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**Quarterly Progress Report
Research
and Development Activities
Waste Fixation Program
October Through December 1977**

J. L. McElroy

March 1979

**Prepared for the U.S. Department of Energy
under Contract EY-76-C-06-1830**

**Pacific Northwest Laboratory
Operated for the U.S. Department of Energy
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QUARTERLY PROGRESS REPORT
RESEARCH AND DEVELOPMENT ACTIVITIES,
WASTE FIXATION PROGRAM²
OCTOBER THROUGH DECEMBER 1977

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SUMMARY

The objective of the Waste Fixation Program is to develop processes for converting high-level waste to solid forms that are demonstrated to be physically, chemically, and radiologically stable. This progress report describes the research and development activities for October through December of 1977:

- Foaming was experienced in the joule-heated ceramic melter during conversion of simulated Savannah River Plant waste to glass. The foaming tendency was decreased by adding the glass-making additives as chemical compounds rather than as pre-formed glass.
- Operation of the 36-in.-dia spray calciner with a 300 l/hr feed rate was demonstrated. Over 300 hr of operating experience has now been accumulated with this unit.
- The variation of leach rate with pH was measured on four typical waste glasses over the pH range of 1 to 13. All four glasses had uniformly low leach rates over the range of pH 4 to pH 9--the pH range of most natural waters.
- A series of waste glass samples were prepared in bead form and were spiked individually with ^{99}Tc , ^{233}U , ^{237}Np , ^{239}Am , and ^{244}Cm . These will be used in the Waste Isolation Safety Assessment Program to study the effect of various naturally occurring waters on leach rate.
- Demonstration laboratory encapsulations of four multibarrier waste forms on a 1-liter scale were completed. The waste forms were: 1) uncoated supercalcine in a vacuum-cast Al-12Si matrix, 2) $\text{PyC}/\text{Al}_2\text{O}_3$ -coated supercalcine in a gravity-sintered copper matrix, 3) glass-coated supercalcine in a vacuum-cast Al-12Si matrix, and 4) simulated waste glass marbles in a vacuum-cast Pb-10Sn matrix.

PREVIOUS REPORTS

Previous reports in this series were BNWL-1699, 1741, 1761, 1788, 1809, 1826, 1841, 1893, 1908, 1932, 1949, 1994, 2070, 2242, 2243, 2264, and PNL-2265-1, 2265-2, and 2265-3.

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QUARTERLY PROGRESS REPORT
RESEARCH AND DEVELOPMENT ACTIVITIES
WASTE FIXATION PROGRAM
OCTOBER THROUGH DECEMBER 1977

INTRODUCTION

The Waste Fixation Program (WFP) is conducted by the Pacific Northwest Laboratory (PNL), operated by Battelle Memorial Institute for the Department of Energy (DOE). Under this program, PNL is conducting research to convert high-level radioactive waste to stable, nondispersible forms. Candidate waste forms include silicate glasses and various crystalline and multibarrier waste forms. The WFP is designed to be a means through which the government and users of the technology can cooperate to effectively handle nuclear waste. Objectives of the comprehensive program include: the development and characterization of glass formulations; equipment and process development; and design, construction, and demonstration of full-scale process equipment. The following sections describe research and development activities in radioactive waste fixation for the past quarterly reporting period.

SECTION 1 - WASTE FIXATION PROCESS DEVELOPMENT

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

JOULE-HEATED CERAMIC MELTER - C. C. Chapman and J. L. Buelt

A glass melter capable of processing 2 tonnes of glass per day has been designed, and fabrication/installation is in progress. The unit will couple directly to the large-scale spray calciner. Startup is planned for April of 1978. A conceptual design for a high-temperature ceramic melter that uses tin oxide electrodes is in progress.

The only ceramic melter that was operational during this reporting period is the liquid-fed (LF) ceramic melter. Various aqueous feeding tests have been made. A general conclusion we have reached as a result of these experiments is that the chemical form of the glass formers in the aqueous feed is important to processing. It appears that when glass formers are added as premelted frit, a trend toward bridging and unstable operation results. If the glass formers are added as unmelted chemicals, smoother operation is achieved. Thus it appears that, as with conventional glass melting, "batch chemistry" is also important in an aqueous feeding process.

Since operation with simulated Savannah River Plant (SRP) waste we have experienced foaming in the LF melter both during actual operation and when simply heating the melter up without feeding. This unusual phenomenon may be explained as reboil and/or chemical reaction; for example, it could be caused by iron going from a +3 to +2 oxidation state. An iron-nickel spinel has been discovered on the floor of the melter. Further examination will be required to determine if the foaming is related to the presence of spinel.

SPRAY CALCINATION - W. J. Mikols

Progress this quarter was marked by completion of three more experimental runs on large spray calciner equipment. Successful operation of this equipment has been demonstrated at feed rates of 300 l/hr. Product characteristics and the effects of calciner operation temperature have been studied for this large-scale unit. To date over 300 hr of operating experience have been accumulated, and over 27,500 l of simulated liquid feed have been processed.

IN-CAN MELTING DEVELOPMENT - H. T. Blair

This quarter we maintained operation of the multizone in-can melter (ICM) throughout the continuous ten-day running of the full-scale spray calciner, and produced 5055 kg of vitrified, simulated high-level commercial waste in ten 12-in.-dia cans. The 7-ft-tall cans were bottom-supported and located completely within the 7.5-ft-tall furnace chamber. As a result, there were no problems with batch accumulating at the top of the fins, and the entire content of each can was vitrified. The frit-feeder airlock, the batch diverter, and

the water-cooled spout that conveys batch into the mouth of the can all performed well during the ten days. The gravimetric frit feed drifted from the set point during the last day of operation. This piece of process equipment, which will be located outside the cell in a waste solidification facility, must be cleaned regularly. The load cell system did not perform well because objects that had fallen to the bottom of the furnace had become wedged between the force-transmitting plug and the furnace shell. We modified the furnace design to shield the joint from further obstructions.

The most significant accomplishment with the ICM during the ten-day run was the processing of two cans of vitrified, simulated waste without any instrumentation in or on the can and without the aid of visual observation of the interior of the can. The ICM process was monitored using only a gamma transmission (movable ^{60}Co source) glass level detection system, the furnace control thermocouples, the load cell system, the frit feeder controls, and the spray calciner liquid feed rate controller.

An overall average melting rate of 40 kg/hr was demonstrated with the ten 12-in.-dia cans, and an average rate of 61 kg/hr was attained in one of the 16-in.-dia cans. The frit feeder malfunction occurred during the filling of the other 16-in.-dia can.

We began evaluating application of the ICM process to vitrification of SRP-type defense wastes by running two different batch simulations through pilot-scale equipment using 8-in.-dia cans containing 8 fins each. One batch was a blend of fine ground frit and dry chemicals used to simulate a calcined liquid waste composition, and the other batch was a blend of coarse frit and calcine produced in the pilot-scale spray calciner from a simulated waste sludge. Foaming of the melt was observed for both batches, but it was more severe with the batch made from the dry chemical blend. The foaming of the melt in the ICM caused the melting rates demonstrated to be only 1/3 to 1/2 of the 30 kg/hr demonstrated with commercial waste compositions in 8-in.-dia cans containing fins. However, the foam did melt away from the fins and collapse rapidly when the batch charging rate was reduced or terminated. The product of these two tests was a vitreous material free of bubbles and devitrified phases.

This quarter we also ran a test in the full-scale ICM using a 12-in.-dia can to evaluate the effect on the melting rate caused by adding 1.5 wt% powdered silicon to the batch to prevent the formation of alkaline molybdate phases on the surface of the melt. Because all of the cans produced during the ten-day run were fed batch that contained silicon, this can was fed batch which did not. The results indicate that the contribution made to the ICM melting rate by adding 1.5 wt% powdered silicon to the batch is not significant enough to be detected in the full-scale demonstration equipment. This and other results obtained in the pilot-scale ICM indicate that melting rate is not just a function of heat transfer. It has been suggested that mass transfer may also be a controlling factor.

To demonstrate an ICM capacity sufficient to meet the requirements of a defense solidification facility, we used a 24-in.-dia can that contained a fin assembly. The fin assembly consists of ten equal-length radial heat transfer plates of a special configuration designed to maximize heat transfer into the can contents. The can was filled with 5.25 ft of a simulated commercial waste composition glass in 12 hours. The average melting rate

for the run was 85 kg/hr, and we achieved a maximum rate of approximately 100 kg/hr early in the run. Experience with the 7-ft-high multizone ICM has shown that the melting rate decreases as the melt level rises in the can because the area of hot surfaces upon which melting occurs is being reduced. The new melting rate data obtained this quarter are presented in Figure 1.

FIGURE 1. In-Can Melter Capacity

CANISTER DEVELOPMENT PROGRAM - S. C. Slate

This quarter we began to formulate a canister handling flowsheet. This flowsheet will describe all canister operations, from fabrication through interim storage at the solidification facility. The descriptions will serve as criteria for the design of canister processing equipment. The flowsheet will also include acceptance criteria and testing requirements for the canister.

We completed the first series of impact tests and one overheat test this quarter. In the impact tests, 12 full-scale canisters were dropped from various heights and at different orientations onto a 6-in. steel plate backed by a 4-ft x 10-ft x 10-ft block of concrete. There was no apparent failure of any of the prototypical canisters. The overheat test simulated a loss-of-cooling accident with an open canister. The canister's integrity was maintained, and no glass degradation was noted.

Work has continued on the development of a canister closure system. We received the rotating TIG closure welder this quarter. Acceptance testing at the vendor's plant was successful. Detailed testing of the welder will continue at PNL using actual and mock-up twist-lock lids.

We also investigated the use of inertia welding as a closure process. This process is being studied because of the potential advantages it has over other processes. Some of the advantages are: 1) consistent, reproducible welds on parts that can be oxidized; 2) relaxed dimensional tolerances; and 3) relaxed setup requirements. Twenty sample welds were made in this study. Three joint configurations were made using both stainless and carbon steels. All but one weld was found to be acceptable. The one unacceptable weld, done on heavily oxidized carbon steel, failed because the parameters for welding oxidized parts had not been developed.

Two papers were issued this quarter on our stress analysis work. The first paper describes the results of the high-temperature creep test we made on two candidate canister materials.⁽¹⁾ The second paper describes the analyses made on the twist-lock canister closure design.⁽²⁾ New work this quarter included the development of a computer model of glass/canister interaction. We expect this model to provide insight into the cracking of the glass and the residual stress found in the canister.

EFFLUENT STUDIES - M. S. Hanson

The laboratory-scale spray calciner, designed for use in effluent studies, was successfully put through four shakedown runs. Feed rates of 0.5 to 2 l/hr have been obtained. The low flow rates are obtained by use of a recycle feed loop.

The efficiency of sintered metal filters, as measured with a classical scattering aerosol spectrometer, can be controlled by blowback frequency. Using longer intervals between blowbacks reduces the time the filters are uncoated, and consequently limits particle penetration.

BATCH CHEMISTRY - J. M. Lukacs

This quarter we developed a laboratory technique to evaluate the foaming potential of a glass batch prior to beginning engineering-scale melting trials. Severe foaming was encountered with SRP-1 feed, and was related to the attack of the frit (21-A) component by Na_2CO_3 . A continuous glassy phase that formed at a temperature as low as 650°C entrained most of the CO_2 that is generally released at higher temperatures. Replacing the frit with batch chemicals reduced foaming by making solid-state reactions possible between Na_2CO_3 and SiO_2 , and by raising glass-formation temperature to 800°C .

SECTION 2 - WASTE FORM CHARACTERIZATION

The purpose of waste form characterization is to measure the properties of candidate solidified products and containers (solidified waste and canister) as functions of composition, processing parameters, and storage conditions. The measurements are used to: 1) ensure operability of the vitrification processes and 2) provide data for safety analyses of high-level waste management. The ultimate goal of waste characterization 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 a final disposal site, we may be fully confident that their behavior is understood and that any changes or interactions with their environments are wholly predictable.

MELT DEVELOPMENT - G. B. Mellinger, E. T. Sherman, and S. K. Jantz

This quarter we examined the changes in the chemical durability of a waste glass caused by changes in the waste composition. The waste studied was PW-7c-1. We ran a series of melts in which the amounts of the materials most likely to vary in the waste stream were increased and decreased. The materials and the amounts by which they were varied are shown in Table 1. The glass frit used was 77-269, and a constant waste-to-frit weight ratio of 1 to 2 was maintained.

TABLE 1. Contents of Various Oxides in As-Defined PW-7c-1 and Content Variations Examined in this Study

Oxide	Amount of Oxide, kg/MTU			
	PW-7c-1	.5 (a)	1.5 (a)	2.0 (a)
Na ₂ O	7.97	3.99	11.96	15.94
Fe ₂ O ₃	3.05	1.52	4.57	6.09
P ₂ O ₅	5.90	2.95	8.85	11.80
Gd ₂ O ₃	25.77	12.88	38.65	
MoO ₃	4.90	2.45	7.34	9.79
U ₃ O ₈	13.74	6.87	20.62	27.48
Rare Earth Mixture ^(b)	9.72	4.86	14.57	19.43

(a) Values represent ratio of grams of oxide in calcine to grams of oxide in PW-7c-1.

(b) A commercial rare earth mixture nominally containing, by wt%, 0.2 Y₂O₃, 24.0 La₂O₃, 48.0 CeO₂, 5.0 Pr₆O₁₁, 17.0 Nd₂O₃, 3.0 Sm₂O₃, 0.8 Eu₂O₃, and 2.0 Gd₂O₃.

The appearance and relative fluidity of each melt was noted and recorded. Leach testing in buffered pH 4 and pH 9 solutions and in a Soxhlet apparatus was performed. We measured the weight loss resulting from each leach test.

The compositional variations had a negligible affect upon the pH 4 and pH 9 leach behavior. The only variations that showed significant increases in Soxhlet leach rate were those changing Na₂O, MoO₃, and U₃O₈. It is encouraging to see that most compositional variations of the glass did not affect leaching behavior; rather wide variations in the waste stream makeup can be accommodated in a single glass type.

THOREX PROCESS HIGH-LEVEL WASTE - G. B. Mellinger

An initial simulated thorex waste calcine was formulated, and initial scoping melts were made containing 20 to 33 wt% of this waste. This calcine poses a fluorine volatility problem. Even though 1.9 wt% of fluorine was added during preparation, only 1.1 wt% was present when the prepared calcine was analyzed. We are investigating causes and solutions to this problem. Based upon the work of Johnson and Rutenbroger⁽³⁾ the fluorine losses may be reducible to 10% or less by complexing the fluoride ion in the initial solution. The fluorine contents of some glasses made with this waste are also being measured. Based upon previous work, losses of ~2 wt% are anticipated.⁽⁴⁾

WASTE GLASS BULK AND THERMAL PROPERTIES - G. B. Mellinger

This quarter we measured the densities of four simulated HLW glasses at temperatures of between 950° and 1300°C. These glasses are 72-68, 77-260, 76-68 and 77-107. The results of the measurements are shown in Figures 2 and 3. The higher density of the 72-68 glass is due to this material's higher ZnO content.

FIGURE 2. HLW Glass Densities as a Function of Temperature

FIGURE 3. HLW Glass Densities as a Function of Temperature

We also completed a preliminary experiment for determining the thermal conductivity of a typical waste glass. Measurements were made at temperatures of between 50° and 1000°C. Also, the effects of devitrification and thermal shock were investigated.

The conductivity of the glass was found to increase approximately linearly with temperature. McElroy, et al. found a sharp increase in the conductivity of another glass at temperatures above 650°C.⁽⁵⁾ They attributed this to convective heat transfer. The extent to which convection contributes to the thermal conductivity of a waste glass at a particular temperature will be primarily dependent upon the viscosity of the glass at that temperature. In certain glasses, convection may contribute significantly above 700°C, and may be the dominant mechanism above 800° to 900°C.

Devitrification was induced by holding the sample at 800°C for 24 hours. We detected no change in thermal conductivity. Two quenching experiments were performed. In the first the sample was cooled down from 600°C in air; no change in conductivity occurred. A water quench was then performed, and a 20% drop in conductivity was observed.

THERMAL EFFECTS UPON STORED GLASS - J. H. Westsik, Jr.

77-107 Glass

Long-term anneals of 77-107 [PW-9-3(1:2)77-268] glass for one day, one week, and two months were completed this quarter. Soxhlet leach rates for these samples are unaffected by storage temperature or time. Figure 4 shows the leach rates as a function of the storage temperature for the two-month anneal. For comparison, the as-formed glass had a Soxhlet leach rate of 4.0×10^{-5} g/cm²-day (1150°C, 2 hr).

FIGURE 4. Soxhlet Leach Rate for 77-107 Glass, 2-Month Storage

77-260 Glass

We have also completed one-day, one-week and two-month high-temperature storage test on 77-260 [PW-7c-3(1:2)77-269] glass. As shown in Figure 5, the storage temperature influences glass leachability. For the two-month period, Soxhlet leach rates were relatively high at storage temperatures centered around 800°C. The length of storage does not change the glass durability beyond formation of the peak at 800°C. The as-formed glass leached at 4.4×10^{-5} g/cm²-day (1050°C, 2 hr).

76-68 Glass

Samples of 76-68 glass have been stored for 1 yr at temperatures ranging from 300°C to 900°C.

Soxhlet leach rates for these samples are shown in Figure 6. The leachability is similar to that reported earlier for samples stored for a maximum of two months at the same temperatures. (6)

FIGURE 5. Soxhlet Leach Rate for 77-260 Glass, 2-Month Storage

FIGURE 6. Soxhlet Leach Rate for 76-68 Glass, 1-Year Storage

PHASE BEHAVIOR - J. W. Wald

We have finished our quantitative examinations of crystallization behavior in glass 76-68 for long-term anneals (up to 2 mo anneal time), and have begun some limited short-term, kinetic-type studies. The 76-68 glass composition continues to exhibit very good phase behavior, as no crystalline phases appear above 10 wt%, while the total crystallinity generally remains below 15%. The phases formed are refractory-type oxides, which are expected to be leach resistant and unaffected by radiation damage.

THERMAL AND MECHANICAL SHOCK - L. R. Bunnell

Previous impact testing has involved impact energies in the range 20 to 160 ft-lb on a glass sample 0.44 in. in dia x 0.50 in. high. This quarter we increased the impact energy to 1400 ft-lb, and BET surface areas were determined as a function of impact energy on soda-lime-silica and ICM-11 glasses. The results, shown in Figure 7, indicate that the surface area for soda-lime-silica glass is roughly linear with impact energy, while in ICM-11 the damage seems to have saturated. This result is of potential importance to the Waste Program, and will be verified using the recently purchased surface area analyzer and some other, more current waste glass formulations.

FIGURE 7. BET Surface Area as a Function of Surface Area for Soda-Lime-Silica Glass and ICM-11

The thermal expansion of two Savannah River glass compositions, SRP-ICM-1 and -2, was determined using a push-rod dilatometer. Heating was in air, at 4°C/minute. The resulting data are shown in Figure 8; the expansion coefficients are essentially identical and are 10%-15% higher than those usually measured on proposed commercial waste glass compositions. Small differences exist in T_g , the glass transition temperature, and T_s , the dilatometric softening point. Both of these parameters are lower than typical proposed commercial waste glass ICM compositions by ~40°C.

FIGURE 8. Thermal Expansion Curves for Two Savannah River Glasses

For purposes of general characterization and in support of canister drop tests, we opened and sampled two large canisters of glass melted in the ceramic melter (CM); canister CM-1 was 16 in. in dia, and canister 2-H had a 36-in. diameter. The glass in both cases was cracked into a series of 3 to 4 annular layers immediately adjacent to the can wall, to which the glass was not bonded. The layers were ~1/16 in. thick in the 16-in. can, and ~1/4 in. thick in the larger one. These layers are partially a result of the canister's not being heated during pouring, and might be lessened by heating the can to about the softening point of the glass during pouring. The glass fragments are progressively larger

proceeding toward the can center, and are 4 to 6 in. in dia there. The surface areas of the fragments are estimated to be not more than 50x the original geometric surface area. However, this estimate may be low, because some dust was found in the badly cracked region near the canister wall (especially in the 16-in. can) and no attempt was made to estimate the additional surface area of this small amount of dust. The dust is apparently glass and is quite fine--perhaps 325 mesh. The origin of the dust is unknown, but it may have originated from the abrasion of fresh fracture surfaces, which in glass have very high friction coefficients. Normal handling and transportation could possibly generate an undesirable quantity of this material; consequently, its origins will be studied further.

ENVIRONMENTAL REACTIONS OF WASTE GLASSES - J. H. Westsik, Jr.

This quarter we measured the durabilities of four commercial waste glasses in aqueous solutions with pHs ranging from 1 to 13. The acid solutions were prepared with nitric acid and the basic solutions were made with NaOH. Weight loss was determined on one gram -42 +60 mesh samples suspended 72 hr in 500 ml of solution. Percent weight loss as a function of pH is shown in Figure 9. The glasses all show excellent durability in solutions, with pHs of between 4 and 9--the pH range of most natural ground waters. The zinc borosilicate glass (containing frit 73-1) has poor durability at high hydrogen ion concentrations.

FIGURE 9. Acid-Base Durability of Commercial Waste Glasses

WASTE ISOLATION SAFETY ASSESSMENT PROGRAM TASK #2; WASTE FORM RELEASE
RATE DATA^(a) - D. J. Bradley

As a compromise between the reality of cracked glass monoliths and the powdered glass frequently studied in accelerated leach testing, we chose glass beads as the sample waste form for study. Two methods of preparing these samples were developed. A resistance-heated unit was installed in a glove box for preparation of doped glasses, and an induction-heated system was set up for hot cell use. Both units involve heating a comparatively large volume of glass to the melt temperature, continuously forming droplets at the end of a nozzle, and collecting the solid beads on a heated metal plate. The beads have a flattened, hemispherical shape well-suited for a geometric determination of their surface areas. The major value of this bead-making approach lies in the ease and reproducibility of sample preparation. Doped glass samples containing ^{99}Tc , ^{233}U , ^{237}Np , ^{239}Pu , ^{243}Am , and ^{244}Cm were produced using these techniques.

Characterization comparisons with undoped and doped glasses were made. Metallography, x-ray diffraction, and alpha autoradiography analyses show that the dopants were homogeneously distributed and that the doping/waste form manufacturing process, both resistance and induction, does not significantly change the glass from its original form with the exception of small increases in porosity.

Leaching tests were started on all of the doped glass beads mentioned above. The leaching procedure for the doped glasses is based on the standard IAEA method and uses five different solutions: 1) salt brine based on the analysis of a deionized water-dissolved core from the WIPP facility (300 g/l NaCl), 2) high-ionic-strength calcium groundwater (2.206 g/l $\text{CaCl}_2 \cdot 2\text{H}_2\text{O}$), 3) high-ionic-strength sodium groundwater (1.76 g/l NaCl), 4) high-ionic-strength bicarbonate groundwater (2.52 g/l NaHCO_3), and 5) deionized water.

The information obtained during FY-77 experiments shows that the leach rates of all nuclide dopants are decreasing with time, presumably as the resistance to diffusion of the radionuclides through the waste-depleted glass matrix near the surface of the beads becomes increasingly dominant. The high-pH bicarbonate solution (pH = 9) shows higher leach rates for all nuclides than do the low pH solutions. This is presumably caused by an increased dissolution rate of the glass matrix itself.

Studies on the effects of leaching at temperatures of up to 90°C and with typical aquifer flowrates have been initiated at Lawrence Livermore Laboratories. The waste form will be the glass beads doped with ^{239}Pu and ^{237}Np and made by the resistance-furnace method discussed above.

(a) This task is funded by WISAP, but is included here for completeness.

SECTION 3 - ALTERNATIVE WASTE FIXATION PROCESSES

The goal of this task is to develop alternative waste fixation procedures that will serve as viable backup processes. Cost and safety factors of the alternative processes and products are to be compared to those of the current reference process and product--silicate glass castings in large metal canisters. These alternative processes are being developed on the laboratory scale. In the concept currently emphasized the waste is formed into small granules or pellets that are coated with nonradioactive, inert materials to provide containment and leach resistance. The coated waste shapes are then incorporated into a metal matrix that provides impact resistance and increased thermal conductivity.

SINTERED AND HOT-PRESSED CERAMIC WASTE FORMS - J. M. Lukacs and C. B. Ruhter

Waste composition PW-9 will be the basis for sintered and hot-pressed ceramic waste form development. Over 8 kg of PW-9 were prepared by spray calcining the simulated liquid feed composition given in Table 2 in the developmental spray calciner. The calcine is a fine, dark brown powder, and has a vibrated bulk density of 0.82 g/cc. Thermo-gravimetric analysis shows this material to have 0.5 to 1.5 wt% residual water, 10 to 11 wt% nitrates, and appreciable volatility above 700°C. A screen analysis of the bulk material shows over 95 wt% passing a 63 mesh screen.

TABLE 2. Waste Composition of PW-9 Used for Ceramic Waste Form Development

<u>Element</u>	<u>Molarity</u>	<u>Element</u>	<u>Molarity</u>
Na	0.499	Fe	0.050
Cr	0.012	Ru	0.018
P	0.025	Rb	0.012
Sr	0.031	Cs	0.063
Zr	0.120	Mo	0.133
Ag	0.002	Ce	0.115
Cd	0.003	Y	0.006
Te	0.014	La	0.092
Ba	0.033	Pr	0.020
Co	0.011	Nd	0.065
Ni	0.005	Sm	0.011
Pd	0.011	Gd	0.007
Ni	0.032		

CONSOLIDATION AND PHASE BEHAVIOR OF SUPERCALCINE - G. J. McCarthy and D. E. Pfoertsch

As-prepared supercalcine, SPC-4, and SPC-4 pre-fired from 600° → 900°C over a 90-min period were pressed into 0.5-in. pellets at 32,000 psi and fired for 2 hr at six temperatures between 950° and 1250°C--and also for 4, 8, 24, and 48 hr at 1150°C--in order to determine crystallization behavior as a function of temperature and time. In these pressed pellets the 2-hr results show that temperatures of at least 1150°C are needed for high consolidation, and that increasing temperature by 50° to 100°C for the same firing period did not yield a substantial increase in consolidation. When firings were continued at 1150°C there was a small increase in consolidation.

X-ray diffraction (XRD) characterization of these SPC-4 specimens indicated that even with the lowest-temperature firing (950°C for 2 hr) all of the observable (by XRD) phases had crystallized, even though the F_{SS} phase had broad reflections and a considerably larger cell parameter (5.30 Å) than would be observed for higher firing temperatures. The reflections of the S_{SS} phases, $(Ba,Sr,Ca)MoO_4$, have grown distinctly weaker after the 24- and 48-hr 1150°C firings. This observation is consistent with the observation of a surface coating rich in S_{SS} observed on pellets of SPC-4 subjected to very long high-temperature firings.

Pellets of SPC-4 were placed in a silica glass tube and crystallized in rapidly flowing N_2 for 2 hr at 1150°C. Deposits were formed downstream on the cold end of the tube, and the amount of deposit was distinctly greater than that from SPC-2 crystallized under similar conditions. Energy dispersive x-ray (EDX) spectrometry on the deposits indicated that the bulk was made up of a Mo-containing phase or phases, with lesser amounts of Cs and minor Ru, Cd, and Ag. These results indicate that the scheelite or sodalite (Mo-containing) phases and the pollucite (Cs-containing) phases, or perhaps interactions between them, are responsible for the small amount of waste volatilization that is observed in supercalcine.

We prepared SPC-4 + U by adding $UO_2(NO_3)_2$ solution to SPC-4 calcine. Pellets of this calcined SPC-4 + U were fired at 1100°, 1150°, 1200°, and 1250°C for 2 hr. It is interesting to note that far more shrinkage occurred in the SPC-4 + U at 1100°C than in prefired SPC-4 with the same firing, and that the higher-temperature firings cause essentially the same shrinkage. The addition of the appropriate amount of U increases consolidation and greatly reduces waste volatilization at a given processing temperature.

Comparison of the x-ray diffractograms of SPC-4 and SPC-4 + U showed major differences in the relative intensity of individual phases and in the F_{SS} cell parameters. Pure UO_2 , an F_{SS} phase itself, has a cell parameter near 5.47 Å, so the substitution of UO_2 into F_{SS} would be expected to expand that unit cell. F_{SS} becomes the most intense phase because of the very high scattering power of the U atom. A pellet of SPC-4 + U was also crystallized in N_2 at 1150°C for 4 hours. The phase assemblage was identical to the SPC-4 + U/air @ 1150°C/2 hr crystallization, and the F_{SS} cell parameter had increased to only 5.355 Å. The non-oxidizing conditions in the N_2 crystallization would favor U^{4+} , and the fact that the cell parameter increased only slightly compared to the air crystallization suggests that the U oxidation state in the SPC-4 + U crystallized in air is predominantly 4+.

PHASE EQUILIBRIA IN THE SYSTEM U-Th-Ce-RE-Zr-O - J. G. Pepin and G. J. McCarthy

Thorium oxide, CeO_2 , and UO_2 all crystallize in the fluorite structure. Our literature review yielded 28 references to the bounding binary systems ThO_2 - CeO_2 , ThO_2 - UO_2 , and UO_2 - CeO_2 . The ternary system has apparently not been studied. As expected, the binary phase diagrams all showed complete solid solution between the fluorite structure oxides. We project, then, that the ternary phase diagram would consist of a single F_{SS} field over the complete temperature range of interest.

The unit cell parameters (a_0) of the F_{SS} phase diagram can provide useful compositional information when compared to the a_0 of F_{SS} phases in supercalcines. Many reports and articles contained cell parameter data for the three dioxide binary systems that could be used to extrapolate a_0 isopleths in the ternary system. However, it was first necessary to plot the binary system a_0 data. Because there were data judged to be reliable in 14 of the references, a least-squares fit of all of the data was made for each of three binary systems. The cell parameter variations were almost exactly linear with composition in all three (i.e., the F_{SS} solid solutions followed Vegard's law). These binary cell parameter data were used in the plotting of proposed ternary a_0 isopleths for the F_{SS} phase in Figure 10. Note that a linear interpolation has been used for each of the incremental values.

An experimental survey of phase behavior and unit cell isopleths in the 1200°C isotherm of this ThO_2 - CeO_2 - UO_2 system has been initiated in order to test the validity of this type of literature extrapolation. In the compositions studied to date, the predicted F_{SS} phase was obtained and the measured cell parameters were in close agreement with the interpolations in Figure 10.

FIGURE 10. Proposed Ternary a_0 Isopleths for F_{SS} Phase of the System ThO_2 - UO_2 - CeO_2

METAL MATRIX DEVELOPMENT - R. O. Lokken, K. R. Sump, and C. B. Ruhter

Four 1-liter demonstration encapsulations of multibarrier waste forms have been successfully completed, as shown in Figure 11. The waste forms and metal matrices include:

- uncoated supercalcine in a vacuum-cast Al-12Si matrix;
- $\text{PyC}/\text{Al}_2\text{O}_3$ -coated supercalcine in a gravity-sintered copper matrix;
- glass-coated supercalcine in a vacuum-cast Al-12Si matrix; and
- simulated waste glass marbles in a vacuum-cast Pb-10Sn matrix.

The simulated waste glass marbles were heated to $\sim 350^\circ\text{C}$ in a stainless steel canister under a dynamic vacuum prior to casting at 400°C . Similarly, the glass-coated and

FIGURE 11. One-Liter Demonstration Encapsulations of Multibarrier Waste Forms

uncoated supercalcine waste forms were preheated to $\sim 600^{\circ}\text{C}$ and cast in Al-12Si at 650°C . Encapsulation of CVD-coated supercalcine was accomplished by vibrating -100 mesh spherical copper powder into the voids between the supercalcine particles. The canisters were sealed and sintered at 900°C for 8 hr under a dynamic vacuum.

Sectioning of the canister of glass-coated supercalcine revealed a shrinkage cavity in the Al-12Si matrix. The shrinkage is a result of a finite supply of molten metal. As the metal solidifies (through radial solidification) a cavity develops along the centerline of the canister. Controlled, directional solidification and an excess amount of molten metal would alleviate this problem. Sectioning of the three remaining canisters is in progress.

CHARACTERIZATION - R. O. Lokken

Concrete is considered desirable for radioactive waste solidification because: 1) raw materials are relatively inexpensive, 2) fabrication temperatures are low, and 3) fabrication processes are simple.

Most of the work to date has dealt with low-heat-generating waste, such as the waste stored at Hanford⁽⁷⁾ and Savannah River.^(8,9) The waste is added to cement mixtures in

the form of dried sludge, with the main elements principally in the form of hydrous oxides. The low heat generation from this type of waste is expected to produce normal operating temperatures of $\sim 100^{\circ}\text{C}$.⁽⁹⁾ However, incorporating high-heat-generating waste (128 W/kg) into concrete requires a canister diameter of less than 10 cm to maintain a temperature of less than 100°C because of low thermal conductivities (0.004 to $0.015 \text{ W/cm-}^{\circ}\text{C}$). Besides poor thermal conductivity, concrete waste forms:

- have large volumes of off gas--mainly H_2O that evolves up to 400°C ;
- exhibit large ^{137}Cs and ^{90}Sr leachabilities--nominally 10^4 to 10^5 times that of glass;
- have interconnecting porosity resulting in increased surface areas;
- are limited to $\sim 600^{\circ}\text{C}$ to avoid excessive mechanical deterioration.

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