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MCC

Materials Characterization Center

**Fabrication and Characterization
of MCC Approved Testing
Material—ATM-9 Glass**

J. W. Wald

June 1986

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-9 GLASS

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Pacific Northwest Laboratory
Richland, Washington 99352

PREFACE

This report, prepared by the Materials Characterization Center (MCC), 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 the MCC at the Pacific Northwest Laboratory (PNL). 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, which is issued on an annual basis. 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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SUMMARY

The Materials Characterization Center (MCC) ATM-9 glass is designed to be representative of glass to be produced by the Defense Waste Processing Facility (DWPF) at the Savannah River Plant, Aiken, South Carolina. ATM-9 glass contains all of the major components of the DWPF glass and corresponds to a waste loading of 29 wt%. The feedstock material for this glass was supplied by Savannah River Laboratory (SRL), Aiken, SC, as SRL-165 Black Frit to which was added Ba, Cs, Mo, Nd, Zr, as well as ^{99}Tc , depleted U, ^{237}Np , $^{239+240}\text{Pu}$, and ^{243}Am . The glass was produced under reducing conditions by the addition of 0.7 wt% graphite during the final melting process. The glass was produced from April to May of 1984 at the request of the Office of Nuclear Waste Isolation (ONWI), Columbus, OH (private correspondence R. W. Cote to J. E. Mendel, March 6, 1984) and is the second in a series of glasses produced for ONWI. All of the glass was produced in the form of glass bars.

Three kilograms of ATM-9 glass were produced from a feedstock melted in a nitrogen-atmosphere glove box at 1150°C first in platinum, then Inconel-690, and finally Denver Fire Clay (DFC) crucibles. On final melting, the glass was formed into stress-annealed rectangular bars of two sizes: 1.9 x 1.9 x 10 cm and 1.3 x 1.3 x 10 cm. Seventeen bars of each size were made.

Table 3.2 in Section 3.0, lists the analyzed composition of ATM-9 glass. The nonradioactive elements were analyzed by inductively coupled argon plasma atomic emission spectroscopy (ICP) analysis, uranium was analyzed by pulsed laser fluorometry, technetium by beta counting analysis, and neptunium, plutonium and americium by alpha energy analysis (counting). Examination by optical microscopy of a single transverse section from one bar showed random porosity estimated at 0.36 vol% with nominal pore diameters ranging from ~5 μm to 200 μm . Only one distinct second phase was observed and it was at a low concentration level in the glass matrix. The phase appeared as spherical metallic particles. Although the particles were not specifically analyzed, similar features analyzed in other ATMs were principally Pd metal. X-ray

diffraction (XRD) analysis of this same sample did not show any diffraction peaks from crystalline components, indicating that the glass contained less than 5 wt% of crystalline devitrification products. An autoradiograph of the sample was obtained and used as a guide to the total radionuclide distribution. The even shading on the radiograph exposure indicated a generally uniform distribution of radioactivity throughout the glass matrix, with no distinct high-concentration regions.

ATM-9 glass has been provided to ONWI and to the Basalt Waste Isolation Project (BWIP) in the form of bars and crushed powder. As of October 1985, approximately 1.35 kg of ATM-9 is available for distribution. Requests for materials or services related to this glass should be made to the MCC Program Office, Pacific Northwest Laboratory.

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1.0 INTRODUCTION

One of the functions of the Materials Characterization Center (MCC) is to assist the U.S. Department of Energy (DOE) projects responsible for nuclear waste form production, transportation, and disposal in assembling 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 details of fabrication and characterization of ATM-9 glass. ATM-9 was produced by the MCC at the Pacific Northwest Laboratory (PNL), Richland, Washington, which is operated for the DOE by Battelle Memorial Institute.

ATM-9 glass is designed to be representative of glass which will be produced by the Defense Waste Processing Facility (DWPF) at the Savannah River Plant, Aiken, South Carolina. This glass was produced at the request of ONWI, with the concurrence of the Basalt Waste Isolation Project (BWIP), for use in their repository testing and development programs. ATM-9 simulates glass that contains all of the major nonradioactive components of the DWPF glass and corresponds to a waste loading of 29 wt% (by weight of oxides). The feedstock material for this glass was SRL-165 Black Frit, as provided by SRL, to which five nonradioactive and five radioactive dopants were added. The nonradioactive additives included Ba, Cs, Mo, Nd, and Zr while the radioactive additives included ^{99}Tc , depleted uranium, ^{237}Np , $^{239+240}\text{Pu}$, and ^{243}Am . Graphite was also added to the glass during the melting process to control the oxidation state of the elements in the final glass.

Section 2 of this report provides details of the fabrication of the glass; Section 3 summarizes the analysis and characterization of the final glass product. ATM-9 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. The procedures followed were documented in PNL Laboratory Record Book number BNW-3660 with supporting documentation filed in the PNL Records Center.

2.0 FABRICATION^(a)

The fabrication of ATM-9 glass began on April 5, 1984 and was completed by May 10, 1984. All work involved in the fabrication of this glass took place in Lab 410, 325 Building, 300 Area and Labs 208 and 220, 308 Building, 300 Area.

2.1 STARTING MATERIALS

The glass which served as the feedstock for ATM-9 was SRL-165 Black Frit as provided to the MCC by John Plodinec of the SRL. The material was supplied in a single coarse crushed lot as batch AX581875-G, milling number B5039. This material is a simulated waste glass that contains all the major nonradioactive components of the Defense Waste Processing Facility (DWPF) product, and corresponds to a waste loading of 29 wt% (by weight of oxides). The average analyzed composition of the SRL-165 glass used in ATM-9 is given in Table 2.1. Five nonradioactive elements were added to the SRL-165 glass frit to more closely represent those elements present in the DWPF waste stream above 5×10^{-4} g/g glass. The selection of radioactive dopants was based upon recommendations of the BWIP staff (E. H. Randklev, Cognizant Engineer) to provide optimum conditions for radiochemical analysis of leach-tested specimens. The constituents added to SRL-165 glass frit to form ATM-9 glass are listed in Table 2.2 along with the target compositional levels.

The nonradioactive chemicals used were obtained from on-hand stock of reagent-grade chemicals. 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. ^{99}Tc was obtained as NH_4TcO_4 from Oak Ridge National Laboratory (ORNL) Isotope Sales as ORNL batch number P-30 on isotope order number 37-1174. The source of the depleted uranium was from on-hand stock of UO_2 powder (0.15% ^{235}U) while the neptunium was obtained from ORNL Isotope Sales as $^{237}\text{NpO}_2$ powder in 1977 as batch number 112HB-11 on isotope order number 30-0174-C. The plutonium was obtained from in-house stock solution of weapons-grade plutonium (92.5% ^{239}Pu) last purified by anion

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

TABLE 2.1. Analyzed Composition of SRL-165 Black Frit
(Batch AX581875-G, Milling No. B5039)
Feedstock for ATM-9 Glass

Oxide (a)	Concentration, wt% (b)
Al ₂ O ₃	4.44
B ₂ O ₃	7.00
BaO	0.04
CaO	1.66
Fe ₂ O ₃	11.97
K ₂ O	0.20
Li ₂ O	4.95
MgO	0.67
MnO ₂	2.76
Na ₂ O	11.03
NiO	0.82
P ₂ O ₅	0.37
SiO ₂	53.6
TiO ₂	0.20
ZnO	0.05
ZrO ₂	0.71
TOTAL	100.47

(a) All oxides represent total concentrations of the elements.

(b) Average of separate ICP analyses conducted on March 5, 1985 and August 5, 1985.

TABLE 2.2. Nonradioactive Constituents and Radioactive Dopants for ATM-9 Glass (Matrix: SRL-165 Black Frit, Batch AX581875-G, Milling No. B5039)

<u>Nonradioactive</u>	<u>Target Concentration, g/g glass</u>
Zr	9.4E-4
Mo	5.4E-4
Cs	6.5E-4
Ba	7.0E-4
Nd ^(a)	18.7E-4
<u>Radioactive</u>	
U - Depleted	1.62E-2
²³⁹⁺²⁴⁰ Pu ^(b)	2.2E-5
²⁴³ Am	5.7E-6
²³⁷ Np	1.0E-4
⁹⁹ Tc	6.7E-5

(a) Nd is substituted, on a molar basis, for Ce, Pr, Pm, Sm, and Eu.

(b) Weapons-grade plutonium, nominally 92.5% ²³⁹Pu, 7.5% ²⁴⁰Pu.

exchange in May 1982. The americium was obtained several years ago from ORNL Isotope Sales as ²⁴³AmO₂ powder. No procurement or receiving information was available on this material. Details of the dopant solution preparation from these materials are given in Appendix B.

2.2 BATCH AND GLASS PREPARATION

The targeted composition for ATM-9 glass, defined by the MCC Program office, is listed in Table 2.3. The final glass was melted using a reductant (graphite) specified at a concentration level (0.7 wt%) accepted at the time by SRL. The SRL staff had indicated that the average ferrous-ferric ratio in

TABLE 2.3. Targeted Oxide Composition for ATM-9 Glass

<u>Stable Oxides(a)</u>	<u>Amount, wt%</u>
Al ₂ O ₃	4.29
B ₂ O ₃	6.86
BaO	0.11
CaO	1.60
Cs ₂ O	0.07
Fe ₂ O ₃	11.76
K ₂ O	0.20
Li ₂ O	4.85
MgO	0.56
MnO ₂	2.80
MoO ₃	0.08
Na ₂ O	10.78
Nd ₂ O ₃	0.22
NiO	0.77
P ₂ O ₅	0.29
SiO ₂	51.70
TiO ₂	0.20
ZnO	0.03
ZrO ₂	0.87
<u>Radioactive Oxides(a)</u>	
U ₃ O ₈	1.91
PuO ₂	2.5E-3
Am ₂ O ₃	6.3E-4
NpO ₂	1.1E-2
Tc ₂ O ₇	1.0E-2
TOTAL	<u>99.97</u>

(a) All oxides represent total concentrations of the element

their glass was 0.25 ± 0.1 under similar processing conditions. The procedures used for initial batch preparation and final glass fabrication were as follows:

- A ~3 kg charge of coarse ground SRL-165 Black Frit was blended with 2.43 g of MoO_3 powder in an alumina ball mill until the frit batch passed through a 100 mesh U.S. Standard sieve ($\sim 150 \mu\text{m}$).
- After size reduction and blending by milling, the frit batch containing the Mo additive weighed 2936.7 g. The dopant solutions, as described in Appendix B, along with container rinses were combined and thoroughly mixed in a 3.5-L polyethylene beaker. The frit batch was then added to the dopant liquid and mixed to a heavy slurry. This slurry was thoroughly stirred with a stainless steel stirring rod.
- Approximately half of the total slurry was transferred to a stainless steel beaker for drying and calcining. The material was dried in the beaker with frequent stirring and was then calcined at 500°C for 90 minutes using a Hoskins furnace. After removing this batch from the stainless steel beaker, the remaining slurry batch was dried and calcined in the stainless steel beaker in the same manner.
- After drying and calcining were complete, the two batches were combined into a single batch and mixed by stirring.
- The dried and calcined batch was melted at 1150°C in a ~250-mL platinum crucible in a Deltec furnace located in a nitrogen atmosphere glove box. Soak time at 1150°C was 30 minutes. (Nitrogen is the normal operating atmosphere of the glovebox; the nitrogen atmosphere was not necessarily selected for any ability it might have to control elemental oxidation state in the glass during melting.) Processing of the total batch required approximately ten separate melts. When the appropriate soak time had been completed, each charge of molten glass was poured (quenched) onto a stainless steel plate at glove box temperature ($\sim 30\text{--}35^\circ\text{C}$).

- When the first melt cycle was completed, the quenched glass batch was coarse crushed in a hardened D-2 tool steel punch and die and fine ground in an automatic alumina mortar and pestle to pass a 100 mesh U.S. Standard sieve ($\sim 150\ \mu\text{m}$). The batch mass was recorded at 2827.6 g. Technical grade graphite powder (see Table A.1, Appendix A) was then added to this glass powder batch at the 0.7 wt% level (i.e., 19.79 g).
- The graphite blending process was accomplished in three steps. Approximately 200 g of glass powder was blended with 19.79 g of graphite and sieved to pass an 80 mesh U.S. Standard sieve ($\sim 180\ \mu\text{m}$). Another charge of ~ 1500 g of glass powder was then blended with this batch and the entire mixture then sieved to pass a 40 mesh U.S. Standard sieve ($\sim 425\ \mu\text{m}$). The final step involved blending the remaining glass powder with this batch by hand stirring in a single container.
- The graphite blended glass batch was melted at 1150°C in a shop-made Inconel 690 crucible in a Deltec furnace located in a nitrogen atmosphere glove box using a soak time of 30 minutes. Processing of the total batch required four separate charges of ~ 750 g each. When the appropriate soak time had been completed, each charge of molten glass was poured (quenched) onto a stainless steel plate at glove box temperature ($\sim 30\text{--}35^\circ\text{C}$). The Inconel 690 crucible was chosen for use during this melting cycle to duplicate processing cycles reported by SRL.
- This quenched glass batch was then coarse crushed in a hardened D-2 tool steel punch and die and ground in an automatic alumina mortar and pestle to pass a 10 mesh U.S. Standard sieve ($\sim 2\ \text{mm}$).
- A third melting was conducted because scale and gas bubbles were apparent in the bars poured from the second melting. A likely cause of this inhomogeneity appeared to be degradation of the Inconel 690 crucible, and so, the crucible type was changed to Denver Fire Clay (DFC). A ~ 350 mL DFC crucible, which would conveniently hold about a third of the batch at one time, was selected. The crucible of glass

was placed in a Deltec furnace that is located inside a nitrogen atmosphere glove box. The first crucible load of glass was heated to 1150°C and allowed to soak at that temperature for 45 minutes. Acceptable bars were poured from this melting but the annealing temperature (510°C) caused the product to lose shape. These bars (~900 g of glass) were crushed to pass a 10 mesh U.S. Standard sieve for remelting and casting. The bars produced from this fourth melt cycle were given a new series of identification numbers. The remaining glass batch was processed through a third melt cycle as described above and acceptable bars were produced.

- Cast bars were formed in stainless-steel bar molds in two different sizes: 1.9 x 1.9 x 10 cm and 1.3 x 1.3 x 10 cm. The bar mold was initially preheated to about 350°C on a hot plate, and a single bar cast at a time. When cool enough to handle without slumping, the bar was transferred to the annealing furnace at 450°C. Elapsed time from pour to annealing furnace was about 1.5 to 2 minutes.
- All final product bars were annealed at 450°C in a Hoskins type FD-104 furnace located in a nitrogen atmosphere glove box for a minimum of 2 h (maximum 5 h), followed by a furnace cool (~175°C/h) to 25°C.

2.3 MEASUREMENT AND TESTING EQUIPMENT

Fluke Digital Thermometer, Model No. 2190A, Calib. No. 364-78-06-007

Mettler Balance, B5, Calib. No. 362-06-03-014

Mettler Balance, H-15, Calib. No. 362-06-03-053

Mettler Balance, PC-4400, Calib. No. 368-06-01-008

2.4 OTHER EQUIPMENT

Deltec-DT-31-S Furnace

Hoskins Furnace, Model FD-104

Honeywell Digital Control Programmer, DCP-7700

Love Model 49 Proportional Controller

Fritsch Auto-Mortar and Pestle, Pulverisette II, Al₂O₃

Corning Model PC-354 Hot Plate
Glove Box

2.5 PROCEDURES

MCC-SP-1 (Draft) Glass Compound Preparation
MCC-SP-2 (Draft) Glass Preparation
RWS-325-10 Radiation Work Specification
RCP-325-10 Radiation Control Protocol
RWP-308-1 Radiation Work Procedure
CSS-308-1 Criticality Safety Specification

2.6 IDENTIFICATION SYSTEM

Each final glass bar was uniquely numbered and labeled according to the order in which it was poured, as follows: ATM-9-1, ATM-9-2, etc. Production records for the glass are recorded on page 59 of PNL Laboratory Record Book BNW-3660 and include data on date of fabrication, melt run number, bar identification number, mass, and current disposition. Table 2.4 gives the production summary for ATM-9 glass.

2.7 MATERIAL AVAILABILITY AND STORAGE

ATM-9 glass has been provided to ONWI and BWIP in the form of bars and crushed powder. As of October 1985, ~1.35 kg of ATM-9 glass in bar form is available for distribution. Archive samples were taken from all initial bars distributed. All remaining ATM-9 materials and archive samples are in the custody of the MCC Program Material Custodian and are stored in the 308 Building (PNL) MCC storage vault.

TABLE 2.4. Product Summary for ATM-9 Glass(a)

<u>MCC ID No.</u>	<u>Bar Mass, g</u>	<u>Pour Date</u>	<u>Melt Cycle</u>
ATM-9-1	91.3	5/8/84	3
ATM-9-2	93.8	5/8/84	3
ATM-9-3	97.9	5/8/84	3
ATM-9-4	94.5	5/8/84	4
ATM-9-5	98.3	5/8/84	4
ATM-9-6	98.8	5/8/84	4
ATM-9-7	94.5	5/8/84	4
ATM-9-8	45.4	5/8/84	4
ATM-9-9	39.8	5/8/84	4
ATM-9-10	42.9	5/9/84	4
ATM-9-11	50.2	5/9/84	4
ATM-9-12	51.9	5/9/84	4
ATM-9-13	51.7	5/9/84	4
ATM-9-14	102.2	5/9/84	4
ATM-9-15	99.8	5/9/84	4
ATM-9-16	103.7	5/9/84	4
ATM-9-17	79.4	5/9/84	4
ATM-9-18	101.6	5/9/84	3
ATM-9-19	108.3	5/9/84	3
ATM-9-20	105.3	5/9/84	3
ATM-9-21	105.7	5/9/84	3
ATM-9-22	101.7	5/9/84	3
ATM-9-23	103.8	5/9/84	3
ATM-9-24	48.3	5/10/84	3
ATM-9-25	48.3	5/10/84	3
ATM-9-26	49.8	5/10/84	3
ATM-9-27	38.5	5/10/84	3
ATM-9-28	46.9	5/10/84	3
ATM-9-29	46.4	5/10/84	3
ATM-9-30	45.0	5/10/84	3
ATM-9-31	46.3	5/10/84	3
ATM-9-32	46.7	5/10/84	3
ATM-9-33	46.2	5/10/84	3
ATM-9-34	54.2	5/10/84	3

(a) Nominal bar size 1.3 x 1.3 x 10 cm, mass range 38-54 g;
1.9 x 1.9 x 10 cm, mass range 79-108 g.

3.0 CHARACTERIZATION

Characterization of ATM-9 glass was conducted by bulk density measurement, chemical analysis, optical microscopy, scanning electron microscopy (SEM), x-ray diffraction analysis, and radiochemical analysis. The characterization work was conducted between November 1984 and March 1985. Individual bars representing each day of production were chosen for analysis. In addition, a set of samples were taken from a single bar to allow estimation of axial chemical homogeneity. The types of chemical and physical measurements made on ATM-9 for characterization purposes cover the most important areas for analytical data. The extent of these analyses is admittedly insufficient to establish a statistically complete database for the entire 34 bars of ATM-9. The characterization data presented in most areas is intended as a guide in the use of the ATM. All raw data and compilations from characterization are stored in MCC Fabrication Task files and the PNL Records Center. The results of these analyses are summarized in the following sections.

3.1 BULK DENSITY MEASUREMENT

Apparent density was determined on four samples, one each from end cuts of bars ATM-9-3, ATM-9-13, ATM-9-26, and ATM-9-34. Density was measured using ASTM procedure C 693-74, "Standard Method for Density of Glass by Buoyancy," modified to use a nominal 2.5-g sample instead of the 20-g sample specified. Density samples were prepared from glass bars by removing a section from the end region of the bar 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 2.725 g/cm³ (Table 3.1).

3.2 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 concentrations of radioactive constituents. Uranium was analyzed by laser fluorometry; technetium by beta-counting analysis

TABLE 3.1. Measured Bulk Density of ATM-9 Glass

<u>Glass Bar No.</u>	<u>Specimen ID</u>	<u>Measured Density, g/cm³</u>
3	ED	2.729
13	ED	2.717
26	ED	2.726
34	ED	<u>2.729</u>
Mean Value =		2.725
(std. dev. =		±0.006)

of an isotope-specific separation; and neptunium, plutonium and americium by alpha energy analysis (counting) techniques. A plutonium isotopic analysis was also performed on the initial dopant solution. Uranium analysis was conducted on a Scintrex UA-3 Uranium Analyzer while alpha/beta counting were performed on counting systems based around Ludlum/Tracor Northern components.

Analytical samples were taken from one end of bars number ATM-9-1 and ATM-9-34 and both ends of bar ATM-9-13. Each of the solid samples was ground to pass a 100 mesh U.S. Standard sieve (-150 μ m), 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.

Results of the chemical analysis of the four samples of ATM-9 glass are given in Table 3.2. Details of the development of the radiochemical data are given in Table 3.3.

With the exception of MoO₃, Nd₂O₃, P₂O₅, TiO₂, ZnO, and PuO₂, the average standard deviation in measured oxide concentrations for the four samples evaluated is ±4%. Concentration measurement variations for MoO₃ (±30%), Nd₂O₃ (±144%), TiO₂ (±15%), ZnO (±13%), and PuO₂ (±50%) imply there is a potential for inhomogeneity of these constituents. Only one measurement was obtained for P₂O₅ in the four samples evaluated, which indicates that this constituent could also be considered inhomogeneously distributed. All constituents, with the exception of P₂O₅, were at nominal concentrations well within the ideal

TABLE 3.2. Analyzed Composition of ATM-9 Glass (a)

Stable Oxides (b)	Concentration, wt%	Std. Dev. wt%
Al ₂ O ₃	4.94	0.13
B ₂ O ₃	7.15	0.23
BaO	0.12	0.01
CaO	1.62	0.03
Cs ₂ O	NA	--
Fe ₂ O ₃	11.18	0.56
K ₂ O	NA	--
Li ₂ O	4.81	0.31
MgO	0.77	0.04
MnO ₂	2.61	0.14
MoO ₃	0.09	0.03
Na ₂ O	11.02	0.40
Nd ₂ O ₃	0.34	0.50
NiO	NA	--
P ₂ O ₅	1.57	(c)
SiO ₂	50.43	2.42
TiO ₂	0.24	0.04
ZnO	0.08	0.01
ZrO ₂	0.85	0.02
Radioactive Oxides (b)		
Am ₂ O ₃	5.64E-4	3.61E-6
NpO ₂	1.29E-2	4.50E-4
PuO ₂	5.74E-3	2.85E-3
Tc ₂ O ₇	4.12E-3	2.28E-4
UO ₂	1.96	6.98E-2
TOTAL	99.80	

(a) Average of analysis of four specimens taken from three bars. Standard deviations are for an individual analysis.

(b) All oxides represent total concentrations of the element.

(c) Single analysis, Sample ATM-9-13EB.

NA = Not analyzed at this time.

TABLE 3.3. Radiochemistry Data for ATM-9 Glass: Example of Calculation Methodology

Isotope	(1) Mean Radiochemical Analysis, dis/min/g Glass	(2) Isotope Activity Factor, dis/min/mg Isotope	(3) Element to Oxide Conversion Factor	(4) wt% Oxide	Oxide Form
^{243}Am	2.28×10^6	4.435×10^8	0.9101	5.64×10^{-4}	Am_2O_3
^{237}Np	1.78×10^5	1.556×10^6	0.8811	1.29×10^{-2}	NpO_2
$^{239+240}\text{Pu}$	8.30×10^6	$1.633 \times 10^8(\text{a})$	0.8820	5.74×10^{-3}	PuO_2
^{99}Tc	9.88×10^5	3.771×10^7	0.6385	4.12×10^{-3}	Tc_2O_7
U	1.73×10^{-2} g/g	(NA)	0.8815	1.96	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) = Calculated standard values.

Column (4) = Calculated concentrations.

(a) Weighted average based upon isotopic analysis of 92.51% ^{239}Pu , 7.11% ^{240}Pu .

detection limits of the analytical instrument, which would tend to eliminate measurement variations due to low concentration as a problem. Nominal P_2O_5 concentration was 33% of the ideal detection limit, indicating an analytical problem is possible here. The summation of concentration values is 99.80%, which is considered satisfactory for analytical results, given the complexity of the sample and limitations of the analytical methods.

Compared to the target concentration values given in Table 3.3, MgO (+37%), Nd_2O_3 (+56%), TiO_2 (+20%), ZnO (+150%), PuO_2 (+130%), and Tc_2O_7 (-59%) all deviated significantly from the desired (targeted) levels. Additionally, Al_2O_3 , MoO_3 , Am_2O_3 , and NpO_2 were found to deviate $\pm 15\%$ from the target values. No analyses were obtained for Cs_2O , K_2O , or NiO due to analytical limitations, while the reliability of the single analysis for P_2O_5 remains suspect. All other constituents were within $\pm 5\%$ of the target concentration values. Of the six constituents that showed significant deviation, MoO_3 , Nd_2O_3 , PuO_2 , and Tc_2O_7 were dopant additive constituents. Apparently, too much Mo , Nd , and Pu were added during formulation, while the discrepancy in Tc concentration is due to the inability to accurately predict and adjust for volatility losses during melting. Since ZnO and MgO were not adjusted in the initial glass feedstock, the higher-than-target concentration levels of these constituents can be traced to slightly higher concentrations in the feedstock (see Table 3.1).

3.3 MICROSCOPIC EXAMINATION

Microscopy examination was performed on a single transverse wafer removed from the end of bar number ATM-9-13. Figure 3.1 illustrates the typical microstructure of this glass as seen on the polished cross section examined in the SEM. No major crystalline-appearing features were observed in this sample of the glass matrix with the exception of a few randomly dispersed metallic spheroids. Although not specifically analyzed in this glass, similar features analyzed in other ATMs were found to be composed principally of Pd . Total porosity was measured in this sample by quantitative image analysis at 0.36 vol.%, with the pore size averaging between 5 μm and 200 μm .

FIGURE 3.1. Typical Microstructure of ATM-9 Glass
(SEM image of polished section from bar
ATM-9-13; bar in lower right represents
10 μm)

Autoradiography on this glass sample showed a generally even shading, indicating a uniform distribution of at least the most radioactive constituent. The photographic film technique used to obtain the autoradiograph is not isotope specific and therefore cannot provide individual distributions but only gross indications of high concentration regions or generally uniform distribution. Figure 3.2 illustrates a typical autoradiograph and corresponding photomicrograph for this glass.

3.4 X-RAY DIFFRACTION ANALYSIS

X-ray diffraction (XRD) analysis was performed on a solid wafer cut from bar ATM-9-13 (the same sample as used for microscopic examination). Analysis was performed on a GE/Diano XRD-5 x-ray diffraction system located inside a nitrogen-atmosphere glove box using Cu K α radiation and a continuous scan mode ($1^\circ/2\theta/\text{min}$). No diffraction peaks from crystalline components were observed in the sample examined. Based upon the known characteristics of the XRD system used, the mass of any crystalline phases in the glass matrix is estimated to be less than 5 wt%.

FIGURE 3.2. Typical Autoradiograph (Right) and Corresponding Photomicrograph (Left) of ATM-9 Glass (Bar ATM-9-13)

3.5 OXIDATION STATE ANALYSIS

ATM-9 glass was fabricated using a specific technique prescribed by SRL to control the final oxidation state of elements in the glass. The technique involved the introduction of graphite into the glass before the final melting cycle. The evaluation of the oxidation state of iron in the glass was used as a guide in establishing the oxidation state control point of all affected elements in the glass. The procedure used for this evaluation involved the spectrophotometric determination of reduced and total iron in a fused glass sample using 1,10-Phenanthroline. The technique was developed for specific application to actinide-containing glasses based upon a procedure reported by Jones et al. Results of this analysis permit a determination of the ferrous to ferric ratio. The measured data and calculated ratios are given in Table 3.4 for glass samples from one end of bars ATM-9-1 and ATM-9-34 and both ends of bar ATM-9-13.

As stated previously, the main intent in the fabrication of ATM-9 glass was to produce an ATM representative of the anticipated Savannah River product, although it was not intended to duplicate any specific SRL material. The glass was therefore fabricated using the reductant (graphite) and the concentration level (0.7 wt%) accepted by SRL at the time. SRL staff had indicated that the average ferrous-ferric ratio expected under these conditions would be 0.25 ± 0.1 . The value obtained in the fabrication of ATM-9 glass, 0.31 ± 0.06 , is within the guidelines provided.

TABLE 3.4. Evaluation of Oxidation State in ATM-9 Glass
(ferrous/ferric ratio)

Sample	wt% Fe ²⁺	wt% Fe Total	wt% Fe ³⁺	Fe ²⁺ / Fe ³⁺
ATM-9-1	1.82	7.62	5.80	0.31
ATM-9-13A	1.74	7.86	6.12	0.28
ATM-9-13B	1.58	7.98	6.40	0.25
ATM-9-34	2.12	7.62	5.50	0.39

Avg. = 0.31
(std. dev. ± 0.06)

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APPENDIX A

SOURCE DOCUMENTATION FOR NONRADIOACTIVE CHEMICALS
USED AS ADDITIVES IN ATM-9 GLASS

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SOURCE DOCUMENTATION FOR NONRADIOACTIVE CHEMICALS USED AS ADDITIVES IN ATM-9 GLASS

TABLE A.1. Sources of Nonradioactive Chemical Compounds Used as Additives in ATM-9 Glass

<u>Compound</u>	<u>Manufacturer</u>	<u>Lot No.</u>
CsNO ₃	Noah Chemical	118202
Ba(NO ₃) ₂	MCB	A12H15/478
Nd(NO ₃) ₃ •6H ₂ O	Research Chemical	(n/a)
Zr(NO ₃) ₄ •5H ₂ O	Varlacoid Chemical	G6H-051-55085
MoO ₃	Research Chemical	(n/a)
C(a)	VWR	RC2410-1

(a) Graphite was blended into the glass for oxidation state control during glass fabrication.

n/a = none available

TABLE A.2. Composition of Nonradioactive Chemicals Used as Additives to SRL-165 Black Frit for ATM-9 Glass

<u>Compound Used</u>	<u>Metal/Compound Factor</u>	<u>Mass (g) of Compound Added to 3000 g glass</u>	<u>Concentration Added, Element (g/g glass)</u>
Ba(NO ₃) ₂	5.26E-1	4.001	7.02E-4
CsNO ₃	6.82E-1	2.861	6.50E-4
MoO ₃	6.67E-1	2.43	5.4E-4
Nd(NO ₃) ₃ •6H ₂ O	3.29E-1	17.052	1.87E-3
Zr(NO ₃) ₄ •5H ₂ O	2.12E-1	8.260	5.84E-4

APPENDIX B

PREPARATION OF DOPANT SOLUTIONS FOR ATM-9 GLASS

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PREPARATION OF DOPANT SOLUTIONS FOR ATM-9 GLASS

Radioactive elements were added to the glass batch for ATM-9 as aqueous nitrate solutions to obtain the concentrations given in Table 2.2. Some nonradioactive elements were also added with the radioactive additives, as oxide powder or aqueous nitrate solutions, to increase the concentration of those elements in the SRL-165 Black Frit to the desired levels. The acid and peroxide used in the solution preparations described were of reagent grade quality. Solutions were prepared or obtained as follows:

- Nonradioactive Additives - As outlined in Table 2.2, five nonradioactive additives to SRL-165 Black Frit were required to produce ATM-9 glass. These elements included Ba, Cs, Mo, Nd, and Zr. The Mo was added as MoO_3 powder by ball milling into the glass powder using an Al_2O_3 ball mill. The Ba, Cs and Nd were obtained as nitrate solids, combined into a single batch and dissolved in the minimum required volume (~ 180 mL) of 0.01 M HNO_3 . Zirconyl nitrate solid was dissolved in 75 mL of 1 M HNO_3 and allowed to stand overnight for observation of solids precipitation. All nonradioactive dopant solutions were then combined into a single container for addition to the glass frit.
- Technetium - 0.3980 g of NH_4TcO_4 (as the source of $^{99}\text{Tc}_2\text{O}_7$) was weighed out and dissolved in 50.00 mL of distilled water containing $\sim 0.5\% \text{ H}_2\text{O}_2$ to produce a clear solution. From this solution, 50.0 μL was removed and added to 10.00 mL $\sim 0.5\% \text{ H}_2\text{O}_2$ in water forming a 10.05 mL analytical aliquot. This analytical aliquot was labeled #41183 Tc.
- Uranium - 55.130 g of UO_2 powder was weighed out and dissolved by adding alternately 30% H_2O_2 , water, and 15.8 M HNO_3 , to avoid reducing HNO_3 to NO and/or NO_2 . By this route, the powder was dissolved as an essentially neutral solution of uranyl nitrate, without excess acid. After heating to destroy all residual peroxide, the solution

was cooled and brought to 200.0 mL with water. An analytical aliquot of 0.400 mL was removed and diluted into 10.00 mL forming a 10.40 mL analytical aliquot. This aliquot was labeled #41184U.

- Neptunium - 0.3420 g of $^{237}\text{NpO}_2$ powder was weighed out and dissolved with 2 mL 0.05 M HF - 15.8 M HNO_3 . This process converted the material to Np(VI), estimated based upon the solution color. The entire solution was then diluted up to 25.00 mL using water. From this solution a 0.500 mL aliquot was removed and diluted into 10.00 mL of HNO_3 forming a 10.50 mL analytical aliquot. This analytical aliquot was labeled #41183 Np.
- Plutonium - The plutonium used as the dopant source was obtained from an on-hand supply of anion exchange purified stock solution, last purified in May 1982. Based upon a 1982 analysis of the solution, which indicated a concentration of 29.1 g Pu/L, 2.30 mL of solution was removed and diluted to 25.0 mL with 0.3 M HNO_3 . An analytical aliquot of 0.500 mL was removed from this solution and diluted into 10.00 mL of HNO_3 , forming a 10.50 mL analytical aliquot. This aliquot was labeled #41183 Pu.
- Americium - 0.0204 g of $^{243}\text{AmO}_2$ powder was weighed out and dissolved in 1 mL of 0.05 M HF - 15.8 M HNO_3 and then diluted with water to 25.00 mL. A 50 μL aliquot of this solution was removed and diluted into 10.00 mL of 1 M HNO_3 forming a 10.05 mL analytical aliquot. This analytical aliquot was labeled #41183 Am.

Results of the radiochemical analyses for the radioactive dopant solutions described above are given in Table B.1. The last column in the table is the result of the quotient of Column 3/Column 4 multiplied by Column 5. The dilution factor is the multiplier that corrects the reported data for the analytical dilution as described above. The mass spectrometry isotopic analysis for the plutonium dopant is given in Table B.2.

TABLE B.1. Radiochemical Analysis of Actinide Dopant Solutions for ATM-9 Glass

<u>Solution No.</u>	<u>Isotope</u>	<u>Radiochemical Analysis, dis/min/mL</u>	<u>Activity Factor, dis/min/mg</u>	<u>Dilution Factor</u>	<u>Isotope Solution Concentration, mg/mL</u>
41183 Tc	^{99}Tc	8.85×10^5	3.771×10^7	201	4.72
41183 U	U-0ep.	9.25 (g/L)	(n/a)	26	240.50
41183 Np	^{237}Np	9.54×10^4	1.566×10^6	201	12.25
41183 Pu	$^{239+240}\text{Pu}$	1.97×10^7	$1.633 \times 10^8(a)$	21	2.53
	^{238}Pu	1.41×10^6	3.803×10^{10}	21	7.79×10^{-4}
41183 Am	^{243}Am	1.55×10^6	4.435×10^8	201	0.70
	^{241}Am	4.9×10^4	7.618×10^9	201	1.29×10^{-3}
	^{244}Cm	1.95×10^5	1.797×10^{11}	201	2.18×10^{-4}

(a) Weighted average based upon isotopic analysis of 92.51% ^{239}Pu , 7.11% ^{240}Pu .
n/a = not applicable

TABLE B.2. Mass Spectrometry Isotopic Analysis of Plutonium
Solution 41183 Pu (dopant solution for ATM-9 glass)

<u>Isotope</u>	<u>wt%</u>	<u>Std. Dev., wt%</u>
^{238}Pu	0.025	0.003
^{239}Pu	92.51	0.04
^{240}Pu	7.11	0.04
^{241}Pu	0.299	0.002
^{242}Pu	0.053	0.002

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