find a source other than $AlCl_3$ for the deposition of Al_2O_3. Potential candidates involve the thermal decomposition of materials of the type aluminum ethylate, propylate, and butylate in the range 550 to 650°C. Other possibilities include the reaction of trimethylaluminum with nitrous oxide at $\sim$650°C and the hydrolysis or oxidation of triisobutyaluminum in the range 400 to 600°C.

CHARACTERIZATION OF COATED WASTE PARTICLES (J. M. Rusin)

Coated cores of Lots 96-44 (pure calcine) and 98-45 (supercalcine), as described in the last section, have been heat-treated at 900°C for 72 hr in air. After 24 hr at 900°C, approximately 10% of Lot 96-44 either cracked or displayed a reaction between the core and coating; after 72 hr, over 90% of the coatings failed. The surface coating of Lot 98-45 was not affected by the heat treatment. Both lots were also heat-treated at 1050°C. Coatings on Lot 96-44 failed within 2 hr, whereas only 5% of Lot 98-45 failed at 72 hr. This attests to the high thermal stability afforded by the supercalcine cores.

Compressive strength has been determined for Lots 98-45 and 96-44, for uncoated cores, and for PyC- and SiC-coated simulated waste. Individual cores were tested on the Instron using a ram speed of 0.002 in./min. For comparison, cores were assumed spherical using the ram gap as the approximate diameter. The results are listed in Table 6.

TABLE 6. Compressive Strength of Particles

Core	Coating	Diameter, in.	Compressive Strength, psi
Calcine	Uncoated	0.0503 ± 0.0060	3050 ± 1121
Calcine	33 μ Al_2O_3	0.0312 ± 0.0027	6175 ± 1672
Supercalcine	Uncoated	0.0576 ± 0.0032	4409 ± 3877
Supercalcine	39 μ Al_2O_3	0.0625 ± 0.0023	7290 ± 3473
OR-2340	30 μ Pyc	0.0170 ± 0.0018	25807 ± 4738
Ga 5768-141	200 μ PyC 18 μ SiC	0.0110 ± 0.0020	10599 ± 6196
Ga 6285-101	60 μ PyC	0.0126 ± 0.0027	21006 ± 10377

Coating the particles with Al_2O_3 increases their strength by nearly a factor of two. The compressive strength of PyC- and SiC-coated waste is greater than Al_2O_3-coated cores. In comparing strengths, one must note the differences in coating thickness and core diameter (cores were assumed spherical). Further strength studies will be conducted on PyC- and PyC+Al_2O_3-coated supercalcine cores and Al_2O_3-coated supercalcine cores impregnated in a metal matrix.

The Al_2O_3-coated cores were leach tested using three solutions: distilled water at 100°C (Soxhlet), basic (pH9) and acid (pH4) at 25°C. One-gram samples of the basic and acidic solutions were agitated and weight measurements were taken at three 25-hr periods. Percent weight change is shown in Table 7.

TABLE 7. Leach Rates of Al_2O_3-Coated Particles

		% Weight Loss		
	Time Period, hr	pH9, 25°C	pH4, 25°C	Soxhlet, 100°C
SUPERCALCINE CORE 31917-98-45				
	1st 24	+0.05	0	-0.02
	2nd 24	0	0	0
	3rd 24	+0.01	0	+0.07
	72 total	+0.06	0	+0.05
CALCINE CORE 31917-96-44				
	1st 24	+0.37	+0.60	+0.50
	2nd 24	+0.13	+0.14	+0.21
	3rd 24	+0.08	+0.25	+0.21
	72 total	+0.48	+0.99	+0.92

The weight gain for the supercalcine is insignificant (0.05% = 0.5 mg), whereas the gain for the calcine is significant. SEM photomicrographs illustrate a total loss of Al_2O_3 coating surface structure on the calcine upon leaching. Besides differences in core materials, the coatings differ in application temperature. The 825°C coating contains kappa and gamma alumina, whereas the 1025°C coating contains only alpha alumina.

GLASS MARBLE PRODUCTION (J. M. Rusin)

The purpose of this work is to develop methods to produce and encapsulate approximately 3 kg of HLW glass marbles in a metal matrix.

Small quantities of marbles have been produced at PNL and by an outside subcontractor. The marbles were formed by vibrating remelted ICM glass in graphite molds. The marbles produced at PNL were formed from glass remelted at 1200°C, which resulted in a spherical marble approximately 3/8 in. diameter. Lower melting temperatures (∿1100°C) were used in some cases which resulted in an ellipsoidal-shaped marble approximately 5/8 by 3/8 in.

The marbles will be encapsulated in a metal matrix by gravity sintering or casting. Optimum marble diameter, matrix material selection, and matrix loading will be determined from heat flow calculations. Major factors will be waste loading and softening point of the glass, and thermal conductivity of the composite. Preliminary data will be obtained on marbles in a copper metal matrix, and impact strength and thermal conductivity of the single components and composite will be measured.

METAL MATRIX (K. R. Sump)

The purpose of this study is to develop a metal matrix for increasing the thermal conductivity and impact strength of containers of HLW.

Gravity sintering of metal powder is being studied as a means of fabricating the metal matrix around HLW particles. Matrices fabricated by this process can withstand higher temperatures than those prepared by casting. In addition, this process is readily adaptable to hot cell use.

Sintered compression specimens of brass, copper, electrolytic iron, steel, and stainless steel were fabricated and tested. The 53% dense stainless steel was the strongest. These specimens did not contain any simulated waste particles.

Pure Al_2O_3 spheres, uncoated supercalcine, uncoated calcine-frit particles, and Al_2O_3-coated supercalcine materials were put in matrices.

CALCINE CORE (PELLET) DEVELOPMENT (A. A. Garrett)

The purpose of this task is to evaluate the disc pelletizer as a means to form and coat pellets (cores) of calcine powder which can be incorporated in a metal matrix.

Approximately 100 lb of various sized cores (pellets) were produced using the disc pelletizer during April, May, and June 1976. PW-7a and PW-4b calcine powders mixed with different glass frits were used to form the cores. Several liquid and solid binders were evaluated, but water appears to be an acceptable binder in most instances.

Waste oxide loading up to about 80 wt% produced good, high density cores when sintered to 1100°C.

About 5 lb of the sintered cores were fed back into a clean disc pelletizer along with Pyrex frit. The frit formed a uniform coating on each core. Coating thicknesses between 200 and 1000 μ were obtained. The coated cores were sintered to partly vitrify the coatings. The resulting coated cores are quite rugged and leach resistant.

Figure 8 shows a cross section of one core with a 1000 μ pyrex frit coating. There appears to be a good bond between the core and the coating. Thermal expansion characteristics have not been fully investigated, but, in general, the coating and core appear to be compatible in terms of thermal expansion. Preliminary findings also indicate the coatings have a low porosity.

SUPERCALCINE (G. J. McCarthy)[a]

The Materials Research Laboratory at Pennsylvania State University, University Park, PA, is participating in this program through an investigation of crystalline ceramic waste forms, which are potential advanced options

(a) Pennsylvania State University, University Park, PA

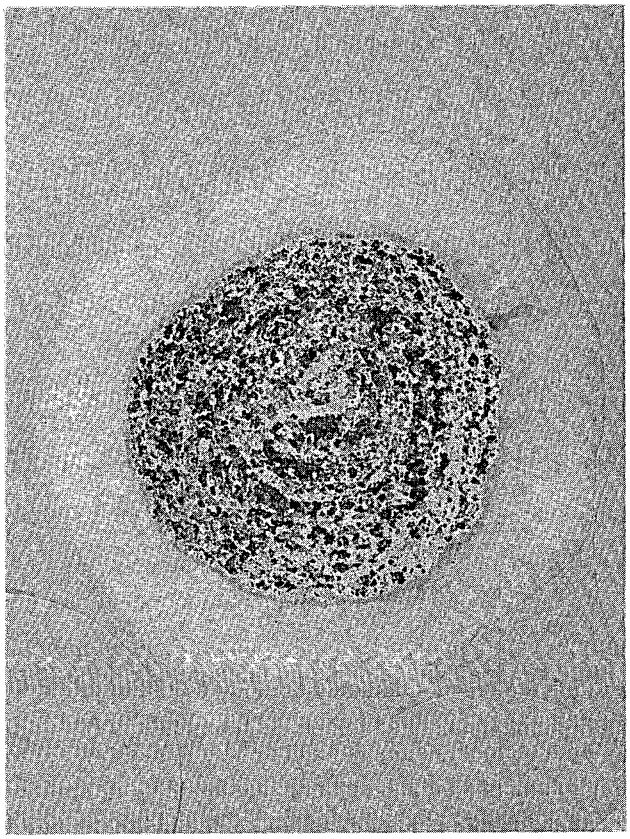

FIGURE 8. PW-4b Pellet Coated with Pyrex Frit (8X)

to melt-formed HLW glass or glass-ceramics. The objective of this study is to develop a waste form in which all of the radioactive atoms will be isolated in thermally and chemically stable crystalline phases. Because this waste form uses the existing calcining processes with a compositionally modified waste liquid it has been termed "supercalcine." Although supercalcine could be considered alone as an improved calcine product, current emphasis is on its use as high stability "cores" in the multibarrier waste form.

CRYSTALLINE PHASES FOR CESIUM AND RUTHENIUM FIXATION (S. A. Gallagher, R. G. Johnston and G. J. McCarthy)

The purpose of this study is to determine the best crystalline phases for cesium, ruthenium and tellurium fixation.

Emphasis during the last quarter was on further critical examination of cesium and ruthenium fixation phases for supercalcine and on initiation of work on developing supercalcine formulations for high-sodium wastes such as PW-7a and PW-8a.

A cesium fixation phase must prevent cesium volatilization during supercalcine crystallization firings. Thermogravimetric analysis on cesium aluminosilicates was performed by S. A. Gallagher during her 3-month stay at PNL. Pollucite, $CsAlSi_2O_6$, appears to have more than adequate thermal stability for cesium fixation in supercalcine.

Based on thermal stability characterization of HLW calcine and glass products, it appeared that ruthenium volatilization during supercalcine crystallization was going to be a major problem. We thus set the development of a ruthenium phase as one of the first objectives in supercalcine studies. After an extensive survey of candidate phases and solid solutions, $SrRuO_3$ was chosen as the best possibility. However, in spite of earlier promising, but preliminary results, $SrRuO_3$ is not suitable as a ruthenium fixation phase. Strontium oxide has a greater affinity to form aluminosilicates than to form $SrRuO_3$. This means that added SrO can prevent complete cesium fixation as pollucite during the supercalcine crystallization firing. The possibility of leaving ruthenium as RuO_2 during supercalcine crystallization is being assessed, since residual RuO_2 was observed routinely in most phase assemblages after firings of up to 3 hr at 1200°C.

SUPERCALCINE FOR HIGH SODIUM WASTES (G. J. McCarthy, D. E. Pfoertsch and M. T. Davidson)

The purpose of this study is to develop supercalcine formulations containing all nonradioactive commercial waste atoms.

A technical progress report on supercalcine development was prepared and issued during the quarter.(8) With the very promising results from the cold engineering-scale demonstration of a supercalcine formulation based on PW-4b,(9) we have been asked to investigate the application of supercalcine concepts to the high sodium, "dirty", wastes PW-7a and PW-8a. Nepheline, $NaAlSiO_4$, looks like the best candidate for sodium fixation. Extensive modifications to the phase formation models will be necessary because of the high sodium concentration. For example, in the presence of high nepheline concentrations, the scheelite structure phase, $SrMoO_4$, reacts to form a cubic sodalite structure phase, $Sr_2Na_6Al_6Si_6O_{24}(MoO_4)_2$ or $(6NaAlSiO_4 \cdot 2SrMoO_4)$. This type of phase has also been observed as one of the products in the devitrified glass-ceramic HLW product under investigation at the Hahn-Meitner Institute.[10] The rare earths still crystallize in an apatite structure phase, but this phase appears to have a substantial sodium concentration.

Based on observations during several complex compatibility studies, it appears that good overall thermal stability at as high as 1200°C might be attainable for nepheline-based supercalcine formulations.

ROLE OF TELLURIUM IN SUPERCALCINE (D. E. Pfoertsch and G. J. McCarthy)

An extensive literature search for relevant oxide crystal chemistry of tellurium has been completed. At high temperatures, tellurium is usually hexavalent and almost always in sixfold coordination with oxygen. No information was found on the potential solid solubility of tellurium in apatite or scheelite structure oxide phases. A study of this possibility has been initiated.

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3 Exxon Nuclear Company

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60 Battelle-Northwest

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