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**DEVELOPMENT AND TESTING OF A GLASS WASTE FORM
FOR THE IMMOBILIZATION OF PLUTONIUM**

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ABSTRACT

Two alkali-tin-silicate (ATS) glasses have been prepared at Argonne National Laboratory (ANL) as part of our ongoing research in radioactive waste glass development. These glasses dissolved 5% and approximately 7% Pu. Early corrosion test results indicate that Pu-bearing ATS glass is extremely durable. The initial goal in this project concerned equally both the solubility of Pu and the durability of the ATS glasses; however, our primary emphasis has changed recently to maximizing the loading of Pu in the glass. ATS-based glasses, using Th(VI) and Ce(III) as surrogates for Pu(IV), are now being investigated to increase the solubility of Pu without substantially sacrificing the durability of the current ATS formulations. The solution data from various corrosion tests on the original Pu-containing ATS glasses are also presented.

INTRODUCTION

The United States has declared about 50 metric tons of weapons-grade Pu surplus to national security needs. The President has directed that this Pu be placed in a form that provides a high degree of proliferation resistance in which the surplus Pu is both unattractive and inaccessible for use by others [1]. Three alternatives are being evaluated for the disposal of this material: (1) use of the Pu as a fuel source for commercial reactors; (2) immobilization, where Pu is fixed in a glass or ceramic matrix that also contains or is surrounded by highly radioactive material; and (3) deep bore hole, where Pu is emplaced at depths of several kilometers. The immobilization alternative is being directed by the staff at Lawrence Livermore National Laboratory (LLNL). The staff at ANL are assisting by developing a glass for the immobilization of Pu and in the corrosion testing of glass and ceramic material prepared both at ANL and at other DOE laboratories. As part of this program, we have developed an ATS glass into which 5-7 wt% Pu has been dissolved. The ATS glass was engineered to accommodate high Pu loading and to be durable under conditions likely to accelerate glass reactions in the geological environment during long-term storage.

ATS DEVELOPMENT

Background

The ATS glass was originally designed to be used in a new melter (greenfield) or adjunct melter facility (located next to the Defense Waste Processing Facility, DWPF). With this in mind, three objectives were set for developing this glass. First, keep the pH of the solution in contact with the glass as low as possible. In solutions with a pH greater than 11, the glass will react more rapidly because of the increased solubility of Si; therefore, minimizing the concentrations of ionized silica species such as $H_3SiO_4^-$ in solution will improve durability. Second, reduce the potential for alteration-phase (i.e., clays) formation by reducing or eliminating Al, Ca, Mg, and Fe from the glass. In addition, keep the total silica concentration in the glass as low as possible to help reduce the potential for zeolites or other alumino-silicate phase formation and the potential for increasing the glass corrosion rate. Third, demonstrate a Pu solubility of 10 wt% in the glass. Using this rationale, an ATS glass was prepared and tested as described by Bates [2].

This glass, referred to as "P10", was developed using principles of glass chemistry and the knowledge of the glass reaction-progress pathway [3]. Although this glass met the first two objectives described above, the glass contained both dissolved (~7 wt%) and some residual, undissolved PuO₂ (~3 wt%). Some of the undissolved PuO₂ was dispersed throughout the glass, while some settled to the bottom of the crucible during fusion.

After the development and preparation of this initial ATS glass, the intended use was changed from a greenfield or adjunct melter to a "can-in-can" concept. For can-in-can, the Pu-containing glass would be placed into small cans which would then be located inside a larger DWPF canister. For this disposal method, the need for high glass durability and for the presence of ¹³⁷Cs as a terrorist deterrent became less important; increasing the Pu solubility became a higher priority. Additionally, based upon repository criticality calculations, the Pu-to-Gd mole ratio was increased to 1:1. With this opportunity to improve the glass composition, a more systematic approach was used to gain a better understanding of the ATS glass composition and the interactions between the glass components.

Based upon this approach, a second ATS glass containing 5 wt% Pu was prepared. The Pu content was set at 5 wt% for this glass for two reasons. Based upon initial criticality concerns, the Pu:Gd ratio in the initial ATS glass was set at 2.7:1 (on an atomic % basis). Based upon current criticality calculations, a Pu:Gd ratio of 1:1 is required. Because Gd is expected to compete for Pu sites in the glass, the Pu content was decreased to 5 wt%. Second, a glass without undissolved PuO₂ was needed for corrosion testing. This new formulation, called "P5", successfully dissolved the Pu. The composition of the initial ATS glass (P10) and the nominal composition of this new P5 ATS glass are reported in Table I.

Table I. Composition of Pu ATS Glasses Subjected to MCC-1 and PCT.

Compound	Glass Compositions ^a (element wt%)	
	ATS-P10	ATS-P5
Al	1.38	1.19
B	3.94	2.76
Cs	0.47	0.85
K	3.32	6.41
Li	1.58	1.82
Na	6.68	6.79
Si	20.0	21.2
Sn	1.42	1.74
Ti	1.32	1.24
Zr	3.70	3.66
Zn	0	2.46 ^b
Pu	10.3 ^c	5.00
Gd	2.60	3.29

^aValues in table for ATS-P10 are a bulk sample analysis.

Values for ATS-P5 are the nominal composition of the feed.

^bBecause of Zn's classification as a hazardous metal, it will be eliminated from future ATS formulations.

^cGlass contains approximately 2.8 wt% undissolved Pu as PuO₂ (see reference 2 for more details).

Use of Pu Surrogates

Because of the difficulty of working with Pu-containing materials, either Ce^{3+} (ionic radius: 0.1143 nm) or Th^{4+} (ionic radius: 0.105 nm) are commonly used as chemical surrogates for Pu^{4+} (ionic radius, 0.096 nm). Thorium appears to be an ideal substitute because of its ionic radius and charge. However, our experiments show that Th is approximately two times more soluble than Pu in the ATS glass. This may be explained by the disparate electronic structures of Th^{4+} (with no 5f valence electrons) and Pu^{4+} (which has 5f electrons). This has led some investigators to use Ce^{3+} as a surrogate for Pu^{4+} , despite charge and radius differences, because Ce has an electronic structure more similar to that of Pu (i.e., 4f electrons). The electronic structures affect the structural roles of these elements in the glass, information that is not currently known. Furthermore, uncertainties remain as to the oxidation state of Pu (which may also occur as Pu^{3+} , Pu^{5+} , and Pu^{6+}) and Ce (which may also be 4+).

We chose to use Th^{4+} (as ThO_2) as our primary surrogate for Pu because, based on the chemistry of Th, we know that it exists solely as a 4+ cation, whereas Ce may exist as a mixture of 3+ and 4+. In a study by Hanchar [4], which utilized a highly oxidizing environment to synthesize zircons doped with rare earth elements, Ce was found to occur as Ce^{3+} . Therefore, it is likely that most, if not all, of the Ce in ATS is in the 3+ oxidation state. Knowledge of the exact extent to which this occurs in ATS glass awaits further work.

Elemental Roles in ATS Glass

The ATS glass currently includes the following component oxides: Li_2O , Na_2O , K_2O , Cs_2O , ZnO , B_2O_3 , Al_2O_3 , SiO_2 , TiO_2 , SnO_2 , ZrO_2 , and Gd_2O_3 (see Table I). In the ATS glass, B, Al, Si, and possibly Zr are "network formers". The Ti, Al, Zr, Th, and Zn (if two-fold coordinated by oxygen) have intermediate structural roles depending on their concentrations. The Sn, Th, Li, Zn, Na, K, and Cs act as "network modifiers" [5]. The exact role an element plays in the ATS glass depends in part on the concentration of each element and on the interactions between the element of interest and the other elements in the glass [6].

Elements are added to the ATS glass for the following reasons: Li(I) increases glass durability (because of its strong bonding with oxygen [7]) and, along with the other alkalis and Al, increases the solubility of the 4+ cations in the glass [7]. In addition to increasing the solubility of the 4+ cations, the alkali elements (with Li having the greatest effect [7]) tend to decrease the viscosity of the melt. Also, both Na(I) and K(I) can reduce glass durability. The ^{137}Cs , replaced by a small amount of nonradioactive Cs in our tests, has little impact on melt viscosity [7].

Zinc(II) increases glass durability and lowers the melting point. Boron(III) lowers the melt viscosity and improves the ease with which the glass components react to form a homogeneous melt [7]. Aluminum(III), in conjunction with the alkalis, stabilizes the 4+ cations [7,8] and is present in the ATS glass at a low concentration (~1.2wt%). Both Ti(IV) and Sn(IV) improve glass durability; SnO_2 significantly decreases the melt viscosity. The benefit of adding Sn to increase durability outweighs the disadvantage caused by decreased melt viscosity. The ZrO_2 increases durability owing to the strong Zr-O bond [6]. The ZrO_2 also increases the viscosity of the melt, although not to the extent that Ti or Th do in similar amounts. Gadolinium(III) is added as a neutron absorber at a 1:1 atomic percent basis with Pu.

Glass Development Rationale for Increasing Pu Loading

As noted above, the primary objective for the continued development of the ATS glass formulation is to increase the Pu content of the glass. To decide how to increase the Pu loading, information on the structural environment of the elements in the glass at their respective concentrations would be useful. Important questions are: (1) what is the most suitable surrogate for Pu in ATS glass, and (2) how do the structural environments of Th, Ce, and Pu change as a function of concentration? Our preliminary results indicate that Th^{4+} is approximately 1.8 times as soluble in ATS glass as Pu. This is in general agreement with the work by Veal et al. [8], who determined that Th was 2.5 times as soluble as Pu. Since their glass composition was a simple

alkali silicate glass, the difference is not too surprising. Knowledge of structural environments may help us to explain observed solubility differences among these elements. The structural environments of other elements in the ATS glass are also uncertain, and how these components may affect the solubilities of Th and Pu in the glass is of interest. Thus, a long-term goal in the development of Pu-containing ATS glass is to obtain information on the structural environments of the elements in these glasses using atomic-absorbance methods, such as X-ray absorption spectroscopy (XAS).

In addition to removing Zn, we will attempt to increase the solubility of the surrogate elements Th and Ce, as well as Pu, by taking advantage of the alkali/aluminum ratio and its effect on the solubility of 4+ cations. Studies by Watson [9] and Ellison and Hess [10] demonstrated the importance of this ratio on the solubility of 4+ cations in silicate melts.

Until spectroscopic data are available, however, we are proceeding in the following manner. The 4+ elements originally added to increase the durability are being systematically removed in an attempt to increase the Pu solubility, with the hope that removing those 4+ elements that have structural roles similar to Pu(IV) will free these atomic sites for Pu. We believe that Th(IV), Ce(III and possibly IV or a mixture of oxidation states) or Pu(IV) can serve a dual role: that of fulfilling the intended task of increased loading as well as maintaining (or increasing) the glass durability.

The consequences of removing the 4+ cations Zr, Sn, and Ti to increase the Pu loading will be evaluated by measuring both viscosity and changes in Ce, Th, and Pu solubilities. The viscosity of the melt must be at least 2-10 Pa•s (20-100 poise) at 1200°C [3], so a value of 2 Pa•s (20 poise) is being used as a benchmark to which the glasses will be compared. Our preliminary viscosity measurements (Table II) on the ATS base glass (i.e., no Pu, Th, Ce, or Gd) indicate that the viscosity at 1200°C is 3.1 Pa•s (30.6 poise), a factor of approximately 1.5 times greater than required.

ATS GLASS TESTING

Corrosion testing of the ATS glass (samples P10 and P5) includes the following tests. These tests are done in deionized water (DIW) to provide a consistent, comparable set of data to demonstrate whether the waste form may undergo catastrophic reaction under test conditions that limit the reaction of the waste form with water.

- (1) MCC-1. A 3-day test in DIW at 90°C and a surface area-to-volume ratio (S/V) of 10 m², which provides an estimate of the forward rate of waste form reaction. This information bounds the rate of the waste-form reaction under a specific test condition but does not provide useful information on the release of Pu or the neutron absorber Gd.
- (2) PCT-B. A variable time test in DIW at 90°C and a S/V of 20,000 m², which allows evaluation of the waste form reaction at high S/V ratios and static conditions known to increase the concentration of glass components in the leachate (DIW) and to accelerate the onset of alteration phase formation. This information provides an indication of initial Pu and Gd release to solution and also can be used to infer the mechanism(s) of waste-form reaction.

Table II. ATS Base Glass Viscosity Data

Temperature (°C)	Temperature 10000/T(K)	Viscosity (Poise)	Viscosity (log ₁₀ Pa•s)
1200	6.789	30.6	0.486
1150	7.027	47.6	0.678
1100	7.283	77.8	0.891
1050	7.558	133.6	1.126
1000	7.855	244.1	1.387
950	8.177	487.6	1.688
900	8.525	1093	2.039

- (3) PCT-A. A 7 day test in DIW at 90°C and a S/V of 2,000 m⁻¹, which provides comparative information (batch to batch) required by the Waste Acceptance Product Specifications used for DWPF glass.
- (4) Vapor Hydration Test (VHT). A 7-56 day test at 200°C provides qualitative information on the tendency of a waste form to form alteration phases [3], and allows us to evaluate the distribution of Pu and Gd within the alteration phases, if applicable.

Normalized release values were calculated for the MCC-1 and product consistency tests (PCT) based upon the analyzed (P10) and as-prepared (P5) glass compositions (Table 1), the geometric surface area, and the solution-analyses from the terminated tests. The equation used to calculate normalized releases is

$$NL(i) = \frac{(C_i - C_i^o)}{\left(\left(\frac{S}{V}\right) \cdot f_i\right)} \quad (1)$$

where NL(i) is the normalized release in g/m², C_i is the concentration of element i in the leachant in g/m³, C_i^o is the concentration of element i in the initial leachant solution in g/m³, S is the surface area of the glass in m², V is the volume of the glass in m³, and f_i is the weight fraction of element i in the glass.

MCC-1 Results

Three-day MCC-1 tests were completed in triplicate for the P5 ATS glass. Based on a comparison of various test methods performed earlier [11], these tests give an estimate of the forward rate of glass reaction. The precision of short term MCC-1 is generally less than that obtained for tests run at higher S/V for longer times, and the concentrations of the least-soluble elements are known with correspondingly low precision. Thus the measured releases of B, Si, and the alkalis are considered reliable and are used to estimate the forward reaction rate. The release of Pu and Gd are not used to estimate the release of these elements under repository conditions. Three-day MCC-1 tests were not completed for P10 because, at the time of testing, the test matrix was not designed to obtain this information.

The glass monoliths were polished using 240-grit abrasive paper, then placed in Teflon vessels with DIW. Solutions were collected at-temperature after 3 days at 90°C and were analyzed using inductively coupled plasma-mass spectrometry (ICP-MS); the pH of each solution was measured within 5 minutes of termination of the experiment. Specific details of the experimental method used to conduct MCC-1 tests are reported elsewhere [12]. Because Teflon vessels were used, acid strips of the vessel walls were not performed. The results are reported in Table III. All reported concentrations were blank-corrected by subtracting the average concentrations obtained from 24 blanks from the test solution results¹. The data for each sample for each glass component are in good agreement; the scatter is typical of this type of test.

PCT Results

Samples of both P10 and P5 glasses were subjected to PCT-A and PCT-B. The 7-day PCT-A were completed in duplicate with 1 gram of glass [13]; sufficient glass required for triplicate testing was not available. These tests were completed in stainless-steel vessels. Acid rinses were performed and analyzed using ICP-MS. Normalized releases, based upon the sum of both solutions, are reported in Table IV.

¹ In addition to conducting tests with glass monoliths (MCC-1) or powders (PCT), a blank test is completed for the same time period and temperature as the glass samples. The solutions from the blank tests are analyzed, and a running average is calculated.

Table III. Results from Triplicate MCC-1 Testing of the P5 ATS Glass

Element	Normalized Release (g/m ²)			
	Sample P5M3A1	Sample P5M3B1	Sample P5M3C1	Average
Li	4.4	4.4	3.9	4.2
B	3.6	2.7	3.1	3.2
Na	6.5	6.4	6.5	6.5
Al	3.6	0.7	2.3	2.2
Si	4.2	2.2	3.5	3.3
K	3.9	3.2	3.5	3.5
Ti	<MDL ^a	<MDL	<MDL	<MDL
Zn	0.45	1.6	0.49	0.8
Zr	0.01	0.03	0.007	0.02
Sn	0.02	0.03	0.005	0.02
Cs	4.6	2.8	3.5	3.6
Gd	0.03	0.05	0.01	0.03
Pu	0.06	0.10	0.05	0.07
pH	9.2	7.4	8.4	8.3

^a<MDL = less than minimum detection limit.

Table IV. Normalized PCT-A Releases from of the ATS Glasses P10 and P5

Element	Normalized Release (g/m ²) ^a			
	P10 ATS Glass		P5 ATS Glass	
	Sample A	Sample B	Sample A	Sample B ^b
Al	0.007	0.008	0.014	0.13
B	1.6	1.6	0.98	1.3
Cs	0.23	0.23	0.20	0.33
K	0.66	0.71	0.66	0.94
Li	1.6	1.7	1.2	1.6
Na	1.1	1.2	0.98	1.3
Si	0.38	0.40	0.53	0.55
Sn	0.017	0.017	0.024	0.092
Ti	0.001	0.0006	0.009	0.088
Zr	0.0006	0.0008	0.006	0.064
Zn	0	0	0.007	0.12
Pu	0.003	0.003	0.015	0.15
Gd	0.001	0.012	0.018	0.21
pH	10.2	10.2	11.1	11.0

^aTests performed at 2,000 m⁻¹ and 90°C, and (NL)_i values are calculated using the total elemental contribution released from the glass. Concentrations were blank-corrected.

^bThe elemental concentrations for this sample are suspect because of possible contamination of the acid-strip solution.

Results from PCT-B were obtained using the same procedures as PCT-A; a single sample was used for each period. These tests were also performed in stainless-steel vessels, which were acid stripped after termination of the tests. Results are reported in Table V for both P10 and P5. For the P5 ATS glass, only the 3-and 28-day tests have been terminated and analyzed to date. The Si and B data reported in Table V are plotted in Fig. 1.

Table V. Normalized Release Data for PCT-B Tests for the Pu ATS Glasses P10 and P5

Component	Normalized Release (g/m ²) ^a						
	P10 ATS Glass					P5 ATS Glass	
	3 day	28 day	98 day	182 day	364 day	3 day	28 day
Al	0.001	0.002	<MDL ^b	<MDL	0.004	0.001	0.005
B	0.35	0.59	0.99	1.2	1.4	0.26	0.48
Cs	0.037	0.065	0.076	0.079	0.20	0.045	0.058
K	0.12	0.21	0.24	0.28	0.40	0.16	0.22
Li	0.37	0.80	1.0	1.1	1.4	0.35	0.56
Na	0.25	0.76	0.69	0.76	0.92	0.32	0.57
Si	0.057	0.054	0.086	0.072	0.088	0.14	0.26
Sn	0.002	0.001	0.0004	0.0007	0.035	0.002	0.0008
Ti	0.001	0.003	0.0007	<MDL	0.010	0.002	0.003
Zr	0.0001	<MDL	0.0002	0.0005	0.024	0.0005	0.001
Zn	na ^c	na	na	na	na	0.002	0.002
Pu	0.0009	0.002	0.0014	0.001	0.002	0.002	0.002
Gd	0.003	0.007	0.005	0.004	0.008	0.003	0.003
pH	10.4	10.3	10.5	10.7	10.7	11.2	11.3

^aTests performed at 20,000 m⁻¹ and 90°C, and (NL), values are calculated using the total elemental contribution released from the glass. All concentrations were blank-corrected.

^b<MDL = less than the minimum detection limit.

^cna = not analyzed; zinc was not included in the P10 formulation.

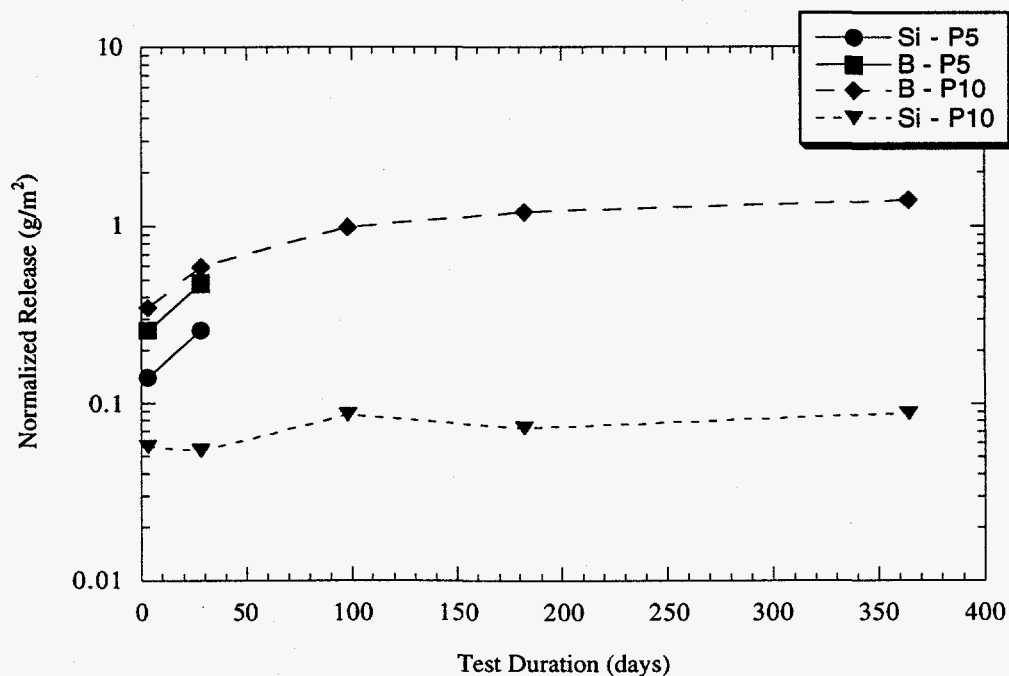


Fig. 1. Plot of Si and B Calculated Normalized Release for PCT-B

CONCLUSIONS

A systematic approach is being used to examine the solubility of Pu in an ATS glass, with the aim of increasing Pu loading while maintaining glass durability. Two ATS glasses containing Pu have been prepared and are currently being examined by a variety of standard leach tests. Also, several glasses containing the Pu surrogates Th and Ce have been prepared. The solubility of Th in ATS glass was determined to be approximately two times that of Pu, which is consistent with a previous study. The viscosity of the ATS base glass was measured to be 3.06 Pa·s (30.6 Poise) at 1200°C, which is a factor approximately 1.5 times the minimum required for a Pu waste glass. Normalized releases for several elements were determined from 3-day MCC-1 testing of the P5 ATS glass. The Si- and B-based normalized releases are 3.3 g/m² and 3.2 g/m², respectively. Results from the PCT-B testing of the initial ATS glass (P10) have been compiled through 364 days. For the revised composition (P5), 3- and 28-day tests have been completed. Results appear to be somewhat similar for the two glasses; however, for the P5 glass, the pH is much higher as is the Si release. We caution that long-term data are still needed to evaluate the release of Pu and Gd from the glass. In addition, how these elements are distributed between the solution and the acid strip are relevant to the potential segregation of each element in a repository environment.

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