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MCC

Materials Characterization Center

**Fabrication and Characterization
of MCC Approved Testing
Material—ATM-11 Glass**

J.W. Wald

J.L. Daniel

August 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-11 GLASS

J. W. Wald
J. L. Daniel

August 1986

Prepared for
the U.S. Department of Energy
under Contract DE-AC06-76RLO 183D

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

ACKNOWLEDGMENT OF CONTRIBUTIONS TO ATM-11

The authors would like to acknowledge and thank those who assisted with the fabrication of the MCC's ATM-11 glass, including Charles A. Church, Jack L. Ryan and Elmer H. Shade. The radionuclide dopant specifications for ATM-11 were established by Edward H. Randklev, BWIP, Rockwell-Hanford. Technical reviews of this report were conducted by Gary D. Maupin and Monty R. Smith for PNL, and Edward H. Randklev and Lance N. Peterson for BWIP, Rockwell-Hanford. The Quality Assurance review of this report was provided by Kyle S. Webster.

SUMMARY

The Materials Characterization Center (MCC) ATM-11 glass is designed to be representative of defense high-level waste glasses that will be produced by the Defense Waste Processing Facility at the Savannah River Plant in Aiken, South Carolina. ATM-11 glass is representative of a 300-year-old nuclear waste glass and was intended as a conservative compromise between 10-year-old waste (i.e., earliest emplacement) and 1000-year-old waste (i.e., as per NRC 10 CFR 60 and the criteria on radionuclide release). The feedstock material for this glass was supplied by Savannah River Laboratory, Aiken, SC, as SRL-165 black frit to which was added Ba, Cs, Mo, Nd, Ni, Pd, Rb, Ru, Sr, Te, Y, and Zr, as well as ^{241}Am , ^{237}Np , $^{239+240}\text{Pu}$, ^{151}Sm , ^{99}Tc , and depleted U. The glass was melted under the reducing conditions that resulted from the addition of 0.7 wt% graphite during the final melting process. Preparation of the glass began December 17, 1984 and was completed February 11, 1985. It was prepared in response to the request of the Basalt Waste Isolation Project, Engineered Barriers Department in letter R84-2835 from M. J. Smith (E. H. Randklev) to J. E. Mendel, August 7, 1984, and corresponds to Glass D of that request. All of the glass was produced in the form of glass bars.

Nearly three kilograms of ATM-11 glass were produced from a feedstock melted in a nitrogen-atmosphere glove box at 1250°C in Denver Fire Clay crucibles. After final melting, the glass was formed into stress-annealed rectangular bars 1.9 x 1.9 x 10 cm nominal size. A total of 26 bars were cast with a nominal weight of about 100 g each.

Table 3.2 in Section 3.0 lists the analyzed composition of ATM-11 glass. The nonradioactive elements were analyzed by inductively coupled argon plasma atomic emission spectroscopy (ICP), uranium was analyzed by pulsed laser fluorimetry, technetium by beta counting analysis, and neptunium, plutonium, and americium by alpha energy analysis (counting). Examination of a single transverse section from one bar by reflected light microscopy showed random porosity estimated at 0.4 vol% with nominal pore diameters ranging from ~5 μm to 175 μm . A distinct randomly distributed second phase was observed at a very low concentration in the glass matrix, as agglomerated, metallic-like clusters.

One form of the aggregates contained mainly a high concentration of iron, while a second form had regions of high nickel concentration, and of high palladium concentration. All the aggregates also contained a low concentration of technetium and/or ruthenium, as indicated by the occurrence of the corresponding overlapping energy dispersive x-ray peaks. An autoradiograph of the sample was obtained to provide an indication of the total radionuclide distribution. The radiograph showed generally uniform shading, with scattered areas of higher exposure frequently associated with the void or pore regions. X-ray diffraction analysis of this same sample did not show any diffraction peaks from crystalline compounds, indicating that the glass probably contained less than about 5 wt% crystalline material.

ATM-11 glass has been provided to the BWIP in the form of crushed and sized powder. As of July 1986, approximately 0.8 kg of ATM-11 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 objectives of the Materials Characterization Center (MCC) is to assist the nuclear waste form projects of the U.S. Department of Energy (DOE) 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 Approved Testing Materials (ATMs) 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-11 glass. ATM-11 was produced by the MCC at the Pacific Northwest Laboratory (PNL), Richland, Washington, which is operated for the DOE by Battelle Memorial Institute.

ATM-11 glass is designed to be representative of defense high-level waste (DHLW) glasses which will be produced by the Defense Waste Processing Facility (DWPF) at Savannah River Plant, Aiken, South Carolina. This glass was produced at the request of the Basalt Waste Isolation Project (BWIP), Engineered Barriers Department, for use in their repository testing and development programs. It corresponds to Glass D described in letter R84-2835, M. J. Smith (E. H. Randklev) to J. E. Mendel, August 7, 1984. ATM-11 is a borosilicate glass made using the reference frit SRL-165 as provided by Savannah River Laboratory (SRL), to which twelve nonradioactive and six radioactive dopants were added. The nonradioactive dopants added were Ba, Cs, Mo, Ni, Nd, Pd, Rb, Ru, Sr, Te, Y, and Zr. The radioactive dopants included ^{241}Am , ^{237}Np , $^{239+240}\text{Pu}$, ^{151}Sm , ^{99}Tc , and depleted U. This combination of nonradioactive and radioactive dopants was selected by the BWIP to produce a glass representative of a 300-year-old waste glass. The glass composition at 300 years was chosen as a conservative compromise between 10-year-old waste (i.e., earliest emplacement) and 1000-year-old waste (i.e., as per NRC 10 CFR 60 and the criteria on radionuclide release). During final glass melting, graphite was also added to the dry batch similar to the method used at the SRL to control the oxidation state of 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-11 was produced in accordance with the requirements of the MCC Program Quality Assurance (QA) Plan PGM-4, Rev. 2 (including CT-74, Rev. 0) and PNL QA Manual PNL-MA-65 to March 27, 1985, and thereafter by QA Plan NWTP-1, Rev. 0 and PNL QA Manual PNL-MA-60. The work is documented in PNL Laboratory Record Books number BNW-3660 and 8NW-5501, with supporting documentation filed in the PNL Records Center.

2.0 FABRICATION^(a)

The fabrication of ATM-11 began on December 17, 1984, and was completed by February 11, 1985. All work involved in the fabrication of this glass took place in Laboratories 410 and 500, 325 Building, 300 Area, and Laboratories 208 and 220, 308 Building, 300 Area.

2.1 STARTING MATERIALS

The glass that served as the feedstock for ATM-11 was SRL-165 Black Frit 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 DWPF product, and corresponds to a waste loading of 29 wt% (by weight of oxides). The average analyzed composition of the SRL-165 black frit used in ATM-11 is given in Table 2.1. Twelve nonradioactive constituents were added to the SRL-165 glass frit to more closely represent the concentration levels predicted for the glass at an age of 300 years. The specifications for the radioactive dopants were established by the BWIP staff (E. H. Randklev, Cognizant Engineer) to provide a waste form material containing the radionuclides needed for testing. The specifications reflect the BWIP objective of obtaining a material which contains these isotopes in concentrations that reflect a suitable compromise between the concentrations identified in the reference design composition for the Defense Waste Processing Facility (DWPF) waste form, and the levels needed to meet the requirements for radiochemical analysis of fluid samples taken from hydrothermal tests. The constituents added to the base SRL-165 frit to form ATM-11 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, wherever available, to provide traceability to the original source; this information is given in Table A.1, Appendix A. The radioactive chemicals

(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) Feed-
stock for ATM-11 Glass^(a)

Oxide	As Analyzed Concentration, ^(b) wt%	Standard Deviation, wt%	Relative SD, %	90% Conf. Interval, ± wt%	Normalized Concentration, ^(c) wt%
Al ₂ O ₃	4.50	0.07	1.6	0.12	4.44
B ₂ O ₃	6.99	0.04	0.6	0.07	6.90
BaO	0.04	0.01	25.0	0.02	0.04
CaO	1.69	0.03	1.8	0.05	1.67
CeO ₂	0.09	0.02	22.2	0.03	0.09
CuO	0.02	0.0	0	0	0.02
Fe ₂ O ₃	11.95	0.05	0.4	0.08	11.80
Li ₂ O	4.94	0.04	0.8	0.07	4.88
MgO	0.77	0.02	2.6	0.03	0.76
MnO ₂	2.65	0.08	3.0	0.14	2.62
Na ₂ O	11.07	0.03	0.3	0.05	10.91
Nd ₂ O ₃	0.05	0.02	40.0	0.03	0.05
NiO	0.84	0.0	0	0	0.83
P ₂ O ₅	0.44	0.05	11.4	0.08	0.43
SiO ₂	54.3	0.22	0.4	0.37	53.63
TiO ₂	0.20	0.0	0	0	0.20
ZnO	0.07	0.04	57.1	0.07	0.07
ZrO ₂	0.67	0.02	3.0	0.03	0.66
Total	101.28				100.00

(a) Each element assumed to be present entirely as oxide shown.

(b) Average of analysis of three samples conducted on 8/5/84.

(c) Normalized values used to calculate target concentrations shown in Table 2.3.

TABLE 2.2. Specified Dopant Constituents (Nonradioactive and Radioactive) for ATM-11 Glass
(Matrix: SRL-165 Black Frit, Batch AX581875-G, Milling No. B5039)

Nonradioactive	Element Conc., g/g Glass ^(a)	Corresponding Oxide	wt% Dopant Oxide in Glass
Ba	9.56E-4	BaO	0.11
Cs	3.98E-4	Cs ₂ O	0.042
Mo	5.39E-4	MoO ₃	0.081
Nd	20.71E-4	Nd ₂ O ₃	0.242
Ni	0.15E-4	NiO	0.0019
Pd	0.83E-4	PdO	0.0095
Rb	0.67E-4	Rb ₂ O	0.0073
Ru	3.74E-4	RuO ₂	0.049
Sr	1.33E-4	SrO	0.016
Te	0.58E-4	TeO ₂	0.0073
Y	1.05E-4	Y ₂ O ₃	0.013
Zr	11.45E-4	ZrO ₂	0.15
<hr/>			
Radioactive			
²⁴¹ Am	5.8E-7	Am ₂ O ₃	6.4 x 10 ⁻⁵
²³⁷ Np	1.5E-4	NpO ₂	0.017
²³⁹⁺²⁴⁰ Pu	2.2E-5	PuO ₂	0.0025
¹⁵¹ Sm	8.2E-8	Sm ₂ O ₃	9.5 x 10 ⁻⁶
⁹⁹ Tc	1.3E-4	Tc ₂ O ₇	0.020
U (depleted)	1.6E-2	UO ₂	1.81

(a) Values given in BWIP Letter R84-2835, M. J. Smith (E. H. Randklev) to J. E. Mendel, 8/7/84.

were from stock already on hand at PNL or were obtained from Oak Ridge National Laboratory (ORNL); sources are listed in Table B.1, Appendix B. Details of the preparation of dopants from these materials are also given in Appendix B.

2.2 BATCH AND GLASS PREPARATION

The batch and glass fabrication were conducted according to the procedures described in the Run Plans prepared for this purpose: "PNL Batch Preparation Run Plan for ATM-11" and "PNL Glass Preparation Run Plan for ATM-11" (see this report, Section 2.5).

The target composition for ATM-11 is given in Table 2.3. The final glass was obtained by remelting the powdered glass from the initial melt after a 0.7 wt% graphite addition had been made for processing under reducing conditions. The procedures used for initial batch preparation and final glass fabrication were as follows:

- The nonradioactive dopants were weighed out in the compound and the quantity listed in Appendix A, Table A.1. All nonradioactive chemicals and the UO_2 powder were added to a 2922.3 g batch of the SRL-165 black frit and the entire mixture was placed into a 12 L hardened white porcelain jar mill along with ~150 Burrundum grinding balls. The entire batch, including balls, occupied ~67% of the jar mill volume. The mixture was rolled at moderate speed for a total time of 3 h, after which the grinding balls were separated from the powder mixture by screening.
- The radioactive dopant solutions were prepared as described in Appendix B. Prior to use, an aliquot was taken from each dopant solution for analysis of each solution separately (Sm-151 was not analyzed, however). A combined dopant solution was made by mixing all of the separate solutions together, just prior to blending with the dry powder.
- The combined radioisotope dopant solution was diluted to ~750 mL volume using deionized water and blended with the entire glass frit batch in a 4 L Pyrex beaker. This slurry was then thoroughly stirred with a glass stir rod, assuring that all dry powder was wetted with dopant solution.

TABLE 2.3 Target Oxide Composition for ATM-11 Glass^(a)

<u>Stable Oxides^(b)</u>	<u>Amount, wt%</u>
Al ₂ O ₃	4.32
B ₂ O ₃	6.72
BaO	0.15
CaO	1.63
CeO ₂	0.09
Cs ₂ O	4.2E-2
CuO	0.02
Fe ₂ O ₃	11.49
Li ₂ O	4.75
MgO	0.74
MnO ₂	2.55
MoO ₃	8.1E-2
Na ₂ O	10.63
Nd ₂ O ₃	0.29
NiO	0.81
P ₂ O ₅	0.42
PdO	9.5E-3
Rb ₂ O	7.3E-3
RuO ₂	4.9E-2
SiO ₂	52.25
SrO	1.6E-2
TeO ₂	7.3E-3
TiO ₂	0.19
Y ₂ O ₃	1.3E-2
ZnO	0.07
ZrO ₂	0.80
<u>Radioactive Oxides^(b)</u>	
Am ₂ O ₃	6.4E-5
NpO ₂	1.7E-2
PuO ₂	2.5E-3
Sm ₂ O ₃	9.5E-6
Tc ₂ O ₇	2.0E-2
UO ₂	1.82
Total	100.00

(a) Calculated from the sum of the normalized Analytical Composition of the matrix frit (Table 2.1) and the Specified Dopant Constituents (Table 2.2), normalized to total 100%.

(b) Each element assumed to be present entirely as oxide shown.

- The slurry was then dried in ~250 to 400 mL batches by pouring into a 600 mL stainless steel beaker and heating at ~120° to 130°C for 2 h in a pot furnace. During this drying operation the mixture was stirred frequently to promote the drying process. When the entire slurry batch had been run through this drying step, the resultant dry batches were mixed together by hand stirring in a single container.
- A second drying step was then applied by holding portions of the dried batch at 300°C for 30 min, again using a stainless steel beaker in a pot furnace. After the second drying step had been applied to the entire batch, the material was blended together again by hand mixing in a single container.
- The dried mixed material was then melted in nine approximately equal batches using Denver Fire Clay (DFC) crucibles.^(a) The batches were melted at a control temperature of 1150°C and held for 1 h in a Deltec furnace located inside a nitrogen atmosphere glove box.^(b) After 1 h at 1150°C the glass was quenched to ambient glove box conditions by pouring onto a stainless steel plate. After each quench, the crucible was recharged and the cycle repeated until all the batches had been processed.
- All the glass was coarse crushed using a hardened tool steel punch and die, and then ground using an automatic Al₂O₃ mortar and pestle until all material passed through a 100 mesh U.S. sieve size screen (150 μm). The net mass of the glass powder was measured and enough technical grade graphite was weighed out to give a 0.7 wt% loading when mixed with the glass powder.^(c) All the graphite was mixed initially with 150 g of the glass powder and blended by hand shaking in

-
- (a) Catalog No. 99D-210-08, size 30 g, supplied by DFC Ceramics, Inc., Canon City, Colorado 81212.
- (b) The graphite reductant concentration was specified by the BWIP to duplicate SRL fabrication procedures, not to obtain a specific redox condition.
- (c) The use of the nitrogen atmosphere glove box was a normal operating condition, and was not used especially for its possible contribution to reducing conditions.

a closed container. This mixture was then blended in a similar fashion with an additional 500 g of the glass powder. This glass-graphite blended batch was then sieved through a 60 mesh U.S. sieve size screen (250 μm) for additional blending. Uniform color distribution indicated that a homogeneous mixture had been obtained. This batch was then blended with the remaining glass powder by hand shaking in a closed container for ~5 min.

- The graphite-blended glass batch was melted in eight separate portions at a control temperature of 1150°C for 1 h using a DFC crucible in a Deltec furnace located inside a nitrogen atmosphere glove box. Test bars cast from the initial melts were porous and of unsatisfactory quality. Therefore, the remainder of the powder was melted by the same procedure and then quenched to ambient glove box conditions by pouring onto a stainless steel plate. When the entire graphite blended batch had been processed as described, the entire melted and quenched batch (test bars and plate-quenched glass) was coarsely crushed and sieved through a 20 mesh U.S. sieve size screen (850 μm) and blended by hand stirring in a single container.
- Final melting, from which bars were cast, was conducted in DFC crucibles at a control temperature of 1250°C in a Deltec furnace located inside a nitrogen atmosphere glove box. The crucible containing a powdered glass charge of about 350 g was placed into the preheated furnace, held for 20 min, removed from the furnace and stirred by agitating the crucible, and then returned to the furnace for an additional 20 min period. At the conclusion of the second period, bars were poured from the melt.
- Cast bars were formed in a stainless-steel bar mold of nominal size 1.9 x 1.9 x 10 cm. The bar mold was initially preheated to about 350°C on a hot plate, and the bars were cast one at a time. When cool enough to handle without slumping, each bar was transferred to the annealing furnace at 480°C. Elapsed time from pour to annealing furnace was about 1.5 to 2 min. Three bars of approximately 100 g each were poured from each crucible load before the crucible was recharged.

- All final product bars were annealed at 480°C in a Hoskins type FD-104 furnace located in a nitrogen atmosphere glove box for a minimum of 2 hours (maximum of 5 hours), followed by a furnace cool (~110°C/h) to 25°C.

2.3 MEASUREMENT AND TESTING EQUIPMENT

Mettler Balance, PC400, Calib. No. 362-06-01-017
Mettler Balance, PC4400, Calib. No. 368-06-01-008
Mettler Balance, B5, Calib. No. 362-06-03-014
Mettler Balance, H-15, Calib. No. 362-06-03-053
Fluke Digital Thermometer, Model 2190A, Calib. No. 364-79-06-003

2.4 OTHER EQUIPMENT

Norton Jar Mill, 12 L Capacity
Hoskins Furnace, Type FD-104
Love Model 49 Proportioning Controller
Deltec DT-31-S Furnace
Honeywell Digital Control Programmer, DCP-7700
Fritsch Auto-Mortar and Pestle, Pulverisette II, Al₂O₃
Corning Model PC-354 Hot Plate
Nitrogen Atmosphere Glove Box

2.5 PROCEDURES

MCC-SP-1 (PNL approved 10/18/84): Glass Component Preparation
MCC-SP-2 (PNL approved 10/19/84): Glass Preparation
MCC-SP-4 (PNL approved 10/19/84): Product Characterization
MCC-AP-5 (PNL approved 10/22/84): Sample/Data Traceability
PNL Batch Preparation Run Plan for ATM-11, Rev. 1, 12/17/84
PNL Glass Preparation Run Plan for ATM-11, Rev. 1, 11/27/84

2.6 IDENTIFICATION SYSTEM

Each final glass bar was uniquely numbered and labeled (using appropriately inscribed sticky-back tape) according to the order in which it was poured, as follows: ATM-11-1, ATM-11-2, etc. Production records for the glass are recorded on page 88 of PNL Laboratory Record Book BNW-3660, and include data on date of fabrication, bar identification number, and mass. The product summary for ATM-11 glass is given in Table 2.4 .

TABLE 2.4. Product Summary for ATM-11 Glass(a)

<u>MCC ID No.</u>	<u>Bar Mass, g</u>	<u>Pour Date</u>
ATM-11-1	91.0	2/7/85
ATM-11-2	98.3	2/7/85
ATM-11-3	92.9	2/7/85
ATM-11-4	97.4	2/7/85
ATM-11-5	98.3	2/7/85
ATM-11-6	100.1	2/7/85
ATM-11-7	103.3	2/7/85
ATM-11-8	106.9	2/7/85
ATM-11-9	94.7	2/8/85
ATM-11-10	100.2	2/8/85
ATM-11-11	101.2	2/8/85
ATM-11-12	100.0	2/8/85
ATM-11-13	96.9	2/8/85
ATM-11-14	104.2	2/8/85
ATM-11-15	103.1	2/8/85
ATM-11-16	98.7	2/8/85
ATM-11-17	104.4	2/8/85
ATM-11-18	102.1	2/8/85
ATM-11-19	102.7	2/11/85
ATM-11-20	103.5	2/11/85
ATM-11-21	101.6	2/11/85
ATM-11-22	101.4	2/11/85
ATM-11-23	103.9	2/11/85
ATM-11-24	101.2	2/11/85
ATM-11-25	108.5	2/11/85
<u>ATM-11-26</u>	<u>39.5</u>	2/11/85
Total Mass:	2556	

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

2.7 MATERIAL AVAILABILITY AND STORAGE

ATM-11 glass has been provided to the BWIP in the form of crushed and sized powder. As of July 1986, ~0.8 kg of ATM-11 glass in bar form is available for distribution. Archive samples were taken from all initial bars distributed. All remaining ATM-11 materials and archive samples are in the custody of the MCC ATMs custodian and are stored in the 308 Building (PNL) MCC storage vault.

3.0 CHARACTERIZATION

Characterization of ATM-11 glass was conducted according to "Characterization Plan for Glass MCC-ATM-11," May 23, 1985. Characterization included bulk density measurement, chemical analysis, reflected light microscopy, scanning electron microscopy (SEM) and associated energy-dispersive x-ray fluorescence analysis (EDX), x-ray diffraction analysis (XRD), and radiochemical analysis. Individual bars representing each day of production were chosen for analysis. In addition, a set of samples was taken from a single bar to allow estimation of axial chemical homogeneity. All raw data and compilations from characterization are stored in MCC fabrication task files and the PNL Records Center. The results of these analyses conducted between February 1985 and March 1986 are summarized in the following sections.

3.1 BULK DENSITY MEASUREMENT

Density was determined on four samples from three bars (Table 3.1). Density was measured using the MCC-12 test method, which is based on ASTM procedure C 693-74, "Standard Method for Density of Glass by Buoyancy," modified to use a nominal 3.6-g sample instead of the 20-g sample specified. Density samples were prepared from glass bars by removing a section from the bar using an Isomet cutoff 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 2.729 g/cm³.

TABLE 3.1. Measured Bulk Density of ATM-11 Glass

<u>Sample</u>	<u>Measured Density, g/cm³</u>
ATM-11-5 (end)	2.726
ATM-11-14 (end)	2.730
ATM-11-14 (middle)	2.730
ATM-11-25 (end)	<u>2.728</u>

Mean Value = 2.729
(std. dev. = ±0.002)

3.2 CHEMICAL ANALYSIS

Analytical samples were taken as wafers from one end of bars ATM-11-5 and ATM-11-25 and from the end and middle of bar ATM-11-14. The wafer samples were each separately crushed and ground to pass a 200 mesh U.S. sieve size screen (75 μ m) and subdivided into three sub-batches of approximately 1 g each. The individual 1 g batches were assigned a unique, randomly selected run number, traceable by the analytical lab only to the batch and not to the bar, so as to assure a randomized analytical order. One set of these samples was submitted for ICP analysis, one set for radiochemical analysis, and the third set for oxidation state 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. The ICP analyses followed Procedures PNL-SP-7 and PNL-SP-7B [ICP Analysis (325 Building)]. The procedure specifies that instrument calibration checks must agree with standard values to within $\pm 3\%$ relative ($\pm 10\%$ for Na, K and Li). Details of application of the procedures and documentation of results are contained in the records for the analytical laboratory. Separate powder aliquots from the same analytical sample were used for radiochemical analysis of the concentrations of the radioactive constituents except samarium, for which no analytical procedure was available. Uranium was analyzed by laser fluorimetry using a Scintrex UA-3 uranium analyzer (HEDL Procedure HTA-4-16); technetium by beta-counting analysis of an isotope specific separation (HEDL Procedure HTA-4-12); and neptunium, plutonium and americium by alpha energy analysis (counting) techniques (HEDL Procedures HTA-4-13, -14 and -15). A plutonium isotopic analysis was also performed on the initial dopant solution. The alpha/beta counting was performed with counting systems based on Ludlum/Tracor Northern components.

Results of the chemical analysis of the four samples of ATM-11 glass are given in Table 3.2. The average count rate data for Am, Np, Pu and Tc, as well as the method of calculation to determine radioisotope oxide concentrations, are given in Table 3.3.

The average bar-to-bar relative standard deviation in concentration for 18 of the 29 measured constituents was about $\pm 3\%$. Six constituents (Ce, P, Pd,

TABLE 3.2. Composition of ATM-11 Glass

Stable Oxides ^(b)	Analysis ^(a)		Relative SD, %	90% Conf. Interval, ± wt%	Target Concentration, wt%	Difference of ATM-11 From Target Value	
	Concentration, wt%	Std. Dev., wt%				wt%	Relative %
Al ₂ O ₃	4.64	0.11	2.4	0.13	4.32	+0.32	+7.4
B ₂ O ₃	6.58	0.07	1.1	0.08	6.72	-0.14	-2.1
BaO	0.14	0.01	7.1	0.01	0.15	-0.01	-6.7
CaO	1.77	0.03	1.7	0.04	1.63	+0.14	+8.6
CeO ₂	<0.04(DL)	-	-	-	0.09	-	-
Cs ₂ O	NA	-	-	-	0.04	-	-
CuO	NA	-	-	-	0.02	-	-
Fe ₂ O ₃	11.13	0.10	0.9	0.12	11.49	-0.36	-3.1
Li ₂ O	4.80	0.05	1.0	0.06	4.75	+0.05	+1.1
MgO	0.53	0.01	1.9	0.01	0.74	-0.21	-28.4
MnO ₂	2.57	0.04	1.6	0.05	2.55	+0.02	+0.8
MoO ₃	0.09	0.03	33.3	0.04	0.08	+0.01	+12.5
Na ₂ O	10.48	0.13	1.2	0.15	10.63	-0.15	-1.4
Nd ₂ O ₃	0.17	0.02	11.8	0.02	0.29	-0.12	-41.4
NiO	0.69	0.05	7.3	0.06	0.81	-0.12	-14.8
P ₂ O ₅	<0.5(DL)	-	-	-	0.42	-	-
PdO	<0.2(DL)	-	-	-	0.01	-	-
Rb ₂ O	<1.0(DL)	-	-	-	0.01	-	-
RuO ₂	0.10	0.01	10.0	0.01	0.05	+0.05	+100.0
SiO ₂	52.23	0.61	1.2	0.72	52.25	-0.02	-0.0
SrO	0.02	-	-	-	0.02	0	0
TeO ₂	<0.03(DL)	-	-	-	0.01	-	-
TiO ₂	0.21	0.01	4.8	0.01	0.19	+0.02	+10.5
Y ₂ O ₃	<0.01(DL)	-	-	-	0.01	-	-
ZnO	0.03	0.01	33.3	0.01	0.07	-0.04	-57.1
ZrO ₂	0.80	0.08	10.0	0.09	0.80	0	0
Radioactive							
Oxides ^(b)							
²⁴¹ Am ₂ O ₃	6.28E-4	1.05E-5	1.7	1.23E-5	6.4E-5	+56.4E-5	+881.0
²³⁷ NpO ₂	2.37E-2	8.98E-4	3.8	1.06E-3	1.7E-2	+0.7E-2	+39.4
²³⁹⁺²⁴⁰ PuO ₂	1.49E-2	5.53E-4	3.7	6.50E-4	2.5E-3	+12.4E-3	+496.0
¹⁵¹ Sm ₂ O ₃	9.80E-6 ^(c)	-	-	-	9.5E-6	-	-
⁹⁹ Tc ₂ O ₇	1.28E-2	9.98E-4	7.8	1.17E-3	2.0E-2	-0.72E-2	-36.0
²³⁵⁺²³⁸ UO ₂	1.76	6.93E-2	3.9	8.14E-2	1.82	-0.06	-3.3
Total	98.79						

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

(b) Each element assumed to be present entirely as oxide shown.

(c) Calculated concentration of Sm₂O₃; see Table 3.3.

DL = Detection limit.

TABLE 3.3. Calculation of Radioisotope Concentrations

A. Concentrations Based on Count Rate Data

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
^{241}Am	4.35×10^7	7.618×10^9	0.9101	6.28×10^{-4}	Am_2O_3
^{237}Np	3.27×10^5	1.566×10^6	0.8811	2.37×10^{-2}	NpO_2
$^{239+240}\text{Pu}$	2.15×10^7	$1.632 \times 10^8(a)$	0.8820	1.49×10^{-2}	PuO_2
^{99}Tc	3.07×10^6	3.771×10^7	0.6385	1.28×10^{-2}	Tc_2O_7

Column (1) = measured and reported values,

Columns (2) and (3) = standard values.

Column (4) = calculated concentrations = $[\text{Col}(1)/\text{Col}(2)/\text{Col}(3)] \times 10^{-3} \times 100$.

B. ^{151}Sm (not analyzed)

Calculated concentrations of Sm_2O_3 based on known additions of ^{151}Sm to batch: (b)

$$\left[\frac{0.8628}{0.8624} \right] \left[\left(\frac{2.95}{10.0} \right) \left(\frac{24.9}{25.0} \right) \right] \left[\frac{10^{-3}}{3 \times 10^3} \right] 100 = 9.80 \times 10^{-6}\%$$

(available mass of Sm_2O_3) (Sm_2O_3 mass added to glass batch) (g/g of glass batch) (100)

(a) Weighted average based upon isotopic analysis of 92.48 wt% ^{239}Pu , 7.14 wt% ^{240}Pu .

(b) See Appendix B.

Rb, Te, and Y) were below the detection limits of the ICP unit used, while three constituents (Cs, Cu, and ^{151}Sm) were not analyzed because no suitable methods of analysis applicable to radioactive materials were available at the time this work was done. The analyzed concentrations of Nd, Ru, and Zr, which were present in the glass at a concentration level only slightly above the analytical detection limits, differed from bar to bar about $\pm 10\text{--}12\%$ relative, while Mo and Zn, which were also very low concentration levels, were found to differ $\pm 33\%$ relative from bar to bar. Relative errors of this magnitude associated with concentration levels near detection limits are considered acceptable because analytical imprecision increases as analysis concentration approaches the lower limit of detection. The summation of concentration values is 98.79 wt% which, when coupled with the reported bar-to-bar standard deviation, is considered satisfactory for analytical results at the 90% confidence interval.

Table 3.2 shows the difference between analyzed concentrations and the target values. The difference between actual and target values never exceeds 0.4 wt%, and relative differences are considered satisfactory for all elements except for Am and Pu. In those latter cases the oxide concentration exceeds the target value by 880% and 500%, respectively. The concentration for Am was verified by separate analyses in another laboratory, and found to be correct as reported here. No explanation has been found for the large deviations from target values; they must be assumed to be due to undetected laboratory process errors, perhaps in the original dopant solution preparation. Additional analyses of intermediate materials in the batching and glass fabrication process should be applied in future preparations to avoid similar errors.

3.3 MICROSCOPY AND AUTORADIOGRAPHY

Microscopy was conducted on a single transverse wafer removed from the end region of bar number ATM-11-5. Figure 3.1 shows the reflected light micrograph and the corresponding autoradiograph, prepared from the polished cross-section. Two microstructural features can be seen here: scattered porosity, and

a. Reflected Light Micrograph

b. Autoradiograph

FIGURE 3.1. Reflected Light Microscopy and Autoradiography of ATM-11 Glass. (Polished Section ATM-11-5. The cracks seen in the light-micrograph occurred during the mounting and polishing of the thin glass wafer, and were not present in the original glass bar.)

metallic-appearing agglomerates. The porosity was randomly dispersed throughout the wafer, with pore sizes ranging between $\sim 5 \mu\text{m}$ and $175 \mu\text{m}$. The total porosity is estimated to be $\sim 0.4 \text{ vol\%}$, based upon a visual comparison with other similar glasses of known porosity.

Scanning electron microscopy and associated analyses by energy dispersive x-ray fluorescence (EDX) were conducted on the same polished section (Figure 3.2). These examinations confirmed the largely featureless uniformity of the glass, and EDX analyses revealed no measurable differences in overall matrix composition at edge-to-edge or center locations. The only exception was the occasional occurrence of metallic aggregates noted on the light micrographs; a typical example is shown in Figure 3.2b. The crystalline-appearing agglomerates were observed in two forms. One was analyzed by EDX as principally iron. The second form was a cluster of at least two features, the major one being analyzed as principally nickel and the secondary feature as principally palladium. All of the aggregates also contained a low concentration of technetium and/or ruthenium as indicated by the occurrence of the corresponding overlapping EDX peaks.

Autoradiography on this glass sample (Figure 3.1B) showed a generally even shading, indicating a uniform distribution of at least the most radioactive constituents. The higher exposure associated with areas such as some pore regions is consistent with the EDX analysis showing possible presence of Tc. Other workers (Bradley et al.) have reported segregation of ^{99}Tc in the void regions of the glass during melting and casting. The photographic film technique used to obtain the radiograph is not isotope specific, and therefore individual isotope distributions cannot be identified.

3.4 X-RAY DIFFRACTION ANALYSIS

X-ray diffraction (XRD) analysis was performed on a solid wafer cut from bar ATM-11-5 (the same sample as used for microscopy examination). Analysis was performed on a GE/Diano XRD-5 x-ray diffraction system located inside a nitrogen-atmosphere glove box using $\text{Cu K}\alpha$ radiation and a continuous scan mode ($1^\circ 2\theta/\text{min}$). No diffraction peaks from crystalline components were detected in

a. Typical Glass Matrix Area

b. Agglomerated Second Phase

FIGURE 3.2. Scanning Electron Micrographs of ATM-11 Glass
(Polished Section ATM-11-5)

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%.

3.5 OXIDATION STATE ANALYSIS

The fabrication of ATM-11 glass included the introduction of graphite into the glass before the final melting cycle, as specified by SRL to control the final oxidation state of elements in the glass. The oxidation state of iron in the glass was used as an indication of the oxidation state of the glass. The analytical technique was developed for specific application to actinide-containing glasses based upon a procedure reported by Jones, Janheski and Goldman (1981), involves the spectrophotometric determination of reduced and total iron in a fused glass sample using 1,10-Phenanthroline. Results of this analysis permit a determination of the ferrous to ferric iron ratio. The measured data and calculated ratios for ATM-11 are given in Table 3.4 for glass samples from one end of bars ATM-11-5 and ATM-11-25, and the end and middle of bar ATM-11-14.

TABLE 3.4. Evaluation of Oxidation State in ATM-11 Glass
(ferrous/ferric ratio)

Sample	<u>Analyzed</u>		<u>Calculated</u>	
	<u>wt% Fe²⁺</u>	<u>wt% Fe Total</u>	<u>wt% Fe³⁺</u>	<u>Fe²⁺/Fe³⁺</u>
ATM-11-25	3.79	8.05	4.26	0.89
ATM-11-5	4.42	7.92	3.50	1.26
ATM-11-14 (middle)	4.50	8.07	3.57	1.26
ATM-11-14 (end)	4.46	7.92	3.46	<u>1.29</u>
Average =				1.18
				(std. dev. = ±0.19)

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APPENDIX A

SOURCE DOCUMENTATION FOR NONRADIOACTIVE CHEMICALS
AND URANIUM DIOXIDE USED AS DRY DOPANTS IN ATM-11 GLASS

APPENDIX A

SOURCE DOCUMENTATION FOR NONRADIOACTIVE CHEMICALS AND URANIUM DIOXIDE USED AS DRY DOPANTS IN ATM-11 GLASS

TABLE A.1. Sources and Quantities of Nonradioactive Chemical Compounds and Uranium Dioxide Used as Dopants in ATM-11 Glass

Compound	Manufacturer	Lot No.	Quantity Used ^(a) , g	Equivalent Oxide, g
Ba(NO ₃) ₂	MCB	10G11	5.442	3.193
CsNO ₃	Noah Chemical	118 202	1.747	1.263
MoO ₃	Research Chemical	n/a	2.427	2.427
Nd(NO ₃) ₃ ·6H ₂ O	Research Chemical	n/a	18.917	7.261
Ni ₂ O ₃ ·xH ₂ O	MCB	C8 918 NX345	0.056	0.056 ^(b)
PdO	n/a	n/a	0.285	0.285
RbNO ₃	Research Chemical	507-111	0.348	0.221
RuO ₂	Noah Chemical	S-11901	1.476	1.476
Sr(NO ₃) ₂	MCB	A4H28	0.961	0.471
TeO ₂	Fairmount	n/a	0.219	0.219
Y(NO ₃) ₃ ·6H ₂ O	Research Chemical	Y-N-4-021	0.678	0.200 ^(c)
ZrO ₂	Mackay	n/a	4.641	4.641
Depleted UO ₂ (0.15 wt% ²³⁵ U)	n/a	n/a	54.60	54.60
C ^(d)	VWR	RC2410-1	19.562	-

(a) Quantity of compound added to 2922.3 g of SRL-165 Black Frit to produce 3000 g of glass batch oxides.

(b) Assumes compound used is Ni₂O₃·1 H₂O.

(c) Quantity used was sufficient for 0.0067% Y₂O₃ in glass.

(d) Graphite blended into glass powder during glass fabrication; quantity added to the 2794.6 g of glass powder used for final glass production.

n/a = Information not available.

APPENDIX B

SOURCES AND PREPARATION OF RADIOACTIVE DOPANT
SOLUTIONS FOR ATM-11 GLASS

APPENDIX B

SOURCES AND PREPARATION OF RADIOACTIVE DOPANT SOLUTIONS FOR ATM-11 GLASS

Radioactive elements (except uranium) were added to the glass batch for ATM-11 as aqueous nitrate solutions to obtain the concentrations given in Table 2.2. Uranium dioxide and nonradioactive dopant constituents were added as solid compounds as described in Section 2.2 and listed in Table A.1. The sources of the radioactive dopants are listed in Table B.1. The acids and peroxide used in the solution preparations described below were of reagent grade quality. The radioisotope dopant solutions were prepared as follows:

- Americium - 0.0185 g of $^{241}\text{AmO}_2$ was dissolved in 8.2 mL of 4 M HNO_3 containing 0.05 M HF . A 1.0 mL aliquot of this solution was diluted to 25.00 ± 0.03 mL with deionized water and served as the base dopant solution. A 0.100 mL sample of this solution was removed and diluted to 100.1 mL for analysis (sample #A11-AM). The remaining 24.90 ± 0.03 mL and flask rinses were combined as the final stock solution.
- Neptunium - Approximately 11.4 mL of stock solution previously analyzed as 39.4 g Np/L was diluted to 25.00 ± 0.03 mL and served as the base dopant solution for ^{237}Np . A 0.100 mL sample of this solution was diluted to 10.10 mL for analysis (sample #A11-NP). The remaining 24.90 ± 0.03 mL and flask rinses were combined as the final stock solution.
- Plutonium - The plutonium used as the dopant source was obtained from an on-hand supply of anion exchanged 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, 2.3 mL of stock solution was diluted to 50.00 ± 0.02 mL with 0.03 M HNO_3 . A 0.100 mL

sample of this solution was diluted to 10.10 mL for analysis (sample #A11-PU). The remaining 49.90 ± 0.02 mL and flask rinses were combined as the final stock solution.

- Samarium - The entire 0.8628 mg (^{151}Sm mass) of the shipment from Oak Ridge National Laboratory Isotope Sales (No. 37-1245-A) was dissolved in 10 mL of 4 M HNO_3 . About 2.95 mL of this was diluted to 25.00 ± 0.03 mL. A 0.100 mL sample of this solution was diluted to 100.1 mL for analysis (sample #A11-SM), and the remaining 24.90 ± 0.03 mL was combined with flask rinses for the final stock solution.
- Technetium - 0.7456 g of NH_4TcO_4 (as the source of $^{99}\text{Tc}_2\text{O}_7$) was weighed out and dissolved in H_2O with a sufficient amount of H_2O_2 to dissolve black radiolysis products present in the NH_4TcO_4 . The solution was diluted to 25.00 ± 0.03 mL with H_2O and a 0.100 mL sample of this solution was removed and diluted to 100.1 mL for analysis (sample #A11-TC). The remaining 24.90 ± 0.03 mL plus flask rinses were combined to serve as the final stock solution.

Results of the radiochemical analyses for the radioactive dopant solutions described above are given in Table B.2. The dilution factors in that table are the multipliers needed to calculate actual dopant solution concentrations from the analysis of the small aliquots removed as described above. The mass spectrometry isotope analysis for the plutonium dopant is given in Table B.3.

TABLE B.1. Sources of Radioactive Chemical Compounds Used
for Dopant Solution in ATM-11 Glass

<u>Radioisotope</u>	<u>Material Used</u>	<u>Source</u>	<u>Order No./Batch</u>
^{241}Am	AmO_2 powder	ORNL(a)	Info. not available
^{237}Np	Nitrate solution	PNL stock solution	Info. not available
$^{239+240}\text{Pu}$	Nitrate solution	PNL stock solution	Info. not available
^{151}Sm	Sm_2O_3 powder	ORNL(a)	37-1245-A
^{99}Tc	NH_4TcO_4 powder	ORNL(a)	37-1174/P-30

(a) Oak Ridge National Laboratory.

TABLE B.2. Radiochemical Analysis of Actinide Dopant Solutions for ATM-11 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>
A11-AM	^{241}Am	4.76×10^5	7.618×10^9	1001	0.0625
A11-NP	^{237}Np	3.69×10^5	1.566×10^6	101	23.8
A11-PU	$^{239+240}\text{Pu}$	2.15×10^6	$1.632 \times 10^8(a)$	101	1.33
A11-SM	^{151}Sm	not analyzed			
A11-TC	^{99}Tc	6.05×10^5	3.771×10^7	1001	16.1

(a) Weighted average based upon isotopic analysis of 92.48 wt% ^{239}Pu , 7.14 wt% ^{240}Pu ; see Table 8.3.

TABLE B.3. Mass Spectrometry Isotopic Analysis of Plutonium Solution All-PU (dopant solution for ATM-11 glass)^(a)

<u>Isotope</u>	<u>wt%</u>	<u>Std. Dev., wt%</u>
238 _{pu}	0.024	0.03
239 _{pu}	92.48	0.04
240 _{pu}	7.14	0.04
241 _{pu}	0.299	0.002
242 _{pu}	0.052	0.002

(a) Analysis of one sample.

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