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MCC MATERIALS
CHARACTERIZATION CENTER

Fabrication and Characterization of MCC Approved Testing Material—ATM-12 Glass

J. W. Wald

October 1985

Prepared for the U.S. Department of Energy
under Contract DE-AC06-76RLO 1830

Pacific Northwest Laboratory
Operated for the U.S. Department of Energy
by Battelle Memorial Institute

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FABRICATION AND CHARACTERIZATION OF MCC
APPROVED TESTING MATERIAL - ATM-12 GLASS

J. W. Wald

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Pacific Northwest Laboratory
Richland, Washington 99352

PREFACE

This report, prepared by the Materials Characterization Center, is one in a series of PNL-5577 reports on the MCC's nuclear waste glass Approved Testing Materials (ATMs). The suffix number shows the specific ATM described (e.g., ATM-1 is the subject of PNL-5577-1; ATMs -2, -3 and -4 are included in PNL-5577-2,3,4; etc.). Additional reports will be added to the series as the corresponding glass ATMs are produced by MCC. For a summary of existing ATMs, see the most recent issue of PNL-4955, Approved Reference and Testing Materials for Use in Nuclear Waste Management Research and Development Programs. PNL-4955-2 (Mellinger and Daniel) was issued in December 1984.

The series currently planned includes the following reports.

<u>Report No.</u>	<u>Subject Glass</u>
PNL-5577-1	ATM-1
PNL-5577-2,3,4	ATM-2, ATM-3, ATM-4
PNL-5557-5	ATM-5
PNL-5577-6	ATM-6
PNL-5577-8	ATM-8
PNL-5577-9	ATM-9
PNL-5577-10	ATM-10
PNL-5577-11	ATM-11
PNL-5577-12	ATM-12
PNL-5577-18	ATM-18

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1.0 SUMMARY

The Materials Characterization Center (MCC) Approved Testing Material ATM-8 is a borosilicate glass that incorporates elements typical of high-level waste (HLW) resulting from the reprocessing of commercial nuclear reactor fuel. Its composition is based upon the simulated HLW glass type 76-68 (Mendel et al. 1977) to which depleted uranium, technetium-99, neptunium-237 and plutonium-239 have been added at moderate to low levels. The glass was requested by the Nevada Nuclear Waste Storage Investigations (NNWSI) Project. It was produced by the MCC at the Pacific Northwest Laboratory (PNL) operated for the Department of Energy (DOE) by Battelle Memorial Institute. ATM-8 glass was produced in April of 1984, and is the second in a series of testing materials for NNWSI.

Three kilograms of ATM-8 glass were produced from a feed stock melted in a nitrogen atmosphere glove box at 1175°C in platinum crucibles, and formed into stress annealed rectangular bars. Bars of ATM-8 were nominally 1.9 x 1.9 x 10 cm, with an average weight of ~106 g for a 10-cm bar. Twenty-five bars were made during a 3-day fabrication period.

Table 1.1 lists the analyzed composition of ATM-8. Uranium was analyzed by pulsed laser fluorometry, technetium by beta counting analysis and neptunium and plutonium by alpha energy (counting) analysis. All other elements listed were analyzed by inductively coupled argon plasma atomic emission spectroscopy (ICP). Microscopy was conducted on a polished section of a transverse wafer from one bar. Quantitative image analysis indicated random porosity of ~0.2 vol% with nominal pore diameters ranging from ~5 to 450 μm . Three distinct crystalline phases were also observed as finely dispersed phases located randomly throughout the sample. Spherical particles of metallic appearance were characterized as high in palladium by Energy Dispersive Spectroscopy (EDS) on the scanning electron microscope (SEM) and were probably reduced to the metal phase during glass melting. A fine needle-like structure high in ruthenium is likely RuO_2 , and a rectangular feature high in chrome and zinc is likely CrZn -spinel.

TABLE 1.1. Analyzed Composition of ATM-12 Glass(a)

Oxide	wt%	Standard Deviation
<u>Stable Constituents</u>		
Ag ₂ O	NA	NA
Al ₂ O ₃	0.20	0.03
B ₂ O ₃	9.45	0.48
BaO	1.00	0.01
CaO	2.02	0.03
CdO	0.03	0.01
CeO ₂	0.84	0.03
Cr ₂ O ₃	0.39	0.01
Cs ₂ O	NA	NA
Eu ₂ O ₃	0.03	0.01
Fe ₂ O ₃	9.67	0.17
Gd ₂ O ₃	<DL, 0.05	--
La ₂ O ₃	0.45	0
MnO ₂	0.02	0.001
MoO ₃	1.93	0.01
Na ₂ O	12.68	0.13
Nd ₂ O ₃	1.64	0.02
NiO	NA	NA
P ₂ O ₅	NA	NA
PdO	0.09	0.01
Pr ₆ O ₁₁	<DL, 1.0	--
RbO	<DL, 1.14	--
Rh ₂ O ₃	NA	NA
RuO ₂	0.91	0.27
SiO ₂	40.35	0.39
Sm ₂ O ₃	0.32	0.04
SnO ₂	0.23	0.07
SrO	0.15	0
TeO ₂	0.15	0.03
TiO ₂	2.94	0.02
Y ₂ O ₃	0.14	0
ZnO	4.92	0.07
ZrO ₂	1.91	0.01
<u>Radioactive Constituents</u>		
Am ₂ O ₃	0.091	0.001
MpO ₂	0.198	0.007
PuO ₂	0.043	0.004
Tc ₂ O ₇	0.208	0.009
UO ₂	4.15	0.08
TOTAL	97.15	

(a) Based on analysis of 4 specimens taken from 3 bars. Standard deviations are for an individual analysis.

NA = Not analyzed at this time.

<DL = Below the detection limit indicated.

(probably RuO_2) was also observed. A third feature observed consisted of rectangular crystals. Rectangular microstructure of this type has typically been associated with spinel crystals in glass; in ATM-12 it was tentatively found to be high in Ru and Np. X-ray diffraction (XRD) analysis on the same sample used in the SEM examination revealed no distinct crystalline features, indicating that the crystalline content is probably less than ~5% by weight. An autoradiograph was obtained of the polished sample used for microscopy, as a guide to total radionuclide distribution. Exposure shading indicated a generally uniform distribution of activity throughout the bulk, with no distinct high-concentration regions.

ATM-12 glass has been provided to the NNWSI Program, in the form of bars, crushed powder and special castings. As of August 1985 approximately 590 g of ATM-12 is available for distribution. Requests for materials or services related to this glass should be directed to the Materials Characterization Center Program Office, PNL.

2.0 INTRODUCTION

The objective of the MCC is to assist the DDE projects responsible for nuclear waste form production, transportation, and disposal to assemble a material database of defensible precision and accuracy. A major responsibility of the MCC is to arrange for the formulation, acquisition and distribution of reference and testing materials essential for quantitative evaluation of nuclear waste forms and repository systems under development in the U.S. The purpose of this document is to provide the fabrication and characterization details of the Approved Testing Material (ATM) ATM-12 glass, which was produced under the MCC charter.

ATM-12 is a borosilicate glass that incorporates elements typical of HLW resulting from the reprocessing of commercial nuclear reactor fuels. The composition has been adjusted to match that predicted for HLW type 76-68 glass (Mendel et al. 1977) at an age of 300 years. Radioactive constituents contained in this glass include depleted uranium, technetium-99, neptunium-237, plutonium-239, and americium-241. The glass was requested by NNWSI staff and produced at PNL July to November 1984. ATM-12 glass is the third in a series of testing materials produced for NNWSI, where it was designated as "Glass III."

Section 3 of this report provides details of the fabrication of the glass and the special forms; Section 4 summarizes the analysis and characterization of the final glass product. ATM-12 was produced under the guidance of the MCC Program Quality Assurance Plan PGM-4, Rev. 2, and PNL QA Plan PNL-MA-65, which were in effect during the course of the work. Specific run plans were prepared for batch preparation and glass preparation. These run plans were supplemented by full documentation in official laboratory records books, as required by the QA plans.

3.0 FABRICATION^(a)

The fabrication of ATM-12 glass began on July 7, 1984 and was completed by November 1, 1984. All work involved in the fabrication of this glass took place in Labs 500 and 410, 325 Building, 300 Area and Labs 208, 212, and 220, 308 Building, 300 Area. ATM-12 glass was prepared from batched base chemicals (oxides, nitrates, carbonates) to which actinide solutions were added as dopants.

3.1 STARTING MATERIALS

ATM-12 glass was batched from base chemicals because the unique composition requested ("300 years old 76-68") could not be obtained by additives to existing stocks of MCC-76-68 glass, as in the case of ATM-1 (Wald 1985a) and ATM-8 (Wald 1985b). Table 3.1 lists the oxide composition requested for ATM-12 by NNWSI. All chemicals were obtained from in-house stock of reagent grade chemicals available specifically for glass-making operations. Chemical stock/lot numbers and manufacturers were recorded so that traceability to the original source was maintained; this information is given in Table A.1, Appendix A.

The radioisotope additives to this glass include ^{99}Tc , depleted U, ^{237}Np , ^{239}Pu , and ^{241}Am . Technetium-99 as NH_4TcO_4 was obtained from Oak Ridge National Laboratory (ORNL) Isotope Sales from their Batch P-50 as isotope order number 37-1174. The source of the depleted uranium was from on-hand stock of 0.15% ^{235}U UO_2 powder. The ^{237}Np was from on-hand quantities of anion-exchange purified stock solution, while the ^{239}Pu was obtained from on-hand "weapons-grade" stock solution (i.e., 7.5% ^{240}Pu). The larger fraction of ^{241}Am used in this glass (i.e., 92%; 3.40 g as oxide) was purchased from ORNL as AmO_2 powder in PNL Purchase Order Number J-7174 and received as ORNL Batch Number 80AMB-1. A smaller fraction of americium oxide (8%; 0.31 g as oxide) was obtained from on-hand stock estimated at several years old. Further information regarding the dopant solutions is given in Appendix B.

(a) The use of trade and manufacturers' names in this report is for documentation purposes only and does not imply endorsement by PNL.

TABLE 3.1. Requested Oxide Composition of ATM-12 Glass

Oxide	wt%
<u>Inerts</u>	
B ₂ O ₃	9.5
CaO	2.0
Cr ₂ O ₃	0.41
Fe ₂ O ₃	9.72
Na ₂ O	12.5
NiO	0.20
P ₂ O ₅	0.48
SiO ₂	40.1
TiO ₂	3.0
ZnO	5.0
<u>Fission Products</u>	
Ag ₂ O	0.03
BaO	1.00
CdO	0.04
CeO ₂	1.01
Cs ₂ O	0.53
Eu ₂ O ₃	0.05
Gd ₂ O ₃	0.06
La ₂ O ₃	0.50
MoO ₂	1.75
Nd ₂ O ₃	1.66
PdO	0.57
Pr ₆ O ₁₁	0.48
RbO	0.13
Rh ₂ O ₃	0.21
RuO ₂	1.01
Sm ₂ O ₃	0.37
SnO ₂	0.04
SrO	0.14
Tc ₂ O ₇	0.84
TeO ₂	0.21
Y ₂ O ₃	0.20
ZrO ₂	1.87
<u>Actinides</u>	
Am ₂ O ₃	0.12
NpO ₂	0.23
PuO ₂	0.04
UO ₂	4.00
TOTAL	100.00

3.2 BATCH AND GLASS PREPARATION

The procedures for preparation of the precursor batch from the base-chemical constituents and the final glass from this batch were described in the Batch and Glass Preparation Run Plans. The batch preparation process was as follows:

- All nonradioactive chemicals (except CdO) whose individual net batch size was less than 50 g (Table A.2, Appendix A) were weighed out and blended collectively in an alumina mortar and pestle. This blend, along with the other nonradioactive (>50 g) chemicals, was placed into a large "hardened white porcelain" jar mill along with approximately 150 burundum grinding cylinders. Net volume occupancy of the contents was ~67% of the grinding chamber.
- The chemical batch was milled by rolling at moderate speed for a total time of 3 h; the contents were inspected visually for blending progress at 30-min intervals. At the conclusion of the milling operation, the grinding cylinders were removed and the milled batch recovered from the jar.
- Portions of the batch material were placed into two large platinum crucibles (~500 mL) and put into the melting furnace preheated to 1150°C. The melt atmosphere was air. Charges were added to the melting batches at half-hour intervals until all had been added; three additions were made to the initial charges. After the final charge was added, the mixture was held at temperature for 3 h.
- At the conclusion of the 3-h hold period, the mixture was visually checked for surface foam and melt consolidation. Mechanical stirring or agitation was applied and the crucibles returned to the furnace for an additional 15-min.
- When the mixture was visually "glassified," the entire contents of the crucibles were poured (quenched) onto a cleaned stainless-steel plate at room temperature (~25°C).
- The quenched-glass batch was then crushed and ground to -100 mesh (-150 μ m) using an Angstrom disc mill and a random sample was taken

for chemical analysis. The chemical analysis indicated that the batch was too low in ruthenium and cesium. Additional ruthenium oxide and cesium nitrate were blended with the defined quantities of cadmium oxide and uranium oxide and the entire four component mixture was added to the previously prepared crushed batch. The mixture was blended together using standard ball-mill technique, as described previously, for a 3-h period in a Norton jar mill.

- A collective dopant solution was prepared containing the actinides plus additional palladium, all as required to achieve the specified concentrations (see Appendix B). The dopant solution volumes were adjusted to keep the liquid-to-glass-powder ratio at approximately 750 mL/3000 g; the ratio actually achieved was approximately 850 mL/2915 g. A main objective in this procedure was to keep nitrate concentrations of the liquid as dilute as possible to reduce nitrate off-gas and glass-product cementitious reactions associated with high-nitric concentrations.
- The collective dopant solution, as described above, was added to the batch feedstock powder (in one lot) in a 4-L Pyrex beaker and stirred by hand until thoroughly mixed and wetted. The mixed slurry was then dried/calcined in 250- to 500-mL batches contained in a stainless-steel beaker at 150° to 200°C for about 2 h.
- After drying, the entire batch was collected in a single container and blended by hand mixing for a few minutes for homogeneity. The batching step was now completed and the material was ready for glass preparation.
- Doped-glass powder was melted in ~600-g batches in a single Pt crucible at 1150°C for 1 h in a nitrogen-atmosphere glove box. After the designated hold time, each batch in this melt run was quenched on a stainless-steel plate at approximately 25°C. Five melt/quench cycles were required. When all material had been melted and quenched, the glass batch was crushed and ground in an Al₂O₃ mortar

and pestle and sieved to -8 mesh (-2.36 mm). The resulting glass was then mixed by hand in a single container.

- The glass powder was melted a second time in ~600 g batches in a single Pt crucible at 1150°C for 1 h in a nitrogen-atmosphere glove box and cast into bars or the special form cylinders. Cast bars were formed in stainless-steel bar molds to a nominal dimension of 1.9 x 1.9 x 10 cm in size. The bar mold was initially preheated to about 350°C, a single bar cast at a time, and when cool enough to handle without slumping, the bar was transferred to the annealing furnace at 500°C. Elapsed time from pour to annealing furnace was about 1.5 to 2 min.
- All cast materials were annealed at 500°C in a Hoskins type FD-104 furnace for a minimum of 2 h (maximum 5 h), followed by a furnace cool (~175°C/h) to 25°C. The atmosphere of the anneal furnace was nitrogen (glove-box atmosphere).

3.2.1 Preparation of Special Forms

Six special forms were requested for delivery to NNWSI (Table 3.2). The production of these special forms from the glass fabricated above is described in the following sections. All glass was processed in a nitrogen-atmosphere glove box.

3.2.1.1 Crushed and Sized Powder

Crushed glass was produced from the pieces of glass remaining after removal of core drilled cylinders (Section 3.2.1.2) from ATM-12 bars 1, 2, 3, 4, and 5. Bar pieces and coring remains were coarse crushed using a hardened steel punch and die and then ground to size using an automatic Al₂O₃ mortar and pestle. Material recovered from the -40 to +80 (U.S. sieve designation) mesh fraction was packaged for shipment. Sizing was performed by manually working the dry crushed material through the stainless-steel sieves with brushing and tapping actions.

TABLE 3.2. Special Form Requirements Requested by NNWSI
for MCC ATM-12 Glass

<u>Form</u>	<u>Physical Description</u>	<u>Quantity</u>
A	Crushed glass, -40 to +80 mesh size	50 g
B	Discs, 1-cm dia. x 3-mm thick	80 discs
C	Discs, dimensions as in "B" but with 304L SS rims	30 discs
D	Cylinder, 1-cm dia. x 1-cm long	10 cylinders
E	Cylinders, dimensions as in "D" but in 304L SS casing	10 cylinders
F	Tapered cast cylinders (1° Taper) 1.6-cm dia. x 2-cm long	20 cylinders

3.2.1.2 Uncased Discs and Cylinders

Cylindrical cores were removed from ATM-12 bars 1, 2, 3, 4, and 5 by diamond core drilling with a water cooled Starlite Industries core-drill assembly. These cores were then cut to the specified lengths using a Buehler Isomet low-speed saw with a low-concentration diamond wafering blade and water as the cutting media. A saw-speed setting of "6" was used; the actual blade velocity was not determined. Discs were nominally 3-mm thick by 1.25-cm diameter, while cylinders were 1-cm long by 1.25-cm diameter. Note that the diameter is 1.25 cm and not the 1.0 cm as specified in Table 3.2. Eighty-six discs and 10 cylinders were produced.

3.2.1.3 Stainless-Steel Clad Discs and Cylinders

Stainless-steel clad cylinders, nominally 3.5-cm long, were cast at the same time and from the same melt source as the final bar forms. The tubular molds were fabricated by cutting lengths from seamless 304L stainless-steel tubing 1 cm in outside diameter (mill specification MFG (SS-SMLS) CQ-2-523238-1A, ASTM-A269/A213, ASME-6A213). The nominal wall thickness of this tubing was 0.9 mm. Prior to the actual pour, the glass-batch temperature was increased to 1250°C and held for 10 minutes. Glass was poured into the tubular sections which were preheated to 500°C. When casting was complete, the tubes were placed into the annealing furnace and held at 500°C for a minimum of 2 hours, followed by a furnace cool (~175°C/h) to 25°C. The tubular castings were cut to size using a Buehler Isomet low speed saw with a low-concentration diamond

wafering blade and Isocut fluid as the cutting media. A saw-speed setting of "10" was used; the actual blade velocity was not determined. Discs were nominally 3-mm thick by 1-cm diameter, while cylinders were 1-cm long by 1-cm diameter. Thirty discs and 11 cylinders were produced. Note that the 1-cm diameter is the outside dimension of the stainless steel cladding. The tube's inside diameter (diameter of the glass cylinders and discs) is 0.82 cm.

3.2.1.4 Tapered Cast Cylinders

The following fabrication outline lists the steps that were followed to produce the tapered cast cylinders (Form F) of ATM-12 glass. The required cast-glass dimensions were specified by NNWSI (private correspondence, Bates to Mellinger, May 17, 1984). The casting crucibles were specified by NNWSI as "a Pt 5% Au mold, ...a modified J. Lawrence Smith crucible from Engelhard Industries, per ANL purchase order D55090." The crucibles for this work were obtained from Johnson Matthey Co. on PNL P.O. J 7160 ARO using the same specification as above with the concurrence of NNWSI staff.

- Form F samples were poured directly from the second glass-melting cycle, between the pouring of bars ATM-12-8 and ATM-12-9. Melting was conducted in a Pt crucible in a nitrogen-atmosphere glove box at 1150-1200°C, a temperature range considered sufficient to allow adequate casting behavior. Melt temperature was adjusted to ~1200°C and held for about 5-10 minutes just before the casting of the cylinders.
- Molten glass was poured into the casting crucible mold which had been preheated to 500°C; the mold was completely enclosed in a mold/firebrick assembly. The assembly was transferred to an annealing furnace heated to 500°C and held for 30 minutes; cooled at ~100°C/h to 350°C; removed from the firebrick and allowed to cool to 25°C.
- The glass casting was removed from the crucible by inverting the crucible and gently shaking and tapping the bottom. Twisting of the glass rod in the crucible was avoided, and care was exercised to minimize surface scratching during this operation.

- Finished samples were a tapered right-circular cylinder 2.0 ± 0.1 cm in length with parallel cut ends normal to the cylinder axis. Cutting was performed on a Buehler low speed cut-off saw using a low-concentration diamond blade (No. 11-4254), at a speed setting of "6", and using water as the cutting fluid. A specially padded double-jaw chuck was used to hold the samples for the entire cutting operation. A ring gauge was used to determine the location of the first cut with a dimensional shift made to locate the second cut.

The special form platinum crucibles described earlier and used in this fabrication were the same crucibles that had been used on two previous glass batches (ATM-1c and ATM-8). The number of crucibles used (6) were initially expected to provide adequate service for the fabrication of Form F of ATM-1, -8, and -12 samples; however, this did not prove to be the case. The crucible surface condition was noticeably degraded by the time ATM-12 was cast and it was anticipated that this batch might have a high rejection fraction. Obvious surface imperfections (discoloration) were present and glass castings were increasingly more difficult to remove from the mold. More than 60 castings had been made prior to ATM-12 Form F, and only four crucibles remained usable for ATM-12 casting. After twenty additional castings of ATM-12 the crucibles were deemed completely unsuitable for further use.

3.3 MEASUREMENT AND TESTING EQUIPMENT

Fluke Digital Thermometer, Model No. 2190A, Calib. No. 364-79-06-007
 Fluke Digital Thermometer, Model No. 2100A, Calib. No. 362-79-06-005
 Mettler Balance, PC-4400, Calib. No. 368-06-01-008
 Mettler Balance, PC-400, Calib. No. 362-06-01-017
 Mettler Balance, B5, Calib. No. 362-06-03-014
 Mettler Balance, H-15, Calib. No. 362-06-03-053

3.4 OTHER EQUIPMENT

Deltec DT-31-1255 Furnace
 Deltec DT-31-S Furnace
 Hoskins Furnace, Model FO-104

Honeywell Digital Control Programmer DCP-7700
Love Model 49 Proportional Controller
Norton Jar Mill
Angstrom Disc Mill w/Tungsten Carbide Chamber
Fritsch Auto-Mortar and Pestle, Pulverisette II, Al_2O_3
Buehler Isomet Cut-off Saw
Starlite Industries Core Drill
Corning Model PC-354 Hot Plate
Glove Box, N_2 Atmosphere

3.5 PROCEDURES

MCC-SP-1 (Draft): Glass Component Preparation
MCC-SP-2 (Draft): Glass Preparation
Batch Preparation Run Plan - ATM-12
Glass Preparation Run Plan - ATM-12
RWS-325-10: Radiation Work Specification
RCP-325-10: Radiation Control Protocol
RWP-308-1 Rev. 14: Lab, Specific Radiation Work Procedure
CSS-308-1: Lab Specific, Criticality Safety Specification

3.6 IDENTIFICATION SYSTEM

Each final glass bar was uniquely numbered and labeled according to the order in which it was poured, as follows: ATM-12-1, ATM-12-2, ...etc. Production records for the glass are recorded on page 71 of PNL Laboratory Record Book BNW-3660, and include data on date of manufacture, bar ID number, mass, and current disposition. Table 3.3 gives the production summary for ATM-12 glass.

3.7 MATERIAL AVAILABILITY AND STORAGE

ATM-12 has been provided to NNWSI in the form of bars, crushed powder and special castings. As of August 1985, ~590 g of ATM-12 glass in bar form are available for distribution. Archive samples were taken from all initial bars.

TABLE 3.3. Product Summary for ATM-12 Glass^(a)

<u>MCC ID No.</u>	<u>Bar Mass, g</u>	<u>Pour Date</u>
ATM-12-1	112.2	10/15/84
ATM-12-2	101.4	10/15/84
ATM-12-3	113.0	10/15/84
ATM-12-4	115.3	10/16/84
ATM-12-5	99.5	10/16/84
ATM-12-6	119.7	10/16/84
ATM-12-7	120.2	10/16/84
ATM-12-8	113.6	10/16/84
NNWSI forms C, E, & F	15 SS tubes and 22 tapered cylinders	10/16-19/84
ATM-12-9	114.1	11/1/84
ATM-12-10	114.4	11/1/84
ATM-12-11	114.2	11/1/84
ATM-12-12	117.4	11/1/84
ATM-12-13	110.1	11/1/84
ATM-12-14	94.9	11/1/84
ATM-12-15	108.8	11/1/84
ATM-12-16	104.1	11/1/84
ATM-12-17	113.3	11/1/84
ATM-12-18	115.2	11/1/84
ATM-12-19	100.3	11/1/84

(a) Nominal bar size 1.9 x 1.9 x 10 cm.

All remaining MCC ATM-12 materials and archive samples are in the custody of the MCC Program Material Custodian and are physically stored in the 308 Building (PNL) Storage Vault.

4.0 CHARACTERIZATION

Characterization of ATM-12 glass was conducted by bulk density measurement, chemical analysis, light optical and scanning electron microscopy (SEM), X-ray diffraction analysis and radiochemical analysis. The characterization work was conducted between the time periods of November 1984 and March 1985. Individual bars chosen for analysis provided data from each day of production, and a set of samples from a single bar that allowed estimation of axial homogeneity. The results of these analyses are summarized in the following sections.

4.1 BULK DENSITY MEASUREMENT

Apparent density was determined on four samples, two samples each from end cuts of bars ATM-12-18 and ATM-12-19. Density was measured using ASTM procedure C 693-74, "Standard Method for Density of Glass by Buoyancy," modified to use 2.5-g samples instead of the 20-g sample specified. Density samples were prepared from the glass bars by removing a section from the end region of a bar, and trimming off all cast surfaces using an Isomet saw. Before measurement all samples were cleaned twice in alcohol in an ultrasonic cleaner and dried at 25°C. The average density for this glass was determined to be $3.0052 \pm 0.0044 \text{ g/cm}^3$ (Table 4.1).

TABLE 4.1. Measured Bulk Density of ATM-12 Glass

<u>Glass Bar No.</u>	<u>Specimen No.</u>	<u>Measured Density, g/cm³</u>
18	E10	3.0018
18	E20	3.0012
19	E10	3.0102
19	E20	<u>3.0077</u>
		Mean value = 3.0052 (std. dev. = 0.0044)

4.2 CHEMICAL ANALYSIS

Chemical analysis was performed by inductively coupled argon plasma atomic emission spectroscopy (ICP) for the majority of nonradioactive elements, using a Bausch and Lomb ARL-34000 unit. Aliquots of the same samples were used for radiochemical analysis of the radioactive constituents. Uranium was analyzed by laser fluorometry, technetium by beta-counting analysis of an isotope-specific separation, while neptunium, plutonium and americium were analyzed by alpha energy (counting) analysis techniques. A plutonium isotopic analysis was also performed on the initial dopant solution. Uranium analysis was conducted on a Scintrex UA-3 Uranium Analyzer, and alpha/beta counting were performed on counting systems based around Ludlum/Tracor Northern components.

Analytical samples were taken from an end sample of bars number ATM-12-1 and ATM-12-19, and the center and end of bar ATM-12-8. Each of the solid samples was ground to -100 mesh (-150 μm), fused with KOH in nickel crucibles and dissolved in dilute HCl to a known volume for analysis. Aliquots of these solutions were submitted for both ICP and radiochemical analysis.

The results of the chemical analysis of the four samples of ATM-12 glass are given in Table 4.2. Radiochemical data for the analyses of ATM-12 specimens are given in Table 4.3.

The standard deviation shown for most of the individual element analyses indicates that all 3 glass bars are homogeneous. When considering the probable concentrations of the elements not analyzed the summation of concentrations is above 98% which is considered satisfactory given the complexity of the sample and limitations of the analytical methods. Additional analyses by independent laboratories and methods are in progress for verification and supporting data.

In comparing the composition of ATM-12 with the composition requested, all major constituents were observed to match the target values with a few exceptions. Concentration of CeO_2 in ATM-12 was about 20% higher than requested, and SnO_2 was also observed to be much higher than target value. PdO was measured at about 15% of the requested value; the reason for this low value is unexplained but may be related to the difficulty in completely fusing metallic Pd, present in the glass, during analytical sample preparation. The final

TABLE 4.2. Analyzed Composition of ATM-12 Glass(a)

Oxide	wt%	Standard Deviation
<u>Stable Constituents</u>		
Ag ₂ O	NA	NA
Al ₂ O ₃	0.20	0.03
B ₂ O ₃	9.45	0.48
BaO	1.00	0.01
CaO	2.02	0.03
CdO	0.03	0.01
CeO ₂	0.84	0.03
Cr ₂ O ₃	0.39	0.01
Cs ₂ O	NA	NA
Eu ₂ O ₃	0.03	0.01
Fe ₂ O ₃	9.67	0.17
Gd ₂ O ₃	<DL, 0.05	--
La ₂ O ₃	0.45	0
MnO ₂	0.02	0.001
MoO ₃	1.93	0.01
Na ₂ O	12.68	0.13
Nd ₂ O ₃	1.64	0.02
NiO	NA	NA
P ₂ O ₅	NA	NA
PdO	0.09	0.01
Pr ₆ O ₁₁	<DL, 1.0	--
RbO	<DL, 1.14	--
Rh ₂ O ₃	NA	NA
RuO ₂	0.91	0.27
SiO ₂	40.35	0.39
Sm ₂ O ₃	0.32	0.04
SnO ₂	0.23	0.07
SrO	0.15	0
TeO ₂	0.15	0.03
TiO ₂	2.94	0.02
Y ₂ O ₃	0.14	0
ZnO	4.92	0.07
ZrO ₂	1.91	0.01
<u>Radioactive Constituents</u>		
Am ₂ O ₃	0.091	0.001
NpO ₂	0.198	0.007
PuO ₂	0.043	0.004
Tc ₂ O ₇	0.208	0.009
UO ₂	4.15	0.08
TOTAL	97.15	

(a) Based on analysis of 4 specimens taken from 3 bars. Standard deviations are for an individual analysis.

NA = Not analyzed at this time.

<DL = Below the detection limit indicated.

TABLE 4.3. Radiochemistry Data for ATM-12 Glass: Example of Calculation Methodology

Isotope	(1)	(2)	(3)	(4)	Oxide Form
	Mean Radiochemical Analysis, d/m/g glass	Isotope Activity Factor, d/m/mg Isotope	Element to Oxide Conversion Factor	wt% Oxide	
^{241}Am	6.30×10^9	7.618×10^9	0.9101	0.091	Am_2O_3
^{237}Np	2.74×10^6	1.566×10^6	0.8810	0.198	NpO_2
^{239}Pu	5.28×10^7	$1.377 \times 10^8(a)$	0.8832	0.043	PuO_2
^{99}Tc	4.98×10^7	3.771×10^7	0.6385	0.208	Tc_2O_7
U	3.66×10^{-2} g/g	NA	0.8815	4.15	UO_2

$$\text{Column (4)} = ([\text{col}(1)/\text{col}(2)/\text{col}(3)] \times 10^{-3} \text{ g/mg}) \times 100\%$$

Column (1) = Measured and reported values

Columns (2) and (3) = Published values

Column (4) = Calculated concentrations

(a) Assumed to be 100% ^{239}Pu .

technetium concentration was about 33% of the requested value which likely resulted from the inability to accurately predict and adjust for volatility losses during preparation. Americium and neptunium concentrations were slightly on the low side while plutonium concentration hit target and uranium was slightly higher than target value.

4.3. MICROSCOPIC EXAMINATION

Microscopy was performed on a transverse wafer removed from the center of bar number ATM-12-8. Figure 4.1 illustrates the typical microstructure of this glass. Three major features are present in this glass: metallic spheroids, fine needle-like agglomerations, and rectangular crystals. Energy dispersive spectroscopy (EDS) analysis on the SEM indicated the metallic spheroids to be primarily palladium; the needle-like structure and rectangular crystals were high in ruthenium. Neptunium was a major constituent in all EDS analyses, and may be an indication of interference due to the relatively high radioactivity of this sample.

Some porosity was also noted in the section examined. Porosity was estimated from a quantitative image analysis of a low-magnification optical photograph to be approximately 0.04% in the region photographed, compared to the measured open porosity of 0.03% as evaluated by saturated weight measurements taken during density evaluation. Pore sizes observed varied from about 5 μm to 250 μm in diameter.

Palladium-rich metallic particles are common in this type glass as Pd-Rh spheroids and have been reported previously (Turcotte et al.). Palladium will generally reduce to the metallic state during melting and cooling, and ruthenium will come out of glass solution as RuO_2 during the same process. Another phase suspected to exist but not positively verified by EDS analysis is an iron-rich spinel. The inability to positively identify this phase by EDS may be the result of interference by the relatively high sample radioactivity level previously mentioned.

Light Microscopy

5 μ m

Scanning Electron Microscopy

FIGURE 4.1. Typical Microstructure of ATM-12 Glass (polished section from center of Bar No. ATM-12-8; 5 μ m in upper photo and 10 μ m in lower photo)

Autoradiography on this glass showed a generally uniform shading indicating a uniform distribution of at least the most active constituent; the method used is not isotope-specific. Figure 4.2 illustrates a typical autoradiograph and corresponding photomicrograph for this glass.

4.4 X-RAY DIFFRACTION ANALYSIS

X-ray diffraction analysis was performed on a solid wafer cut from the center of bar ATM-12-8. Analysis was performed on a GE/Diano XRD-5 x-ray diffraction system located inside a nitrogen-atmosphere glove box. No crystalline peaks were observed in the sample examined. Based on the known characteristics of the XRD system used, the content of crystalline phases is estimated to be less than ~5% by weight.

FIGURE 4.2. Typical Autoradiograph (Right) and Corresponding Photomicrograph (Left) of ATM-12 Glass (Bar ATM-12-8, middle section)

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APPENDIX A

SOURCE DOCUMENTATION FOR NONRADIOACTIVE BASE CHEMICALS USED
TO BATCH ATM-12 GLASS

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SOURCE DOCUMENTATION FOR NONRADIOACTIVE BASE CHEMICALS USED TO BATCH ATM-12 GLASS

TABLE A.1. Sources of Nonradioactive Chemical Compounds Used
to Batch ATM-12 Glass

<u>Compound</u>	<u>Manufacturer</u>	<u>Lot #</u>
AgNO ₃	Baker	726364
Ba(NO ₃) ₂	MCB	10611
CaCO ₃	Mallinckrodt	KPXR
CdO	Argent	NA
Ce(NO ₃) ₃ ·6H ₂ O	Research Chemicals	NA
Cr ₂ O ₃	MCB	A10J25
CsNO ₃	Noah	118202
Eu(NO ₃) ₃ ·6H ₂ O	Argent	NA
Fe ₂ O ₃	Fisher	732511
Gd(NO ₃) ₃ ·6H ₂ O	Noah	11956-3213
H ₃ BO ₃	Fisher	711494
La ₂ O ₃	American Potash & Chem.	A0428
MoO ₃	Research Chemicals	NA
Na ₂ CO ₃	Fisher	724017
Na ₄ P ₂ O ₇ ·10H ₂ O	Baker	148094
Nd(NO ₃) ₃ ·6H ₂ O	Research Chemicals	NA
Ni ₂ O ₃ ·xH ₂ O	MCB	CB918NX345
PdO	Alfa	NA
Pd(NO ₃) ₂	Mathey-Bishop	32909
Pr(NO ₃) ₃ ·6H ₂ O	Research Chemicals	PRN3019
RbNO ₃	Research Chemicals	507-111
Rh ₂ O ₃	Alfa	NA
RuO ₂	Noah	5-11901
SiO ₂	Fisher	741764
Sm(NO ₃) ₃ ·6H ₂ O	Research Chemicals	SMN3017
SnO	Baker	322858
Sr(NO ₃) ₂	MCB	A4H28
TeO ₂	Fairmount	NA
TiO ₂	Sargent Welch	4128A
Y(NO ₃) ₃ ·6H ₂ O	Research Chemicals	Y-N-4-021
ZnO	Mallinckrodt	BVY
ZrO ₂	A. D. Mackay	NA

Note: NA indicates that no Lot # was given or available
on the material container.

TABLE A.2. Composition Worksheet for ATM-12 Glass
(Nonradioactive Components)

Oxide	Nominal wt%	Formula of Compound Used	Mult. Factor	Weight compound/ 3500 g Glass
Ag ₂ O	0.03	AgNO ₃	1.47	1.54
B ₂ O ₃	10.02	H ₃ BO ₃	1.78	624.25
BaO	1.06	Ba(NO ₃) ₂	1.70	63.07
CaO	2.11	CaCO ₃	1.78	131.45
CdO	0.04	CdO	1	1.40
CeO ₂	1.07	Ce(NO ₃) ₃ ·6H ₂ O	2.52	94.37
Cr ₂ O ₃	0.43	Cr ₂ O ₃	1	15.05
Cs ₂ O	0.56	CsNO ₃	1.38	27.05
Eu ₂ O ₃	0.05	Eu(NO ₃) ₃ ·6H ₂ O	2.54	4.45
Fe ₂ O ₃	10.26	Fe ₂ O ₃	1	359.10
Gd ₂ O ₃	0.06	Gd(NO ₃) ₃ ·6H ₂ O	2.49	5.23
La ₂ O ₃	0.53	La ₂ O ₃	1	18.55
MoO ₂	1.85	MoO ₃	1.13	73.17
Na ₂ O	13.19	Na ₂ CO ₃	1.71	774.40
Nd ₂ O ₃	1.75	Nd(NO ₃) ₃ ·6H ₂ O	2.61	159.86
NiO	0.21	Ni ₂ O ₃ ·xH ₂ O	0.98 ^(a)	7.20
P ₂ O ₅	0.51	Na ₄ P ₂ O ₇ ·10H ₂ O	0.296 P ₂ O ₅ /0.249 Na ₂ O	60.30
PdO	0.60	PdO	1	17.00
		Pd(NO ₃) ₂ [soln.]	0.0793 (g Pd/g soln.)	37.27 ^(b)
Pr ₆ O ₁₁	0.51	Pr(NO ₃) ₃ ·6H ₂ O	2.55	45.52
RbO	0.14	RbNO ₃	1.45	7.11
Rh ₂ O ₃	0.22	Rh ₂ O ₃	1	7.70
RuO ₂	1.07	RuO ₂	1	37.45
SiO ₂	42.30	SiO ₂	1	1480.50
Sm ₂ O ₃	0.39	Sm(NO ₃) ₃ ·6H ₂ O	2.55	34.81
SnO ₂	0.04	SnO	0.89	1.25
SrO	0.15	Sr(NO ₃) ₂	2.04	10.71
TeO ₂	0.22	TeO ₂	1	7.70
TiO ₂	3.17	TiO ₂	1	110.95
Y ₂ O ₃	0.21	Y(NO ₃) ₃ ·6H ₂ O	1.70	12.50
ZnO	5.28	ZnO	1	184.80
ZrO ₂	1.97	ZrO ₂	1.00	68.95

(a) Multiplication factor determined by analysis of actual Ni content of the stock oxide used.

(b) See text, p. B.1.

APPENDIX B

PREPARATION OF DOPANT SOLUTIONS FOR ATM-12 GLASS

APPENDIX B

PREPARATION OF DOPANT SOLUTIONS FOR ATM-12 GLASS

Actinides were added to the glass batch for ATM-12 as aqueous nitrate solutions to obtain the concentrations given in Table 2.1. Nonradioactive palladium was also added with the actinides as a nitrate solution to increase the concentration in the glass to the desired levels. Solutions were prepared or obtained as follows:

- Palladium - Palladium nitrate solution was used directly from commercially available stock solution (Mathey-Bishop, Inc., Lot No. 32909, 79.3 g metal/kg solution). Sufficient solution was weighed out to add 3.4 g of PdO to ~2900 g of glass batch. The total palladium oxide content of the final batch (from both dry powder and solution additions) was 21 g oxide/3500 g glass.
- Technetium - 30.78 g of NH_4TcO_4 was weighed out and dissolved in 500 mL of H_2O using sufficient 30% H_2O_2 to dissolve black radiolytic decomposition products (presumably TcO_2). The solution was allowed to stand for a two-day period for complete H_2O_2 decomposition before final adjustment to 500.0 ± 0.15 mL. A sample of this solution (0.100 mL) was diluted to 100.1 mL in H_2O and submitted for radiochemical analysis (Sample No. A12-TC). The remaining 499.9 ± 0.15 mL and flask rinses were used as the dopant source for ^{99}Tc .
- Neptunium - The ^{237}Np source was from on-hand stock of anion exchange purified stock solution. Approximately 154 mL of solution, previously analyzed as 39.4 g Np/L, was diluted to 200.0 ± 0.13 mL with 0.1 M HNO_3 . A sample of this solution (0.100 mL) was diluted to 10.1 mL in 0.3 M HNO_3 and submitted for radiochemical analysis (Sample No. A12-NP). The remaining 199.9 ± 0.13 mL plus flask rinses were used as the dopant source for ^{237}Np .

- Plutonium - The ^{239}Pu source was from on-hand stock of "weapons grade" (i.e., 7.5% ^{240}Pu) stock solution. About 36 mL of stock solution, previously analyzed as 29.1 g Pu/L, was diluted to 50.00 ± 0.02 mL with 0.1 M HNO_3 . A sample of this solution (0.100 mL) was diluted to 10.1 mL in 0.3 M HNO_3 and 0.100 mL of this dilution was again diluted to 10.1 in 0.3 M HNO_3 . This second dilution was submitted for radiochemical analysis (Sample No. A12-PU). The remaining 49.90 ± 0.02 mL plus flask rinses were used as the dopant source for ^{239}Pu .
- Americium - The major fraction of the ^{241}Am used as the dopant was purchased directly from ORNL as AmO_2 for this work. Of the ORNL AmO_2 , 3.40 g was added to 0.31 g of AmO_2 from on-hand oxide material (several years old) and the mixture was reacted with 5.0 mL of 15.8 M HNO_3 - 0.05 M HF. This solution was boiled to the viscous stage and diluted to 50.00 ± 0.02 mL with H_2O . A sample of the solution (0.100 mL) was diluted to 100.1 mL with 0.3 M HNO_3 and 0.100 mL of this dilution was again diluted to 100.1 mL with 0.3 M HNO_3 . This second dilution was submitted for radiochemical analysis (Sample No. A12-AM). The remaining 49.90 ± 0.02 mL of the Am solution plus flask rinses were used as the dopant source for ^{241}Am .

Results of the radiochemical analyses for the actinide dopant solutions described above are given in Table B.1. The last column in the table results from the quotient of Column 3/Column 4 multiplied by Column 5. The "dilution factor" is the multiplier which corrects the reported data for the analytical dilutions as described above. Mass Spectrometry isotopic analyses of plutonium in solution A12-Pu is given in Table B.2.

TABLE B.1. Radiochemical Analysis of Actinide Dopant Solutions for ATM-12 Glass

<u>Solution No.</u>	<u>Isotope</u>	<u>Radiochemical Analysis, d/m/mL</u>	<u>Activity Factor, d/m/mg</u>	<u>Dilution Factor</u>	<u>Isotopic Solution Concentration, mg/mL</u>
A12-TC	^{99}Tc	1.29×10^6	3.771×10^7	1001	34.24
A12-NP	^{237}Np	5.93×10^5	1.566×10^6	101	38.25
A12-PU	$^{239+240}\text{Pu}$	3.05×10^5	$1.640 \times 10^8(\text{a})$	1.01×10^4	18.78
A12-AM	^{241}Am	6.76×10^5	7.618×10^9	1.001×10^6	88.83

(a) Weighted average based upon isotopic analysis of 92.52% ^{239}Pu , 7.10% ^{240}Pu .

TABLE B.2. Mass Spectrometry Isotopic Analysis of Plutonium Solution A12-PU (dopant solution for fabrication of ATM-12 glass)

<u>Isotope</u>	
^{238}Pu	$0.021 \pm 0.003 \text{ wt\%}$
^{239}Pu	92.52 ± 0.04
^{240}Pu	7.10 ± 0.04
^{241}Pu	0.310 ± 0.002
^{242}Pu	0.050 ± 0.002

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