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**Quarterly Progress Report
Research and Development Activities
Waste Fixation Program
July through September 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

BNWL-2243

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QUARTERLY PROGRESS REPORT
RESEARCH AND DEVELOPMENT ACTIVITIES
WASTE FIXATION PROGRAM
JULY THROUGH SEPTEMBER 1976

J. L. McElroy

August 1977

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PREVIOUS REPORTS

This progress report is the sixteenth 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, 2070, and 2242.

SUMMARY

Research and development activities of the Waste Fixation Program for July through September 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.

- Two engineering-scale in-can melting (ICM) runs were completed. One run evaluated the melting of PW-8a-2 type waste containing cesium and ruthenium with 76-101 type frit. The other run established a melting rate of 30.9 kg/hr for an 8-in. diameter can containing a drop-in fin assembly made from an alloy having an expansion coefficient similar to that of the glass. Agglomeration of batch in the frit line and the connecting section between the calciner and melter was eliminated by insulating the lines. A new multizone furnace was received which will make in-can melting possible in cans as large as 8 ft high by 2 ft in diameter.
- High sodium feed was successfully spray calcined by lowering the calciner chamber temperature and using an additive in the feed material.
- The joule-heated ceramic melter has been at operating temperature for eight continuous months. The unit recovered from an unplanned power outage, demonstrating that concern regarding restart capability may not be warranted.
- The fluidized-bed calciner testing continued with emphasis on developing several mechanical components and engineering a hot cell installation and a full-scale prototype.
- A variable speed, weigh belt-type solids feeder was evaluated as a frit feeding device. An air lock device was evaluated for transferring frit into an in-can melting system. A strain gage-type load cell system with a maximum rated load capacity of 3000 kg was evaluated for a canister weighing system.

- A 76-183 melt composition was developed for PW-7a waste, and preliminary tests indicate that the composition will be satisfactory.
- Leach testing of thermally treated 72-68 glass was continued and the results were calculated through 259 days.
- Comprehensive examination of 72-68 glass samples annealed for 1 year has been completed and has indicated no significant change over the results observed at 2 months. Additionally, characterization and identification work performed on the high-neodymium rare earth phase has indicated that an apatite-type structure exists within the temperature range of 500 to 750°C but only after a minimum time of 1 week at temperature.
- Preliminary impact tests on a gravity-sintered 304 stainless steel matrix demonstrate that the majority of the cracks go around, not through, the Al_2O_3 supercalcine cores.
- Gravity sintering parameters for stainless steel matrices have been reduced to 1050°C for 8 hr in a stagnant atmosphere, and both stainless steel Al_2O_3 and pure stainless steel spheres have been successfully fabricated.
- A promising plan for feeding the disc pelletizer directly from an operating spray calciner and recycling dust back to the calciner is being evaluated.

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QUARTERLY PROGRESS REPORT
RESEARCH AND DEVELOPMENT ACTIVITIES
WASTE FIXATION PROGRAM
JULY THROUGH SEPTEMBER 1976

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 July through September 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 non-dispersible 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 using the storage can as the melting crucible.

Engineering-scale in-can melting (ICM) runs, using a single zone furnace and connected directly to the heated wall spray calciner, continue to be made to further the development of this HLLW solidification process. The product of run ICM-18, which was described in the last quarterly report,⁽¹⁾ is a very homogeneous glass exhibiting no phase separation or settling except for a thin layer of slag on top of the glass. A successful ICM run was made in an 8-in. diameter can without fins to evaluate the melting behavior of calcined PW-8a-2, a simulated Nuclear Fuel Services (NFS) combined HLLW and ILLW containing cesium and ruthenium. A new frit composition (76-101) designed especially for use with PW-8a-2 was used. During another ICM run using an 8-in. diameter can containing a drop-in fin assembly, a melting rate of 30 kg/hr was sustained until the can was full. The fin assembly was made from a stainless steel alloy which is resistant to attack by the melt but has an expansion coefficient almost the same as that of the glass.

During the last two runs, the line that carries frit from the metering device into the cone below the calciner chamber and the line that carries the batch from this cone into the melter were both stripped of heat sinks and well insulated. This resulted in no more agglomerations of batch accumulated in these lines because of condensation.

A resistance-heated furnace was received. This furnace has a heated chamber 28 in. in diameter by 7-1/2 ft high that is divided into six 15-in. high zones. The furnace has many control features including individual zone-controlled heating and cooling to permit maximum flexibility of operation for further development of the ICM process.

DEVELOPMENT SPRAY CALCINER (L. S. Romero)

The objective of this work is to provide a reliable system for conversion of HLLW-ILLW to calcine for devitrification.

Several runs were conducted during the process of developing a method of calcining high sodium feed of approximately 90 wt% sodium nitrate. The high sodium feed was successfully spray-calcined by lowering the chamber temperature and using an additive in the feed to avoid melting sodium nitrate on the chamber.

Several runs were conducted to study ruthenium volatility during spray calcination. Material balances indicate that negligible ruthenium was volatilized during calcination as a result of varying the chamber temperature and the filter blowback cycle.

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 mid-January of this year, the scale model ceramic melter has been at operating temperature for 8 months. This brings the total exposure of the melter to molten glass to 19 months. A 4-by 6-in. section of one sidewall has slowly collapsed into the melting cavity. This occurred directly above the portion that was undermined during a much earlier experiment. This failure exposed the backup refractory directly to the molten glass and the condition has existed for 2 months without loss of containment. It is believed that corrosion will be limited by the water-cooled side plates.

An unplanned power outage yielded an interesting result. Power to the melter was completely off for 62 min; however, when power was reinstated, the unit recovered to the normal idling condition. This unplanned event demonstrated that this unit is capable of recovering even after a long power outage. With actual waste glass the safe time without power might be several hours. Thus, the concern with periodic restart of the melter because of power outages may not be warranted.

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.

Testing of the calciner has continued on an intermittent basis during this reporting period and components tested include:

- a conical distributor plate
- feed and fuel nozzle
- an air jet solids feeder to replace the mechanical slide valve presently used.

Engineering efforts are continuing toward future installation of a pilot-scale calciner in the hot cell as well as scope designs for a full-scale prototypic design.

Electron microscope photos of various calcine product forms at magnification ranges from 200 to 20,000 X are shown in Figures 1 to 7.

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 high-level waste vitrification equipment.

FIGURE 1. Silica Sand Used as Inert Bed Material

FIGURE 2. Agglomerated Particles Caused by Higher Sodium (>1 m) in the Feed

FIGURE 3. Calcine Particles from Conventional Operation (Noninert Bed)-No Sodium

FIGURE 4. Calcine Coated Inert Bed Particle (~ 0.6 m Na)

FIGURE 5. Surface of "Conventional"
Calcine Particle

FIGURE 6. Surface of Coated Inert
Bed Particle

FIGURE 7. Filter Fines

A Merrick, Model 950 DSC-1, digital weigh-belt feeder system was evaluated for application as a frit feeder. Operation with a -20 +80 frit material was exceptionally smooth for delivery rates ranging from 1 to 50 kg/hr. The unit showed a discharge rate repeatability of $\pm 1\%$ over this range and, while a 2% bias was also observed, this can be reduced to zero with appropriate controller adjustments.

A double-acting, air cylinder-actuated air lock device was evaluated for its applicability in conducting frit from an out-of-cell location into an ICM system while maintaining the integrity of the cell wall barrier. The double-acting valve, using rubber valves that close onto steel seats, transferred a -20 +80 sized frit without incident into a spray calciner maintained at its normal operating pressure of 10 in. of water vacuum. The amount of air leaking in through the operating valve was negligible and did not significantly affect the operation of the calciner pressure control system.

A Baldwin-Lima-Hamilton strain gage-type load cell system was evaluated for use in a canister weighing system. The instrumentation for weight indication was accurate to ± 0.5 kg from zero to its maximum rated load of 2725 kg. However, a companion instrument system intended to indicate the rate of weight increase of the total load in kg/hr, while accurate to ± 1 kg/hr at loads up to 500 kg, became erratic in its performance at loads above 1727 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.

A 76-183 [PW-7a-1: (2.0) 53 SiO₂, 14 B₂O₃, 9 TiO₂, 1.5 Al₂O₃, 12 Na₂O, 3 K₂O, 3 CaO, 4.5 CuO] composition has been developed for Allied General Nuclear Services (AGNS) HLW-ILW combined waste. Initial tests indicate that the calcine waste loading can be varied from 0 to 40 wt% without major changes in viscosity. The CuO addition allows non-ZnO glasses to be formulated and still have moderately low Soxhlet leach rates. Durability in both mild acid and base is satisfactory. Longer term tests are in progress.

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.

Full-level radioactive glass, 72-68 formulation, devitrification samples are being prepared in C-Cell. One small sample (approximately 1/4 in.

diameter by 1/8 in. thick) was devitrified at 800°C for 1 week. This sample is now being cast into a predrilled plastic mount along with five as-vitrified samples. These five additional samples include: 1) a 72-68 nonradioactive standard glass, 2) a sample of HLW glass from the bottom section of a canister, 3) a sample of HLW glass from the top section of a canister, 4) a sample of HLW glass from the same section as the devitrified sample, and 5) a sample of canister-wall-HLW glass interface from the same section as the devitrified sample.

A larger sample of HLW glass (approximately 1 x 1 x 1/8 in. thick) is now being devitrified for 1 week at 800°C. This larger sample will be used for x-ray diffraction analysis and the smaller samples for ceramographic examination and microprobe analysis.

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.

The leach testing of irradiated LWR fuel pellet fragments in Hanford ground water and building distilled water was terminated July 29, 1976 after 345 days of cumulative leach. The weight losses of air-dried fuel fragments during the leach test are listed in Table 1.

TABLE 1. Weight Losses of the Air-Dried Fuel Fragments
During the Leach Test

<u>Fragment</u>	<u>g</u>	<u>wt%</u>
Z-2, Hanford Ground Water	0.0099	0.19
Z-3, Hanford Ground Water	0.0101	0.20
Z-4, Building Distilled Water	0.0047	0.11
Z-5, Building Distilled Water	0.0177	0.33

The deionized water leach tests will be continued until the leach equipment is needed for other tests.

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

A series of leach solutions generated using distilled water in contact with actual HLW glass wafers has been analyzed by gamma spectroscopy. The radioisotope concentrations were higher for the glass originating from the middle of the waste glass cylinder. A second series of leach solutions has been submitted for analysis.

To analyze the HLW glass wafers in-cell, a gamma radiation collimating tube has been designed and is being fabricated. This will enable the use of an out-of-cell detector to analyze the glass prior to leaching or devitrification experiments.

Work has begun on the source term characterization task of the program plan to define HLW glass leaching and leachate behavior in geologic storage media. Area-use planning of the hot cell has been started and isotopes needed for the study are on order.

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

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

Leach testing of 72-68 glass stored at elevated temperatures for a maximum of 2 months is completed through the monthly sampling phase of the modified IAEA leach test procedure. Results through 259 days of exposure to deionized water show that leach rates have remained constant at approximately 1×10^{-7} g/cm²/day for the 1000°C reference samples and 1×10^{-6} g/cm²/day for the samples stored 2 months at 700°C. The leach rates of the latter samples are the highest measured; however, they are only a factor of ten higher than those of the reference glass.

Figures 8 and 9 show the glass leachability expressed as cumulative penetration in centimeters. Figure 8 shows curves based on cesium for the reference samples and the 700°C samples, and Figure 9 shows curves based on strontium behavior for the same samples. Presenting leach data as cumulative penetration permits extrapolation of leachabilities to long time periods. The slopes of the curves are indicative of the mechanism(s) involved in the leaching reaction.

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

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.

During this quarter, the principal effort has been a comprehensive examination of the 1-year anneal, simulated waste glass samples (composition 72-68). In addition, work has been performed to characterize and identify a high neodymium, rare earth phase which exists under certain conditions of time and temperature in the 72-68 glass.

Overall examination of the 72-68 glass after annealing for 1 year has yielded no new information; for all purposes, the microstructures observed are identical to those seen at 2 months. All familiar phases [i.e., Zn_2SiO_4 , $(\text{Sr}, \text{Ba}, \text{Ca})\text{MoO}_4$, CeO_2 , and RuO_2] were present and to approximately the same extent as at the 2-month period. Analysis of the glass matrix did tend to indicate that a slightly larger percentage of constituents were dissolved in the matrix at 1 year than at 2 months. This result would tend to indicate that redissolution, to a small extent, is taking place during long-term anneals.

SEM examination has identified a rare earth phase which exists in the 72-68 glass that is high in neodymium, lanthanum, and calcium. X-ray diffraction analysis has also confirmed the existence of some unknown phase(s) existing within the temperature range of 500 to 750°C and only after a

FIGURE 8. Cumulative Penetration Based on Cesium Behavior

FIGURE 9. Cumulative Penetration Based on Strontium Behavior

minimum time of 1 week at temperature. This phase is believed to be of an apatite-type structure. The diffraction intensities and peak overlap with higher concentration phases preclude an unambiguous structural identification.

STANDARD TESTS DEVELOPMENT (J. M. Rusin)

The purpose of this work is to develop standard tests for intra- and interlaboratory comparisons of waste form properties for potential use by the regulatory agency to demonstrate conformance to established standards.

Test procedures to evaluate impact strength and leach rate have been outlined. The test result is in the form of a risk value. For initial development, the risk value has been defined as:

$$R = S \cdot G$$

where S = % particles less than 500 μ

G = % weight loss as determined by the Soxhlet test.

Modifications of the risk value may include the additional factors of:

- total surface area
- leach rate of specific fission products
- thermal stress cracking based upon thermal expansion.

The impact test consists of dropping a weight onto a cylindrical sample approximately 0.50-in. diameter by 0.50-in. high contained in a diamond mortar. Particle size distribution is obtained by sieve analysis. The exact weight and height of the drop will be determined from preliminary runs to insure that the sample is impacted with excessive energy.

The Soxhlet test is presently used, although a test based upon leaching of specific ions would be desired since upon devitrification certain phases which do not contain fission products may have a high leach rate. Annealed and heat-treated samples will be tested. Temperatures selected will vary depending upon the type of glass tested; the annealing temperature will be based upon glass transition temperature and the devitrification temperature upon maximum crystal yield. An alternative to this test is to directly leach the impacted sample.

Thus, the leachability would be both a function of particle size produced upon impact and durability. Both tests will be evaluated.

Thermal and Mechanical Shock (L. R. Bunnell)

During the period between processing and final storage, waste-containing glass must undergo handling. Various accident scenarios involve impact, and knowledge of the resultant dispersible or respirable fines is required. Work has already been conducted⁽³⁾ in which canisters have been dropped or impacted and the resulting material analyzed. That type of full-scale testing, involving the rather complex interaction of canister and glass, will continue to be necessary, but it is an expensive way to evaluate the effects of month-to-month process changes. The purpose of our work is to develop an impact test amenable to small-scale laboratory testing, sufficiently convenient to perform on a large number of samples, and sensitive enough to detect the defects of composition or processing changes. The methods and device used should also be amenable to a small amount of eventual hot-cell testing.

Recent work from two sources^(4,5) is directed toward a test which should satisfy the objectives above. The test involves placing a sample, in the shape of a right cylinder, into an oversized cavity in a hardened steel block and placing a hardened plunger on top of the sample. A known mass falling through a measured distance provides the impact energy. The resultant powder is then screened to determine the size distribution, from which parameters such as geometric surface area can be calculated, and from which respirable fines can be determined.

A suitable impacting device was located; it has the capacity to impart ~120 ft-lb of energy to the sample, which is substantially more than the machine used in the above reference. The reserve energy will be needed to investigate devitrified products, since these are substantially tougher than vitreous glass.

Some initial testing has been done using soda-lime-silica plate glass. Two fairly typical curves are shown in Figure 10. We are now in the process

of determining the sample fracture as a function of applied energy. A two-stage effect is expected at a low energy level in that a sample will only produce a few large fragments; this has been seen in tests involving 17 ft-lb of energy. The second stage, requiring more energy, is where the glass cylinder collapses and fracture is much more extensive, such as shown in Figure 10. Surface area will be measured as a function of applied energy and used to determine the most appropriate test energy for a given glass type, as well as to determine sample response to higher energies. As might be expected for particles produced by a single impact, the resultant glass is extremely angular, and might have surface area (and leaching behavior) different from a ground sample. Some BET surface area measurements will be made, and eventual leach testing will be performed.

FIGURE 10. Particle Size Distribution for Two Impact Energies

Thermal Expansion Measurements (J. L. Bates)

Thermal expansion measurements are required for any glass used as a storage medium for waste materials. In addition to providing data for calculating thermal stresses in both glass and canister, several important parameters can be obtained from the shape of the curve, as was done in a previous quarterly report.⁽⁶⁾ The samples measured this quarter were intended to assess the effects of incomplete annealing.

These measurements were made in air, in an aluminum oxide pushrod dilatometer. The heating rate was 4°C/min, and a digitized data point was taken every 8°C. The data were then corrected for the expansion of the dilatometer itself (as determined by using an NBS tungsten standard) and plotted. In each case, heating was done only to the point of obvious softening. Two curves for 72-68 glass are shown in Figure 11; glass 2 was poorly annealed, glass 3B was well annealed. Comments relevant to the remaining glasses, and factors which could be determined from the dilatometer curves, are included in Table 2. The annealing state of the glass obviously has a large influence on the amount of data that can be determined for a glass sample. Since annealing schedules are usually designed by use of expansion data, a problem exists. However, annealing schedules can be determined that will do an adequate job on a particular class of glasses.

FIGURE 11. Thermal Expansion of Two Species of 76-68 Glass

TABLE 2. Thermal Expansion Results

Sample No.	Previous Thermal Treatment	Dilatometric Softening Pt, °C	Glass Transition Temp, °C	Comments
1. 76-68 (Pellet)	Glass powder, sintered at 650°C, annealed at 350°C 2 hr	562	551	Curve had general features of an annealed glass
2. 76-68	Melted at 1050°C, annealed at 350°C 2 hr	566	514	Appeared quite well annealed
3A. 76-68	1200°C melt, 350°C anneal 2 hr	576	Not Determinate	Not well annealed. Contracted at 490°C showing densification, typical of glass in quenched state
3B. 76-68	1200°C melt, 500°C anneal 2 hr, slow cool to 450°C, furnace cool to R.T.	570	511	Well annealed
4. 76-76	1200°C melt, 350°C anneal 2 hr	554	500	Appeared well annealed
5. 75-232	European Borosilicate melted at 1250°C, annealed at 350°C 2 hr	551	Not Determinate	Not well annealed, contracted at ~450°C
6. Lead Borosilicate	Melted at 1250°C, annealed 350°C, 2 hr	506	462	Appeared well annealed
7. 76-183	1250°C melt annealed at 350°C 2 hr	568	Not Determinate	Not well annealed, contracted at ~450°C
8. 76-179	1250°C melt annealed at 350°C 2 hr	572	Not Determinate	Not well annealed, contracted at ~450°C

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 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/Al₂O₃, coatings on 1- to 10-mm pellets.

In the last quarterly report⁽¹⁾ the problems encountered with the duplex coating applied at that time were discussed in considerable detail. Briefly, impervious protective Al₂O₃ coatings and crack-free PyC coatings could each be applied to bare supercalcined waste, but Al₂O₃ coatings applied over PyC on the same substrate resulted in a portion of the coated particles having cracked Al₂O₃ coatings. It was hypothesized that the most probable cause for the cracking was a thermal expansion mismatch problem between the Al₂O₃ coating and the supercalcine simulated waste, especially after it was strengthened by infiltration of its porous structure during the PyC-coating operation. The principal approach explored to circumvent the cracking problem was to retain the porosity (crushability)

(a) Battelle Columbus Laboratories

of the supercalcined simulated waste by applying the PyC coating rapidly, at least initially, to close the pores at the surface before gross infiltration occurred.

To coat 2- to 3-mm supercalcine waste particles, it was necessary to modify the existing system since the vibrating/fluidized-bed technique used earlier to coat the 1-mm particle did not result in as high a quality Al_2O_3 coating when used in coating the larger particles. The modified system that was developed consisted of mixing 10 cm^3 of the larger particles and 10 cm^3 of an appropriate bed diluent, and then coating this mixture by a conventional fluidized-bed technique. A 420 to 710 μm ZrO_2 particle diluent was determined to be of a size and density that resulted in the larger waste particles and the diluent particles circulating in the fluidized bed in a similar manner.

To preserve the porosity of the simulated waste for the reasons and by the techniques discussed above, a batch of the relatively large (2- to 3-mm) supercalcined simulated waste particles was coated initially with 3.5 μm of PyC at 1200°C from a 50:50 mole % C_3H_8 mixture at a rate of $\sim 0.5\text{ }\mu\text{m}/\text{min}$ (>10 times as fast as the PyC coating on the earlier material evaluated). Coating of the PyC was then continued to a thickness of $\sim 35\text{ }\mu\text{m}$ at only $\sim 0.3\text{ }\mu\text{m}/\text{min}$ by reducing the concentration of propane to ~ 17 mole %. This technique accomplished the objective of reducing the PyC infiltration into the porous substrate structure. The PyC-coated material was then overcoated with Al_2O_3 by the hydrolysis of AlCl_3 at 1025°C and the duplex coated product is being examined at Battelle-Columbus. It is suspected that equipment problems encountered, unrelated to the basic techniques, have resulted in an Al_2O_3 coating of less than optimum quality.

In an effort to identify a suitable nonhalide system for applying Al_2O_3 coatings to waste particles, work was initiated on the evaluation of the pyrolysis of $\text{Al}_2(\text{OC}_2\text{H}_5)_3$ system. A second portion of the PyC-coated material discussed above was coated with Al_2O_3 at 600°C using the $\text{Al}_2(\text{OC}_2\text{H}_5)_3$ system and the fluidized-bed technique devised for coating the 2 to 3 mm particles. The quality of the Al_2O_3 coating applied has yet to be ascertained, but no major operational difficulties were encountered.

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.

Encapsulation of waste glass marbles in a metal matrix is limited by the softening point of the glass. Gravity sintering in a copper matrix was unsuccessful because of excessive flow of the glass at 900°C. Conventional casting of the marbles in molten lead was successful except for a few voids near the top of the casting. Vacuum casting should eliminate any voids.

Sufficient 3/8-in. diameter marbles (over 1000) have been produced from remelted ICM glass to encapsulate larger samples by vacuum casting with various lead alloys.

Commercial suppliers of marbles have been contacted and will be visited to discuss application of their processes to remote facility operation.

MULTIBARRIER WASTE FORM CHARACTERIZATION (J. M. Rusin)

The purpose of this study is to characterize the thermal, mechanical, and physical properties of all components of the multibarrier waste form (e.g., core, coating, and metal matrix).

Three multibarrier waste forms will be produced and characterized in FY-1977:

- supercalcine cores coated with $\text{PyC}/\text{Al}_2\text{O}_3$ encapsulated in stainless steel
- supercalcine cores coated with glass frit encapsulated in copper or bronze
- uncoated glass marbles cast in lead alloy matrix.

Preliminary impact tests on gravity sintered 304 stainless steel matrix demonstrate that the majority of the cracks go around the Al_2O_3 supercalcine cores. Only a few cores were cracked. The samples were 0.52-in. diameter rods and were impacted 0.87 in. from the clamping point with 155 ft-lb.

Sintered supercalcine cores (6 to 30 hr, 1100°C) produced by disc pelletizing SPC-2 supercalcine powder have typical open porosity of 42% and bulk density of 2.41 gm/cm³. Average specific gravity of the calcined and sintered supercalcine is 3.78 and 4.09, respectively.

Phenolic resin doped with ¹⁴C has been pyrolyzed and finely ground to determine the leachability of PyC; the pyrolyzed resin contains 0.77 mCi/gm ¹⁴C.

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.

The gravity sintering parameters for stainless steel matrices have been reduced to 1050°C for 8 hr in a stagnant atmosphere. Both pure stainless steel and stainless steel-Al₂O₃ spheres have been successfully fabricated using the above parameters. These fabrications have also been done with 410 stainless steel, as well as the 300 series stainless.

Vacuum conditions further reduce the required temperature for gravity sintering. Copper was adequately bonded at 700°C for 16 hr in a dynamic vacuum.

Lead was cast around glass marbles in a quartz tube at 400 to 450°C. The molten lead bonded to the quartz tube making removal impossible. Some of the elongated waste composition glass marbles were cast in lead at 450°C.

DISC PELLETIZER EVALUATION (A. A. Garrett)

The disc pelletizer is being evaluated to determine if it can provide a simple way to make pellets from a mixture of calcine and frit powders. The pellets would then be sintered, coated and incorporated in a metal matrix prior to terminal storage.

During this quarter, 100 kg of 6 to 8 mm diameter PW-4b cores and 10 kg of 0.5 to 5.0 mm diameter PW-7 supercalcine cores were produced by a 16-in. disc pelletizer. These cores were sintered at 900 and 1100°C,

respectively. The sintered cores are quite rugged and leach resistant. Additionally, about 2 kg of 6 to 8 mm diameter PW-4b cores were coated with a borosilicate glass frit using a disc pelletizer, then sintered at 775°C. The sintered frit produced an impermeable, uncontaminated glass coating around the calcine cores.

A scheme for feeding the disc pelletizer directly from an operating spray calciner and for recycling dust from the pelletizing operation back to the spray calciner is being evaluated. Currently, the scheme appears feasible and quite promising.

Core green strength for both PW-4b and supercalcine has been significantly improved by increasing core moisture content and by adding dry binders such as clay to the calcine powder.

Dust rising from the pelletizing operation is being contained, collected, and recycled by providing a flexible "boot" for the calcine feeder input line and by providing a close-fitting exhaust hood over the lower half of the pelletizer pan. Exhaust from the hood is piped to the feed hopper where a filter removes the particulates.

CHARACTERIZATION OF SUPERCALCINE 76-3 (SPC-2) (G. J. McGarthy, M. T. Davidson, D. E. Pfoertsch and R. G. Johnston)^(a)

Current objectives of the Supercalcine Task include: 1) refining existing formulations for PW-4b and PW-7 waste, 2) developing formulations for the high Na wastes PW-7a and -8a, 3) determining crystalline phase behavior of cesium, ruthenium, tellurium, palladium, rhodium and the actinides in supercalcine, and 4) considering the effects of radiation and transmutation on supercalcine stability.

The engineering-scale spray calcination of this formulation has been previously described. The simulated waste was a simplified PW-7 without the very expensive elements (palladium, rhodium), the radioactive elements (technetium, uranium, plutonium, neptunium, americium, curium,

(a) Pennsylvania State University, University Park, PA.

promethium) and tellurium; the ruthenium concentration was only 10% of its value in PW-7. The additives required for formation of the crystalline phases specified by the phase formation model were nitrate solutions of calcium, strontium and aluminum and a colloidal suspension of SiO_2 . Waste oxide loading was 71 wt%.

Crystalline Phase Formation Model

Based on many compatibility studies and on laboratory simulations of supercalcines, the phase formation model specified by 76-3(SPC-2) was

Crystallization and X-Ray Phase Analysis

After spray calcination, the product is very fine powder which is x-ray amorphous. It must be fired at temperatures from 1000 to 1200°C to give both crystallization of the specified phases and sintering into the desired shapes. Firing tests lasting from 2 to 8 hr at temperatures from 800 to 1250°C have been performed on the "as received" powder and on 1/2-in. diameter pellets cold pressed at 74,000 psi. The shrinkage in pellet diameter was recorded as a measure of the degree of sintering. X-ray powder diffractometers were used for phase identification.

Good crystallization and sintering were obtained with firing temperatures as low as 1050°C for 2 hr. The crystalline phase assemblage and shrinkage in pellet diameter ($24 \pm 1\%$) remained constant for all firings between 1050 and 1250°C.

Each of the phases specified in the phase formation model was identified in the firing products. The reflections of A_{ss} , M_{ss} , T_{ss} , S_{ss} , P , and $(Fe,Cr)_2O_3$ were distinct in the x-ray diffractograms, but the amounts of RuO_2 and Sp_{ss} were below the level of detection. The identification of RuO_2 is described later. The ferrimagnetic Sp_{ss} was clearly identified by the action of a hand magnet on the product.

One additional phase was also in the product. It was a fluorite structure solid solution [F_{ss}] phase with the approximate composition $(Ce_{0.8}, Zr_{0.2})O_2$, as estimated from its cubic cell parameter. The phase formation model had called for cerium to be fixed in A_{ss} . However, the amount of cerium contained in the less expensive "rare earth mixes" used in engineering scale processing tests at PNL exceeded by a factor of three the actual cerium concentration of PW-7. As we demonstrated earlier this year,⁽⁶⁾ only a limited amount of cerium can be accommodated in A_{ss} . Thus, in 76-3, the excess cerium reacts with part of the free ZrO_2 to form the F_{ss} phase.

Thermal Stability

Data on this property are both preliminary and qualitative. The lack of staining of the firebricks used in all of the crystallization tests (800 to 1250°C) is one qualitative indication that there is no major waste oxides volatilization. The silica tube test for cesium volatilization indicated a very small amount of cesium loss at 1150°C after 2 hr, but little or none after 2 hr at 1100°C. Pure pollucite ($CsAlSi_2O_6$) shows a corresponding behavior. There was a light deposit of a black substance near the cooler end of the silica glass tubes after both firings. We have previously found that this indicates the presence of ruthenium and its volatilization, at least in part, during crystallization. However, we found that this ruthenium volatilization deposit can be virtually eliminated by crystallization of 76-3 in a nitrogen rather than an air atmosphere.

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.

Crystalline phase formation in supercalcines designed for high sodium wastes (PW-7a, PW-8a) will be quite different from that just described for the low sodium PW-7 waste. Two of the major differences will be the crystallization of a sodalite structure solid solution (So_{ss}) and an A_{ss} with significant sodium substitution. Fifty-four possible solid solution models for these two phases have been studied with the following results.

- The molybdate nosean variety of So_{ss} is $AE_2[NaAlSiO_4]_6(MoO_4)_2$, where $AE=Ca, Sr$. The calcium phase is by far more stable and crystallizes completely at 800°C in a few hours, whereas the strontium phase is still not fully reacted after 6 hr at 1200°C. Thus, calcium would be a better additive to promote So_{ss} crystallization.
- There is also a chromate nosean variety of So_{ss} $AE_2[NaAlSiO_4]_6(CrO_4)_2$ in which calcium again appears to form a more stable phase than strontium. Compatibility tests will be needed to find whether chromium would crystallize in this phase or in the Fe_2O_3 and Sp_{ss} phases.
- Cesium cannot be substituted for any of the sodium in the various So_{ss} phases. It reacts with aluminum and silicon instead to form pollucite.
- Sodium and coupled sodium-aluminum substitutions are possible in silicate A_{ss} . Simple Na-RE-silicates have been crystallized in the apatite structure, but only after lengthy firings at 1050°C. Some solid solution models containing sodium, calcium, RE and aluminum crystallized in apatite structures after only brief firings in the 800 to 1000°C range suitable for high sodium supercalcines. Some examples are $Na_2Al_2Nd_6[SiO_4]_6O$, $Na_{1.5}Al_{1.5}CaNd_6[SiO_4]_6O$ and $Ca_2Na_2RE_6[SiO_4]_6$.
- Phosphate appears to be incompatible with sodium substitutions in A_{ss} . $REPO_4$ [M_{ss}] impurity phases were noted in the firing products of all phosphate containing solid solution models. Thus, specify $[PO_4]$ in M_{ss} rather than A_{ss} in the high sodium supercalcines.

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