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Distribution Category:
Nuclear Waste Management
(UC-70)

ANL--82-10

DE83 003340

ANL-82-10

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THE MEASUREMENT OF LEACHING FROM SIMULATED
NUCLEAR-WASTE GLASS USING RADIOTRACERS

by

J. K. Bates, L. J. Jardine,* and M. J. Steindler

Chemical Engineering Division

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September 1982

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TABLE OF CONTENTS

	<u>Page</u>
ABSTRACT	1
I. INTRODUCTION	1
II. EXPERIMENTAL PROCEDURES	4
A. Experimental Approach	4
B. Glass Preparation	6
C. Leach Test Procedure	13
III. RESULTS AND DISCUSSION	13
A. SRL 211** Glass	13
B. SRL 211*** Glass	18
C. SRL 131* Glass	22
D. Utility and Sensitivity of the Test Methods	26
IV. CONCLUSIONS	27
REFERENCES	29
APPENDIX	30
1.0 TEST CONDITIONS, SPECIMEN PREPARATION, AND DESCRIPTION	30
1.1 Materials Composition	30
1.2 Test Equipment	30
1.3 Analytical Techniques	33
1.4 Calibrations and Standards	34
1.5 Leachant Preparation	34
1.6 Specimen Preparation and Description	34
2.0 LEACHANT ANALYSIS	34
3.0 LEACHATE ANALYSIS	34
4.0 TABULATED AND PLOTTED DATA	53
5.0 DISCUSSION	53
5.1 Counting Error	53
5.2 Procedural Error	53

LIST OF FIGURES

<u>No.</u>	<u>Title</u>	<u>Page</u>
1.	Radionuclide Composition of High-Level Liquid Waste from Reprocessing	3
2.	Flow Diagram of the General Approach Being Used for Com- paring Leach Rate Results for SRL Frit 211** Waste Glass by Neutron Activation and Radioactive-Tracer Addition	7
3.	Schematic of 50-g Ingot of Radioactive Glass, Illustrating the Coding of the Wafers Cut from the Original Ingot	11
4.	Multielement Leach Results from SRL 211** Cold Glass	15
5.	Multielement Leach Results from SRL 211** Spiked/Activated Glass	16
6.	Multielement Leach Results from SRL 211** Spiked Glass	17
7.	Comparison of Cesium Leach Rates from SRL 211** Glass	18
8.	Multielement Leach Results from SRL 211*** Cold Glass	20
9.	Multielement Leach Results from SRL 211*** Activated Glass	20
10.	Multielement Leach Results from SRL 211*** Spiked Glass	21
11.	Multielement Leach Results from SRL 131* Cold Glass	22
12.	Multielement Leach Results from SRL 131* Activated Glass	24
13.	Multielement Leach Results from SRL 131* Spiked Glass	25

LIST OF TABLES

<u>No.</u>	<u>Title</u>	<u>Page</u>
1.	Estimated Total Activity and Half-Lives of the Radionuclides in the High-Level Wastes at Savannah River Plant by 1985	2
2.	Compositions of Simulated SRL Waste Glasses Used in Leach Tests	5
3.	Radioactive Isotopes Used and their Radiation Properties	8
4.	"Mass" Balance of Radioactivity Added to SRL Frit 211** Glass and Amount of Radioactivity Measured after Melting	9
5.	Consolidated Leaching Results for SRL 211** Glass	14
6.	Consolidated Leaching Results for SRL 211*** Glass	19
7.	Consolidated Leaching Results for SRL 131* Glass	23
8.	Sensitivity Limits Obtained Using Radiotracers to Measure Leach Rates from SRL Glasses	27
A1-1.	Specific Activities for Radionuclides Used in Spiked/Leach Tests	31
A3-1.	Analyses of Blanks	35
A3-2.	Leachate Analysis for DI Water Leachant, SRL 211** C Glass	36
A3-3.	Leachate Analysis for DI Water Leachant, SRL 211** A Glass	37
A3-4.	Leachate Analysis for DI Water Leachant, SRL 211** S/A Glass	38
A3-5.	Leachate Analysis for DI Water Leachant, SRL 211** S Glass	39
A3-6.	Leachate Analysis for DI Water Leachant, SRL 211*** C Glass	40
A3-7.	Leachate Analysis for DI Water Leachant, SRL 211*** A Glass	41
A3-8.	Leachate Analysis for DI Water Leachant, SRL 211*** S Glass	43

LIST OF TABLES (contd)

<u>No.</u>	<u>Title</u>	<u>Page</u>
A3-9.	Leachate Analysis for DI Water Leachant, SRL 211*** S Glass (Repeat)	45
A3-10.	Leachate Analysis for DI Water Leachant, SRL 131* C Glass	46
A3-11.	Leachate Analysis for DI Water Leachant, SRL 131* A Glass	48
A3-12.	Leachate Analysis for DI Water Leachant, SRL 131* S Glass	49
A3-13.	Leachate Analysis for DI Water Leachant, SRL 131* S Glass (Repeat)	51
A4-1.	Normalized Elemental Mass Losses, (NL) _i , for DI Water Leachant and SRL 211** C Glass	54
A4-2.	Normalized Elemental Mass Losses, (NL) _i , for DI Water Leachant and SRL 211** A Glass	55
A4-3.	Normalized Elemental Mass Losses, (NL) _i , for DI Water Leachant and SRL 211** S/A Glass	56
A4-4.	Normalized Elemental Mass Losses, (NL) _i , for DI Water Leachant and SRL 211** S Glass	57
A4-5.	Normalized Elemental Mass Loss, (NL) _i , for DI Water Leachant and SRL 211*** C Glass	58
A4-6.	Normalized Elemental Mass Loss, (NL) _i , for DI Water Leachant and SRL 211*** A Glass	59
A4-7.	Normalized Elemental Mass Loss, (NL) _i , for DI Water Leachant and SRL 211*** S Glass	60
A4-8.	Normalized Elemental Mass Loss, (NL) _i , for DI Water Leachant and SRL 211*** S Glass (Repeat)	61
A4-9.	Normalized Elemental Mass Loss, (NL) _i , for DI Water Leachant and SRL 131* C Glass	62
A4-10.	Normalized Elemental Mass Loss, (NL) _i , for DI Water Leachant and SRL 131* A Glass	63
A4-11.	Normalized Elemental Mass Loss, (NL) _i , for DI Water Leachant and SRL 131* S Glass	64

LIST OF TABLES (contd)

<u>No.</u>	<u>Title</u>	<u>Page</u>
A4-12.	Normalized Elemental Mass Loss, $(NL)_i$, for DI Water Leachant and SRL 131* S Glass (Repeat)	65
A5-1.	Observed Activities in Neutron-Activated Glass Sample L-243, 10 Observations	66

THE MEASUREMENT OF LEACHING FROM SIMULATED NUCLEAR WASTE GLASS USING RADIOTRACERS

by

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ABSTRACT

The use of radiotracer spiking as a method of measuring the leaching from simulated nuclear waste glass is shown to give results comparable with other analytical detection methods. The leaching behavior of ^{85}Sr , ^{106}Ru , ^{133}Ba , ^{137}Cs , ^{141}Ce , ^{152}Eu , and other isotopes is measured for several defense waste glasses. These tests show that radiotracer spiking is a sensitive, multielement technique that can provide leaching data, for actual waste elements, that are difficult to obtain by other methods. Additionally, a detailed procedure is described that allows spiked glass to be prepared with a suitable distribution of radionuclides.

I. INTRODUCTION

Nuclear wastes are currently being produced through the fuel cycles of (1) commercial nuclear power reactors and (2) military weapons and propulsion (e.g., nuclear submarines). Presently, it is estimated that there are over 80 million cubic feet of long-lived radioactive nuclear wastes stored at interim sites throughout the United States [DEUTCH].

The safe disposal of these wastes through long-term repository storage is of paramount importance to closing the nuclear cycle. Until repository sites are selected and ultimately placed in operation, the major concern is the release of radionuclides directly into the biosphere during interim storage, processing, and transportation. After the wastes have been enclosed in the repository, the most likely scenario for waste-constituent release is through leaching and transport by groundwater. The time frame of concern depends on the half-lives of the radionuclides, but is generally on the order of ten thousand years.

Currently, the majority of nuclear waste has evolved from weapons programs, and Savannah River Laboratory (SRL) has been charged with the expedient solution to the waste disposal problem for wastes now at that site. Although considerable research in this area is ongoing, the manner in which the waste package (i.e., "envelope" in which waste is stored) will be affected by processing and long-term storage has not been established, nor have the choices of geologic media for burial or waste form (e.g., glass pellets) been made.

Because of these uncertainties, the Nuclear Regulatory Commission has proposed regulations [NRC] that depend on the waste form and the total waste package to establish part of the necessary containment. Proposed NRC regulations for high-level waste require that there be no release of activity from the waste package for the first 1000 years. For defense waste such as that

at the Savannah River Plant, the radionuclides of interest, their half-lives, and the estimated total activity are shown in Table 1. Figure 1 illustrates the estimated radionuclide composition of such waste after various decay periods. As is evident in Fig. 1, a 1000-year time period would allow ^{137}Cs and ^{90}Sr to decay to acceptable levels. After that period, the acceptable release of activity would be no greater than ten parts per million per year of the activity remaining at a thousand years.

In order to detect such low rates of element release, sensitive analytical methods have been developed. The ideal method would incorporate simultaneous detection of the elements of interest with sufficient sensitivity, reliability, ease of use, and economy. Elements of interest include both hazardous waste nuclides and solid matrix elements. Of the methods in use, spectroscopic techniques such as inductively coupled plasma (ICP), direct-current plasma (DCP), and atomic absorption (AA) are popular because they meet most of the aforementioned criteria. However, they do not possess adequate sensitivity for certain classes of elements, including rare earths and some transition (noble) metals. Additionally, in order to obtain leach rates, these methods require an independent analysis of the solid waste form, thus possibly introducing bias to the results. Neutron activation analysis (NAA)

Table 1. Estimated Total Activity and Half-Lives of the Radionuclides in the High-Level Wastes at Savannah River Plant by 1985 [SRL-1]

Radionuclide	Total Activity, Ci	Half Life, y
^{90}Sr	1.3×10^8	29
^{137}Cs	1.3×10^8	30
^{147}Pm	4.6×10^7	2.6
$^{144}\text{Ce} - ^{144}\text{Pr}$	1.1×10^7	0.78
^{151}Sm	4.2×10^6	93
$^{106}\text{Ru} - ^{106}\text{Rh}$	1.8×10^6	1
^{238}Pu	6.0×10^5	88
^{241}Am	6.0×10^4	433
^{244}Cm	6.0×10^4	18
^{239}Pu	2.4×10^4	2.4×10^4

Fig. 1. Radionuclide Composition of High-Level Liquid Waste from Reprocessing (1 to 10^6 years after irradiation) [SRL-1]

has also been applied to measure leach rates and can be used by activating either the solid before leaching or the dried leachate. Advantages and drawbacks of this method have been presented in detail elsewhere [BATES-1; BATES-2]. While NAA makes increased sensitivity possible for many nuclides, difficulties may arise due to either sample size limitations in solid activation or γ -ray interferences that decrease the limits of detection during

counting under certain circumstances. These difficulties may be resolved by the use of radiotracer spiking of the waste form with elements of interest. Radiotracers have been used extensively in biology and chemistry to study reaction mechanisms and chemical equilibria and in industry to investigate corrosion and diffusion processes. Thus, radiotracer techniques are well established [BOWEN, DAUDEL].

In this report, a method using radiotracers to measure leaching from glass is described. Radionuclides of interest are identified, a method for making spiked glass is described, and the results of leach tests, which demonstrate the limits and sensitivity of the method, are presented. Finally, potential problems and applications are discussed.

II. EXPERIMENTAL PROCEDURES

A. Experimental Approach

A series of experiments has been completed to qualify the use of radiotracers in measuring the leach rates from glasses made with SRL frit 211 and SRL frit 131. These frits contain glass matrix elements and waste sludge components that result from the SRL reprocessing and waste storage process. These frits were chosen because they (1) represent the most current SRL reference glass compositions and, accordingly, will provide meaningful leach results, and (2) already have a good data base for leach rate comparison. Experiments were also done on simulated waste glass, which is a combination of a frit plus additives typical of SRL waste. These glasses are designated by the basic frit number and one or more stars. The compositions of all the frits used are listed in Table 2.

These experiments focused on four classes of chemical elements present as minor (<5 wt %) elements in SRL defense waste. These elements are of concern to waste management either because of their near-term toxicity or difficulties in their measurement by other techniques. The four classes are alkali metals (Cs), alkaline earths (Sr, Ba), rare earths (Eu, Ce) and noble metals (Ru). Simulated waste glass containing these radiotracers as spikes was prepared and the leaching of these elements monitored. Additionally, to demonstrate the validity of using radiotracers to measure leach rates, tests on both nonradioactive (cold) and neutron-activated glasses were done. Because microcompositional differences in the glass structure may exist from batch to batch, and since these differences may affect elemental leach rates, samples of spiked glass were also subjected to neutron activation so that the leaching behavior of the same element could be determined by an activation product and by a radiotracer spike for the same batch of glass (i.e., $^{139,141}\text{Ce}$, $^{152,154}\text{Eu}$, and $^{134,137}\text{Cs}$). Leach rates from these samples were measured using well-established measurement methods, ICP spectroscopy, and NAA [FLYNN, BATES-1]. These rates provided the base data to which leach rates from spiked glass could be compared.

Thus, leach rates were measured to allow comparisons among four different series of tests: (1) conventional chemical solution analyses (ICP, AA, DCP, and colorimetry) of leachates from nonradioactive (cold) glass; (2) γ -ray spectroscopy, AA, and colorimetric analyses of leachate solutions from neutron-activated glass; (3) γ -ray spectroscopy, AA, and colorimetric analyses of

Table 2. Compositions of Simulated SRL Waste Glasses Used in Leach Tests

Element	SRL Frit Composition, wt %				
	211 ^{a,b}	211** ^b	211*** ^c	131 ^c	131* ^c
Si	19.9	19.5	19.6	19.9	19.4
Na	11.0	10.8	11.1	9.9	9.7
Fe	8.5	8.0	9.9	9.7	9.5
B	2.5	2.4	2.4	3.3	3.2
Ca	3.6	3.4	3.6	0.9	0.9
Mn	2.7	2.2	2.4	2.3	2.2
Al	2.2	1.8	1.9	1.8	1.7
Li	1.2	1.3	1.4	1.9	1.8
Ni	1.4	1.3	1.3	1.3	1.3
Mg	-	-	-	0.9	0.9
Ti	-	-	-	0.4	0.4
Zr	-	-	-	0.3	0.3
La	-	-	-	0.3	0.3
Additives:					
Cs	-	0.1	0	-	0
Sr	-	0.05	0.3	-	0.3
Ba	-	0.1	0.3	-	0.3
Ce	-	0.7	0.7	-	0.7
Nd	-	0.8	0.8	-	0.8
Eu	-	0.01	0.1	-	0.1
Ru	-	0.05	0.4	-	0.4
K	-	-	0.05	-	0

^aReference [SRL-2].

^bComposition determined by ICP/AA analyses.

^cNominal composition.

leachate solutions from glass specimens containing radioactive tracers; and (4) γ -ray spectroscopy and colorimetric analyses of leachate solution from neutron-activated glass specimens containing radioactive tracers. In each case, analyses for both waste and matrix elements were obtained.

The objective of these measurements was to define the reproducibility, sensitivity, limitations, and precision of leach rate measurements for spiked glass.

B. Glass Preparation

A flow diagram illustrating the experimental approach used for SRL frit 211** glass is shown in Fig. 2.

Because of the importance of demonstrating the homogeneous distribution of radiotracers, extensive details of the glassmaking, sectioning, and counting procedures are given below for SRL 211** glass. Other glass preparations followed the same procedures, but the details are not presented.

A batch (300 g) of SRL frit 211 nonradioactive simulated waste glass, obtained as cast marbles from Pacific Northwest Laboratory (PNL), was melted for 4 h at 1050°C in a high-purity alumina crucible, together with additives of Cs, Ba, Sr, Ce, Nd, Eu, and Ru. The melt was then poured into water to make a frit. This 300-g batch of SRL 211** glass was ground and used as a source of base frit for the latter comparison tests. The compositions of the simulated waste glasses used in leach tests are given in Table 2. The compositions of SRL 211 and SRL 211** glasses were determined using ICP and AA spectroscopy; the values for the other glasses represent nominal compositions. The initial melt in the alumina crucible caused the actual aluminum concentration to be increased.

A portion (~50 g) of the initial 300-g batch of SRL 211** glass was melted in a platinum crucible for about 2 h at 1050°C and then poured into a Pt-5 wt % Au mold. The glass was then annealed in the mold for two hours at 500°C, producing a 10-g button (~40-mm dia by ~7-mm-thick disk). This non-radioactive molded glass button was sectioned with a low-speed Isomet saw (Model 11-1180) into pieces (~1/2 g of 15 mm by 15 mm by 1 mm) having a ~400 mm² surface area. Water was used as the cutting lubricant. Half of the cut specimens were prepared for leach tests, and the remainder were leach tested after neutron activation at the University of Illinois reactor. Leach tests of these two types of glasses provide a data base that allows direct comparisons with the leach results from spiked glass.

About 300 µCi of radioactive isotopes were added to another 50-g batch of SRL frit 211**, prepared from the initial 300-g batch. The radioactive isotopes, ¹³⁷Cs, ¹³³Ba, ¹³⁹Ce, ¹⁵²Eu, ⁸⁸Y and ¹⁰⁶Ru, were selected as tracers (~1 µCi per element per gram of glass) to be added to the SRL 211**. The radioactive properties of these isotopes (along with the other tracers used in the experiments are summarized in Table 3. Strontium-85 was not used in these initial experiments because of interfering γ-rays from ¹⁰⁶Rh. The tracers were purchased as HCl solutions, with ¹³⁹Ce and ⁸⁸Y being the only carrier-free isotopes. After dilution, all solutions of these vendor-supplied materials were stored in Pyrex volumetric flasks, with the exception of the ¹³⁷Cs, which was stored in a polypropylene vessel.

Aliquots (sizes given in Table 4) of these six stock solutions were then added to a rectangular platinum crucible, approximately 3.2 cm by 5.1 cm by 3.2 cm. The platinum crucible had been re-formed into a rectangular shape by gentle hammering of a 50-cm³ oval platinum dish crucible. This shape facilitated the cutting of glass specimens for leach testing with tolerable losses. Table 4 shows the proportions and the calculated activity levels loaded into the platinum crucible, based on vendor-supplied values. From the

Fig. 2. Flow Diagram of the General Approach Being Used for Comparing Leach Rate Results for SRL Frit 211 Waste Glass by Neutron Activation and Radioactive-Tracer Addition**

Table 3. Radioactive Isotopes Used and their Radiation Properties

Radioactive Tracer	Half-Life	Decay Mode	Primary γ -radiation, keV (photons/100 decays)
<u>Alkali Metal</u>			
^{137}Cs	30.2 y	β^-	661.7(85.0)
^{134}Cs	2.1 y	β^-	604.7(97.6), 795.8(85.4)
<u>Alkaline Earth</u>			
^{85}Sr	64.7 d	EC ^a	514.0(99.3)
^{133}Ba	10.7 y	EC	356.0(62), 81.0(31.7), 302.8(18.1), 383.8(8.0), 276.4(7.0)
^{131}Ba	11.7 d	EC	496.3(46.8), 216.1(19.7)
<u>Rare Earth</u>			
^{139}Ce	137.5 d	EC	165.9(79.9)
^{141}Ce	32.5 d	β^-	145.4(48.4)
^{152}Eu	14 y	73% EC (β^+), 27% β^-	1408.0(21.3), 121.8(29.1), 344.3(27.2), 964(14.8), 1112.1(13.8), 778.9(13.2)
^{88}Y	106.6 d	EC, β^+	898.0(91.3), 1836.1(99.3)
<u>Noble Metal</u>			
^{106}Ru (^{106}Rh 30 s)	368 d	β^-	511.8(19), 622.2(9.8), 1050.5(1.6), 616.3(0.82), 873.7(0.45), 1128.2(0.42)
^{103}Ru	39.4 d	β^-	497.1(89.5), 610.4(5.6)

^aElectron capture.

9300 λ in the platinum crucible, two small aliquots (10 λ and 20 λ) of each radioactive mixture were taken, evaporated to dryness, and analyzed by γ -ray counting. The average μCi in the crucible based on measured results from these two aliquots are shown in Table 4 in the "Before Melt" column.

Except for ^{137}Cs and ^{106}Ru , the measured μCi loaded into the crucible agrees with the μCi calculated from the vendor-supplied values. The 33% lower measured value for ^{137}Cs is assumed to be due to an error in the vendor-supplied value of μCi present in the purchased stock tracer solution. The 20% higher measured value for ^{106}Ru is believed to be primarily due to uncertainty in the absolute intensity of the two photons (i.e., 512 and 622 keV) associated

Table 4. "Mass" Balance of Radioactivity Added to SRL Frit 211**
Glass and Amount of Radioactivity Measured after Melting

Isotope	Aliquot, ^a λ	Carrier, μg	Calculated Activity Level, ^b μCi	Measured, Before Melt ^c		Measured, After Melt ^d		Loss, %
				μCi	(average)	μCi	(average)	
¹³⁷ Cs	300	4.5	60	40.1 ± 0.5 ^e		37.3 ± 0.4 ^f		
				38.9 ± 0.5 ^f	(40 ± 2)	37.8 ± 0.4 ^g	(38 ± 2)	-
						39.9 ± 0.4 ^h		
¹³³ Ba	2000	9.6	40	46.1 ± 0.5 ^e		43.1 ± 0.5 ^f		
				44.2 ± 0.5 ⁱ	(45 ± 2)	43.7 ± 0.5 ^g	(44 ± 3)	-
						46.5 ± 0.5 ^h		
¹³⁹ Ce	2000	1	40	41.9 ± 0.6 ^e		39.5 ± 0.6 ^f		
				41.4 ± 0.6 ⁱ	(42 ± 2)	41.9 ± 0.6 ^g	(42 ± 3)	-
						43.5 ± 0.6 ^h		
¹⁵² Eu	2000	40	48	50.3 ± 1.0 ^e		49.9 ± 0.5 ^f		
				48.2 ± 1.0 ⁱ	(49 ± 2)	50.0 ± 0.5 ^g	(50 ± 3)	-
						52.5 ± 0.5 ^h		
⁸⁸ Y	1000	<0.04	51	49.0 ± 0.2 ^e		47.4 ± 1.0 ^f		
				48.1 ± 0.2 ⁱ	(49 ± 2)	50.5 ± 1.0 ^g	(50 ± 3)	-
						51.5 ± 1.0 ^h		
¹⁰⁶ Ru	2000	40	50.4	67.9 ± 4.0 ^e		48.5 ± 2.0 ^f		
				63.0 ± 4.0 ⁱ	(66 ± 7) ^j	52.6 ± 2.0 ^g	(52 ± 5) ^j	<25
						53.6 ± 2.0 ^h		
				58.9 ± 3.8 ^e		43.3 ± 3.0 ^f		
				56.0 ± 3.9 ⁱ	(58 ± 6) ^k	47.7 ± 3.0 ^g	(45 ± 4) ^k	<25
						47.7 ± 3.0 ^h		
Total	9300	95	289					<25

^a1λ = 0.001 mL of solution.

^bBased on vendor-supplied values and initial dilutions used to prepare tracer stock solutions.

^cThe measured average values based on γ-ray analyses of two separate aliquots of 10λ and 20λ, each taken from platinum crucible before any glass frit additions or melting.

^dThe measured average values based on γ-ray analyses of three representative glass specimens, about 0.5 g, sectioned from the cooled melted glass. Based on analyses normalized to entire mass (50 g) of glass melt.

^eValue derived from 10λ aliquot.

^fValue from wafer (1,1).

^gValue from wafer (2,6).

^hValue from wafer (3,11).

ⁱValue derived from 20λ aliquot.

^jBased on absolute intensity value of 0.19 for 512-keV γ-ray.

^kBased on absolute intensity value of 0.095 for 622-keV γ-ray.

with the β^- decay; the disagreement with the vendor-supplied value is not regarded as significant. In addition, because of the long counting times required for these small aliquots of ^{106}Ru activity, there is more uncertainty about 512-keV transition (due to room background variations) than the 622-keV transition. Thus, two values, derived from the two different transitions, are given in Table 4 to allow better estimation of the uncertainty.

About 30 g of the 50-g batch of SRL frit 211** (previously sifted to -14 mesh or <1.2 mm) was mixed with the about 9.3 mL of the above-described tracer solution directly in the platinum crucible, using a glass stirring rod. The loaded crucible was then heated at about 140°C under a heat lamp and on a hot plate for several hours to evaporate the water, after which it was cooled overnight exposed to the atmosphere.

The remaining 20 g of SRL frit 211** was then added as a top layer to the platinum crucible, and the entire crucible was placed in a 500-mL Al_2O_3 secondary crucible with an Al_2O_3 cover. The mixture and the secondary crucible were placed in a furnace, and its temperature was held at 1050°C for 3 1/2 h in air so that the glass melted. The glass was then cooled to 500°C and held at that temperature for two hours, after which the furnace and crucible were allowed to cool overnight.

Weighing of the cooled ingot and platinum crucible indicated an uncertain and negligible weight gain (i.e., ~20 mg in comparison to the 67-g total weight). The platinum was then cut and stripped from the 50-g glass ingot. All outer edges of the glass ingot were removed by sectioning the ingot with an Isomet saw into six major pieces. These six pieces were further sectioned into 27 minor (1 cm by 1 cm by ~0.1 cm) pieces or wafers of about 0.5 g each. Figure 3 illustrates the sectioning and coding used to label the wafers cut from the ingot.

Of the 27 wafers cut, six were not usable, as indicated by the notes in Fig. 3. Ten wafers--(1,2), (1,3), (2,4), (2,7), (3,9), (4,14), (4,17), (5,21), (6,24), and (6,27)--from locations throughout the ingot were cleaned, weighed, measured, wrapped in aluminum foil, and submitted for neutron-activation analysis. Three wafers--(1,1), (2,6), and (3,11)--were submitted for (1) γ -ray analyses to establish the initial homogeneity of the tracers in the glass and (2) 7- and 14-day leach tests at 90°C in deionized water to establish whether sufficient tracer had been added for measuring leach rates. The remaining eight wafers--(2,8), (3,10), (4,15), (4,16), (5,22), (5,23), (6,25), and (6,26)--were prepared for γ -ray analyses to establish both the initial radioactivity prior to leaching and the degree of homogeneity of the radioactive tracers added to the glass. These eight wafers and the ten neutron-activated wafers were leached at 90°C in deionized water using the modified MCC-1 leaching procedure* for radioactivity [BATES-1].

The results of the gamma-ray analyses of wafers (1,1), (2,6), and (3,11) agreed to $\pm 5\%$ for all six isotopes (see Table 4). This agreement means that the tracers were homogeneously distributed throughout the glass. These results were then used to convert measured activity in the three 0.5-g pieces of glass to that present in the entire 50-g batch (the "Measured After Melt" column of

* Developed by the Materials Characterization Center at Battelle Pacific Northwest Laboratory.

Fig. 3. Schematic of 50-g Ingot (~ 5 -cm long by ~ 3.8 -cm wide by ~ 1.5 -cm thick) of Radioactive Glass, Illustrating the Coding of the Wafers Cut from the Original Ingot.

- Notes:
1. The positions of major pieces 1 and 3 could be interchanged because the precise location was not monitored; similarly, the major pieces 4 and 6 could be interchanged.
 2. For two cut wafers, (1,12) and (3,13), associated with pieces 1 or 2 or 3, precise locations could not be assigned. These two wafers are assigned arbitrary locations shown as (1,12) and (3,13).
 3. Wafer (2,5) had a small (~ 0.3 cm by 0.2 cm) triangular chip which was lost during cutting.
 4. Wafers (4,18), (5,19), and (5,20) were fractured during the cutting and cleaning procedures.

Table 4). Direct comparison of these results with the "Measured Before Melt" column allows estimation of the amount of radioactive tracers lost from the melt during glass fabrication. This comparison shows that, within the sensitivity of the measurements, only some ^{106}Ru was lost and that this loss was no greater than 25%. Counting of the Al_2O_3 secondary crucible indicated very small amounts of radioactivity present, namely, ^{137}Cs and ^{106}Ru , but in the same relative amounts as the sample. After removal of most of the glass from the platinum crucible with boiling acid solutions, the crucible contained a larger ($\sim 100\times$) amount of ^{106}Ru than ^{137}Cs . Thus, the ^{106}Ru was lost, at least in part, due to reaction with the platinum crucible during the melting.

As will be described in Section III.A, the amount of cesium in SRL 211** glass was too great to allow multielement detection using NAA. Additionally, several radiotracers were difficult to detect in the spiked leachates. A new glass composition, SRL 211***, was used to overcome these difficulties. SRL frit 211*** contained no added cesium and increased amounts of strontium ($\times 6$), barium ($\times 3$), europium ($\times 10$), and ruthenium ($\times 8$) compared to the previous 211 glass. The frit was made using the procedure described for SRL 211** glass, and the nominal composition of this frit is shown in Table 2.

A 41.6-g batch of the SRL 211*** glass, powdered and pulverized, was placed in a platinum crucible to which 7010 λ of four radioactive tracers had been added. The tracers totaled $\sim 2621 \mu\text{Ci}$ and consisted of ^{141}Ce (820 μCi in 1000 λ), ^{152}Eu (1000 μCi in 5000 λ), ^{106}Ru (800 μCi in 1000 λ), and ^{137}Cs (1.3 μCi in 10 λ). The concentration of activity per gram of glass was increased by a factor of ~ 40 compared to the SRL frit 211** glass. The amount of increased activity was selected based on previous leach rate measurements of neutron-activated SRL frit 211** glass containing detectable amounts of ^{141}Ce , ^{152}Eu , and ^{106}Ru .

A 5- λ aliquot of the above tracer solution was prepared for γ -ray analysis to verify the levels of tracers added to the glass. A glass stirring rod was used to mix the powdered SRL frit 211*** glass with the tracer solutions; this mixture was then left to dry overnight. The glass was melted at 1050°C for four hours, poured into a Pt-5 wt % Au mold, and the ingot was annealed at $\sim 500^\circ\text{C}$ for ~ 2 hours and allowed to cool overnight. The resulting ~ 30 g ingot of SRL 211*** was then sectioned into 21 mm by 7 mm by 1 mm pieces for leaching studies, detailed below.

A batch of modified SRL frit 131* glass was also prepared. The nominal composition of this frit is shown in Table 2, where it can be compared to SRL 131 and to the SRL 211 glasses used in these studies. SRL frit 131* glass contains no cesium additives and is similar in additives to the SRL frit 211***; the SRL 131 glass contains Mg, Ti, Zr, and La, which are not in the basic SRL frit 211 glass.

A 40.1-g batch of the SRL 131* glass, powdered and pulverized, was placed in a platinum crucible to which 12,005 λ of five radioactive tracers had been added. The tracers totaled $\sim 3786 \mu\text{Ci}$ and consisted of ^{141}Ce (1640 μCi in 2000 λ), ^{152}Eu (2000 μCi in 4000 λ), ^{85}Sr (100 μCi in 4000 λ), ^{133}Ba (45 μCi in 2000 λ), and ^{137}Cs (0.65 μCi in 5 λ). The amount of activity per gram of glass was increased by a factor of ~ 40 for these experiments compared to the first experiments in this series with SRL frit 211**. The amount of increased

activity was selected based on previous leach rate measurements of neutron-activated SRL frit 211** glass containing detectable amounts of ^{141}Ce , ^{152}Eu , ^{133}Ba , and ^{137}Cs . No leach rate data had yet been obtained for ^{85}Sr .

A 5- λ aliquot of the tracer solution was prepared for γ -ray analysis to verify the composition of the tracers and the levels of tracers added to the glass. After a measurement of the γ spectrum, the amount of ^{85}Sr in the glass was doubled to the 100 μCi total reported above.

The powdered SRL frit 131* glass was then mixed with the tracer solutions by stirring with a glass rod and left to dry overnight. The glass was melted at 1050°C for four hours, poured into a Pt-5 wt % Au mold, annealed at 500°C for 2 hours and allowed to cool overnight. The resulting 30 gram ingot was then sectioned into 21 mm by 7 mm by 1 mm pieces for leaching studies.

C. Leach Test Procedure

The leach tests were done following a modified version of the MCC-1 static leach test [BATES-1, MCC]. The exact MCC-1 procedure was not followed because the purpose of the tests was not to provide quantitative leach results for different waste forms under all MCC-1 conditions, but to demonstrate the use of radiotracers as an analytical technique. Therefore, it was not necessary to follow the complete MCC-1 test procedure.

The procedural details are given in Section 1.0 of the Appendix. Briefly, the glass specimen was immersed in static deionized water for specified periods of time through 42 days, each time period representing a separate leach test. Tests were done at 90°C and replicate tests were done at either 28 or 42 days. Blanks were run for each series of tests.

Leachate analyses were done using γ -ray counting and both AA and DCP spectroscopy. Techniques established previously [BATES-1] were used in leachate preparation and counting. Generally, the radioactive leachates were split, with γ -ray counting being done for radioactive elements and colorimetric analysis being done for silicon. Nonradioactive leachates were analyzed using DCP (by Trace Elements, Inc.) and AA (by Analytical Chemistry Laboratory/Chemical Engineering Division) and provide a complete description of matrix-element leaching behavior; DCP analyses of radioactive samples could not be done.

γ -ray counting was done by the Analytical Chemistry Laboratory/Chemical Engineering Division (ACL/CEN), the University of Missouri (U of M), and the Fuel Cycle Section/Chemical Engineering Division (FCS/CEN). Selected samples counted by ACL/CEN and U of M were recounted by FCS/CEN to assure that systems errors between sites were less than 10%.

III. RESULTS AND DISCUSSION

A. SRL 211** Glass

Four series of leach tests were done on SRL 211** glass--cold (C), activated (A), spiked (S), and spiked/activated (S/A). The detailed experimental conditions and results are presented in the Appendix. In Table 5, the test results have been consolidated for comparison purposes.

Table 5. Consolidated Leaching Results for SRL 211** Glass^a

211** Glass Type	Test Dura- tion, d	Δ Mass, SA g/m ²	Normalized Elemental Loss, g/m ²																
			Determined by γ -ray Analysis								Determined by AA or DCP								
			51Cr	124Sb	133Ba	134Cs	137Cs	139Cs	141Ce	152Eu	Si	Cs	Na	Li	B	Mn	Ca	Al	Fe
C	3	7.6									10.3	11.4	11.1	11.2	10.7	0.2	7.0	9.9	0.3
	5	7.4									9.5	11.5	12.8	13.1	11.9	<0.2	7.5	9.6	0.06
	7	7.9									10.2	13.7	15.3	15.7	14.2	<0.2	7.3	10.3	0.10
	14	9.3									10.8	16.1	17.1	17.6	16.0	0.2	7.5	10.2	0.2
	28	9.6									11.4	17.6	19.1	21.0	17.9	0.3	7.6	9.5	<0.06
	42	11.4									13.0	19.0	21.5	22.3	19.0	0.2	7.4	10.2	0.1
A	3	8.1																	
	7	10.5																	
	14	12.0																	
	28	14.5				20.1													
	42	15.8																	
S	3	6.7			0.9		14.3												
	7	9.7			0.5		18.1												
	14	11.1			0.5		21.7		0.06										
	28	ND ^b			0.3		22.1												
	42	10.8			0.3		24.2	0.02		0.10	15.4								
S/A	3	5.9		7.8	0.7	11.4	13.2		0.01	0.06									
	7	8.9	4.4	9.7	0.4	16.0	18.7												
	14	10.9	3.9	13.7	0.5	18.9	21.9		0.01										
	28	ND ^b	3.5	14.1	0.4	21.0	24.2			0.07									
	42	12.0	7.0	16.2	0.3	21.1	24.5			0.02	14.9								

^aComplete analytical results are given in the Appendix.^bNot determined.

The base leaching behavior of 211** glass was established through tests on A and C glass. The tests on C glass established the leaching behavior of the matrix elements and cesium, while the tests on A glass were to establish the leaching behavior of the waste elements.

The leaching of the matrix elements for 211** C glass is shown in Fig. 4; three different types of leaching are observed. Several elements, e.g., Cs, and Na (Li and B are not shown), leach at the highest rate and are still being released through 42 days. Silicon, calcium, and aluminum have initial rates comparable to the first group, but beyond seven days of leaching show little additional release. Finally, iron and manganese have leach rates that are 30-70 times lower than those of the other elements. The scatter in the normalized elemental mass loss $[(NL)_i]$ values for these elements may be because their detection is near the analytical limit.

Fig. 4. Multielement Leach Results from SRL 211** Cold Glass

When the leachates for A glass were analyzed, only ^{134}Cs could be readily detected. The cause of this problem was discovered when the amount of cesium added to the SRL frit 211** was determined. A delicate balance must be achieved between the amount of cesium in the glass and the amount of cesium that leaches from the glass [BATES-1]. The amount was chosen based on leach results [FLYNN] done at 25 and 70°C on a different glass composition. Determining the amount of cesium to add is further complicated because (1) cesium release varies more than that of other elements as a function of temperature and (2) some cesium may be lost during glass formation. Unfortunately, the composition used in

the SRL 211** glass resulted in too much cesium; consequently, other activated elements could not be directly detected by γ -ray analysis. Since cesium leach rates had already been determined for SRL 211** C glass, they were not repeated for any other than the 28-day leachate of SRL 211** A glass.

In the S/A glass, leach rates for ^{134}Cs , ^{137}Cs and ^{133}Ba could be determined for all samples (Fig. 5). The ^{141}Ce and ^{152}Eu could be detected in some samples; however, because concentrations were near the detection limits, a complete description of the leaching process for these elements was not obtained. Leach rates for ^{51}Cr and ^{124}Sb were also obtained due to the activation process; however, the leach rates of ^{139}Ce and ^{154}Eu could not be obtained.

Fig. 5. Multielement Leach Results from SRL 211** Spiked/Activated Glass

In the S glass, only ^{133}Ba and ^{137}Cs could be consistently detected (Fig. 6). In some samples, ^{139}Ce and ^{152}Eu could be detected, but again they were near the detection limit. A silicon analysis was done at 42 days for both the S/A and S samples.

Although a complete description of the leaching process cannot be obtained from these studies on SRL 211** glass, several useful observations can be made:

- (1) The overall leaching behavior of all the glass samples is the same, as measured by weight loss. For the four glasses, the normalized

Fig. 6. Multielement Leach Results from SRL 211** Spiked Glass

weight loss data at 42 days give an average of 12.5 g/m^2 , with a standard deviation of 2.3. This consistency in the leaching of the major glass components is confirmed by the silicon analysis. The silicon leach rates have been determined at 42 days for each glass, except the activated one. Triplicate samples were analyzed and, within experimental error, no difference in $(\text{NL})_{\text{Si}}$ was observed. The average $(\text{NL})_{\text{Si}}$ is $14.4 \pm 1.3 \text{ g/m}^2$. These data indicate that the matrix for each glass type is leaching at the same rate.

- (2) Cesium leach rates were determined for ^{134}Cs , ^{137}Cs , and elemental cesium. The leach rates for ^{137}Cs , as measured in S/A and S glass, are the same. This is also the case for ^{134}Cs from S/A and A glass. Comparison of cesium leaching from all four glasses is shown in Fig. 7. While the cesium leaching as measured by all methods is within $\pm 12\%$, the ^{137}Cs appears to exhibit consistently higher leach rates than ^{134}Cs , even from the same glass.
- (3) Leach rates for ^{133}Ba as measured in S/A and S glass are similar. However, neither NAA (^{131}Ba) nor DCP spectroscopy was sensitive enough to measure a base rate for comparison with the radiotracer.

In conclusion, the leach studies of SRL 211** glass demonstrate that cesium and barium leach rates can be measured successfully using radiotracer spiking, but they also demonstrate that care must be taken when loading the glass. For SRL 211** glass, not one of the other spikes was observed in the

Fig. 7. Comparison of Cesium Leach Rates from SRL 211** Glass

leachate due to (1) the low leach rates of these elements and (2) the high background produced by the large amount of cesium in the leachate. The behavior of the other radiotracers had to be investigated using another glass to which less cesium was added.

B. SRL 211*** Glass

Since SRL 211** glass contained too much cesium to detect most other γ -emitters, glass SRL 211*** was made. This glass contains nominally no added cesium. Tests were done on cold, activated, and spiked (^{137}Cs , ^{141}Ce , ^{152}Eu , and ^{106}Ru) samples of this glass. Condensed leaching results are given in Table 6; complete results are in the Appendix.

As was done for SRL 211** glass, the base leaching behavior was determined using C and A glass. The leaching of the matrix elements from SRL 211*** C glass is shown in Fig. 8, and within experimental error the results are identical to those of SRL 211** glass. Weight loss measurements are also identical.

For A glass, ^{51}Cr , ^{59}Fe , ^{85}Sr , ^{103}Ru , ^{124}Sb , ^{131}Ba , ^{134}Cs , ^{141}Ce , ^{147}Nd , ^{152}Eu , and Si were analyzed in the leachate (Fig. 9). However, no leach rates for ^{134}Cs were determined since (1) no cesium was added to the solid and (2) no γ -ray peaks attributable to ^{134}Cs could be observed above the background. Leach rates for ^{59}Fe , ^{85}Sr , ^{124}Sb , ^{141}Ce , and ^{152}Eu were determined for each

Table 6. Consolidated Leaching Results for SRL 211*** Glass

Normalized Elemental Loss, g. m ²																						
211*** Glass Type	Test Dura- tion, d	Δ Mass, SA g/m ²	Determined by γ -ray Analysis											Determined by AA or DCP								
			⁵¹ Cr	⁵⁹ Fe	⁸⁵ Sr	¹⁰³ Ru	¹⁰⁶ Ru	¹²⁴ Sb	¹³¹ Ba	¹³⁷ Cs	¹³⁹ Ce	¹⁴¹ Ce	¹⁴⁷ Nd	¹⁵² Eu	Si	Na	Li	B	Mn	Ca	Al	Fe
C	3	8.0													8.5	11.4	12.8	11.4	0.3	7.6	9.1	0.2
	7	8.5													10.2	15.0	16.1	14.1	0.3	7.1	10.1	0.2
	14	10.1													11.2	16.4	17.6	16.0	0.3	7.8	9.6	0.2
	28	11.3													12.6	18.9	21.4	18.9	0.3	7.8	10.6	0.2
	42	12.1													14.6	20.8	23.6	20.8	0.3	8.2	8.8	0.2
A	3		1.3	0.12	1.9			7.9	0.9		0.04	0.07	0.04									
	7	8.4	1.6	0.09	1.8			12.4	0.5		0.03	0.05	0.03									
	14	13.3	2.9	0.06	1.8			12.0			0.03		0.03									
	28	11.9	6.6	0.14	1.3	0.03		18.8			0.08		0.06	13.6								
	42			0.18	0.9			17.0			0.08		0.08									
S	3	5.9					0.008			13.5		0.002		0.004								
	7	8.8					0.006			17.7		0.003		0.004								
	14	9.5					0.002			16.4		ND		0.004								
	28	9.9					0.02			20.1	0.15	0.11 ^a		0.008	13.3							
	42	9.8					0.02			22.1		0.02		0.01								

^aAnomalous value.

Fig. 8. Multielement Leach Results from SRL 211*** Cold Glass

Fig. 9. Multielement Leach Results from SRL 211*** Activated Glass

sampling period. However, for the other elements, leach rates were calculated from a limited number of sampling periods due to short half-lives or very low leach rates. These rate determinations provide the necessary base to which leach rates from S glass can be compared.

For S glass, each of the radiotracers could be detected in the leachates (Fig. 10). The rare earths and ruthenium have similar (NL)_i values, increasing with time, while cesium has a (NL)_i value at least 1000 times greater. Because the amounts of each radiotracer in SRL-211*** had been decided upon based on our experience with SRL 211** glass, γ-ray interferences were minimized and more effective data collection and assimilation was achieved for these S- or A-glass experiments.

Fig. 10. Multielement Leach Results from SRL 211*** Spiked Glass

Comparison of the data indicates the following:

- (1) Leach rates for ¹³⁷Cs are identical in both SRL 211** and SRL 211*** glass. No comparison with the release of ¹³⁴Cs or elemental cesium from SRL 211*** glass was possible.
- (2) When compared at 28 days, ¹⁰³Ru (A) and ¹⁰⁶Ru (S) leach rates are similar; however, ¹⁰³Ru leach rate determination was tenuous because it was near the detection limits.
- (3) Leach rates for ¹⁴¹Ce and ¹⁵²Eu mirror each other, but are about one order of magnitude greater in A glass than in S glass.

C. SRL 131* Glass

Using the information and experience gained in studying the SRL 211 series of glasses, radiotracer leaching tests were done with SRL 131* glass. The SRL 131 glass is the current reference frit, and it was modified for these experiments through the addition of ~3 wt % waste elements (Table 2). Tests were done on cold, activated, and spiked (^{85}Sr , ^{137}Cs , ^{133}Ba , ^{141}Ce , ^{152}Eu) glass, and results are given in Table 7 and the Appendix. The noble metal group (^{106}Ru) was not studied because there was interference between the γ -rays from ^{85}Sr and ^{106}Ru .

The base leaching behavior for SRL 131* glass was established using C and A glass. Elemental leaching from C glass is shown in Fig. 11. The release of Na, Si, and B is the most rapid, and the (NL)_i values appear to be still increasing after 37 days. The Si release and weight loss are less than the alkali metals, but are still increasing slowly after 37 days. The Al release is constant beyond 7 days and, at 7 days, calcium leaching has stopped, with the calcium concentration actually less after 37 days than initially. In the

Fig. 11. Multielement Leach Results from SRL 131* Cold Glass

final tests, no Ca could be detected in solution. Iron and manganese releases are low, and little additional release of these elements occurs after the third day of leaching. Barium and titanium could not be detected in solution using DCP spectroscopy. Except for the marked decrease in calcium with time, the leaching trends observed for the matrix or matrix-modifying elements are similar for SRL 131* and SRL 211-type glasses.

Table 7. Consolidated Leaching Results for SRL 131* Glass

131* Glass Type	Test Dura- tion, d	A Mass, SA g/m ²	Normalized Elemental Loss, g/m ²																				
			Determined by γ-ray Analysis										Determined by AA or DCP										
			51Cr	59Fe	85Sr	124Sb	131Ba	133Ba	134Cs	137Cs	141Ce	152Eu	Si	Na	Li	B	Mn	Ca	Al	Fe	Ba	Ti	Mg
C	3	7.0											8.5	ND	ND	11.8	<0.2	3.8	8.4	0.1	<1.7	<1.2	
	7	11.8											14.2	ND	ND	19.2	0.3	1.4	12.9	0.08	<1.7	<1.2	
	14	12.8											16.0	ND	ND	23.3	0.3	0.7	12.6	0.07	<1.7	<1.2	
	28	16.6											19.5	28.9	29.5	30.5	0.3	<0.5	12.5	<0.06	<1.7	<1.2	
	37	18.0											22.3	31.0	32.0	30.3	0.4	<0.5	13.0	<0.06	<1.7	<1.2	0.7
A ^a	2.25	5.6	1.5	0.13	1.6	ND	2.5				0.04	0.06											
	7	ND ^b	1.1	ND	0.4	7.1	ND		8.2		0.008	0.10											
	14	6.2	3.2	ND	0.5	7.2	0.3		7.9		0.007	0.005											
	28	10.3	3.4	0.08			ND		22.7		0.01	0.01	17.7										
S	3	4.3			4.0			0.8		9.0	0.004	0.005											
	7	8.1			1.7			0.3		15.5	0.02	0.005											
	14	11.5			1.6			0.4		21.6	ND	0.003											
	28	13.1			1.3			0.3		20.0	0.04	0.01											
	42	11.9			1.1			0.2		22.5	0.04	ND	17.3										

^a Large errors due to counting statistics and waste-form alteration during irradiation.^b ND = Not determined.

The base leaching rates for the waste elements for SRL 131* A glass were established with NAA and are shown in Fig. 12. The leach rates of ^{51}Cr , ^{59}Fe , ^{134}Cs , ^{141}Ce , and ^{152}Eu could be determined through 28 days, while the leach rates of ^{85}Sr and ^{131}Ba could be determined for shorter periods of time. Thus, the leaching of five waste elements-- ^{85}Sr , ^{131}Ba , ^{134}Cs , ^{141}Ce , and ^{152}Eu --was determined and provides data to which the leaching of radiotracers can be compared. Unfortunately, some difficulty in applying the NAA technique occurred (see Appendix); therefore, the test matrix was not completed, and the results are not as consistent as those normally achieved. Cesium-134 has the highest leach rate, and it is still being released into solution after 28 days. The leach rates of ^{85}Sr and ^{131}Ba are about 10 times less than ^{134}Cs , and the amount of these nuclides in solution decreases with time. The leach rates of the rare earths are about 100 times lower still and, after a possibly spurious rapid initial release, show a slow increase after 28 days.

Fig. 12. Multi-element Leach Results from SRL 131* Activated Glass

Each of the radionuclides could be detected in the leachates from the S glass; the leaching trends are shown in Fig. 13. As was observed in the SRL 211 glasses, the rare earths have low but increasing (NL)_i values, while ^{137}Cs has a high and increasing (NL)_i value. Concentrations of ^{85}Sr and ^{133}Ba in the leachate decrease with time. The ^{137}Cs , ^{85}Sr and ^{133}Ba (NL)_i values display a smoothly increasing or decreasing trend; however, the behavior of the rare earths is more erratic, showing leach rate variations between leach periods of up to a factor of 10. A possible cause for these variations is discussed in the Appendix.

Fig. 13. Multielement Leach Results from SRL 131* Spiked Glass

The leach tests on SRL 131* glass provide the most complete evaluation of the radiotracer spiking method of measuring leach rates. For each radio-tracer added to the glass, an independent leach-rate determination for that element was made using a different method of analysis. Additionally, the leaching behavior of the glass matrix elements was monitored for each glass type, using weight loss measurements and silicon analyses. The following observations are made:

- (1) The leaching of the glass matrix as measured by the silicon leach rate is, within experimental error, identical for each glass type. The average silicon elemental loss for all glass types after 28 days is $18.2 \pm 0.9 \text{ g}/\text{m}^2$.
- (2) The cesium leach rates as measured by the radiotracer ^{137}Cs and the activation product ^{134}Cs are equal within experimental error after 28 days.
- (3) The leach rates of the alkaline earth elements, Sr and Ba, as measured by the radiotracers ^{85}Sr and ^{133}Ba and by the activation products ^{85}Sr and ^{131}Ba vary over the testing period by a factor of three for Sr and by far less for Ba, but the trends are the same. Because of the error in the NAA measurements [BATES-1], this variation may be within experimental error.
- (4) The leach rates of the rare earths, ^{141}Ce and ^{152}Eu , are very similar and, despite the significant variations in the data for the initial test periods, the averaged results for the 28-day leach period indicate that the $(\text{NL})_i$ values are identical for spiked and for activated glass.

D. Utility and Sensitivity of the Test Methods

Radiotracers can be used to measure the leach rates of elements from at least the four chemical groups, i.e., alkali metals, alkaline earths, rare earths, and noble metals. Since most individual waste elements of interest have commercially available radionuclides, the leaching of any number of elements, theoretically, can be studied simultaneously. However, the effective use of the method is tempered by several factors.

(1) It must be possible to incorporate the radiotracer in the waste form. For glass, careful safety procedures must be followed, but the process is not difficult. For ceramic waste forms such as SYNROC, greater difficulty may be encountered because of the complexity of the manufacturing process.

(2) The half-life of a radionuclide must be long enough so that it can be detected throughout the entire period (leaching plus analysis) of the experiment. If the half-life is too short, the large amount of activity that must initially be added to the sample may cause interferences in detecting other γ -rays (elements) of interest. Additionally, problems due to high radiation levels may influence handling procedures which require significant contact during sample preparation. As a general rule, the half-life of a radionuclide should not be shorter than the total duration of the testing period.

(3) Radionuclides must be chosen so that γ -ray energy interferences are minimized. For example, in the present experiments leach tests could not be done simultaneously with ^{85}Sr and ^{106}Ru because of interference between the 514 keV γ -ray of ^{85}Sr and the 512 keV γ -ray of ^{106}Ru . Both γ -rays are the primary analytical energy peaks for each element. Because of these interferences, it was useful to focus on chemical groups instead of individual elements, since alternative radiotracers may be available, e.g., ^{133}Ba for ^{85}Sr .

(4) The amount of each radionuclide initially added to the waste form must be balanced so that interference-free detection is possible in both the solid and the leachate. This balance is a problem because of the wide range ($\sim 10^5$) of leach rates for waste elements of interest. For example, the amount of ^{137}Cs added to the waste form must be carefully controlled because cesium has a high leach rate and can easily become the only detectable nuclide in the leachate.* However, obtaining a satisfactorily reduced amount of cesium in the leachate may require adding so small an amount of ^{137}Cs that it is difficult to detect in the solid initially. For example, solids that require only 30 min of counting to determine most radiotracers would require 50 times longer to determine the amount of ^{137}Cs . It is also likely that the ratio of radionuclides added to the solid must be adjusted according to the conditions of the leach test. Cesium leach rates increase more rapidly with temperature than many other elements; consequently, a radionuclide ratio that was satisfactory at 90°C may not provide complete results at 40°C. Some estimate of

* In most instances, chemical separation followed by γ -ray analysis could resolve the interferences; the added complication of the separation was avoided to retain the ease and speed of γ -ray analysis coupled to computer-aided spectrum resolution.

the leaching behavior of the waste form is necessary when making the initial mix of radiotracers.

The detailed description of the glass-making process (Section II.B) provides a starting point for determining the amounts of radionuclides needed to provide adequate elemental detection in both solid and leachate while maintaining convenient handling (<100 mR/h).

The sensitivity of γ -ray spectroscopic techniques as applied to leach rate measurements depends on several factors which have been discussed in detail elsewhere [BATES-1, FLYNN]. In this context, sensitivity is the level at which the leach rate of an element can be measured. Generally, it is dependent on counting factors and on the nature of the spectrum being analyzed. The sensitivity for one radionuclide could be greatly increased by increasing the initial amount of that nuclide in the solid. However, this increase would likely decrease the sensitivity for other elements.

For the present studies, a practical sensitivity limit [based on a reasonable count time of 3000 min and an ability to obtain 500 counts above background for the γ -ray of interest (5% counting error)] is given in Table 8 for each radionuclide in terms of minimum measurable leach rate, $(NL)_i$. As is evident, the radiotracer mix has been optimized to give the greatest sensitivity for the rare earths and noble metals and the least sensitivity for the alkali metals and alkaline earths. This is in line with the expected leaching behavior of the elements. However, the sensitivity for any one element can be increased, possibly at the expense of other elements, simply by increasing the amount of initial activity.

Table 8. Sensitivity Limits Obtained Using Radiotracers to Measure Leach Rates from SRL Glasses

	^{85}Sr	^{106}Ru	^{133}Ba	^{137}Cs	^{141}Ce	^{152}Eu
Sensitivity ^a $(NL)_i$, g/m ²	5×10^{-2}	5×10^{-3}	1×10^{-2}	5×10^{-1}	1×10^{-3}	1×10^{-3}

^aThe values, $(NL)_i$, are based on 3000 min count times and a 5% counting error.

IV. CONCLUSIONS

Radiotracers belonging to four chemical groups have been used to measure the leaching of typical waste elements from defense waste glass. These results have been compared to leach rates of the same elements measured by other established techniques to determine if any differences exist between the methods. The conclusions for each group are as follows.

Alkali metals. Cesium leach rates, as measured by the radiotracer ^{137}Cs , were similar to those measured by NAA (^{134}Cs) and AA (elemental cesium). This is best indicated by the results for SRL 211** glass, which was the only glass to contain added nonradioactive cesium. Leach rates for ^{137}Cs and ^{134}Cs from

the same glass batch, SRL 211** S/A, varied by about 15%, while the leach rate for cesium (AA) measured for SRL 211** C, the same glass but a different batch, also varied ~15% from the average value for all cesium determinations.

Alkaline Earths. The same leaching trends are observed for both barium and strontium. The normalized elemental loss is initially at a moderate level (1 to 10 g/cm²), but the amount of each element in solution decreases with time. Barium leach rates as measured by ¹³³Ba were compared to the activation product, ¹³¹Ba, and the results are equal within experimental error. Barium leach rates as determined by DCP were near the detection limit and did not provide a useful comparison. A more-sensitive analytical technique, such as ICP spectroscopy, would have yielded a better comparison. ¹³³Ba is a more-effective method for measuring barium leach rates than ¹³¹Ba because of its longer half-life.

⁸⁵Sr added as a radiotracer was compared to ⁸⁵Sr produced by neutron activation. The larger difference between the strontium radiotracer and NAA results, as compared to barium, is attributed to a greater error in the NAA value, which is due to the small amount of strontium in the activated solid. The small amount of ⁸⁵Sr, coupled with its 65-day half-life, produced difficulties in measuring ⁸⁵Sr in both the activated solid and its leachate. Using ⁸⁵Sr as a radiotracer provided better control over the amount of activity, and resulted in more reliable leach rate measurement.

Rare Earths. The consistency of rare earth leach rates was less than expected, mainly due to procedural problems (Appendix Section 5.1). Nevertheless, results obtained using NAA and radiotracers agreed, at worst, within a factor of ten, and usually much better. In this study the leaching behavior of the rare earths, as represented by ^{139,141}Ce, ¹⁴⁷Nd, and ¹⁵²Eu, was similar among the elements, and it is suggested that ¹⁵²Eu be used as the rare earth radiotracer. With proper handling and glass-making procedures, radiotracers offer a sensitive means of measuring the leach rates of this group of elements.

Noble Metals. The leach rates were measured for ¹⁰⁶Ru and ¹⁰³Ru in SRL 211*** S and A glasses, respectively. The leach rates were difficult to measure due to the slow release of both isotopes, the short half-life of ¹⁰³Ru, and interferences with the primary γ -rays of both isotopes. However, similar leach rates were observed in the spiked and in the activated samples. For the radiotracers to be more effective, a greater amount of the ruthenium spike should be used. Radiotracer measurement of ruthenium is better than measurement by NAA because of the greater control over the amount of activity present in the sample.

The utility of radiotracers in measuring the leach rate from nuclear waste forms is limited by the nature of the solid, the conditions of the leach test, and the choice of radionuclides. However, radiotracer analysis is easier to use and more sensitive than NAA because there is more control over the activity in the solid and, hence, in the leachate. Gamma-ray detection can be refined to give fewer interferences and lower detection limits. Radiotracer analysis is of greater use than conventional spectroscopic methods in measuring leach rates of the rare earths and noble metals. Overall, the use of radiotracers is a reliable and sensitive method for simultaneously measuring the leaching of many elements from simulated nuclear waste forms.

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APPENDIX

In this Appendix, detailed experimental procedures and results are presented following, in part, the format suggested by the Materials Characterization Center (MCC). In Section 5.0, a discussion of deviations from the procedure and of inadvertent happenings that affected the significance of the results is presented.

1.0 TEST CONDITIONS, SPECIMEN PREPARATION, AND DESCRIPTION

1.1 Materials Composition

Three glasses were used in the experiments described in this report, SRL 211**, 211***, and 131*. The glasses were made using a starting frit supplied by PNL for the SRL 211 glasses and by SRL for the SRL 131* glass. To each of these frits were added various amounts of simulated waste. The procedure used in making the glasses and the composition of the glasses are given in Section II of this report. The actual composition of the SRL 211 glasses was determined using ICP analysis done at Ames Laboratory, but no analyses were done for SRL 131* glass, due to prohibitive cost. For this glass (SRL 131*), leach rate calculations are based, where necessary, on nominal composition. For NAA or spike leach rate determinations, the relative elemental concentrations are determined for each specimen by γ -counting.

The homogeneity of the glass samples was determined by calculating the specific activity for each nuclide for every piece of glass. The results for each set of spiked glasses are shown in Table A1-1. Generally, the standard deviation found for each average specific activity is $\leq 5\%$ of the average, which is the same as the counting error. Thus, there is no evidence of gross inhomogeneities between specimens of the same glass. However, the polished glass samples were also studied using scanning electron microscopy (SEM) and energy dispersive analysis of X-rays (EDAX) and inclusions in the glass were observed. These were identified as Pd and Ru metal. The amount of Pd or Ru was not determined quantitatively but was estimated to be $<2\%$ volume. The distribution of crystals was random throughout the samples. Before sectioning, the as-cast glass ingot generally had a darker, less transparent appearance on the edges of the sample. The edge regions were removed during the sectioning process.

1.2 Test Equipment

- Environmental chamber--Blue M Mechanical Connection Oven, Model OV-490 A-2, Blue M. Co., Blue Island, IL.
- The temperature uniformity was verified using precalibrated thermocouples placed near the corners, the wall centers, and the middle of the chamber. During a test period of two days, none of the recorded temperatures deviated from 90°C by more than 0.5°C . The temperature in the oven was set using a thermometer from the Brooklyn Thermometer Company that had been calibrated to NBS standards.
- Leach containers--For nonradioactive samples, teflon PFA containers with a 60-mL capacity were used. These were obtained from Savillex, #0102-53 MOD and #0102, Savillex Corp., Minnetonka, MN. Specimens

Table A1-1. Specific Activities for Radionuclides Used in Spike/Leach Tests

Test Number	L-257	L-258	L-259	L-260	L-261	L-262	L-263	L-264	L-265	Average of Specific Activities	
Sample mass, g	0.4240	0.4511	0.4253	0.4108	0.4452	0.5477	0.4552	0.4389	0.4436		
SRL 211** S Activity, normalized $\mu\text{Ci/g}^a$										Average	S.D.
88Y	3.3 E-1 ^b	3.5 E-1	3.53 E-1	3.63 E-1	3.23 E-1	3.73 E-1	3.27 E-1	3.01 E-1	2.61 E-1	3.31 E-1	3 E-2
106Ru	4.36 E-1	5.17 E-1	5.08 E-1	5.21 E-1	4.31 E-1	5.04 E-1	4.94 E-1	4.63 E-1	3.97 E-1	4.75 E-1	4 E-2
133Ba	8.54 E-1	8.45 E-1	9.19 E-1	8.69 E-1	8.56 E-1	8.99 E-1	8.77 E-1	8.43 E-1	7.62 E-1	8.58 E-1	4 E-2
137Cs	6.60 E-1	6.61 E-1	6.87 E-1	6.69 E-1	6.60 E-1	6.93 E-1	6.85 E-1	8.82 E-1	8.30 E-1	7.14 E-1	8 E-2
139Ce	3.21 E-1	3.46 E-1	3.55 E-1	3.46 E-1	3.19 E-1	3.58 E-1	3.16 E-1	3.05 E-1	2.57 E-1	3.25 E-1	3 E-2
152Eu	1.04	1.06	1.14	1.05	9.32 E-1	9.72 E-1	9.14 E-1	8.36 E-1	6.85 E-1	9.59 E-1	1 E-1
Test Number	L-222	L-223	L-224	L-225	L-226	L-227	L-228	L-229	L-230		
Sample mass, g	0.4360	0.4471	0.4535	0.4491	0.4486	0.4491	0.4496	0.4460	0.4525		
SRL 211** S/A Activity, normalized $\mu\text{Ci/g}$											
133Ba	8.17 E-1	8.03 E-1	8.29 E-1	8.19 E-1	8.52 E-1	8.82 E-1	8.10 E-1	8.18 E-1	7.65 E-1	8.22 E-1	3 E-2
134Cs	5.21	5.37	5.29	5.19	5.35	4.92	4.92	5.13	5.22	5.18	1.6 E-1
137Cs	6.24 E-1	6.20 E-1	6.35 E-1	6.35 E-1	6.42 E-1	6.72 E-1	6.29 E-1	6.37 E-1	5.92 E-1	6.32 E-1	2 E-2
141Ce	2.45	2.59	2.43	2.65	2.47	2.12	1.91	2.33	2.50	2.38	2 E-1
152Eu	2.87	2.89	2.84	2.85	3.01	2.63	2.51	2.65	2.76	2.78	2 E-1

(contd)

Table A1-1. (contd)

Test Number	L-287	L-288	L-289	L-290	L-291	L-292	L-293			
Sample mass, g	0.4517	0.4394	0.4447	0.4462	0.4577	0.4569	0.4367			
SRL 211*** S Activity, normalized μCi/g								Average of Specific Activities		
								Average	S.D.	
	¹⁰⁶ Ru	7.90	8.12	8.27	8.26	8.19	8.34	8.18	0.16	
	¹³⁷ Cs	6.6 E-2	1.19 E-1	1.64 E-1	1.22 E-1	1.27 E-1	1.31 E-1	1.22 E-1	0.03	
	¹⁴¹ Ce	9.33	13.0	12.6	13.1	13.4	12.8	12.4	1.5	
	¹⁵² Eu	22.1	22.8	22.3	22.9	22.9	22.7	22.6	0.3	
Test Number	L-339	L-340	L-341	L-342	L-343	L-344	L-345			
Sample mass, g	0.5157	0.4837	0.4788	0.4716	0.4816	0.4907	0.4721			
SRL 131* S Activity, normalized μCi/g										
	⁸⁵ Sr	4.81 E-1	6.0 E-1	4.8 E-1	4.8 E-1	5.0 E-1	5.7 E-1	5.9 E-1	5.3 E-1	5 E-2
	¹³³ Ba	1.2	1.3	1.2	1.3	1.3	1.3	1.3	1.3	5 E-2
	¹³⁷ Cs	8.7	8.7	9.2	6.1	8.1	8.6	7.8	8.1	9 E-1
	¹⁴¹ Ce	17.5	17.8	17.8	18.0	18.1	17.3	17.6	17.7	3 E-1
	¹⁵² Eu	47.1	47.1	47.0	47.0	47.1	46.9	47.4	47.1	2 E-1

*Activity has been decayed to T₀, the beginning of the leach test series.

^b3.3 E-1 reads as 3.3 × 10⁻¹.

were suspended from the center of the lid or from a rod that was molded into the lid by use of a teflon FEP, 0.125-mm monofilament. For radioactive samples, 125 mL-capacity polypropylene containers (Nalge 2105 from Scientific Products #B7533-4) were used. The samples rested on polypropylene mesh located 1/2 in. from the bottom of the container. The sample surface area contacted by the mesh was <5% of the total sample surface area.

- Balances--Sartorius 1602 MP (± 0.1 mg), Mettler PL 1200 (± 0.01 g).

1.3 Analytical Techniques

- pH was measured by a Cole Parmer #5987 digphase digital pH meter.
- γ -ray counting of radioactive samples was done by the University of Missouri, the Analytical Chemistry Laboratory (ACL)/CEN, and by the Fuel Cycle Section (FCS)/CEN. All results were calibrated against the counting system used by FCS/CEN and found to agree within 10%. The FCS/CEN detection and counting system includes an Ortec high-purity germanium detector, #1112-13180-S, coupled to a Tracer Northern TN-1710 multichannel analyzer. The Ge(Li) detector was calibrated by determining the system linearity and relative detector efficiency from the known gamma-ray energies and branching ratios of ^{152}Eu . The relative efficiencies are converted to absolute values by normalizing to point source amounts of ^{57}Co , ^{137}Cs , ^{134}Cs , ^{54}Mn , and ^{60}Co . Absolute standards for the calibration were obtained from the Amersham Radiochemical Center and the National Bureau of Standards.
- Silicon analysis of radioactive samples was done spectrophotometrically. Silicon (as silicate) reacts with ammonium molybdate in acid solution to form a silico-molybdate acid. This complex is reduced (reductant reagent is a solution of NaHSO_3 , NaOH and 1-amino-2-naphthol-4-sulfuric acid), forming a blue colloidal solution. The method has a relative standard deviation of 5%.
- Nonradioactive samples were analyzed for B, Cd, Mg, Zn, Si, Cu, Ni, Mn, Ca, Mo, Co, Al, Ti, V, Cl, Fe, Pb, and Ba using DCP spectroscopy. The analyses, which have a relative standard deviation of 5%, were done by Trace Element, Inc. of Park Ridge, IL.
- Nonradioactive samples were analyzed for sodium using atomic absorption (589.0 nm) with an air-acetylene flame. The asperated solution contains 1 mg/mL potassium and the relative standard deviation is 5%.
- Nonradioactive samples were analyzed for cesium using atomic absorption (852.1 nm) with a nitrous oxide-acetylene flame. The asperated solution contains 4 mg/mL potassium. The standard additions technique is used to analyze at least one sample in each leaching matrix, and the relative standard deviation is 5%.
- SEM analyses were done using an International Scientific Instruments Super II microscope, and EDAX analyses were done using a Princeton Gamma Tech System III energy-dispersive X-ray spectrometer. The samples were either carbon or gold coated.

1.4 Calibrations and Standards

Micrometer

- MG Tool Co., calibrated with a standard gauge block set supplied by the ACL/CEN.
- Calibrated at 6-mo. intervals.

Thermocouples

- Type K (chromel-alumel).
- Calibrated against a Chemical Thermometer #22640 made by the Brooklyn Thermometer Co., Farmingdale, NY, and calibrated factor certified Reference NBS Test No. 216642.
- Calibrated at 6-mo. intervals.

Balance

- Sartorius #1602 MP
- Calibrated by manufacturer with standard weights at 6 mo. intervals.

Direct Current Plasma

- Calibrated by Trace Elements, Inc. before running each set of leachates using their internal standards. Their calibration was checked using a standard leachate solution prepared by the ACL/CEN and standard solution SL-1 supplied by the MCC. Additionally, blanks containing fresh deionized (DI) water were periodically submitted for analysis.

1.5 Leachant Preparation

Deionized water produced by a Synbron/Barnstead #D2770 water purifier.

1.6 Specimen Preparation and Description

See Section II of the main report for a complete description of this process.

2.0 LEACHANT ANALYSIS

Analyses were done for each test series, and no impurities were detected in the DI water using DCP spectroscopy.

3.0 LEACHATE ANALYSIS

Analyses are presented in Tables A3-1 to A3-13.

Table A3-1. Analyses of Blanks

	L-286	L-297	L-310	L-324
Date run began	2/19/81	3/5/81	2/19/81	2/25/81
Initial pH	6.3	5.9	6.3	6.0
@ Temp, °C	22	22	22	22
Date run ended	3/19/81	4/2/81	3/19/81	3/23/81
Final pH	4.7	6.0	4.3	4.1
@ Temp, °C	22	23	22	22
V, mm ³	46	408	445	454
Wt leachant, g	46.0	40.8	44.5	45.4
Exp. init. wt, g	124.1	64.8	124.3	123.2
Exp. final wt, g	123.6	58.8	112.8 ^a	122.6
Exp. wt _i - exp. wt _f , ^b g	0.5	6.0	0	0.6
Blank loss, %	1.1	14.7	0	1.3
Init. pH - Final pH	1.6	-0.1	2.0	1.9

Constituent^c
Analysis, mg/L

B	<0.05	<0.05	d	<0.05
Cd				
Mg				
Zn				
Si				
Cu				
Ni				
Mn				
Ca				
Mo				
Co				
Al				
Ti				
V				
Cr				
Fe				
Pb				
Ba				

^aSome leachant spilled.

^bWt_i = initial weight, wt_f = final weight.

^cDetection limit for all elements is 0.05 mg/L.

^dNo analysis done.

Table A3-2. Leachate Analysis for DI Water Leachant, SRL 211** C Glass

	Specimen ID No.										
	L-154	L-154a	L-155	L-156	L-157	L-158	L-159	L-160	L-161	L-162	L-163
Run duration, d	3	3	5	7	14	14	14	28	42	42	42
Date run began	1/2/81	1/6/81	12/31/80	12/29/80	12/22/80	12/22/80	12/22/80	12/22/80	12/22/80	12/22/80	12/22/80
Initial pH	6.4	6.4	6.4	6.4	6.4	6.4	6.4	6.4	6.4	6.4	6.4
@ Temp, °C	19	20	19	19	20	20	20	20	20	20	20
Date run ended	1/5/81	1/9/81	1/5/81	1/5/81	1/5/81	1/5/81	1/5/81	1/19/81	2/2/81	2/2/81	2/2/81
Spec. SA, ^a mm ²	422	421	419	420	420	419	418	420	420	419	420
Wt leachant, g	42.2	42.2	41.9	42.1	41.8	41.9	42.0	42.2	42.0	41.8	41.9
Final pH	9.2	9.1	9.7	9.7	9.7	9.7	9.6	9.6	9.5	9.5	9.5
@ Temp, °C	20	20	20	20	20	20	20	21	22	22	22
Exp. initial wt, g	122.2	120.7	121.9	122.7	120.3	120.7	121.4	119.6	121.6	120.5	120.9
Exp. final wt, g	121.8	120.6	121.8	122.5	120.1	-	-	119.2	120.9	119.9	120.4
Exp. wt _i - exp wt _f ^b , g	0.4	0.1	0.1	0.2	0.2	-	-	0.4	0.7	0.6	0.5
Leachant loss, %	1.0	0.2	0.2	0.5	0.5	-	-	1.0	1.7	1.4	1.2
Init. pH-final pH	-2.8	-2.7	-3.3	-3.3	-3.3	-3.3	-3.2	-3.2	-3.1	3.1	3.1
Specimen											
Init. wt, g	0.3866	0.3655	0.3776	0.3833	0.3735	0.3889	0.3576	0.3760	0.3817	0.3634	0.3803
Final wt, g	0.3835	0.3623	0.3743	0.3795	0.3695	0.3850	0.3536	0.3712	0.3766	0.3589	0.3755
Wt loss, g×10 ⁻⁴	31	32	33	38	40	39	40	48	51	45	48
Constituent Analysis, mg/LC											
Na (0.003)	-	12.1	13.5	15.8	19.1	18.7	18.1	21.5	23.6	22.2	24.7
Li (0.003)	-	1.40	1.60	1.88	2.28	2.22	2.14	2.72	2.93	2.71	2.82
Ca (0.05)	-	0.12	0.12	0.14	0.17	0.17	0.16	0.19	0.21	0.19	0.20
Si (0.05)	-	20	18	19	21	21	21	23	26.2	25.2	25.5
B (0.05)	-	2.6	2.8	3.3	4.0	3.9	3.8	4.5	4.76	4.54	4.59
Mn (0.05)	-	0.05	<0.05	<0.05	<0.05	0.06	0.05	0.06	0.05	0.05	0.06
Cu (0.05)	-	2.4	2.5	2.4	2.6	2.6	2.6	2.7	2.48	2.65	2.61
Al (0.05)	-	1.8	1.6	1.8	1.9	1.8	1.9	1.8	1.87	1.85	1.88
Fe (0.05)	-	0.28	0.05	0.09	0.12	0.17	0.12	<0.05	0.09	0.08	0.13
Ba (0.05)	-	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05

^aSpec. SA = Specimen surface area.^bwt_i = Initial weight; wt_f = final weight.^cDetection limit is given in parenthesis.

Table A3-3. Leachate Analysis for DI Water Leachant, SRL 211** A Glass

	Specimen ID No.								
	L-134	L-135	L-136	L-137	L-138	L-139	L-140	L-141	L-142
Run duration, d	3.6	7	14.6	14.6	14.6	28	42.7	42.7	42.7
Date run began	12/29/80	12/24/80	12/18/80	12/18/80	12/18/80	12/18/80	12/18/80	12/18/80	12/18/80
Initial pH	6.4	6.3	6.3	6.3	6.3	6.3	6.3	6.3	6.3
@ Temp, °C	19	20	20	20	20	20	20	20	20
Date run ended	1/2/81	12/31/81	1/2/81	1/2/81	1/2/81	1/15/81	1/30/81	1/30/81	1/30/81
Spec. SA, ^a mm ²	369.1	363.8	361.4	367.4	367.4	379.5	386.8	372.5	374.2
Wt leachant, g	37.1	36.4	36.3	36.7	36.5	38.0	38.8	37.3	37.2
Final pH	9.8	9.7	9.6	9.6	9.8	9.7	9.8	9.7	9.8
@ Temp, °C	19	19	19	19	19	20	21	21	21
Exp. initial wt, g	68.1	68.2	69.0	66.7	65.1	68.1	69.0	65.5	64.5
Exp. final wt, g	68.1	68.0	62.9	65.2	58.3	65.5	62.0	63.4	60.8
Exp. wt _i - exp wt _f ^b	0	0.2	6.1	1.5	6.8	2.6	7.0	2.1	3.7
Leachant loss, %	0	0.6	16.8	4.1	18.6	6.8	18.0	5.6	10.0
Init. pH-final pH	-3.4	-3.4	-3.3	-3.3	-3.5	-3.4	-3.5	-3.4	-3.5
Specimen									
Init. wt, g	0.5874	0.5746	0.5473	0.5839	0.5525	0.6276	0.5273	0.5824	0.6147
Final wt, g	0.5844	0.5708	0.5430	0.5791	0.5485	0.6221	0.5213	0.5767	0.6085
Wt loss, g×10 ⁻⁴	30	38	43	48	40	55	60	57	62
Constituent Analysis, mg/L ^c									
Si (.005)	-	21.48	-	-	-	-	-	-	-

^aSpec. SA = Specimen surface area.^bwt_i = Initial weight; wt_f = final weight.^cDetection limit in parenthesis.

Table A3-4. Leachate Analysis for DI Water Leachant, SRL 211** S/A Glass

	Specimen ID No.								
	L-222	L-223	L-224	L-225	L-226	L-227	L-228	L-229	L-230
Run duration, d	3	7	14	14	14	28	42	42	42
Date run began	1/30/81	1/30/81	1/30/81	1/30/81	1/30/81	1/30/81	1/30/81	1/30/81	1/30/81
Initial pH	6.0	6.0	6.0	6.0	6.0	6.0	6.0	6.0	6.0
@ Temp, °C	21	21	21	21	21	21	21	21	21
Date run ended	2/2/81	2/6/81	2/13/81	2/13/81	2/13/81	2/27/81	3/13/81	3/13/81	3/13/81
Spec. SA, cm ²	390.0	392.7	392.6	385.8	391.3	396.9	395.0	394.8	397.0
Wt leachant, g	40.4	39.4	40.1	40.6	40.8	40.4	39.6	39.9	40.4
Final pH	9.5	9.7	9.4	9.4	9.4	9.3	9.7	9.5	9.7
@ Temp, °C	22	21	22	22	22	20	21	21	21
Exp. initial wt, g	64.7	63.4	64.1	64.3	64.4	64.3	63.9	63.7	64.6
Exp. final wt, g	64.0	61.9	61.5	61.6	61.7	59.1	56.6	56.2	57.3
Exp. wt _i - exp wt _f	0.7	1.5	2.6	2.7	2.7	5.2	7.3	7.5	7.3
Leachant loss, %	1.7	3.8	6.5	6.7	6.6	12.9	18.4	18.8	18.1
Init. pH-final pH	-3.5	-3.7	-3.4	-3.4	-3.4	-3.3	-3.7	-3.5	-3.7
Specimen									
Init. wt, g	0.4383	0.4505	0.4577	0.4537	0.4528	0.4532	0.4542	0.4504	0.4572
Final wt, g	0.4360	0.4471	0.4535	0.4491	0.4486	-	0.4496	0.4460	0.4525
Wt loss, g x 10 ⁻⁴	23	35	42	46	42	-	46	44	47
Constituent Analysis ^{a,b,c,d,e}									
⁵¹ Cr A _l (310)	<1.58 E-4	4.84 E-4	4.01 E-4	3.54 E-4	5.22 E-4	9.93 E-4	1.02 E-3	7.82 E-4	1.45 E-3
A _s (310)	3.46 E-1	2.29 E-1	2.58 E-1	2.90 E-1	2.17 E-1	5.60 E-1	2.19 E-1	2.43 E-1	
¹²⁴ Sb A _l (1691)	1.13 E-3	1.71 E-3	1.90 E-3	2.32 E-3	2.49 E-3	2.52 E-3	3.46 E-3	3.23 E-3	1.65 E-3
A _s (1691)	3.45 E-1	3.50 E-1	3.58 E-1	3.45 E-1	3.79 E-1	3.59 E-1	3.68 E-1	3.69 E-1	3.76 E-1
¹³³ Ba A _l (356)	1.09 E-4	7.47 E-5	5.70 E-5	8.02 E-5	9.67 E-5	7.53 E-5	5.77 E-5	5.06 E-5	5.47 E-5
A _s (356)	3.56 E-1	3.59 E-1	3.76 E-1	3.68 E-1	3.82 E-1	3.96 E-1	3.64 E-1	3.65 E-1	3.46 E-1
¹³⁴ Cs A _l (795)	1.10 E-2	1.83 E-2	1.87 E-2	1.99 E-2	2.10 E-2	2.32 E-2	2.80 E-2	2.63 E-2	2.51 E-2
A _s (795)	2.27	2.40	2.40	2.33	2.40	2.21	2.21	2.29	2.36
¹³⁷ Cs A _l (661)	1.51 E-3	2.46 E-3	2.60 E-3	2.83 E-3	2.89 E-3	3.65 E-3	4.13 E-3	3.76 E-3	3.33 E-3
A _s (661)	2.72 E-1	2.77 E-1	2.88 E-1	2.85 E-1	2.88 E-1	3.02 E-1	2.83 E-1	2.84 E-1	2.68 E-1
¹⁴¹ Ce A _l (145)	5.6 E-6 ^f		6.4 E-6 ^f	8.4 E-6 ^f					
A _s (145)	1.07	1.16	1.10	1.19	1.11	9.5 E-1	8.6 E-1	1.04	1.13
¹⁵² Eu A _s	3.4 E-5 ^f			3 E-5 ^f		4.1 E-5 ^f		1.1 E-5 ^f	
A _l	1.25	1.29	1.29	1.28	1.35	1.18	1.13	1.18	1.25

^aγ-counting done by University of Missouri. Count time for leachates = 54,000 s; for solids = 3,600 s.^bPrincipal γ-ray energy (keV) used in analysis in parenthesis, A_l = activity leachate, A_s = activity solid.^cSolids were counted two times and results are averaged.^dUnits are μCi.^eBackground corrections = 0.^fError too large to use number in (NL)_i calculations.^gPrincipal γ-rays used were 121, 964, and 1408 keV.

Table A3-5. Leachate Analysis for DI Water Leachant, SRL 211** S Glass

	Specimen ID No.								
	L-257	L-258	L-259	L-260	L-261	L-262	L-263	L-264	L-265
Run duration, d	3	7	14	14	14	28	42	42	42
Date run began	1/30/81	1/30/81	1/30/81	1/30/81	1/30/81	1/30/81	1/30/81	1/30/81	1/30/81
Initial pH	6.0	6.0	6.0	6.0	6.0	6.0	6.0	6.0	6.0
@ Temp, °C	21	21	21	21	21	21	21	21	21
Date run ended	2/2/81	2/6/81	2/13/81	2/13/81	2/13/81	2/27/81	3/13/81	3/13/81	3/13/81
Spec. SA, mm ²	373.0	391.2	373.6	394.4	399.2	388.6	398.4	397.9	392.0
Wt leachant, g	39.8	40.4	40.1	39.7	40.4	40.8	40.3	40.8	40.1
Final pH	9.5	9.5	9.4	9.4	9.4	9.4	9.6	9.6	9.4
@ Temp, °C	22	21	22	22	22	20	21	21	21
Exp. initial wt, g	63.9	64.2	64.1	63.6	64.2	64.8	64.1	64.4	64.4
Exp. final wt, g	63.1	62.8	61.5	60.9	61.7	59.9	56.4	56.7	57.1
Exp. wt _i - exp wt _f	0.8	1.4	2.6	2.7	2.5	4.9	7.7	7.7	7.3
Leachant loss, %	2.0	3.5	6.5	6.8	6.2	12.0	19.1	18.9	18.2
Init. pH-final pH	-3.5	-3.5	-3.4	-3.4	-3.4	-3.4	-3.6	-3.6	-3.4
Specimen									
Init. wt, g	0.4265	0.4549	0.4297	0.4152	0.4493	0.5513	0.4597	0.4433	0.4483
Final wt, g	0.4240	0.4511	0.4253	0.4108	0.4452	-	0.4552	0.4389	0.4436
Wt loss, g x 10 ⁻⁴	25	38	44	44	41	-	45	44	47
Constituent Analysis ^{a,b,c,d,e}									
88Y A ₁ (898)						3 E-6 ^f			2 E-6 ^f
A _s (898)	1.40 E-1	1.58 E-1	1.50 E-1	1.49 E-1	1.44 E-1	2.04 E-1	1.49 E-1	1.32 E-1	1.16 E-1
106Ru A ₁ (622)									1.6 E-5 ^f
A _s (622)	1.85 E-1	2.33 E-1	2.16 E-1	2.14 E-1	1.92 E-1	2.76 E-1	2.25 E-1	2.03 E-1	1.76 E-1
133Ba A ₁ (356)	1.49 E-4	7.51 E-5	7.28 E-5	7.50 E-5	8.96 E-5	6.75 E-5	6.12 E-5	5.66 E-5	4.77 E-5
A _s (356)	3.62 E-1	3.81 E-1	3.91 E-1	3.57 E-1	3.81 E-1	4.92 E-1	3.99 E-1	3.70 E-1	3.38 E-1
137Cs A ₁ (661)	1.78 E-3	2.20 E-3	2.85 E-3	2.81 E-3	3.03 E-3	3.36 E-3	4.02 E-3	3.78 E-3	3.65 E-3
A _s (661)	2.80 E-1	2.98 E-1	2.92 E-1	2.75 E-1	2.94 E-1	3.79 E-1	3.12 E-1	3.87 E-1	3.68 E-1
139Ce A ₁ (165)									1.6 E-6 ^f
A _s (165)	1.36 E-1	1.56 E-1	1.51 E-1	1.42 E-1	1.42 E-1	1.96 E-1	1.44 E-1	1.34 E-1	1.14 E-1
152Eu ^g A ₁	<1.78 E-5	<1.77 E-5	1.21 E-5	<7.26 E-6	<1.93 E-5	4.38 E-4	<1.8 E-5	<1.8 E-5	1.65 E-5
A _s	4.40 E-1	4.77 E-1	4.86 E-1	4.33 E-1	4.15 E-1	5.32 E-1	4.16 E-1	3.67 E-1	3.04 E-1
Si ^h , mg/L							36.2	35.3	37.2

^aγ-counting done by University of Missouri, count time for leachates 54,000 seconds, for solids 3,600 seconds.

^bPrincipal γ-ray energy (keV) used in analysis in parenthesis; A₁ = activity leachate, A_s = activity solid.

^cSolids were counted two times and results are averaged.

^dUnits are μCi.

^eBackground corrections = 0.

^fError too large to use number in (NL)_i calculations.

^gPrincipal γ-rays used were 121, 964, and 1408 keV.

^hAnalysis by colorimetry, units mg/L.

Table A3-6. Leachate Analysis for DI Water Leachant, SRL 211*** C Glass

	Specimen ID No.						
	L-276	L-277	L-278	L-279	L-280	L-281	L-292
Run duration, d	3	7	14	28	28	28	42
Date run began	3/2/81	2/26/81	2/19/81	2/19/81	2/19/81	2/19/81	2/19/81
Initial pH	6.3	6.3	6.3	6.3	6.3	6.3	6.3
@ Temp, °C	22	21	20	20	20	20	20
Date run ended	3/5/81	3/5/81	3/5/81	3/19/81	3/19/81	3/19/81	4/2/81
Spec. SA, mm ²	413	422	424	428	417	427	421
Wt leachant, g	44.5	42.5	42.3	45.3	44.6	45.3	44.9
Final pH	9.7	9.7	9.7	9.8	10.0	9.8	9.5
@ Temp, °C	22	22	22	22	22	22	22
Exp. initial wt, g	124.7	-	122.6	125.2	124.8	125.2	124.9
Exp. final wt, g	124.6	121.7	122.2	124.6	124.3	124.7	124.2
Exp. wt _i - exp wt _f	0.1	-	0.4	0.6	0.5	0.5	0.7
Leachant loss, %	0.2	-	0.9	1.3	1.1	1.1	1.6
Init. pH-final pH	-3.4	-3.4	-3.4	-3.5	-3.7	-3.5	-3.2
Specimen							
Init. wt, g	0.4056	0.3974	0.3970	0.4048	0.3835	0.4129	0.3748
Final wt, g	0.4023	0.3938	0.3027	0.3999	0.3786	0.4081	0.3697
Wt loss, g x 10 ⁻⁴	33	36	43	47	49	48	51
Constituent							
Analysis, mg/L ^{a,b}							
Na (0.003)	11.5	16.4	18.0	19.2	19.8	19.6	21.4
Li (0.003)	1.49	2.03	2.23	2.53	2.60	2.52	2.81
Si (0.05)	15.4	20.2	22.0	21.4	24.3	24.3	27.0
B (0.05)	2.56	3.45	3.91	4.50	4.49	4.01	4.76
Mn (0.05)	0.06	0.06	0.05	0.06	0.05	0.06	0.06
Ca (0.05)	2.43	2.47	2.72	2.76	2.66	2.23	2.68
Al (0.05)	1.54	1.86	1.76	1.82	1.76	1.95	1.53
Fe (0.05)	0.12	0.11	0.13	0.16	0.09	0.12	0.11
Ba (0.05)	0.07	0.06	0.06	0.05	<0.05	0.05	<0.05

^aDetection limits in parenthesis.^bNo correction for blank is necessary.

Table A3-7. Leachate Analysis for DI Water Leachant, SRL 211*** A Glass

		Specimen ID No.						
		L-237	L-238	L-239	L-240	L-241	L-242	L-243
Run duration, d		3	7	14	28	28	28	43
Date run began		2/9/81	2/9/81	2/9/81	2/9/81	2/9/81	2/9/81	2/9/81
Initial pH		5.6	5.6	5.6	5.6	5.6	5.6	5.6
@ Temp, °C		21	21	21	21	21	21	21
Date run ended		2/12/81	2/16/81	2/23/81	3/9/81	3/9/81	3/9/81	3/24/81
Spec. SA, mm ²		430	428	436	426	430	430	434
Wt leachant, g		40.2	40.2	39.8	45.0	40.2	40.0	40.2
Final pH		9.4	9.4	9.6	9.6	9.6	9.6	7.2
@ Temp, °C		21	21	20	22	22	22	22
Exp. initial wt, g		64.7	64.1	63.9	68.6	64.4	63.9	64.3
Exp. final wt, g		64.0	62.6	61.2	63.1	58.8	58.3	56.6
Exp. wt _i - exp wt _f		0.7	1.5	2.7	5.5	5.6	5.6	7.7
Leachant loss, %		1.7	3.7	6.8	12.2	13.9	14.0	19.2
Init. pH-final pH		-3.8	-3.8	-4.0	-4.0	-4.0	-4.0	-1.6
Specimen								
Init. wt, g		0.4194	0.3878	0.3993	0.3712	0.4119	0.3694	0.4054
Final wt, g		0.4117	0.3842	0.3935	0.3659	0.4070	0.3609	0.3942
Wt loss, gx10 ⁻⁴		77	36	58	53	49	85	112
Constituent								
Analysis ^{a,b,c,d,e}								
⁵⁹ Fe	A ₁ (1099)	4.2 E+2	3.8 E+2	2.6 E+2	1.8 E-4	3.0 E-4	1.3 E-4	3.1 E-4
	A _s (1099)	7.3 E+6	6.8 E+6	6.9 E+6	2.00	2.13	2.48	2.8
⁸⁵ Sr	A ₁ (514)	3.0 E+2	3.4 E+2	3.4 E+2	1.3 E-4	8.7 E-5	1.1 E-4	8.5 E-5
	A _s (514)	3.1 E+5	3.3 E+5	3.0 E+5	1.3 E-1	1.4 E-1	1.3 E-1	1.4 E-1

(contd)

Table A3-7. (contd)

			Specimen ID No.													
			L-237		L-238		L-239		L-240		L-241		L-242		L-243	
¹²⁴ Sb	A ₁	(1691)	1.8	E+4	2.7	E+4	3.4	E+4	1.9	E-2	1.6	E-2	1.7	E-2	1.7	E-2
	A _S	(1691)	4.5	E+6	3.7	E+6	4.7	E+6	1.3		1.3		1.3		1.6	
¹³¹ Ba	A ₁	(216,496)	3	E+3	2.0	E+3	-		-		-		-		-	
	A _S	(216,496)	6.5	E+6	6.0	E+6	-		-		-		-		-	
¹³⁴ Cs	A ₁	(795)	1.2	E+3	1.5	E+3	1.9	E+3	9.9	E-4	9.2	E-4	9.4	E-4	1.0	E-3
	A _S	(795)	5.3	E+5	4.9	E+5	5	E+5	6	E-2	6.5	E-2	6.0	E-2	8.0	E-2
¹⁴¹ Ce	A ₁	(145)	9.8	E+2	9.6	E+2	8.4	E+2	3.5	E-4	1.4	E-3	4.2	E-4	1.0	E-3
	A _S	(145)	4.5	E+7	4.0	E+7	4.2	E+7	11.5		14.0		10.5		19.3	
¹⁴⁷ Nd	A ₁	(91,531)	3.2	E+3	2.6	E+3	3.0	E+3	-		-		-		-	
	A _S	(91,531)	8.8	E+7	8.0	E+7	1.7	E+8	-		-		-		-	
¹⁵² Eu	A ₁	(121,344, 1408)	3.5	E+3	2.9	E+3	3.0	E+3	1.2	E-3	4.4	E-3	1.4	E-3	3.6	E-3
	A _S	(121, 344, 1408)	1.8	E+8	1.6	E+8	1.7	E+8	51.4		57.2		57.2		70.5	
Si (0.005) ^f			-		-		-		-		32.5		33.2		-	

^a γ -counting for samples L-237, L-238, and L-239 done by ACL/CEN; leachates counted for 29,000 s, solids for 3,600 s; the remaining samples were counted by the University of Missouri.

^b Principal γ -ray energy (keV) used in analysis in parenthesis.

^c For L-237, L-238, L-239 units are dpm at T₀; the rest of the units are μ Ci at T₀.

^d Solids counted by the University of Missouri were counted two or three times and an average was taken.

^e Background corrections are zero.

^f Analysis by calorimetry, mg/L. Detection limit in parenthesis.

Table A3-8. Leachate Analysis for DI Water Leachant, SRL 211*** S Glass

	Specimen ID No.						
	L-287	L-288	L-289	L-290	L-291	L-292	L-293
Run duration, d	3	7	14	28	28	28	42
Date run began	3/6/81	3/5/81	3/5/81	3/5/81	3/5/81	3/5/81	3/5/81
Initial pH	5.9	5.9	5.9	5.9	5.9	5.9	5.9
@ Temp, °C	22	22	22	22	22	22	22
Date run ended	3/9/81	3/12/81	3/19/81	4/2/81	4/2/81	4/2/81	4/16/81
Spec. SA, mm ²	411	408	409	409	412	411	408
Wt leachant, g	40.7	40.9	39.3	40.4	43.4	40.0	39.7
Final pH	9.6	9.8	9.5	9.5	9.0	9.4	9.5
@ Temp, °C	21	22	22	22	22	22	22
Exp. initial wt, g	64.7	64.8	63.6	64.4	67.5	64.0	63.5
Exp. final wt, g	63.9	63.3	60.8	58.9	62.0	58.6	55.2
Exp. wt _i - exp wt _f	0.6	1.5	2.8	5.5	5.5	5.4	8.3
Leachant loss, %	1.5	3.7	7.1	13.6	12.7	13.5	20.9
Init. pH-final pH	-3.7	-3.9	-3.6	-3.6	-3.1	-3.5	-3.6
Specimen							
Init. wt, g	0.4541	0.4430	0.4482	0.4501	0.4618	0.4607	0.4407
Final wt, g	0.4517	0.4394	0.4447	0.4462	0.4577	0.4569	0.4367
Wt loss, gx10 ⁻⁴	24	36	35	39	41	38	40
Constituent							
Analysis ^{a,b,c,e,g}							
¹⁰⁶ Ru A ₁ (622)	-	-	-	2.46 E-5 ^f	1.42 E-4 ^f	-	4.65 E-5 ^f
A _g (622)	-	3.47	3.61	3.69	3.78	3.74	3.64

(contd)

Table A3-8. (contd)

		Specimen ID No.						
		L-287	L-288	L-289	L-290	L-291	L-292	L-293
¹³⁷ Cs	A ₁ (661)	-	4.92 E-4	2.80 E-4	2.56 E-4	2.95 E-4	3.22 E-4	3.94 E-4
	A _s (661)	-	2.9 E-2	5.3 E-2	7.3 E-2	5.6 E-2	5.8 E-2	5.7 E-2
¹³⁹ Cs	A ₁ (165)	-	-	-	1.19 E-5	6.97 E-6	-	-
	A _s (165)	-	1.1 E-1	1.3 E-1	1.3 E-1	1.4 E-1	1.4 E-1	1.4 E-1
¹⁴¹ Ce	A ₁ (145)	-	1.37 E-5	-	3.09 E-4	2.75 E-4	-	7.6 E-5
	A _s (145)	-	4.1	5.8	5.6	6.0	6.1	5.6
¹⁵² Eu ^g	A ₁	-	4.0 E-5	1.5 E-5	4.64 E-4	4.07 E-4	2.5 E-5	1.25 E-4
	A _s	-	9.73	10.12	9.94	10.50	10.48	9.92
Si (0.005) ^h		-	-	-	-	-	30.8	-

^aγ-counting done by University of Missouri, except for L-287, which was done by ACL/CEN. Count time for leachates = 54,000 s, for solids = 3,600 s.

^bPrincipal γ-ray energy (KeV) used in analysis in parenthesis.

^cSolids were counted two times and results are averaged; error for ¹³⁷Cs solid is larger than for previous counts due to lower amount of Cs in the solid, est. error ~40%.

^dPrincipal γ-rays used were 121, 964, and 1408 keV.

^eUnits are μCi.

^fError too large to use number in (NL)_i calculation.

^gBackground corrections = 0

^hAnalysis by colorimetry, mg/L. Detection limits in parenthesis.

Table A3-9. Leachate Analysis for DI Water Leachant, SRL 211*** S Glass (Repeat)

	Specimen ID No.						
	L-475	L-476	L-477	L-478	L-479	L-480	L-481
Run duration, d	3	7	14	28	28	28	42
Date run began	7/21/81	7/21/81	7/21/81	7/21/81	7/21/81	7/21/81	7/21/81
Initial pH	5.6	5.6	5.6	5.6	5.6	5.6	5.6
@ Temp, °C	24	24	24	24	24	24	24
Date run ended	7/24/81	7/28/81	8/5/81	8/17/81	8/17/81	8/17/81	8/31/81
Spec. SA, mm ²	373.1	374.3	373.8	370.9	369.9	372.8	370.3
Wt leachant, g	38.1	37.6	37.5	37.2	37.0	36.9	37.3
Final pH	9.2	9.4	9.1	9.3	9.6	9.4	9.4
@ Temp, °C	25	25	24	25	25	25	24
Exp. initial wt, g	63.5	64.7	63.7	64.7	64.2	64.3	64.4
Exp. final wt, g	62.8	63.2	60.9	61.8	61.6	62.8	56.7
Exp. wt _i - exp wt _f	0.7	1.5	2.8	2.9	2.6	1.5	7.7
Leachant loss, %	1.8	4.0	7.5	7.8	7.0	4.1	20.6
Init. pH-final pH	-3.6	-3.8	-3.5	-3.7	-4.0	-3.8	-3.8
Specimen							
Init. wt, g	0.4235	0.4228	0.4964	0.4362	0.4255	0.4376	0.4325
Final wt, g	0.4211	0.4193	0.4921	0.4319	0.4215	0.4327	0.4286
Wt loss, g x 10 ⁻⁴	24	35	43	43	40	49	39
Constituent Analysis ^{a,b,c,d}							
¹³⁷ Cs A ₁ (661)	1.1 E+1	1.4 E+1	1.7 E+1	1.9 E+1	2.7 E+1	1.8 E+1	1.7 E+1
A ₂ (661)	8.4 E+2	8.7 E+2	1.2 E+3	1.0 E+3	9.4 E+2	9.7 E+2	8.5 E+2
¹⁵² Eue A ₁ (121)	5.2 E-1	2.4 E-1	3.6 E-1	8.0 E-1	6.7 E-1	3.1 E-1	6.9 E-1
A ₂ (121)	1.0 E+5	1.0 E+5	1.2 E+5	1.0 E+5	1.0 E+5	1.0 E+5	1.0 E+5

^a γ-counting done by FCS; count time for leachates = ~150,000 s, for solids = ~3,600 s.

^b Principal γ-ray energy (keV) used in analysis in parenthesis.

^c Units are cps adjusted for detector efficiency.

^d Background corrections are included.

^e Principal γ-rays used were 121, 344, and 1408 keV, but only cps for 121 are reported, because of efficiency differences.

Table A3-10. Leachate Analysis for DI Water Leachant, SRL 131* C Glass

	Specimen ID No.						
	L-311	L-312	L-313	L-314	L-315	L-316	L-317
Run duration, d	3	7	14	28	28	28	37
Date run began	3/6/81	3/2/81	2/23/81	2/23/81	2/23/81	2/23/81	2/23/81
Initial pH	6.3	6.1	6.0	6.0	6.0	6.0	6.0
@ Temp, °C	22	22	21	21	21	21	21
Date run ended	3/9/81	3/9/81	3/9/81	3/23/81	3/23/81	3/23/81	4/2/81
Spec. SA, mm ²	415	332	429	414	443	364	412
Wt leachant, g	43.0	37.9	46.7	45.4	45.5	46.5	46.1
Final pH	9.7	9.7	9.7	9.8	9.8	9.8	9.6
@ Temp, °C	23	23	23	23	23	23	23
Exp. initial wt, g	120.9	116.3	126.9	123.6	123.7	126.7	126.3
Exp. final wt, g	120.9	116.1	126.6	123.1	123.1	126.1	125.6
Exp. wt _i - exp wt _f	0	0.2	0.3	0.5	0.6	0.6	0.9
Leachant loss, %	0	0.5	0.7	1.2	1.4	1.7	2.2
Init. pH-final pH	-3.4	-3.6	-3.7	-3.8	-3.8	-3.8	-3.6
Specimen							
Init. wt, g	0.4527	0.3030	0.4025	0.3693	0.4887	0.3891	0.4556
Final wt, g	0.4498	0.2991	0.3970	0.3626	0.4818	0.3826	0.4482
Wt loss, gx10 ⁻⁴	29	39	55	67	69	65	74

(contd)

Table A3-10. (contd)

Constituent Analysis, mg/L ^{a,b}	Specimen ID No.						
	L-311	L-312	L-313	L-314	L-315	L-316	L-317
Na (0.003)	-	-	-	25.5	25.2	24.5	27.2
Li (0.003)	-	-	-	4.87	4.91	4.51	5.22
Si (0.05)	16.0	24.1	28.7	34.2	34.6	32.6	39.2
B (0.05)	3.68	5.36	6.88	8.86	8.96	8.42	8.79
Mn (0.05)	<0.05	0.05	0.05	0.06	0.06	0.07	0.08
Ca (0.05)	0.33	0.11	0.06	<0.05	<0.05	<0.05	<0.05
Al (0.05)	1.39	1.91	1.98	4.14	1.87	1.77	2.00
Fe (0.05)	0.09	0.07	0.05	<0.05	<0.05	<0.05	<0.05
Ba (0.05)	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05
Ti (0.05)	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05
Mg (0.05)	<0.05	<0.05	<0.05	<0.05	<0.05	0.07	0.06

^aDetection limits are in parenthesis.

^bNo correction for blank is necessary.

Table A3-11. Leachate Analysis for DI Water Leachant, SRL 131* A Glass

		Specimen ID No.				
		L-327	L-328	L-329	L-330	L-331
Run duration, d		2.25	7	14	28	28
Date run began		3/18/81	3/18/81	3/18/81	3/18/81	3/18/81
Initial pH		6.2	6.2	6.2	6.2	6.2
@ Temp, °C		23	23	23	23	23
Date run ended		3/20/81	3/25/81	4/1/81	4/15/81	4/15/81
Spec. SA, mm ²		425	390	386	433	421
Wt leachant, g		37.7	39.1	39.3	40.6	39.1
Final pH		9.4	9.5	9.6	9.8	9.7
@ Temp, °C		23	23	23	24	24
Exp. initial wt, g		61.7	63.3	63.2	64.5	63.2
Exp. final wt, g		60.7	61.8	59.8	59.3	58.1
Exp. wt _i - exp wt _f		1.0	1.5	3.4	5.2	5.1
Leachant loss, %		2.7	3.8	8.7	12.8	13.0
Init. pH-final pH		-3.2	-3.3	-3.4	-3.6	-3.5
Specimen						
Init. wt, g		0.4632	0.3986	0.3989	0.4595	0.4067
Final wt, g		0.3091	0.3971	0.3965	0.4546	0.4018
Wt loss, g x 10 ⁻⁴		21	15	24	39	49
Constituent Analysis ^{a,b,c}						
⁵⁹ Fe	A ₁ (1099)	4.0 E+2	-	-	1.5 E-4	1.5 E-4
	A _s (1099)	6.6 E+6	-	-	3.8	3.6
⁸⁵ Sr	A ₁ (1099)	3.2 E+2	1.1 E+2	1.7 E+2	-	-
	A _s (1099)	4.3 E+5 ^d	6.0 E+5 ^d	6.7 E+5 ^d	-	-
¹³⁴ Cs	A ₁ (795)	5.3 E+3	3.2 E+3	3.7 E+3	3.7 E-3	4.3 E-3
	A _s (795)	6.4 E+5 ^d	7.6 E+5 ^d	9.0 E+5 ^d	3.2 E-1 ^d	4.1 E-1 ^d
¹⁴¹ Ce	A ₁ (145)	7.2 E+2	3.1 E+2	1.6 E+2	1.6 E-4	1.0 E-4
	A _s (145)	4.3 E+7	5.0 E+7	4.4 E+7	23.1	20.3
¹⁵² Eu	A ₁ ^e (121)	5.0 E+3	1.1 E+3	4.9 E+2	3.5 E-4	3.2 E-4
	A _s (121)	1.7 E+8	2.0 E+8	1.7 E+8	83.6	75.4
Si (0.005) ^f		-	-	-	41.2	43.2

^a γ-counting done by ACL/CEN for samples L-327, L-328, L-329 with units dpm at T₀. Samples L-330 and L-331 were counted by University of Missouri with units of μCi.

^b Principal γ-ray energy (keV) used in analysis in parenthesis.

^c Background corrections are included.

^d Significant counting error (>25%).

^e Principal γ-rays used in analysis were 121, 344, and 1408 keV.

^f Analysis by colorimetry, mg/L. Detection limit in parenthesis.

Table A3-12. Leachate Analysis for DI Water Leachant, SRL 131* S Glass

	Specimen ID No.						
	L-339	L-340	L-341	L-342	L-343	L-344	L-345
Run duration, d	3	7	14	28	28	28	42
Date run began	3/23/81	3/26/81	3/26/81	3/26/81	3/26/81	3/26/81	3/26/81
Initial pH	5.8	5.8	5.8	5.8	5.8	5.8	5.8
@ Temp, °C	23	23	23	23	23	23	23
Date run ended	3/26/81	4/2/81	4/9/81	4/23/81	4/23/81	4/27/81	5/7/81
Spec. SA, mm ²	435	426	427	428	429	429	427
Wt leachant, g	40.0	40.7	40.9	40.0	39.7	40.2	40.0
Final pH	9.5	9.3	9.4	9.2	9.0	9.3	9.2
@ Temp, °C	23	23	23	23	23	23	23
Exp. initial wt, g	63.6	64.7	64.7	63.7	63.8	63.9	64.2
Exp. final wt, g	63.2	63.0	61.5	58.1	58.3	58.2	56.1
Exp. wt _i - exp wt _f	0.4	1.7	3.2	4.6	5.5	5.7	8.1
Leachant loss, %	1.0	4.2	7.8	11.5	13.9	14.2	20.3
Init. pH-final pH	-3.7	-3.5	-3.6	-3.4	-3.2	-3.5	-3.4
Specimen							
Init. wt, g	0.5174	0.4869	0.4842	0.4772	0.4866	0.4965	0.4773
Final wt, g	0.5157	0.4837	0.4788	0.4416	0.4816	0.4907	0.4721
Wt loss, g $\times 10^{-4}$	17	22	54	56	50	58	51

(contd)

Table A3-12. (contd)

		Specimen ID No.						
		L-339	L-340	L-341	L-342	L-343	L-344	L-345
Constituent Analysis ^{a,b,c,d,e}								
⁸⁵ Sr	A ₁ (514)	4.1E-1	1.8E-4	1.8E-4	1.8E-4	3.2E-4	1.9E-4	1.7E-4
	A _S (514)	2.5E-1	2.9E-1	2.3E-1	2.3E-1	2.4E-1	2.8E-1	2.8E-1
¹³³ Ba	A ₁ (356)	2.0E-4	5.8E-5	1.4E-4	8.7E-5	1.4E-4	2.0E-4	1.2E-4
	A _S (356)	6.0E-1	6.2E-1	5.9E-1	6.0E-1	6.2E-1	6.4E-1	5.9E-1
¹³⁷ Cs	A ₁ (661)	7.2E-5	1.3E-4	2.0E-4	2.2E-4	1.9E-4	2.1E-4	2.4E-4
	A _S (661)	4.3	4.2	4.4	2.9 ^f	2.9 ^f	4.2 ^f	3.7 ^f
¹⁴¹ Ce	A ₁ (145)	1.4 ^f	5.7E-5	2.0E-3	2.7E-4	8.5E-4	2.4E-3	5.7E-4
	A _S (145)	9.0	8.6	8.5	8.6	8.7	8.5	8.3
¹⁵² Eu ^g	A ₁	5.2E-5	1.7E-4	4.4E-3	6.1E-4	2.0E-3	5.5E-3	1.4E-3
	A _S	24.3	22.8	22.5	22.5	22.7	23.0	22.4
Si ^h (0.005)		-	-	-	43.3	36.2	46.5	-

^a γ -counting done by University of Missouri. Count time for leachates = 54,000 s, for solids = 3,600 s.

^b Principal γ -ray energy (KeV) used in analysis in parenthesis.

^c Solids were counted two times and results are averaged.

^d Units are μ Ci.

^e Background corrections = 0.

^f Counting error too large to use number in (NL)_i calculation.

^g Principal γ -rays used were 121, 964, and 1408 KeV.

^h Analysis by colorimetry, mg/L. Detection limit in parenthesis.

Table A3-13. Leachate Analysis for DI Water Leachant, SRL 131* S Glass (Repeat)

	Specimen ID No.						
	L-468	L-469	L-470	L-471	L-472	L-473	L-474
Run duration, d	3	7	14	28	28	28	56
Date run began	7/17/81	7/16/81	7/16/81	7/16/81	7/16/81	7/16/81	7/16/81
Initial pH	5.6	5.6	5.6	5.6	5.6	5.6	5.6
@ Temp, °C	25	25	25	25	25	25	25
Date run ended	7/20/81	7/23/81	7/30/81	8/13/81	8/13/81	8/13/81	9/10/81
Spec. SA, mm ²	373.1	374.3	373.8	370.9	369.9	372.8	370.3
Wt leachant, g	40.1	39.7	40.5	40.1	39.9	40.7	40.8
Final pH	9.2	8.8	9.6	9.0	9.0	5.3	-
@ Temp, °C	25	24	24	24	24	24	23
Exp. initial wt, g	63.9	63.5	64.5	64.3	64.0	119.5	64.7
Exp. final wt, g	63.3	62.1	61.9	59.3	59.0	118.4	-
Exp. wt _i - exp wt _f	0.6	1.4	2.6	5.0	5.0	1.1	-
Leachant loss, %	1.5	3.5	6.4	12.5	12.5	2.7	-
Init. pH-final pH	-3.6	-3.2	-4.0	-3.4	-3.4	+0.3	-
Specimen							
Init. wt, g	0.3989	0.4074	0.3993	0.3792	0.3833	0.4019	0.3888
Final wt, g	0.3972	0.4042	0.3955	0.3741	0.3783	0.3988	-
Wt loss, gx10 ⁻⁴	17	32	38	51	50	51	-

(contd)

Table A3-13. (contd)

			Specimen ID No.						
			L-468	L-469	L-470	L-471	L-472	L-473	L-474
Constituent Analysis ^a									
⁸⁵ Sr	A ₁	(514)	5.8	4.7	2.9	1.6	1.5	-	-
	A _S	(514)	1.6 E+3	2.8 E+3	1.9 E+3	1.7 E+3	1.3 E+3	-	-
¹³³ Ba	A ₁	(356)	7.3	3.1	1.5	1.1	1.1	-	6.9 E-1
	A _S	(356)	1.0 E+4	1.0 E+4	1.0 E+4	9.5 E+3	9.5 E+3	-	9.6 E+3
¹³⁷ Cs	A ₁	(661)	4.7	7.2	9.8	11.2	11.3	-	11.1
	A _S	(661)	5.7 E+2 ^b	5.1 E+2 ^b	4.8 E+2 ^b	6.1 E+2 ^b	5.0 E+2 ^b	-	5.2 E+2 ^b
¹⁵² Eu ^c	A ₁	(344)	3.0 E-1	1.1	2.9 E-1	1.5	2.4	-	4.2 E-1
	A _S	(344)	1.8 E+5	1.8 E+5	1.8 E+5	1.7 E-5	1.7 E+5	-	1.7 E+5

^a γ -counting done by FCS/CEN and units are counts per second. Solid counts were for 3,600-8,200 s and leachate counts were for 160,000 to 231,000 s. Background corrections are negligible and principal γ -ray energies used in analyses are given in parenthesis (keV).

^b Significant counting error (>25%) exists.

^c Principal γ -rays used were 121, 344 and 1408 keV.

4.0 TABULATED AND PLOTTED DATA

The data are tabulated as $(NL)_i$, normalized leach rate, in Tables A4-1 to A4-12. The data are plotted in Section III of the main report.

5.0 DISCUSSION

Error was introduced into the results via two avenues--counting error and procedural error. The possibility for significant error is raised because of the uneven leaching trends for cesium (in some experiments) and for the rare earths.

5.1 Counting Error

Statistical errors due to counting are discussed in detail in [BATES-1], but some indication of the error in the present experiments was obtained by the University of Missouri when they did repeated counts on a sample (L-243) of neutron-activated glass. The results are shown in Table A5-1.

Standard deviations range from 3 to 13% of the absolute activity, depending on the element being analyzed. Similar errors exist for the counting of leachates, and care must be taken when reporting results for γ -ray peaks containing a statistically insufficient number of counts. In the present experiments, larger errors existed for those elements present in small amounts in either the solid or the leachate, and these errors are so indicated in the tables. The nonuniform results for cesium leaching in some experiments are due to the small number of counts. More counts could be obtained by counting those samples in question for longer periods of time, but this was not possible for counts done at the University of Missouri.

5.2 Procedural Error

Procedural error evolved from the small amounts of the radioactivity handled, inhomogeneity of the glass, and problems with the neutron-activation process. Cross contamination of samples also was possible because much of the same equipment was used in successive experiments, (i.e., pH meter, drying lamp, tweezers, work area, etc.) However, a good check for cross contamination was obtained by comparing activation products and radiotracers. For example, there should be no ^{134}Cs in the leachates containing only radiotracers, and there should be no ^{137}Cs in leachates containing only activation products. Checks for such cross contamination were made and, in most cases none could be found. In cases where there was cross contamination, it was usually <5% of the total amount.

The removal of the radioactive samples from the leach container was another source of procedural error. The procedure was to remove the sample from the container with tweezers, being careful not to drop the sample or to remove parts of the leached layer through abrasion. If small pieces of the leached layer were included in the leachate, an increase in the rare earth and noble metal activity levels may be expected without a corresponding increase in the cesium, strontium, or barium levels. This increase would result because the leached layer would be depleted in the more leachable

Table A4-1. Normalized Elemental Mass Losses, $(NL)_i$, for DI Water Leachant and SRL 211** C Glass

	Specimen ID No.											14-d		42-d	
	L-154	L-154a	L-155	L-156	L-157	L-158	L-159	L-160	L-161	L-162	L-163	Ave.	S.D.	Ave.	S.D.
Length of run, d	3	3	5	7	14	14	14	28	42	42	42				
Final pH	9.2	9.1	9.7	9.7	9.7	9.7	9.6	9.6	9.5	9.5	9.5				
Init. pH - final pH	-2.8	-2.7	-3.3	-3.3	-3.3	-3.3	-3.2	-3.2	-3.1	-3.1	-3.1				
Leachant loss, %	1.0	0.2	0.2	0.5	0.5	-	-	1.0	1.7	1.4	1.2				
Spec. wt loss, g x 10 ⁻⁴	31	32	33	38	40	39	40	48	51	45	48				
Normalized Elemental Mass Loss, g/m ²															
Na		11.1	12.8	15.3	17.4	17.0	17.0	19.1	21.8	20.2	22.5	17.1	0.2	21.5	1.2
Li		11.2	13.1	15.7	18.0	17.5	17.4	21.0	23.4	21.3	22.3	17.6	0.3	22.3	1.1
Cs		11.4	11.5	13.7	16.0	16.1	16.1	17.6	20.0	18.1	18.8	16.1	0.1	19.0	1.0
Si		10.3	9.5	10.2	10.7	10.6	11.0	11.4	13.5	12.7	12.9	10.8	0.2	13.0	0.4
B		10.7	11.9	14.2	16.3	15.8	16.0	17.9	19.7	18.4	18.7	16.0	0.3	18.9	0.7
Mn		0.2	<0.2	<0.2	<0.2	0.3	0.2	0.3	0.2	0.2	0.3	0.2	0.1	0.2	0.1
Ca		7.0	7.5	7.3	7.5	7.5	7.7	7.6	7.2	7.6	7.5	7.6	0.1	7.4	0.2
Al		9.9	9.6	10.3	10.3	9.7	10.6	9.5	10.3	10.0	10.2	10.2	0.5	10.2	0.2
Fe		0.3	0.06	0.1	0.1	0.2	0.2	<0.06	0.1	0.1	0.2	0.2	0.1	0.1	0.1
Ba															
Wt loss	8.8	7.6	7.4	7.9	9.1	9.5	9.3	9.6	11.4	12.1	10.7	9.3	0.2	11.4	0.7

55

	Specimen ID No.												
	L-134	L-135	L-136	L-137	L-138	L-139	L-140	L-141	L-142				
Length of run, d	3.6	7	14.6	14.6	14.6	28	42.7	42.7	42.7				
Final pH	9.8	9.7	9.6	9.6	9.8	9.7	9.8	9.7	9.8				
Init. pH - final pH	-3.4	-3.4	-3.3	-3.3	-3.5	-3.4	-3.5	-3.4	-3.5				
Leachant loss, %	0	0.6	16.8	4.1	18.6	6.8	18.0	5.6	10.0				
Spec. wt loss, g x 10 ⁻⁴	30	38	43	48	40	55	60	57	62 ²				
Normalized Elemental Mass Loss, g/m ²													
										14-d		28-d	
										Ave.	S.D.	Ave.	S.D.
Si		15.3											
Wt loss	8.1	10.5	11.9	13.1	10.9	14.5	15.5	15.3	16.6	12.0	1.1	15.8	0.7

Table A4-3. Normalized Elemental Mass Losses, (NL)_i, for
DI Water Leachant and SRL 21i** S/A Glass

	Specimen ID No.												
	L-222	L-223	L-224	L-225	L-226	L-227	L-228	L-229	L-230	14-d		42-d	
										Ave.	S.D.	Ave.	S.D.
Length of run, d	3	7	14	14	14	28	42	42	42				
Final pH	9.5	9.7	9.4	9.4	9.4	9.3	9.7	9.5	9.7				
Init. pH - final pH	-3.5	-3.7	-3.4	-3.4	-3.4	-3.3	-3.7	-3.5	-3.7				
Leachant loss, %	1.7	3.8	6.5	6.7	6.6	12.9	18.4	18.8	18.1				
Spec. wt loss, g x 10 ⁻⁴	23	35	42	46	42	-	46	44	47				
Normalized Elemental Mass Loss, g/m ²													
51Cr	<0.85	4.4	3.7	2.8	5.2	3.5	8.1	5.9		3.9	1.2	7.0	1.6
88Y	-	-	-	-	-	-	-	-					
106Ru	-	-	-	-	-	-	-	-					
124Sb	7.8	9.7	12.1	15.0	13.9	14.1	16.5	15.8	8.4	13.7	1.5	13.6	4.5
133Ba	0.72	0.44	0.36	0.49	0.55	0.38	0.28	0.25	0.30	0.47	0.10	0.28	0.03
134Cs	11.4	16.0	18.7	19.3	18.8	21.0	22.0	21.0	20.4	18.9	0.3	21.1	0.8
137Cs	13.2	18.7	21.7	22.4	21.6	24.2	25.3	24.3	23.8	21.9	0.4	24.5	0.8
139Ce	-	-	-	-	-	-	-	-	-				
141Ce ^a	0.01	-	0.01	0.02	-	-	-	-	-				
152Eu ^a	0.06	-	-	0.05	-	0.07	-	0.02	-				
Si	-	-	-	-	-	-	14.9	14.9	14.7			14.8	0.1
Wt loss	5.9	8.9	10.7	11.2	10.7	-	11.7	12.4	11.8	10.9	0.3	11.7	0.8

^a(NL)_i values are based on data containing large errors.

Table A4-4. Normalized Elemental Mass Losses, (NL)_i, for
DI Water Leachant and SRL 211** S Glass

	Specimen ID No.												
	L-257	L-258	L-259	L-260	L-261	L-262	L-263	L-264	L-265	14-d		42-d	
										Ave.	S.D.	Ave.	S.D.
Length of run, d	3	7	14	14	14	28	42	42	42				
Final pH	9.5	9.5	9.4	9.4	9.4	9.4	9.6	9.6	9.4				
Init. pH - final pH	-3.5	-3.5	-3.4	-3.4	-3.4	-3.4	-3.6	-3.6	-3.4				
Leachant loss, %	2.0	3.5	6.5	6.8	6.2	12.0	19.1	18.9	18.2				
Spec. wt loss, g x 10 ⁻⁴	25	38	44	44	41	-	45	44	47				
Normalized Elemental Mass Loss, g/m ²													
88 _Y	-	-	-	-	-	-	-	-	-				
106 _{Ru}	-	-	-	-	-	-	-	-	-				
133 _{Ba}	0.92	0.48	0.43	0.44	0.48	0.34	0.29	0.38	0.36	0.45	0.03	0.28	0.02
137 _{Cs}	14.3	18.1	22.7	21.4	21.1	22.1	23.9	24.2	24.6	21.7	0.9	24.2	0.4
139 _{Ce}	-	-	-	-	-	-	-	-	0.02				
152 _{Eu}	<0.09	<0.09	0.06	<0.04	<0.10	2.04	<0.10	<0.10	0.10				
Si	-	-	-	-	-	-	15.2	15.1	16.0			15.4	0.5
Wt loss	6.7	9.7	11.8	11.2	10.3	-	11.3	11.1	12.0	11.1	0.8	11.5	0.5

Table A4-5. Normalized Elemental Mass Loss, $(NL)_i$, for DI Water Leachant and SRL 211*** C Glass

	Specimen ID No.								
	L-276	L-277	L-278	L-279	L-280	L-281	L-282		
Length of run, d	3	7	14	28	28	28	42		
Final pH	9.7	9.7	9.7	9.8	10.0	9.8	9.5		
Init. pH - final pH	-3.4	-3.4	-3.4	-3.5	-3.7	-3.5	-3.2		
Leachant loss, %	0.2	-	0.9	1.3	1.1	1.1	1.6		
Spec. wt loss, g x 10 ⁻⁴	33	36	43	47	49	48	51		
Normalized Elemental Mass Loss, g/m ²								28-d	
								Ave.	S.D.
Na	11.4	15.0	16.4	18.5	19.3	19.0	20.8	18.9	0.4
Li	12.8	16.1	17.6	21.1	22.0	21.2	23.6	21.4	0.5
Si	8.5	10.2	11.2	11.5	13.2	13.1	14.6	12.6	1.0
B	11.4	14.1	16.0	19.5	19.7	17.5	20.8	18.9	1.2
Mn	0.3	0.3	0.2	0.3	0.2	0.3	0.3	0.3	0.1
Ca	7.6	7.1	7.8	8.4	8.2	6.8	8.2	7.8	0.9
Al	9.1	10.1	9.6	10.4	10.2	11.2	8.8	10.6	0.5
Fe	0.2	0.2	0.2	0.2	0.1	0.2	0.2	0.2	0.1
Ba	2.6	2.0	2.0	1.7	<1.7	<1.7	<1.7	<1.7	
Wt loss	8.0	8.5	10.1	11.0	11.8	11.2	12.1	11.3	0.5

Table A4-6. Normalized Elemental Mass Loss, (NL)_i, for
DI Water Leachant and SRL 211*** A Glass

	Specimen ID No.								
	L-237	L-238	L-239	L-240	L-241	L-242	L-243	Ave. ^{b,c}	S.D.
Length of run, d	3	7	14	28	28	28	43		
Final pH	9.4	9.4	9.6	9.6	9.6	9.6	7.2		
Init. pH - final pH	-3.8	-3.8	-4.0	-4.0	-4.0	-4.0	-1.6		
Leachant loss, %	1.7	3.7	6.8	12.2	13.9	14.0	19.2		
Spec. wt loss, g x 10 ^{-4a}	77	36	58	53	49	85	112		
Normalized Elemental Mass Loss, g/m ²									
⁵⁹ Fe	1.2 E-1	9 E-2	6 E-2	1.2 E-1	2.2 E-1	7 E-2	1.8 E-2	1.4 E-1	8 E-2
⁸⁵ Sr	1.9	1.9	1.9	1.4	1.1	1.3	0.9	1.3	1 E-1
¹³¹ Ba	9 E-1	5 E-1	-	-	-	-	-	-	-
¹³⁴ Cs	15.4	18.5	21.7	22.1	20.9	21.9	20.6	21.6	6 E-1
¹⁴¹ Ce	4 E-2	4 E-2	3 E-2	4 E-2	1.5 E-1	4 E-2	8 E-2	4 E-2	-
¹⁴⁷ Nd	7 E-2	5 E-2	-	-	-	-	-	-	-
¹⁵² Eu	4 E-2	3 E-2	3 E-2	3 E-2	1.2 E-1	3 E-2	8 E-2	3 E-2	-
Si	-	-	-	-	13.6	13.6	-	13.6	-
Wt loss ^a	-	8.4	13.3	12.4	11.4	19.8	25.8	11.9	7 E-1

^aWt loss values may be affected due to chipping during irradiation.

^bValues for L-241 (Eu and Ce) were not used in averaging.

^cValue for L-242 wt loss was not used in averaging.

Table A4-7. Normalized Elemental Mass Loss, (NL)_i, for DI Water Leachant and SRL 211*** S Glass

	Specimen ID No.								
	L-287	L-288	L-289	L-290	L-291	L-292	L-293		
Length of run, d	3	7	14	28	28	28	42		
Final pH	9.6	9.8	9.5	9.5	9.0	9.4	9.5		
Init. pH - final pH	-3.7	-3.9	-3.6	-3.6	-3.1	-3.5	-3.6		
Leachant loss, %	1.5	3.7	7.1	13.6	12.7	13.5	20.9		
Spec. wt loss, g x 10 ⁻⁴	24	36	35	39	41	38	40		
Normalized Elemental Mass Loss, g/m ²							28-d		60
							Ave.	S.D.	
¹⁰⁶ Ru	-	-	-	1 E-2	8 E-2	-	2 E-2	5 E-2	5 E-2
¹³⁷ Cs	9.3	18.5	10.7	8.9	11.3	11.2	12.0	11.5	0.4
¹³⁹ Ce	-	-	-	1.9 E-1	1.1 E-1	-	-	-	-
¹⁴¹ Ce	1.7 E-3	3 E-3	-	1.1 E-1	1 E-1	-	2 E-2	6 E-2	6 E-2
¹⁵² Eu	1.8 E-3	4 E-3	3 E-3	9 E-2	8 E-2	5 E-3	2 E-2	4 E-2	4 E-2
Si	-	-	-	-	-	13.3	-	13.3	-
Wt loss	5.8	8.8	8.6	9.5	10.0	9.3	9.8	9.7	0.4

Table A4-8. Normalized Elemental Mass Loss, (NL)_i, for
DI Water Leachant and SRL 211*** S Glass (Repeat)

	Specimen ID No.								
	L-475	L-476	L-477	L-478	L-479	L-480	L-481		
Length of run, d	3	7	14	28	28	28	42		
Final pH	9.2	9.4	9.1	9.3	9.6	9.4	9.4		
Init. pH - final pH	-3.6	-3.8	-3.5	-3.7	-4.0	-3.8	-3.8		
Leachant loss, %	1.8	4.0	7.5	7.8	7.0	4.1	20.6		
Spec. wt loss, g x 10 ⁻⁴	24	35	43	43	40	49	39		
Normalized Elemental Mass Loss, g/m ²								28-d Ave. ^a	S.D.
¹³⁷ Cs	13.5	17.7	16.4	19.9	34.2	20.3	22.7	20.1	0.3
¹⁵² Eu	7 E-3	3 E-3	4 E-3	9 E-3	8 E-3	4 E-3	7 E-3	6 E-3	2 E-3
Wt loss	5.9	8.7	10.4	10.7	10.0	12.2	9.7	11.0	1.1

^aValue for L-479 (¹³⁷Cs) not used in averaging.

Table A4-9. Normalized Elemental Mass Loss, (NL)_i, for DI Water Leachant and SRL 131* C Glass

	Specimen ID No.								
	L-311	L-312	L-313	L-314	L-315	L-316	L-317		
Length of run, d	3	7	14	28	28	28	37		
Final pH	9.4	9.7	9.7	9.8	9.8	9.8	9.6		
Init. pH - final pH	-3.4	-3.6	-3.7	-3.8	-3.8	-3.8	-3.6		
Leachant loss, %	0	0.2	0.3	0.5	0.6	0.6	0.9		
Spec. wt loss, g x 10 ⁻⁴	29	39	55	67	69	65	74		
Normalized Elemental Mass Loss, g/m ²								28-d	
								Ave.	S.D.
Na	-	-	-	28.5	26.3	31.8	31.0	28.9	2.8
Li	-	-	-	29.3	27.7	31.6	32.0	29.5	2.0
Si	8.5	14.2	16.0	19.1	18.1	21.2	22.3	19.5	1.6
B	11.8	19.2	23.3	30.0	28.4	33.2	30.0	30.5	2.4
Mn	<0.2	0.3	0.3	0.3	0.3	0.4	0.4	0.3	0.1
Ca	3.8	1.4	0.7	<0.5	<0.5	<0.5	<0.5	<0.5	
Al	8.4	12.9	12.6	26.4	11.1	13.8	13.0	12.5 ^a	1.9
Fe	0.1	0.08	0.07	<0.06	<0.06	<0.06	<0.06	<0.06	
Ba	<1.7	<1.7	<1.7	<1.7	<1.7	<1.7	<1.7	<1.7	
Ti	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2	
Mg	-	-	-	-	-	0.7	0.6	0.7	0.1
Wt loss	7.0	11.8	12.8	16.2	15.6	17.9	18.0	16.6	1.2

^aValue for L-314 not used in averaging.

Table A4-10. Normalized Elemental Mass Loss, (NL)_i, for DI
Water Leachant and SRL 131* A Glass

	Specimen ID No.						
	L-327	L-328	L-329	L-330	L-331		
Length of run, d	3	7	14	28	28		
Final pH	9.4	9.5	9.6	9.8	9.7		
Init. pH - final pH	-3.2	-3.3	-3.4	-3.6	-3.5		
Leachant loss, %	2.7	3.8	8.7	12.8	13.0		
Spec. wt loss, ^a g x 10 ⁻⁴	21	15	24	39	49		
Normalized Elemental ^a Mass Loss, g/m ²						28-d Ave.	S.D.
⁵⁹ Fe	0.13	-	-	0.07	0.08	0.08	0.01
⁸⁵ Sr	1.6	0.4	0.5	-	-		
¹³⁴ Cs	17.9	8.2	7.9	22.0	23.4	22.7	1.0
¹⁴¹ Ce	0.04	0.008	0.007	0.01	0.01	0.01	0
¹⁵² Eu	0.06	0.10	0.005	0.01	0.01	0.01	0
Si	-	-	-	17.4	18.0	17.7	0.4
Wt loss	5.6	-	6.2	9.0	11.6	10.3	1.8

^a Considerable error may exist in all results due to irradiation problems, see
Section 5.1.

Table A4-11. Normalized Elemental Mass Loss, $(NL)_i$, for DI Water Leachant and SRL 131* S Glass

	Specimen ID No.								
	L-339	L-340	L-341	L-342	L-343	L-344	L-345		
Length of run, d	3	7	14	28	28	28	42		
Final pH	9.5	9.3	9.4	9.2	9.0	9.3	9.2		
Init. pH - final pH	-3.7	-3.5	-3.6	-3.4	-3.2	-3.5	-3.4		
Leachant loss, %	1.0	4.2	7.8	11.5	13.9	14.2	20.3		
Spec. wt loss, g x 10 ⁻⁴	17	22	54	56	50	58	51		
Normalized Elemental Mass Loss, g/m ²								28-d Ave.	S.D.
⁸⁵ Sr	4.1	1.5	1.8	1.4	2.5	1.3	1.1	1.7	0.7
¹³³ Ba	0.8	0.2	0.5	0.3	0.4	0.6	0.4	0.4	0.1
¹³⁷ Cs ^a	4.2	7.4	9.9	13.9	9.0	9.7	11.6	10.9	2.7
¹⁴¹ Ce	4 E-3 ^a	2 E-2	5.2 E-1	6 E-2	1.8 E-1	5.4 E-1	1.2 E-1	0.3	0.3
¹⁵² Eu	1 E-2	2 E-2	4.3 E-1	5 E-2	1.6 E-1	4.6 E-1	1.1 E-1	0.2	0.2
Si	-	-	-	17.7	14.9	19.3	-	17.3	2.2
Wt loss	3.9	7.5	12.7	13.1	11.7	13.5	11.9	12.8	0.9

^aConsiderable error exists due to the small amount of ¹³⁷Cs in the solid and the count times used.

Table A4-12. Normalized Elemental Mass Loss, (NL)_i, for DI Water Leachant and SRL 131* S Glass (Repeat)

	Specimen ID No.								
	L-468	L-469	L-470	L-471	L-472	L-473	L-474		
Length of run, d	3	7	14	28	28	28	56		
Final pH	9.2	8.8	9.6	9.0	9.0	5.3	ND ^a		
Init. pH - final pH	-3.6	-3.2	-4.0	-3.4	-3.4	+0.3	ND		
Leachant loss, %	1.5	3.5	6.4	12.5	12.5	2.7	ND		
Spec. wt loss, g x 10 ⁻⁴	17	32	38	51	50	51	ND		
Normalized Elemental Mass Loss, g/m ²								28-d	
								Ave.	S.D.
⁸⁵ Sr	3.9	1.9	1.6	1.0	1.2	-	-	1.1	0.1
¹³³ Ba	0.8	0.3	0.2	0.1	0.1	-	8 E-2	0.1	0
¹³⁷ Cs	9.0	15.5	21.6	18.7	21.0	-	22.5	19.9	1.5
¹⁵² Eu	2 E-3	6 E-3	3 E-3	9 E-3	1 E-2	-	-	1 E-2	7 E-4
Wt loss	4.6	8.6	10.2	13.8	13.5	8.3	-	13.7	0.2

^aNot determined.

Table A5-1. Observed Activities in Neutron-Activated Glass Sample L-243, 10 Observations

Nuclide	Mean Activity, μCi	Std. Deviation
^{152}Eu	72.4	2.02
^{60}Co	0.21	0.04
^{59}Fe	5.08	0.21
^{134}Cs	0.10	0.01
^{58}Co	1.35	0.076
^{103}Ru	2.32	0.12
^{95}Zr	3.53	0.18
^{95}Nb	2.60	0.33

elements (Cs, Sr, Ba) and enriched in the less-leachable elements (rare earths and Ru). Such handling problems may have occurred for the first sets of leachates. For this reason, some of the spiked experiments were repeated using more careful handling procedures, and more consistent results were achieved.

Large deviations in rare earth and noble metal leach rates would also occur if small pockets of these elements existed in the glass. If these pockets of activity were spread evenly throughout the glass, a gross counting of each sample would not detect the inhomogeneity. There is no experimental evidence that such inhomogeneity exists, but it would explain the varied rare earth leach results.

Problems also existed in the neutron-activation process. Activations were done by both the University of Illinois and the University of Missouri. Care must be taken in activating the samples so that no chipping or heating will occur. Significant problems arose with the SRL 131* batch of activated glass because, due to overheating, the surface of the glass actually softened. This caused a decrease in the surface area of the as-cut samples and caused the Al foil, which surrounds the sample during irradiation, to become bonded to the solid. In many cases, it was difficult to remove all of the Al foil from the glass before leaching. These problems caused the SRL 131* A leach tests to be reduced in number and produced an uncertainty in the results that is difficult to measure.