Distribution Category:
High-Level Radioactive Waste
Disposal in Tuff (UC-814)

ANL--89/24

DE91 000583

ANL-89/24

ARGONNE NATIONAL LABORATORY
9700 South Cass Avenue
Argonne, Illinois 60439

APPLICATION OF THE NNWSI UNSATURATED TEST METHOD
TO ACTINIDE DOPED SRL 165 TYPE GLASS

by

John K. Bates and Thomas J. Gerding

Chemical Technology Division

August 1990

MASTER
DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED
pe

TABLE OF CONTENTS

	<u>Page</u>
ABSTRACT	1
EXECUTIVE SUMMARY	2
I. INTRODUCTION	3
A. Background	3
II. EXPERIMENTAL	4
A. Overview	4
B. Starting Components	7
1. Glass Composition and Homogeneity	7
2. Oxidation State	11
3. Crystallinity	12
4. Porosity	12
5. Equilibrated J-13 (EJ-13) Water	13
6. Presensitized 304L Stainless Steel Waste Form Holders	13
III. RESULTS AND DISCUSSION	16
A. N2 Unsaturated Test	16
1. General Observations	16
2. Solution Analysis	23
3. Analysis of Test Components	32
4. Glass	34
5. Retainer Sections	43
IV. DISCUSSION	50
V. CONCLUSIONS	57
ACKNOWLEDGMENTS	58
REFERENCES	59
APPENDIX I. Compositional Homogeneity Measurements of SRL A Glass	61
APPENDIX II. Complete Solution Analytical Results from the N2 Test	62

LIST OF FIGURES

<u>No.</u>	<u>Title</u>	<u>Page</u>
1.	Schematic Drawing of Unsaturated Test Apparatus	6
2.	Photomicrograph of the Surface of Sample #11-1	12
3.	Micrographs of Polished Cross Sections of Etched Stainless Steel	17
4.	Photographs of the Reacted Top Glass Surfaces from N2 Tests	22
5.	Normalized Releases for the N2 Batch Tests	27
6.	Normalized Releases for the N2 Continuous Tests	28
7.	Log of the Concentrations in the Test Solutions for the Three Continuous Tests in the N2 Unsaturated Test Series	31
8.	Photomicrograph of the Bottom Glass Surface from Test N2 #8	35
9.	Photomicrographs and EDS Spectra of Alteration Products Shown in Fig. 8	36
10.	Photomicrograph and EDS Spectra of Alteration Products that Formed on the Top Glass Surface from Test N2 #6	37
11.	Photomicrographs and EDS Spectrum of the Top Glass Surface from Test N2 #6	37
12.	Photomicrographs and EDS Spectra of the Glass Surface from Test N2 #2	39
13.	Photomicrograph and EDS Spectra on the Top Glass Surface from Test N2 #6	39
14.	Photomicrographs and EDS Spectra of the Solid Solution that Formed on the Retainer Surface	40
15.	Photomicrographs and EDS Spectra of Alteration Products that Form on the Retainer Sections	44
16.	Photomicrographs and EDS Spectra that Were Formed on the Bottom Retainer Sections	45

LIST OF FIGURES - Cont'd

<u>No.</u>	<u>Title</u>	<u>Page</u>
17.	Photomicrographs and EDS Spectra of Amorphous Reaction Products that Form on the Retainer Sections	46
18.	Photomicrographs and EDS Spectra of Pre-Sensitized but Non-Reacted Surface of 304L ss, Heat 699960	48
19.	Photomicrographs and EDS Spectra of Reaction Products that Formed on the Surface of the Top Retainer Section from Test N2 #4	49
20.	Comparison of Normalized Elemental Releases from the N2 Test, the F1 Test, and the P-VIII Experiments	54

LIST OF TABLES

<u>No.</u>	<u>Title</u>	<u>Page</u>
1.	Results from the F-1 Unsaturated Test Series	5
2.	Test Matrix for Unsaturated Tests	8
3.	List of Procedures Used in the Unsaturated Test	9
4.	Composition of Glasses Used in the N2 Tests	10
5.	Amount of Fe ²⁺ in Glass Samples	11
6.	Analyses of Equilibrated J-13 Water and J-13 Water Used in Unsaturated Testing	14
7.	Composition of 304L Stainless Steel Used in the N2 Unsaturated Test Series	15
8.	Test Matrix and Component Weights for the N2 Test Series	18
9.	Blank Corrected Solution Results from the N2 Tests	25
10.	Normalized Release from the N2 Unsaturated Tests	26
11.	Normalized Lithium Release through 52 Weeks of Testing	29
12.	Measured Elemental Concentrations of Test Solution in Contact with Glass during the Unsaturated Test	30
13.	Concentration of Np, Pu, and Am Associated with Test Solutions in the N2 Tests	33
14.	XRD Results for Iron Containing Phases Formed during the Unsaturated Test	38
15.	XRD Results for the Solid Solution Ca _{0.74} (Mn,Mg) _{0.26} CO ₃ Plus CaSO ₄ •2H ₂ O	41
16.	XRD Results for the Group of Precipitates Containing Sodium Feldspar, Orthoclase, and Cristobalite	42
17.	Interactions and Sensitization C served in Unsaturated Tests and Experiments	55

LIST OF TABLES (Cont'd)

<u>No.</u>	<u>Title</u>	<u>Page</u>
AI-1.	Relative Radionuclide Concentrations in N2 Glass Samples (Gamma Counting)	61
AI-2.	Relative Radionuclide Concentrations in N2 Glass Samples (Total Alpha Counting)	61
AII-1.	Solution Analyses (Cations) for the N2 Test Series	63
AII-2a.	Solution Results (Neptunium) for the N2 Test Series	65
AII-2b.	Solution Results (Plutonium) for the N2 Test Series	70
AII-2c.	Solution Results (Americium) for the N2 Test Series	75
AII-3a.	(Neptunium)	80
AII-3b.	(Plutonium)	81
AII-3c.	(Americium)	82

APPLICATION OF THE NNWSI UNSATURATED TEST METHOD TO ACTINIDE DOPED SRL 165 TYPE GLASS

by

John K. Bates and Thomas J. Gerding

ABSTRACT

The results of tests done using the Unsaturated Test Method are presented. These tests, done to determine the suitability of glass in a potential high-level waste repository as developed by the Nevada Nuclear Waste Storage Investigations Project, simulate conditions anticipated for the post-containment phase of the repository when only limited contact between the waste form and water is expected. The reaction of glass occurs via processes that are initiated due to glass/water vapor and glass/liquid water contact. Vapor interaction results in the initiation of an exchange process between water and the more mobile species (alkalis and boron) in the glass. The liquid reaction produces interactions similar to those seen in standard leaching tests, except due to the limited amount of water present and the presence of partially sensitized 304L stainless steel, the formation of reaction products greatly exceeds that found in MCC-1 type leach tests. The effect of sensitized stainless steel on the reaction is to enhance breakdown of the glass matrix thereby increasing the release of the transuranic elements from the glass. However, most of the Pu and Am released is entrained by either the metal components of the test or by the reaction phases, and is not released to solution.

EXECUTIVE SUMMARY

The conditions anticipated for a repository located in the unsaturated zone of the tuff beds of Yucca Mountain, Nevada, are such that large amounts of flowing water are not expected to contact the waste package or accumulate in contact with the waste form. For this reason the Nevada Nuclear Waste Storage Investigations (NNWSI) Project has developed the Unsaturated Test Method to monitor the release of radionuclides from the waste form and to study physical interactions that are important under unsaturated conditions.

The Unsaturated Test Method has previously been performed using non-transuranic containing Savannah River Laboratory (SRL) 165 type glass [BATES-1, -2]. In the present report the application of the test method to SRL 165 type glass containing ^{237}Np , ^{239}Pu , and ^{241}Am is described.

The test consists of a waste package assemblage (WPA) containing the radioactive glass and 304L stainless steel as used in the Defense Waste Processing Facility (DWPF) glass pour canister. The WPA is contained in a steel reaction vessel, the air of which is saturated in water vapor, and is contacted periodically by a small amount of tuff-equilibrated repository water that is introduced through the top of the reaction vessel. During the reaction period the water, after first contacting the top glass surface, percolates over the sides of the glass, collects on the bottom of the WPA, and eventually drips to the bottom of the test vessel where it is periodically collected for analysis. The test is performed at 90°C .

Three types of interactions that affect the reaction of the glass are anticipated to occur under unsaturated conditions. These include (1) contact between the glass and moist air, followed by periodic rinsing of the glass surface with flowing water; (2) contact between the glass and standing water; and (3) contact between the glass and standing water in close contact with partially sensitized 304L stainless steel. The Unsaturated Test provides for the possibility that these interactions will occur, and specifies analyses be performed that are designed to judge the importance of each one.

In the present Unsaturated Test series (N2), duplicate batch tests were terminated at 13-week intervals through 52 weeks, and separate continuous tests were sampled at 6.5-week intervals and are still ongoing. Analyses were performed on the solutions for all the tests and on the test components for the batch tests. The solution results provide information regarding the amount of radionuclides that are actually released from the WPA, while the component analyses allow the nature of the interactions to be investigated.

The most striking observation in the present test series, compared to previous testing, was in the visually observed amount of reaction that occurred. The glass and metal components were covered with a rust-colored reaction product. This strong interaction occurred wherever the metal was in contact with the glass and could be correlated with the extent of sensitization observed in the metal. The metal sections were partially sensitized to duplicate conditions that may exist in the glass pour

canister. The strong interaction resulted in the release of elements such as Pu and Am, whose release is normally associated with breakdown of the glass matrix, but only slightly affected the release of alkali metals, whose release is usually associated with a leaching process.

The reaction processes proposed to describe the observed results include (1) interdiffusional exchange of alkali with water, which occurs over the entire glass surface, (2) the formation of a reaction layer on the surface of the glass, which forms in regions where standing water accumulates, and (3) complete breakdown of the reaction layer, which occurs in regions of strong interaction with the metal.

The actual elemental releases after 65 weeks of continuous testing, normalized to the surface area and composition of the glass, were $(NL)_{Li} = \sim 2 \text{ g/m}^2$, $(NL)_B = \sim 1 \text{ g/m}^2$, and $(NL)_{Pu,Am} = \sim 0.01 \text{ g/m}^2$. The actual amount of Pu and Am released into solution was less than that released from the glass. Most of the released Pu and Am remained associated with either the reaction products or metal components in the system.

Numerous reaction products were observed both on the metal and glass components. These included FeOOH and ferrihydrate ($5\text{Fe}_2\text{O}_3 \cdot 9\text{H}_2\text{O}$) as the major iron-containing phases that gave the rust appearance to the samples, together with calcite ($\text{Ca}_{.74}(\text{Mn,Mg})_{.26}\text{CO}_3$), gypsum ($\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$), cristobalite (SiO_2), orthoclase (KAlSi_3O_8), and albite ($\text{NaAlSi}_3\text{O}_8$). Other phases were found that had unique compositions, but gave no distinct XRD pattern.

The results from the N2 Unsaturated Test are compared to those from the parametric experiments which use a similar experimental design as the N2 Test except that the metal retainer sections are not sensitized. These comparisons indicate that in the absence of sensitized metal sections the extent of glass reaction is reduced.

I. INTRODUCTION

Investigation of the volcanic tuff beds of Yucca Mountain, Nevada, as a potential site for a high-level waste repository is a function of the Nevada Nuclear Waste Storage Investigations (NNWSI) project. As part of this study, the NNWSI Unsaturated Test has been developed as a method of obtaining data on the release of waste components from the waste package under conditions currently envisioned in the NNWSI repository environment after the 300/1000-year containment period has elapsed. Specifically, the test will provide information to be used by the NNWSI project in licensing the NNWSI repository site. The test method will provide (1) data that describe the release of radionuclides from a specifically designed glass/container assemblage under strictly controlled test conditions, and (2) information concerning synergistic effects that may occur between waste package components. The test results can be used to determine interactions that are important when glass leaching occurs in an environment of air, water vapor, and liquid water.

A. Background

The results of an Unsaturated Test matrix (F series) performed using a glass based on Savannah River Laboratory (SRL) frit 165 have been reported

[BATES-1, -2]. These initial results demonstrated that the test method was capable of producing repository-relevant results and that the extent of glass reaction depended strongly on interactions that occurred between waste package components. The greatest degree of reaction occurred when glass, standing water, and weld-affected 304L stainless steel were in contact. The extent of glass reaction, based on glass weight loss and the release of Li and B, is shown in Table 1. When there was a strong interaction in the weld-affected area, the glass reaction was greater (tests F1, F2, F3, F4, and F5) than when there was a weak interaction (tests F6, F7, F8, F9, F10, and F11). The glass reaction was described as occurring via (1) a diffusional process that resulted in a fairly constant release of alkali metals from the entire glass surface, and by (2) breakdown of the remaining gel layer that resulted in the release of all glass components from the affected area.

This first Unsaturated Test matrix served as a trial run of the test method, and two important changes to the procedure were made to make the results more consistent and easier to interpret. These were (1) the waste form holder was subjected to a pre-test heat treatment process that would promote a strong glass/water/metal interaction uniformly over the sample surface, and (2) the tuff cup was eliminated so that the total amount of radionuclides and glass components released from the WPA could be easily and accurately determined. These procedural changes are described in more detail in Section II.

To demonstrate repository relevance of the Unsaturated Test method and to help interpret the effect of varying selected test parameters, laboratory analog and parametric experimentation was performed in conjunction with the Unsaturated Test. Results of these types of experiments have been discussed previously [BATES-1, -2, -3, -4].

In this report the results of the second Unsaturated Test matrix are presented. This matrix was done using the same SRL 165 type frit as used in the F series of tests, except for the addition of ^{237}Np , ^{239}Pu , and ^{241}Am to the present glass. The batch tests have been completed through 52 weeks, while the continuous tests are ongoing having been sampled through 65 weeks. Additional laboratory analog and parametric experiments, using the same glass composition as used in the N2 Test series, are being conducted and the results will be compared with those from the Unsaturated Test in a separate report [BATES-5].

II. EXPERIMENTAL

A. Overview

The apparatus used in the Unsaturated Test has been described previously [BATES-1] and a schematic diagram of the system is shown in Fig. 1. The components of the test apparatus are the test vessel, which provides for collection and containment of liquid and support of the waste package; the waste package assemblage (WPA), which consists of the waste form and presensitized metallic components representing the canister (retainer bottom and retainer top); and a solution feed system to inject test water.

Table 1. Results from the F-1 Unsaturated Test Series

Test No.	Test Description	Weight Change (NL) _{wt} , g/m ²	Total Release, μg or (g/m ²)			
			Li	(NL) _{Li}	B	(NL) _B
F-1	13-week batch	0.26 ^b	5.9 ^b		3.9 ^b	
F-2	13-week batch	0.25 ^b	6.2 ^b		5.9 ^b	
Sum of Two	6.5-week continuous ^a		10.0	0.4	3.3	0.1
F-3	26-week batch	0.47 ^b	10.9 ^b		15.1 ^b	
F-4	26-week batch	0.40 ^b	11.1 ^b		15.5 ^b	
Sum of Four	6.5-week continuous ^a		20.8	0.8	5.0	0.2
F-5	39-week batch	0.71 ^b	14.4 ^b		22.2 ^b	
F-6	39-week batch	0.25	10.8		5.4	
Sum of Six	6.5-week continuous ^a		27.5	1.1	6.3	0.2
F-7	52-week batch	0.28	10.5		4.7	
F-8	52-week batch	0.06	4.4		7.1	
Sum of Eight	6.5-week continuous ^a		37.9	1.3	7.2	0.3
F-9	52-week continuous	0.05				
F-10	52-week continuous	0.06				
F-11	52-week continuous	0.11				

^aAverage of tests F-10 and F-11.^bThese tests had a strong glass/metal interaction.

1. Body
2. Nut
3. Cap
4. Retainer Top
5. Retainer Bottom
6. Ethylene Propylene Gasket
7. Swagelok Fitting

Fig. 1. Schematic Drawing of
Unsaturated Test
Apparatus, 10 mm =
19.8 mm full-scale.

This configuration is the same as used in the F1 Tests, except that the tuff cup and tuff cup supporting ring present in the F1 Test series has been removed. This allows the liquid that is released from the WPA to be directly collected and analyzed. Additionally, the 304L stainless steel components of the waste form holder have been presensitized. This was done to reduce the localization of strong glass/stainless steel interaction that occurred previously near the weld-affected region of the metal. It was hoped that with uniformly sensitized metal, the strong glass/stainless steel interaction would occur throughout regions of glass/stainless steel contact. Such contact would be anticipated between the DWPF pour canisters and the glass, and by instituting a standardized heat treatment to the metal, more consistent release results should be achieved.

The WPA is contacted every 3.5 days by 0.075 mL (drops) repository water that has been preequilibrated with tuff at 90°C. The test is conducted at 90°C and the inject system is configured so that the water attains a temperature of 90°C before contacting the glass. The nature and degree of radionuclide release from the assemblage are determined by collection and analysis of the water that has contacted the package and by surface analysis of the assemblage components. The test solution has cooled to ambient temperature when collected, however, the water that is collected in the bottom of the vessel has dripped from the WPA at temperature. Solution samples are collected immediately upon termination of the test and after the vessel has been acid stripped for removal of elements that may have precipitated from solution or sorbed onto the stainless steel. Materials interactions are then noted, and secondary alteration products which influence the nuclide release from the assemblage, are identified.

The test schedule incorporates batch and continuous testing. In the batch mode, tests are terminated at 13-week time intervals through 52 weeks. In the batch mode, the test apparatus is disassembled, and analyses of both the solution and components are performed. In the continuous mode, the WPA (including liquid associated with the WPA) is transferred to a new test vessel; and the test is continued. For the first 52 weeks the continuous tests are sampled at 6.5-week intervals. After 52 weeks, 13-week intervals are used. Analyses are done on the solution in the old vessel. With the continuous-testing matrix, replication of solution analysis can be achieved. In addition, investigation of the test components is possible at the termination points, and yet the test can continue for an unspecified number of test periods or until information most useful to repository evaluation is obtained. The Test matrix is shown in Table 2.

This matrix allows for two batch tests to be terminated at the 13-, 26-, 39-, and 52-week intervals and for three continuous tests to be sampled periodically. Thus, for each 13-week batch interval there will be five sets of solution results. After the batch tests have terminated, there will be three sets of solution results.

B. Starting Components

The procedures used in setting up, performing, and analyzing the Test are listed in Table 3 and provided in detail in [BATES-1, -2]. Based on these procedures, a description of the starting Test components is provided below.

1. Glass Composition and Homogeneity

The basis for the glass composition is SRL 16, black frit doped with Cs, Sr, U [CHANDLER] and the transuranic elements ^{237}Np , ^{239}Pu , and ^{241}Am . This composition was chosen because of the data available as part of the SRL testing program [CHANDLER] and because a single quantity of SRL black frit obtained from SRL has formed the basis for all Unsaturated Testing of SRL glass and of all recent parametric experimentation done at ANL.

The radionuclide content of the glass was determined by complete dissolution of the glass followed by radiochemical counting techniques and isotopic mass spectroscopy. The remaining cations were determined using inductively coupled plasma spectroscopy (ICP), gravimetric titration for Si, atomic absorption for Cs, fluorescence for U, mass spectrometric isotope dilution for B, and colorimetry for P. The glass composition is given in Table 4.

The compositional homogeneity of the glass was determined for the radionuclides and other glass-forming elements following the procedures listed in Table 3. The results of the homogeneity measurements shown in

Table 2. Test Matrix for Unsaturated Tests

Test No.	Components	Test Duration, weeks						
		0	6.5	13	19.5	28	39	52
N2 #1 Batch	glass, 304L ss holder							↓ = periodic change
#2 Batch	glass, 304L ss holder							= termination
#3 Batch	glass, 304L ss holder							
#4 Batch	glass, 304L ss holder							
#5 Batch	glass, 304L ss holder							
#6 Batch	glass, 304L ss holder							
#7 Batch	glass, 304L ss holder							
#8 Batch	glass, 304L ss holder							
#9,10,12 Continuous	glass, 304L ss holder							
#11 Continuous	blank							

ANALYSES REQUIRED^b

Total radionuclide (solution and metal)	✓✓	✓✓✓✓	✓✓	✓✓✓✓	✓✓	✓✓✓✓	✓✓	✓✓✓✓	✓✓
Total matrix element (solution Li, B)	✓✓	✓✓✓✓	✓✓	✓✓	✓✓✓✓	✓✓	✓✓✓✓	✓✓	✓✓
Wt loss and surface analyses	✓✓	✓✓	✓✓	✓✓	✓✓	✓✓	✓✓	✓✓	✓✓
Interaction of stainless steel		✓✓		✓✓	✓✓	✓✓	✓✓	✓✓	✓✓

^aThe continuous tests are sampled every 13 weeks after the 52-week interval.^bA check means this analysis is required; the number of checks indicate replicate analyses possible. The wt loss and surface analyses for the continuous test will be done when the tests are terminated.

Table 3. List of Procedures Used in the Unsaturated Test

NNWSI-	Title
05-003	Current Standard Operating Procedures for the Determination of Total Metals in Water by Inductively Coupled Plasma-Atomic Emission Spectrometry (ICP-AES)
05-004	Procedure for Uranium in Leachate Solutions
05-005	Acceptance Specifications and Production Method for NNWSI Glass Forms to be Used in the Unsaturated Test Method
05-007	Modification and Setup of Blue-M Oven Model OV490A-2 for NNWSI Tests
05-008	Procedure for Making Actinide-Doped Glass for NNWSI Unsaturated Testing
05-009	Equilibration of J-13 Water
05-010	Cleaning Procedures for Test Vessels, Waste Form Holders, O-Rings, and Polyethylene Containers Used in the NNWSI Unsaturated Test Method
05-011	NNWSI Unsaturated Test Procedure
05-012	Acceptance Specifications and Production Method for Test Vessels, Waste Form Holders, and O-Rings Used in the NNWSI Unsaturated Test Method
05-013	Sensitization of Waste Form Holders
05-018	Procedure for Alpha Spectrometry Counting Actinide Samples
05-021	Procedure for Qualitative Description of Test Components
05-024	Dissolution of Glass for Metals Analysis
05-025	Determination of Relative Radionuclide Concentration Using Gamma Spectroscopy
05-026	Homogeneity of Glass Surfaces Using Alpha Spectroscopy
05-027	Determination of Ferrous Iron in Glasses
05-029	Comparative Quantitative Analysis of Glasses Using SEM/EDS/WDS

Table 4. Composition of Glasses Used in the N2 Tests

Formula	Oxide wt %	SRL A Element wt %	Black Frit Oxide, wt %	
			Ferro ^a	MCC ^b
Al ₂ O ₃	4.08	2.16	4.1	4.3
Am ₂ O ₃	3.9E-4	6.21E-4		
B ₂ O ₃	6.76	2.09	6.8	6.8
BaO	0.06	0.05		<0.1
CaO	1.62	1.16	1.5	1.6
CeO ₂	<0.05	<0.04		
Cr ₂ O ₃	<0.01	<0.007		
Cs ₂ O	0.072	0.07		
Fe ₂ O ₃	11.74	8.20	12.3	11.7
K	NA ^c			
La ₂ O ₃	<0.05	<0.004		0.2
Li ₂ O	4.18	1.94	4.7	4.8
MgO	0.70	0.42	0.8	0.6
MnO ₂	2.79	1.76	2.9	2.8
MoO ₃	<0.01	<0.007		
Na ₂ O	10.85	8.05	10.3	10.8
Nd ₂ O ₃	<0.05	<0.005		
NiO	0.85	0.67	0.9	0.8
NpO ₂	0.008	2.39E-2		
P ₂ O ₅	0.29	0.13		0.3
PuO ₂ ^d	0.022	1.98E-2		
SiO ₂	52.86	24.71	54.1	51.6
SrO ₂	0.11	0.10		0.2
TiO ₂	0.14	0.08		<0.1
UO ₂	0.92	0.81		
ZnO	0.04	0.03		
ZrO ₂	0.66	0.48	1.2	0.7
Tc ₂ O ₇	-	<0.001		
F			0.06	
Cl			0.05	
Pb			0.05	

^aBlack frit supplied to ANL by SRL, composition as determined by Ferro Corp.

^bBlack frit supplied by SRL to the MCC, composition as determined by MCC.

^cNA = not analyzed.

^dIsotopic breakdown is ²³⁹Pu (97.44%), ²⁴⁰Pu (2.52%), ²⁴¹Pu (0.04%), and ²⁴²Pu (0.003%).

Appendix I, indicate that all components of the glass fall within acceptable limits, i.e., radionuclides and elements making up more than 5 wt % of the glass fall within $\pm 5\%$ of the mean, elements making up 1 to 5% of the glass within $\pm 10\%$ of the mean, and other elements within $\pm 50\%$ of the mean.

2. Oxidation State

The redox state of the glass was regulated to some degree by the addition of graphite to the melt. The purpose of this was to simulate conditions anticipated for the DWPF melter; Fe, Mn, and U are the most abundant species in the glass whose oxidation state would be affected. The effectiveness to which the redox state of the melt was controlled was determined by measuring the Fe^{2+}/Fe ratio for four samples. The results are shown in Table 5 where the average ratio is 0.31 ± 0.09 . By comparison, waste forms cast in an unregulated air environment (parametric samples) [BATES-5] were found to have an average Fe^{2+}/Fe ratio of 0.14, ranging from 0.22 to 0.07. The addition of graphite to the melt resulted in additional foaming, and the variance in the amount of Fe^{2+} in individual samples was likely a result of remelting different amounts of batch material to cast the individual forms. However, the addition of graphite did result in more reducing casting conditions.

Table 5. Amount of Fe^{2+} in Glass Samples

Sample Description	Wt % Fe^{2+} in Glass
N2 #9	1.65
N2 #5	2.40
N2 #8	3.40
N2 #12	2.80
Parametric ^a	
P-VIII #1	0.80
#2	1.65
#3	0.90
#4	2.10

^aThe parametric samples were cast using the same method as was used for the Unsaturated Test samples except the air environment was uncontrolled [BATES-5].

3. Crystallinity

The glass was examined by x-ray diffraction (XRD) and SEM/EDS to characterize crystalline material present in the sample. The XRD pattern indicated that no crystalline material was present which, based on past experience, indicates there is less than 5% crystalline material present in the sample. However, with SEM/EDS small inclusions were observed. There were diffuse circular areas of approximately 2 μm in diameter that were slightly enriched in S and Cl, and small phases rich in Fe or Cu.

4. Porosity

A concern in accepting samples for use in the Unsaturated Test is the size and position of bubbles that may form in the glass during the casting process. Imperfections that result from bubbles located on the edges of the surface could direct water flow in a preferential pathway which is undesired. Alternatively, imperfections on the flat surfaces could act as reservoirs to hold water, thereby promoting reactions with a lower SA/V (surface area of glass to volume of solution) ratio than present over the majority of the surface. The samples were carefully examined to assure that no strategically located large imperfections were present on the surfaces, and micrographs were taken using reflected-light optical microscopy and SEM to characterize the glass surface. Figure 2 is a SEM micrograph of a polished surface which indicates voids ranging from

Fig. 2. Photomicrograph of the Surface of Sample #11-1 Archive Showing the Maximum Degree of Bubble Formation (Dark Circular Areas) Found on the Glass Surface. The pores represent less than 0.4% of the displayed surface area. The scale bar represents 100 μm .

5 to 50 μm in diameter are present on the surface. These voids are randomly spaced throughout the glass and surfaces and constitute less than 0.4% of the total surface area. This void area was measured using the image processing capabilities of the SEM.

5. Equilibrated J-13 (EJ-13) Water

J-13 water from the J-13 well located near the repository horizon was equilibrated with tuff at 90°C following procedure NNWSI-05-009 to produce EJ-13 water. The composition of the water as analyzed throughout the test period is invariant and is given in Table 6. This is the same batch of water that was used in the F series of tests [BATES-1, -2].

6. Presensitized 304L Stainless Steel Waste Form Holders

During the process of filling the DWPF pour canisters with glass, the 304L stainless steel will be subjected to temperatures ranging from 1150°C to several hundred degrees centigrade as the glass cools. During this initial cooling period the possibility exists that the microstructure of the pour canister could become sensitized. Sensitization could also occur during fabrication or welding of the canister or during the long period of storage in the repository. Sensitization of stainless steel is a process by which grain boundary precipitation of chromium carbide occurs. This process depletes localized areas of the steel in chromium and the steel becomes susceptible to intergranular stress corrosion cracking and other methods of corrosion.

Because of the possibility of sensitization, the F Test series incorporated a weld-affected region of steel in contact with the glass. However, no control was maintained over the weld conditions, and a non-uniform interaction occurred in this weld-affected area. Enhanced reaction was observed in the weld-affected region which resulted in the formation of iron/manganese/chromium silicate reaction products. This reaction indicated both the glass and the metal were affected, and the test procedure was changed so that the entire waste form holder was subjected to a pre-test heat treatment process. The new procedure was to heat the metal at 550°C for 24 hours and to cool slowly to room temperature. This procedure was provided by the corrosion group of NNWS and was based on the time, temperature, sensitization (TTS) diagram for 304L stainless steel [BINDER]. The procedure was tried on a sample of the 304L stainless steel stock (heat 699960) and complete sensitization of the metal was found.

All of the waste form holders used in the N2 test (composition given in Table 7) were sensitized simultaneously according to procedure NNWSI-05-013. The resulting metal had a gray-blue to dark blue color in most areas, with some sections of the holders appearing gold in color. Color photographs of all the heat-treated metal sections were taken prior to testing. It is important to note that the coloration on all the samples was not identical. It was not determined prior to testing to what degree the color of the specimen could quantitatively be used to predict the

Table 6. Analyses of Equilibrated J-13 Water and J-13 Water used in Unsaturated Testing

Date	Concentration ($\mu\text{g/L}$, ppb)																		
	Al	B	Ba	Be	Ca	Co	Cr	Cu	Fe	Li	Mg	Mn	Mo	Na	Ni	Si	Sr	Ti	V
Zn																			
U																			
F ⁻																			
Cl ⁻																			
NO ₃ ⁻																			
SO ₄ ²⁻																			
Detection Limits ^a	50	30	10	5	100	20	10	10	10	20	5	5	30	100	20	100	10	5	20
J-13 Water ^b																			
11/22/85	50	130	ND	ND	11800	ND	ND	ND	<10	40	1900	<5	ND	44000	20	28700	39	ND	ND
11/21/86	50	130	ND	ND	12000	ND	ND	ND	<10	41	1920	<5	ND	46100	20	29300	32	ND	ND
Equilibrated J-13 Water																			
8/13/84	700	100	<10	<5	4500	20	<10	<10	10	64	300	<5	<30	40000	20	30300	30	<5	20
12/19/84	720	200	ND ^c	ND	4550	ND	ND	ND	ND	75	415	ND	ND	47000	92	37900	39	ND	ND
5/29/85	600	100	ND	ND	4550	ND	ND	ND	ND	70	420	ND	ND	49000	57	38000	44	ND	ND
6/26/85	600	170	ND	ND	4300	ND	ND	ND	<10	79	400	<5	ND	48200	ND	37800	47	ND	ND
11/22/85	720	100	ND	ND	4370	ND	ND	ND	<10	71	400	<5	ND	48500	ND	36900	41	ND	ND
5/14/86	750	100	ND	ND	4150	ND	ND	ND	17	60	390	<5	ND	47000	ND	36200	41	ND	ND
8/05/86	710	170	ND	ND	4100	ND	ND	ND	<10	71	390	<5	ND	48000	ND	36900	37	ND	ND
11/18/86	700	100	ND	ND	4100	ND	ND	ND	<10	79	400	<5	ND	48700	ND	36200	37	ND	ND
Calculational Values ^d	179				4255				73	400				40276		37172	39		4.0

^aDetection limits are reported as 3X noise, all numbers have been rounded to at most three significant figures.

^bJ-13 water is used only as the starting water for equilibration. Procedures governing the Unsaturated Test place specifications only on equilibrated J-13 water. However, the original batch of J-13 water, received 5/23/84, has been periodically analyzed following approved analytical procedures.

^cND = not determined.

^dThe calculational value is the composition of EJ-13 used in later calculations. This value is derived by averaging the unrounded values presented in this table for the period of analysis from 5/29/85 to 11/18/86. This is true except for Sr for which the time spanned 11/22/85 to 11/18/86.

Table 7. Composition of 304L Stainless Steel Used in the N2 Unsaturated Test Series

Component	Heat #	Element %											
		C	Mn	P	S	Si	Cr	Ni	Cu	Co	N	Mo	V
Vessel Tops	0A2682	0.020	1.87	0.025	0.026	0.27	18.05	10.01	0.28	0.15	0.058	0.41	0.12
Vessels Bottoms	9A1929	0.013	1.39	0.028	0.018	0.300	18.23	8.86	0.31	0.10	0.078	0.27	NA
Retainer N2	699960	0.022	1.49	0.025	0.003	0.40	18.22	10.02	0.12	NA	0.036	0.24	NA
Posts	70815	0.027	1.61	0.028	0.021	0.26	18.50	10.62	NA	NA	NA	NA	NA
Retainer P-VIII	22841	0.016	1.33	0.029	0.003	0.51	18.15	9.22	0.36	0.11	0.082	0.28	NA

NA = not reported in certificate of analysis.

degree of sensitization, if indeed, color was related to sensitization at all. However, subsequently it was demonstrated that if the specimen was blue after heat treatment, sensitization was likely to be present, while if it was gold, little sensitization had occurred.

The TTS diagram for 304L stainless steel indicates sensitization will occur between ~ 550 and 850°C if the metal (>0.03 wt % C) is held at temperature for between ~ 1 and 48 hours. The conditions used for sensitization in the present test are near the sensitization boundary and were chosen in an effort to impart a partial degree of sensitization to the metal. Subsequent experimentation, using a different temperature, $535 \pm 10^{\circ}\text{C}$ for 24 hours, indicated that no sensitization was imparted to the metal (Fig. 3c). Additionally, the heat treated yet unsensitized metal components were completely gold in color. Thus, it is possible, due to temperature variations and compositional inhomogeneities in the metal, that different degrees of sensitization were imparted to the retainer sections used in the N2 Test series. To determine whether this was the case the retainers, after undergoing other surface analytical studies, were cleaned and etched to examine the extent of sensitization present in each individual component. The results of these post-test studies are discussed in Section V.

III. RESULTS AND DISCUSSION

The N2 Unsaturated Test series has been in progress for over 65 weeks. The results of batch tests completed through 52 weeks are presented in conjunction with the results of continuous tests completed through 65 weeks.

A. N2 Unsaturated Test

The detailed N2 Test matrix, together with selected results, is presented in Table 8. Tests N2-1 through N2-8 are the batch tests, and tests N2-9 through N2-12 are the continuous tests. Three types of results are presented: general observations, solution analyses, and component analyses. A combination of each type of result is required to provide a concerted description of the reaction process. Eventually, the results presented here will be combined with a modeling effort to predict the long-term interactions in the system observed in this Test.

1. General Observations

Unlike the F Test series, where there was a marked disparity in the post-reaction appearance of the components from individual tests, the components from the N2 Test series were quite similar in appearance. General observations were made of the test components prior to, during, and after testing. These observations included color photographs of the pre- and post-test retainer and glass samples and visual appearance of the test components during the sampling periods.

(a)

(b)

(c)

(d)

— ■ 20 μm

Fig. 3. Micrographs of Polished Cross Sections of Etched Stainless Steel (a) N2 #4 bottom canister treated at 550°C; (b) N2 #8 top canister treated at 550°C; (c) retainer section treated at 535°C; and (d) "ditch" appearance generated by more rigorous heat treatment conditions. All micrographs are 375X and the scale bar equals 20 μm .

Table 8. Test Matrix and Component Weights for N2 Test Series

Description	Test #	Vessel #	Oven Position	Test Period (weeks)	Date Started	Date Stopped	Sample #	Weight Glass IN (gm)	Weight Glass OUT (gm)	Δ Mass (μ g)	SA Glass (mm ²)
Batch	N2-1	ANL-36	3	13	2/03/86	5/05/86	23	10.56620	10.56606	(140)	1389
Batch	N2-2	ANL-37	4	13	2/03/86	5/05/86	24	10.21957	10.21915	(420)	1359
Batch	N2-3	ANL-38	5	26	2/03/86	8/04/86	31	10.32225	10.32131	(940)	1367
Batch	N2-4	ANL-39	6	26	2/03/86	8/04/86	32	9.85644	9.85644	(880)	1317
Batch	N2-5	ANL-40	7	39	2/03/86	11/03/86	33	10.19810	10.19689	(1210)	1363
Batch	N2-6	ANL-41	8	39	2/03/86	11/03/86	34	10.20494	10.20453	(410)	1354
Batch	N2-7	ANL-42	9	52	2/03/86	1/31/87	111	10.59013	10.58971	(420)	1384
Batch	N2-8	ANL-43	10	52	2/03/86	1/31/87	131	10.82178	10.39635	(440)	1376
Continuous	N2-9	ANL-34	1	6.5	2/03/86	3/20/86	21	10.27493			1364
		ANL-45		13	3/20/86	5/05/86					
		ANL-34		19.5	5/05/86	6/19/86					
		ANL-45		26	6/19/86	8/04/86					
		ANL-34		32.5	8/04/86	9/18/86					
		ANL-45		39	9/18/86	11/03/86					
		ANL-34		45.5	11/03/86	12/18/86					
		ANL-45		52	12/18/86	1/31/87					
		ANL-34		65	1/31/87	5/04/87					
		ANL-45		78	5/04/87	8/03/87					
Continuous	N2-10	ANL-35	2	6.5	2/03/86	3/20/86	22	10.46979			1386
		ANL-46		13	3/20/86	5/05/86					
		ANL-35		19.5	5/05/86	6/19/86					
		ANL-46		26	6/19/86	8/04/86					
		ANL-35		32.5	8/04/86	9/18/86					
		ANL-46		39	9/18/86	11/03/86					
		ANL-35		45.5	11/03/86	12/18/86					
		ANL-46		52	12/18/86	1/31/87					
		ANL-34		65	1/31/87	5/04/87					
		ANL-46		78	5/04/87	8/03/87					

Cont'd

Table 8 (Cont'd)

Top Canister IN (gm)	Top Canister OUT (gm)	Δ Mass (μ gm)	Bottom Canister IN (gm)	Bottom Canister OUT (gm)	Δ Mass (μ gm)	Total Vessel IN (gm)	Total Vessel OUT (gm)	Δ Mass (gm)	Water Added During Testing (mL)
3.13942	3.14933	910	4.10280	4.10391	1110	1349.4	1341.7	1.3	1.95
3.17688	3.17758	920	4.08610	4.08729	1190	1335.6	1336.9	1.3	1.95
3.14779	3.14869	900	4.07221	4.07326	1050	1338.4	1340.8	2.4	3.90
3.16517	3.16609	920	4.09671	4.09671	880	1335.8	1338.8	3.0	3.90
3.17319	3.17431	1120	4.08299	4.08452	1530	1330.0	1334.0	4.0	5.85
3.17702	3.17800	980	4.12878	4.12988	1100	1339.1	1343.1	4.0	5.85
3.15554	3.15643	890	4.07967	4.08089	1200	1329.6	1335.2	5.6	7.80
3.13933	3.13933	1130	4.10057	4.10182	1250	1329.1	1335.6	6.5	7.80
3.15980			4.09704			1336.5	1337.2	+0.7	0.975
						1329.4	1330.0	+0.6	0.975
						1336.5	1337.2	+0.7	0.975
						1329.5	1330.2	+0.7	0.975
						1338.7	1337.8	+1.1	0.975
						1329.5	1330.2	+0.7	0.975
						1336.6	1337.1	+0.5	0.975
						1329.4	1229.9	+0.5	0.975
						1336.4	1337.9	+1.5	1.95
						1329.3			
3.16255			4.12307			1337.9	1338.5	+0.6	0.975
						1341.4	1342.1	+0.7	0.975
						1338.0	1338.6	+0.6	0.975
						1341.6	1342.2	+0.6	0.975
						1338.0	1338.8	+0.8	0.975
						1341.6	1342.3	+0.7	0.975
						1338.0	1338.7	+0.7	0.975
						1341.5	1342.2	+0.7	0.975
						1337.9	1339.4	+1.5	1.95
						1341.6			

Cont'd

Table 8 (Cont'd)

Description	Test #	Vessel #	Oven Position	Test Period (weeks)	Date Started	Date Stopped	Sample #	Weight Glass IN (gm)	Weight Glass OUT (gm)	Δ Mass (μ g)	SA Glass (mm ²)
Blank	N2-11	ANL-44	11	6.5	2/03/86	3/20/86	none				
		ANL-44		13	3/20/86	5/05/86					
		ANL-44		19.5	5/05/86	6/19/86					
		ANL-44		26	6/19/86	8/04/86					
		ANL-44		32.5	8/04/86	9/18/86					
		ANL-44		39	9/18/86	11/03/86					
		ANL-44		45.5	11/03/86	12/18/86					
		ANL-44		52	12/18/86	1/31/87					
		ANL-44		58.5	1/31/87	3/19/87					
		ANL-44		65	3/19/87	5/04/87					
		ANL-44		78	5/04/87	8/03/87					
Continuous	N2-12	ANL-36	3	6.5	5/06/86	6/19/86	132	10.94878		1384	
		ANL-37		13	6/19/86	8/04/86					
		ANL-36		19.5	8/04/86	9/18/86					
		ANL-37		26	9/18/86	11/03/86					
		ANL-36		32.5	11/03/86	12/18/86					
		ANL-37		39	12/18/86	1/31/87					
		ANL-36		45.5	1/31/87	3/19/87					
		ANL-37		52	3/19/87	5/04/87					
		ANL-36		65	5/04/87	8/03/87					
		ANL-36									
		ANL-36									

Cont'd

Table 8 (Cont'd)

Top Canister IN (gm)	Top Canister OUT (gm)	Δ Mass	Bottom Canister IN (gm)	Bottom Canister OUT (gm)	Δ Mass	Total Vessel IN (gm)	Total Vessel OUT (gm)	Δ Mass (gm)	Water Added During Testing (mL)
3.16770						1320.0	1320.6	+0.6	0.975
						1328.0	1328.7	+0.7	0.975
						1328.0	1328.8	+0.8	0.975
						1328.0	1328.6	+0.6	0.975
						1328.1	1328.9	+0.8	0.975
						1328.5	1329.0	+0.5	0.975 ^a
						1328.3	1328.6	+0.3	0.975 ^a
						1320.0	1320.3	+0.3	0.975 ^a
						1320.0	1320.7	+0.7	0.975
						1320.1	1320.5	+0.4	0.975
						1320.0			
			4.09051			1348.4	1349.1	+0.7	0.975
						1336.1	1336.7	+0.6	0.975
						1340.5	1341.1	+0.6	0.975
						1336.1	1336.8	+0.7	0.975
						1340.5	1341.2	+0.7	0.975
						1336.1	1336.8	+0.7	0.975
						1340.5	1341.4	+0.9	0.975
						1336.2	1336.8	+0.6	0.975
						1340.6			

^aInlet line plugged during the test period.

The as-prepared glass specimens met the appearance requirements given in procedure NNWSI-05-005. The as-cut sections were uniform in appearance and the sides were reflective and smooth. After testing, the top and bottom surfaces were noticeably reacted as shown in Fig. 4. The majority of the reaction products were found surrounding the circumference of the holes in the retainer sections. This is where standing water was observed in some of the tests upon opening the vessels. The regions of glass/metal contact showed little evidence of coverage by alteration products except in some localized cases where channels appeared between open areas. The extent of coverage by the rust-colored alteration products appeared to increase between the 13- and 26-week samples, but was fairly constant thereafter.

The side surfaces of the glass retained their reflective appearance, but were covered with random watermarks as shown previously (see Fig. 2 of [BATES-2]). This appearance suggests that some water flowed over the sides but that the strong interactions present on the top and bottom surfaces did not occur on the sides. During the sampling and termination periods, water was observed standing on the bottom retainer section at the interface between the glass and the metal. Only in a few samples was standing water observed on the top surface of the glass in the holes of the retainer. It is believed that water collects in this area during the test but evaporates during the cool down period before the vessels are opened. Condensed water is always observed on the inner sides and tops of the vessels.

Fig. 4. Photographs of the Reacted Top Glass Surfaces from N2 Tests (a) N2 #1 and (b) N2 #5, Magnification ~4X.

The pre-test appearance of the top and bottom retainer sections ranged from a uniform deep blue to a mix of blue and gold. No measure of the extent of sensitization was made on the actual test specimens prior to testing. Such studies were done only after the test periods were complete (Section 4).

After testing, the top and bottom retainers were discolored in the glass contact region, attaining a dark brown to rust-colored appearance. The interface where the glass contact ended was clearly marked by a buildup of precipitates. However, the extent of precipitate coverage did not visually increase during the 52-week testing period. This observation agrees with the measured weight gains of the top and bottom canister sections which are constant throughout the 52-week test period.

2. Solution Analysis

The test solutions were analyzed either at the end of a test period or, for the continuous tests, at the sampling period. At the termination of a test the WPA was rinsed with 10 mL of high purity water, while for the continued tests, only the water collected in the bottom of the vessel was analyzed. Thus, the terminated tests contained a rinsed component that was not present in the continuous tests. A blank test was also run in the continuous mode. The purpose of this test was to evaluate whether any cross-contamination of radionuclides occurred during sample handling and to serve as a check of the EJ-13 water composition collected during a test period. The solutions were analyzed for components of the glass frit, the EJ-13 water, and for radionuclides.

The as-analyzed solution compositions are presented in Appendix II. These tables present the total amount of each element detected in solution and the total volume of solution measured in the vessel upon sampling. The sampled volume differs from the total amount of liquid added during testing. This difference could be due to evaporative losses during handling or losses that occurred during injection. Analyses of the blank solutions suggest that most of the water loss occurred due to evaporation. This is because the amount of Na, Ca, and Mg detected in solution was near that predicted from the amount of EJ-13 water injected during a test period. For Si, the measured amount is usually less than predicted. The reason for this is not known, but the same observation was made during the F Test series.

The Ca and B levels in the blank solutions were larger than the input EJ-13 values for some sampling periods. This probably resulted from leaching of the ethylene propylene gasket material, which during pre-test experimentation was shown to release small amounts of these elements when soaked in water. At no time were any significant amounts of U, ^{237}Np , ^{239}Pu , or ^{241}Am detected in the blank solutions, indicating that a minimal risk of cross-contamination existed due to handling procedures.

The raw solution results can be used in two ways: (1) after correction for components present in the EJ-13 water, the extent of glass reaction can be monitored, and (2) for the continuous tests, an estimate of the composition of the water in contact with the WPA can be made. This compositional information, in combination with the identification of alteration products formed on the test components, can be used to describe the interactions that occurred during the test.

a. Glass Reaction

The extent of glass reaction is best measured by monitoring elements whose presence in solution can be associated mainly with the glass, as opposed to the EJ-13 water or the metal components of the system. Such elements include Li, U, ^{237}Np , ^{239}Pu , ^{241}Am , and B. Boron is present in the starting EJ-13 water at an insignificant level. As noted previously, some B may be introduced into solution due to interaction with the gasket material, however, the data are presented assuming that most of the B comes from the glass.

Blank-corrected solution results, given in Table 9, lead to the following observations:

- (1) All elements except Si show a net release from the glass in the batch tests. Silicon shows a net release in Tests N2 #3, 4, and 5, but is relatively deficient in solutions from the remaining tests.
- (2) An intercomparison of the batch test results indicates an apparent trend of increasing reaction through 26 weeks, with little additional release to solution thereafter. This trend is supported by a consistency in the release of all elements. The trend may be real, but more likely, when interpreted in conjunction with all the test data, is skewed by increased reaction in tests N2 #3, 4, and 5. The continuous tests give a better representation of temporal trends in element release than the batch tests do as described in Section IV.
- (3) Normalized release values (Table 10 and Figs. 5 and 6), calculated to compare the release behavior of different elements, indicate the alkali elements, Li and Na, are released from the glass at nearly equivalent rates which are larger than that calculated for B or the actinide elements. Note the normalized release of B is nearly equivalent to that of Np which is greater than U which is greater than Pu and Am. These plotted releases are calculated based on the total surface area of the glass and on the measured amount of each element released from the WPA after rinsing. However, if the amount of Pu, Am, and U remaining with the metal components of the WPA is measured via complete stripping (Table 10) of the metal, and it is assumed that the release of these elements is associated mainly with the glass that is in contact with metal, then modified $(NL)_{\text{Pu,Am,U}}$ values can be calculated which are much nearer to the values of B and Np.

Table 9. Blank Corrected Solution Results^a from the N2 Tests

Test #	Duration (weeks)	Total Elemental Release to Solution, gm x 10 ⁻⁹										gm x 10 ^{-8b}			gm x 10 ⁻¹¹	
		B	Ca	Li	Mg	Na	Si	Sr	U			237Np	239Pu		241Am	
N2-1	13	6100	14900	17700	900	48000	(47000)	130	120			6.2	22.7		6.8	
N2-2	13	13400	29700	24300	3200	95100	(79000)	440	700			13.9	40.4		14.9	
N2-3	26	37500	53800	54300	8100	207600	144800	1200	2300			43.6	63.7		18.4	
N2-4	26	34800	61300	52100	6200	211900	153000	1200	2000			62.0	73.0		22.9	
N2-5	39	38800	54100	62800	6800	224400	142000	1400	4800			42.3	64.2		16.6	
N2-6	39	23500	44200	36400	3100	120400	(41600)	600	400			14.3	22.0		6.4	
N2-7	52	21700	36300	50600	3200	204800	(41300)	800	2000			19.4	37.7		10.4	
N2-8	52	24300	28500	54700	3400	222400	(28800)	800	1300			26.7	70.0		12.7	
N2-9	13	7700	(600)	15400	(400)	24800	(54900)	140	70			3.9	12.3		3.1	
	26	13400	18200	25500	1100	39700	(105300)	400	300			11.8	18.8		5.1	
	39	15700	30400	33100	2300	62400	(88800)	600	700			15.8	22.7		6.3	
	52	23700	43300	41700	4200	107800	(122400)	950	2200			30.8	26.5		9.0	
	65	29200	45400	46900	4700	105700	(163600)	1040	2350			33.5	27.9		9.6	
N2-10	13	6500	(6000)	300	(500)	(83800)	(69000)	140	10			0	0		0	
	26	9000	8000	15800	100	(30500)	(106600)	290	140			3.0	5.6		2.0	
	39	11300	24600	24700	1200	(3100)	(140700)	460	640			6.3	9.5		3.8	
	52	13500	27800	29700	1600	(22400)	(197100)	570	1080			8.6	11.6		4.6	
	65	16100	36500	36600	2400	39600	(231300)	740	1400			10.9	13.4		5.1	
N2-12	13	7200	(6700)	9400	(800)	(30200)	(50400)	180	50			2.9	22.4		4.8	
	26	8500	(4600)	17700	(1200)	(31200)	(101500)	240	80			4.6	29.1		6.9	
	39	9500	8000	29000	(700)	27700	(129300)	380	230			7.3	35.9		8.3	
	52	11300	14300	36300	(400)	59100	(154500)	480	430			10.1	39.6		9.3	

^aNumbers in parentheses ( ) indicate less of the element was present in the test solution than in EJ-13 water used during the test. Blank corrections were made by subtracting from the total of element i in solution (Appendix AII-1) the total amount of element i added during the test period. The amount of element i added during the test period is the sum of the water injected plus the 0.5 m³ added at the beginning of each period. The concentrations for each element used for background subtraction are given in the table. These concentration values are the average of the unrounded numbers present in Table 6 for the periods of analysis from 5/29/85 to 11/18/86.

^bThe meaning of this notation is that the number in the column (6.2 for entry #1) is 6.2 x 10⁻⁸ gm. This meaning is used in all subsequent tables.

Table 10. Normalized Release^a (g/m²) from the N2 Unsaturated Tests

Test #	Duration (weeks)	Solution Only						Solution + Acid Strip			
		(NL)Li	(NL)B	(NL)Ns	(NL)Np	(NL)Pu	(NL)Am	(NL)U	(NL)P ₂	(NL)Am	(NL)U
N2-1	13	0.7	0.2	0.4	0.2	0.008	0.008	0.01			
N2-2	13	0.9	0.5	0.9	0.4	0.015	0.018	0.06	0.3	0.4	2.9
N2-3	26	2.0	1.3	1.9	1.3	0.024	0.022	0.21			
N2-4	26	2.0	1.3	2.0	2.0	0.028	0.028	0.19	0.7	1.1	3.0
N2-5	39	2.4	1.4	2.1	1.3	0.024	0.020	0.42			
N2-6	39	1.4	0.8	1.1	0.4	0.008	0.008	0.004	0.5	0.7	2.3
N2-7	52	1.9	0.8	1.9	0.6	0.014	0.012	0.18			
N2-8	52	2.0	0.8	2.1	0.8	0.026	0.015	0.12	0.6	0.7	2.6
N2-9	13	0.6	0.3		0.1	0.004	0.004	0.01			
	26	1.0	0.5		0.4	0.007	0.006	0.03			
	39	1.2	0.6		0.5	0.008	0.008	0.06			
	52	1.6	0.8		0.9	0.009	0.011	0.20			
	65	1.8	1.0		1.0	0.010	0.011	0.21			
N2-10	13	0	0.2		0	0	0	0			
	26	0.6	0.3		0.1	0.002	0.002	0.01			
	39	0.9	0.4		0.2	0.003	0.004	0.06			
	52	1.1	0.5		0.2	0.004	0.005	0.10			
	65	1.4	0.6		0.2	0.005	0.006	0.12			
N2-12	13	0.3	0.2		0.1	0.008	0.006	0			
	26	0.7	0.3		0.1	0.011	0.008	0.01			
	39	1.1	0.3		0.2	0.013	0.010	0.02			
	52	1.3	0.4		0.3	0.015	0.011	0.04			

^aThe entire surface area of the glass was used to calculate (NL)_i for the Solution Only results, while only the surface area of the end sections of the glass were used in the Solution + Acid Strip calculations. Normalized release = $M_i / (SA \cdot f_i)$ where M_i is the total measured release of element i , SA is the surface area as described above, and f_i is the mass fraction of element i in the bulk glass.

Fig. 5a. Normalized Release of (Δ) Li, (□) Na, (○) B, and (◇) wt loss for the N2 Batch Tests.

Fig. 5b. Normalized Release of (○) Np, (□) U, (▽) Am, (Δ) Pu, and (◇) wt loss for the N2 Batch Tests.

Fig. 6a. Normalized Release of Li for Three Continuous Tests in the N2 Test Series

Fig. 6b. Normalized Release of (O) Np, (◇) U, (▽) Am, and (Δ) Pu for the N2 Continuous Tests (average of three tests).

In the continuous tests, where the components have not been rinsed before the solution is analyzed, several elements, including Ca, Mg, Na, and Si, are depleted in the analyzed solution compared to the starting EJ-13 water. Normalized release values for Li in the continuous tests are plotted in Fig. 6. While these values should be slightly lower than the batch tests due to the lack of a rinse component, it is seen that there is continuous release from the glass through the 65-week time period. For Li, the element that apparently is least affected by interaction with test components, the normalized release values after 52 weeks of testing are highlighted in Table 11. The values for the continuous tests have been calculated assuming that 7000 ngm of Li will be rinsed from the WPA to make the continuous values comparable with the batch tests. The 7000 value is taken from measured differences between batch and continuous tests in the F-1 Test series [BATES-1]. The consistency of these values through 52 weeks, combined with the continually increasing reaction trend observed in the continuous tests, suggests that the plateau in reaction progress seen in the results from the batch tests is due to a physical effect that accelerated the reaction in tests N2, 3, 4, and 5 compared with the other tests. This possibility will be discussed later.

b. Solution Composition

The composition of the solution that is in contact with the WPA can be estimated from the results of the continuous tests. This solution composition is important because it is the interaction between this solution and the waste package components that results in the formation of alteration products that are more stable than the glass. In the continuous tests, the volume of water collected in the test vessel after it has contacted the WPA is measured and its composition is determined. While the process by which the water leaves the WPA is a combination of evaporation

Table 11. Normalized Lithium Release
through 52 Weeks of
Testing

Test #	(NL) _{Li} , (g/m ²)
N2 #7	1.9
N2 #8	2.0
N2 #9	1.8
N2 #10	1.4
N2 #12	1.6

and dripping, the water collected in the test vessel should be nearly representative of the water on the WPA. Indeed, it is unlikely that the composition of the water that collects on the top and bottom sections of the WPA has exactly the same composition due to the flow characteristics of the test. Regardless, the elemental concentrations in the continuous tests are given in Table 12 and Fig. 7. These values have been calculated assuming that all the water injected during a test period has contacted the WPA, reacted, and been released and collected in the test vessel. Thus, the solution in the test vessels consists of injected EJ-13 water that has reacted with the WPA and 0.5 mL of EJ-13 water that was placed in the bottom of the test vessel before the test began and at each sampling period. The elemental concentrations are determined by subtracting the volume and compositional effect of the initial EJ-13 water from the analyzed results and then dividing the remaining elemental values by the volume of injected liquid. Values have been combined for two 6.5-week

Table 12. Measured Elemental Concentrations of Test Solution in Contact with Glass during the Unsaturated Test^a

Test #	Sampling Period (weeks)	Concentration ($\mu\text{gm/mL}$)							
		B	Ca	Li	Mg	Na	Si	Sr	U
Initial EJ-13		0.2	4.5	0.1	0.4	48	37	0.04	0.004
N2-9	13	4.1	4.0	8.0	0.2	61	9.0	0.1	0.04
	26	3.1	13.9	5.2	1.2	56	11	0.2	0.12
	39	1.3	10.6	4.0	1.0	60	46	0.1	0.21
	52	4.2	10.8	4.5	1.4	71	20	0.2	0.77
	65	3.0	5.3	2.8	0.7	47	16	0.1	0.08
N2-10	13	3.5	1.2	0.2	0.2	5.3	1.8	0.1	0
	26	1.5	11.4	8.0	0.7	76	18	0.1	0.07
	39	1.3	12.8	4.6	0.9	62	20	0.1	0.26
	52	1.3	5.8	2.6	0.6	38	8.2	0.1	0.22
	65	1.5	8.8	3.6	0.8	75	20	0.1	0.17
N2-12	13	3.9	0.8	4.8	0	33	11	0	0.03
	26	0.9	5.3	4.3	0.2	48	11	0.1	0.02
	39	0.8	10.7	5.8	0.7	79	23	0.1	0.08
	52	1.0	7.5	3.9	0.6	64	24	0.1	0.11

^aThe concentration is calculated by taking the total amount of element i in solution (Appendix AII-1) for a given 13-week period and subtracting the amount of element i present in the EJ-13 water that is added to the test vessel at the beginning of the test periods (e.g., 1 mL for the 13-, 26-, 39-, and 52-week values, 0.5 mL for the 65-week value). The resultant value for element i is then divided by the amount of EJ-13 water injected during a 13-week period (1.95 mL).

Fig. 7. Log of the Concentrations of
 (□) Na, (◇) Si, (▽) Ca,
 (△) Li, and (○) B in the
 Test Solutions for the Three
 Continuous Tests in the N2
 Unsaturated Test Series.

periods because of inconsistent release from the WPA for one 6.5-week period. For example, in the N2-10 test no liquid dripped from the WPA during the first two test periods (ref. Li and actinide solution values), although the WPA was visually reacted to the same degree as the other tests.

While there is scatter in the values for the individual tests, the average elemental concentration, except for Li, are nearly constant for at least the last two sampling periods. Lithium shows a gradual decrease in concentration throughout the testing cycle.

The concentrations of the transuranic elements were determined in the batch tests for the unfiltered and filtered solutions. The unfiltered solutions were sampled directly upon opening the test vessels while the filtered samples were taken from the rinse solutions and thereby contained both an added actinide fraction from the WPA and an additional 10 mL of DIW. The unfiltered fraction is representative of a truly soluble component, plus material suspended in solution (e.g., colloids, etc.) after it has left the WPA and been consolidated in the vessel. The filtered fraction represents what is actually in solution after rinsing. Because of the potential for changing speciation and solubility of the actinide component during rinsing, the solution soluble fraction may bear no relationship to repository conditions. No fraction of the non-rinse solutions was collected for filtering because of lack of solution volume.

As shown in Table 13 the unfiltered component varies randomly throughout the test periods by about a factor of ten. The order as detected in solution is $Np > Pu > Am$. For the filtered samples the detected values are quite constant with time.

3. Analysis of Test Components

To characterize the interactions that have occurred during testing, the components have been examined using SEM/EDS and XRD. The purpose of such characterization is to (1) in combination with the solution results, provide a description of the reaction progress; and (2) to catalog and identify, if possible, alteration products that form during the course of the reaction. Both of these issue factors must be quantified in order to project the performance of the waste form to repository time frames.

At the end of each batch test period, the components were separately rinsed with high-purity water and placed in a dust-free environment to dry. The weight change for each component was determined and then components from selected tests (N2 #s 2, 4, 6, and 8) were analyzed. The components from the remaining tests (N2 #s 1, 3, 5, and 7) were maintained in storage to be analyzed using additional techniques as they become available. The methods and rationale used to perform and interpret the SEM/EDS analyses have been described previously [BATES-1, -2]. The specimens were photographed using a low resolution optical microscope in order to map the surface and to identify regions of interest. The glass samples were then examined using SEM/EDS to obtain a qualitative description of the phases present. Micrographs and EDS spectra were taken extensively. After removal from the SEM, phases with unique composition, and crystalline structure, and of suitable size were picked from the surface and analyzed using x-ray powder diffraction analysis.

SEM/EDS can be used to provide a description of the phases and alteration products, but cannot be used for absolute identification. Additionally, after exposure to the coating process and vacuum conditions of the SEM, the sample may be altered to make examination by other spectroscopic methods, such as infrared or Raman spectroscopy, difficult, if not misleading. For this reason, not all of the samples were immediately analyzed.

The canister components were also photographed and examined via SEM/EDS and XRD. They were then acid-stripped to remove all residual radioactivity, mounted in epoxy, polished, and subjected to an oxalic acid etch process to measure the extent of sensitization that occurred during the heat treatment step.

These component analyses have resulted in an extensive collection of micrographs and spectra. In the following discussions, only a representative selection of these analyses will be presented to provide a description of the reaction process. A complete compilation of the data set is available for review at ANL.

Table 13. Concentration (moles/liter) of Np, Pu, and Am Associated with Test Solutions in the N2 Test

Test # N2	Duration (weeks)	Np		Pu		Am	
		Unfiltered	Filtered	Unfiltered	Filtered	Unfiltered	Filtered
#1	13	3E-8	3E-9	2E-9	4E-12	4E-11	1E-13
#2	13	2E-7	1E-8	7E-10	<1E-12	9E-12	2E-13
#3	26	5E-7	3E-8	3E-9	5E-12	7E-11	5E-13
#4	26	5E-7	4E-8	2E-9	<1E-12	3E-11	2E-13
#5	39	3E-7	4E-8	3E-10	6E-12	5E-12	3E-13
#6	39	1E-7	2E-8	2E-10	3E-12	9E-12	1E-13
#7	52	9E-8	2E-8	9E-10	<1E-12	3E-11	1E-13
#8	52	1E-7	2E-8	2E-9	<1E-12	5E-11	<5E-14

4. Glass

The weight change measurements for the glass samples are given in Table 8. All samples lost weight, although, as shown in Fig. 4, the glass samples were covered extensively with reaction products. The weight loss normalized to the surface area of the specimens, $(NL)_{wt}$, closely followed the reactivity of the glass as determined by the solution results (see Figs. 5a,b), i.e., those tests in which a large weight loss was observed also had a concomitant release of glass components to solution. The $(NL)_{wt}$ in all tests was about 50% the value for $(NL)_B$.

All of the glass surfaces (top and bottom) had the general appearance shown in Fig. 4. The top surfaces had the appearance of being slightly more reacted than the bottom mainly due to an apparently greater coverage of the rust-colored precipitates and the lighter appearance of the glass where there was contact between glass and metal. This appearance of reduced reaction likely resulted because the interface between the glass and the metal was better maintained on the bottom section due to the weight of the glass, while a looser interface existed at the top. However, the nature of the reaction products was the same for both the top and bottom sections.

The greatest extent of precipitate buildup occurred around the circumference of the holes where the metal contacted the glass. Precipitate buildup typical of the non-metal contact regions of the glass is shown in Fig. 8. The semi-circular regions are where standing water collected during the test, and the bulk of the precipitates in these regions were composed of Fe or Fe/Ni/Cr/Mn (Fig. 9a) combined with Si. The next most prominent phase, as determined by SEM/EDS, consisted of Si with lesser amounts of Al and Fe (Figs. 10 and 11). The compositions of the Fe-rich alteration products, varied over a range between pure Fe and Fe-enriched Si/Al. XRD (Table 14) of the main regions of precipitate buildup (Fig. 8) indicated both ferrihydrate ($5Fe_2O_3 \cdot 7H_2O$) and $FeO(OH)$ to be present; however, no pattern was obtained that could be attributed to the Si/Al phase.

In addition to these prominent regions of precipitate buildup, other alteration products were located in a random fashion over the glass surface. In several instances, only a limited number of each phase was found on the surface. In many cases, the phases were of micron size and were too small for XRD analysis, and only SEM/EDS information has been collected to date. Several representative micrographs of alteration products are shown in Figs. 9b-d, 12, and 13. Note, that in many cases, Cl and S are identified in the EDS spectra as associated with the alteration products (Fig. 13).

A few of the precipitated phases were large enough to be identified using XRD. These included gypsum ($CaSO_4 \cdot 2H_2O$) and a solid solution of the composition $Ca_{.74}(Mn,Mg)_{.26}CO_3$ (Fig. 14a, Table 15), and a group of phases (Fig. 14b, Table 16) that was tentatively identified as containing orthoclase ($KAlSi_3O_8$), cristobalite (SiO_2), and possibly albite ($NaAlSi_3O_8$). EDS spectra are shown with the associated micrographs supporting the XRD identities (Fig. 14b,d).

Fig. 8. Photomicrograph of the Bottom Glass Surface from Test N2 #8. Magnification 19X.

———— = 1 mm

— = 10 μm

(a)

— = 10 μm

(b)

— = 20 μm

(c)

— = 40 μm

(d)

Fig. 9. Photomicrographs and EDS Spectra of Alteration Products Shown in (a) the Upper Center of Fig. 8, Magnification 500X; (b) the Upper Center of Fig. 8, Magnification 700X; (c) the Center of Fig. 8, Magnification 350X; and (d) the Lower Center of Fig. 8, Magnification 170X.

— = 10 μm

— = 10 μm

— = 1 μm

— = 1 μm

Fig. 10. Photomicrograph and EDS Spectra of Alteration Products that Formed on the Top Glass Surface from Test N2 #6, Magnification 500 and 1500X.

(a)

(c)

(b)

Fig. 11. Photomicrographs of the Top Glass Surface from Test N2 #6 (a) Magnification = 5,000X, (b) Magnification = 1,500X, and (c) EDS Spectrum of the Surface.

Table 14. XRD Results for Iron Containing Phases Formed during the Unsaturated Test

Observed XRD		FeOOH		Ferrihydrate $5\text{Fe}_2\text{O}_3 \cdot 9\text{H}_2\text{O}$		d-FeOOH	
I	d	I	d	I	d	I	d
100	4.92	100	4.94				
1	4.5 vd					20	4.61
30	4.15 grease	80	2.94				
10	2.51 vd			100	2.50	100	2.545
5	2.22 vd			80	2.21	100	2.255 vd
5	1.98 vd			80	1.96		
				50	1.72	100	1.685 vd
5	1.515 vd			70	1.51		
10	1.47 vd			80	1.48	100	1.471

vd = very diffuse

— = 10 μm

— = 1 μm

(a)

(b)

(c)

(d)

Fig. 12. Photomicrographs of the Glass Surface from Test N2 #2 (a) General Region of Non ss Contact, Magnification 500X; (b) Spherical Precipitate Shown in Fig. 12a, Magnification 5,000X; (c) EDS Spectrum of Spherical Precipitates; and (d) EDS Spectrum of Irregularly Shaped Precipitate in the Center of Fig. 12a.

— = 40 μm

(a)

(b)

(c)

Fig. 13. Photomicrograph and EDS Spectra of (a) Cl-Rich Alteration Products that Form on the Top Glass Surface from Test N2 #6, Magnification 170X; (b) EDS Spectrum of White Specs on the Dark Background; and (c) EDS Spectrum of the Dark Background.

— = 10 μm

(a)

(b)

— = 10 μm

(c)

(d)

Fig. 14. (a) Photomicrograph of the Solid Solution that Formed on the Retainer Surface, Magnification 250X; (b) EDS Spectra of the Solid Solution; (c) Photomicrograph of Phases Identified as Orthoclase, Cristobalite, and Albite. EDS analysis of the top precipitate indicates a U inclusion near the lower right corner with a Si substrate, while the remainder of the precipitate gives the EDS spectrum shown in (d). The bottom precipitate is composed of orthoclase (d) and albite. Magnification is 1,250X.

Table 15. XRD Results for the Solid Solution
 $\text{Ca}_{.74}(\text{Mn},\text{Mg})_{.26}\text{CO}_3$ Plus $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$

Observed XRD		Gypsum $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$		Calcite Type $\text{Ca}_{.74}(\text{Mn},\text{Mg})_{.26}\text{CO}_3$	
I	d	I	d	I	d
30	7.6	77	7.59		
30	4.28	100	4.279		
30	4.16			10	4.13
30	3.78	13	3.795	40	3.78
30	3.07	55	3.061		
100	2.98			100	2.981
30	2.87	42	2.875		
10	2.79t	8	2.789	10	2.77
20	2.68	31	2.683		
5	2.60d	3	2.595		
5	2.49	8	2.499		
40	2.46			40	2.462
30	2.25			50	2.248
10	2.22	9	2.215		
		11	2.087		
40	2.07t	8	2.079		
		9	2.075	50	2.062
50	1.87	9	1.897	50	1.869
		8	1.878		
50	1.84			60	1.840

t = thick line, possible doublet.

d = diffuse line.

Table 16. XRD Results for the Group of Precipitates Containing Sodium Feldspar, Orthoclase, and Cristobalite

Observed XRD Pattern		(Major lines only) KAlSi ₃ O ₈ JCPDS 19-931	(Major lines) SiO ₂ Cristobalite JCPDS 11-695	(Major lines only) NaAlSi ₃ O ₈ JCPDS 20-572	
I	d	I	d	I	d
5	4.22	70	4.22		
100	4.04			100	4.05
5	3.7 vsp	80	3.77	84	4.03
5	3.45 vsp	45	3.47	61	3.75
5	3.3 vsp	100	3.331		
		65	3.24		
20	3.20			100	3.21
				8	3.18
5	2.83			14	2.84
10	2.49			20	2.485

The surface of the glass had the appearance shown in Fig. 11, consisting of the honeycombed pattern depleted in Na. This appearance existed for all time periods and was typical for regions of ss and non-ss contact. The lack of Na in the EDS spectra indicates a depletion of Na from at least the first micron of the surface.

5. Retainer Sections

As with the glass, the greatest buildup of reaction products on the retainer sections was in regions where standing water could accumulate. Such regions existed around the rims of the punched holes and at the edge of the glass/metal interface. Typical regions of reaction product buildup are shown in Fig. 15. Iron oxides and hydroxide together with an Al/Si mat (Fig. 15) were the prominent reaction products.

In all the tests, the top and bottom retainer sections gained weight. The weight gain was ~1 mg for each retainer section and was nearly constant for all test periods.

As with the glass sections, the same reaction products were not found on all retainer sections, and indeed individual phases may exist only as isolated examples on a single sample. Photomicrographs and EDS spectra of such reaction products are shown in Fig. 16 and include several apparently different U-rich phases. Also, comparatively large agglomerates with varying compositions were found scattered over both the metal and glass surfaces. These agglomerates were composed mainly of typically insoluble elements and elements that showed a net extraction from solution, e.g., Ti, Fe, Mg, Ca, Si, and gave no crystalline XRD pattern. Many regions had intense S and/or Cl peaks in the EDS spectra. Several examples are shown in Fig. 17.

After the canister sections were examined using SEM/EDS, they were acid stripped, mounted in epoxy, and subjected to an oxalic acid etch to determine the extent of sensitization for samples N2 #s 2, 4, 6, 8, a reference sample, and the canister section heat treated at 535°C for 24 hours. Typical micrographs of the metal sections are shown in Fig. 3. Different degrees of sensitization were observed for different groups of samples. Samples from tests N2 #6 and N2 #8 showed partial sensitization of the bulk metal, but significantly less sensitization near the flat surfaces of the canister that were in contact with the glass (Fig. 3b). The near-surface region where there was less complete sensitization ranges between 30 and 40 μm for these samples. Samples from tests N2 #2 and N2 #4 also showed partial sensitization of the bulk, but showed a non-sensitized gap of only 10-15 μm (Fig. 3a). Note that the gap only existed on the surfaces of the steel that had been subjected to some processing (cold working) during manufacturing and which were not altered during the component preparation procedures. Only where the edges of the retainer sections had been machined did the sensitization extend to the surface of the metal (not shown).

(a)

— = 10 μm

(b)

— = 1 μm

(c)

— = 10 μm

(d)

— = 0.5 μm

(f)

(g)

(h)

(i)

(e)

Fig. 15. Photomicrographs and EDS Spectra of Alteration Products that Form on the Retainer Sections. (a) Bottom retainer from test N2 #2, mag = 250X; (b) central region of Fig. 15a, mag = 1200X; (c) top retainer from test N2 #6, mag = 400X; (d) top retainer glass/non-glass interface region, mag = 6500X; (e) test N2 #2 bottom retainer, ss in the glass contact region; (f) platelets shown in Fig. 15b; (g) spherical precipitates in Fig. 15b; (h) irregular precipitate in Fig. 15a; and (i) clumps of precipitates in Fig. 15c.

(a)

(e)

— = 10 μm

— = 10 μm

(b)

(f)

— = 10 μm

— = 10 μm

(c)

— = 1 μm

(d)

Fig. 16. Photomicrographs and EDS Spectra that were formed on the Bottom Retainer Sections (a) U-Rich N2 #2 bottom, mag = 2500X; (b) U-rich, mag = 1000X, the white spec is U-rich; (c) U-rich analog, mag = 7000X; (d) N2 #4 bottom, mag = 3400X; (e) N2 #4 bottom, mag = 1000X; and (f) N2 #6 bottom, mag = 2000X.

Fig. 17. Photomicrographs and EDS Spectra of Amorphous Reaction Products that Form on the Retainer Sections (a) Top Retainer N2 #8, Magnification = 200X; (b) Top Retainer N2 #6, Magnification = 1600X; (c) Substrate of Precipitate Shown in Fig. 16a; (d) Substrate of Precipitate Shown in Fig. 16b, the Elemental Ratios are not Constant throughout the Precipitate; (e) Large White Inclusion in the Top Center of the Precipitate Shown in Fig. 16b.

The entire cross-section of the retainer sections heat treated at 535°C showed only random pitting that was not associated with grain boundaries, and represented non-sensitized metal (Fig. 3c).

Pre- and post-test retainer section surfaces were carefully examined to see if any inhomogeneities in the composition of the metal exists that would help explain the strong glass/metal interaction that was observed. The pre-test retainer section was a sample of punched 304L ss sheet, heat #699960, from which the N2 retainers were fabricated. The specimen was heated at 550°C for 24 hours and cooled slowly to room temperature. The heat treatment was not done simultaneously with the N2 retainers, but did follow the same procedure. The etched cross-section of the sample indicated partial sensitization as observed for tests N2 #6 and #8 was imparted to the specimen.

The general surface of the unreacted metal is shown in Fig. 18a. Note that the grains are well defined and that few Si-rich inclusions are present, in comparison with previously examined archive sections of 304L ss from heat #22841 [BATES-2]. EDS spectra were taken at several locations over the surface including the general surface area, the middle of the grain boundaries, and the region of metal just adjacent to the grain boundaries. The spectra were taken using varying electron beam conditions so as to optimize spatial resolution of the sampled area. The estimated resolution at 10 keV in ss is approximately 0.5 μ m. At this scale, the regions listed above showed no variance in elemental composition, except for possible (speculative) enrichment of Cr in the grain boundary regions.

Other regions of the unreacted surface, however, did show compositional inhomogeneities. These included the Cr-rich and Fe-depleted regions shown in Fig. 18b, and Cr-depleted areas associated mainly with physically altered regions (scratches) on the surface (not shown).

The reacted surfaces were also examined for areas of localized reaction. Many areas that appear darker in the photomicrographs (see Fig. 19g,h) are Cr enriched and may correspond to the Cr-rich regions noted in the unreacted retainer section (Fig. 18c). Note the darker areas also have more Si than the lighter areas of glass contact (Fig. 19e,f). The regions of the reacted containers that are not in contact with the glass have little Si (Fig. 19i). Also shown in Fig. 19e is a pitted area. No areas similar to this were found on the non-reacted retainer section.

In many examples, the reacted retainer surfaces show accumulation of reaction products around regions that are Cr rich. On many of the retainer sections, precipitates of the type shown in Fig. 19a were found. These precipitates gave very low count rates with EDS, which could indicate they are composed of elements lighter than Na. The EDS spectra (Fig. 19b), which is also indicative of the underlying metal surface, indicates the precipitates are composed of Ca and Na with S and Cl. During exposure to the electron beam, these precipitates are easily blown from the surface leaving the appearance shown in Fig. 19c. EDS spectra of the underlying surface indicates (Fig. 19d) a definite enrichment in Cr exists in these areas.

(a)

— = 10 μm

(b)

— = 10 μm

(c)

— = 10 μm

(d)

(e)

(f)

Fig. 18. Photomicrographs and EDS Spectra of Pre-Sensitized but Non-Reacted Surface of 304L ss, Heat 699960. (a & d) General surface, magnification - 500X; (b & c) Cr-rich inclusions, magnification = 300/1500X; (d) EDS of general surface; (e) EDS of grain region; and (f) EDS of Cr-rich inclusion shown in Fig. 18c.

a)

— = 10 μ m

(f)

(b)

(g)

— = 10 μ m

(h)

(c)

— = 10 μ m

(d)

(i)

(e)

— = 10 μ m

Fig. 19. Photomicrographs of (a) Reaction Products that Formed on the Surface of the Top Retainer Section from Test N2 #4, Mag = 2200X; (b) EDS Spectrum of Reaction Products Shown in Fig. 19a; (c) Surface under the Reaction Products Shown in Fig. 19a, Mag = 2200X; (d) EDS Spectrum of Surface Shown in Fig. 19c; (e) Top Retainer Section from Test N2 #8, Mag = 7000X; (f) EDS Spectrum of the Light Streaked Areas; (g) Top Retainer Section from Test N2 #8, Mag = 500X; (h) EDS Spectrum of the Dark Regions Shown in Fig. 19e; and (i) EDS Spectrum of ss in the Region of Non-Glass Contact.

IV. DISCUSSION

The best method of measuring the extent of glass reaction is to monitor the performance of glass matrix or modifying elements that do not interact with components in the system and which are not present in either the EJ-13 water or the metal test components to a significant degree. Lithium, boron, and neptunium are the best examples of such elements based on past experience [EDERT]. The other actinide elements, U, Pu, and Am, while not initially present in the water or metal, strongly interact with the metal and are not released into solution.

The normalized release of Li is the largest of all elements reaching $\sim 2 \text{ g/m}^2$ after one year. In the batch tests, $(\text{NL})_{\text{Li}}$ is largest in the 39-week sample N2 #5. Test N2 #6, the other 39-week sample, and tests N2 #7, #8, the 52-week samples, had Li releases about equal to that observed for 26 weeks (Fig. 5a).

For B the release is also largest in test N2 #5, but drops by about 40% in tests N2 #6, #7, and #8. Neptunium release follows the same trend as Li and B, but drops about 50% in tests N2 #6, #7, and #8.

Such release trends are not expected for glasses that react in a continuous fashion and, as discussed later, are a result of non-equivalent interaction processes. The results of the batch tests can be used to bracket the releases that may occur, but without better control over the interactions, or a larger number of samples per test period, do not provide a complete indication of the reaction trend.

A more meaningful measure of the reaction trends can be obtained from the individual continuous tests where the test components, and therefore the interactions, are constant throughout the testing period. In the continuous tests, the release of all three elements (Li, B, Np) increases with time, reaching values after 52 weeks only slightly less than observed in the batch tests. The difference in part is due to the rinse component present in the batch tests that is not included in the continuous tests.

The release trends observed in the N2 tests, e.g., discontinuous release for the batch tests, increasing release in the continuous tests, are similar to those noted in the F1 tests, and the explanation is similar; the extent of glass reaction is strongly affected by the interaction with the retainer sections. The heat treatment to which the retainer sections were subjected was intended to impart an equal and uniform degree of sensitization to all metal samples. However, as shown in Fig. 3 the heat treatment process was not constant, as intended, and slightly different degrees of sensitization were imparted to the retainers. It is not known whether slight variations in the temperature of the oven (a 20°C range in temperature was allowed in the sensitization procedure) or differences in the constitution of the metal caused this effect.

Sensitization of austenitic stainless steels is most often considered within the context of the alloy's thermal history. The literature contains considerable documentation supporting a chromium depletion mechanism where chromium carbides are formed along the grain boundaries when the alloy is heated in the range 550-850°C. Formation of these chromium-rich carbides can cause depletion of chromium (to <12%) in the immediate vicinity of the grain boundaries. The depleted material is no longer "stainless" and is quite susceptible to attack at the grain boundaries, e.g., intergranular corrosion. Both theoretical and experimental studies have shown that chromium depletion is indeed the primary cause of sensitization.

Secondary contributors to sensitization of austenitic alloys or susceptibility to intergranular corrosion are the amount and extent of cold work, the presence of the martensite phase, the grain size and the compositional variability from heat to heat, particularly of carbon and to a lesser extent the other austenite stabilizers.

There are several standard chemical tests of the pass-fail type to determine whether a material is sensitized to the extent that it is susceptible to intergranular corrosion. These chemical tests are, by their design, not quantitatively useful for determining the degree of sensitization in those samples containing only partial sensitization. Yet partial sensitization may contribute to reaction(s) occurring in laboratory-scale tests, as is apparently the case in the N2 test series.

A standard test is the oxalic acid etch test, which is used to determine if an alloy is not sensitized. Briefly, the test consists of exposing the sample to an oxalic acid solution and passing a specified amount of electric current through the test sample. The etching solution selectively attacks various carbides and produces a surface that can be classified as "step," "ditch," or "dual." If a surface has the "step" appearance (Fig. 3c), the sample is not sensitized. If it has a "ditch" appearance (Fig. 3d), considerable carbide precipitation has occurred and further testing is required to determine if the sample is sensitized; and if it has "dual" surface structure (Fig. 3a,b), it contains, to some extent, both "step" and "ditch" features and is classified as not sensitized. However, it is obvious from examining samples with "dual" structure that some carbide precipitation has occurred and that the sample is indeed partially sensitized. However, it is not possible to quantify the degree of sensitization with this method. In this partially sensitized condition, there exist grain boundaries with continuous chromium carbides, or more likely $M_{23}C_6$, where up to 25% of M can be Fe, as well as boundary regions where carbide particles exist as small clusters in a discontinuous configuration. It is likely that some of the boundary region displaying continuous carbide formation has discrete areas where chromium depletion has reduced the chromium content to below 12%. It is likely that if these samples were exposed to a reactive environment, some corrosion would occur in the regions of chromium depletion. This reaction would not be the extensive intergranular corrosion reaction that would be exhibited by a fully sensitized material, but a more restricted, localized corrosion reaction. This is because the passive condition of the alloy has been disturbed due to chromium depletion.

The use of partially sensitized stainless alloys provides for localized sites for reaction to occur. However, previous workers [BRIANT] have noted that the partial sensitization is very sensitive to local variations in carbon content of the alloy and large errors can be associated with experimental measurements because of non-uniformity in the starting material.

Characterizing the partially sensitized metal samples to describe the number of reactive sites, i.e., those areas containing chromium concentrations below 12 wt %, and total area of these sites is very difficult and probably is subject to large error limits. [SCHMIDT] has used SEM/EDS to examine areas for Cr enrichment or depletion and was, for the most part, unsuccessful, concluding that the beam resolution was not small enough to differentiate regions of varying Cr concentrations. This is essentially what was observed in the present study.

Tests done with partially sensitized austenitic alloys may display significant differences in results due to variable sensitization. This effect precludes the direct and meaningful comparison of replicate or even similar samples unless special consideration is provided for establishing the reactivity differences between test samples. Examples of this are also seen in the N2 batch test results where varying degrees of sensitization and reaction were observed although the retainer sections were subjected to the same heat treatment process.

The extent to which any alloy has been subjected to cold working may affect its susceptibility to sensitization. To examine the effect of cold work on austenitic alloys, specimens were deformed [BRIANT] by bending to produce a room temperature cold worked microstructure. Time-to-sensitization tests were then performed on the deformed and pristine samples of low carbon stainless steel. The results showed dramatic differences in the time-temperature sensitivity (TTS) relationship. For example, heating the samples at 550°C required 24-100 hours to sensitize a nondeformed sample whereas it required only 0.08-0.12 hours to sensitize the deformed sample [BRIANT]. Examination of the microstructure of these samples revealed the corrosion was completely intergranular in the undeformed samples and predominantly transgranular in the deformed samples.

Further examination of the microstructure of the deformed samples showed that the deformation had produced a martensite phase in the 304 stainless steel. Sensitization is a diffusion-controlled process and the diffusion coefficients of carbon and chromium are both higher in martensite than in austenite. Therefore, chromium carbides should precipitate more rapidly in martensite than austenite. Additionally, carbon is less soluble in martensite than austenite, providing an added driving force for carbide precipitation. Transgranular corrosion results from martensite introduced by cold deformation causing lath formation within the grains where the carbides precipitate. Samples from the N2 test certainly had some degree of cold work introduced during manufacture of the sheet and during punching of the holes. The surfaces seem to resist sensitization more than the bulk, as shown in Fig. 3, while the punched surfaces have very deformed grains, but show no obvious signs of localized reaction.

The effect of sensitization on glass reaction was examined more closely in a set of parametric experiments (P-VIII) [BATES-4] done in concert with the N2 Test series. The purpose of these experiments was to duplicate the N2 series using non-transuranic containing glass such that more extensive surface profiling techniques could be used to study the glass reaction. The conditions and components of these experiments were the same as for the N2 tests except that the 304L ss was from a different heat and thereby had a slightly different composition (Table 7); the heat treatment process although intended to be done at 550°C was probably done at a slightly lower temperature, $535 \pm 10^\circ\text{C}$; and that the vessels used were parametric vessels which are smaller than the N2 vessels. These parametric experiments will be discussed in detail elsewhere [BATES-5], but it is instructive to compare the rate of reaction and degree of sensitization in these tests with that observed in the N2 and F1 tests.

The $(\text{NL})_i$ results for Li, B, and U from the batch and continuous P-VIII experiments are shown in Fig. 20a-f. No strong interaction was noticed except in the 52-week batch test. Also, it was found that no noticeable sensitization had been imparted to the retainer sections (Fig. 3) for all experiments except the 52-week batch sample (which has not yet been examined). In the continuous P-VIII experiments (average of two experiments), the $(\text{NL})_{\text{Li}}$ values trended upward, reaching $\sim 0.5 \text{ g/m}^2$ after 26 weeks and 1 g/m^2 after 52 weeks. The normalized release of B and U is much less than that of Li at all time periods reaching $\sim 0.1\text{-}0.2 \text{ g/m}^2$ after 52 weeks. In the batch P-VIII experiments (only one experiment for each time period) the results were more erratic, with $(\text{NL})_{\text{Li}}$ being $\sim 1 \text{ g/m}^2$ at 26 weeks and $\sim 2 \text{ g/m}^2$ at 52 weeks. The batch experiments contain a rinse fraction and the (NL) values are expected to be larger than the corresponding values for the continuous experiments. For B and U the batch results are close to those of the continuous experiments through 39 weeks, but jump dramatically to $\sim 1.2 \text{ g/m}^2$ at 52 weeks when a strong localized interaction was observed. Thus, the strong localized interaction affects the release of B and U to a much larger extent than for Li.

The same effect of localized interaction, i.e., increased release of elements associated with the glass matrix, is seen in all tests and experiments done to date using the Unsaturated Test design. Table 17 reviews the tests that have been performed and Fig. 20 shows comparative results.

All of the N2 tests, the F1 batch tests #s 1-5, and the 52-week batch P-VIII experiment showed evidence of strong glass/metal interaction. When strong interaction is observed, the release of Li is marginally increased (10 to 50%) compared to tests where there is no strong interaction. Under the same conditions the releases of B and U are increased by 300 to 1000% in samples where a strong interaction is observed. The release of the transuranic elements has only been measured systematically in the N2 test series. However, comparison can be made with the results of a laboratory experiment (similar configuration to Unsaturated Test, but with water contact rate controlled by flow through a tuff core) [BATES-1, -2] where no strong glass/metal interaction or sensitization of the metal was observed. This comparison indicates that after 52 weeks the release of Pu and Am is about eight times greater in the N2 tests.

Fig. 20. Comparison of Normalized Elemental Releases from the N2 Test (□), the F1 Test (Δ), and the P-VIII Experiments (○). The release of Li, B, and U in the continuous mode are shown in a-c and in the batch mode in d-f.

Table 17. Interactions and Sensitization Observed in Unsaturated Tests and Experiments

Test ID	Test Type	Interaction ^a		Sensitization ^a	
		Strong	Weak	Yes	No
N2 Unsaturated Test	Continuous	X		X	
N2 Unsaturated Test	Batch	X		X	
F1 Unsaturated Test	Continuous		X		X
F1 Unsaturated Test	Batch	X	X	X	X
P-VIII Parametric	Continuous		X		X
P-VIII Parametric	Batch	X	X	X	X

^aIf both choices are marked, this means that both effects were observed for individual samples that comprise the entire set.

In tests where there is no strong interaction, the Li release is much larger than that of B or U after 52 weeks ($1-1.5 \text{ g/m}^2$ vs. $0.1-0.2 \text{ g/m}^2$). When there is a strong interaction, the Li release is in the $1.5-2 \text{ g/m}^2$ range while the B and U releases are in the $1-1.5 \text{ g/m}^2$ range after 52 weeks. The (NL)₁ release values are quite consistent depending on the extent of interaction, and support the conclusions drawn from previous tests [BATES-1, -2] and experiments [ABRAJANO] regarding the processes by which the glass reacts.

The observed release of elements is a result of the different types of interactions that occur during the test period. As has been described previously [BATES-1, -2; ABRAJANO] at all surfaces the first process to occur is an ion exchange process whereby water exchanges with alkali metal cations and to a lesser extent B via a diffusional process. This process is perpetuated by both the thin film and larger amounts of standing water that contact the glass due to saturated vapor conditions. The exchange process results in hydrolysis of the glass, eventually forming a layer near the surface that is depleted in alkali metal cations and B. The layer initially does not act as a diffusion barrier to these elements. The layer does retain other elements such as Pu and Am and to a lesser degree U and Np due to its physical and chemical properties. Such a layer is observed on the top and bottom surfaces (Fig. 11) but not on the side surfaces. Extended studies of the reaction kinetics that occur under vapor contact (side surfaces) and liquid contact (top and bottom surfaces) are in progress [BATES-6, EBERT].

In addition to the processes described above, the top and bottom surfaces are affected by a strong interaction between the metal and the glass. This interaction correlates well with the extent of sensitization that exists in the retainer sections and results in a significant increase in the release of those elements that are not released by ion exchange from the glass. This is likely not a homogeneous process that occurs evenly over the top and bottom surfaces, but probably occurs at interfacial regions where the protective oxide layer on the steel has broken down and elements (iron, chromium, and nickel) from the steel react with silicon from the glass and the groundwater to form insoluble metal silicates. The glass reaction is accelerated because these metal silicates act as a sink to remove silicon from solution.

Some measure of the extent of the breakdown of the reacted layer can be calculated if it is assumed that Pu and Am are released only through complete dissolution of this layer. This assumption has been verified in previous leaching studies of SRL 165 type glass using solution analysis and ion microprobe profiling arguments [ABRAJANO]. For test N2 #8, a reaction layer $\sim 0.2 \mu\text{m}$ thick would have to be dissolved from the top and bottom surfaces to release the measured amount of Pu and Am. This layer does not account for all of the released B or U, which indicates that these elements are in part also released by the other processes discussed above.

A strong interaction between glass and metal has been noted previously when ductile iron and glass were present in the same leaching container as the glass [McVAY]. However, previous studies involving stainless steel and glass using standard leaching conditions showed no enhanced reaction due to the metal [BAZAN, EBERT, McVAY]. The exact nature of the increased interaction is being investigated further, but it is interesting to note that in several regions of strong interaction, chloride containing precipitates are observed (Figs. 9e and 14). Thus, while the extent of glass/metal interaction correlates well with the extent of sensitization found in the metal, it has not been verified that the localized reaction occurs via an intergranular attack process or is related to a pitting or crevice corrosion attack that may be enhanced by the S and Cl present in the EJ-13 water or in the S/Cl-rich pockets associated with the glass.

The release of the actinide elements from the WPA follows the trend $\text{Np} > \text{U} > \text{Pu} \approx \text{Am}$. A similar trend has been observed in leaching studies of SRL 165 type glass under saturated NNWSI conditions [EBERT]. Neptunium was released to only a slightly lesser extent than Li, and showed little interaction with the metal components of the test. Uranium release to solution, although somewhat variable, was about five times less than that of Np. Uranium was the only actinide for which a unique phase was detected using SEM/EDS (Fig. 16). Uranium containing phases were not identified on all samples, which may explain some of the variability in U release. Plutonium and Am were found only sparingly in solution. Previous studies [BART, WALTON] indicate that these elements can sorb strongly to metal surfaces or can either sorb or be incorporated into metal oxide and hydroxide phases. Cleaning the metal test components by an acid stripping process accounts for all the Pu and Am associated with the metal components

(metal plus precipitates), and when combined with the Pu and Am detected in solution provides the total measurable release. Some additional Pu and Am is likely associated with the reaction products that remain on the glass surface.

There is no evidence that the sorption capacity of the metal components and alteration phases formed during the reaction process decreases with time, and the enhanced glass reaction that occurs due to metal sensitization may adequately generate new reaction phases that will incorporate Pu and Am as they are released from the glass.

The reaction products identified in the analyzed samples result from the interaction processes described above, and are the result of saturation of the liquid in contact with the WPA. The composition of this liquid (Table 12) as determined from the continuous tests shows some variance due to the nonuniform processes by which water is released from the WPA (evaporation or dripping). In the N2 #10 test no glass components were detected in the test solution until the 19.5-week sampling period, although the WPA had the same strongly reacted appearance as noted in the other tests.

However, as shown in Fig. 7, the solution composition of most components has become reasonably constant by the 65-week sampling period. Only Li, which shows a slowly decreasing concentration has a non-constant trend. However, the continuous tests are still in progress to see whether the observed trends are continued. The next step in interpreting the reaction process will be to use geochemical modeling codes in conjunction with the solution and phase results reported here to see if any correlation between the modeling theory and results exists.

V. CONCLUSIONS

Unsaturated testing with associated parametric experimentation of SRL 165 type glass has been completed for up to 65 weeks. These tests, which are specific to the NNWSI repository site, result in the formation of several identifiable reaction phases and reasonably constant elemental concentrations in solution. These data, when augmented with pH and anion measurements of the test solutions, should be amenable to modeling using geochemical codes to project the long-term repository performance of glass waste.

Other data presented suggests that sensitized 304L ss interacts strongly with glass in the presence of small volumes of repository water. This strong interaction results in breakdown of the reacted layer on the surface of the glass, thereby increasing elemental release up to ten times the amount observed without the interaction. However, the layer breakdown does not necessarily result in equal release to solution for all the actinide elements. Neptunium is released to solution without interacting significantly with either alteration phases or metal components. However, U interacts moderately with the reaction products and forms unique stable

phases which decreases its release to solution. The releases of Pu and Am to solution are even more reduced as these elements strongly interact with the reaction products and metal components.

The Tests reported here are still in progress and will be continued to gain additional information regarding glass/metal/water interactions under unsaturated conditions.

At time of publication, the continuous tests have been ongoing for 206 weeks. Additionally, advances in characterization of the reacted glass surfaces using analytical electron microscopy have been developed which should provide additional insight into the glass reaction process. AEM will be applied to the archive samples from the batch tests.

ACKNOWLEDGMENTS

Work supported by the U.S. Department of Energy, Office of Geologic Repositories, Nevada Nuclear Waste Storage Investigations Project, under Contract Number W-31-109-38.

REFERENCES

ABRAJANO

T. A. Abrajano, Jr., J. K. Bates, T. J. Gerding, and W. L. Ebert, "The Reaction of Glass in a Gamma Irradiated Saturated Tuff Environment, Part 3: Long-Term Experiments at 1×10^4 Rad/hour," Argonne National Laboratory report ANL-88-14 (1988). NNA.880204.0006

BART

G. Bart et al., "Borosilicate Glass Corrosion in the Presence of Steel Corrosion Products," in Scientific Basis for Nuclear Waste Management X, J. K. Bates and W. B. Seefeldt, eds., Mater. Res. Soc. Proc. 85, Pittsburgh, PA, pp. 613-622 (1987). NNA.891212.0005

BATES-1

J. K. Bates and T. J. Gerding, "One-Year Results of the NNWSI Unsaturated Test Procedure: SRL 165 Glass Application," Argonne National Laboratory Report ANL-85-41 (1986). NNA.870903.0081

BATES-2

J. K. Bates and T. J. Gerding, "NNWSI Phase II Materials Interaction Test Procedure and Preliminary Results," Argonne National Laboratory Report ANL-84-81 (1984). HQS.880517.2388

BATES-3

J. K. Bates et al., "NNWSI Waste Form Testing at Argonne National Laboratory, Semiannual Report July-December 1985," Lawrence Livermore National Laboratory Report UCRL-15801 (1986). HQS.880517.2392

BATES-4

J. K. Bates et al., "NNWSI Waste Form Testing at Argonne National Laboratory, Semiannual Report January-June 1986," Lawrence Livermore National Laboratory Report UCRL-15801-86-1 (1987). NNA.871218.0006

BATES-5

J. K. Bates and T. J. Gerding, "The Reaction of Glass in Unsaturated Conditions," in preparation.

BATES-6

J. K. Bates et al., "NNWSI Waste Form Testing at Argonne National Laboratory, Semiannual Report July-December 1986," Lawrence Livermore National Laboratory Report UCRL-15801-86-2 (1988).

BAZAN

F. Bazan and J. H. Rego, "The Tuff Reaction Vessel Experiment," Lawrence Livermore National Laboratory Report UCRL-53735 (1986). HQS.880517.2395

BINDER

W. O. Binder, C. M. Brown, and R. Franks, ASM Trans. Quant. 41, 1301 (1949). NNA.891120.0006

BRIANT

C. L. Briant, "Effects of Nitrogen and Cold Work on the Sensitization of Austenitic Stainless Steels," EPRI Report NP-2457 (1982).

CHANDLER

G. T. Chandler, G. G. Wicks, and R. M. Wallace, "Effects of SA/V and Saturation on the Chemical Durability of SRP Waste Glass," Savannah River Laboratory report DP-MS-86-56 (1986).

EBERT

W. L. Ebert, J. K. Bates, and T. J. Gerding, "The Reaction of Glass during Gamma Irradiation in a Saturated Tuff Environment: Part 4. SRL 165, ATM-1c, and ATM-8 Glasses at 1E3 R/h and 0 R/h," Argonne National Laboratory report ANL-90/13 (1990). NNA.900509.0197

McVAY

G. L. McVay and G. R. Robinson, "Effects of Tuff Waste Package Components on Release from 76-68 Simulated Waste Glass," Pacific Northwest Laboratory Report PNL-4897 (1984). HQS.880517.2496

SCHMIDT

C. G. Schmidt et al., "Low-Temperature Sensitization of Type 304 Stainless Steel Weld Heat Affected Zone," EPRI Report NP-3368 (1983).

WALTON

F. B. Walton, Atomic Energy of Canada, Limited, Pinawa, Manitoba (private communication, 12/87).

APPENDIX I

COMPOSITIONAL HOMOGENEITY MEASUREMENTS OF SRL A GLASS

Data included in this report are contained in notebooks 390 and 306 (NNWSI-05-016), file elements NNWSI-05-020, NNWSI-05-030, NNWSI-05-031, and in element 07 of the records file.

Table AI-1. Relative Radionuclide Concentrations in N2 Glass Samples (Gamma Counting)

Test #	Sample# ^b	Radionuclide Concentration ^a		
		Am (61 kev)	Np (86 kev)	Pa (311 kev)
N2-1	21	55.4	0.45	0.82
N2-2	22	55.1	0.45	0.84
N2-3	23	53.9	0.46	0.82
N2-4	24	54.8	0.46	0.84
N2-5	31	52.4	0.44	0.83
N2-6	32	56.1	0.46	0.83
N2-7	33	54.6	0.46	0.81
N2-8	34	55.8	0.46	0.82
N2-9	111	55.1	0.45	0.84
N2-10	131	53.6	0.41	0.80
N2-11	132	52.1	0.43	0.79
Avg		54.5 ± 1.3	0.45 ± 0.02	0.82 ± 0.02

^aValues are counts/sec/gm, not corrected for detector efficiency.

^bSamples are identified by the mold in which they were cast (first number), and by the sequence of casting (subscript).

Table AI-2. Relative Radionuclide Concentrations in N2 Glass Samples (Total Alpha Counting)

Test #	Sample #	Position ^a (counts/min) ^b			
		1	2	3	4
N2-9	11 ₁ , top	5755	5645	5568	5624
N2-8	3 ₄ , top	6119	5424	5789	5643
N2-7	3 ₃ , top	5691	5919	5937	5660
N2-12	13 ₂ , top	5816	5732	5924	5728
N2-8	3 ₄ , bottom	5621	5775	5871	5823
N2-9	11 ₁ , bottom	5972	5839	5766	5879
		Avg 5771 ± 151			

^aFour different positions of equal area were counted on the surface of each sample using a mask to screen unwanted surface.

^bTotal alpha. Each value is the average of two determinations.

APPENDIX II

COMPLETE SOLUTION ANALYTICAL RESULTS
FROM THE N2 TEST

Data presented in Table AII-1 were obtained by inductively coupled plasma ignition coupled with atomic emission spectroscopy detection (ICP/AES). The analytical error reported for these analyses is $\pm 10\%$ of the amount analyzed.

Data presented in Table AII-2 were obtained by alpha spectroscopy of test solutions. These solutions were taken directly from the tests sampled and have not been subjected to any separation procedures to remove salts or selected actinides. Analytical errors for these measurements based on counting statistics and quality of spectra are reported to be $\pm 20\%$.

Table AII-1. Solution Analyses (Cations) for the N2 Test Series

Test Type	Test #	Test Period (weeks)	Solution ^b Volume (mL)	Sample ID	Total Element in Solution (nanogram) ^a											
					Al	B	Ca	Fe	Li	Mg	Mn	Na	Ni	Si	Sr	U
Batch	N2-1	13	1.58	TF-304		6500	25300	220	17900	1900	11200	166300	900	44100	230	132
	N2-2	13	1.36	TF-305		13900	40100	470	24500	4200	18000	213400	2600	12100	540	707
	N2-3	26	2.34	TF-340		39300	72500	6200	54600	7900	58900	420000	8800	308400	1400	2309
	N2-4	26	3.70	TF-341		35600	80000	4500	52400	8900	82700	424300	8600	316600	1400	2631
	N2-5	39	4.17	TF-379	<1400	39900	81100	800	63300	9300	45700	531000	NA	378000	1650	4641
	N2-6	39	4.17	TF-380	<1300	24600	71200	800	36900	5600	32900	427000	NA	194500	830	460
	N2-7	52	5.86	TF-408	1900	23200	71600	2400	51200	6500	39500	605500	NA	267200	1100	2823
	N2-8	52	6.73	TF-409	<1600	25800	63800	5000	55300	6700	43000	623100		279700	1100	1368
Continuous ^c	N2-9	6.5	0.8	TF-284	<1600	4800	2100	<160	6300	160	5000	61000	<320	28700	<160	34
		13	0.82	TF-302	<1100	3400	9900	670	9300	640	17400	186200	1600	34100	<110	45
		19.5	0.71	TF-378	<1000	4400	23000	<100	7500	1900	16400	181600	5400	25600	260	95
		26	1.10	TF-336	<1100	1900	8300	230	2800	800	4500	55700	1200	33600	110	148
		32.5	0.88	TF-351	1900	1600	7600	6100	<330	610	11100	31800	6800	75800	<80	<16
		39	1.20	TF-375	<1100	1200	17200	<120	7500	1700	6300	133300	NA	51100	240	391
		45.5	1.00	TF-395	<1100	2800	3700	200	<220	400	1300	24600	NA	15500	<60	<11
		52	1.33	TF-404	<1600	6500	21700	<160	8600	2700	11300	163200	NA	59800	400	1504
Continuous ^c	N2-10	65	1.60 ^d	TF-439	<1200	6000	12500	200	5400	1500	5400	116200	2200	49500	190	148
		6.5	0.8	TF-285	<1600	4700	2800	<160	<320	280	10300	23900	<320	19200	<160	<8
		13	1.00	TF-303	<1100	2300	3800	740	<220	440	14500	24700	740	21500	<100	<10
		19.5	0.93	TF-327	<1100	1900	14600	310	8800	930	8900	102100	4300	34700	140	31
		26	1.05	TF-337		1200	11900	790	6900	830	3500	93600	1100	37300	120	109
		32.5	0.98	TF-352	4000	1600	17300	2100	4200	1200	12200	75700	3600	48900	130	143
		39	1.10	TF-376	<1100	1200	11900	290	4900	1050	5400	94100	NA	34700	160	371
		45.5	1.16	TF-396	<1100	1000	3100	320	<230	300	600	23300	NA	16600	<60	<11
Continuous ^c	N2-12	52	1.16	TF-405	<1700	1700	12400	500	5000	1300	5300	109900	NA	36600	165	435
		65	1.67	TF-440	<1200	3100	19300	120	7100	1800	7600	171200	3300	58900	270	334
		6.5	0.96	TF-329	<1100	5300	3000	160	4400	170	1300	53800	340	27600	<50	52
		13	1.05	TF-339	<1000	2400	2900	<140	5100	200	1100	68300	510	31700	<70	14
		19.5	1.03	TF-354	<1600	1500	5200	<160	3800	330	900	61300	570	28000	<80	<16
		26	1.16	TF-378	<1200	<360	9360	<120	4800	470	1400	80100	NA	30500	97	29
		32.5	1.10	TF-398	<1100	<340	11300	<110	4500	770	1800	82100	NA	33500	115	41
		39	1.22	TF-407	<1500	1300	13600	<160	7000	930	2300	119200	NA	48400	140	117
Continuous ^c		45.5	1.26	TF-428	<1100	1100	8400	120	3400	660	1600	78800	700	37600	90	57
		52	1.07	TF-442	<1200	1100	10500	220	4200	840	2400	95100	1300	4600	130	156

Cont'd

Table AII-1 (Cont'd)

Test Type	Test #	Test Period (weeks)	Solution ^b Volume (mL)	Sample ID	Total Element in Solution (nanogram) ^a													
					Al	B	Ca	Fe	Li	Mg	Mn	Na	Ni	Si	Sr	U		
Continuuc ^c Blank	N2-11		1.475		1000	260	6170			106	580		70000		53900	57		
		6.5	0.8	TF-286	<1600	<320	6600	190		<160	660	15300		66200	3400	48300	<150	<8
		13	0.89	TF-306	<1100	230	6100	300		<220	570	10900		67100	17200	45400	<100	<10
		19.5	0.88	TF-328	1900	280	9500			<210	770	10600		71100	9400	55200	50	<11
		26	0.88	TF-338	<1000	1400	8000	190		<260	650	17300		70200	6800	38600	<70	<13
		32.5	1.09	TF-353	<1700	1800	8900	<170		<330	690	2100		72200	1800	47800	<80	<16
		39	0.98	TF-377	2000	2300	9100	5600		<220	730	24100		69200	NA	43300	56	<11
		45.5	0.72	TF-397	<1100	<240	6700	3900		<230	420	1200		63200	NA	35200	57	<11
		52 ^e	0.56	TF-406	<1600	<500	5100	<160		<320	360	600		59800	NA	25300	<58	<16
		58.5	0.66		<1200	<350	5900	<150		<150	410	350		66700	970	40800	<58	<12
	65		0.88		<1200	<350	6400	190		<120	390		65000	1100	34800	<58	<12	
EJ-13 Water	11/22/85		1.0		720	180	4400		<10	71	400	<5	46500	<20	36900	41	4	
	5/14/86		1.0		750	180	4200		<10	66	390	<5	47800	<20	36200	41	NA	
	8/05/86		1.0		710	170	4100		<10	71	390	<5	46800	<20	36900	37	4	
	11/03/86	TF-374	1.0		700	175	4100		<10	79	400	<5	48700	NA	36200	37	3	
	1/31/87		1.0		720	176	4200		<10	72	395	<5	47500	<20	36500	39	3	

^aThe solution consists of 0.5 mL added at the beginning of the test plus the volume of EJ-13 added during the test. The elemental values have not been "blank" corrected, but they have been corrected for the volume of solution removed for alpha detection.

^bThe reported volume is that remaining in the test vessel upon sampling or termination. It is not the total solution added during testing.

^cValues are for each individual sampling period.

^dEstimated based on N2-10.

^eInlet line partially plugged thereby reducing the injected solution volume slightly.

NA = not analyzed, elements for which no values are entered have been analyzed but not yet reported.

Table AII-2a. Solution Results (Neptunium) for the N2 Test Series

Aliquot Description	Test #	Sol'n Volume Counted (mL)	Total Solution Volume (mL)	Aliquots ID	Live Time (sec)	Detector Efficiency	Counts	Raw c/s	Background Corrected c/s	Efficiency ^b and Volume Corrected d/s	Sol'n ID	Count ID
Test Solution (TS) wp Rinse + TS Filtered Vessel Acid Soak	N2-1	0.050	1.58	N2-1	142998	0.1907	310	2.2E-3	2E-3	0.33	1.3	None
		0.100	10.86*	N2-1*	86399	0.1907	195	2.3E-3	2.1E-3	1.17	4.5	TF-304
		0.150	10.86*	N2-1**	355110	0.1946	251	7E-4	6E-4	0.21	0.8	TF-304
		0.100	32.85	N2-1***	257068	0.1939	120	5E-4	3E-4	0.45	1.7	TF-309
Test Solution wp Rinse + TS Filtered Vessel Acid Soak	N2-2	0.050	1.36	N2-2	142994	0.1981	2050	1.43E-2	1.42E-2	1.95	7.5	None
		0.100	11.00*	N2-2*	86394	0.1981	409	4.7E-3	4.5E-3	2.55	9.8	TF-305
		0.150	11.00*	N2-2**	248944	0.1938	617	2.5E-3	2.3E-3	0.86	3.3	TF-305
		0.100	32.83	N2-2***	257041	0.1946	202	8E-4	6E-4	1.07	4.1	TF-310
Test Solution wp Rinse + TS Filtered Vessel Acid Soak	N2-3	0.200	2.34	N2-3*	13212	0.1938	1719	1.3E-1	1.30E-1	7.84	30.2	None
		0.200	15.58*	N2-3***	20837	0.1938	451	2.1E-2	2.15E-2	8.58	33.0	TF-340
		0.350	15.58*	N2-3**	59388	0.1938	857	1.44E-2	1.42E-2	3.25	12.5	TF-340
		1.000	29.43	N2-3****	81577	0.1938	1503	1.84E-2	1.82E-2	2.77	10.6	TF-372
Test Solution wp Rinse + TS Filtered Vessel Acid Soak	N2-4	0.200	3.70	N2-4*	15704	0.1938	1728	1.10E-1	1.10E-1	10.49	40.3	None
		0.200	11.22*	N2-4***	12119	0.2007	382	3.15E-2	3.13E-2	8.77	33.7	TF-341
		0.300	11.22*	N2-4**	72842	0.2007	1157	1.59E-2	1.57E-2	2.51	9.7	TF-341
		0.500	28.63	N2-4****	91576	0.2007	2117	2.60E-2	2.58E-2	7.36	20.3	TF-374
Test Solution wp Rinse + TS Filtered Vessel Acid Soak	N2-5	0.200	4.17	N2-5*	54842	0.1925	4314	7.87E-2	7.85E-2	8.50	32.7	None
		0.200	14.39*	N2-5***	25011	0.1925	731	2.92E-2	2.90E-2	10.85	41.7	TF-379
		0.350	14.39*	N2-5**	160261	0.1929	2937	1.77E-2	1.75E-2	3.72	14.3	TF-379
		2.000	32.55	N2-5****	86067	0.1929	190	2.4E-3	2.1E-3	0.17	0.6	TF-385
Test Solution wp Rinse + TS Filtered Vessel Acid Soak	N2-6	0.200	4.17	N2-6*	54843	0.2009	1397	2.55E-2	2.53E-2	2.61	10.0	None
		0.200	14.97*	N2-6***	25011	0.2009	269	1.00E-2	1.00E-2	3.63	13.9	TF-380
		0.400	14.97*	N2-6**	160263	0.1999	1339	8.1E-3	7.9E-3	1.37	5.3	TF-380
		2.000	30.17	N2-6****	86066	0.1999	145	1.8E-3	1.6E-3	0.11	0.4	TF-386
Test Solution wp Rinse + TS Filtered Vessel Acid Soak	N2-7	0.200	5.86	N2-7*	87249	0.1912	1942	2.23E-2	2.2E-2	3.38	13.0	None
		0.200	15.81	N2-7***	79215	0.1912	815	1.03E-2	1.01E-2	4.16	16.0	TF-408
		0.250	15.81	N2-7**	104122	0.1912	576	5.5E-3	5.3E-3	1.75	6.7	TF-408
		2.000	32.93	N2-7****	79031	0.1912	841	1.06E-2	1.04E-2	0.89	3.4	TF-414

Cont'd

Table AII-2a (Cont'd)

Aliquot Description	Test #	Sol'n Volume Counted (mL)	Total Solution Volume (mL)	Aliquot ID	Live Time (sec)	Detector Efficiency	Counts	Raw c/s	Efficiency and Volume Corrected			Solution Count ID
									Back-ground Corrected c/s	d/s	gm x10 ⁻⁸	
Test Solution	N2-8	0.200	6.73	N2-8*	87250	0.1972	2805	3.21E-2	3.2E-2	5.45	21.0	None
np Rinse + TS		0.200	16.25	N2-8***	79210	0.1972	891	1.12E-2	1.11E-2	4.56	17.5	TF-409
Filtered		0.300	16.25	N2-8**	104127	0.1972	754	7.2E-3	7.1E-3	1.94	7.5	TF-409
Vessel Acid Soak		2.000	38.10	N2-8****	79029	0.1972	1998	2.53E-2	2.51E-2	2.4	9.2	TF-415
Test Solution	N2-9	0.100	~0.8	N2-9*	168797	0.1456	895	5.3E-3	5.1E-3	0.28	1.1	None
Test Solution	6.5 weeks	0.100	~0.8	N2-9*	21736	0.1958	134	6.2E-3	6.0E-3	0.25	0.9	None
Dil/Filt TS		0.050	~0.8	N2-9**	334863	0.1456	172	5E-4	3E-4	2.92E-2	0.1	None
Vessel Acid Soak		0.500	31.62	N2-9***	336717	0.1456	81	2E-4	0	0	0	TF-287
Test Solution	N2-9	0.050	0.82	N2-9*	84400	0.1939	369	4.4E-3	4.2E-3	0.35	1.3	None
Vessel Acid Soak	13 weeks	0.100	33.91	N2-9***	172800	0.1939	74	4E-4	2E-4	0.4	1.5	TF-307
Test Solution	N2-9	0.050	0.71	N2-9*	55725	0.1981	620	1.11E-2	1.09E-2	0.78	3.0	None
Rinsed TS	19.5 weeks	0.100	11.26*	N2-9***	110214	0.1926	170	1.5E-3	1.3E-3	0.79	3.0	TF-328
Vessel Acid Soak		0.500	31.41	N2-9****	114777	0.1933	289	2.5E-3	2.3E-3	0.75	2.9	TF-330
Test Solution	N2-9	0.200	1.10	N2-9*	258167	0.2007	2176	8.4E-3	8.2E-3	0.23	0.9	None
Rinsed TS	26 weeks	0.200	13.38*	N2-9***	271218	0.1938	265	1.0E-3	8E-4	0.27	1.0	TF-336
Vessel Acid Soak		1.000	30.51	N2-9****	169503	0.1938	72	4E-4	2E-4	3.54E-2	1.0	TF-368
Test Solution	N2-9	0.050	0.88	N2-9*	171680	0.1984	119	7E-4	5E-4	4.6E-2	0.2	None
Rinsed TS	32.5 weeks	0.200	11.26*	N2-9***	173311	0.1904	50	3E-4	1E-4	2.6E-2	0.1	TF-351
Vessel Acid Soak		1.000	29.89	N2-9****	188386	0.1929	57	3E-4	-	1.1E-2	-	TF-364
Test Solution	N2-9	0.050	1.20	N2-9*	114698	0.1929	835	7.3E-3	7.1E-3	0.88	3.4	None
Rinsed TS	39 weeks	0.200	11.60*	N2-9***	170774	0.1925	555	3.2E-3	3.0E-3	0.90	3.5	TF-375
Vessel Acid Soak		2.000	32.07	N2-9****	142317	0.1923	152	1.1E-3	8E-4	7.0E-2	0.3	TF-381
Test Solution	N2-9	0.050	1.00	N2-9*	198443	0.1909	51	3E-4	-	2.8E-3	-	None
Rinsed TS	45.5 weeks	0.200	11.24*	N2-9***	279527	0.1909	69	2E-4	-	5.0E-3	-	TF-395
Vessel Acid Soak		2.000	31.76	N2-9****	143424	0.1909	50	3E-4	1E-4	9.9E-3	-	TF-399
Test Solution	N2-9	0.050	1.33	N2-9*	258150	0.1912	7097	2.75E-2	2.73E-2	3.79	14.6	None
Rinsed TS	52 weeks	0.200	11.16*	N2-9***	258145	0.1912	1810	7.0E-3	6.8E-3	1.93	7.4	TF-404
Vessel Acid Soak		2.000	36.84	N2-9****	234167	0.1938	273	1.2E-3	9E-4	0.09	0.4	TF-410

Cont'd

Table AII-2a (Cont'd)

Np										
Aliquot Description	Test #	Sol'n Volume Counted (ml.)	Total Solution Volume (ml.)	Aliquot ID	Live Time (sec)	Detector Efficiency	Counts	Raw c/s	Back-ground Corrected c/s	Efficiency and Volume Corrected d/s
										Solution Count ID
Test Solution Rinsed TS Vessel Acid Soak	N2-9	0.100	1.6**	N2-9*	145275	0.1909	1216	8.4E-3	8.2E-3	0.68
	65 weeks	0.200	12.16*	N2-9***	145345	0.1973	330	2.3E-3	2.1E-3	0.64
		2.000	33.68	N2-9****	249221	0.1954	128	5E-4	3E-4	0.03
Test Solution Dil/Filt TS Vessel Acid Soak	N2-10	0.100	~0.8	N2-10*	169042	0.1958	31	2E-4	0	0
	6.5 weeks	0.050	~0.8	N2-10**	334861	0.1958	41	1E-4	0	0
		0.500	32.07	N2-10***	237247	0.1958	28	1E-4	0	0
Test Solution Vessel Acid Soak	N2-10	0.050	1.00	N2-10*	84394	0.1946	8	1E-4	0	0
	13 weeks	0.100	32.82	N2-10***	140929	0.1938	29	2E-4	0	0
		0.050	0.93	N2-10*	89809	0.1926	288	3.2E-3	3.0	0.29
Test Solution Rinsed TS Vessel Acid Soak	N2-10	0.100	11.32*	N2-10***	110223	0.1981	60	5E-4	4E-4	0.22
	19.5 weeks	0.500	32.10	N2-10****	114785	0.1946	55	5E-4	3E-4	0.12
		0.200	1.05	N2-10*	86402	0.1938	916	1.06E-2	1.04E-2	0.28
Test Solution Rinsed TS Vessel Acid Soak	N2-10	0.200	13.33*	N2-10***	344199	0.1938	286	8.31E-4	6E-4	0.22
	26 weeks	1.000	30.95	N2-10****	169501	0.2007	137	8.08E-4	7E-4	0.10
		0.05	0.98	N2-10*	171679	0.1974	356	2.1E-3	1.9E-3	0.19
Test Solution Rinsed TS Vessel Acid Soak	N2-10	0.200	11.43*	N2-10***	173252	0.1974	144	8E-4	7E-4	0.20
	32.5 weeks	1.000	29.22	N2-10****	188392	0.1999	71	5E-4	3E-4	4.5E-2
		0.05	1.10	N2-10*	114686	0.1999	517	4.5E-3	4.3E-3	0.48
Test Solution Rinsed TS Vessel Acid Soak	N2-10	0.200	11.46*	N2-10***	179772	0.2009	361	2.1E-3	1.9E-3	0.55
	39 weeks	2.000	29.88	N2-10****	142312	0.1999	138	1.0E-3	8E-4	5.8E-2
		0.05	1.16	N2-10*	198474	0.1969	35	2E-4	0	0
Test Solution Rinsed TS Vessel Acid Soak	N2-10	0.200	11.42*	N2-10***	279567	0.1969	43	2E-4	0	0
	45.5 weeks	2.000	28.97	N2-10****	156506	0.1940	49	3E-4	-	6.2E-3
		0.05	1.16	N2-10*	159432	0.1912	781	4.9E-3	4.7E-3	0.57
Test Solution Rinsed TS Vessel Acid Soak	N2-10	0.200	11.55	N2-10***	159447	0.1912	241	1.5E-3	1.3E-3	0.39
	52 weeks	2.000	37.41	N2-10****	234166	0.2030	90	4E-4	2E-4	0.02
		0.05	1.16	N2-10*	159432	0.1912	781	4.9E-3	4.7E-3	0.57

Cont'd

Table AII-2a (Cont'd)

Aliquot Description	Test #	Sol'n Volume Counted (ml)	Total Solution Volume (ml)	Aliquot ID	Live Time (sec)	Detector Efficiency	Counts	Raw c/s	Efficiency			Solution Count ID
									Back-ground Corrected c/s	Volume Corrected d/s	gm x10 ⁻⁸	
Test Solution Rinsed TS Vessel Acid Soak	N2-10 65 weeks	0.100	1.67	N2-10*	241803	0.1934	1751	7.2E-3	7.1E-3	0.61	2.3	None
		0.200	12.23*	N2-10****	241802	0.2022	510	2.1E-3	1.9E-3	0.58	2.2	TF-440
		2.000	31.87	N2-10****	249219	0.2018	50	2E-4	-	-	-	TF-444
Test Solution Rinsed TS Vessel Acid Soak	N2-12 6.5 weeks	0.050	0.96	N2-12*	89867	0.1981	78	9E-4	7E-4	6.77E-2	0.3	None
		0.100	10.83*	N2-12***	148007	0.1939	60	4E-4	3E-4	0.17	0.6	TF-329
		0.500	31.96	N2-12****	173493	0.1946	119	7E-4	5E-4	0.18	0.7	TF-333
Test Solution Rinsed TS Vessel Acid Soak	N2-12 13 weeks	0.200	1.05	N2-12*	70523	0.1938	493	7.0E-3	6.8E-3	0.18	0.7	None
		0.200	11.57*	N2-12***	167603	0.1979	97	6E-4	4E-4	0.16	0.6	TF-339
		1.000	30.98	N2-12****	70937	0.1929	116	1.6E-3	1.4E-3	0.23	0.9	TF-371
Test Solution Rinsed TS Vessel Acid Soak	N2-12 19.5 weeks	0.05	1.03	N2-12*	226272	0.1904	392	1.7E-3	1.6E-3	0.17	0.7	None
		0.200	11.39*	N2-12***	226271	0.1974	93	4E-4	3E-4	7.5E-2	0.3	TF-354
		1.000	31.15	N2-12****	94063	0.1929	53	6E-4	3E-4	5.4E-2	0.2	TF-367
Test Solution Rinsed TS Vessel Acid Soak	N2-12 26 weeks	0.05	1.16	N2-12*	230229	0.1929	392	1.7E-3	1.5E-3	0.18	0.7	None
		0.200	12.37*	N2-12***	230227	0.1999	172	7E-4	6E-4	0.17	0.7	TF-378
		2.000	30.38	N2-12****	154556	0.1909	95	6E-4	4E-4	3.1E-2	0.1	TF-384
Test Solution Rinsed TS Vessel Acid Soak	N2-12 32.5 weeks	0.05	1.10	N2-12*	324163	0.1909	687	2.1E-3	1.9E-3	0.22	0.9	None
		0.200	11.48*	N2-12***	324158	0.1909	466	1.4E-3	1.3E-3	0.36	1.4	TF-398
		2.000	27.82	N2-12****	143752	0.1969	165	1.1E-3	1.0E-3	1.0E-3	-	TF-402
Test Solution Rinsed TS Vessel Acid Soak	N2-12 39 weeks	0.05	1.22	N2-12*	257510	0.1912	671	2.6E-3	2.4E-3	0.39	1.2	None
		0.200	10.62*	N2-12***	257525	0.1972	336	1.1E-3	1.1E-3	0.30	1.2	TF-407
		2.000	32.06	N2-12****	173134	0.1938	117	7E-4	4E-4	3.54E-2	0.1	TF-413
Test Solution Rinsed TS Vessel Acid Soak	N2-12 45.5 weeks	0.05	1.26	N2-12*	348705	0.1909	641	1.8E-3	1.6E-3	2.12E-1	0.8	None
		0.200	10.35*	N2-12***	348681	0.1973	293	8E-4	7E-4	1.71E-1	0.7	TF-428
		2.000	32.45	N2-12****	135809	0.1973	223	1.6E-3	1.5E-3	1.19E-1	0.5	TF-430
Test Solution Rinsed TS Vessel Acid Soak	N2-12 52 weeks	0.05	1.07	N2-12*	170800	0.1934	513	3.0E-3	2.8E-3	3.07E-1	1.2	None
		0.200	11.62*	N2-12***	170804	0.2022	240	1.4E-3	1.2E-3	3.49E-1	1.3	TF-442
		2.000	35.13	N2-12****	174860	0.1954	162	8E-4	5E-4	4.4E-2	0.2	TF-446

Cont'd

Table AII-2a (Cont'd)

Aliquot Description	Test #	Sol'n Volume Counted (mL)	Total Solution Volume (mL)	Aliquot ID	Live Time (sec)	Detector Efficiency	Counts	Raw c/s	Np			Solution Count ID	
									Back-ground Corrected c/s	Efficiency and Volume Corrected d/s	gm x10 ⁻⁸		
Test Solution	N2-11	0.100	~0.8	N2-11*	336886	0.1958	52	2E-4	0	0	0	None	499
Vessel Acid Soak	6.5 weeks	0.500	31.29	N2-11***	888246	0.1958	133	2E-4	0	0	0	TF-289	501
Test Solution	N2-11	0.050	0.89	N2-11*								None	513
Vessel Acid Soak	13 weeks	0.100	30.76	N2-11***								TF-311	514
Test Solution	N2-11	0.050	0.88	N2-11*	173573	0.1938	38	2E-4	0	0	0	None	609
Rinsed TS	19.5 weeks	0.100	10.70	N2-11***	248925	0.1946	33	1E-4	0	0	0	TF-328	606
Vessel Acid Soak		0.500	30.60	N2-11***	184243	0.1999	38	2E-4	0	0	0	TF-332	814
Test Solution	N2-11	0.200	0.88	N2-11*	99769	0.2007	11	1E-4	0	0	0	None	626
Rinsed TS	26 weeks	0.200	13.54*	N2-11***	258999	0.2007	44	2E-4	0	0	0	TF-338	630
Vessel Acid Soak		1.000	30.93	N2-11***	184247	0.1929	50	3E-4	0	0	0	TF-370	813
Test Solution	N2-11	0.050	1.09	N2-11*	166350	0.1925	34	2E-4	0	0	0	None	799
Rinsed TS	32.5 weeks	0.200	11.52*	N2-11***	166225	0.2009	25	2E-4	0	0	0	TF-353	800
Vessel Acid Soak		1.000	32.54	N2-11***	94056	0.1999	17	2E-4	0	0	0	TF-366	829
Test Solution	N2-11	0.050	0.98	N2-11*	166108	0.1925	33	2E-4	0	0	0	None	791
Rinsed TS	39 weeks	0.200	11.44*	N2-11***	166271	0.2009	38	2E-4	0	0	0	TF-377	790
Vessel Acid Soak		2.000	31.86	N2-11***	154593	0.1969	32	2E-4	0	0	0	TF-383	837
Test Solution	N2-11	0.05	0.72	N2-11*	166058	0.1940	38	2E-4	0	0	0	None	862
Rinsed TS	45.5 weeks	0.200	11.36*	N2-11***	166059	0.1990	49	3E-4	1E-4	3.0E-2	0.1	TF-397	863
Vessel Acid Soak		2.000		N2-11***	156562	0.1990	27	2E-4	0	0	0	TF-401	861
Test Solution	N2-11	0.05	0.56	N2-11*	173602	0.1938	55	3E-4	1E-4	5E-3	-	None	957
Rinsed TS	52 weeks	0.200	11.51*	N2-11***	173703	0.2030	30	2E-4	0	0	0	TF-406	958
Vessel Acid Soak		2.000	26.81	N2-11***	173214	0.2030	25	1E-4	0	0	0	TF-412	956
Test Solution	N2-11	0.05	0.66	N2-11*	253048	0.1909	46	2E-4	0	0	0	None	1000
Rinsed TS	58.5 weeks	0.200	11.73*	N2-11***	253009	0.1973	43	2E-4	0	0	0	TF-427	1001
Vessel Acid Soak		2.000	34.82	N2-11***	135825	0.1909	44	3E-4	1E-4	9E-3	-	TF-429	1006

aThe asterisks refer to aliquot identification and are not footnotes.

bTo convert counts/sec for Np to gm Np follow the following steps: (1) subtract background counts for Np from raw Np c/s, (2) divide background corrected c/s by detector efficiency and by the solution volume counted and multiply by the total solution volume, and (3) divide Np d/s by $(2.6 \times 10^4 \text{ d/s}) / (\text{gm Np})$.

Table AII-2b. Solution Results (Plutonium) for the N2 Test Series

Aliquot Description	Test #	Sol'n Volume Counted (mL)	Total Solution Volume (mL)	Aliquot ID	Live Time (sec)	Detector Efficiency	Counts	Raw c/s	Background Corrected c/s	Efficiency ^a			Solution Count ID
										Back-ground Corrected	Volume Corrected	gm x10 ⁻¹⁰ d/s	
Test Solution (TS) wp Rinse + TS Filtered Vessel Acid Soak	N2-1	0.050	1.58	N2-1	142998	0.1907	1135	7.9E-3	7.8E-3	1.29	5.7	None	589
		0.100	10.86*	N2-1*	86399	0.1907	523	6.1E-3	6.0E-3	3.38	14.9	TF-304	591
		0.150	10.86*	N2-1**	355110	0.1946	83	2E-4	1E-4	2.4E-2	0.1	TF-304	592
		0.100	32.85	N2-1***	257068	0.1939	301	1.2E-3	1.1E-3	1.78	7.8	TF-309	599
Test Solution wp Rinse + TS Filtered Vessel Acid Soak	N2-2	0.050	1.36	N2-2	142994	0.1981	541	3.8E-3	3.6E-3	0.50	2.2	None	590
		0.100	11.00*	N2-2*	86394	0.1981	532	6.2E-3	6.1E-3	3.33	14.7	TF-305	592
		0.150	11.00*	N2-2**	248944	0.1938	34	1E-4	0	0	0	TF-305	595
		0.100	32.83	N2-2***	257041	0.1946	933	3.6E-3	3.5E-3	5.84	25.7	TF-310	598
Test Solution wp Rinse + TS Filtered Vessel Acid Soak	N2-3	0.200	2.34	N2-3*	13212	0.1938	803	6.0E-2	6.07E-2	3.66	16.1	None	625
		0.200	15.50*	N2-3***	29837	0.1938	542	2.60E-2	2.59E-2	10.3	45.6	TF-340	616
		0.350	15.50*	N2-3**	59388	0.1938	20	3E-4	2E-4	5.0E-2	0.2	TF-340	618
		1.000	29.43	N2-3****	81577	0.1938	2212	2.71E-2	2.70E-2	4.09	18.1	TF-372	635
Test Solution wp Rinse + TS Filtered Vessel Acid Soak	N2-4	0.200	3.70	N2-4*	15704	0.1938	546	3.48E-2	3.46E-2	3.31	14.6	None	619
		0.200	11.22*	N2-4***	12119	0.2007	446	3.68E-2	3.67E-2	10.2	45.1	TF-341	628
		0.300	11.22*	N2-4**	72842	0.2007	15	2E-4	0	0	0	TF-341	623
		0.500	28.63	N2-4****	81576	0.2007	3629	4.45E-2	4.44E-2	6.33	27.9	TF-374	636
Test Solution wp Rinse + TS Filtered Vessel Acid Soak	N2-5	0.200	4.17	N2-5*	54842	0.1925	377	6.9E-3	6.7E-3	0.73	3.2	None	786
		0.200	14.39*	N2-5***	25011	0.1925	765	3.06E-2	3.05E-2	11.4	50.2	TF-379	788
		0.350	14.39*	N2-5**	166261	0.1929	54	3E-4	2E-4	4.2E-2	0.2	TF-379	807
		1.000	32.55	N2-5****	86087	0.1929	3252	4.06E-2	4.05E-2	3.18	14.0	TF-385	832
Test Solution wp Rinse + TS Filtered Vessel Acid Soak	N2-6	0.200	4.17	N2-6*	54843	0.2009	196	3.6E-3	3.4E-3	0.34	1.5	None	787
		0.200	14.97*	N2-6***	25011	0.2009	299	1.2E-2	1.18E-2	4.06	17.9	TF-380	789
		0.400	14.97*	N2-6**	166263	0.1999	60	4E-4	2E-4	3.0E-2	0.1	TF-380	808
		2.000	30.17	N2-6****	86086	0.1999	1086	1.4E-2	1.3E-2	0.94	4.1	TF-386	833
Test Solution wp Rinse + TS Filtered Vessel Acid Soak	N2-7	0.200	5.86	N2-7*	87249	0.1912	1549	1.78E-2	1.76E-2	2.70	11.9	None	939
		0.200	15.81	N2-7***	79215	0.1912	1267	1.60E-2	1.59E-2	6.56	28.9	TF-408	941
		0.250	15.81	N2-7**	104122	0.1912	14	1E-4	0	5E-3	-	TF-408	935
		2.000	32.93	N2-7****	79031	0.1912	1043	2.33E-2	2.32E-2	2.0	8.8	TF-414	945

Cont'd

Table AII-2b (Cont'd)

Aliquot Description	Test #	Sol'n Volume Counted (mL)	Total Solution Volume (mL)	Aliquot ID	Live Time (sec)	Detector Efficiency	Counts	Raw c/s	Pu			Solution Count ID
									Back-ground Corrected c/s	Efficiency and Volume Corrected d/s	gm x10 ⁻¹⁰	
Test Solution	N2-8	0.200	6.73	N2-8*	87250	0.1972	3869	4.43E-2	4.42E-2	7.534	33.2	948
wp Rinse + TS		0.200	16.25	N2-8***	79210	0.1972	1482	1.87E-2	1.85E-2	7.630	33.6	942
Filtered		0.300	16.25	N2-8**	104127	0.1972	19	2E-4	0	2E-3	-	936
Vessel Acid Soak		2.000	38.10	N2-8****	79829	0.1972	3397	4.30E-2	4.28E-2	8.27	36.4	946
Test Solution	N2-9	0.100	~0.8	N2-9*	168797	0.1456	2884	1.66E-2	1.65E-2	0.91	4.0	450
Test Solution	6.5 weeks	0.100	~0.6	N2-9*	21736	0.1956	587	2.33E-2	2.32E-2	0.95	4.2	453
Dil/Filt TS		0.050	~0.8	N2-9**	334863	0.1456	67	2E-4	5E-3	7.3E-3	-	496
Vessel Acid Soak		0.500	31.62	N2-9***	336717	0.1456	63	2E-4	0	0	0	498
Test Solution	N2-9	0.050	0.82	N2-9*	84400	0.1939	197	2.3E-3	2.23E-3	0.19	0.8	595
Vessel Acid Soak	13 weeks	0.100	33.91	N2-9***	172884	0.1939	180	1.0E-3	9E-4	1.65	7.3	600
Test Solution	N2-9	0.050	0.71	N2-9*	55725	0.1981	110	2.0E-3	1.9E-3	0.13	0.6	580
Rinsed TS	19.5 weeks	0.100	11.26*	N2-9***	110714	0.1926	40	3E-4	2E-4	0.14	0.6	587
Vessel Acid Soak		0.500	31.41	N2-9****	114777	0.1938	365	3.2E-3	3.1E-3	0.99	4.4	587
Test Solution	N2-9	0.200	1.10	N2-9*	258167	0.2007	263	1.0E-3	8E-4	2.33E-2	0.1	624
Rinsed TS	26 weeks	0.200	13.38*	N2-9***	271218	0.1938	79	3E-4	2E-4	5.91E-2	0.3	629
Vessel Acid Soak		1.000	30.51	N2-9****	189583	0.1938	387	1.81E-3	1.7E-3	0.27	1.2	633
Test Solution	N2-9	0.050	0.88	N2-9*	171690	0.1984	134	8E-4	7E-4	6.0E-2	0.3	705
Rinsed TS	32.5 weeks	0.200	11.26*	N2-9***	173311	0.1984	25	1E-4	-	4.2E-3	-	703
Vessel Acid Soak		1.000	29.89	N2-9****	188386	0.1929	100	5E-4	4E-4	6.2E-2	0.3	826
Test Solution	N2-9	0.050	1.20	N2-9*	114698	0.1929	294	2.6E-3	2.4E-3	0.31	1.4	809
Rinsed TS	39 weeks	0.200	11.60*	N2-9***	170774	0.1925	290	1.7E-3	1.6E-3	0.47	2.1	796
Vessel Acid Soak		2.000	32.07	N2-9****	142317	0.1929	476	3.3E-3	3.2E-3	0.27	1.2	830
Test Solution	N2-9	0.050	1.00	N2-9*	198443	0.1989	31	2E-4	-	2.7E-3	-	854
Rinsed TS	45.5 weeks	0.200	11.24*	N2-9***	279527	0.1989	46	2E-4	-	1.0E-2	-	850
Vessel Acid Soak		2.000	31.76	N2-9****	143424	0.1989	54	4E-4	2E-4	2.1E-2	0.1	856
Test Solution	N2-9	0.050	1.33	N2-9*	258150	0.1912	450	1.7E-3	1.6E-3	0.23	1.0	937
Rinsed TS	52 weeks	0.200	11.16*	N2-9***	258145	0.1912	219	8E-4	7E-4	0.19	0.8	938
Vessel Acid Soak		2.000	36.84	N2-9****	234167	0.1938	1548	6.6E-3	6.4E-3	0.62	2.7	953

Cont'd

Table AII-2b (Cont'd)

Aliquot Description	Test #	Sol'n Volume Counted (mL)	Total Solution Volume (mL)	Aliquot ID	Live Time (sec)	Detector Efficiency	Counts	Raw c/s	Pu				Solution ID	Count ID
									Back-ground	Corrected	Efficiency and Volume	Corrected		
								c/s	c/s	d/s		d/s		
Test Solution	N2-9	0.100	1.6**	N2-9*	145275	0.1909	103	7E-4	6E-4	0.05	0.2	0.05	None	1026
Rinsed TS	65 weeks	0.200	12.16*	N2-9***	145345	0.1973	42	3E-4	1E-4	0.03	0.1	0.03	TF-439	1027
Vessel Acid Soak		2.000	33.68	N2-9****	249221	0.1954	784	3.2E-3	3.0E-3	0.26	1.2	0.26	TF-443	1081
Test Solution	N2-10	0.100	~0.8	N2-10*	169042	0.1958	27	2E-4	0	0	0	0	None	451
Dil/Filt TS	6.5 weeks	0.050	~0.8	N2-10**	334861	0.1958	58	2E-4	0	0	0	0	None	497
Vessel Acid Soak		0.500	32.07	N2-10***	237247	0.1958	60	2E-4	0	0	0	0	TF-288	479
Test Solution	N2-10	0.053	1.00	N2-10*	84394	0.1946	13	2E-4	0	0	0	0	None	596
Vessel Acid Soak	13 weeks	0.100	32.82	N2-10***	140929	0.1938	16	1E-4	0	0	0	0	TF-358	611
Test Solution	N2-10	0.050	0.93	N2-10*	89809	0.1926	244	2.7E-3	2.6E-3	0.25	1.1	0.25	None	585
Rinsed TS	19.5 weeks	0.00	11.32*	N2-10***	110223	0.1981	49	4E-4	3E-4	0.17	0.8	0.17	TF-327	588
Vessel Acid Soak		0.500	32.10	N2-10****	114785	0.1946	232	2.0E-3	1.9E-3	0.63	2.8	0.63	TF-331	609
Test Solution	N2-10	0.200	1.05	N2-10*	86402	0.1938	329	3.8E-3	3.7E-3	9.9E-3	-	9.9E-3	None	627
Rinsed TS	26 weeks	0.200	13.33*	N2-10***	344199	0.1938	116	3E-4	2E-4	7.46E-2	0.3	7.46E-2	TF-337	621
Vessel Acid Soak		1.000	30.95	N2-10****	169581	0.2007	381	2.2E-3	2.1E-3	0.32	1.4	0.32	TF-359	634
Test Solution	N2-10	0.05	0.98	N2-10*	171679	0.1974	121	7E-4	5E-4	5.3E-2	0.2	5.3E-2	None	705
Rinsed TS	32.5 weeks	0.200	11.43*	N2-10***	173252	0.1974	187	1.1E-3	9E-4	0.26	1.2	0.26	TF-352	704
Vessel Acid Soak		1.000	29.22	N2-10****	188392	0.1999	145	1.0E-3	8E-4	0.12	0.5	0.12	TF-365	827
Test Solution	N2-10	0.05	1.15	N2-10*	114686	0.1999	194	1.7E-3	1.5E-3	0.17	0.7	0.17	None	810
Rinsed TS	39 weeks	0.200	11.46*	N2-10***	170772	0.2009	166	1.0E-3	8E-4	0.22	1.0	0.22	TF-376	797
Vessel Acid Soak		2.000	29.88	N2-10****	142312	0.1999	588	4.0E-3	3.8E-3	0.28	1.2	0.28	TF-382	831
Test Solution	N2-10	0.05	1.16	N2-10*	198474	0.1969	38	2E-4	0	0	0	0	None	855
Rinsed TS	45.5 weeks	0.200	11.42*	N2-10***	279567	0.1969	50	2E-4	0	0	0	0	TF-396	851
Vessel Acid Soak		2.000	28.97	N2-10****	156586	0.1940	104	7E-4	5E-4	4.0E-2	0.2	4.0E-2	TF-400	860
Test Solution	N2-10	0.05	1.15	N2-10*	159432	0.1912	295	1.9E-3	1.7E-3	0.21	0.9	0.21	None	943
Rinsed TS	52 weeks	0.200	11.55	N2-10***	159447	0.1912	101	6E-4	5E-4	0.13	0.6	0.13	TF-425	944
Vessel Acid Soak		2.000	37.41	N2-10****	234166	0.2030	630	2.7E-3	2.5E-3	0.23	1.0	0.23	TF-411	954

Cont'd

Table AII-2b (Cont'd)

Aliquot Description	Test #	Sol'n Volume Counted (ml.)	Total Solution Volume (ml.)	Aliquot ID	Live Time (sec)	Detector Efficiency	Counts	Raw c/s	Pu			Solution Count ID
									Back-ground Corrected c/s	Efficiency and Volume Corrected d/s	gm $\times 10^{-10}$	
Test Solution	N2-10	0.100	1.67	N2-10*	241803	0.1934	90	4E-4	2E-4	2.09E-2	0.1	None
Rinsed TS	65 weeks	0.200	12.23*	N2-10***	241882	0.2072	124	5E-4	3E-4	9.76E-2	0.4	TF-448
Vessel Acid Soak		2.000	31.87	N2-10****	249219	0.2018	1050	4.2E-3	4.0E-3	0.32	1.4	TF-444
Test Solution	N2-12	0.050	0.96	N2-12*	89867	0.1981	1205	1.34E-2	1.33E-2	1.29	5.7	None
Rinsed TS	6.5 weeks	0.100	10.83*	N2-12***	140087	0.1939	531	3.6E-3	3.4E-3	2.02	8.9	TF-329
Vessel Acid Soak		0.500	31.96	N2-12****	173493	0.1946	493	2.8E-3	2.7E-3	0.88	3.9	TF-333
Test Solution	N2-12	0.200	1.05	N2-12*	70523	0.1938	2816	3.99E-2	3.98E-2	1.00	4.8	None
Rinsed TS	13 weeks	0.200	11.57*	N2-12***	167603	0.1979	690	4.1E-3	4.0E-3	1.43	6.3	TF-339
Vessel Acid Soak		1.000	30.98	N2-12****	70937	0.1929	344	4.9E-3	4.7E-3	0.75	3.3	TF-371
Test Solution	N2-12	0.05	1.03	N2-12*	228272	0.1904	486	2.2E-3	2.0E-3	0.22	1.0	None
Rinsed TS	19.5 weeks	0.200	11.39*	N2-12***	228271	0.1974	233	1.0E-3	9E-4	0.25	1.1	TF-354
Vessel Acid Soak		1.000	31.15	N2-12****	94063	0.1929	298	3.2E-3	3.0E-3	0.49	2.2	TF-367
Test Solution	N2-12	0.05	1.16	N2-12*	230729	0.1929	486	2.1E-3	2.0E-3	0.24	1.1	None
Rinsed TS	26 weeks	0.200	12.37*	N2-12***	230727	0.1999	305	1.3E-3	1.1E-3	0.35	1.5	TF-376
Vessel Acid Soak		2.000	30.38	N2-12****	154556	0.1909	834	5.4E-3	5.3E-3	0.42	1.9	TF-384
Test Solution	N2-12	0.05	1.10	N2-12*	324163	0.1909	258	8E-4	7E-4	7.6E-2	0.3	None
Rinsed TS	32.5 weeks	0.200	11.48*	N2-12***	324158	0.1969	734	2.3E-3	2.1E-3	0.61	2.7	TF-398
Vessel Acid Soak		2.000	27.82	N2-12****	143752	0.1969	724	5.0E-3	5.0E-3	0.34	1.5	TF-402
Test Solution	N2-12	0.05	1.22	N2-12*	257510	0.1912	173	7E-4	5E-4	6.9E-2	0.3	None
Rinsed TS	39 weeks	0.200	10.62*	N2-12***	257525	0.1972	194	8E-4	6E-4	1.5E-1	0.7	TF-407
Vessel Acid Soak		2.000	32.06	N2-12****	173134	0.1938	910	5.3E-3	5.1E-3	4.2E-1	1.9	TF-413
Test Solution	N2-12	0.05	1.26	N2-12*	348705	0.1909	167	5E-4	3E-4	4.6E-2	0.2	None
Rinsed TS	45.5 weeks	0.200	10.35*	N2-12***	348681	0.1973	159	5E-4	3E-4	7.0E-2	0.3	TF-428
Vessel Acid Soak		2.000	32.45	N2-12****	135809	0.1973	413	3.0E-3	2.9E-3	2.3E-1	1.0	TF-430
Test Solution	N2-12	0.05	1.07	N2-12*	170800	0.1934	299	1.8E-3	1.6E-3	1.79E-1	0.8	None
Rinsed TS	52 weeks	0.200	11.62*	N2-12***	170804	0.2022	197	1.2E-3	1.0E-3	2.77E-1	1.2	TF-442
Vessel Acid Soak		2.000	35.13	N2-12****	174860	0.1954	571	3.3E-3	3.1E-3	2.82E-1	1.2	TF-446

Cont'd

Table AII-2b (Cont'd)

Pu													
Aliquot Description	Test #	Sol'n Volume Counted (ml.)	Total Solution Volume (ml.)	Aliquot ID	Live Time (sec)	Detector Efficiency	Counts	Raw c/s	Back-ground Corrected c/s	Efficiency and Volume Corrected d/s	gm-10	Solution ID	Count ID
Test Solution	N2-11	0.100	~0.8	N2-11*	336886	0.1958	60	2E-4	0	0	0	None	499
Vessel Acid Soak	6.5 weeks	0.500	31.29	N2-11***	888246	0.1958	154	2E-4	0	0	0	TF-289	501
Test Solution	N2-11	0.050	0.89	N2-11*								None	513
Vessel Acid Soak	13 weeks	0.100	30.76	N2-11***								TF-311	514
Test Solution	N2-11	0.050	0.88	N2-11*	173573	0.1938	24	1E-4	0	0	0	None	609
Rinsed TS	19.5 weeks	0.100	10.70	N2-11***	248925	0.1946	43	2E-4	0	0	0	TF-328	606
Vessel Acid Soak		0.500	30.60	N2-11****	184243	0.1999	30	2E-4	0	0	0	TF-332	814
Test Solution	N2-11	0.200	0.88	N2-11*	99769	0.2007	16	2E-4	2E-4	0	0	None	626
Rinsed TS	26 weeks	0.200	13.54*	N2-11***	258999	0.2007	54	2E-4	2E-4	0	0	TF-338	630
Vessel Acid Soak		1.000	30.93	N2-11****	184247	0.1929	24	1E-4	1E-4	0	0	TF-370	813
Test Solution	N2-11	0.050	1.09	N2-11*	166353	0.1925	48	3E-4	2E-4	1.8E-2	0.1	None	799
Rinsed TS	32.5 weeks	0.200	11.52*	N2-11***	166225	0.2009	36	2E-4	0	0	0	TF-353	800
Vessel Acid Soak		1.000	32.54	N2-11****	94056	0.1999	15	2E-4	0	0	0	TF-366	829
Test Solution	N2-11	0.050	0.98	N2-11*	166108	0.1925	19	1E-4	0	0	0	None	791
Rinsed TS	36 weeks	0.200	11.44*	N2-11***	166271	0.2009	26	2E-4	0	0	0	TF-377	790
Vessel Acid Soak		2.000	31.86	N2-11****	154593	0.1969	32	2E-4	0	0	0	TF-383	837
Test Solution	N2-11	0.05	0.72	N2-11*	166056	0.1946	19	1E-4	0	0	0	None	862
Rinsed TS	45.5 weeks	0.200	11.36*	N2-11***	166059	0.1950	36	2E-4	0	0	0	TF-397	863
Vessel Acid Soak		2.000	29.84	N2-11****	156562	0.1950	27	2E-4	0	0	0	TF-401	861
Test Solution	N2-11	0.50	0.56	N2-11*	173602	0.1938	32	2E-4	1E-4	3E-3	0.01	None	957
Rinsed TS	52 weeks	0.200	11.51*	N2-11***	173703	0.2036	34	2E-4	0	0	0	TF-406	958
Vessel Acid Soak		2.000	26.81	N2-11****	173214	0.2030	25	1E-4	0	0	0	TF-412	956
Test Solution	N2-11	0.05	0.66	N2-11*	253048	0.1909	53	2E-4	0	0	0	None	1000
Rinsed TS	58.5 weeks	0.200	11.73*	N2-11***	253009	0.1973	40	2E-4	0	0	0	TF-427	1001
Vessel Acid Soak		2.000	34.82	N2-11****	135825	0.1909	29	2E-4	1E-4	8E-3	4E-2	TF-428	1006

Follow the same procedure as described for Nb but use $(2.27 \times 10^5 \text{ d/s}) / (\text{gm Pu})$ during the conversion from d/s to gm of Pu.

Table AII-2c. Solution Results (Americium) for the N2 Test Series

		Am									
Aliquot Description	Test #	Sol'n Volume Counted (mL)	Total Solution Volume (mL)	Aliquot ID	Live Time (sec)	Detector Efficiency	Counts	Raw c/s	Background Corrected c/s	Efficiency ^a and Volume Corrected d/s	Solution Count ID
Test Solution (TS)	N2-1	0.850	1.58	N2-1	142998	0.1907	1678	1.17E-2	1.07E-2	1.77	589
wp Rinse + TS		0.100	10.86*	N2-1*	86399	0.1907	915	1.06E-2	9.6E-3	5.43	591
Filtered		0.150	10.86*	N2-1**	355110	0.1948	161	5E-4	1E-4	0	602
Vessel Acid Soak		0.100	32.85	N2-1***	257068	0.1939	749	2.9E-3	1.9E-3	3.16	599
Test Solution	N2-2	0.850	1.36	N2-2	142994	0.1981	445	3.1E-3	2.7E-3	0.38	590
wp Rinse + TS		0.100	11.00*	N2-2*	86394	0.1981	1332	1.54E-2	1.50E-2	8.37	592
Filtered		0.150	11.00*	N2-2**	248944	0.1938	309	1.2E-3	2E-4	7.2E-2	605
Vessel Acid Soak		0.100	32.83	N2-2***	257041	0.1946	1589	6.8E-3	6.1E-3	10.5	600
Test Solution	N2-3	0.200	2.34	N2-3*	13212	0.1938	878	6.5E-2	6.54E-2	3.95	625
wp Rinse + TS		0.200	15.50*	N2-3**	20837	0.1938	1003	4.8E-3	4.7E-3	18.8	616
Filtered		0.350	15.50*	N2-3**	59388	0.1938	138	2.3E-3	1.3E-3	0.29	618
Vessel Acid Soak		1.000	25.43	N2-3****	81577	0.1938	2542	3.12E-2	3.01E-2	4.57	635
Test Solution	N2-4	0.200	3.70	N2-4*	15704	0.1938	499	3.18E-2	3.14E-2	2.93	619
wp Rinse + TS		0.200	11.22*	N2-4***	12119	0.2007	852	7.03E-2	6.99E-2	19.5	628
Filtered		0.300	11.22*	N2-4**	72842	0.2007	47	6E-4	3E-4	4.7E-2	623
Vessel Acid Soak		0.500	28.63	N2-4****	81576	0.2007	5487	6.73E-2	6.69E-2	9.54	636
Test Solution	N2-5	0.200	4.17	N2-5*	54812	0.1925	315	5.7E-3	4.7E-3	0.51	786
wp Rinse + TS		0.200	14.39*	N2-5***	25011	0.1925	1171	4.68E-2	4.53E-2	17.1	788
Filtered		0.350	14.39*	N2-5**	166261	0.1929	292	7.8E-3	7E-4	0.15	807
Vessel Acid Soak		1.000	32.55	N2-5****	86087	0.1929	4152	5.2E-2	5.09E-2	4.9	832
Test Solution	N2-6	0.200	4.17	N2-6*	54843	0.2009	507	9.2E-3	8.8E-3	0.91	787
wp Rinse + TS		0.200	13.97*	N2-6***	25011	0.2009	431	1.72E-2	1.68E-2	5.83	789
Filtered		0.400	13.97*	N2-6**	166263	0.1999	109	7E-4	3E-4	4.3E-2	808
Vessel Acid Soak		2.000	30.17	N2-6****	86086	0.1999	2449	3.06E-2	3.02E-2	2.12	833
Test Solution	N2-7	0.200	5.86	N2-7*	87249	0.1912	2719	3.12E-2	3.01E-2	4.61	939
wp Rinse + TS		0.200	15.81	N2-7***	79215	0.1912	2128	2.69E-2	2.58E-2	10.65	941
Filtered		0.250	15.81	N2-7**	104122	0.1912	129	1.2E-3	1E-4	4.6E-2	935
Vessel Acid Soak		2.000	32.93	N2-7****	79031	0.1912	2413	3.05E-2	2.94E-2	2.53	945

Cont'd

Table AII-2c (Cont'd)

Am										Efficiency and Volume			
Aliquot Description	Test #	Sol'n Volume Counted (mL)	Total Solution Volume (mL)	Aliquot ID	Live Time (sec)	Detector Efficiency	Counts	Raw c/s	Back-ground Corrected c/s	Corrected d/s	gm x10 ⁻¹¹	Solution ID	Count ID
Test Solution w/ Rinse + TS Filtered	N2-8	0.200	6.73	N2-8*	87250	0.1972	4380	5.07E-2	4.98E-2	8.50	6.7	None	940
		0.200	16.25	N2-8***	79210	0.1972	2179	2.75E-2	2.71E-2	11.17	8.8	TF-409	942
		0.300	16.25	N2-8***	104127	0.1972	23	2E-4	0	0	0	TF-409	936
Vessel Acid Soak		2.000	38.10	N2-8****	79029	0.1972	4886	5.17E-2	5.13E-2	4.96	3.9	TF-415	946
Test Solution	N2-9	0.100	~0.8	N2-9*	168797	0.1456	2878	1.23E-2	1.14E-2	0.63	0.5	None	459
Test Solution	6.5 weeks	0.100	~0.8	N2-9*	21736	0.1958	374	1.72E-2	1.68E-2	0.69	0.5	None	453
Dil/Filt TS		0.050	~0.8	N2-9**	334863	0.1456	232	7E-4	0	0	0	None	496
Vessel Acid Soak		0.500	31.62	N2-9***	336717	0.1456	262	~E-4	0	0	0	TF-287	498
Test Solution	N2-9	0.050	0.82	N2-9*	84400	0.1939	271	3.2E-3	2.2E-3	0.19	0.8	None	595
Vessel Acid Soak	13 weeks	0.100	33.91	N2-9***	172804	0.1939	406	2.3E-3	1.3E-3	2.32	1.8	TF-307	600
Test Solution	N2-9	0.050	0.71	N2-9*	55725	0.1981	119	2.1E-3	1.7E-3	0.12	0.1	None	580
Rinsed TS	19.5 weeks	0.100	11.26*	N2-9***	110214	0.1926	158	1.4E-3	4E-4	0.26	0.2	TF-326	587
Vessel Acid Soak		0.500	31.41	N2-9****	114777	0.1938	696	6.1E-3	5.1E-3	1.65	1.3	TF-336	607
Test Solution	N2-9	0.200	1.10	N2-9*	258167	0.2007	1340	5.2E-3	4.8E-3	0.13	0.1	None	624
Rinsed TS	26 weeks	0.200	13.38*	N2-9***	271218	0.1938	444	1.6E-3	6E-4	0.20	0.2	TF-336	629
Vessel Acid Soak		1.000	31.51	N2-9****	169503	0.1938	581	3.4E-3	2.4E-3	0.37	0.3	TF-368	633
Test Solution	N2-9	0.050	0.88	N2-9*	171680	0.1904	296	1.7E-3	7E-4	6.29E-2	-	None	705
Rinsed TS	32.5 weeks	0.200	11.26*	N2-9***	173311	0.1904	237	1.4E-3	3E-4	9.4E-2	0.1	TF-351	703
Vessel Acid Soak		1.000	29.89	N2-9****	188386	0.1929	417	2.2E-3	1.2E-2	0.18	0.1	TF-354	626
Test Solution	N2-9	0.050	1.20	N2-9*	114698	0.1929	684	5.9E-3	4.9E-3	0.61	0.5	None	809
Rinsed TS	39 weeks	0.200	11.60*	N2-9***	170774	0.1925	585	3.4E-3	2.4E-3	0.72	0.6	TF-375	796
Vessel Acid Soak		2.000	32.07	N2-9****	142317	0.1929	919	6.5E-3	5.4E-3	0.45	0.4	TF-381	830
Test Solution	N2-9	0.050	1.00	N2-9*	198443	0.1909	213	1.1E-3	0	0	0	None	854
Rinsed TS	45.5 weeks	0.200	11.24*	N2-9***	279527	0.1909	338	1.2E-3	2E-4	4.7E-2	-	TF-395	850
Vessel Acid Soak		2.000	31.76	N2-9****	143424	0.1909	231	1.6E-3	6E-4	4.7E-2	-	TF-399	856
Test Solution	N2-9	0.050	1.33	N2-9*	258150	0.1912	4767	1.85E-2	1.74E-2	2.42	1.9	None	937
Rinsed TS	52 weeks	0.200	11.16*	N2-9***	258145	0.1912	1696	6.6E-3	6.2E-3	1.75	1.4	TF-404	938
Vessel Acid Soak		2.000	36.84	N2-9****	234167	0.1938	2802	1.2E-2	1.09E-2	1.03	0.8	TF-410	953

Cont'd

Table AII-2c (Cont'd)

Aliquot Description	Test #	Sol'n Volume Counted (mL)	Total Solution Volume (mL)	Aliquot ID	Live Time (sec)	Detector Efficiency	Counts	Raw c/s	Efficiency and Volume			Solution Count ID
									Back-ground Corrected c/s	Corrected d/s	gm x10 ⁻¹¹	
Test: Solution	N2-9	0.100	1.0**	N2-9*	115928	0.1930	564	4.9E-3	3.8E-3	0.31	0.2	None
Rinsed TS	65 weeks	0.200	12.16*	N2-9***	145345	0.1973	190	1.3E-3	9E-4	0.28	0.2	TF-439
Vessel Acid Soak		2.000	33.08	N2-9***	249221	0.1954	1635	6.6E-3	5.5E-3	0.47	0.4	TF-443
Test: Solution	N2-10	0.100	~0.8	N2-10*	169042	0.1958	76	4E-4	0	0	0	None
Dil/Filt TS	6.5 weeks	0.050	~0.8	N2-10**	334861	0.1958	123	4E-4	0	0	0	None
Vessel Acid Soak		0.500	32.97	N2-10***	237247	0.1958	92	4E-4	0	0	0	TF-288
Test: Solution	N2-10	0.050	1.00	N2-10*	84394	0.1946	39	4E-4	0	0	0	None
Vessel Acid Soak	13 weeks	0.190	32.82	N2-10***	140929	0.1938	152	1.1E-3	0	0	0	TF-308
Test: Solution	N2-10	0.050	0.93	N2-10*	89089	0.1926	400	4.5E-3	4.1E-3	0.39	0.3	None
Rinsed TS	19.5 weeks	0.100	11.32*	N2-10***	110723	0.1981	71	6E-4	3E-4	0.16	0.1	TF-327
Vessel Acid Soak		0.500	32.10	N2-10***	114785	0.1946	555	4.8E-3	4.4E-3	1.47	1.2	TF-331
Test: Solution	N2-10	0.200	1.05	N2-10*	86402	0.1938	909	1.05E-2	9.5E-3	0.26	0.2	None
Rinsed TS	26 weeks	0.200	13.33*	N2-10***	344199	0.1938	539	1.6E-3	5E-4	0.18	0.1	TF-337
Vessel Acid Soak		1.000	30.95	N2-10***	169501	0.2007	501	3.0E-3	2.6E-3	0.40	0.3	TF-369
Test: Solution	N2-10	0.05	0.98	N2-10*	171679	0.1974	567	3.3E-3	3.0E-3	0.29	0.3	None
Rinsed TS	32.5 weeks	0.200	11.43*	N2-10***	173252	0.1974	435	2.5E-3	2.2E-3	0.63	0.5	TF-352
Vessel Acid Soak		1.000	29.22	N2-10***	188392	0.1999	284	2.0E-3	1.6E-3	0.23	0.2	TF-365
Test: Solution	N2-10	0.05	1.10	N2-10*	114686	0.1999	938	8.7E-3	8.3E-3	0.91	0.7	None
Rinsed TS	39 weeks	0.200	11.46*	N2-10***	170772	0.2009	561	3.3E-3	2.9E-3	0.82	0.6	TF-376
Vessel Acid Soak		2.000	29.88	N2-10***	142312	0.1999	912	6.4E-3	6.0E-3	0.45	0.4	TF-382
Test: Solution	N2-10	0.05	1.16	N2-10*	198474	0.1969	82	4E-4	0	0	0	None
Rinsed TS	45.5 weeks	0.200	11.42*	N2-10***	279567	0.1969	102	4E-4	0	0	0	TF-396
Vessel Acid Soak		2.000	28.97	N2-10***	156506	0.1940	304	1.9E-3	9E-4	6.7E-2	-	TF-400
Test: Solution	N2-10	0.05	1.16	N2-10*	159432	0.1912	1226	7.7E-3	6.6E-3	0.80	0.6	None
Rinsed TS	52 weeks	0.200	11.55	N2-10***	159447	0.1912	397	2.5E-3	2.1E-3	0.61	0.5	TF-405
Vessel Acid Soak		2.000	37.41	N2-10***	234166	0.2030	256	3.2E-3	7.8E-3	0.26	0.2	TF-411

Cont'd

Table AII-2c (Cont'd)

Aliquot Description	Test #	Sol'n Volume Counted (mL)	Total Solution Volume (mL)	Aliquot ID	Live Time (sec)	Detector Efficiency	Counts	Raw c/s	Efficiency and Volume Corrected			Solution ID	Count ID
									Back-ground Corrected c/s	d/s	gm x10 ⁻¹¹		
Test Solution	N2-10	0.100	1.67	N2-10*	241803	0.1934	362	1.5E-3	4E-4	3.4E-2	0.03	None	1036
Rinsed TS	65 weeks	0.200	12.23*	N2-10***	241802	0.2022	180	7E-4	3E-4	1.04E-1	0.1	TF-440	1037
Vessel Acid Soak		2.000	31.87	N2-10***	249219	0.2018	1857	7.5E-3	7.1E-3	0.56	0.4	TF-444	1082
Test Solution	N2-12	0.050	0.96	N2-12*	89867	0.1981	1434	1.60E-2	1.58E-2	1.51	1.2	None	586
Rinsed TS	6.5 weeks	0.100	10.83*	N2-12***	148007	0.1939	735	4.9E-3	4.01E-3	2.23	1.8	TF-329	597
Vessel Acid Soak		0.500	31.96	N2-12***	173493	0.1946	727	4.2E-3	3.8E-3	1.26	1.0	TF-333	610
Test Solution	N2-12	0.200	1.05	N2-12*	70523	0.1938	3381	4.79E-2	4.69E-2	1.27	1.0	None	620
Rinsed TS	13 weeks	0.200	11.57*	N2-12***	167003	0.1979	822	4.9E-3	4.6E-3	1.04	1.3	TF-339	617
Vessel Acid Soak		1.000	30.98	N2-12***	70037	0.1929	459	6.5E-3	5.4E-3	0.87	0.7	TF-371	815
Test Solution	N2-12	0.05	1.03	N2-12*	226272	0.1904	873	3.9E-3	2.8E-3	0.36	0.2	None	707
Rinsed TS	19.5 weeks	0.200	11.39*	N2-12***	226271	0.1974	340	1.5E-3	1.2E-3	0.33	0.3	TF-354	708
Vessel Acid Soak		1.000	31.15	N2-12***	94063	0.1929	719	7.6E-3	6.6E-3	1.06	0.8	TF-367	828
Test Solution	N2-12	0.05	1.16	N2-12*	230229	0.1929	973	3.8E-3	2.7E-3	0.33	0.3	None	811
Rinsed TS	26 weeks	0.200	12.37*	N2-12***	230227	0.1999	453	2.0E-3	1.6E-3	0.50	0.4	TF-378	812
Vessel Acid Soak		2.000	30.38	N2-12***	154556	0.1909	1598	1.03E-2	9.3E-3	0.74	0.6	TF-384	836
Test Solution	N2-12	0.05	1.10	N2-12*	324163	0.1909	693	2.1E-3	1.1E-3	0.13	0.1	None	852
Rinsed TS	32.5 weeks	0.200	11.48*	N2-12***	324158	0.1909	891	2.8E-3	2.4E-3	0.69	0.5	TF-398	853
Vessel Acid Soak		2.000	27.82	N2-12***	143752	0.1969	1273	8.8E-3	8.4E-3	0.60	0.5	TF-402	857
Test Solution	N2-12	0.05	1.22	N2-12*	257510	0.1912	582	2.3E-3	1.2E-3	0.15	0.1	None	947
Rinsed TS	39 weeks	0.200	10.62*	N2-12***	257525	0.1972	370	1.4E-3	1.0E-3	0.28	0.2	TF-407	948
Vessel Acid Soak		2.000	32.06	N2-12***	173134	0.1938	747	4.3E-3	3.2E-3	0.27	0.2	TF-413	955
Test Solution	N2-12	0.05	1.26	N2-12*	348705	0.1909	623	1.8E-3	7E-4	9.1E-2	0.1	None	998
Rinsed TS	45.5 weeks	0.200	10.35*	N2-12***	348681	0.1973	265	8E-4	4E-4	9.4E-2	0.1	TF-428	999
Vessel Acid Soak		2.000	32.45	N2-12***	135909	0.1973	573	4.2E-3	3.8E-3	3.14E-1	0.2	TF-430	1007
Test Solution	N2-12	0.05	1.07	N2-12*	170800	0.1934	741	4.3E-3	3.2E-3	3.58E-1	0.3	None	1038
Rinsed TS	52 weeks	0.200	11.62*	N2-12***	170804	0.2022	343	2.0E-3	1.6E-3	4.62E-1	0.4	TF-442	1039
Vessel Acid Soak		2.000	35.13	N2-12***	170800	0.1954	1045	6.0E-3	4.9E-3	4.40E-1	0.3	TF-446	1083

Cont'd

Table AII-2c (Cont'd)

A ₀													
Aliquot Description	Test #	Sol'n Volume Counted (mL)	Total Solution Volume (mL)	Aliquot ID	Live Time (sec)	Detector Efficiency	Counts	Raw c/s	Back-ground Corrected c/s	Efficiency and Volume Corrected d/s	gm x10 ⁻¹¹	Solution ID	Count ID
Test Solution	N2-11	0.100	0.8	N2-11*	336886	0.1958	113	3E-4	0	0	0	None	499
Vessel Acid Soak	6.5 weeks	?	31.29	N2-11***	888246	0.1958	310	4E-4	0	0	0	TF-289	501
Test Solution	N2-11	0.050	0.89	N2-11*								None	513
Vessel Acid Soak	13 weeks	0.100	30.76	N2-11***								TF-311	514
Test Solution	N2-11	0.050	0.88	N2-11*	173573	0.1938	178	1.0E-3	0	0	0	None	609
Rinsed TS	19.5 weeks	0.100	10.70	N2-11***	248925	0.1946	94	4E-4	0	0	0	TF-328	606
Vessel Acid Soak		0.500	30.60	N2-11****	184243	0.1999	82	4E-4	0	0	0	TF-332	814
Test Solution	N2-11	0.200	0.88	N2-11*	99769	0.2007	36	4E-4	0	0	0	None	626
Rinsed TS	26 weeks	0.200	13.54*	N2-11***	253999	0.2007	103	4E-4	0	0	0	TF-338	630
Vessel Acid Soak		1.000	30.93	N2-11****	184247	0.1929	183	1.0E-3	0	0	0	TF-370	813
Test Solution	N2-11	0.050	1.09	N2-11*	166350	0.1925	177	1.1E-3	0	0	0	None	799
Rinsed TS	32.5 weeks	0.200	11.52*	N2-11***	166225	0.2009	87	5E-4	1E-4	3.5E-2	-	TF-353	800
Vessel Acid Soak		1.000	32.54	N2-11****	94056	0.1999	32	3E-4	0	0	0	TF-366	829
Test Solution	N2-11	0.050	0.98	N2-11*	166108	0.1925	166	1.0E-3	0	0	0	None	791
Rinsed TS	39 weeks	0.200	11.44*	N2-11***	166271	0.2009	52	3E-4	0	0	0	TF-377	793
Vessel Acid Soak		2.000	31.86	N2-11****	154593	0.1969	64	4E-4	0	0	0	TF-383	837
Test Solution	N2-11	0.05	0.72	N2-11*	166056	0.1940	168	1.0E-3	0	0	0	None	862
Rinsed TS	45.5 weeks	0.200	11.36*	N2-11***	166059	0.1990	59	4E-4	0	0	0	TF-397	863
Vessel Acid Soak		2.000	29.84	N2-11****	156562	0.1990	66	4E-4	0	0	0	TF-401	861
Test Solution	N2-11	0.05		N2-11*	173602	0.1938	180	1.0E-3	0	0	0	None	957
Rinsed TS	52 weeks	0.200		N2-11***	173703	0.2030	65	4E-4	0	0	0	TF-406	958
Vessel Acid Soak		2.000		N2-11****	173214	0.2030	68	4E-4	0	0	0	TF-412	956
Test Solution	N2-11	0.05	0.66	N2-11*	253048	0.1909	260	1.0E-3	0	0	0	None	1000
Rinsed TS	58.5 weeks	0.200	11.73*	N2-11***	253009	0.1973	114	5E-4	1E-4	1.5E-2	1E-2	TF-427	1001
Vessel Acid Soak		2.000	34.82	N2-11****	135825	0.1909	171	1.3E-1	2E-4	1.5E-2	1E-2	TF-429	1006

a) Follow the same procedure as described for Np but use $(1.27 \text{ d/s}) / (\text{gm A})$ during the conversion from d/s to gm for A.

Table AII-3a. (Neptunium)

Aliquot Description	Test #	Sol'n Volume Counted (mL)	Total Sol'n Volume (mL)	Aliquot ID	Live Time (sec)	Detector Efficiency	Counts	Raw c/s	BGC c/s	Np		Solution ID	Count #
										Effcy & Vol Corrected d/s	g x 10 ⁻⁸		
N2 #2 btm can 1st	N2 #2	0.1	11.42		148732	0.1930	150						1068
N2 #2 top can 1st	N2 #2	0.1	7.47		86062	0.1930	60						1064
N2 #2 top can 2nd	N2 #2	0.1	9.68		142369	0.2018	115						1066
N2 #4 btm can 1st	N2 #4	0.1	11.98		7029	0.2018	20						1080
N2 #4 top can 1st	N2 #4	0.1	12.16		148728	0.2018	200						1069
N2 #6 btm can 1st	N2 #6	0.1	13.57		143966	0.1930	20						1061
N2 #6 top can 1st	N2 #6	0.1	35.65		33304	0.1930	15						1059
N2 #6 top can 2nd	N2 #6	0.1	11.90		26933	0.2018	10						1054
N2 #8 btm can 1st	N2 #8	0.1	32.50		33302	0.2018	30						1060
N2 #8 top can 1st	N2 #8	0.1	12.04		26934	0.1930	10						1053
N2 #8 top can 2nd	N2 #8	0.1	31.78		81751	0.1930	40						1057
N2 #8 top can 2nd	N2 #8	0.1	10.79		59528	0.1930	20						1052
N2 #8 top can 2nd	N2 #8	0.1	35.03		81748	0.2018	50						1058
Analogs III #1 btm can		0.1	21.20		55187	0.2018	20						1056
top can		0.1	19.16		88184	0.1930	20						1055

Table AII-3b. (Plutonium)

Aliquot Description	Test #	Sol'n Volume Counted (mL)	Total Sol'n Volume (mL)	Aliquot ID	Live Time (sec)	Detector Efficiency	Counts	Raw c/s	BGC c/s	Pu			Solution ID	Count #
										Effcy %	Vol Corrected d/s	g x 10 ⁻⁸		
N2 #2 btm can 1st	N2 #2	0.1	11.42		148732	0.1930	6021	4.05E-2	4.04E-2	23.9	105			1068
N2 #2 top can 1st	N2 #2	0.1	7.47		86062	0.1930	2933	3.41E-2	3.40E-2	13.1	57.7			1064
N2 #2 top can 2nd	N2 #2	0.1	9.68		142369	0.2018	5879	4.13E-2	4.11E-2	19.7	86.9			1066
N2 #4 btm can 1st	N2 #4	0.1	11.98		7029	0.2018	744	1.06E-1	1.06E-1	62.7	276			1080
N2 #4 top can 1st	N2 #4	0.1	12.16		148728	0.2018	9828	6.61E-2	6.59E-2	39.7	175			1069
N2 #6 btm can 1st	N2 #6	0.1	13.57		143966	0.1930	2015	1.04E-2	1.39E-2	9.7	42.9			1061
N2 #6 top can 1st	N2 #6	0.1	35.65		33304	0.1930	733	2.20E-2	2.19E-2	40.4	178			1059
N2 #6 top can 2nd	N2 #6	0.1	11.90		26933	0.2018	1664	6.18E-2	6.16E-2	36.3	160			1054
N2 #8 btm can 1st	N2 #8	0.1	32.50		33302	0.2018	785	2.36E-2	2.34E-2	37.6	166			1060
N2 #8 top can 1st	N2 #8	0.1	12.04		26934	0.1930	1115	4.14E-2	4.13E-2	25.7	113			1053
N2 #8 top can 2nd	N2 #8	0.1	31.78		81751	0.1930	1878	2.3E-2	2.28E-2	37.6	166			1057
N2 #8 top can 1st	N2 #8	0.1	10.79		59528	0.1930	3696	6.21E-2	6.20E-2	34.6	153			1052
N2 #8 top can 2nd	N2 #8	0.1	35.03		81748	0.2018	2300	2.81E-2	2.79E-2	48.5	214			1058
Analogs III #1														
btm can		0.1	21.20		55187	0.2018	399	7.2E-3	7.0E-3	7.4	32.5			1056
top can		0.1	19.16		88184	0.1930	406	7.4E-3	7.2E-3	7.2	31.6			1055

Table AII-3c. (Americium)

Aliquot Description	Test #	Sol'n Volume Counted (mL)	Total Sol'n Volume (mL)	Aliquot ID	Live Time (sec)	Detector Efficiency	Counts	Raw c/s	BGC c/s	Effcy & Vol Corrected d/s	g x 10 ⁻⁸	Solution ID	Count #
N2 #2 bta can 1st	N2 #2	0.1	11.42		148732	0.1930	17062	1.15E-1	1.14E-1	67.3	53.0		1068
N2 #2 top can 1st	N2 #2	0.1	7.47		88062	0.1930	6207	7.21E-2	7.11E-2	27.5	21.7		1064
N2 #2 top can 2nd	N2 #2	0.1	9.68		142369	0.2018	11372	7.99E-2	7.95E-2	38.1	30.0		1066
N2 #4 bta can 1st	N2 #4	0.1	11.98		7029	0.2018	1920	2.73E-1	2.73E-1	162	127		1080
N2 #4 top can 1st	N2 #4	0.1	12.16		148728	0.2018	34726	2.33E-1	2.33E-1	140	111		1069
N2 #6 bta can 1st	N2 #6	0.1	13.57		143966	0.1930	3399	2.36E-2	2.26E-2	15.9	12.5		1061
N2 #6 top can 1st	N2 #6	0.1	35.65		33304	0.1930	1699	5.10E-2	5.00E-2	92.3	72.7		1059
N2 #6 top can 2nd	N2 #6	0.1	11.90		26933	0.2018	3921	1.46E-1	1.45E-1	85.6	67.4		1054
N2 #8 bta can 1st	N2 #8	0.1	32.50		33302	0.2018	2129	6.39E-2	6.35E-2	102.3	80.6		1060
N2 #8 top can 1st	N2 #8	0.1	12.04		26934	0.1930	1923	7.14E-2	7.03E-2	43.9	34.6		1053
N2 #8 top can 2nd	N2 #8	0.1	31.78		81751	0.1930	4496	5.5E-2	5.39E-2	88.8	69.9		1057
N2 #8 top can 1st	N2 #8	0.1	10.79		59528	0.1930	8386	1.41E-1	1.40E-1	78.2	61.6		1052
N2 #8 top can 2nd	N2 #8	0.1	35.03		81748	0.2018	5166	6.32E-2	6.28E-2	109	85.8		1058
Analog III #1 bta can top can		0.1	21.20		55187	0.2018	593	1.07E-2	1.03E-1	10.9	8.6		1056
		0.1	19.16		88184	0.1930	895	1.62E-2	1.52E-2	15.1	11.9		1055

- END -

DATE FILMED

11 / 02 / 90