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A LEACHING STUDY OF NUCLEAR MELT GLASS: PART I

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ABSTRACT

Ground samples of three nuclear melt glasses from underground nuclear explosions at the Nevada Test Site (NTS) were leached at 25°C with natural ground water from NTS. Using our dynamic single-pass flow-through leaching system we monitored the release of radionuclides from the glasses during 420 days of leaching. We continually flowed the ground water over the melt glass at flow rates of 185 ml/day for half of the samples and 34 ml/day for the rest. Leachate solutions were collected continuously, and composite samples, collected on days 1, 2, 3, 6, 11, 32, 38, 70, 120, 230 and 420, were analyzed using low-background Ge(Li) gamma spectrometers.

For most of the radionuclides the leach rate decreased smoothly throughout the experiment. Except for ^{95}Zr , ^{144}Ce , and ^{155}Eu , there was no difference between the fast (185 ml/day) and slow (34 ml/day) flow-rate leach rates. The measurable leach rates ranged from a high of 1×10^{-2} g-glass/m² day for ^{22}Na (slow flow-rate, day 1 in glass #2) to a low of 1×10^{-6} g-glass/m² day for ^{54}Mn (slow flow-rate, day 420 in glass #2). Most of the leach-rate values were approximately 5×10^{-4} g-glass/m² day initially, decreasing to 5×10^{-6} g-glass/m² day after 420 days of leaching. The leach rates had not leveled off by the end of the experiment and were, in general, continuing to decrease.

From the detected activities in the leachate solutions, we determined the percent of the pre-leach activity leached from the melt glasses during

the experiment. Only ^{124}Sb , in one fast flow-rate channel of glass #2, exceeded 3% of the initial activity leached. The majority of the samples released less than 1% of the pre-leach activity for a given radionuclide. The percent activity released from the samples leached at the fast and slow flow rates were nearly equal.

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INTRODUCTION

The first underground nuclear explosion at the Nevada Test Site was detonated in 1957, followed by continued underground testing of nuclear devices. The underground tests deposit radionuclides in the detonation cavity and chimney formed by the cavity collapse. The chemical and physical distribution of the radioactive products is complex and dependent on the composition of the surrounding rock and characteristics of the nuclear device (Borg, 1975; Dupuis, 1970). Borg estimates that within a minute after the explosion greater than 75% of the fission activity is associated with the molten rock (called the melt or puddle). This percentage decreases as volatile radionuclides, those with gaseous precursors, and those with high vapor pressures escape from the melt. The escaped radionuclides condense on the chimney rubble.

The melt vitrifies as it cools. The refractory elements generally remain in the resulting melt glass. One year after the detonation over half the residual activity usually remains associated with the melt glass (Borg, 1975). Our experiment studied the leaching of this radioactive melt glass by natural ground water. Chimney rubble was not included in this experiment primarily because of the expense involved in obtaining representative, homogeneous samples.

There is a great deal of interest in the leaching of deeply-buried radioactive glass by ground water due to the potential for ground-water contamination. An ongoing field test at NTS monitors the level of radio-

nuclides in ground water from a well 90 meters from the chimney of the Cambrian event (Hoffman et al., 1977, Coles and Ramspott, 1982). The leaching of radioactive melt glass buried in a nuclear explosion cavity is analogous to leaching vitrified high-level nuclear waste buried in an underground repository. In the present plan for nuclear waste disposal, the radioactive material, immobilized in a vitrified waste form, will be placed into canisters and buried in deep underground repositories. If ground water could leach radionuclides from the vitrified waste, radioactivity might leave the repository and enter the biosphere. Hence, this study relates directly to the geologic disposal of vitrified nuclear waste.

Borg et al. (1976) reviewed the existing leach tests on nuclear melt glass. Most of these tests used the static or batch procedure where the sample remains in contact with the leachant for a set period of time. The leachant is then removed for analysis and may be replaced with fresh leachant for another leaching period.

Our leach study was a dynamic test, continually flowing fresh leachant over the melt glass for 420 days. In the dynamic system, a steady-state solution composition is not achieved, and the deviation from steady-state can vary with the flow-rate. This single-pass flow-through leaching system has been used in a number of other studies. Coles et al. (1982) used it to study the leaching of PNL 76-68 borosilicate glass, a potential matrix for encapsulating nuclear waste. A second, more extensive study, examined the leaching characteristics of mock reactor fuel pellets (UO_2), as well as PNL 76-68 glass (Bazan et al., 1982).

We also completed a second dynamic leaching study of nuclear melt glass in 1982 (Rego, 1983).

In the melt glass leaching study discussed in this report, replicate samples of melt glass from three nuclear explosions were leached at 25°C for 420 days. The leachant was ground water from Well-5B at NTS. The flow-rates during the leaching were 34 or 185 ml/day.

The interim report of Coles (Coles et al., 1979) covers the details of the melt glass characterization and results from the first 120 days of leaching for melt glass #3. This final report includes data for all three glasses for the full 420 day leaching period and the post-leach results.

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METHODS

We chose melt glass from three nuclear tests which had been detonated 0.5 to 1.5 years prior to the start of the leaching and had high fission-fusion ratios. This resulted in samples containing a variety of radionuclides at high levels of activity. The pieces of melt glass were handpicked from material cored from each explosion cavity. This glass material was then carefully cleaned, ground, sieved, mixed, and analyzed to ensure it contained primarily vitreous material with a homogeneous size and radioactivity distribution. Coles et al. (1979) gives a detailed characterization of the glasses and describes the sample preparation. The melt glasses' surface areas were measured by nitrogen adsorption (BET). Particle sizing was performed by sieve analysis. The glass samples used in the leaching cells were the (-100 +325) mesh fraction.

Weed and Jackson (1979) designed the single-pass flow-through leaching apparatus as an alternative to static and multi-pass leach methods. The system design is described in detail in Weed and Jackson (1979). A block diagram of this leaching system is shown in Fig. 1. The experimental matrix and numbering of the cells is shown in Fig. 2. Triplicates of each combination of glass and flow-rate were included in the experiment. For melt glass #2 an additional cell was run at each flow-rate with the direction of the flow reversed (cells #7 and #17). This was done to see if the direction of flow affected the results; it did not.

Fig. 1. Single-Pass Flow-Through Leaching System
Functional Diagram

Fig. 2: Experimental Matrix

The leaching cell which contained the ground melt glass samples is shown in Fig. 3. Prew weighed samples ($0.58 \pm .11\text{g}$) were loaded into each cell by plugging the inlet opening, pulling a vacuum on the exit opening, and pouring the ground melt glass through the funnel into the side filler opening. This procedure greatly reduced the chances of spilling the sample or spreading any radioactive dust.

Fig. 3: Single Pass Continuous Flow Leaching Cell

Approximately 200 liters of NTS Well - 5B ground water were shipped to LLNL every two months to supply the necessary leachant solution. The pH was measured one week after each delivery. The elemental composition was determined by inductively coupled plasma-optical emission spectrometry (ICP-OES). (See Table 2.) The leachant was held in a reservoir and gravity fed through a filter, the supply manifolds, and another filter. At this point, the leachant flow-rate was metered by peristaltic pumps. For ten cells the flow-rate was 185 ml/day $\pm$ 10% and for the remaining ten cells the rate was 34 ml/day $\pm$ 5%. The leachant flowed into the cells via connecting lines of small-bore polyethylene tubing and Luer connectors. The leaching cells were immersed in constant temperature baths to maintain them at $25 \pm 2^\circ\text{C}$ throughout the experiment. Experience from other leaching experiments has shown that dry constant temperature units (ovens or refrigerators) reduce the equipment maintenance and cell corrosion and leakage problems.

A side effect of using natural ground water as the leachant was algal growth in the system. The algae was determined to be chaetomorpha (Spies, 1978). This resulted in clogging of the leachant filters and the tubing into and out of the cells which caused variations in the flow-rates at times. More importantly, it affected the composition of the leachant. From tests made following the second melt glass leaching study (Rego, 1983), the amount of dissolved organic carbon (DOC) in the leachates and water used to flush the system was significant (5-9 ppm DOC). The possible effects of the algae on the leaching results are considered in the next section. The algae growth can be controlled by keeping the leachant solutions in the dark.

The leaching began on November 11, 1977. The highest leach-rates and greatest change in leach-rate were expected in the first few days of leaching. For this reason the leachate was collected for each twenty-four hour period during the first three days of the experiment. Thereafter the leachates were collected and combined throughout the sampling interval (e.g., the day 6 sample was a composite of the leachate from days 4, 5 and 6).

The leachates were collected on days 1, 2, 3, 6, 11, 32, 38, 70, 120, 230 and 420. This sampling sequence maintains approximately equal separation of data points on a logarithmic time scale. The day 32 sample was planned for day 20, but a scheduling error occurred.

To accommodate the large volumes of leachate from the long collection periods the collection bottles were periodically emptied into one liter bottles lined with a 5 x 10 x 30 cm polyethylene bag. We then dried the sample at 85°C leaving only the residual salts in the bag. This procedure was repeated as often as necessary to handle all the sample. For sampling intervals greater than 100 days the initial drying was done in beakers and the wet salts were transferred to the polyethylene bags.

The bags were rolled, keeping the salts in the center, placed in a dye, and compressed into pellets. The pellets were gamma-counted for 24 hours on a low background Compton-suppressed Ge(Li) spectrometer. Repetitive tests with ^{137}Cs -spiked ground water indicated this procedure resulted in less than 5% uncertainty in the activity determination.

In Appendices A and B we indicate when the gamma-counts reported no detected activity for a certain radionuclide in the replicate leachate samples. There are several explanations for a "not detected" result. The first, most obvious one, is that the radionuclide was not present in the leachate or was present but below our detection limit. Gamma-ray [Ge(Li)] spectroscopy detection limits are nontrivial functions of count duration, sample size, and mostly the concentration of the other radionuclides present. For this reason we do not list a specific detection limit for each radionuclide.

Another explanation is the short half-life of some of the radionuclides, causing them to decay beyond detectability by the time the leachate was collected, processed and counted. When the extent of the decay results in "not detected" values, we can expect the radionuclide will not be detected in leachates collected late in the experiment. The measured activities of all the detected radionuclides were decay corrected to the start of the leaching experiment, however, this can not account for activity that has decayed beyond detection. Interference from other radionuclides, Compton scattering, background, and other gamma-counting anomalies are probably responsible for the remainder of the "not detected" values. The spectral analysis program (GAMANAL, Gunnink and Niday, 1972) used to interpret the gamma-ray spectra is well-known for its reliability. However, no analysis or program is infallible, and so the output was checked for obvious omissions and errors. Any of the possible explanations for a radionuclide being "not detected" should be considered when examining the data.

We calculated the leach-rate (Eq. 1), activity loss (Eq. 2) and cumulative leach (Eq. 3) values as follows:

$$\text{leach-rate} = R = (Q/Q_0)(1/\Delta t)(1/S) \quad \text{Eq. 1}$$

(units: g-glass/m² day)

where:

R = fraction leached per day per m² surface area

Q = total activity leached during Δt

Q₀ = pre-leach activity in melt glass sample

Δt = interval of leaching, days

S = specific glass surface area, m²/g-glass

$$\text{activity loss} = \left(\frac{Q_0 - Q_f}{Q_0} \right) 100 \quad \text{Eq. 2}$$

(units: % of pre-leach activity)

where:

Q_f = post-leach activity

$$\text{cumulative leach} = \sum_i Q_i \left(\frac{1}{Q_0} \right) 100 \quad \text{Eq. 3}$$

(units: % of pre-leach activity)

where:

Q_i = activity in the leachate collected during interval i

The leach-rate values are listed in Appendix B and the activity loss and cumulative leach values in Tables 1a-c.

We analyzed melt glass #2 and six leachate samples for ^{239}Pu using the following procedure (Reed et al., 1981). After the "leachate pellets" and melt glass samples were dissolved, the plutonium was converted to Pu(IV) using NaNO_2 . The plutonium was separated on a 1-X8 anion exchange column and eluted with freshly prepared $\text{NH}_4\text{I-HCl}$ (1.5g $\text{NH}_4\text{I}/100\text{ml}$ HCl). After treating the elutant with H_2O_2 , it was further purified on a Dowex anion exchange column and eluted with the $\text{NH}_4\text{I-HCl}$ solution. The plutonium was then electroplated onto a stainless steel planchet (Mitchell, 1960). The $[^{239}, ^{240}]\text{Pu}$ activity was measured using Si(Li) alpha detectors which have a detection limit of approximately 9×10^{-3} dpm.

RESULTS

The leaching experiment began 0.5 to 1.5 years after the nuclear detonations. Glass #3 came from the most recent of the three tests (1977). Glass #1 was from the oldest test (late 1975). The difference in the decay time can explain some of the variation in the pre-leach activities (Table 1a-c). Differences in the type of device detonated can explain other variations, in the ^{22}Na and ^{137}Cs activities. Triplicate samples were counted to determine the pre-leach activities of the melt glasses. The uncertainties (%) reported are the larger of the standard deviation about the mean value or the compounded counting uncertainties. Of the radionuclides observed in the melt glasses, we did not detect ^{59}Fe or ^{65}Zn in the leachates. Ruthenium-103, ^{134}Cs , ^{141}Ce , ^{168}Tm and ^{182}Ta were observed only sporadically in the leachates. Short half-lives and low initial activities explain the absence of ^{59}Fe , ^{65}Zn , ^{103}Ru , ^{134}Cs , and ^{141}Ce in the leachates. The ^{168}Tm and ^{182}Ta appear to be retained in the solid melt glass.

The post-leach activities (Table 1a-c) were determined for a single sample of each glass. Due to a change in project priorities and personnel, the gamma-count was made years after the end of the experiment. At count time, the activity for many of the radionuclides was at or below our level of detection. This explains the large uncertainties and the "not detected" results for many of the radionuclides. We decay corrected the measured activities to the same zero time as the earlier gamma-counts, the start of the leaching experiment. The percent activity loss was calculated from Eq. 2. Due to the uncertainties in the data, these values only give an assurance that the amount of activity leached from the

glasses was small. Except for a few cases (i.e., Na-22 glass # 1), the percent loss was about 10%. The very large uncertainties arise from calculating small differences in large numbers which also have large uncertainties. If the uncertainty in the percent activity loss was greater than 150%, an upper limit was calculated and reported.

The cumulative leach values (Eq. 3) provide a better measure of the percent of the pre-leached activity removed from the melt glasses than these activity loss values. The percent cumulative leach values in Tables 1a-c are reported as median and range (with the uncertainty) instead of the average and standard deviation because of the wide variation in some of the replicates. Anomalously high or "not detected" activities obviously cause errors in the cumulative leach values. Attempts were made to account for this by estimating activities from replicate samples or by smoothing out the leach-rate curve. "Less than" values for the cumulative leach were calculated by extrapolation when several "not detected" values were reported. The uncertainties were based on the counting uncertainties only.

By comparing the fast and slow flow-rate cumulative leach data (Tables 1a-1c) we can determine the effect of flow-rate on the leaching. The samples leached at the fast flow-rate were exposed to over five times more ground water than the slow flow-rate samples. But we find the cumulative leach values for the fast flow-rate samples were nearly equivalent to the values for the associated slow flow-rate samples. The

Table 1a: Activity and Cumulative Leach Values, $\pm 1 \sigma$ (%)
Melt Glass #1

Radio-nuclide	Pre-Leach Activity ^a dpm/g	Post-Leach Activity ^b dpm/g	Activity loss ^c %	Cumulative Leach ^d median(%) range	
				FAST	SLOW
²² Na	3.45×10^2 ± 15	2.20×10^2 ± 40	35 ± 83	ND ^e	ND
⁵⁴ Mn	1.06×10^5 ± 0.4	9.14×10^4 ± 3.9	14 ± 24	.13 .12-.22 ± 4.2	.13 .083-.17 ± 12
⁵⁷ Co	9.79×10^2 ± 9.0	ND	--	SD ^e	SD
⁵⁸ Co	ND	ND	--	ND	ND
⁶⁰ Co	2.61×10^4 ± 0.7	2.3×10^4 ± 3.7	10 ± 33	.099 .0079-.18 $\pm 5.$	.099 .042-.12 $\pm 5.$
⁶⁵ Zn	1.32×10^3 ± 17	ND	--	ND	ND
⁹⁵ Zr	4.03×10^4 ± 0.5	ND	--	SD	SD
¹⁰³ Ru	ND	ND	--	ND	ND
¹⁰⁶ Ru	4.28×10^5 ± 2.6	3.92×10^5 ± 3.4	8 ± 48	SD	SD
¹²⁴ Sb	ND	ND	--	ND	ND
¹²⁵ Sb	7.35×10^4 ± 0.7	6.65×10^4 ± 3.2	10 ± 31	.095 .078-.16 ± 25	.085 .061-.12 ± 12
¹³⁴ Cs	5.30×10^4 ± 2.6	ND	ND	ND	ND
¹³⁷ Cs	5.67×10^4 ± 0.7	5.10×10^4 ± 3.3	10 ± 30	.28 .16-.31 ± 2.6	.26 .24-.26 3.4

Table 1a, cont.

Radio-nuclide	Pre-Leach Activity ^a dpm/g	Post-Leach Activity ^b dpm/g	Activity loss ^c %	Cumulative Leach ^d median(%) range	
				FAST	SLOW
¹⁴¹ Ce	ND	ND		ND	ND
¹⁴⁴ Ce	2.93 × 10 ⁶ +0.6	2.56 × 10 ⁶ +3.3	13 +23	.089 .081-.13 +2.0	.055 .0031-.11 +25
¹⁵⁵ Eu	1.17 × 10 ⁵ +2.1	1.04 × 10 ⁵ +4.0	11 +37	.14 .11-.19 +7.1	.10 .077-.15 +4.3
¹⁶⁸ Tm	ND	ND		ND	ND
¹⁸² Ta	1.10 × 10 ³ +18	ND		SD	SD

Notes:

- Average activity of three replicate samples; uncertainties are the larger of one standard deviation or the compounded counting uncertainties in percent.
- Activity of one sample; uncertainties (%) are the counting uncertainties.
- Difference in pre-leach and post-leach activities normalized to the pre-leach value.
- Sum of activities measured in leachates (see text); uncertainties (%) are the counting uncertainties.
- ND = not detected; SD = sporadically detected.

Table 1b: Activity and Cumulative Leach Values, $\pm 1 \sigma$ (%)
Melt Glass # 2

Radio-nuclide	Pre-Leach Activity ^a dpm/g	Post-Leach Activity ^b dpm/g	Activity loss ^c %	Cumulative Leach ^d median(%) range	
				FAST	SLOW
²² Na	9.22×10^3 ± 9.9	9.11×10^3 ± 4.9	$<14.5^f$	1.7 1.7-1.7 ± 12	1.6 1.3-1.6 ± 11
⁵⁴ Mn	5.16×10^6 ± 0.4	4.93×10^6 ± 3.7	4 ± 80	.24 .19-.24 $\pm .62$	.13 .13-.13 $\pm .67$
⁵⁷ Co	7.18×10^5 ± 0.7	6.82×10^5 ± 3.9	5 ± 100	1.3 1.3-1.4 $\pm .94$	.84 .81-.99 $\pm .88$
⁵⁸ Co	1.76×10^5 ± 1.2	ND ^e	--	1.1 1.1-1.2 ± 2.3	.65 .54-.82 ± 4.8
⁶⁰ Co	1.98×10^5 ± 0.5	1.85×10^5 ± 3.7	7 ± 53	1.7 1.1-1.2 $\pm .95$	.65 .68-.79 $\pm .99$
⁶⁵ Zn	8.39×10^3 ± 26	ND	--	ND	ND
⁹⁵ Zr	5.36×10^5 ± 0.5	ND	--	.18 1.1-.20 ± 29	SD
¹⁰³ Ru	4.06×10^4 ± 3.6	ND	--	SD ^e	SD
¹⁰⁶ Ru	1.19×10^6 ± 4.2	1.12×10^6 ± 5.0	6 ± 100	.53 .40-.62 ± 4.6	.38 .36-.43 ± 4.9
¹²⁴ Sb	8.96×10^3 ± 5.5	ND	--	<8.5	<3.5
¹²⁵ Sb	5.04×10^4 ± 5.3	4.58×10^4 ± 4.0	9 ± 70	1.3 1.3-1.4 ± 11	1.0 .81-1.0 ± 6.2
¹³⁴ Cs	4.32×10^3 ± 20	ND	--	SD	SD

Table 1b, cont.

Radio-nuclide	Pre-Leach Activity ^a dpm/g	Post-Leach Activity ^b dpm/g	Activity loss ^c %	Cumulative Leach ^d median(%) range	
				FAST	SLOW
¹³⁷ Cs	1.70×10^4 <u>+5.3</u>	1.61×10^4 <u>+3.8</u>	5 <u>+120</u>	<1.6	<2.2
¹⁴¹ Ce	9.16×10^3 <u>+10</u>	ND	--	SD	SD
¹⁴⁴ Ce	2.70×10^6 <u>+9.7</u>	2.60×10^6 <u>+3.9</u>	<15.6 ^f	.20 .14-.22 <u>+9.8</u>	<.08
¹⁵⁵ Eu	5.60×10^4 <u>+4.1</u>	5.40×10^4 <u>+3.9</u>	4 <u>+150</u>	<.77	<.62
¹⁶⁸ Tm	8.76×10^4 <u>+3.0</u>	ND	--	SD	SD
¹⁸² Ta	5.29×10^5 <u>+0.6</u>	ND	--	SD	SD

Notes:

- Average activity of three replicate samples; uncertainties are the larger of one standard deviation or the compounded counting uncertainties in percent.
- Activity of one sample; uncertainties (%) are the counting uncertainties.
- Difference in pre-leach and post-leach activities normalized to the pre-leach value.
- Sum of activities measured in leachates (see text); uncertainties (%) are the counting uncertainties.
- ND = not detected; SD = sporadically detected.
- The upper limit is reported for activity loss values with greater than 150% uncertainty.

Table 1c: Activity and Cumulative Leach Values, $\pm 1 \sigma$ (%)
Melt Glass #3

Radio-nuclide	Pre-Leach Activity ^a dpm/g	Post-Leach Activity ^b dpm/g	Activity loss ^c %	Cumulative Leach ^d median(%) range	
				FAST	SLOW
²² Na	1.96 x 10 ⁴ +4.3	1.70 x 10 ⁴ +8.0	13 +62	2.0 1.9-2.1 +5.0	1.9 1.8-2.0 +5.2
⁵⁴ Mn	8.63 x 10 ⁶ +0.4	8.45 x 10 ⁶ +3.2	2. +150	.39 .39-.46 +0.5	.20 .19-.21 +6.3
⁵⁷ Co	1.32 x 10 ⁶ +6.3	1.22 x 10 ⁶ +4.7	8 +100	.39 .34-.42 +6.3	.24 .21-.26 +6.3
⁵⁸ Co	4.70 x 10 ⁵ +0.6	ND ^e	--	.30 .27-.30 +6.0	.18 .18-.25 +13.
⁶⁰ Co	7.66 x 10 ⁵ +1.0	7.43 x 10 ⁵ +3.1	3 +125	.39 .30-.38 +1.3	.18 .18-.24 +1.2
⁶⁵ Zn	1.59 x 10 ⁴ +25.	ND	--	ND	ND
⁹⁵ Zr	8.34 x 10 ⁵ +0.4	ND	--	.15 .12-.17 +10.	<.15
¹⁰³ Ru	5.79 x 10 ⁴ +0.5	ND	--	SD ^e	SD
¹⁰⁶ Ru	6.95 x 10 ⁵ +18.	6.73 x 10 ⁵ +9.7	<26 ^f	.47 .47-.56 +20.	.32 .28-.43 +20.
¹²⁴ Sb	4.93 x 10 ⁴ +3.7	ND	--	<2.4	<2.7
¹²⁵ Sb	5.21 x 10 ⁴ +7.8	4.84 x 10 ⁴ +3.7	7 +120	.47 .40-.51 +15.	.29 .27-.33 +23.
¹³⁴ Cs	2.09 x 10 ⁴ +8.9	ND	--	SD	SD

Table 1c, cont.

Radio-nuclide	Pre-Leach Activity ^a dpm/g	Post-Leach Activity ^b dpm/g	Activity loss ^c %	Cumulative Leach ^d median(%) range	
				FAST	SLOW
¹³⁷ Cs	1.56 x 10 ⁴ +7.7	1.59 x 10 ⁴ +3.5	<9 ^f	<.95	<.82
¹⁴¹ Ce	2.65 x 10 ⁴ +4.7	ND	--	SD	SD
¹⁴⁴ Ce	2.89 x 10 ⁶ +1.0	2.70 x 10 ⁶ +3.4	6 +50	.30 .29-.37 +1.7	.090 .086-.10 2.1
¹⁵⁵ Eu	3.34 x 10 ⁴ +13.	3.05 x 10 ⁴ +5.8	<24 ^f	<.73	<.24
¹⁶⁸ Tm	3.52 x 10 ⁺⁴ +1.7	ND	--	SD	SD
¹⁸² Ta	2.98 x 10 ⁴ +9.8	ND	--	SD	SD

Notes:

- Average activity of three replicate samples; uncertainties are the larger of one standard deviation or the compounded counting uncertainties in percent.
- Activity of one sample; uncertainties (%) are the counting uncertainties.
- Difference in pre-leach and post-leach activities normalized to the pre-leach value.
- Sum of activities measured in leachates (see text); uncertainties (%) are the counting uncertainties.
- ND = not detected; SD = sporadically detected.
- The upper limit is reported for activity loss values with greater than 150% uncertainty.

ratio of the median cumulative leach values at different flow-rates, within the uncertainties, was <2 for nearly every radionuclide (the exceptions being ^{144}Ce in glass #2 and #3 and ^{95}Zr in glass #2). This ratio never approached the factor of five difference in the ground water volume. Therefore, we conclude from these results that the cumulative leach for most of the radionuclides in these melt glasses is independent of flow-rate. This agrees with other leaching studies (Coles et al., 1982) where flow-rate effects are seen only at elevated temperatures (75°C).

The actual percent cumulative leach is very low, with generally $<1.5\%$ of the pre-leach activity detected in the leachates (Tables 1a-c). For ^{22}Na , in glasses #2 and #3, 1.3 to 2.1% of the pre-leach activity was detected in the leachates. For a given melt glass the three cobalt isotopes (^{57}Co , ^{58}Co , and ^{60}Co) each have the same cumulative leach values (Tables 1b and 1c) and leach-rates (Fig. A-8 - A-9); however, the antimony isotopes (^{124}Sb and ^{125}Sb) behave differently (Fig. A-11 - A-12). This is explained by comparing the method of formation of the radionuclides. The three cobalt isotopes are formed from the same precursor, Ni, but the antimony isotopes have different precursors. This is discussed in greater detail later in this section.

The ^{239}Pu cumulative leach from glass #2 is apparently higher than would be expected from other studies (Wolfsberg, 1978 and Coles et al., 1982). To obtain our cumulative leach value, an estimated ^{239}Pu activity

(for the leachate samples not analyzed for ^{239}Pu) was determined by linearly interpolating between the experimental leach-rates and then calculating the activity. We estimate 0.14% of the original ^{239}Pu was leached from glass #2. In Wolfsberg's (1978) batch leaching study of nuclear test debris the percent of initial ^{239}Pu activity leached after 277 days was $<10^{-2}$. In a doped borosilicate glass bead dynamic leaching study (Coles et al., 1982), at comparable temperature and flow-rates, the cumulative percentage leach was 2×10^{-2} over 420 days.

Our higher ^{239}Pu cumulative leach value may be due to the algal growth, mentioned in the Experimental section, which elevated the dissolved organic carbon (DOC) levels in the leachant. The distribution coefficient, K_d , for Pu shows a marked decrease when DOC levels increase. In the study by Nelson et al., (1981) the K_d dropped by almost an order of magnitude in most cases when the DOC increased from 1 to 10 ppm. The decrease in K_d is attributed to the increase in plutonium solubility with increased DOC. If the leach rate of ^{239}Pu from the melt glass is controlled by plutonium solubility, then elevated DOC levels will result in higher ^{239}Pu leach rates. Insufficient data was collected in our experiment to determine the exact effect of the algal growth on our Pu leaching.

The ground water from Well 5-B at NTS was analyzed to determine the elemental composition by ICP-OES at LLNL. The anion values are from determinations by the United States Geological Survey (USGS, 1973). The radionuclides were determined by gamma-counting at LLNL. The initial composition is typical of NTS ground water (Table 2). No elemental analysis was made of the leachate solutions.

Table 2: Composition of Well-5B Water, NTS

Element	($\mu\text{g}/\text{ml}$)	Element	($\mu\text{g}/\text{ml}$)	Element	($\mu\text{g}/\text{ml}$)
Al	<0.004	K	12.	Pb	0.032
As	<0.05	Li	0.043	Si	28.
B	0.023	Mg	2.5	Sr	0.030
Ca	7.9	Mn	0.007	Ti	<0.005
Cd	0.004	Mo	0.015	V	0.011
Co	0.004	Na	97.	Zn	<0.001
Cu	<0.001	Ni	<0.004	Zr	<0.005
Fe	0.13	P	<0.07		
CO_3^{-2}	5	HCO_3^-	174	F^-	0.8
Cl^-	22	SO_4^{-2}	56	NO_3^-	11
PO_4^{-3}	<0.01				
^{106}Ru	<0.1dpm/l	^{125}Sb	<0.01dpm/l	^{137}Cs	<0.04dpm/l

Elemental composition determined by spectrochemical analysis, LLNL, all uncertainties <10%.

Anion composition determined by USGS (USGS, 1973).

Radionuclide composition determined by γ -counting, LLNL.

The pH of the NTS Well 5-B ground water was very consistent, 8.13 ± 0.01 . After flowing over the melt glass, the pH increased. the pH was 8.40 ± 0.06 for the fast flow-rate leachates and 8.65 ± 0.03 for the slow flow-rate leachates. Apparently the longer interaction time for the slow flow-rate solutions resulted in a greater alteration of the ground water composition.

The leach-rate (Eq. 1) is a measure of the loss of a radionuclide from the melt glass. It is dependent on the leaching time and the surface area exposed to the leachant. As displayed in Fig. 2, we ran triplicate samples of each glass at each flow-rate. The leach-rates reported in Tables B-1 - B-13 are averages of the replicate values (see Notes, Appendix B).

The surface area is used as a measure of the amount of melt glass in contact with ground water. The leach-rate will vary with the available surface area of the sample. The leach-rate expression (Eq. 1) includes the surface area. However, it is not clear what is the appropriate surface area value to use in calculating leach-rates. Generally, the choice is between BET gas (N_2 or Kr) adsorption values or geometric calculations.

Others (Oversby, 1982; Coles and Bazan, 1981) have shown that N_2 and Kr BET surface areas are 5 to 27 times larger than geometric calculations of surface areas. Assuming cubic particles with a particle size equivalent to the 50th percentile of the cumulative weight distribution from the sieve size analyses (Coles et al. 1979) and a density of 2.3 g/cm^3 (Borg, 1982) we calculated geometric surface areas for the ground melt glass samples (Table 3). As predicted the N_2 BET surface area is larger than the calculated geometric surface area.

Table 3: Comparison of Surface Area Values

	<u>Glass 1</u>	<u>Glass 2</u>	<u>Glass 3</u>
Particle Size (50%) (μm)	85	80	78
Estimated Particle Density (g/cm^3)	2.3	2.3	2.3
Geometric Surface Area (m^2/g)	0.031	0.033	0.033
N_2 BET Surface Area (m^2/g)	0.396	0.465	0.457
BET/Geometric	12.9	14.3	13.7

From the BET/geometric ratio and Eq. 1, we see that using the BET surface area underestimates the leach-rates for these melt glasses. However, to remain consistent with data presented in the earlier report on this experiment (Coles et al. 1979) the leach-rates reported in this report (Appendices A and B) used the N_2 BET surface areas. When comparing our leach-rates from glass to glass and over time it is immaterial which surface area method was used, as long as we are consistent.

Using Eq. 1 to calculate the leach-rate, we assume the samples have a constant surface area, a constant mass, and a constant source term. The data verifying the constant surface area assumption comes from a second melt glass leaching experiment (Rego, 1983) which included ground samples of melt glass #3. The surface area per gram was measured before and after 420 days of leaching. Within experimental uncertainty the surface area per gram of the glass did not change. This supports our assumption of constant surface area. Assuming a constant source term is reasonable considering the low cumulative leach values (generally <1.5%) we measured (Tables 1a-c).

To verify a constant sample mass, we tried to obtain post-leach weights on the melt glass samples. A gelatinous layer, from decomposition of the melt glass, had formed around the samples and filters. This made quantitative transfer difficult. However, on all but two cells (#6 and #8), the weight of the dried, post-leach sample was from 96 to 99% of the pre-leach sample weight. A variation in sample mass of 4% over 420 days is a small change. When we consider that sample loss in transfer

may account for about 4% of the original mass and comparing this to the relative magnitude of the other experimental uncertainties (counting statistics, time measurements, etc.), we feel comfortable in assuming a constant sample mass in our calculations.

As described in the experimental section, the leachate samples were composites of all the ground-water leachate which passed through the cell during a given leaching interval. The composite leach-rate calculated from Eq. 1 is not the value at the starting or ending of the leaching interval. It is an integrated leach-rate for the period. The leach-rates were plotted against the log of the mean leaching time in Figures A-1 - A-25. This is the common logarithm of elapsed time from the start of the leaching to the median of each collection interval. By displaying the data in this way, we give a more clear picture of the change in the leach-rate with time.

The leach-rate for each radionuclide varied as the leaching progressed. The specific pattern of leach-rate versus time is slightly different for each radionuclide, flow-rate, melt glass combination. But in general, the leach-rate drops most quickly in the first 11 days of leaching, after which the decrease is more gradual. The majority of the radionuclides (^{22}Na , ^{54}Mn , $^{57,58}\text{Co}$, ^{60}Co , ^{106}Ru , 124 and ^{125}Sb , ^{137}Cs , and ^{239}Pu) showed this smooth pattern. The final rates were from 1.5 to 2.5 orders of magnitude less than the initial leach-rates. The leach-rate for these radionuclides show little or no difference between the fast and slow flow-rate values.

For the radionuclides ^{155}Eu , ^{144}Ce , and ^{95}Zr the leach-rate was not as smooth or predictable. The initial leach-rates were often lower than those measured later in the experiment. The leach-rate for these radionuclides showed definite differences between the fast and slow flow-rate results. The following section contains a discussion of the trends, similarities, and differences between the leach-rates of radionuclides consistently detected in the leachates. Appendix A contains the plots of leach-rate versus time and Appendix B contains tables of the calculated leach-rates.

Sodium-22

Sodium-22 had a higher leach-rate than the other radionuclides. The leach-rate variation with time in both melt glass #2 and #3 show a smooth decline with no difference between the fast and slow flow-rates (Fig. A-1 and A-2). The leach-rates were continuing to decline after 420 days of leaching. The "not detected" value for day 2 in glass #2, fast flow-rate, is most likely due to a problem with the gamma-counting. Melt glass #1 originally contained far less ^{22}Na than the other two melt glass samples (Table 1a). As a result, the leachates of melt glass #1 did not contain detectable amounts of ^{22}Na .

Manganese-54

Manganese-54 was originally present in all three melt glass samples (Tables 1a-c). The general leach-rate pattern for ^{54}Mn is very similar to the ^{22}Na . For all three glasses the fast flow-rate leach-rates show a steady decline over the 420 days of leaching (Fig. A-3 - A-5). However, the slow flow-rate channels show a slightly different pattern. The initial

decline in the slow flow-rate leach-rate was not as rapid as in the fast flow-rate channels. For melt glass #1 the slow flow-rate channels show an increase in leach-rate for day 38 and day 70 samples. This has not been explained and is possibly due to contamination or a counting/calculation error. For melt glass #3 the slow flow-rate leach-rate increases initially and then begins to decrease. The slow flow-rate leach-rates begin a rapid decline after 100 days of leaching. The leach-rates have not leveled off after 420 days of leaching for either flow-rate.

Ruthenium-106

The general trend of the ^{106}Ru leaching for melt glasses #2 and #3 followed the ^{22}Na pattern with smooth decrease from the beginning of the experiment. (Fig. A-6 - A-7) The fast and slow flow-rate results agree within experimental uncertainty. Ruthenium-106 was only sporadically detected in the melt glass #1 leachates. The initial ^{106}Ru activity in melt glass #1 (4.28×10^5 dpm/g) was comparable to the melt glass #3 activity (6.95×10^5 dpm/g), yet the leaching behavior was very different. Ruthenium-103 has a short half-life (40 days) and was not detected in the glass #2 or #3 leachates after day 1, or in the pre-leach melt glass #1 samples. Therefore, no comparison in leaching behavior between ^{106}Ru and ^{103}Ru can be made.

Cobalt-57, -58 and -60

All three cobalt isotopes detected in the melt glasses (^{57}Co , ^{58}Co and ^{60}Co) were detected in the leachates of glasses #2 and #3. The leach-rates are nearly identical for all three isotopes in a given glass

and flow-rate combination (Fig. A-8 - A-9). All three isotopes have a nickel precursor:

- ^{57}Ni (n,p) ^{57}Co
- ^{58}Ni (n,p) ^{58}Co
- ^{60}Ni (n,p) ^{60}Co

Cobalt-60 is also an activation product of ^{59}Co (^{59}Co (n, γ) ^{60}Co). The behavior of the three isotopes during the cooling and vitrification of the glasses is evidently similar and is reflected in their identical leaching behavior.

The leach-rate pattern for the cobalt isotopes is similar to that for ^{54}Mn . The difference between the fast and slow flow-rate leaching for ^{54}Mn is also seen in the cobalt leaching (Fig. A-3 - A-5 and A-8 - A-9). This is not too surprising since Fe (^{54}Fe (n,p) ^{54}Mn), Ni and Co are all first row transition metals.

Due to the short half-lives of ^{57}Co and ^{58}Co , only ^{60}Co was routinely detected in the melt glass #1 leachates (Fig. A-10). The quality of the ^{60}Co gamma-counts on these leachates is poor but the leach-rate data follows the general trend for glasses #2 and #3, and the variations in the data should not be considered significant.

Antimony-124 and -125

The leach-rate pattern for both of the antimony isotopes follows the general pattern exhibited in ^{22}Na , ^{54}Mn , ^{106}Ru and the cobalt isotopes. However, the agreement in the actual leach-rates seen with the three cobalt isotopes is not evident for ^{124}Sb and ^{125}Sb (Fig. A-11 and A-12). The

^{124}Sb leach-rates are slightly higher than those of ^{125}Sb for both glasses 2 and #3. Due to the rapid decay of the ^{124}Sb ($t_{1/2}=60$ days), it was not detected in the later leachate samples. Antimony-124 was not detected in the pre-leach samples of melt glass #1; therefore, we detected only ^{125}Sb in the melt glass #1 leachates (Table B-9 and Fig. A-13).

The difference in ^{124}Sb and ^{125}Sb leach-rates can be explained by the different ways they are produced by the nuclear explosion. Antimony-124 is produced by activation of stable ^{123}Sb ($^{123}\text{Sb}(n,\gamma)^{124}\text{Sb}$) while ^{125}Sb is produced by beta decay along the mass 125 fission product decay chain:

Comparing the boiling points of these precursors (In - 2000°C ; Sn - 2270°C ; Sb - 1380°C), we expect that ^{125}Sb probably condensed in the melt, as its In or Sn precursor, before ^{124}Sb . The earlier condensation of the ^{125}Sb precursors implies a more homogeneous distribution in the glasses compared to ^{124}Sb due to mixing after condensation. This could account for the lower ^{125}Sb leach-rates.

Cesium-137

Overall, the leach-rate pattern for ^{137}Cs is the same as that of the previous radionuclides. Lower leach-rate values in melt glass # 1 are obtained in the fast flow-rate channels only during days 2-6, and after that the fast and slow flow-rate leach-rates agree. All 20 channels show anomalously high ^{137}Cs activity on day 120. The leach-rate values

from these data are plotted in Fig. A-14 - A-16 but are not connected to the other points. The reason for these high ^{137}Cs values is not clear, but we expect that they are due to either a gamma-counting/calculational error or to contamination. Other anomalous values exist in the melt glass #2 and #3 data. The "not detected" values are suspect. The ^{137}Cs activity reported for the day 38 fast flow-rate leachate of glass #3 is suspect because it is a result from only one of the triplicate samples. Cesium-134 was not detected in any leachate after day 1, probably due to its low activity in the pre-leach samples. Therefore no comparison can be made of the ^{134}Cs and ^{137}Cs leaching properties.

Plutonium-239

The smooth leach-rate pattern was also observed for ^{239}Pu in melt glass #2 (Fig. A-17). Due to the major effort required for each ^{239}Pu analysis, only one fast flow-rate channel containing melt glass #2 (channel #5) was chosen. Six of the eleven composite leachates (day 1, 6, 32, 120, 230, and 420) from this channel were then analyzed for ^{239}Pu .

The leach-rate and cumulative leach values for the radionuclides just discussed, ^{22}Na , ^{54}Mn , ^{106}Ru , $^{57,58,60}\text{Co}$, $^{124,125}\text{Sb}$, ^{137}Cs , and ^{239}Pu , are independent of the measured leachant flow-rate. The flow pattern of the leachant through the ground glass sample determines the actual flow-rate at different points within the sample. If even piston flow occurs, the flow-rate will be the same for the entire sample.

However, if flow channels are established within the sample, the flowing leachant will interact with only the small portion of the glass along the channels. The remainder of the glass will be in contact with leachant which is static with respect to the flow in the channels. Following the initial leaching of the "channel glass", the diffusion of this "static leachant" solution from the remainder of the sample into the channels would be the major source of activity in the leachate. This diffusion process would be independent of the flow-rate through the channel. This idea is a possible explanation for the lack of flow-rate dependence of the leach behavior.

Three radionuclides, ^{95}Zr , and the two rare earths, ^{144}Ce , and ^{155}Eu , have flow-rate dependent leach behavior. These radionuclides may be bound in the melt glass in a form different from the radionuclides which show no flow-rate dependence. Melt glass is often found to be highly vesiculated, even on the microscopic scale (Borg, 1982). We expect a different leach behavior from radionuclides associated with the vesicles compared to those bound in the glass. The glass bound radionuclides would have leach-rates dependent on the glass dissolution rate, which is dependent on the volume and, hence, flow-rate of leachant. Other causes for flow-rate dependent leaching are rate differences in dissolution versus diffusion processes, and resorption of leach radionuclide on the gel layer surrounding the melt glass or the walls of the leaching apparatus.

Zirconium-95

The ^{95}Zr data is very sporadic in melt glass #1, and in melt glass #2 ^{95}Zr was only detected in the fast flow-rate channels (Fig. A-18). (The high day 38 value is suspect and is probably due to contamination or a counting/calculational error.) In melt glass #3 ^{95}Zr was detected in the leachates of both the fast and slow flow-rate channels (Fig. A-19). However, the slow flow-rate leach-rates are below the fast flow-rate values. (Again the high day 38 value is suspect.) Though the data is not of good quality, it indicates a difference in the fast and slow flow-rate leaching.

Cerium-144

The effect of flow-rate on the leach-rate is most pronounced in the ^{144}Ce data. The fast flow-rate data for all three melt glasses show a fairly smooth decline, however this decline is very slight. (Fig A-20 - A-22). For most of the radionuclides, the leach-rates decrease over two orders of magnitude during the 420 days of leaching. However the decrease for the fast flow-rate leach of ^{144}Ce is only 30, 10 and 8 for glasses #1, 2 and 3, respectively. In contrast the slow flow-rate data for ^{144}Ce is very scattered. Each melt glass shows a different slow flow-rate pattern (Fig. A-20 A-22).

The mechanism of the cerium leach is apparently different than for the majority of radionuclides. If the cerium is very well integrated into the glass matrix its release into solution would be controlled by the glass dissolution since cerium is known to be very insoluble. This

is expected to be a step which is controlled by flow-rate. Further work will be necessary to understand the ^{144}Ce results. Though ^{141}Ce was originally present in melt glass #2 and #3 it rapidly decayed away ($t_{1/2} = 32$ days) and was not detected in the leachates after day 1. Therefore, no comparison can be made of the ^{141}Ce and ^{144}Ce leaching properties.

Europium-155

Europium-155 displayed an unusual leaching pattern. For melt glass #1, which had the lowest pre-leach ^{155}Eu activity, ^{155}Eu was consistently detected in the fast flow-rate leachates. In the melt glass #1 slow flow-rate leachates ^{155}Eu was not consistently detected until day 11 (a single channel, #13, contained a large amount of ^{155}Eu on day 1 but was not detected again until day 11). Following day 11 the slow flow-rate leach-rates were similar to but less than the fast flow-rate leach-rates (Fig. A-23).

For melt glass #2 and #3 ^{155}Eu was not detected until day 11 in the fast flow-rate leachates (Fig. A-24 - A-25). Europium-155 was not detected until day 70 and day 120 in the slow flow-rate leachates of melt glass #2 and #3, respectively. Since the sample volume from the first few collection intervals is small, very low leach-rates do not produce detectable levels of activity. The low initial ^{155}Eu leach-rate may be due to a hold-up after it has leached from the glass or a resistance to leaching until the glass has undergone a change. Apparently, the ^{155}Eu was either held in the glass until the matrix had undergone a change, or the europium was being held-up after being leached from the glass.

Radiation damage and glass dissolution are two time dependant processes which could result in the degradation of the glass. Melt glass #1, as the "oldest" sample, probably had the most extensive radiation damage at the start of the leaching. This could account for the smooth ^{155}Eu leach-rate pattern in these samples (Fig. A-23). The slow flow-rate glass #1 channels show ^{155}Eu leaching beginning day 11, perhaps due to slower dissolution of the glass. Similar arguments can explain the glass #2 and #3 results.

The second possible explanation is that the release of the ^{155}Eu from the glass was followed by its reabsorption on the surrounding gel layer or the walls of the leaching apparatus. The ^{155}Eu would be detected in the leachates after its re-release. We have no data to support this mechanism, so it is purely conjecture.

SUMMARY

Melt glass material from the explosion chimneys of three nuclear tests was leached in a single-pass flow-through system for 420 days. All samples were held at 25°C and natural ground water was flowed over the samples at one of two flow-rates (34 or 185 ml/day). Gamma-counting detected eighteen radionuclides in the pre-leach melt glasses. Of these, twelve were routinely detected in composite leachates collected on days 1, 2, 3, 6, 11, 32, 38, 70, 120, 230, and 420.

The leach-rates for the radionuclides ^{22}Na , ^{54}Mn , ^{57}Co , ^{58}Co , ^{60}Co , ^{106}Ru , ^{124}Sb , ^{125}Sb , ^{137}Cs , and ^{239}Pu generally decreased with time giving a smooth pattern. There was no appreciable difference between the fast and slow flow-rate leach-rates for these radionuclides. Any differences for these radionuclides between the three glass samples is generally due to the differences in pre-leach activities.

Three radionuclides, ^{95}Zr , ^{144}Ce , and ^{155}Eu , showed a different leaching behavior. For these radionuclides different leach-rate patterns were seen at each flow-rate. Europium-155 was not detected in the initial slow flow-rate leachates but was detected later in the experiment. The slow flow-rate leach-rate pattern for ^{144}Ce was very different from the fast flow-rate leach-rate. In glass #2 ^{95}Zr was detected in the fast flow-rate leachates but not in the slow flow-rate leachates.

By summing the activity of each radionuclide detected in the leachates, we determined the percent cumulative leach. The cumulative leach is not directly related to the volume of water passed over the glass samples. The fast flow-rate cumulative leach was nearly equivalent to the associated slow flow-rate value. Again ^{95}Zr and ^{144}Ce show a greater difference between the fast and slow flow-rate results than the other radionuclides.

We calculated that 0.14% of the ^{239}Pu pre-leach activity was leached from melt glass #2. This is a higher percentage than reported by others for ^{239}Pu leached from nuclear test debris (Wolfsberg 1978) and doped borosilicate glass beads (Coles et al., 1982). Algal growth in the leachate may have caused the higher ^{239}Pu leach. Further study is needed to better understand ^{239}Pu leaching from melt glass.

The small percentages of activity leached from these samples leads us to conclude that the possibility of ground water contamination due to nuclear melt glass leaching is very small. This is particularly true when we consider the difference in available surface area between the bulk melt glass in the cavity and the finely ground samples used in this study. Even though the amount of the ^{239}Pu released from the melt glass is very small, it still exhibited a larger leach-rate than expected. Plutonium mobility should not present a hazard because the soils and rocks will remove much of the plutonium from the ground water by surface absorption (Hajek, 1966; Rydberg 1982).

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APPENDIX A

Leach-Rate Plots

This Appendix contains log-log plots of leach-rate, calculated from Eq. 1 and tabulated in Appendix B, versus mean leach time for each radio-nuclide routinely detected in the melt glass leachates.

Symbols placed on the abscissa (i.e., Fig. A-1) indicate a "not detected" activity in the corresponding leachates (see text for further explanation). Circled points (i.e., Fig. A-10) indicate anomalously high activity in the corresponding leachates (see text for further explanation). We are reporting these values but do not include them in the overall leach-rate trends.

LEACH RATE G/CM²-DAY

Fig. A-1

LEACH RATE G/CM²-DAY

Fig. A-2

LEACH RATE G/CM²·HR

LEACH RATE PLOTS
MELT GLASS #1

Fig. A-3

LEACH RATE PLOTS
MELT GLASS #2

LEACH RATE G/CM²·HR

Fig. A-4

Fig. A-5

LEACH RATE PLOTS
MELT GLASS 42

Fig. A-6

LEACH RATE PLOTS
MELT GLASS 43

Fig. A-7

LEACH RATE PLOTS
MELT GLASS #2

Fig. A-8

LEACH RATE PLOTS
MELT GLASS #3

Fig. A-9

LEACH RATE PLOTS
MELT CLASS 02

Fig. A-11

LEACH RATE PLOTS
MELT CLASS 03

Fig. A-12

Fig. A-13

LEACH RATE PLOTS
MELT GLASS #1

Fig. A-14

LEACH RATE PLOTS
MELT GLASS #2

Fig. A-15

LEACH RATE PLOTS
MELT GLASS 03

Fig. A-16

Fig. A-17

LEACH RATE PLOTS
MELT GLASS #2

Fig. A-18

LEACH RATE PLOTS
MELT GLASS #3

Fig. A-19

LEACH RATE PLOTS
MELT GLASS #1

Fig. A-20

LEACH RATE PLOTS
MELT GLASS #2

Fig. A-21

LEACH RATE PLOTS
MELT GLASS 49

Fig. A-22

LEACH RATE PLOTS
MELT GLASS #1

Fig. A-23

LEACH RATE PLOTS
MELT GLASS #2

Fig. A-24

LEACH RATE PLOTS
MELT GLASS 43

Fig. A-25

APPENDIX B

Melt Glass Leaching Rate Values

Unless otherwise noted, the leach-rates in this appendix are the average value of the replicate samples. If no activity was detected in one or more of the samples, the remaining values were used to determine the average. The footnote letters indicate when this has occurred. The uncertainties, in parenthesis, are one standard deviation in percent.

a = average value $\pm$ 1 σ (%) of 2 samples

b = data from one sample only $\pm$ counting uncertainty

ND = not detected in any sample

SD = sporadically detected

Table B-1: ^{22}Na Leach Rates
(g-glass/m² day)

Collection day	MELT GLASS #1	MELT GLASS #2	MELT GLASS #3
		<u>Fast Flow Rate</u>	
1	ND	9.83×10^{-3} (12)	5.82×10^{-3} (24)
2	"	1.28×10^{-3} (21)	1.42×10^{-3} (25)
3	"	ND	7.35×10^{-4} (13)
6	"	4.02×10^{-4} (21)	5.88×10^{-4} (22)
11	"	2.66×10^{-4} (16)	4.01×10^{-4} (7)
32	"	1.44×10^{-4} (38)	2.34×10^{-4} (9)
38	"	1.25×10^{-4} (20)	2.09×10^{-4} (17)
70	"	1.07×10^{-4} (14)	1.44×10^{-4} (17)
120	"	6.82×10^{-5} (20)	1.01×10^{-4} (9)
230	"	4.58×10^{-5} (10)	6.93×10^{-5} (10)
420	"	3.78×10^{-5} (8)	4.61×10^{-5} (15)
		<u>Slow Flow Rate</u>	
1	ND	1.17×10^{-2} (5)	8.03×10^{-3} (12)
2	"	b 1.35×10^{-3} (34)	1.59×10^{-3} (20)
3	"	b 6.93×10^{-4} (43)	8.32×10^{-4} (25)
6	"	3.50×10^{-4} (22)	6.10×10^{-4} (16)
11	"	2.08×10^{-4} (26)	4.00×10^{-4} (7)
32	"	1.62×10^{-4} (16)	2.11×10^{-4} (6)
38	"	a 1.56×10^{-4} (29)	1.57×10^{-4} (11)
70	"	1.22×10^{-4} (19)	1.28×10^{-4} (7)
120	"	6.40×10^{-5} (17)	9.28×10^{-5} (7)
230	"	4.69×10^{-5} (6)	5.85×10^{-5} (5)
420	"	2.97×10^{-5} (6)	4.39×10^{-5} (10)

Table B-2: ^{54}Mn Leach Rates
(g-glass/m²day)

Collection day	MELT GLASS #1	MELT GLASS #2		MELT GLASS #3
		Fast	Flow Rate	
1	1.01×10^{-3} (9)		4.70×10^{-4} (18)	8.24×10^{-4} (4)
2	1.68×10^{-4} (10)		3.80×10^{-4} (7)	5.11×10^{-4} (6)
3	1.06×10^{-4} (30)		2.44×10^{-4} (4)	2.69×10^{-4} (4)
6	7.39×10^{-5} (27)		1.44×10^{-4} (4)	1.43×10^{-4} (4)
11	4.06×10^{-5} (9)		6.75×10^{-5} (13)	7.61×10^{-5} (9)
32	a 1.64×10^{-5} (9)		2.77×10^{-5} (28)	4.44×10^{-5} (6)
38	a 1.25×10^{-5} (24)		1.90×10^{-5} (27)	4.22×10^{-5} (5)
70	7.05×10^{-6} (8)		1.47×10^{-5} (35)	3.60×10^{-5} (21)
120	a 7.06×10^{-6} (21)		8.06×10^{-6} (27)	2.30×10^{-5} (4)
230	9.32×10^{-6} (30)		4.49×10^{-6} (31)	1.30×10^{-5} (27)
420	6.45×10^{-6} (25)		3.27×10^{-6} (30)	9.40×10^{-6} (5)
<u>Slow Flow Rate</u>				
1	a 1.14×10^{-3} (46)		8.12×10^{-5} (25)	1.03×10^{-4} (24)
2	4.88×10^{-4} (47)		8.94×10^{-5} (19)	1.91×10^{-4} (4)
3	1.68×10^{-4} (30)		1.03×10^{-4} (15)	2.07×10^{-4} (18)
6	7.38×10^{-5} (10)		7.92×10^{-5} (15)	1.35×10^{-4} (17)
11	3.87×10^{-5} (16)		4.50×10^{-5} (24)	5.42×10^{-5} (30)
32	1.35×10^{-5} (17)		2.92×10^{-5} (9)	2.75×10^{-5} (21)
38	a 1.72×10^{-5} (34)		1.75×10^{-5} (12)	2.14×10^{-5} (7)
70	2.89×10^{-5} (15)		1.23×10^{-5} (5)	1.63×10^{-5} (4)
120	5.23×10^{-6} (31)		6.84×10^{-6} (14)	1.52×10^{-5} (7)
230	b 3.20×10^{-6} (11)		3.15×10^{-6} (10)	7.73×10^{-6} (5)
420	2.66×10^{-6} (40)		1.03×10^{-6} (10)	2.31×10^{-6} (4)

Table B-3: ^{57}Co Leach Rates
(g-glass/m²day)

Collection day	MELT GLASS #1	MELT GLASS #2	MELT GLASS #3
		<u>Fast Flow Rate</u>	
1	SD	1.41×10^{-3} (21)	6.69×10^{-4} (27)
2	"	2.40×10^{-3} (5)	4.03×10^{-4} (4)
3	"	2.15×10^{-3} (4)	2.46×10^{-4} (4)
6	"	1.51×10^{-3} (18)	1.32×10^{-4} (7)
11	"	6.50×10^{-4} (18)	7.43×10^{-5} (12)
32	"	2.86×10^{-4} (25)	4.70×10^{-5} (4)
38	"	1.84×10^{-4} (25)	4.61×10^{-5} (15)
70	"	8.11×10^{-5} (25)	3.59×10^{-5} (18)
120	"	5.66×10^{-5} (25)	2.53×10^{-5} (17)
230	"	1.87×10^{-5} (6)	1.32×10^{-5} (23)
420	"	9.01×10^{-6} (23)	7.08×10^{-6} (27)
		<u>Slow Flow Rate</u>	
1	SD	1.48×10^{-4} (9)	1.54×10^{-4} (42)
2	"	1.88×10^{-4} (25)	1.23×10^{-4} (5)
3	"	2.54×10^{-4} (12)	1.23×10^{-4} (14)
6	"	4.08×10^{-4} (25)	9.96×10^{-5} (4)
11	"	4.30×10^{-4} (25)	5.20×10^{-5} (19)
32	"	1.92×10^{-4} (14)	2.76×10^{-5} (16)
38	"	1.22×10^{-4} (11)	2.41×10^{-5} (5)
70	"	9.44×10^{-5} (9)	1.90×10^{-5} (4)
120	"	6.00×10^{-5} (17)	1.68×10^{-5} (10)
230	"	2.64×10^{-5} (12)	1.09×10^{-5} (25)
420	"	6.62×10^{-6} (9)	4.56×10^{-6} (40)

Table B-4: ^{58}Co Leach Rates
(g-glass/m²day)

Collection day	MELT GLASS #1	MELT GLASS #2		MELT GLASS #3
		Fast	Flow Rate	
1	ND		1.13x10 ⁻³ (23)	5.33x10 ⁻⁴ (26)
2	"		2.39x10 ⁻³ (5)	3.65x10 ⁻⁴ (11)
3	"		2.13x10 ⁻³ (5)	2.16x10 ⁻⁴ (9)
6	"		1.39x10 ⁻³ (19)	1.19x10 ⁻⁴ (10)
11	"		5.99x10 ⁻⁴ (18)	6.39x10 ⁻⁵ (20)
32	"		2.56x10 ⁻⁴ (40)	4.05x10 ⁻⁵ (4)
38	"		1.61x10 ⁻⁴ (40)	4.37x10 ⁻⁵ (20)
70	"		7.76x10 ⁻⁵ (24)	3.36x10 ⁻⁵ (17)
120	"		4.93x10 ⁻⁵ (32)	2.35x10 ⁻⁵ (21)
230	"		1.73x10 ⁻⁵ (19)	1.18x10 ⁻⁵ (20)
420	"		ND	ND
		Slow	Flow Rate	
1	ND		1.00x10 ⁻⁴ (10)	9.96x10 ⁻⁵ (40)
2	"		1.66x10 ⁻⁴ (18)	1.15x10 ⁻⁴ (18)
3	"		2.77x10 ⁻⁴ (15)	1.13x10 ⁻⁴ (25)
6	"		3.88x10 ⁻⁴ (33)	9.23x10 ⁻⁵ (18)
11	"		4.18x10 ⁻⁴ (33)	4.52x10 ⁻⁵ (26)
32	"		1.74x10 ⁻⁴ (14)	2.26x10 ⁻⁵ (18)
38	"		1.01x10 ⁻⁴ (11)	2.22x10 ⁻⁵ (14)
70	"		6.71x10 ⁻⁵ (20)	1.67x10 ⁻⁵ (5)
120	"		5.11x10 ⁻⁵ (20)	1.56x10 ⁻⁵ (12)
230	"		1.70x10 ⁻⁵ (5)	1.23x10 ⁻⁵ (17)
420	"		b 1.15x10 ⁻⁶ (40)	b 6.87x10 ⁻⁶ (50)

Table B-5: ^{60}Co Leach Rate
(g-glass/m² day)

Collection day	MELT GLASS #1	MELT GLASS #2	MELT GLASS #3
	<u>Fast Flow Rate</u>		
1	5.51x10 ⁻⁴ (13)	1.23x10 ⁻³ (23)	5.54x10 ⁻⁴ (25)
2	3.77x10 ⁻⁴ (23)	2.04x10 ⁻³ (5)	3.39x10 ⁻⁴ (7)
3	b 2.55x10 ⁻⁴ (66)	1.83x10 ⁻³ (4)	2.10x10 ⁻⁴ (4)
6	ND	1.46x10 ⁻³ (27)	1.11x10 ⁻⁴ (4)
11	a 7.90x10 ⁻⁵ (12)	5.48x10 ⁻⁴ (18)	6.30x10 ⁻⁵ (9)
32	b 2.40x10 ⁻⁵ (14)	2.39x10 ⁻⁴ (65)	3.94x10 ⁻⁵ (6)
38	5.17x10 ⁻⁵ (27)	1.46x10 ⁻⁴ (55)	3.95x10 ⁻⁵ (7)
70	a 1.09x10 ⁻⁵ (21)	7.21x10 ⁻⁵ (24)	3.29x10 ⁻⁵ (15)
120	a 6.81x10 ⁻⁶ (20)	4.58x10 ⁻⁵ (35)	2.21x10 ⁻⁵ (23)
230	a 6.60x10 ⁻⁶ (25)	1.48x10 ⁻⁵ (5)	1.19x10 ⁻⁵ (22)
420	a 6.53x10 ⁻⁶ (25)	8.88x10 ⁻⁶ (26)	7.43x10 ⁻⁶ (24)
<u>Slow Flow Rate</u>			
1	a 5.11x10 ⁻⁴ (50)	1.32x10 ⁻⁴ (4)	1.45x10 ⁻⁴ (44)
2	a 3.58x10 ⁻⁴ (24)	1.84x10 ⁻⁴ (23)	1.02x10 ⁻⁴ (30)
3	b 2.24x10 ⁻⁴ (48)	2.39x10 ⁻⁴ (15)	1.06x10 ⁻⁴ (11)
6	b 1.59x10 ⁻⁴ (22)	3.54x10 ⁻⁴ (27)	8.34x10 ⁻⁵ (18)
11	a 5.11x10 ⁻⁵ (19)	3.59x10 ⁻⁴ (33)	4.44x10 ⁻⁵ (20)
32	b 1.29x10 ⁻⁵ (39)	1.57x10 ⁻⁴ (13)	2.24x10 ⁻⁵ (16)
38	5.10x10 ⁻⁵ (29)	1.00x10 ⁻⁴ (13)	1.91x10 ⁻⁵ (7)
70	a 9.86x10 ⁻⁶ (23)	7.43x10 ⁻⁵ (8)	1.55x10 ⁻⁵ (4)
120	7.33x10 ⁻⁶ (38)	4.74x10 ⁻⁵ (15)	1.42x10 ⁻⁵ (9)
230	b 4.31x10 ⁻⁶ (13)	2.10x10 ⁻⁵ (15)	9.54x10 ⁻⁶ (28)
420	b 2.31x10 ⁻⁶ (20)	5.87x10 ⁻⁶ (13)	4.50x10 ⁻⁶ (44)

Table B-6: ^{95}Zr Leach Rates
(g-glass/m²day)

Collection day	MELT GLASS #1	MELT GLASS #2	MELT GLASS #3
		<u>Fast Flow Rate</u>	
1	SD	1.25×10^{-4} (8)	1.12×10^{-4} (10)
2	"	8.37×10^{-5} (30)	5.44×10^{-5} (18)
3	"	8.01×10^{-5} (30)	4.25×10^{-5} (38)
6	"	ND	1.79×10^{-5} (13)
11	"	ND	1.72×10^{-5} (22)
32	"	8.49×10^{-6} (28)	1.11×10^{-5} (22)
38	"	a 1.35×10^{-4} (5)	1.18×10^{-5} (31)
70	"	7.69×10^{-6} (35)	1.36×10^{-5} (32)
120	"	8.53×10^{-6} (35)	1.24×10^{-5} (12)
230	"	1.41×10^{-5} (35)	1.40×10^{-5} (29)
420	"	b 1.12×10^{-5} (51)	ND
		<u>Slow Flow Rate</u>	
1	SD	SD	3.62×10^{-5} (47)
2	"	"	a 3.17×10^{-5} (35)
3	"	"	a 3.55×10^{-5} (33)
6	"	"	ND
11	"	"	b 7.58×10^{-6} (45)
32	"	"	a 5.28×10^{-5} (26)
38	"	"	b 1.68×10^{-5} (20)
70	"	"	a 4.59×10^{-6} (22)
120	"	"	a 6.88×10^{-6} (15)
230	"	"	ND
420	"	"	ND

Table B-7: ^{106}Ru Leach Rates
(g-glass/m²day)

Collection day	MELT GLASS #1	MELT GLASS #2	MELT GLASS #3
		<u>Fast Flow Rate</u>	
1	SD	5.82×10^{-4} (8)	7.66×10^{-4} (24)
2	"	1.83×10^{-4} (34)	a 2.06×10^{-4} (50)
3	"	1.16×10^{-4} (21)	b 1.38×10^{-4} (50)
6	"	9.49×10^{-5} (13)	7.66×10^{-5} (21)
11	"	6.50×10^{-5} (17)	6.61×10^{-5} (19)
32	"	6.07×10^{-5} (21)	4.20×10^{-5} (10)
38	"	5.69×10^{-5} (28)	5.00×10^{-5} (40)
70	"	4.58×10^{-5} (22)	3.15×10^{-5} (19)
120	"	3.16×10^{-5} (21)	1.78×10^{-5} (23)
230	"	1.90×10^{-5} (28)	1.25×10^{-5} (19)
420	"	1.52×10^{-5} (40)	8.77×10^{-6} (21)
		<u>Slow Flow Rate</u>	
1	SD	4.48×10^{-4} (15)	4.86×10^{-4} (12)
2	"	1.72×10^{-4} (41)	ND
3	"	8.51×10^{-5} (19)	b 1.12×10^{-4} (54)
6	"	5.48×10^{-5} (45)	b 6.48×10^{-5} (55)
11	"	4.22×10^{-5} (45)	4.76×10^{-5} (30)
32	"	2.84×10^{-5} (31)	2.62×10^{-5} (28)
38	"	3.01×10^{-5} (27)	b 3.63×10^{-5} (46)
70	"	3.55×10^{-5} (39)	1.28×10^{-5} (19)
120	"	2.62×10^{-5} (19)	1.28×10^{-5} (27)
230	"	1.84×10^{-5} (7)	1.51×10^{-5} (30)
420	"	1.02×10^{-5} (13)	6.23×10^{-6} (21)

Table B-8: ^{124}Sb Leach Rates
(g-glass/m²day)

Collection day	MELT GLASS #1	MELT GLASS #2	MELT GLASS #3
		<u>Fast Flow Rate</u>	
1	ND	4.85×10^{-3} (21)	3.69×10^{-3} (26)
2	"	4.33×10^{-3} (18)	1.34×10^{-3} (12)
3	"	a 4.34×10^{-3} (67)	6.80×10^{-4} (21)
6	"	2.41×10^{-3} (37)	a 3.03×10^{-4} (39)
11	"	1.38×10^{-3} (16)	2.45×10^{-4} (15)
32	"	8.79×10^{-4} (12)	1.33×10^{-4} (30)
38	"	a 6.78×10^{-4} (25)	ND
70	"	4.00×10^{-4} (45)	a 7.69×10^{-5} (46)
120	"	a 2.26×10^{-4} (40)	ND
230	"	ND	ND
420	"	ND	ND
		<u>Slow Flow Rate</u>	
1	ND	1.43×10^{-3} (30)	1.70×10^{-3} (12)
2	"	a 1.98×10^{-3} (40)	1.29×10^{-3} (27)
3	"	b 4.07×10^{-3} (56)	7.19×10^{-4} (30)
6	"	b 1.49×10^{-4} (26)	5.27×10^{-4} (18)
11	"	a 7.02×10^{-4} (40)	2.07×10^{-4} (33)
32	"	6.06×10^{-4} (20)	2.22×10^{-4} (63)
38	"	ND	ND
70	"	2.90×10^{-4} (20)	9.86×10^{-5} (21)
120	"	ND	ND
230	"	ND	ND
420	"	ND	ND

Table B-9: ^{125}Sb Leach Rates
(g-glass/m²day)

Collection day	MELT GLASS #1	MELT GLASS #2	MELT GLASS #3
	<u>Fast Flow Rate</u>		
1	2.39×10^{-4} (18)	2.24×10^{-3} (23)	1.64×10^{-3} (28)
2	a 1.56×10^{-4} (29)	1.34×10^{-3} (8)	4.71×10^{-4} (28)
3	ND	1.01×10^{-3} (11)	b 4.05×10^{-4} (62)
6	b 5.37×10^{-5} (43)	6.79×10^{-4} (10)	a 1.39×10^{-4} (36)
11	2.52×10^{-5} (13)	3.81×10^{-4} (13)	8.45×10^{-5} (36)
32	a 2.04×10^{-5} (13)	1.76×10^{-4} (44)	b 5.85×10^{-5} (67)
38	ND	1.60×10^{-4} (38)	b 3.02×10^{-5} (51)
70	8.35×10^{-6} (26)	9.85×10^{-5} (13)	4.40×10^{-5} (17)
120	a 8.24×10^{-6} (34)	5.82×10^{-5} (21)	2.18×10^{-5} (21)
230	4.16×10^{-6} (38)	4.04×10^{-5} (20)	1.73×10^{-5} (36)
420	5.19×10^{-6} (40)	2.01×10^{-5} (13)	1.27×10^{-5} (45)
<u>Slow Flow Rate</u>			
1	a 6.15×10^{-4} (54)	1.44×10^{-3} (12)	1.16×10^{-3} (10)
2	a 3.95×10^{-4} (30)	6.75×10^{-4} (10)	7.75×10^{-4} (12)
3	ND	5.31×10^{-4} (21)	a 4.28×10^{-4} (16)
6	b 8.13×10^{-5} (34)	4.11×10^{-4} (11)	1.53×10^{-4} (20)
11	a 4.20×10^{-5} (31)	3.73×10^{-4} (14)	a 6.05×10^{-5} (25)
32	1.28×10^{-5} (14)	1.56×10^{-4} (19)	3.93×10^{-5} (36)
38	ND	1.27×10^{-4} (16)	ND
70	6.76×10^{-6} (24)	8.35×10^{-5} (19)	a 2.16×10^{-5} (29)
120	b 6.36×10^{-6} (26)	4.41×10^{-5} (34)	a 1.45×10^{-5} (29)
230	3.30×10^{-6} (50)	2.94×10^{-5} (10)	7.97×10^{-6} (23)
420	3.27×10^{-6} (40)	1.62×10^{-5} (16)	6.51×10^{-6} (16)

Table B-10: ^{144}Ce Leach Rates
(g-glass/m²day)

Collection day	MELT GLASS #1	MELT GLASS #2		MELT GLASS #3
		Fast	Flow Rate	
1	8.17x10 ⁻⁵ (24)	4.90x10 ⁻⁵ (20)	5.98x10 ⁻⁵ (25)	
2	7.68x10 ⁻⁵ (22)	5.15x10 ⁻⁵ (19)	5.52x10 ⁻⁵ (22)	
3	a 7.93x10 ⁻⁵ (11)	4.94x10 ⁻⁵ (36)	6.60x10 ⁻⁵ (26)	
6	6.95x10 ⁻⁵ (40)	4.54x10 ⁻⁵ (35)	5.55x10 ⁻⁵ (29)	
11	3.13x10 ⁻⁵ (4)	3.64x10 ⁻⁵ (13)	4.29x10 ⁻⁵ (5)	
32	a 1.58x10 ⁻⁵ (4)	2.82x10 ⁻⁵ (39)	3.84x10 ⁻⁵ (9)	
38	1.25x10 ⁻⁵ (35)	2.97x10 ⁻⁵ (30)	4.11x10 ⁻⁵ (3)	
70	1.07x10 ⁻⁵ (8)	1.86x10 ⁻⁵ (26)	3.48x10 ⁻⁵ (28)	
120	a 7.39x10 ⁻⁶ (8)	1.08x10 ⁻⁵ (34)	2.64x10 ⁻⁵ (10)	
230	3.03x10 ⁻⁶ (4)	5.60x10 ⁻⁶ (36)	1.28x10 ⁻⁵ (20)	
420	5.01x10 ⁻⁶ (5)	4.25x10 ⁻⁶ (21)	8.37x10 ⁻⁶ (25)	
		Slow	Flow Rate	
1	3.88x10 ⁻⁴ (4)	1.26x10 ⁻⁵ (21)	8.45x10 ⁻⁶ (30)	
2	b 6.01x10 ⁻⁵ (11)	ND	ND	
3	b 9.88x10 ⁻⁶ (60)	a 8.45x10 ⁻⁶ (34)	ND	
6	b 3.54x10 ⁻⁶ (52)	ND	ND	
11	1.38x10 ⁻⁵ (10)	a 1.31x10 ⁻⁶ (35)	b 8.62x10 ⁻⁷ (38)	
32	8.26x10 ⁻⁶ (10)	1.68x10 ⁻⁶ (34)	2.04x10 ⁻⁶ (20)	
38	1.78x10 ⁻⁵ (10)	ND	a 5.12x10 ⁻⁶ (33)	
70	5.84x10 ⁻⁶ (55)	3.10x10 ⁻⁶ (49)	3.21x10 ⁻⁶ (20)	
120	5.18x10 ⁻⁶ (31)	5.45x10 ⁻⁶ (30)	6.85x10 ⁻⁶ (10)	
230	3.47x10 ⁻⁶ (70)	6.40x10 ⁻⁶ (36)	9.80x10 ⁻⁶ (10)	
420	1.98x10 ⁻⁶ (50)	2.65x10 ⁻⁶ (38)	3.68x10 ⁻⁶ (10)	

Table B-11: ^{137}Cs Leach Rates
(g-glass/m²day)

Collection day	MELT GLASS #1	MELT GLASS #2	MELT GLASS #3
	<u>Fast Flow Rate</u>		
1	1.54x10 ⁻³ (20)	1.64x10 ⁻³ (11)	2.09x10 ⁻³ (31)
2	1.56x10 ⁻⁴ (32)	ND	b 6.68x10 ⁻⁴ (56)
3	b 8.89x10 ⁻⁵ (34)	ND	ND
6	a 5.32x10 ⁻⁵ (19)	ND	b 2.79x10 ⁻⁴ (41)
11	2.78x10 ⁻⁵ (29)	b 9.16x10 ⁻⁵ (40)	a 8.78x10 ⁻⁵ (22)
32	a 1.47x10 ⁻⁵ (17)	b 2.38x10 ⁻⁵ (40)	a 3.89x10 ⁻⁵ (14)
38	a 1.38x10 ⁻⁵ (50)	ND	b 1.25x10 ⁻⁴ (32)
70	1.24x10 ⁻⁵ (19)	1.12x10 ⁻⁵ (29)	b 2.14x10 ⁻⁵ (47)
120	a 4.72x10 ⁻⁵ (20)	1.72x10 ⁻⁴ (50)	1.64x10 ⁻⁴ (35)
230	5.63x10 ⁻⁶ (30)	9.25x10 ⁻⁶ (16)	8.86x10 ⁻⁶ (12)
420	3.87x10 ⁻⁶ (70)	6.94x10 ⁻⁶ (19)	7.49x10 ⁻⁶ (41)
	<u>Slow Flow Rate</u>		
1	a 1.00x10 ⁻³ (20)	1.05x10 ⁻³ (10)	1.80x10 ⁻³ (20)
2	6.44x10 ⁻⁴ (16)	7.62x10 ⁻⁴ (26)	1.18x10 ⁻³ (15)
3	3.90x10 ⁻⁴ (36)	b 3.76x10 ⁻⁴ (68)	b 5.83x10 ⁻⁴ (54)
6	1.43x10 ⁻⁴ (28)	ND	a 1.70x10 ⁻⁴ (14)
11	4.04x10 ⁻⁵ (15)	ND	a 8.64x10 ⁻⁵ (33)
32	1.39x10 ⁻⁵ (9)	b 1.75x10 ⁻⁵ (50)	b 2.75x10 ⁻⁵ (59)
38	b 1.44x10 ⁻⁵ (40)	ND	ND
70	8.38x10 ⁻⁶ (23)	b 1.30x10 ⁻⁵ (28)	a 2.19x10 ⁻⁵ (29)
120	4.68x10 ⁻⁵ (16)	3.58x10 ⁻⁴ (75)	1.80x10 ⁻⁴ (33)
230	4.99x10 ⁻⁶ (20)	6.04x10 ⁻⁶ (38)	6.13x10 ⁻⁶ (20)
420	5.09x10 ⁻⁶ (15)	3.95x10 ⁻⁶ (19)	4.02x10 ⁻⁶ (24)

Table B-12: ^{155}Eu Leach Rates
(g-glass/m²day)

Collection day	MELT GLASS #1	MELT GLASS #2	MELT GLASS #3
	<u>Fast Flow Rate</u>		
1	1.55×10^{-4} (18)	ND	ND
2	1.32×10^{-4} (25)	ND	ND
3	1.51×10^{-4} (20)	ND	ND
6	9.42×10^{-4} (20)	ND	ND
11	4.55×10^{-5} (14)	a 8.50×10^{-5} (30)	b 1.21×10^{-4} (49)
32	a 2.52×10^{-5} (23)	a 3.49×10^{-5} (20)	7.01×10^{-5} (37)
38	b 1.95×10^{-5} (34)	a 7.87×10^{-5} (40)	b 6.49×10^{-5} (28)
70	1.57×10^{-5} (10)	b 1.92×10^{-6} (20)	4.17×10^{-5} (26)
120	a 1.39×10^{-5} (17)	9.76×10^{-6} (20)	3.16×10^{-5} (30)
230	5.14×10^{-6} (34)	9.41×10^{-6} (20)	1.90×10^{-5} (17)
420	6.02×10^{-6} (43)	6.49×10^{-6} (20)	1.21×10^{-5} (21)
	<u>Slow Flow Rate</u>		
1	a 4.56×10^{-4} (15)	ND	ND
2	ND	ND	ND
3	ND	ND	ND
6	ND	ND	ND
11	2.65×10^{-5} (32)	ND	ND
32	1.42×10^{-5} (19)	ND	ND
38	b 3.02×10^{-5} (44)	ND	ND
70	1.23×10^{-5} (25)	a 8.70×10^{-6} (38)	ND
120	8.14×10^{-6} (15)	a 1.30×10^{-5} (25)	a 2.09×10^{-5} (59)
230	7.47×10^{-6} (5)	8.45×10^{-6} (28)	1.35×10^{-5} (34)
420	5.09×10^{-6} (15)	4.33×10^{-6} (35)	6.62×10^{-6} (24)

Table B-13: ^{239}Pu Leach-Rates
(g-glass/m²day)

Collection day	MELT GLASS #2	
	<u>Fast</u>	<u>Flow Rate</u>
1		1.53×10^{-4}
2		$6.03 \times 10^{-5*}$
3		$3.86 \times 10^{-5*}$
6		2.36×10^{-5}
11		$1.87 \times 10^{-5*}$
32		1.33×10^{-5}
38		$7.09 \times 10^{-5*}$
70		$9.10 \times 10^{-6*}$
120		7.21×10^{-6}
230		6.09×10^{-6}
420		5.08×10^{-6}

* linearly interpolated value
uncertainty is $\pm 10\%$