MCC MATERIALS
CHARACTERIZATION CENTER

**Fabrication and Characterization
of MCC Approved Testing
Material—ATM-8 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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PACIFIC NORTHWEST LABORATORY
operated by
BATTELLE
for the
UNITED STATES DEPARTMENT OF ENERGY
under Contract DE-AC06-76RLO 1830

Printed in the United States of America
Available from
National Technical Information Service
United States Department of Commerce
5285 Port Royal Road
Springfield, Virginia 22161

NTIS Price Codes
Microfiche A01

Printed Copy	
Pages	Price Codes
001-025	A02
026-050	A03
051-075	A04
076-100	A05
101-125	A06
126-150	A07
151-175	A08
176-200	A09
201-225	A010
226-250	A011
251-275	A012
276-300	A013

FABRICATION AND CHARACTERIZATION OF MCC
APPROVED TESTING MATERIAL - ATM-8 GLASS

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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-5577-5	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-8 Glass^(a)

Oxide	wt%	Standard Deviation
<u>Stable Constituents</u>		
Ag ₂ O	NA	--
Al ₂ O ₃	1.06	0.05
B ₂ O ₃	8.18	0.28 ^(b)
BaO	0.61	0.02
CaO	2.13	0.06 ^(b)
CdO	<DL, 0.005	--
CeO ₂	0.11	0.03
Cr ₂ O ₃	0.48	0.02
Cs ₂ O	NA	--
Dy ₂ O ₃	0.01	0.00
Eu ₂ O ₃	<DL, 0.01	--
Fe ₂ O ₃	8.18	0.24
Gd ₂ O ₃	<DL, 0.05	--
K ₂ O	NA	--
La ₂ O ₃	4.87	0.15
MgO	0.16	0.01
MnO ₂	0.06	0.01
MoO ₃	1.98	0.04
Na ₂ O	12.2	0.2 ^(b)
Nd ₂ O ₃	1.66	0.06
NiO	NA	--
P ₂ O ₅	0.57	0.07 ^(b)
PdO	0.37	0.10 ^(c)
Rh ₂ O ₃	NA	--
RuO ₂	0.58	0.10
SiO ₂	37.8	0.7 ^(b)
Sm ₂ O ₃	<DL, 0.05	--
SrO	0.47	0.01
TeO ₂	<DL, 0.1	--
TiO ₂	2.61	0.10
Y ₂ O ₃	<DL, 0.1	--
ZnO	4.37	0.12
ZrO ₂	1.99	0.05
<u>Radioactive Constituents</u>		
NpO ₂	0.38	0.02 ^(b)
PuO ₂	0.11	0.01 ^(b)
Tc ₂ O ₇	0.18	0.01 ^(b)
UO ₂	4.24	0.07 ^(b)
TOTAL	95.36	

(a) Based on analyses of 16 specimens taken from 9 different bars unless otherwise indicated. Standard deviations are for an individual analysis.

(b) From analyses of 4 specimens taken from 3 different bars.

(c) From analyses of 12 specimens taken from 9 different bars.

NA = Not analyzed.

ND = Analyzed but not detected.

<DL = Below detection limit indicated.

These are materials which have come out of glass solution during glass melting and and cooling. X-ray diffraction analysis on the same four samples that were submitted for ICP analysis did not indicate the presence of any distinct crystalline peaks, so it is estimated that the crystalline level in this glass is less than ~5 wt%.

Autoradiographs were taken of the same sample examined by optical and scanning electron microscopy. Exposure shading indicated a generally uniform distribution of activity throughout the bulk with higher activity levels associated with pore or void regions. The technique used is not isotope specific, so the result presented is only a guide to radionuclide distribution. The high concentrations of activity around pore regions are probably associated with technetium concentration.

ATM-8 glass has been provided to the NNWSI Project in the form of bars, crushed powder and special castings. As of August 1985, approximately 550 g of ATM-8 glass 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 DOE 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-8 glass, which was prepared under the MCC charter.

ATM-8 is a borosilicate glass that incorporates elements typical of high-level waste (HLW) resulting from the reprocessing of commercial nuclear reactor fuels. 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 were added at moderate to low levels. The glass was produced in April of 1984 at the request of the NNWSI at PNL, and is the second in a series of glasses produced for NNWSI. Most of the glass produced was in the form of cast bars; special castings and crushed material were also produced. Information and instructions on these special forms were provided to the MCC by NNWSI staff in private correspondence. At NNWSI, ATM-8 was designated as "Glass II."

Section 3 of this report describes the fabrication of the glass and the special forms; Section 4 summarizes the analysis and characterization of the final glass product. ATM-8 was produced under the guidance of the MCC Program Quality Assurance (QA) Plan PGM-4, Rev. 2, and PNL QA Plan PNL-MA-65, which were in effect during the course of the work. No written technical procedures were prepared specifically for the fabrication and characterization of this glass. The procedures followed were fully documented in official laboratory record books, as required in the previously listed QA plans.

3.0 FABRICATION^(a)

The fabrication of ATM-8 glass began on March 5, 1984 and was completed by April 13, 1984. All work involved in the fabrication of this glass took place in Labs 208 and 220, 308 Building, 300 Area. Actinide dopant solutions were prepared in Lab 410, 325 Building, 300 Area and transferred to the 308 Building for the fabrication work. The ATM-8 glass was prepared with crushed type 76-68 glass.

3.1 STARTING MATERIALS

The glass which served as the feedstock for ATM-8 was MCC-76-68, Lot 1, bar numbers 52, 53, 54, 55, 56, and 57. The MCC-76-68 glass was produced for PNL by Penberthy Electromelt of Seattle, Washington under purchase order number A2820/A2823 AF and received at PNL in September 1980. The average analyzed composition of the MCC-76-68, Lot 1 glass used for ATM-8 is given in Table 3.1. The radioisotopes added to this glass to produce ATM-8 included ⁹⁹Tc, depleted U, ²³⁷Np and ²³⁹Pu. The technetium was received from Oak Ridge National Laboratory (ORNL) Isotope Sales as NH₄TcO₄ from their Batch P-30 as isotope order number 37-0496. The uranium was obtained from on-hand stock of 0.15% ²³⁵U UO₂ powder, while the ²³⁷Np was also from on-hand supply of oxide powder. The plutonium was also obtained from in-house stock solution of weapons-grade plutonium (92.5% Pu-239) which was last purified by anion exchange in May 1982. Further information for the radioisotope additives and their preparation for use as dopants is given in Appendix A.

Additional nonradioactive compounds were added to the glass as nitrate solutions of Ru, Rh, and Pd, with the aim of achieving the requested concentration of these elements. These nitrate solutions were obtained from a commercial vendor (Mathey-Bishop, Inc.) a number of years before the present work. No other historical or analytical information is available for these solutions.

(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. Analysis of MCC-76-68 Glass, Lot 1^(a)
(Feedstock for ATM-8 Glass)

Oxide ^(b)	Concentration, wt% ^(c)	Standard Deviation ^(d)
Al ₂ O ₃	0.84	0.03
B ₂ O ₃	8.65	0.06
BaO	0.62	0.01
CaO	2.29	0.04
CdO	0.05	0.01
CeO ₂	<0.02	--
Cr ₂ O ₃	0.50	0.02
Cs ₂ O	1.21	0.08
Oy ₂ O ₃	0.01	0.00
Eu ₂ O ₃	0.01	0.00
Fe ₂ O ₃	9.08	0.12
Gd ₂ O ₃	0.03	0.01
K ₂ O	0.09	0.03
La ₂ O ₃	4.89	0.06
MgO	0.18	0.01
MnO ₂	0.06	0.00
MoO ₃	2.17	0.01
Na ₂ O	11.60	0.95
Nd ₂ O ₃	1.65	0.01
NiO	0.25	0.03
P ₂ O ₅	0.56	0.08
SiO ₂	40.33	0.81
SrO	0.49	0.01
TiO ₂	2.86	0.04
ZnO	4.75	0.08
ZrO ₂	2.33	0.04
TOTAL	95.50	

(a) See Table 12, Mellinger and Daniel, 1984.

(b) Oxides listed were components of fabrication specifications for MCC-76-68. Other oxides of interest for ATM-8 glass, neither included in the MCC-76-68 nor analyzed, include Ag₂O, PdO, Rh₂O₃, RuO₂, Sm₂O₃, TeO₂, and Y₂O₃, and radio-isotopes NpO₂, PuO₂, Tc₂O₇ and UO₂.

(c) Based on ICP analysis of three bars from the 100 bars comprising MCC-76-68, Lot 1.

(d) Standard deviation of an individual analysis.

3.2 BATCH AND GLASS PREPARATION

Table 3.2 lists the oxide composition requested for ATM-8 by NNWSI. The batching process was as follows:

- Six bars of glass MCC-76-68, Lot 1, as listed above, were crushed, ground and sieved to -100 mesh ($\sim 150\ \mu\text{m}$) using a tungsten carbide disc mill.
- Nitrate dopant solutions of Np, Pu, Tc and U were prepared according to the procedure outlined in Appendix A. A collective dopant solution was made up of the individual constituent solutions so that the final concentration in the glass would be as given in Table 3.2 when added to the crushed MCC-76-68. The dopant solutions' concentrations were adjusted to keep the liquid to glass powder ratio of approximately 750 mL/3000 g. A main objective in this procedure was to keep the nitrate concentration of the liquid as dilute as reasonably possible to reduce nitrate offgas and glass-product cementitious reactions associated with high nitric concentrations.
- All operations involved in the glass manufacture from this point were carried out in a nitrogen atmosphere glove box. Dopant solution was added to the -100 mesh glass powder and stirred until thoroughly mixed and wetted. The slurry was dried in 250-mL batches in Pyrex beakers at 150-200°C for 2 hours.
- The dried powder was crushed, ground for ~ 5 minutes in an Al_2O_3 mortar and pestle, and sieved to -100 mesh. Grinding and sieving were repeated until all the doped glass was -100 mesh.
- Nitrate solutions of Ru, Rh, and Pd at approximately 0.1 g/mL concentration were added to the -100 mesh actinide-doped powder to bring the concentration of these constituents to the requested level. The slurry was stirred and blended as above.
- The slurry was dried in 250-mL batches in a stainless-steel beaker at 150-200°C for 2 hours.

TABLE 3.2. Requested Oxide Composition of ATM-8 Glass

<u>Oxide</u>	<u>wt%</u>
<u>Inerts</u>	
Al ₂ O ₃	0.48
B ₂ O ₃	9.08
CaO	2.28
Cr ₂ O ₃	0.34
Fe ₂ O ₃	8.86
K ₂ O	0.20
MnO ₂	0.08
Na ₂ O	12.77
NiO	0.20
P ₂ O ₅	0.67
SiO ₂	40.89
TiO ₂	2.92
ZnO	4.40
<u>Fission Products</u>	
Ag ₂ O	0.03
BaO	0.52
CdO	0.03
CeO ₂	0.99
Cs ₂ O	0.99
La ₂ O ₃	1.83
MoO ₂	1.74
Nd ₂ O ₃	1.28
PdO	0.56
Rh ₂ O ₃	0.17
RuO ₂	0.99
Sm ₂ O ₃	0.12
SrO	0.40
Tc ₂ O ₇	0.50
TeO ₂	0.28
Y ₂ O ₃	0.21
ZrO ₂	1.59
<u>Actinides</u>	
NpO ₂	0.40
PuO ₂	0.10
UO ₂	4.10
TOTAL	100.00

- The dried powder was crushed, ground in an Al_2O_3 mortar and pestle, sieved to -10 mesh (-2 mm), then returned to the furnace for final drying at 375-400°C for 2-1/2 hours. Analytical samples were taken and analyzed for archive purposes after the initial and secondary drying of the doped mixtures.
- The doped glass powder was melted in ~200 g batches in a Pt crucible at 1175°C for 10 minutes, stirred, melted for 5 minutes longer, and cast (quenched) onto a stainless steel plate at ambient glove box temperature (~30°C). When all material had been processed, the quenched glass batch was crushed, ground in an Al_2O_3 mortar and pestle and sieved to -10 mesh. The resulting glass was then blended by hand mixing in a single container.
- Glass powder was melted in ~200 g batches in a single Pt crucible heated to 1175°C and held for 15 minutes in a Deltec DT-31-S furnace in a nitrogen atmosphere glove box. The melt was visually checked for quality after the 15-minute hold period to verify that no unmelted cap or bubbles existed. If acceptable, the melt was poured into a stainless steel bar mold (1.9 x 1.9 x 10 cm) that had been preheated to ~350°C. If the visual check indicated an unacceptable melt, the crucible contents were mixed by swirling and returned to the furnace for an additional ~10-minute hold period. A single bar was cast from each crucible melt and when cool enough to handle without slumping, the bar was transferred to the annealing furnace at 525°C. Elapsed time from the pour to transfer into the annealing furnace was about 1.5 minutes.
- All cast glass bars were annealed at 525°C in a Hoskins FD-104 furnace from 2 to 5 hours, followed by a furnace cool (~175°C/h) to 25°C. All temperatures for melting and annealing were measured with a Fluke Model 2190A Digital Thermometer.

3.2.1 Preparation of Special Forms

Six special forms were requested for delivery to NNWSI (Table 3.3). The production of these special forms from the glass fabricated above is described in the following sections.

3.2.1.1 Crushed and Sized Powder

Crushed glass was produced from the pieces of glass remaining after removal of core-drilled cylinders from ATM-8 bars 1, 2, 3 and 4, . Bar pieces and coring remains were coarse crushed using a hardened steel punch and die and then ground to size using an automatic Al_2O_3 mortar and pestle. The -40 to +80 (U.S. sieve designation) mesh fraction was collected and packaged for shipment. Sizing was performed by manually working the crushed material through the stainless steel sieves with brushing and tapping actions.

3.2.1.2 Uncased Discs and Cylinders

Cylindrical cores were removed from ATM-8 bars 1, 2, 3 and 4 by diamond core drilling with a water cooled Starlite Industries core drill assembly. These cores were then cut to size 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-cm diameter, while cylinders were 1-cm long by 1-cm diameter. Eighty discs and 10 cylinders were produced.

TABLE 3.3. Special Form Requirements Requested by NNWSI
for MCC ATM-8 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

3.2.1.3 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 for the bars described in Section 3.2. 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, ASMF-6A-213]. 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 525°C. When casting was complete the tubes were placed into the annealing furnace and held at 525°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-three discs and 13 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 cylinder 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-8 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 055090." 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.

- Previously melted glass bars (ATM-8 bars 5, 6, 7, 8 and 9) were remelted to form the feed material for the present castings. Remelting was done in a Pt crucible in an nitrogen-atmosphere glove box at 1150-1200°C, a temperature sufficient to allow adequate casting behavior. Residence time at temperature was ~30 minutes.

- 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 room temperature.
- 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 cut to a length of 2.0 ± 0.1 cm with parallel cut ends normal to the cylinder axis. Cutting to dimensions 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, and a dimensional shift of the chuck and sample was made to position the sample for the second cut.

3.3 MEASUREMENT AND TESTING EQUIPMENT

Fluke Digital Thermometer, Model No. 2190A, Calib. No. 364-79-06-007

Mettler Balance, Model No. 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-S Furnace

Honeywell Digital Control Programmer DCP-7700

Hoskins Furnace, Model FD-104

Love Model 49 Proportional Controller

Fritsch Auto-Mortar and Pestle, Pulverisette II, Al₂O₃

Buehler Isomet Cut-off Saw
Starlite Industries Core Drill
Corning Model PC-354 Hot Plate
Glove Box, N₂ Atmosphere

3.5 PROCEDURES

MCC-SP-1 (Draft): Glass Component Preparation
MCC-SP-2 (Draft): Glass Preparation
RWP-308-1 Rev. 14: Lab Specific, General Radiation Work Procedure
CSS-308-1: Lab Specific, Criticality Safety Specification

3.6 IDENTIFICATION SYSTEM

Each final glass bar was uniquely numbered and labelled according to the order in which it was poured, as follows: ATM-8-1, ATM-8-2, etc. Production records for this glass are recorded on page 48 of PNL Laboratory Record Book BNW-3660, and include data on date of manufacture, bar ID number, weight, and current disposition. Table 3.4 gives the production summary for ATM-8 glass.

3.7 MATERIAL AVAILABILITY AND STORAGE

ATM-8 glass has been provided to NNWSI in the form of bars, crushed powder and special castings. As of August 1985, ~550 g of ATM-8 glass in bar form are available for distribution. Archive samples were taken from all initial bars. All remaining MCC-ATM-8 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.

TABLE 3.4. Product Summary for ATM-8 Glass^(a)

<u>MCC ID No.</u>	<u>Bar Weight, g</u>	<u>Pour Date</u>
ATM-8-1	102.85	4/2/84
ATM-8-2	104.81	4/2/84
ATM-8-3	101.42	4/2/84
ATM-8-4	100.90	4/2/84
ATM-8-5	107.38	4/2/84
ATM-8-6	94.00	4/2/84
ATM-8-7	108.25	4/2/84
ATM-8-8	59.21	4/2/84
ATM-8-9	108.11	4/4/84
ATM-8-10	114.52	4/4/84
ATM-8-11	97.31	4/4/84
ATM-8-12	102.85	4/4/84
ATM-8-13	112.92	4/4/84
ATM-8-14	108.53	4/4/84
ATM-8-15	106.51	4/4/84
ATM-8-16	110.25	4/4/84
ATM-8-17	103.69	4/13/84
ATM-8-18	109.83	4/13/84
ATM-8-19	113.07	4/13/84
ATM-8-20	119.47	4/13/84
ATM-8-21	102.76	4/13/84
ATM-8-22	111.34	4/13/84
ATM-8-23	103.43	4/13/84
ATM-8-24	100.08	4/13/84
ATM-8-25	27.00	4/13/84

(a) Nominal bar size 1.9 x 1.9 x 10 cm; bars No. 8 and 25 are significantly shorter.

4.0 CHARACTERIZATION

Characterization of ATM-8 glass was conducted by bulk density measurement, chemical analysis, light optical and scanning electron microscopy (SEM), X-ray diffraction (XRD) analysis and radiochemical analysis. Individual bars chosen for chemical analysis provided data from each production day as well as a set of samples on a single bar that allowed estimation of axial homogeneity. A summary of these analyses is presented in the following sections according to analytical procedure.

4.1 BULK DENSITY MEASUREMENTS

Bulk density was determined on four samples, two each taken from bars number ATM-8-20 and ATM-8-22. Density was measured using ASTM Procedure C 693-74, "Standard Method for Density of Glass by Buoyancy." The technique was modified by using ~3-g samples instead of the 20-g samples specified. Density samples were prepared by removing a section from the end and the middle of the two bars listed, and trimming off all cast surfaces using a Buehler Isomet saw with a low concentration diamond wafering blade and water as the cutting media. Before measurement, all samples were cleaned in alcohol in an ultrasonic cleaner and dried at ambient glove box temperature (~25°C). The average density for this glass was determined to be $3.0493 \pm 0.0091 \text{ g/cm}^3$ (Table 4.1).

TABLE 4.1. Measured Bulk Density of ATM-8 Glass

<u>Glass Bar No.</u>	<u>Specimen No.</u>	<u>Measured Density, g/cm³</u>
20	ED	3.0547
20	MD	3.0541
22	ED	3.0526
22	MD	<u>3.0357</u>
		Mean value: 3.0493 (std. dev. = 0.0091)

4.2 CHEMICAL ANALYSIS

Chemical analysis for most of the nonradioactive elements was performed by inductively coupled argon plasma atomic emission spectroscopy (ICP). Separate preparations of the same samples used for ICP were made for radiochemical analysis of the radioactive constituents. Uranium was analyzed for by laser fluorometry, technetium by beta counting analysis of an isotope-specific separation, and neptunium and plutonium were analyzed for by alpha energy analysis (counting) techniques. Additionally, radiochemical and plutonium isotope analyses was performed on the initial dopant solution (see Tables A.1 and A.2 in Appendix A). ICP analyses were performed on a B&L ARL-34000 unit, uranium was analyzed 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 one end of bars ATM-8-7 and ATM-8-13, and from the center and end of bar ATM-8-20. The solid samples were 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 this solution from each of the samples were submitted for both ICP and radiochemical analysis.

The results of the chemical analysis of the four samples from ATM-8 glass are given in Table 4.2. Radiochemistry data for the analyses of ATM-8 specimens are given in Table 4.3.

The satisfactory precision shown for most of the individual element analyses indicates that all 3 glass bars are homogeneous, but the summation of concentrations is significantly less than 100%, even considering probable concentrations of elements not analyzed. Therefore, the accuracy of individual values is uncertain, and simple normalization of all data equally to achieve 100% is not justified. Additional analyses by independent laboratories and methods are in progress.

In comparing the composition of ATM-8 with the composition requested (Table 3.2), the user should recognize that the use of MCC-76-68 feedstock actually determined the product composition except for the specific additions made. For that reason, the content of Al_2O_3 and La_2O_3 (originally present in MCC-76-68) is significantly higher than requested. Similarly, since only a low

TABLE 4.2. Analyzed Composition of ATM-8 Glass^(a)

Oxide	wt%	Standard Deviation
<u>Stable Constituents</u>		
Ag ₂ O	NA	--
Al ₂ O ₃	1.06	0.05
B ₂ O ₃	8.18	0.28 ^(b)
BaO	0.61	0.02
CaO	2.13	0.06 ^(b)
CdO	<DL, 0.005	--
CeO ₂	0.11	0.03
Cr ₂ O ₃	0.48	0.02
Cs ₂ O	NA	--
Dy ₂ O ₃	0.01	0.00
Eu ₂ O ₃	<DL, 0.01	--
Fe ₂ O ₃	8.18	0.24
Gd ₂ O ₃	<DL, 0.05	--
K ₂ O	NA	--
La ₂ O ₃	4.87	0.15
MgO	0.16	0.01
MnO ₂	0.06	0.01
MoO ₃	1.98	0.04
Na ₂ O	12.2	0.2 ^(b)
Nd ₂ O ₃	1.66	0.06
NiO	NA	--
P ₂ O ₅	0.57	0.07 ^(b)
PdO	0.37	0.10 ^(c)
Rh ₂ O ₃	NA	--
RuO ₂	0.58	0.10
SiO ₂	37.8	0.7 ^(b)
Sm ₂ O ₃	<DL, 0.05	--
SrO	0.47	0.01
TeO ₂	<DL, 0.1	--
TiO ₂	2.61	0.10
Y ₂ O ₃	<DL, 0.1	--
ZnO	4.37	0.12
ZrO ₂	1.99	0.05
<u>Radioactive Constituents</u>		
NpO ₂	0.38	0.02 ^(b)
PuO ₂	0.11	0.01 ^(b)
Tc ₂ O ₇	0.18	0.01 ^(b)
UO ₂	4.24	0.07 ^(b)
TOTAL	95.36	

(a) Based on analyses of 16 specimens taken from 9 different bars unless otherwise indicated. Standard deviations are for an individual analysis.

(b) From analyses of 4 specimens taken from 3 different bars.

(c) From analyses of 12 specimens taken from 9 different bars.

NA = Not analyzed.

ND = Analyzed but not detected.

<DL = Below detection limit indicated.

TABLE 4.3. Radiochemistry Data for ATM-8 Glass: Example of Calculation Methodology

Isotope	(1) Mean Radiochemical Analysis	(2) Analytical Dilution Factor, mg Glass/mL	(3) Isotope Activity Factor, d/m/mg Isotope	(4) Element to Oxide Conversion Factor	(5) wt% Oxide	Oxide Form
^{237}Np	6.31×10^3 d/m/mL	1.22	1.566×10^6 0.8810	0.375	NpO ₂	
^{239}Pu	1.56×10^5	1.22	1.377×10^8 (a)	0.8832	0.105	PuO ₂
^{99}Tc	5.23×10^4	1.22	3.771×10^7 0.6385	0.178	Tc ₂ O ₇	
U	45.6 μg U/mL	1.22	(N/A) 0.8815	4.24	UO ₂	

$$\text{Column (5)} = [\text{Col. (1)} / \text{Col. (2)} / \text{Col. (3)} / \text{Col. (4)}] \times 100\%$$

Column (1) = Measured and reported values.
 Column (2) = Laboratory preparation values.
 Columns (3) and (4) = Published values.
 Column (5) = Calculated concentrations.

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

concentration of CeO_2 was present originally and no addition was made, the CeO_2 concentration in ATM-8 is lower than requested.

Of the additions, the concentration of the actinides (NpO_2 , PuO_2 and UO_2) is very close to that requested. PdO is somewhat low, and Rh_2O_3 was not analyzed in ATM-8. The concentrations of Tc_2O_7 and RuO_2 are significantly low, probably as a result of the 2-step doping process used during glass batch preparation. Both Tc and Ru can occur readily as volatile compounds or as precipitates or agglomerates in glass; that condition is aggravated by prolonged heating or presence of degassing nitrate solutions, both of which existed during preparation of ATM-8.

4.3 MICROSCOPIC EXAMINATION

Scanning electron and light microscopy were conducted on a polished section of a transverse wafer removed from the center of bar number ATM-8-10. Figure 4.1 illustrates a typical microstructure of this glass. Four features are evident in this glass system: metallic spheroids, fine-needle structures, rectangular crystals, and voids or porosity. Energy dispersive x-ray spectroscopy analysis on the SEM indicated the metallic spheroids to be Pd, the needle structure to be high in ruthenium and the rectangular crystals to be high in chromium and zinc. Other work on glasses of this general type (Turcott et al. 1980) has shown that some palladium will reduce to the metal state during melting and cool down, and ruthenium may come out of glass solution as the oxide RuO_2 . The high chromium-zinc phase is probably a cubic spinel, another precipitated phase common in this glass. Porosity was estimated from an areal analysis of a low magnification optical photograph to be ~0.2% of the region photographed. Pore size was found to vary from about 5 μm to 450 μm in diameter. The amount of porosity of this glass is greater than expected, probably the result of a low residence time (15 minutes) during batch melting. This short melt time was an attempt to minimize the losses of volatile constituents (e.g., Tc, Ru, Cs) during preparation of the glass.

Autoradiography on this sample showed a generally uniform shading indicating a homogeneous distribution of at least the most active constituent. Figure 4.2 illustrates a typical autoradiograph for this glass. There was some

FIGURE 4.1. Typical Microstructure of ATM-8 Glass (Polished section from middle of Bar No. ATM-8-10; bar at bottom of each photo represents 10 μm)

FIGURE 4.2. Typical Autoradiograph (right) and Corresponding Photomicrograph (left) of ATM-8 Glass (Bar No. ATM-8-10, middle section)

indication of higher radioactivity regions roughly corresponding to void or pore areas. In past work this condition has generally been found to be associated with technetium concentration in bubbles within the glass (Bradley et al. 1979). Technetium volatility remains one of the major difficulties in the fabrication of this type glass, and pore generation/migration may be related to this problem.

4.4 X-RAY DIFFRACTION ANALYSIS

X-ray diffraction analysis was performed on solid wafers cut from the end of bars ATM-8-7 and ATM-8-20, and the middle of bars ATM-8-10 and ATM-8-20. 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 any of the samples examined. Therefore, based on the known detection limits of the XRD system used, the crystalline content of ATM-8 glass is estimated to be less than ~5% by weight.

5.0 REFERENCES

- Bradley, D. J., C. O. Harvey, and R. P. Turcotte. 1979. Leaching of Actinides and Technetium from Simulated High-Level Waste Glasses. PNL-3152, Pacific Northwest Laboratory, Richland, Washington.
- Mellinger, G. B., and J. L. Daniel. 1984. Approved Reference and Testing Materials for Use in Nuclear Waste Management Research and Development Programs. PNL-4955-2, Pacific Northwest Laboratory, Richland, Washington.
- Mendel, J. E. et al. 1977. Annual Report of the Characteristics of High-Level Waste Glasses. BNWL-2252, Pacific Northwest Laboratory, Richland, Washington.
- Turcotte, R. P., J. W. Wald and R. P. May. 1980. "Devitrification of Nuclear Waste Glasses." In Scientific Basis for Nuclear Waste Management. Vol. 2, CJM Northrup, ed., Plenum Publishing Co., New York, pp. 141-146.

APPENDIX A

PREPARATION OF DOPANT SOLUTIONS FOR ATM-8 GLASS

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PREPARATION OF DOPANT SOLUTIONS FOR ATM-8 GLASS

Actinides were added to the glass batch for ATM-8 as liquid (nitrate) solutions to obtain the concentrations given in Table 3.2. Solutions were prepared/obtained as follows:

- Technetium - ^{99}Tc was obtained from ORNL Isotope Sales as NH_4TcO_4 . This material was black because of partial decomposition to TcO_2 . The shipping order received from ORNL with the material indicated that the powder received was 50.5% ^{99}Tc (versus 54% theoretical for NH_4TcO_4). Using the ORNL supplied factor, 18.983 g of solid was weighed out, dissolved in 200 mL of hot water and allowed to cool to 25°C over a 12-hour period. The resultant liquid was decanted from the TcO_2 decomposition product precipitate and set aside for later use. The TcO_2 was then treated with dilute H_2O_2 to dissolve it as HTcO_4 . When the last signs of solid were gone, the solution was heated to destroy the H_2O_2 . This solution and beaker rinses were then added to the decanted liquid along with some additional water to dissolve a small quantity of white NH_4TcO_4 which had crystallized out. The final solution was about 300-350 mL.
- Uranium - Depleted (0.15% ^{235}U) uranium dioxide powder at -325 mesh ($\sim 44\ \mu\text{m}$) was obtained from on-hand stock. 123 g of the oxide powder was weighed out and placed into a 500 mL pyrex beaker and wetted with 75-100 mL of deionized water. Concentrated nitric acid was slowly added to the wetted powder until most of the powder appeared to be in solution (~ 200 -250 mL total solution volume). Nitric acid was added to the oxide powder inside a chemical fume hood as large volumes of NO_2 were given off and a significant amount of self heating was observed. Although no external heating was used, the solution was placed on a hot plate/stirrer and stirred for approximately 1 hour at a low stirring rate to dissolve any remaining flecks of oxide. When

cool, the solution was transferred to a 250-mL volumetric flask and adjusted to volume using deionized water.

- Neptunium - The ^{237}Np solution was prepared from on-hand supplies of NpO_2 powder. A 12.002 g aliquot of the oxide was dissolved in 50 mL of 15.7 M HNO_3 - 0.05 M HF which resulted in the evolution of copious quantities of NO_2 fumes. The Np at this point had converted to brown Np(VI) . The solution was evaporated to 20 mL, transferred to a storage bottle, and diluted up to 50 mL using water washes of the preparation beaker. The final acidity was estimated at <5 M.
- Plutonium - The ^{239}Pu was obtained directly from on-hand stock solution of weapons grade Pu (92.5% ^{239}Pu). The stock solution was last purified by anion exchange in nitric acid in May 1982. Since it was old Pu, the ingrowth of ^{241}Am would be expected to be less than with newer material of this isotopic composition. Ninety-one mL of solution was withdrawn from the stock solution of 29.1 gPu/L.

Before mixing the solutions, analytical aliquots ranging in size from 0.25 to 1 mL (depending upon initial dopant solution volume) were removed for concentration and radioisotope impurity analyses. These results are presented in Table A.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 dilution of each aliquot to a larger volume (i.e., 0.25 mL of dopant added to 10 mL of H_2O gives a dilution factor 10.25/0.25 = 41). Mass spectrometry isotopic analysis of plutonium in solution ATM-8SP is given in Table A.2.

TABLE A.1. Radiochemical Analysis of Actinide Dopant Solutions for ATM-8 Glass

Solution No.	Isotope	Radiochemical Analysis	Activity Factor, d/m/mg	Dilution Factor	Isotopic Solution Concentration, $\mu\text{g/mL}$
ATM-8ST	^{99}Tc	1.41×10^8 d/ m/mL	3.771×10^7	11	41.1
ATM-8SU	U	38.8 g/L	--	11	426.8
ATM-8SN	^{237}Np	6.72×10^6 d/ m/mL	1.566×10^6	41	175.9
	$^{239+240}\text{Pu}$	3.1×10^4	1.377×10^8 (a)	41	0.0092
	$^{238}\text{Pu} + ^{241}\text{Am}$	6.30×10^5	2.282×10^{10} (b)	41	0.0011
	^{244}Cm	3.9×10^4	1.797×10^{11}	41	8.9×10^{-6}
ATM-8SP	$^{239+240}\text{Pu}$	1.73×10^8	1.642×10^8 (c)	21	22.1
	$^{238}\text{Pu} + ^{241}\text{Am}$	1.36×10^7	2.282×10^{10} (b)	21	0.013

(a) Assumed to be 100% ^{239}Pu .(b) Mean value for $^{238}\text{Pu} + ^{241}\text{Am}$.(c) Weighted average based upon isotopic analysis of 92.48% ^{239}Pu , 7.14% ^{240}Pu .

TABLE A.2. Mass Spectrometry Isotopic Analysis of Plutonium
Solution ATM-8SP (Dopant solution for ATM-8 Glass)

<u>Isotope</u>	
^{238}Pu	$0.024 \pm 0.001 \text{ wt\%}$
^{239}Pu	92.48 ± 0.03
^{240}Pu	7.14 ± 0.03
^{241}Pu	0.311 ± 0.002
^{242}Pu	0.0521 ± 0.0006

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