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MCC
Materials Characterization Center

**Fabrication and Characterization
of MCC Approved Testing
Material—ATM-2, ATM-3, and
ATM-4 Glasses**

J.W. Wald

March 1988

**Prepared for the U.S. Department of Energy
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**Pacific Northwest Laboratory
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FABRICATION AND CHARACTERIZATION OF MCC
APPROVED TESTING MATERIAL - ATM-2, ATM-3,
AND ATM-4 GLASSES

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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; ATM-2, ATM-3, and ATM-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. 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>	<u>Issued</u>
PNL-5577-1	ATM-1	October 1985
PNL-5577-2,3,4	ATM-2, ATM-3, ATM-4	August 1987
PNL-5577-5	ATM-5	
PNL-5577-6	ATM-6	
PNL-5577-8	ATM-8	October 1985
PNL-5577-9	ATM-9	June 1986
PNL-5577-10	ATM-10	
PNL-5577-11	ATM-11	August 1986
PNL-5577-12	ATM-12	October 1985
PNL-5577-18	ATM-18	

SUMMARY

Materials Characterization Center glasses ATM-2, ATM-3, and ATM-4 are designed to simulate high-level waste glasses that are likely to result from the reprocessing of commercial nuclear reactor fuels. The three Approved Testing Materials (ATMs) are borosilicate glasses based upon the MCC-76-68 glass composition. One radioisotope was added to form each ATM. The radioisotopes added to form ATM-2, ATM-3, and ATM-4 were ^{241}Am , ^{237}Np , and ^{239}Pu , respectively. These ATM glasses were produced in January and February 1983 at PNL at the request of staff from the Tuff Repository Investigations Project for use in testing in support of the Nevada Nuclear Waste Storage Investigations Project. All of the glass was produced in the form of stress annealed bars, but the MCC recommends that these glasses be used as testing materials only in the form of powder at 250 μm (60 mesh U.S. sieve size) or smaller, prepared by grinding and blending at least 35 to 40 g (nominally one bar) of the glass.

Each of the ATM lots described in this report was produced in a nominal lot size of 450 g. Each was produced from feed stock melted in a nitrogen-atmosphere glove box at 1200°C in a platinum crucible. Each ATM was cast into bars using a rectangular stainless steel bar mold of nominal size 1.3 x 1.3 x 7.6 cm; bars were annealed at 500°C for 2 h for stress relief.

The analyzed compositions of these three glasses are listed in Table 3.2 in Section 3.0. The nonradioactive elements were analyzed by inductively coupled argon plasma atomic emission spectroscopy (ICP), and the radioisotope analyses were done by alpha energy analysis. Chemical analysis indicated that the glasses were very similar in composition (with the exception of the radioisotopes) and did not deviate significantly from MCC-76-68. The average concentration (wt%) of radioisotope in each ATM was:

$$\text{ATM-2 } (^{241}\text{Am}_2\text{O}_3) = 5.66\text{E-2}$$

$$\text{ATM-3 } (^{237}\text{NpO}_2) = 2.45\text{E-1}$$

$$\text{ATM-4 } (^{239}\text{PuO}_2) = 6.20\text{E-2}$$

Scanning electron microscopy of these ATMs showed the presence of only a few small ($\sim 1\ \mu\text{m}$) pores and scattered crystalline-appearing particles. Alpha autoradiographs indicated fairly uniform ^{241}Am and ^{239}Pu distribution

throughout ATM-2 and ATM-4, respectively, with a slight indication of localized inhomogeneity. Swirl patterns in the autoradiograph for ATM-3 indicated that significant layering of ^{237}Np occurred, probably during bar pouring. X-ray diffraction analysis produced no crystal peaks in any of the glass spectra, indicating that the crystalline material content is probably less than 5 wt%. Concerns regarding the radioisotopic homogeneity in ATM-2, ATM-3, and ATM-4 (as outlined in Section 3.0) have led the MCC to recommend that these ATMs be used in testing only as-crushed material.

These ATM glasses have been used in chemical durability testing conducted at PNL for the Tuff Repository Investigations Project and for the Materials Characterization Center. As of July 1986, approximately 298 g of ATM-2, 218 g of ATM-3, and 215 g of ATM-4 remained available for distribution. Requests for materials or services related to these glasses should be made to the MCC Program Office, Pacific Northwest Laboratory.

ACKNOWLEDGMENT OF CONTRIBUTORS TO ATM-2, ATM-3, AND ATM-4

The author would like to to acknowledge and thank those who assisted with the fabrication of the ATM-2, ATM-3, and ATM-4 glasses, including Jack L. Ryan, Elmer H. Shade and Ron W. Stephens.

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1.0 INTRODUCTION

One of the objectives 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 under development in the United States. The purpose of this document is to provide the details of fabrication and characterization of ATM-2, ATM-3, and ATM-4 glasses. These glasses were produced by the MCC at the Pacific Northwest Laboratory (PNL), Richland, Washington, which is operated for the DOE by Battelle Memorial Institute.

ATM-2, ATM-3, and ATM-4 are borosilicate glasses that incorporate the radioisotope dopants ^{241}Am , ^{237}Np , and ^{239}Pu , respectively, to form simulated high-level waste (HLW) glasses that might result from the reprocessing of commercial nuclear reactor fuels. Their compositions are based on the simulated HLW glass type MCC-76-68 (Mendel et al. 1977) to which the singular actinide dopants were added. The glasses were prepared sequentially during January and February 1983 at PNL. They were made at the request of the Tuff Repository Investigations Project at PNL for use initially in testing at PNL in support of the Nevada Nuclear Waste Storage Investigations (NNWSI) Project. These glasses were the first in the ATM series of actinide doped glasses to be produced for MCC distribution. As such, they were not produced under the same detailed fabrication and characterization specifications that were incorporated into later ATM materials such as ATMs-8, -9, -10, -11, or -12. Although ATM-2, ATM-3, and ATM-4 are felt to be suitable testing materials when used as recommended, they are not typical of the uniform material produced in more recent fabrication efforts.

Section 2 of this report provides the details of the fabrication of the glasses; Section 3 summarizes the analyses and characterization of the final glass products. The glasses reported here were produced under the guidance

of the appropriate MCC Program quality assurance (QA) plan and PNL QA Manual. The work was documented in an official PNL Laboratory Record Book, and supporting documentation was filed in the PNL Records Center.

2.0 FABRICATION^(a)

The three ATM glasses reported here were made sequentially over a period of 2 months and were processed similarly except for the addition of the different actinide dopants. ATM-2 fabrication work began on February 10, 1983, and was completed by February 16, 1983. ATM-3 fabrication work began on January 3, 1983, and was completed by January 14, 1983. ATM-4 fabrication work began on January 12, 1983, and was completed by January 28, 1983. Actinide dopant solution preparation was conducted in Lab 410, 325 Building, 300 Area. The fabrication of the glasses took place in Lab 208, 308 Building, 300 Area. All three glasses were prepared from crushed type MCC-76-68 glass to which the individual dopant was added as a nitrate in solution.

2.1 STARTING MATERIALS

The glass that served as the feedstock for ATM-2, ATM-3, and ATM-4 was MCC-76-68, Lot 3, bar numbers 265, 276, 281, 307, and 309. The MCC-76-68 glass was produced for PNL by Penberthy Electromelt of Seattle, Washington, and received at PNL in September 1980. The average analyzed composition of the MCC-76-68 Lot 3 glass used for these ATMs, as reported by Mellinger and Daniel (1984), is given in Table 2.1; no additional analyses were conducted on the glass used as part of this effort. The actinide dopants were added as dilute nitrate solutions prepared from supplies of oxide powders on hand at PNL.

The selection of the actinide dopants, the dopant concentration levels, and glass batch sizes were defined by G. L. McVay for the Tuff Repository Investigations Project in December 1982. The intended dopant concentration levels were: ATM-2, 0.064 wt% $^{241}\text{Am}_2\text{O}_3$; ATM-3, 0.306 wt% $^{237}\text{NpO}_2$; and ATM-4, 0.062 wt% $^{239}\text{PuO}_2$. The target nominal mass for each batch of glass was 450 g.

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

TABLE 2.1. Analyzed Composition MCC-76-68 Lot 3 Glass from Mellinger and Daniel (1984, Table 12)

(Feedstock for Glasses ATM-2, ATM-3, and ATM-4)

Oxide(a)	Concentration, wt%(b)	Std. Dev., wt%(c)
Al ₂ O ₃	0.59	0.02
B ₂ O ₃	9.06	0.01
BaO	0.55	0.00
CaO	2.37	0.04
CdO	0.05	0.01
CeO ₂	0.92	0.01
Cr ₂ O ₃	0.47	0.01
Cs ₂ O	1.11	0.06
Dy ₂ O ₃	0.01	0.00
Eu ₂ O ₃	0.01	0.00
Fe ₂ O ₃	9.56	0.13
Gd ₂ O ₃	0.02	0.00
K ₂ O	<0.06	--
La ₂ O ₃	4.16	0.05
MgO	0.16	0.01
MnO ₂	0.06	0.01
MoO ₃	1.97	0.03
Na ₂ O	12.77	1.21
Nd ₂ O ₃	1.41	0.02
NiO	0.23	0.01
P ₂ O ₅	0.80	0.16
SiO ₂	42.00	0.26
SrO	0.42	0.01
TiO ₂	3.08	0.05
ZnO	5.10	0.46
ZrO ₂	1.89	0.04
TOTAL	98.77	

(a) Oxides listed were components of fabrication specifications for MCC-76-68.

(b) Based on ICP analysis of three bars from the 100 bars comprising Lot 3 (Cs analysis by AA).

(c) Standard deviation of an individual analysis.

2.2 BATCH AND GLASS PREPARATION

All three ATM glasses were prepared by the same general method, differing only in the specific dopant added. The method was as follows:

- Five bars of MCC-76-68, Lot 3, glass were crushed, ground, and sieved to pass a 100 mesh screen ($\sim 150\ \mu\text{m}$, U.S. sieve size) using an agate chamber laboratory disc mill (Angstrom) and Newark stainless steel sieves. All the ~ 100 mesh material was combined into a single container and tumble blended to assure uniformity of mixture.
- Four hundred fifty grams of crushed glass was weighed out for each ATM and mixed with the appropriate actinide dopant solution. The dopant solutions were prepared from the respective oxides as dilute nitrate solutions using standard wet chemistry techniques. The solutions were diluted to a final solution volume of 120 mL using deionized water to achieve a ratio of 30 mL dopant solution per 100 g of glass powder. Isotopic purity was analyzed only on the plutonium dopant solution. The isotopic analysis of the plutonium was conducted by mass spectrometry, which indicated the values given in Table 3.3.
- The slurry composed of glass powder and dopant solution was blended by hand stirring for approximately 5 min. The slurry mixture then was dried as a single batch contained in a Pyrex beaker on a conventional hot plate. Total drying time was approximately 4 h at about 200°C .
- When dry, each doped batch was then crushed in a hardened 0-2 tool steel punch and die to break up any agglomerated particles and sieved to pass through a 20 mesh screen ($\sim 850\ \mu\text{m}$, U.S. sieve size).
- Each batch was melted in a single Pt crucible at 1200°C in a nitrogen atmosphere glove box and held at that temperature for ~ 20 min prior to bar pouring. Each melt was mixed, prior to pouring, by swirling and agitating the crucible.
- The molten glass batch was poured into a preheated ($\sim 400^\circ\text{C}$) stainless steel bar mold (nominal dimensions $1.3 \times 1.3 \times 7.6$ cm), allowed to

cool enough to facilitate handling, placed into an annealing furnace at 500°C, held for 2 h, and cooled in the furnace at ~100°C/h to room temperature. Three separate melting runs, producing three to four bars each, were required to process each ATM glass batch. Eleven bars of ATM-2, 13 bars of ATM-3, and 12 bars of ATM-4 were produced.

2.3 MEASUREMENT AND TESTING EQUIPMENT

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

2.4 OTHER EQUIPMENT

Fluke Digital Thermometer, 2190^(a)

Deltec DT-31 Furnace

Honeywell Digital Control Programmer, DCP-7700

Hoskins Furnace, Model FD-104

Love Model 49 Proportional Controller

Angstrom Laboratory Disc Mill, with agate grinding chamber

Corning PC-354 Hot Plate

Glove Box

2.5 IDENTIFICATION SYSTEM

Glass bars poured in this fabrication work were individually numbered, independent of pouring order. Production records for these glass batches are recorded in an official PNL Laboratory Record Book. Table 2.2 describes the glass bars produced in this work.

2.6 MATERIAL AVAILABILITY AND STORAGE

As of July 1986, approximately 298 g of ATM-2, 218 g of ATM-3, and 215 g of ATM-4 remained available for distribution. Archive samples of all materials dispensed to date have been retained. The glass and archive materials are controlled by the MCC Material Custodian and are stored in the MCC assigned storage vault in Corridor 7, 308 Building, 300 Area.

(a) This instrument was verified to be correctly calibrated subsequent to this work.

TABLE 2.2. Product Summary for ATM-2, ATM-3, and ATM-4 Glass

<u>ATM No.</u>	<u>Bar Identification</u>	<u>Mass, g</u>	<u>Pour Date</u>
ATM-2 (²⁴¹ Am ₂ O ₃)	A-1	46.22	2/16/83
	A-2	37.11	2/16/83
	A-3	37.14	2/16/83
	A-4	39.94	2/16/83
	A-5	37.41	2/16/83
	A-6	35.82	2/16/83
	A-7	37.58	2/16/83
	A-8	40.66	2/16/83
	A-9	36.98	2/16/83
	A-10	34.97	2/16/83
	A-11	27.69	2/16/83
ATM-3 (²³⁷ NpO ₂)	N-1	39.20	1/13/83
	N-2	36.14	1/13/83
	N-3	36.15	1/13/83
	N-4	37.77	1/13/83
	N-5	37.24	1/13/83
	N-6	36.53	1/13/83
	N-7	36.47	1/13/83
	N-8	35.56	1/13/83
	N-9	36.04	1/13/83
	N-10	38.09	1/13/83
	N-11	36.87	1/13/83
	N-12	21.62	1/13/83
	N-13	10.24	1/13/83
ATM-4 (²³⁹ PuO ₂)	P-1	36.71	1/27/83
	P-2	37.15	1/27/83
	P-3	37.94	1/27/83
	P-4	41.87	1/27/83
	P-5	37.63	1/27/83
	P-6	37.34	1/27/83
	P-7	38.25	1/27/83
	P-8	37.18	1/27/83
	P-9	38.50	1/27/83
	P-10	18.00	1/27/83
	P-11	37.51	1/27/83
	P-12	38.61	1/27/83

3.0 CHARACTERIZATION

ATM-2, ATM-3, and ATM-4 glasses were characterized by chemical analyses, reflected light microscopy, scanning electron microscopy (SEM), alpha autoradiography, and x-ray diffraction (XRD) analysis. The characterization work was conducted between April 1984 and January 1985. Samples were taken that represented an end and middle of selected bars to estimate axial homogeneity. The characterization data presented are intended as a guide in the use of the ATMs. All original data and compilations from the characterization efforts are stored in MCC Fabrication Task files and the PNL Records Center. The results of these analyses are summarized in the following sections. The sample selection used for various analyses is summarized in Table 3.1.

The average composition of these glasses appears to be satisfactory for use as a waste form testing material when compared with the analysis reported for MCC-76-68 (lot 3) and considering the variabilities in measured values (i.e., standard deviations). The total weight percentages of the analyzed oxide components are less than the lower limits normally expected for similar types of analyses, however. Generally, acceptable total weight percentages will be between 98 and 102 wt% for homogeneous, non-hydrous solid materials such as these. A comparison of the analytical data indicates that the analyses for Fe_2O_3 and SiO_2 in ATM-2, ATM-3, and ATM-4 glasses (Table 3.2) are systematically 0.6 and 2.0 wt%, respectively, lower than the values listed for the same two oxide components in Mellinger and Daniel (1984) for MCC-76-68, lot 3 glass. This result is consistent with more recent analyses where the inability to fuse high concentrations of SiO_2 completely, or moderate concentrations of insolubles such as Fe_2O_3 , was identified as a major difficulty in the preparation technique. The radioisotope dopants also appear to show some inhomogeneous distribution in the glasses, especially in ATM-3. Because of the uncertainties in the radioisotope homogeneity, the MCC recommends that these ATM glasses be used as crushed and blended material rather than in monolithic form. Specifically, MCC recommends that ATM-2, ATM-3, or ATM-4 testing material be used only in the form of powder at 250 μm (60 mesh U.S. sieve size) or smaller, prepared by grinding and blending at least 35 to 40 g (nominally one bar) of the glass. The crushing and blending of larger bar

TABLE 3.1. Sample Selection for Characterization Analyses of ATM-2, ATM-3, and ATM-4(a)

Bar Number	ATM-2 ($^{241}\text{Am}_2\text{O}_3$)					ATM-3 ($^{237}\text{NpO}_2$)					ATM-4 ($^{239}\text{PuO}_2$)				
	ICP	Radio-chem	Micros-copy	Auto-radiog.	X-ray Dif.	ICP	Radio-chem	Micros-copy	Auto-radiog.	X-ray Dif.	ICP	Radio-chem	Micros-copy	Auto-radiog.	X-ray Dif.
1															
2	E	E													
3		E				E	E		X		E	E			
4									X			E			
5		E					E					E			
6	E,M	E,M	E,M		E		E		X	E		E,M			M
7		E,M					E				E,M	E,M	M	(b)	
8		E,M	E			E,M	E,M		X			E,M	E,M		
9		E	E				E					E	E		
10	M	E,M					E,M	E,M	X			E			
11				X			E	E			E	E			
12						M	E,M	M	X						
13															

(a) Samples taken from end (E), middle (M) and/or unspecified location (X) in the respective bars.

(b) Bar and location used were not specified.

TABLE 3.2. Analyzed Composition of ATM-2, ATM-3, and ATM-4 Glasses

Oxide	ATM-2		ATM-3		ATM-4	
	Concentration, wt%	Std. Dev., wt%	Concentration, wt%	Std. Dev., wt%	Concentration, wt%	Std. Dev., wt%
<u>Stable Isotopes (a)</u>						
Al ₂ O ₃	0.54	0.05	0.56	0.03	0.52	0.02
B ₂ O ₃	9.54	0.21	9.38	0.03	8.94	0.03
BaO	0.54	0.01	0.54	0.01	0.54	0.00
CaO	2.38	0.01	2.38	0.02	2.39	0.05
CdO	0.05(b)	-	0.05(b)	-	0.05(b)	-
CeO ₂	0.64	0.08	0.72	0.13	0.92	0.05
Cr ₂ O ₃	0.41	0.01	0.42	0.01	0.41	0.01
Cs ₂ O	1.11(b)	-	1.11(b)	-	1.11(b)	-
Dy ₂ O ₃	0.01(b)	-	0.01(b)	-	0.01(b)	-
Eu ₂ O ₃	0.01(b)	-	0.01(b)	-	0.01(b)	-
Fe ₂ O ₃	8.97	0.05	8.98	0.10	8.93	0.03
K ₂ O	<0.06(DL)(b)	-	<0.06(DL)(b)	-	<0.06(DL)(b)	-
La ₂ O ₃	4.79	0.03	4.29	0.07	4.27	0.01
MgO	0.13	0.01	0.13	0.01	0.13	0.01
MnO ₂	0.07	0.01	0.06	0.00	0.06	0.00
MoO ₃	1.82	0.02	1.82	0.02	1.79	0.01
Na ₂ O	13.3	0.10	13.3	0.2	13.0	0.1
Nd ₂ O ₃	1.3	0.05	1.35	0.04	1.43	0.07
NiO	0.23(b)	-	0.23(b)	-	0.23(b)	-
P ₂ O ₅	0.64	0.14	0.44	0.02	0.55	0.11
SiO ₂	40.1	0.3	40.1	0.40	39.6	0.40
SrO	0.41	0.01	0.41	0.00	0.41	0.00
TiO ₂	2.91	0.01	2.91	0.03	2.88	0.02
ZnO	4.66	0.02	4.64	0.03	4.61	0.02
ZrO ₂	1.67	0.11	1.73	0.02	1.73	0.01
<u>Radioisotopes (c)</u>						
²⁴¹ Am ₂ O ₃	5.66E-2	3.81E-3	-	-	-	-
²³⁷ NpO ₂	-	-	2.45E-1	2.17E-2	-	-
²³⁹ PuO ₂ (d)	-	-	-	-	6.20E-2	4.23E-3
TOTAL	96.29		95.82		94.58	

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

(b) Nominal value from feedstock analysis, Table 2.1; element not analyzed in the ATM glass.

(c) Average of analysis of 12 specimens taken from 8 or 9 bars. Standard deviations are for an individual analysis.

(d) See plutonium isotopic distribution, Table 3.3.

DL = Detection Limit.

fractions should even out any macro-inhomogeneities. It is further recommended that additional radioisotopic analyses be conducted on any crushed material designed for testing, to verify that homogeneity exists in the material used.

3.1 CHEMICAL ANALYSIS

Chemical analysis was performed by inductively coupled argon plasma atomic emission spectroscopy (ICP) for the nonradioactive elements, using a Bausch and Lomb ARL-34000 unit. Aliquots of the same samples were used for radiochemical analysis of the radioactive constituents. Neptunium, plutonium, and americium were analyzed by alpha energy analysis using an alpha/beta counting system based around Ludlum/Tracor Northern components. Results of these analyses are shown in Table 3.2. Mass spectrometry isotopic analysis of the plutonium dopant solution is given in Table 3.3.

Chemical analysis of these ATM glasses indicated that weight percentage totals were significantly lower than what might be considered acceptable for a reasonable measurement accuracy. Examination of the reported data shows that SiO_2 and Fe_2O_3 values are consistently lower, in all cases, than those measured in the original starting glass. It is believed that an inability to completely dissolve all of the SiO_2 in the analytical fusion, and/or a precipitation from fusion solution prior to measurement, resulted in the low SiO_2 values. Fe_2O_3 is also believed to exhibit similar behavior during fusion preparation. This observation is significant in the overall totals because SiO_2 is the dominant

TABLE 3.3. Isotopic Distribution of Plutonium
Contained in ATM-4

<u>Pu Isotope</u>	<u>wt%(a)</u>	<u>Uncertainty, wt%</u>
238	0.004	0.002
239	99.09	0.01
240	0.897	0.008
241	0.004	0.002
242	0.004	0.002

(a) Average of analysis of two samples.

oxide constituent in these glasses, and Fe_2O_3 has the third highest concentration value. Values measured for the other constituents in these ATMs compare somewhat better with the starting glass composition. The variability in measured values (i.e., standard deviation) also appears comparable to the reported values in the starting glass. The average actinide dopant concentration for ATM-2 ($^{241}\text{Am}_2\text{O}_3$) was about 12% lower than the target value. The dopant concentration for ATM-3 ($^{237}\text{NpO}_2$) was about 20% lower, and in ATM-4 the dopant ($^{239}\text{PuO}_2$) concentration was essentially at the target level. The radiographic images (Section 3.3) have indicated a tendency towards inhomogeneous distribution of the actinides, which might account for these deviations. However, the analytical samples were prepared from entire wafers that were powdered and blended prior to analysis. The radiochemical results represent analysis of twelve separate wafers taken from among nine bars for each ATM. It is felt that the sampling size and methodology allow confidence in the accuracy of the radiochemical data, although the measurement precision (± 7 to 9%) follows the observation of a tendency toward inhomogeneous distribution. This deviation is not considered excessive though.

3.2 MICROSCOPY

Typical micrographs for ATM-2, ATM-3, and ATM-4 are shown in Figure 3.1. Microscopy examination was conducted on four samples from each ATM, using polished samples for reflected light microscopy and fracture surfaces in the SEM. Optical microscopy was conducted on a Ziess Axiomat metallograph, and SEM was conducted on a JEOL JSM-35C microscope with a TN-2000 analyzing system. All three glasses appeared essentially the same by both microscopy methods, with no unusual features noted. Although some pores were noted in the glass samples, they were minimal and not a major measurable feature. Some of the samples exhibited a scattering of discrete second-phase inclusions in the glass matrix. Energy dispersive x-ray analysis of these features, concurrent with the SEM examination, showed a high concentration of iron, chromium, and zinc. Based on the morphology and the chemical analyses, these inclusions appear to be a spinel that formed during the melting and/or cooling process. Their concentration in the glasses is estimated from visual examination to be less than 1.0 wt%.

83843A

83844A

83845A

FIGURE 3.1. Typical Microstructure of Polished Surface of ATM Glasses by Reflected Light Microscopy [top, ATM-2 ($^{241}\text{Am}_2\text{O}_3$); middle, ATM-3 ($^{237}\text{NpO}_3$); bottom, ATM-4 ($^{239}\text{PuO}_2$)]

3.3 AUTORADIOGRAPHY

Alpha autoradiographs were taken of single samples of ATM-2 ($^{241}\text{Am}_2\text{O}_3$) and ATM-4 ($^{239}\text{PuO}_2$) and of several samples of ATM-3 ($^{237}\text{NpO}_2$). Autoradiographic exposures were obtained by placing a piece of photographic film in contact with a polished cross section of the sample for a period of time sufficient to obtain a suitably resolved image. The film was then processed using conventional processing techniques and printed in negative image to produce darkening on the print as indication of regions of high concentration. Shading of the radiographs indicated slight localized radioisotope inhomogeneity in ATM-2 and indications of layering in ATM-4. The radiographs of ATM-3 (Figure 3.3) indicated a wide variability in the ^{237}Np distribution from bar to bar, ranging from rather uniform distribution (i.e., bars 3, 6, and 8) to gross striations, or layering, indicating nonuniform distribution (i.e., bars 4, 10, and 12). This result is likely due to poor effective blending of the dopant into the melt prior to bar pouring but may not necessarily be indicative of inhomogeneities in the nonradioactive constituents since they were part of a uniform initial matrix and not discretely added components.

3.4 X-RAY DIFFRACTION ANALYSIS

X-ray diffraction (XRD) analysis was performed on a solid wafer cut from a single bar from each ATM. Analysis was performed on a GE/Diano XRD-5 x-ray diffraction system located inside a nitrogen-atmosphere glove box using $\text{CuK}\alpha$ radiation and a continuous scan mode ($1^\circ 2\theta/\text{min}$). No diffraction peaks from crystalline components were observed in the samples 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 5.0 wt%.

83843B

83845B

FIGURE 3.2. Typical Autoradiographs of ATM-2 and ATM-4 Glasses
 [top, ATM-2 (^{241}Am); bottom, ATM-4 (^{239}Pu)]

838108A

Bar 3

838109B

Bar 4

838113A

Bar 12

FIGURE 3.3. Typical Autoradiographs of ATM-3 (^{237}Np)

FIGURE 3.3. Optical microphotographs of ATN-3 (SSV no.)

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