LEACHING STUDIES USING PNL 76-68
GLASS BEADS AND UO₂ RODS WITH
UMTANUM BASALT AND NUGGET SANDSTONE

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ABSTRACT

We have performed a 440-day leaching experiment, Bead Leach II, using PNL 76-68 glass beads and simulated uranium fuel rods in the presence of repository host rocks. The experiment was conducted in a single pass continuous-flow apparatus consisting of 72 channels. The experimental conditions were: 25°C and 75°C, flow rates of 1, 10, and 300 ml/d, and leachant solutions consisting of simulated basalt groundwater, brine, and sodium bicarbonate solution. The two host rocks studied were Umtanum basalt and Nugget sandstone.

The Bead Leach II experiment began in late 1980 and the leaching phase was concluded in December, 1981. Analysis of the leachates and of the two rock types was carried out subsequently and the results are presented in two reports. Part I was published in March, 1983, and it included results of the leaching of PNL 76-68 glass beads with the basalt groundwater. Part II (this report) includes results of the leaching of PNL 76-68 glass beads with brine and bicarbonate solution and the leaching of UO₂ pellets with basalt groundwater, brine, and bicarbonate solution. Results are in the form of leach rates, cumulative fractions leached, and adsorption profiles on basalt and sandstone. The radio-nuclides studied were Pu and Np in the case of the glass beads and uranium in the case of the simulated uranium fuel rods.

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I. INTRODUCTION

The Bead Leach II experiment was a long-term single-pass continuous flow leaching experiment which studied the leaching of PNL 76-68 glass beads and simulated UO_2 fuel rods, both in the presence and in the absence of two rock types - Umtanum basalt and Nugget sandstone. There were three leachants used in the experiment: a simulated basalt groundwater, a NaCl brine and a 0.03 M sodium bicarbonate solution. The leaching experiments were conducted at two temperatures (25°C and 75°C) and at three flow rates (1 ml/day, 10 ml/day, and 300 ml/day).

The Bead Leach II experiment was started in September, 1980, and was completed in December, 1981. Since then, we have been analyzing the leachates and the material adsorbed on the rock. Because of the complexity of the experiment and the great number of samples selected for analysis, there are two reports on the completed Bead Leach II experiment. Part I¹, which was published in March, 1983, described the experiment in detail and summarized the data obtained on the leaching of PNL 76-68 glass beads with a simulated basalt groundwater in the presence and absence of crushed basalt.

The present report is Part II, and it contains data obtained on the leaching of:

1. PNL 76-68 glass beads with a NaCl brine.
2. UO_2 simulated fuel rods with a NaCl brine.
3. UO_2 simulated fuel rods with simulated basalt groundwater.
4. UO_2 simulated fuel rods with bicarbonate solution.
5. PNL 76-68 glass beads with bicarbonate solution.

Each of the above will be treated in a separate section of this report.

II. PNL-76-68 GLASS BEADS WITH BRINE SOLUTION

Fourteen cells in the Bead Leach II experiment were assigned to the leaching of glass beads with brine. Six of these cells contained glass beads only, whereas the remaining eight cells contained both glass beads and crushed sandstone. Figure 1 shows the configuration of beads and rock.

The experimental matrix of channel number, temperature, flow rate, and cell contents is shown in Table 1. The glass beads were those described in Part I.¹ The chemical composition of the beads is given in Table 2. The bulk Nugget sandstone was crushed at LLNL with a chipmunk crusher and ground further with a mill grinder. The crushed material was sieved with various screens and only the grain size fraction passing through an 8-mesh screen (2.4 mm) was retained for use. The physical, mineralogical, and cation exchange capacity for this sandstone has been established.²

Both the beads and the crushed rock were cleaned in an ultrasonic bath by rinsing first with distilled water for five minutes, and, then with ethanol for another five minutes. Finally, they were allowed to dry overnight at room temperature.

The solution used for this portion of the Bead Leach II study was based on a scenario of groundwater intrusion from a local sandstone aquifer through the salt which constitutes the repository and finally migrating out of the repository and back into the sandstone aquifer. In retrospect, this scenario was overly simplistic. A more credible scenario would require the slow intrusion of dissolved-silica and bicarbonate-containing groundwater into the salt. This water would then dissolve the salt and, upon reaching the repository, would consist

Fig. 1 Leaching cells

TABLE 1. Experimental Matrix

(Glass Beads - Brine)

Channel No. (N°)	Temperature (°C)	Flow Rate (ml/d)	Leaching (Beads)	Cell Configuration (Rock) (Beads-Rock)
1	25	10	X	
4	25	300	X	
5	25	300	X	
7	25	10		X
8	25	10		X
10	25	300		X
25	25	10		X
29	25	300		X
33	75	10	X	
34	75	10	X	
36	75	300	X	
39	75	10		X
42	75	300		X
43	75	300		X
57	75	10		X
59	75	300		X
65	75	1		X
69	75	1		X

Table 2.

Composition of PNL 76-68 Simulated High Level Waste Glass in wt. %*

SiO ₂	40.0	Pr ₆ O ₁₁	0.53
Na ₂ O	12.5	²³⁷ NpO ₂	0.46
Fe ₂ O ₃	9.6	P ₂ O ₅	0.46
B ₂ O ₃	9.5	Cr ₂ O ₃	0.40
ZnO	5.4	SrO	0.37
²³⁸ UO ₂	4.2	Sm ₂ O ₃	0.32
TiO ₂	3.0	TeO ₂	0.26
MoO ₃	2.2	Y ₂ O ₃	0.21
CaO	2.0	NiO	0.20
ZrO ₂	1.7	Rh ₂ O ₃	0.17
Nd ₂ O ₃	1.65	Rb ₂ O	0.13
CeO ₂	1.19	Eu ₂ O ₃	0.070
RuO ₂	1.07	Gd ₂ O ₃	0.050
Cs ₂ O	1.03	²³⁹ PuO ₂	0.046
BaO	0.56	CdO	0.033
PdO	0.53	Ag ₂ O	0.031
La ₂ O ₃	0.53		

*PNL 3152

of a saturated brine with little, if any, of the characteristics of the original sandstone aquifer groundwater. Because many salt deposits do contain silicate mineral contamination³ as clays and silt, the salt brine could still include dissolved silica. An even more credible breach scenario would require human intrusion through salt-solution mining. Salt-solution mining requires that fresh groundwater be pumped down into a salt bed and the resultant brine is pumped up to the surface where it is processed to recover the salt. Due to the expected presence of silica in the fresh groundwater and likely presence of silicate-mineral contamination in the salt, dissolved silica would be expected in these solution-mined brines as well.

The composition of the synthetic Wipp Brine "B" does not include silica³. Because of the potential for dissolved silica in actual repository brines, we felt compelled to modify the Brine "B" composition to include dissolved silica. Initially, this was done by adding crushed sandstone to the Brine "B" solution; later, silica was added to the brine solution during its preparation. The addition of silica was important because it can affect the dissolution rate of the glass. If the leachant is nearly saturated with respect to dissolved silica before contacting the glass waste-form, it could limit the rate of glass dissolution. The actual effect is not clearly understood.

The Brine "B" composition was further modified by adding bicarbonate ion (groundwaters usually contain bicarbonate) and by diluting the brine to a 20% saturated composition to facilitate flow through the tubing and peristaltic pumps. In the Bead Leach I experiment, an 80% saturated brine was used; this brine caused continual tube clogging and leaking problems which resulted in considerable equipment damage from metal corrosion. The Bead Leach II 20% brine caused no such problems. While

the use of a diluted brine was a necessary experimental compromise, it was a departure from expected repository conditions because it is most likely that the repository would be invaded by a saturated brine rather than a diluted one. In view of these modifications on the leachant used in this section, the results of this experiment should be interpreted with these limitations in mind.

The chemical composition of the brine leachant is shown in Table 3.

Leaching Cells

The leaching cells and contents were assembled following the same procedure used with the basalt channels.¹ Cells with numbers 1, 4, 5, 33, 34, and 36 contained beads only, whereas cells with numbers 7, 8, 10, 39, 42, 43, 65, and 69 contained both beads and rock. In addition, cells with numbers 25, 29, 57, and 59 contained rock only for background or blank corrections. Hereafter, in this section, these cells will be referred to as brine channels or simply channel 1, 4, 5, etc., as a matter of convenience.

Leaching System

The single-pass continuous flow system used with the "brine" channels was the same as that used with the basalt channels referred to in Part I¹ and patterned after the system assembled for the Read Leach I experiment.⁴

The procedures used for the installation of the system, start-up, monitoring during the length of the experiment, sampling schedule, and termination of the leaching phase were the same as those used for the basalt channels.

TABLE 3.

Modified WIPP Brine "B" Chemical Composition

Ion	mg/l
Na ⁺	23,000
K ⁺	4.5
Mg ⁺	3.5
Ca ⁺	188
Sr ⁺	0.3
Cs ⁺	0.2
SiO ₂	26.
Cl ⁻	35,000
SO ₄ ⁼	700
BO ₃ ⁼	2
HCO ₃ ⁼	50
Br ⁻	80

Analytical Procedures

Leachate samples from all channels were analyzed for Pu and Np by alpha spectrometry. The analytical procedures were the same as those described in Part I. Analysis for other glass components was not obtained due to the high concentration of brine in the leachates. When ICP analysis was performed in diluted samples, most of the minor elements in solution were no longer detectable. Therefore, no further attempts were made to obtain data other than Pu and Np on the brine leachates.

Pu and Np were also analyzed in the rock samples using an acid-wash procedure¹ followed by alpha spectrometry. At the conclusion of the leaching phase, each cell containing the crushed sandstone was disassembled by removing four separate successive layers of rock. These layers were labelled A through D with the D layer being farthest away from the beads in the direction of the leachant flow; approximately equal amounts of rock were taken in each layer.

Calculations

Two types of calculations were carried out using the measured Pu and Np concentrations. First, incremental leach rates and cumulative fractions leached were calculated for the fourteen leachate samples analyzed for each channel. Secondly, adsorption calculations were done on the four rock layers for each channel containing crushed rock. The algorithms used for all calculations are shown below.

Leach Rate Calculations

Leach rate data were calculated from the equation:

$$R_1 = \frac{(a_1 V_1)}{(A_0)(SA)}$$

where:

a_i = dpm/cm³ leachate ($\mu\text{g}/\text{cm}^3$ for nonradiochemical analyses)

A_0 = initial activity (dpm/gram glass)

V_i = volume leachate collected per day (cm^3/day)

SA = average geometric surface area of beads (cm^2) = 11.128 cm^2
per 8-bead sample

i = channel number

R_i = grams glass leached/ $\text{cm}^2 \cdot \text{day}$, based on the element or radionuclide analyzed.

Cumulative Fraction Leached Calculations

Cumulative fraction leached, C_{im} , for the i^{th} channel up to and including the m^{th} day, was calculated from the equation:

$$C_{im} = \sum_{n \leq m} \frac{(SA) \bar{R}_{in} \Delta T_n}{W_{oi}}$$

where:

SA = geometric surface area of beads (11.128 cm^2),

W_{oi} = total weight of glass beads in i^{th} cell (grams),

$\bar{R}_{in}$ = average leach rate between n^{th} and $(n-1)^{\text{th}}$ days
(grams glass/ $\text{cm}^2 \text{ day}$), and

ΔT_n = time (days) between n^{th} and $(n-1)^{\text{th}}$ days.

Rock Adsorption Calculations

First, the total amount of radionuclide or element absorbed in each layer of rock was determined by using:

$$N_i = A_i \times V_i$$

where:

A_i = dpm/cm³ or µg/cm³ of "acid wash" for layer i

V_i = volume of "acid wash" for layer i

N_i = total dpm in layer i

Next, F_i , the percent of the total amount leached over 440 days found in layer i was calculated from

$$F_i = \frac{N_i \times 100}{\sum_i (N_i + L)}$$

where:

N_i = total dpm in layer i

L = dpm in the leachate

F_i = percent of the total leached activity in layer i

Results and Discussion

Bead Weights

The glass beads were weighed at the beginning and at the end of the leaching experiment. The weight losses in each cell are recorded in Table 4. It appears that the beads in cells 1, 4, 5, 7, 8 and 10, which were leached at 25°C with either 10 or 300 ml/d, lost less than 1% of their original weight. Next, there is a 1-4% weight loss from cells 33, 34, 39, 65, and 69 (the first three are 10 ml/day and the last two are 1 ml/day) which were leached at 75°C. Finally, the beads in cells 42 and 43 (75°C and 300 ml/day) lost about 10% of their original weight. Weight loss phenomena in any leaching experiment should be treated with caution because reprecipitation of the leached species and/or formation of hydrous silica gels on the surface of the glass beads can occur.

TABLE 4.

Glass Beads Weights (Before and After Leaching)

Channel	Temp. (°C)	Flow Rate (ml)	Original Weight (g)	Final Weight (g)	ΔW (g)	ΔW (%)
1	25	10	2.7685	2.7675	-0.0010	0.036
4	25	300	2.8902	2.8900	-0.0002	0.007
5	25	300	2.8276	2.8308	+0.0032	0.11
7	25	10	2.7602	2.7574	-0.0028	0.10
8	25	10	2.8848	2.8833	-0.0015	0.052
10	25	300	2.8064	2.8063	-0.0001	0.004
33	75	10	2.8189	2.7204	-0.0985	3.49
34	75	10	2.9726	2.8905	-0.0821	2.76
36	75	300	2.8658	2.6467	-0.2191	7.65
39	75	10	2.7158	2.6223	-0.0935	3.44
42	75	300	2.8437	2.5297	-0.3140	11.04
43	75	300	2.8102	2.5214	-0.2888	10.28
65	75	1	2.9009	2.8561	-0.0448	1.54
69	75	1	2.8165	2.7700	-0.0465	1.65

Incremental Leach Rates

The Pu and Np concentrations (dpm/cm^3) in the leachates are given in Tables 5 and 6, respectively. The data pertaining to the beads-rock channels were not blank-corrected since the blank values obtained on the rock-only channels (channels 25, 29, 57, and 59) were essentially background values. The average Pu blank value for 23 leachate samples was 0.0015 cpm compared to a counter background of 0.0013 cpm. The average Np blank value for the same 23 leachate samples was 0.0044 cpm compared to a background value of 0.0041 cpm. The Pu and Np concentrations were used to calculate leach rates at the prescribed collection days during the experiment. The leach rate values are shown in Tables 7 and 8. In both the concentration tables and the incremental leach rate tables, the number underneath the measured value is the percent uncertainty due to counting statistics only. In these Pu tables, the letter C indicates that the value was obtained from a leachate sample that was chemically processed to separate Pu from Np as opposed to the majority of the results which were obtained from counting a 100 μl aliquot of the leachate for both Pu and Np.

The incremental leach rates for channels 1, 4, 5, 33, 34, and 36 (beads only) are the actual leach rates since these channels did not contain rock. These are shown in Figs. 2 and 3 for Pu and Np, respectively. On the other hand, the incremental leach rates for channels 7, 8, 10, 39, 42, 43, 65, and 69 (beads and rock channels) are pseudo-leach rates in the sense that these values reflect only a fraction of the leached species because the remaining fraction is adsorbed on the surfaces of the crushed sandstone. These leach rates are shown in Figs. 4 and 5 for Pu and Np, respectively. Identification of channels is shown with numbers on the right hand side of each graph.

Table 5

BEADLEACH 11

TABLE OF PLUTONIUM CONCENTRATION (DPH/CM3)

CHAN TEMP FLOW SAMPLING DAYS

	1	2	3	6	11	20	37	70	120	181	213
1 COLD MED	2.51E+01	3.24E+01	2.02E+01	6.93E+00	7.26E+00	1.18E+01	1.82E+00	7.89E+01	1.66E+01	3.60E+01	2.88E+01
4 COLD FAST	1.4E+00*	1.4E+00	1.4E+00	1.7E+00	2.4E+00	1.5E+00	5.1E+00	6.5E+00	1.2E+01	9.6E+00	1.1E+01
5 COLD FAST	3.41E+00	1.34E+00	5.17E+01	1.64E+01	1.24E+01	9.47E+01	1.31E+01	3.41E+02	6.6E+02	4.88E+02	3.00E+02
7 COLD MED	3.7E+00	7.0E+00	C 9.4E+01	1.4E+01	1.5E+01	C 2.3E+00	1.6E+01	C 3.9E+00	C 2.8E+00	2.0E+01	2.1E+01
8 COLD MED	5.14E+00	1.05E+00	4.95E+01	3.53E+01	1.11E+01	1.49E+01	3.41E+02	6.70E+02	1.14E+01	2.03E+02	0.
10 COLD FAST	2.8E+00	7.9E+00	8.7E+00	1.0E+01	1.6E+01	1.3E+00	2.0E+01	1.8E+01	C 3.9E+00	2.4E+01	3.3E+01
25 COLD MED	4.62E+00	7.55E+00	7.13E+00	2.54E+00	1.92E+00	1.60E+00	7.31E+01	2.51E+01	2.01E+02	4.50E+01	1.11E+01
29 COLD FAST	3.2E+00	2.9E+00	2.6E+00	4.3E+00	4.3E+00	5.0E+00	7.6E+00	1.1E+01	2.4E+01	8.5E+00	2.0E+01
33 HOT MED	4.55E+00	1.12E+01	1.85E+01	3.89E+00	2.26E+00	2.78E+00	8.15E+01	1.28E+01	1.06E+01	1.66E+01	3.62E+01
34 HOT MED	3.2E+00	2.5E+00	C 2.6E+01	3.0E+00	4.2E+00	C 7.7E+01	7.4E+00	C 2.8E+00	C 3.0E+00	1.3E+01	1.3E+01
36 HOT FAST	2.53E+00	5.83E+01	4.32E+01	7.30E+02	8.25E+02	1.27E+01	0.	2.87E+02	1.28E+01	9.01E+02	4.09E+02
39 HOT MED	4.1E+00	6.8E+00	1.7E+01	1.7E+01	1.7E+01	1.3E+01	2.7E+01	2.2E+01	1.5E+01	1.6E+01	2.2E+01
42 HOT FAST	0.	8.37E+02	0.	0.	0.	0.	0.	6.14E+02	3.57E+02	2.26E+02	0.
43 HOT FAST	0.	1.8E+01	0.	0.	0.	0.	0.	2.6E+01	2.0E+01	4.1E+01	0.
44 HOT FAST	0.	1.10E+01	0.	0.	0.	0.	0.	4.04E+02	2.82E+02	5.66E+02	0.
45 HOT FAST	0.	1.5E+01	0.	0.	0.	0.	0.	3.2E+01	2.2E+01	2.7E+01	0.
46 HOT FAST	1.72E+01	5.94E+00	4.66E+00	1.04E+00	1.90E+01	9.87E+02	4.63E+02	4.62E+02	9.66E+02	2.07E+01	3.23E+01
47 HOT FAST	1.9E+03	2.1E+00	C 4.6E+01	6.1E+00	1.3E+01	C 3.1E+00	2.2E+01	C 4.6E+00	C 3.1E+00	1.3E+01	1.3E+01
48 HOT FAST	1.62E+01	6.68E+00	3.26E+00	1.24E+00	2.68E+01	5.02E+02	5.69E+02	7.35E+02	1.90E+01	2.36E+02	4.95E+02
49 HOT FAST	2.1E+00	1.7E+00	3.1E+00	5.6E+00	1.1E+01	2.1E+01	1.9E+01	2.1E+01	1.3E+01	2.1E+01	1.1E+01
50 HOT FAST	1.1E+00	2.98E+01	2.42E+01	3.24E+01	3.08E+01	2.93E+01	1.01E+01	7.80E+02	7.05E+01	6.21E+01	8.88E+01
51 HOT FAST	8.8E+00	8.3E+00	1.2E+01	1.0E+01	1.1E+01	9.5E+00	1.6E+01	1.7E+01	7.2E+00	7.4E+00	5.2E+00
52 HOT FAST	5.51E+01	1.69E+01	2.35E+01	1.13E+01	5.25E+02	4.48E+02	0.	2.16E+02	8.10E+03	2.60E+02	1.76E+01
53 HOT FAST	C 3.0E+00	4.3E+00	C 9.0E+00	C 3.0E+00	C 5.4E+00	C 5.4E+00	3.2E+01	2.4E+01	2.8E+01	2.4E+01	1.4E+01
54 HOT FAST	1.09E+00	1.16E+01	1.03E+01	9.38E+02	1.33E+01	2.60E+01	1.64E+01	1.16E+01	6.6E+01	7.62E+01	1.61E+00
55 HOT FAST	5.0E+00	1.7E+01	C 2.4E+00	1.7E+01	1.4E+01	C 1.4E+00	C 2.1E+00	C 7.9E+01	C 7.9E+01	7.9E+00	4.4E+00
56 HOT FAST	8.71E+01	1.03E+01	8.14E+02	1.62E+01	2.01E+01	1.86E+01	1.59E+01	9.18E+02	9.02E+01	8.20E+01	2.62E+00
57 HOT MED	3.5E+00	1.8E+01	1.7E+01	1.4E+01	1.3E+01	1.2E+01	0.4E+01	1.7E+01	7.5E+00	7.5E+00	3.4E+00
58 HOT MED	0.	6.2E+02	0.	0.	0.	0.	0.	3.8E+01	1.05E+02	4.16E+02	0.
59 HOT FAST	0.	2.9E+01	0.	0.	0.	0.	0.	1.8E+01	5.8E+01	3.2E+01	0.
60 HOT FAST	0.	8.21E+02	0.	0.	0.	0.	0.	4.01E+03	6.99E+02	3.68E+02	0.
61 HOT FAST	0.	2.4E+01	0.	0.	0.	0.	0.	1.0E+02	1.4E+01	3.3E+01	0.
1- 10											
65+ HOT SLOW	3.59E+01	0.	0.	0.	2.62E+02	7.35E+02	2.01E+02	1.49E+02	7.13E+02	1.23E+02	1.03E+01
66+ HOT SLOW	C 1.9E+00	0.	0.	0.	C 8.5E+00	C 1.2E+01	C 1.3E+01	C 1.2E+01	C 6.7E+00	2.5E+01	C 8.6E+00
67+ HOT SLOW	C 3.69E+02	0.	0.	0.	9.26E+01	5.31E+02	6.90E+02	1.74E+01	1.30E+01	0.	3.40E+01
68+ HOT SLOW	C 5.2E+00	0.	0.	0.	C 1.9E+00	C 4.7E+00	C 7.9E+00	C 2.9E+00	C 4.0E+00	3.0E+01	C 2.5E+00

Note: * Indented value under each concentration value is the percent uncertainty based on counting statistics

† Collections were over a cumulative period in order to obtain enough sample for analysis.

Table 5 (continued)

BEADLEACH II

TABLE OF PLUTONIUM CONCENTRATION (DPM/CM3)

CHAN TEMP FLOW SAMPLING DAYS			
		342	435
1	COLD MED 0.	2.64E-01	3.00E-01
4	COLD FAST 0.	1.4E-01	1.2E+01
5	COLD FAST 0.	5.47E-02	1.31E-01
7	COLD MED 0.	2.6E+01	1.5E+01
8	COLD MED 0.	7.76E-02	5.63E-02
10	COLD FAST 0.	2.4E+01	1.9E+01
25	COLD MED 0.	1.06E-01	1.98E-01
29	COLD FAST 0.	2.0E+01	1.2E+01
33	HOT MED 0.	4.11E-01	5.48E-01
34	HOT MED 0.	1.2E+01	7.8E+00
35	HOT FAST 0.	2.52E-02	4.69E-02
39	HOT MED 0.	2.1E+01	1.9E+01
42	HOT FAST 0.	7.79E-02	1.98E-01
43	HOT FAST 0.	2.5E+01	1.6E+01
57	HOT MED 0.	3.78E-02	6.39E-02
59	HOT FAST 0.	2.8E+01	2.1E+01
65	HOT SLOW 0.	3.28E+00	2.46E+00
69	HOT SLOW 0.	3.5E+00	4.0E+00
73	HOT SLOW 0.	1.33E+00	1.97E+00
77	HOT SLOW 0.	5.1E+00	4.5E+00
81	HOT SLOW 0.	9.97E-01	4.21E-01
85	HOT SLOW 0.	5.8E+00	8.9E+00
89	HOT SLOW 0.	1.73E+00	9.37E-01
93	HOT SLOW 0.	C 9.4E-01	C 6.9E-01
97	HOT SLOW 0.	2.41E+00	1.35E+00
101	HOT SLOW 0.	4.0E+00	5.7E+00
105	HOT SLOW 0.	3.44E+00	5.50E-01
109	HOT SLOW 0.	3.4E+00	8.8E+00
113	HOT SLOW 0.	7.89E-02	6.50E-02
117	HOT SLOW 0.	1.7E+01	2.5E+01
121	HOT SLOW 0.	0.	5.03E-02
125	HOT SLOW 0.	0.	2.5E+01
		310-342	408-435
65	HOT SLOW 0.	1.51E-01	1.54E-01
69	HOT SLOW 0.	C 1.9E+00	C 1.8E+00
73	HOT SLOW 0.	8.07E-02	9.12E-02
77	HOT SLOW 0.	C 3.9E+00	C 3.3E+00

Table 6

BEADLEACH 11

TABLE OF NEPTUNIUM CONCENTRATION (DPH/CM3)

CHAN TEMP: FLOW SAMPLING DAYS

	1	2	3	6	11	20	37	70	120	151	213
1 COLD: MED	1.42E+01*	3.59E+01	2.78E+01	7.89E+00	3.94E+00	3.41E+00	1.66E+00	2.56E+00	1.45E+00	8.03E-01	1.67E+00
4 COLD: FAST	1.9E+00	1.3E+00	1.2E+00	2.5E+00	3.2E+00	3.3E+00	5.1E+00	3.6E+00	4.7E+00	6.3E+00	4.7E+00
5 COLD: FAST	3.34E+00	1.30E+00	5.23E-01	1.86E-01	2.00E-01	6.59E-02	2.33E-01	3.39E-02	6.94E-02	1.03E-01	8.55E-02
7 COLD: MED	3.7E+00	6.8E+00	1.0E+00	1.1E+01	1.1E+01	C 3.1E+00	1.1E+01	C 5.7E+00	C 3.4E+00	1.2E+01	1.2E+01
8 COLD: MED	4.16E+00	1.67E+00	1.05E+00	5.09E-01	2.29E-01	1.42E-01	1.32E-01	4.27E-02	1.05E-01	1.8E-02	3.70E-02
10 COLD: FAST	3.1E+00	8.1E+00	5.9E+00	7.7E+00	1.0E+01	1.1E+01	1.1E+01	1.4E+01	1.2E+01	1.5E+01	1.5E+01
25 COLD: MED	4.13E+00	8.47E+00	1.75E+01	8.81E+00	4.64E+00	2.57E+00	2.08E+00	3.04E+00	2.63E+00	1.07E+00	1.19E+00
29 COLD: FAST	3.3E+00	2.7E+00	1.6E+00	2.3E+00	2.7E+00	3.9E+00	4.6E+00	3.4E+00	3.6E+00	4.1E+00	7.1E+00
33 HOT: MED	4.88E+00	1.22E+01	2.19E+01	8.63E+00	4.06E+00	3.89E+00	2.03E+00	6.13E+00	4.46E+00	9.70E-01	1.74E+00
34 HOT: MED	3.1E+00	2.3E+00	C 3.3E-01	2.0E+00	3.1E+00	C 9.4E-01	4.7E+00	C 7.6E-01	C 1.0E+00	6.0E+00	6.0E+00
36 HOT: FAST	2.68E+00	1.81E+00	8.53E-01	2.82E-01	8.52E-02	1.89E-01	7.64E-02	2.16E-01	6.43E-02	4.62E-02	2.07E-02
39 HOT: MED	3.9E+00	5.0E+00	6.2E+00	9.2E+00	1.2E+01	9.7E+00	1.2E+01	1.0E+01	1.3E+01	1.4E+01	1.6E+01
57 HOT: MED	0.	2.04E-01	0.	0.	0.	0.	0.	1.52E-01	1.53E-01	1.06E-01	0.
59 HOT: FAST	0.	1.1E+01	0.	0.	0.	0.	0.	1.17E-01	1.0E-01	1.80E-01	0.
65+ HOT: MED	0.	1.0E+01	0.	0.	0.	0.	0.	1.9E+01	1.2E+01	1.5E+01	0.
69+ HOT: MED	8.35E+01	1.3E+02	1.93E+02	1.37E+02	6.31E+01	8.19E+01	2.89E+01	9.30E+01	1.03E+02	9.57E+01	8.58E+01
33 HOT: MED	8.5E-01	4.4E-01	C 1.1E-01	5.4E-01	8.0E-01	C 1.7E-01	1.3E+00	C 1.6E-01	C 1.9E-01	6.6E-01	8.4E-01
34 HOT: MED	6.80E+01	1.42E+02	1.03E+02	1.60E+02	8.28E+01	5.93E+01	1.98E+01	6.09E+01	9.68E+01	1.69E+00	1.10E+02
36 HOT: FAST	1.0E+00	4.9E-01	5.5E-01	5.1E-01	7.0E-01	8.9E-01	1.4E+00	9.5E-01	6.6E-01	4.2E-00	7.8E-01
39 HOT: MED	1.17E+01	1.70E+01	1.65E+01	1.72E+01	1.38E+01	1.72E+01	1.03E+01	1.28E+01	1.44E+01	1.24E+01	1.28E+01
42 HOT: FAST	1.6E+00	1.2E+00	1.6E+00	1.5E+00	1.8E+00	1.3E+00	1.8E+00	1.7E+00	1.6E+00	1.7E+00	1.7E+00
43 HOT: FAST	2.06E+01	3.20E+01	3.72E+01	3.84E+01	3.87E+01	4.65E+01	4.32E+01	5.73E+01	7.88E+01	7.62E+01	7.17E+01
57 HOT: MED	1.10E+01	1.2E+00	1.0E+00	1.0E+00	9.8E-01	8.3E-01	9.5E-01	8.5E-01	7.9E-01	7.5E-01	8.1E-01
59 HOT: FAST	1.1E+00	1.66E+01	1.84E+01	1.42E+01	1.47E+01	1.89E+01	1.68E+01	8.87E+00	2.02E+01	1.65E+01	2.01E+01
65+ HOT: SLOW	1.8E+00	1.7E+00	C 1.8E-01	1.7E+00	1.6E+00	C 2.2E-01	1.0E+00	C 3.4E-01	C 1.8E-01	1.7E+00	1.3E+00
69+ HOT: SLOW	1.31E+01	1.53E+01	1.68E+01	1.82E+01	1.68E+01	1.34E+01	2.01E+01	6.44E+00	2.07E+01	1.35E+01	1.54E+01
65+ HOT: SLOW	1.5E+00	1.5E+00	1.5E+00	1.5E+00	1.5E+00	1.5E+00	1.4E+00	1.5E+00	1.5E+00	1.5E+00	1.5E+00
57 HOT: MED	0.	1.23E-01	0.	0.	0.	0.	0.	1.67E-01	1.19E-01	2.21E-01	0.
59 HOT: FAST	0.	2.0E+01	0.	0.	0.	0.	0.	8.6E+00	1.7E+01	1.4E+01	0.
65+ HOT: SLOW	0.	2.10E-01	0.	0.	0.	0.	0.	2.93E-01	3.15E-01	1.84E-01	0.
69+ HOT: SLOW	0.	1.5E+01	0.	0.	0.	0.	0.	1.2E+01	6.7E+00	1.6E+01	0.
1-10											
11-20											
21-30											
31-50											
51-80											
81-120											
121-150											
183-213											
65+ HOT: SLOW	2.91E+02	0.	0.	0.	6.51E+01	1.05E+02	1.47E+02	1.48E+02	2.94E+02	1.69E+02	2.74E+02
69+ HOT: SLOW	3.5E+01	0.	0.	0.	1.1E+00	6.9E-01	4.9E-01	7.0E-01	3.9E-01	4.9E-01	4.0E-01
65+ HOT: SLOW	1.8E+02	0.	0.	0.	1.79E+02	1.39E+02	1.78E+02	1.69E+02	2.77E+02	1.94E+02	2.15E+02
69+ HOT: SLOW	4.4E-01	0.	0.	0.	6.3E-01	7.1E-01	4.7E-01	6.1E-01	3.6E-01	4.6E-01	4.7E-01

Note: * Indented value under each concentration value is the percent uncertainty based on counting statistics
 Collections were over a cumulative period in order to obtain enough sample for analysis.

Table 6 (continued)

BEADLEACH II

TABLE OF NEPTUNIUM CONCENTRATION (DPM/CM3)

CHAN TEMP FLOW SAMPLING DAYS

		280	342	435
1	COLD MED	0.	7.47E-01	6.45E-01
		0.	6.4E+00	7.0E+00
4	COLD FAST	0.	3.38E-02	3.01E-01
		0.	1.9E+01	9.2E+00
5	COLD FAST	0.	7.09E-02	1.36E-01
		0.	1.7E+01	1.2E+01
7	COLD MED	0.	4.39E-01	7.63E-01
		0.	1.0E+01	6.4E+00
8	COLD MED	0.	6.53E-01	7.74E-01
		0.	6.4E+00	6.2E+00
10	COLD FAST	0.	6.12E-02	1.92E-01
		0.	1.2E+01	1.0E+01
23	COLD MED	0.	1.46E-01	1.20E-01
		0.	1.8E+01	1.8E+01
29	COLD FAST	0.	1.77E-01	2.55E-01
		0.	1.6E+01	1.2E+01
33	HOT MED	0.	1.51E+02	1.70E+02
		0.	5.2E-01	4.8E-01
34	HOT MED	0.	1.45E+02	1.21E+02
		0.	5.0E-01	5.8E-01
36	HOT FAST	0.	1.17E+01	6.33E+00
		0.	1.7E+00	2.1E+00
39	HOT MED	0.	9.37E-01	1.31E+02
		0.	7.0E-01	5.6E-01
42	HOT FAST	0.	1.43E+01	1.14E+01
		0.	1.7E+00	2.0E+00
43	HOT FAST	0.	1.67E+01	4.77E+00
		0.	1.6E+00	3.1E+00
57	HOT MED	0.	1.54E-01	1.22E-01
		0.	1.2E+01	1.8E+01
59	HOT FAST	0.	0.	1.48E-01
		0.	0.	1.5E+01
65	HOT SLOW	246-280	310-342	408-435
		0.	2.35E+02	3.59E+02
		0.	3.7E-01	3.2E-01
69	HOT SLOW	0.	1.88E+02	7.07E+02
		0.	4.3E-01	2.1E-01

Table 7

BEADLEACH II
PLUTONIUM LEACH RATE (G/CM2.DAY)

CHAN TEMP FLOW SAMPLING DAYS

	1	2	3	6	11	20	37	70	120	151	213
1 COLD MED	1.32E-06 1.4E+00	* 1.35E-06 1.4E+00	4.49E-07 1.4E+00	1.20E-07 2.7E+00	1.23E-07 2.4E+00	2.10E-07 1.6E+00	3.29E-08 5.1E+00	1.17E-08 6.5E+00	3.37E-08 1.2E+01	6.72E-09 9.5E+00	5.04E-09 1.1E+01
4 COLD FAST	1.71E-06 3.7E+00	6.91E-07 7.0E+00	2.61E-07 9.4E-01	0.18E-08 1.4E+01	6.05E-08 1.5E+01	4.73E-08 2.3E+00	5.52E-08 1.6E+01	1.65E-08 3.9E+00	3.23E-08 2.0E+01	2.10E-08 2.0E+01	1.49E-09 2.1E+01
5 COLD FAST	2.71E-06 2.8E+00	5.40E-07 7.9E+00	2.50E-07 8.7E+00	1.76E-07 1.0E+01	5.41E-08 1.6E+01	7.39E-08 1.3E+01	1.70E-08 2.0E+01	3.05E-08 1.0E+01	5.68E-08 3.9E+00	1.01E-08 2.4E+01	0 3.3E+01
7 COLD MED	8.23E-08 3.2E+00	1.38E-07 2.8E+00	1.29E-07 2.6E+00	4.69E-08 4.3E+00	3.46E-08 4.3E+00	3.04E-08 5.0E+00	1.35E-08 7.8E+00	4.53E-09 1.1E+01	3.40E-10 2.4E+01	5.26E-09 8.5E+00	6.25E-10 2.0E+01
8 COLD MED	8.26E-08 3.2E+00	2.10E-07 2.5E+00	3.39E-07 2.6E-01	7.49E-08 3.0E+00	4.02E-08 4.2E+00	5.08E-08 7.7E-01	1.47E-08 7.4E+00	7.79E-10 2.8E+00	1.90E-09 3.0E+00	3.32E-09 1.3E+01	5.65E-09 1.3E+01
10 COLD FAST	1.78E-06 4.1E+00	3.26E-07 8.8E+00	2.33E-07 8.9E+00	3.83E-08 1.7E+01	4.18E-08 1.7E+01	6.51E-08 1.3E+01	0 2.7E+01	8.60E-09 2.2E+01	6.51E-08 1.5E+01	4.61E-08 1.6E+01	2.11E-08 2.2E+01
33 HOT MED	2.92E-07 1.9E+00	9.55E-08 2.1E+00	7.95E-08 4.6E-01	1.72E-08 6.1E+00	2.91E-09 1.9E+01	1.62E-08 3.1E+00	7.65E-10 2.2E+01	7.39E-10 4.6E+00	1.57E-09 3.1E+00	8.25E-09 1.3E+01	5.12E-09 1.3E+01
34 HOT MED	3.08E-07 2.1E+00	1.55E-07 1.7E+00	5.75E-08 3.1E+00	2.27E-08 5.6E+00	4.99E-09 1.1E+01	8.87E-10 2.1E+01	* 0.01E-09 1.9E+01	7.05E-10 2.1E+01	3.28E-09 1.3E+01	4.12E-10 2.1E+01	8.19E-09 1.1E+01
36 HOT FAST	5.92E-07 4.8E+00	1.54E-07 8.3E+00	1.21E-07 1.2E+01	1.62E-07 1.0E+01	1.53E-07 1.1E+01	1.48E-07 9.5E+00	4.00E-08 1.5E+01	3.18E-08 1.7E+01	3.42E-07 1.2E+00	2.96E-07 7.4E+00	4.30E-07 6.2E+00
39 HOT MED	1.15E-08 3.0E+00	3.11E-09 4.3E+00	4.73E-09 3.0E+00	2.06E-09 3.0E+00	9.22E-10 5.1E+00	8.42E-10 5.4E+00	0 3.2E+01	1.61E-10 2.4E+01	1.29E-10 2.6E+01	4.11E-10 2.4E+01	3.34E-09 1.4E+01
42 HOT FAST	5.46E-07 5.0E+00	5.91E-08 1.7E+01	4.96E-08 2.4E+00	4.52E-08 1.7E+01	6.43E-08 1.4E+01	1.25E-07 1.4E+00	7.87E-08 1.4E+01	5.32E-08 2.1E+00	4.51E-07 7.9E-01	8.59E-07 7.7E+00	8.59E-07 4.4E+00
43 HOT FAST	4.30E-07 5.9E+00	5.19E-08 1.8E+01	4.01E-08 1.7E+01	7.92E-08 1.4E+01	9.79E-08 1.3E+01	9.05E-08 1.2E+01	7.69E-08 1.4E+01	4.36E-08 1.7E+01	4.33E-07 1.7E+00	3.91E-07 7.5E+00	1.34E-06 3.4E+00
	1- 10				11- 20	21- 30	31- 50	51- 80	81-120	121-150	161-213
65 [†] HOT SLOW	5.17E-10 1.9E+00	0.	0.	0.	4.55E-11 8.5E+00	1.31E-10 1.2E+01	3.63E-11 1.3E+01	2.65E-11 1.2E+01	8.81E-11 6.7E+00	5.61E-11 2.5E+01	1.75E-10 8.6E+00
69 [†] HOT SLOW	1.67E-10 5.2E+00	0.	0.	0.	1.57E-09 1.9E+00	1.00E-10 4.7E+00	1.25E-10 7.9E+00	2.59E-10 2.9E+00	2.12E-10 4.0E+00	0.	7.62E-10 2.5E+00

Notes: * Indented value under each concentration value is the percent uncertainty based on counting statistics.
[†] Collections were over a cumulative period in order to obtain enough sample for analysis.

Table 7 (continued)

BEADLEACH II

PLUTONIUM LEACH RATE (G/CM².DAY)

CHAN TEMP FLOW SAMPLING DAYS

		280	342	435
1	COLD MED	0. 0.	4.91E-03 1.4E+01	5.58E-09 1.2E+01
4	COLD FAST	0. 0.	3.01E-08 2.6E+01	6.28E-08 1.5E+01
5	COLD FAST	0. 0.	3.99E-08 2.4E+01	2.35E-08 1.9E+01
7	COLD MED	0. 0.	2.17E-09 2.0E+01	3.80E-09 1.2E+01
8	COLD MED	0. 0.	7.21E-09 1.2E+01	9.35E-09 7.9E+00
10	COLD FAST	0. 0.	1.15E-08 2.1E+01	2.25E-08 1.9E+01
33	HOT MED	0. 0.	5.61E-08 3.5E+00	4.15E-08 4.0E+00
34	HOT MED	0. 0.	2.43E-08 5.1E+00	3.55E-08 4.5E+00
36	HOT FAST	0. 0.	5.20E-07 5.8E+00	1.39E-07 8.9E+00
39	HOT MED	0. 0.	3.55E-08 C 9.4E-01	1.91E-08 C 8.9E-01
42	HOT FAST	0. 0.	1.25E-08 4.0E+00	8.38E-07 5.7E+00
45	HOT FAST	0. 0.	1.90E-08 9.4E+00	3.19E-07 8.6E+00
		246-280	310-342	408-435
65	HOT SLOW	0. 0.	7.47E-10 C 1.6E+00	3.79E-10 C 1.8E+00
69	HOT SLOW	0. 0.	1.28E-10 C 3.9E+00	1.15E-10 C 3.3E+00

Table 8

BEADLEACH II

NEPTUNIUM LEACH RATE (G/CM2.DAY)

CHAN TEMP FLOW SAMPLING DAYS

		1	2	3	5	11	20	37	70	120	151	213
1	COLD MED	6.52E-06 1.9E+00*	1.90E-05 1.3E+00	5.41E-06 1.2E+00	1.19E-06 2.5E+00	5.86E-07 3.2E+00	5.39E-07 3.3E+00	2.69E-07 5.1E+00	3.31E-07 3.6E+00	2.90E-07 4.7E+00	1.91E-07 6.3E+00	2.56E-07 4.7E+00
4	COLD FAST	1.45E-05 3.7E+00	5.89E-06 6.8E+00	2.91E-06 C 1.0E+00	9.14E-07 1.1E+01	9.60E-07 1.1E+01	2.87E-07 C 3.1E+00	1.01E-06 1.1E+01	1.42E-07 C 5.7E+00	2.99E-07 C 3.4E+00	4.48E-07 1.2E+01	3.70E-07 1.2E+01
5	COLD FAST	1.92E-05 3.1E+00	7.50E-06 6.1E+00	4.65E-06 5.9E+00	2.22E-06 7.7E+00	9.77E-07 1.0E+01	6.17E-07 1.1E+01	5.74E-07 1.1E+01	1.69E-07 1.4E+01	4.55E-07 1.2E+01	6.75E-08 1.5E+01	1.61E-07 1.5E+01
7	COLD MED	5.43E-07 3.3E+00	1.36E-06 2.7E+00	2.78E-06 1.6E+00	1.42E-06 2.3E+00	7.30E-07 2.7E+00	4.16E-07 3.9E+00	3.98E-07 4.6E+00	4.79E-07 3.4E+00	3.98E-07 3.0E+00	2.01E-07 4.1E+00	5.88E-08 7.1E+00
8	COLD MED	7.74E-07 3.1E+00	2.00E-06 2.3E+00	3.51E-06 C 3.3E-01	1.49E-06 2.0E+00	6.30E-07 3.1E+00	6.20E-07 C 9.4E-01	3.19E-07 4.7E+00	3.25E-07 C 7.6E-01	7.04E-07 C 1.0E+00	1.52E-07 6.0E+00	2.47E-07 8.0E+00
10	COLD FAST	1.64E-05 3.9E+00	8.83E-06 5.0E+00	4.01E-06 6.2E+00	1.29E-06 9.2E+00	3.77E-07 1.2E+01	8.38E-07 9.7E+00	3.39E-07 1.2E+01	4.33E-07 1.0E+01	2.65E-07 1.3E+01	2.07E-07 1.4E+01	9.33E-08 1.6E+01
33	HOT MED	1.24E-05 8.5E-01	1.90E-05 4.4E-01	2.87E-05 C 1.1E-01	1.97E-05 5.4E-01	8.44E-06 8.0E-01	1.18E-05 C 1.7E-01	4.17E-06 1.3E+00	1.30E-05 C 1.6E-01	1.43E-05 C 1.9E-01	1.31E-05 6.6E-01	1.19E-05 8.4E-01
34	HOT MED	1.13E-05 1.0E+00	2.22E-05 4.3E-01	1.58E-05 5.5E-01	2.56E-05 5.1E-01	1.25E-05 7.9E-01	9.16E-06 8.9E-01	3.06E-07 4.4E+00	5.10E-06 7.1E+00	1.48E-05 6.6E-01	2.08E-07 4.2E+00	1.59E-05 7.8E-01
36	HOT FAST	5.16E-05 1.6E+00	7.68E-05 1.6E+00	7.28E-05 1.6E+00	7.49E-05 1.5E+00	6.02E-05 1.8E+00	7.45E-05 1.3E+00	4.39E-05 1.8E+00	4.69E-05 1.6E+00	5.08E-05 1.6E+00	5.23E-05 1.7E+00	5.27E-05 1.7E+00
39	HOT MED	3.75E-06 1.2E+00	5.13E-06 1.2E+00	6.54E-06 1.0E+00	6.08E-06 1.0E+00	5.94E-06 9.8E-01	7.68E-06 8.9E-01	6.90E-06 9.9E-01	8.75E-06 8.5E-01	1.09E-05 7.3E-01	1.05E-05 7.5E-01	1.19E-05 8.1E-01
42	HOT FAST	4.81E-05 1.6E+00	7.40E-05 1.7E+00	7.78E-05 C 1.8E-01	5.99E-05 1.7E+00	6.18E-05 1.6E+00	6.64E-05 C 2.2E-01	7.10E-05 1.9E+00	3.57E-05 C 3.4E-01	8.26E-05 1.7E+00	6.78E-05 1.7E+00	8.34E-05 1.7E+00
43	HOT FAST	5.66E-05 1.5E+00	6.73E-05 1.8E+00	7.12E-05 1.5E+00	7.76E-05 1.5E+00	7.17E-05 1.5E+00	5.70E-05 1.6E+00	8.52E-05 1.4E+00	2.67E-05 1.4E+00	8.68E-05 1.5E+00	5.61E-05 1.9E+00	6.39E-05 1.5E+00

1- 10

11- 20

21- 30

31- 50

51- 60

61-120

121-150

163-213

55 [†]	HOT SLOW	4.37E-06 3.5E-01	0. 0.	0. 0.	0. 0.	9.18E-07 1.1E+00	1.63E-06 6.9E-01	2.31E-06 4.9E-01	2.41E-06 7.0E-01	3.18E-06 3.3E-01	6.75E-06 4.9E-01	4.08E-06 4.0E-01
69 [†]	HOT SLOW	2.52E-06 4.4E-01	0. 0.	0. 0.	0. 0.	2.66E-06 6.3E-01	2.29E-06 7.1E-01	2.83E-06 4.7E-01	2.19E-06 6.1E-01	3.95E-06 3.6E-01	3.29E-06 4.6E-01	4.20E-06 4.7E-01

Notes: * Indented value under each concentration value is the percent uncertainty based on counting statistics.

[†] Collections were over a cumulative period in order to obtain enough sample for analysis.

Table 8 (continued)

BEADLEACH II

NEPTUNIUM LEACH RATE (G/GM2.DAY)

CHAN TEMP FLOW SAMPLING DAYS

	280	342	435
1 COLD MED	0. 0.	1.21E-07 8.4E+00	1.37E-07 7.0E+00
4 COLD FAST	0. 0.	1.62E-07 1.9E+01	1.26E-06 9.2E+00
5 COLD FAST	0. 0.	3.19E-07 1.7E+01	4.97E-07 1.2E+01
7 COLD MED	0. 0.	7.83E-06 1.0E+01	1.26E-07 6.4E+00
8 COLD MED	0. 0.	1.31E-07 8.4E+00	1.19E-07 6.2E+00
10 COLD FAST	0. 0.	3.22E-07 1.2E+01	8.08E-07 1.0E+01
33 HOT MED	0. 0.	2.26E-05 5.2E-01	2.81E-05 4.8E-01
34 HOT MED	0. 0.	2.31E-05 5.0E-01	1.90E-05 5.8E-01
36 HOT FAST	0. 0.	5.32E-05 1.7E+00	2.40E-05 2.1E+00
39 HOT MED	0. 0.	1.79E-05 7.0E-01	2.33E-05 5.6E-01
42 HOT FAST	0. 0.	6.50E-05 1.7E+00	5.17E-05 2.0E+00
43 HOT FAST	0. 0.	8.05E-05 1.6E+00	2.42E-05 3.1E+00
	246-280	310-342	408-435
65 HOT SLOW	0. 0.	1.01E-05 3.7E-01	7.72E-06 3.2E-01
69 HOT SLOW	0. 0.	3.62E-06 4.3E-01	7.79E-06 2.1E-01

Fig. 2. Glass-Brine Leachant

Fig. 3. Glass-Brine Leachant

Fig. 4. Glass-Brine Leachant

Fig. 5. Glass-Brine Leachant

The Pu leach rate behavior is not straightforward. In the case of the "rock-absent" channels, Fig. 2, there appears to be flow rate dependence both at 25°C (solid line) and at 75°C (dotted line); 300 ml/d gives higher leach rates than 10 ml/d. After the first 50 days, the leach rates are somewhat constant except for the 10 ml/d channels (33 and 34); the overall range is from 10^{-9} g/cm²·d to about 5×10^{-7} g/cm²·d except during the initial part of the experiment where some leach rates are on the order of $1-3 \times 10^{-6}$ g/cm²·d for the 25°C - 10 ml/d channels (1, 4, 5). There is not a clear definition of temperature dependence particularly at the medium flow rate of 10 ml/d. At 300 ml/d, the leach rates are higher at 75°C than at 25°C by about one order of magnitude after day 100.

The Pu leach rate patterns obtained are qualitatively similar to those found in the Bead Leach I experiment⁴; however, the quantitative values of the incremental leach rates are lower in this experiment. This probably reflects the differences in chemical composition between the two brines used and, in particular, is consistent with the reduction in glass dissolution rate to be expected from increased levels in dissolved SiO₂ in the leachant. The brine used in the Bead Leach I experiment contained no SiO₂ whereas the one used in Bead Leach II contained 26 ppm SiO₂.

In the case of the "rock present" channels, Fig. 4, the flow rate effect is more obvious. At 75°C, the apparent leach rates are 2-4 orders of magnitude higher at the fast flow than at the medium or slow flows; the range is from 10^{-10} g/cm²·d to 10^{-6} g/cm²·d. Likewise at 25°C, the leach rates are higher at the fast flow than at the medium flow, but the range is narrower, about one order of magnitude. There is a clear temperature effect on the leach rates at the fast flow, but this effect is less obvious at the medium flow. This variation in leach rates, shown by the

behavior of Pu in the presence or absence of rock, may be due to the tendency of Pu to be readily adsorbed on surfaces and a combined effect of both temperature and flow rate on leaching and sorption.

The Np leach rate behavior is more clearly defined than the Pu as shown in Figs. 3 and 5. In the case of the "rock-absent" channels, the leach rates are higher at 75°C than at 25°C, thus showing a temperature effect. There is also a flow rate effect at 75°C, 300 ml/d > 10 ml/d, but this effect is absent at 25°C. The leach rates range from about 10^{-7} g/cm²·d at 25°C to about 10^{-4} g/cm²·d at 75°C. In the case of the rock-present channels, Fig. 5, the leach rate pattern just described does not appear to change. The flow rate effect is more evident at 75°C with the addition of the slow flow channels in the graph. The range in the leach rates is still the same as if no rock were present in the system.

Cumulative Fractions

Cumulative fractions leached were also calculated for Pu and Np at each one of the fourteen collection days during the experiment. These values are shown in Tables 9 and 10, respectively. They are also plotted in Figs. 6 through 9. As pointed out in the case of the incremental leach rates, the cumulative fractions leached for the "rock-absent" channels (1, 4, 5, 33, 34, 36) are actual cumulative values, whereas those for the "rock-present" channels (7, 8, 10, 39, 42, 43, 65, 69) are pseudo-cumulative values because they do not include the fraction of Pu and Np adsorbed on the rock. Identification of channels is shown with numbers on the right hand side of each graph.

A summary of the cumulative fractions leached, expressed in percent, for Pu and Np is presented in Table 11. In this table, the cumulative

Table 9

BEADLEACH II

PLUTONIUM CUMULATIVE FRACTION LEACHED(CFL)

CHAN TEMP FLOW SAMPLING DAYS

		1	2	3	6	11	20	37	70	120	151	213
1	COLD MED	5.33E-06	1.09E-05	1.26E-05	1.55E-05	1.78E-05	2.40E-05	3.20E-05	3.50E-05	3.65E-05	3.71E-05	3.86E-05
4	COLD FAST	6.48E-06	9.18E-06	1.02E-05	1.18E-05	1.31E-05	1.50E-05	1.87E-05	2.38E-05	2.89E-05	3.20E-05	3.63E-05
5	COLD FAST	1.07E-05	1.29E-05	1.38E-05	1.63E-05	1.82E-05	2.05E-05	2.35E-05	2.66E-05	3.58E-05	3.96E-05	0.
7	COLD MED	3.27E-07	8.97E-07	1.40E-06	2.33E-06	3.09E-06	4.26E-06	5.74E-06	6.94E-06	7.42E-06	7.80E-06	8.52E-06
8	COLD MED	3.13E-07	1.15E-06	2.40E-06	4.32E-06	5.32E-06	6.92E-06	9.01E-06	9.97E-06	1.02E-05	1.06E-05	1.17E-05
10	COLD FAST	7.03E-06	8.36E-06	9.25E-06	1.05E-05	1.13E-05	1.32E-05	0.	2.03E-05	2.78E-05	3.45E-05	4.27E-05
33	HOT MED	1.10E-06	1.50E-06	1.82E-06	2.28E-06	2.44E-06	2.52E-06	2.60E-06	2.70E-06	2.94E-06	3.24E-06	4.30E-06
34	HOT MED	1.10E-06	1.73E-06	1.94E-06	2.33E-06	2.55E-06	2.65E-06	2.71E-06	2.81E-06	3.21E-06	3.41E-06	4.47E-06
36	HOT FAST	2.17E-06	2.82E-06	3.29E-06	5.04E-06	7.37E-06	1.31E-05	1.94E-05	2.46E-05	6.46E-05	1.03E-04	1.93E-04
39	HOT MED	4.44E-08	5.82E-08	7.76E-08	1.14E-07	1.42E-07	1.74E-07	0.	2.76E-07	3.07E-07	3.43E-07	8.45E-07
42	HOT FAST	2.04E-06	2.29E-06	2.48E-06	3.04E-06	4.12E-06	7.59E-06	1.43E-05	2.28E-05	7.83E-05	1.27E-04	2.81E-04
43	HOT FAST	1.62E-06	1.84E-06	2.00E-06	2.80E-06	4.55E-06	7.88E-06	1.35E-05	2.13E-05	7.33E-05	1.24E-04	3.45E-04
		1- 10				11- 20	21- 30	31- 50	51- 80	81-120	121-150	163-213
65	HOT SLOW	2.35E-08				2.52E-08	3.03E-08	3.31E-08	3.62E-08	4.94E-08	5.61E-08	9.10E-08
69	HOT SLOW	6.55E-09				6.83E-08	7.24E-08	8.22E-08	1.14E-07	1.47E-07	0.	3.61E-07

Table 9 (continued)

BEADLEACH 11

PLUTONIUM CUMULATIVE FRACTION LEACHED(CFL)

CHAN TEMP FLOW SAMPLING DAYS						
				280	342	435
1	COLD	MED	0.		4.11E-05	4.31E-05
4	COLD	FAST	0.		4.75E-05	6.41E-05
5	COLD	FAST	0.		5.85E-05	7.00E-05
7	COLD	MED	0.		9.32E-06	1.06E-05
8	COLD	MED	0.		1.49E-05	1.81E-05
10	COLD	FAST	0.		5.09E-05	5.73E-05
33	HOT	MED	0.		2.27E-05	3.98E-05
34	HOT	MED	0.		1.30E-05	2.40E-05
36	HOT	FAST	0.		4.29E-04	5.46E-04
39	HOT	MED	0.		1.29E-05	2.25E-05
42	HOT	FAST	0.		8.11E-04	1.19E-03
43	HOT	FAST	0.		1.17E-03	1.57E-03
				246-280	310-342	408-435
65	HOT	SLOW	0.		3.53E-07	5.47E-07
69	HOT	SLOW	0.		5.45E-07	5.92E-07

Table 10

BEADLEACH II

		NEPTUNIUM CUMULATIVE FRACTION LEACHED(CFL)										
CHAN	TEMP FLOW	SAMPLING DAYS										
		1	2	3	6	11	20	37	70	120	151	213
1	COLD MED	2.63E-05	7.99E-05	1.01E-04	1.33E-04	1.49E-04	1.69E-04	1.96E-04	2.37E-04	2.95E-04	3.16E-04	3.66E-04
4	COLD FAST	5.55E-05	7.84E-05	8.71E-05	1.03E-04	1.18E-04	1.37E-04	1.81E-04	2.53E-04	2.99E-04	3.44E-04	4.43E-04
5	COLD FAST	7.56E-05	1.06E-04	1.24E-04	1.61E-04	1.89E-04	2.16E-04	2.56E-04	3.04E-04	3.70E-04	4.00E-04	4.29E-04
7	COLD MED	2.56E-06	8.14E-06	1.89E-05	4.25E-05	6.19E-05	8.20E-05	1.08E-04	1.64E-04	2.55E-04	2.92E-04	3.24E-04
8	COLD MED	2.93E-06	1.09E-05	2.38E-05	4.98E-05	6.77E-05	8.94E-05	1.20E-04	1.62E-04	2.67E-04	3.15E-04	3.64E-04
10	COLD FAST	6.50E-05	1.01E-04	1.16E-04	1.43E-04	1.57E-04	1.80E-04	2.19E-04	2.74E-04	3.45E-04	3.74E-04	4.11E-04
33	HOT MED	4.66E-05	1.28E-04	2.41E-04	5.15E-04	7.65E-04	1.13E-03	1.65E-03	2.84E-03	5.63E-03	7.30E-03	1.04E-02
34	HOT MED	4.01E-05	1.30E-04	1.89E-04	4.44E-04	7.69E-04	1.13E-03	1.41E-03	1.77E-03	3.74E-03	4.52E-03	6.52E-03
36	HOT FAST	1.90E-04	5.12E-04	7.97E-04	1.67E-03	2.58E-03	5.22E-03	9.06E-03	1.49E-02	2.60E-02	3.28E-02	4.56E-02
39	HOT MED	1.45E-05	3.71E-05	6.40E-05	1.42E-04	2.62E-04	5.16E-04	1.02E-03	1.75E-03	3.36E-03	4.72E-03	7.61E-03
42	HOT FAST	1.80E-04	4.92E-04	7.97E-04	1.88E-03	2.75E-03	5.01E-03	9.89E-03	1.64E-02	2.89E-02	3.79E-02	5.67E-02
43	HOT FAST	2.13E-04	5.02E-04	7.83E-04	1.69E-03	3.13E-03	5.38E-03	1.02E-02	1.74E-02	2.96E-02	3.82E-02	5.32E-02
		1- 10				11- 20	21- 30	31- 50	51- 80	81-120	121-150	183-213
65	HOT SLOW	1.67E-04				2.02E-04	2.65E-04	4.40E-04	7.29E-04	1.20E-03	2.01E-03	3.16E-03
69	HOT SLOW	9.91E-05				2.04E-04	2.95E-04	5.16E-04	7.86E-04	1.39E-03	1.80E-03	2.76E-03

Table 10 (continued)

BEADLEACH II

NEPTUNIUM CUMULATIVE FRACTION LEACHED(CFL)			
CHAN TEMP FLOW SAMPLING DAYS			
		280	342 435
1	COLD MED	0.	4.52E-04 5.01E-04
4	COLD FAST	0.	5.73E-04 8.30E-04
5	COLD FAST	0.	5.50E-04 6.99E-04
7	COLD MED	0.	3.59E-04 4.01E-04
8	COLD MED	0.	4.47E-04 4.91E-04
10	COLD FAST	0.	5.18E-04 7.29E-04
33	HOT MED	0.	1.96E-02 2.85E-02
34	HOT MED	0.	1.61E-02 2.32E-02
36	HOT FAST	0.	7.19E-02 8.57E-02
39	HOT MED	0.	1.56E-02 2.39E-02
42	HOT FAST	0.	9.37E-02 1.17E-01
43	HOT FAST	0.	8.90E-02 1.09E-01
		246-280	310-342 408-435
65	HOT SLOW	0.	7.02E-03 1.03E-02
69	HOT SLOW	0.	4.72E-03 7.29E-03

Fig. 6. Glass-Brine Leachant

Fig. 7. Glass-Brine Leachant

Fig. 8. Glass-Brine Leachant

Fig. 9. Glass-Brine Leachant

TABLE 11
Summary of Cumulative Fractions Leached, Day 440

Channel (No.)	Temperature (°C)	Flow Rate (ml/d)	Percent	
			Pu	Np
1	25	10	4.3×10^{-3}	0.050
7*	25	10	2.9×10^{-3}	0.040
8*	25	10	3.4×10^{-3}	0.049
Avg.			$(3.5 \pm 0.7) \times 10^{-3}$	0.046 ± 0.006
4	25	300	6.4×10^{-3}	0.083
5	25	300	7.0×10^{-3}	0.070
10*	25	300	6.2×10^{-3}	0.073
Avg.			$(6.5 \pm 0.4) \times 10^{-3}$	0.075 ± 0.007
65*	75	1	2.5×10^{-3}	1.1
69*	75	1	2.5×10^{-3}	0.83
Avg.			2.5×10^{-3}	0.97 ± 0.19
33	75	10	4.0×10^{-3}	2.9
34	75	10	2.4×10^{-3}	2.3
39*	75	10	10.9×10^{-3}	2.5
Avg.			$(5.8 \pm 4.5) \times 10^{-3}$	2.6 ± 0.3
36	75	300	5.5×10^{-3}	8.6
42*	75	300	3.7×10^{-1}	11.8
43*	75	300	2.2×10^{-1}	11.0
Avg.			$(2.0 \pm 1.8) \times 10^{-1}$	10.5 ± 1.7

*Beads-rock channels: values include Pu and Np concentrations in the rock.

fractions for channels 7, 8, 10, 39, 42, 43, 65, and 69 include the Pu and Np adsorbed on the rock. Furthermore, the channels have been grouped to show reproducibility of the data within each group having the same experimental conditions of temperature and flow rate. An average value is also shown for each group; the uncertainty for this value is one standard deviation from the mean of the three values in each group with the exception of the 1 ml/d channels of which there are only two.

The Pu data indicate that the total amount of Pu released over 440 days is less than 0.01% for all channels and all conditions except the last group which is 0.2%. Including this group, the actual range of cumulative fractions is from $2.5 \times 10^{-3}\%$ to 0.2%, about two orders of magnitude. The reproducibility of the data within each group varies from a few percent in the case of 25°C, 300 ml/d channels to nearly 100% in the case of the 75°C, 300 ml/d channels. This large variation may be due to the unpredictable leaching behavior of Pu under different experimental conditions. The data, however, indicate that Pu leaching is certainly flow rate dependent as evidenced by the 75°C data and to a lesser extent by the 25°C data. It may also be temperature dependent, although the data are too scattered to make a definite conclusion.

The Np cumulative fractions in Table 11 clearly show that both flow rate and temperature affect Np leaching; the values are 300 ml/day > 10 ml/d > 1 ml/d and 75°C > 25°C. The range of cumulative fractions is from 0.046% for the 25°C, 10 ml/d channels to 10.5% for the 75°C, 300 ml/d channels, and the reproducibility of the data within each group is very good, 10 to 20%.

One of the possible leaching mechanisms is congruent dissolution of the glass. In this experiment, the relationship between weight loss of the glass beads and the Np dissolution indicates that congruent dissolution may be occurring. Tabulated in Table 12 are the Np cumulative fractions leached at day 440 in terms of percent leached and the weight losses for the beads in each corresponding channel. These data indicate that flow rate clearly affects both weight loss and Np dissolution at 75°C. This effect is not apparent at 25°C because the data carry large uncertainties. At the lower temperature, the total weight change is 0.1% or less in every case, and the cumulative Np fraction is the sum of fourteen measurements many of which have as much as 20% uncertainties due to counting alone. At 75°C, however, the data are sufficiently reliable to point out that a linear relationship exists between weight loss and Np dissolution as a function of flow rate. The average values in the "weight loss" column at the three flow rates of 1 ml/d, 10 ml/d, and 300 ml/d are 1.55 ± 0.07 , 3.2 ± 0.4 , and 9.7 ± 1.7 , respectively. Likewise, in the "% Np leached" column, the average values for the three flow rates are 0.95 ± 0.21 , 2.6 ± 0.3 , and 10.5 ± 1.7 , respectively. When these data are plotted against each other, a regression line is obtained with a weight loss (%) intercept of 0.91 ± 0.17 , a slope of 1.2 ± 0.2 and a correlation factor of 1.0. This kind of relationship between weight loss and Np cumulative fraction leached has also been observed in the leaching of glass beads with the basalt groundwater¹ and in the Bead Leach I experiment.

TABLE 12

Comparison of Beads Weight Loss vs. Np Leached

Channel (N°)	Temperature (°C)	Flow Rate (ml/d)	Beads Weight Loss (%)	Np Leached (%)
1	25	10	0.04	0.05
4	25	300	0.01	0.08
5	25	300	0.11	0.07
7	25	10	0.10	0.04*
8	25	10	0.05	0.05*
10	25	300	0.0	0.07*
33	75	10	3.5	2.9
34	75	10	2.8	2.3
36	75	300	7.7	8.6
39	75	10	3.4	2.5*
42	75	300	11.0	11.8*
43	75	300	10.3	11.0*
65	75	1	1.5	1.1*
69	75	1	1.6	0.8*

*Values represent sum of leachate and rock.

Pu and Np Adsorption on Sandstone

The Pu and Np concentrations (dpm) in the four layers of sandstone are shown in Tables 13 and 14, respectively. Also shown in these tables is the percent fraction of Pu and Np adsorbed in each of the four layers and the sum of all four layers. These calculated values represent fractions of the total Pu or Np leached from the beads over 440 days. The total amounts leached are the sum of the concentrations found in all four layers of rock and in the leachates collected. The dpm values in the leachates were calculated from the cumulative fractions leached at day 440 (Tables 9 and 10).

In the case of Pu, adsorption on sandstone appears to be more flow rate dependent than temperature dependent. At 75°C, nearly all of the leached Pu, 98%, is retained on the rock at the slow flow (channels 65 and 69), 80% at the medium flow (channel 39), and only 26% at the fast flow (channel 43). The data for channel 42 should be used with caution because sample A (60% adsorption) may have been contaminated with flake-off from the glass beads; black specks were present in the acid treatment of the crushed rock. At 25°C, about 55% of the leached Pu is retained on the rock at the medium flow (channels 7 and 8) and only 8% at the fast flow (channel 10). Comparing channels 7 and 8 with channel 39, we find a difference in adsorption of 55% vs 80%, respectively, and a difference of 8% vs 26% between channel 10 and channel 43, respectively. These data illustrate the temperature effect on adsorption when flow rate is held constant. The Pu adsorption on sandstone is shown in Fig. 10, quantifying the retardation of Pu as it travels through the entire column. In general, more Pu is found in the layer nearest the glass beads, and the distribution is in a progressively declining pattern from layer A to layer D.

TABLE 13. Rock Adsorption
Plutonium

Channel (N°)	Temperature (°C)	Flow Rate (ml/d)	Concentration (dpm)		Percent Adsorption
			Rock	Leachate	
7A	25	10	1246		28
7B	25	10	686		15
7C	25	10	444		10
7D	25	10	<u>497</u>		<u>11</u>
TOTAL			2873	1653	64
8A	25	10	611		11
8B	25	10	816		15
8C	25	10	604		11
8D	25	10	<u>520</u>		<u>10</u>
TOTAL			2551	2950	47
10A	25	300	422		4
10B	25	300	144		2
10C	25	300	109		1
10D	25	300	<u>129</u>		<u>1</u>
TOTAL			804	9086	8
39A	75	10	4573		28
39B	75	10	3496		21
39C	75	10	2958		18
39D	75	10	<u>2101</u>		<u>13</u>
TOTAL			13128	3453	80

TABLE 13. Rock Adsorption (continued)
Plutonium

Channel (N°)	Temperature (°C)	Flow Rate (ml/d)	Concentration (dpm)		Percent Adsorption
			Rock	Leachate	
42A*	75	300	351300		60
42B	75	300	13950		2
42C	75	300	16160		3
42D	75	300	<u>15370</u>		<u>3</u>
TOTAL			396780	191200	68
43A	75	300	41130		12
43B	75	300	30340		9
43C	75	300	10490		3
43D	75	300	<u>6390</u>		<u>2</u>
TOTAL			88350	249300	26
65A	75	1	2183		53
65B	75	1	886		22
65C	75	1	459		11
65D	75	1	<u>487</u>		<u>12</u>
TOTAL			4015	90	98
69A	75	1	2044		51
69B	75	1	1106		28
69C	75	1	416		10
69D	75	1	<u>342</u>		<u>9</u>
TOTAL			3908	94	98

*Sample contains black glass flakes.

TABLE 14. Rock Adsorption
Neptunium

Channel (N°)	Temperature (°C)	Flow Rate (ml/d)	Concentration (dpm)		Percent Adsorption
			Rock	Leachate	
7A	25	10	< 36		ND
7B	25	10	< 46		ND
7C	25	10	< 43		ND
7D	25	10	< 43		ND
TOTAL				6896	
8A	25	10	< 85		ND
8B	25	10	< 500		ND
8C	25	10	< 43		ND
8D	25	10	< 43		ND
TOTAL				8824	
10A	25	300	< 36		ND
10B	25	300	< 36		ND
10C	25	300	< 43		ND
10D	25	300	< 43		ND
TOTAL				12746	
39A	75	10	2940		0.7
39B	75	10	3860		0.9
39C	75	10	3570		0.9
39D	75	10	2600		0.6
TOTAL			12970	4.04×10^5	3.1

ND Not detected

TABLE 14. Rock Adsorption (continued)
Neptunium

Channel (N°)	Temperature (°C)	Flow Rate (ml/d)	Concentration (dpm)		Percent Adsorption
			Rock	Leachate	
42A*	75	300	19700		0.9
42B	75	300	1460		0.1
42C	75	300	3960		0.2
<u>42D</u>	75	300	<u>1685</u>		<u>0.1</u>
TOTAL			26805	2.07×10^6	1.3
43A	75	300	4230		0.2
43B	75	300	2410		0.1
43C	75	300	2180		0.1
<u>43D</u>	75	300	<u>1650</u>		<u>0.1</u>
TOTAL			10470	1.91×10^6	0.5
65A	75	1	3470		1.7
65B	75	1	4570		2.2
65C	75	1	5780		2.8
<u>65D</u>	75	1	<u>5170</u>		<u>2.5</u>
TOTAL			18990	1.86×10^5	9.2
69A	75	1	2920		2.0
69B	75	1	3880		2.7
69C	75	1	3820		2.7
<u>69D</u>	75	1	<u>5750</u>		<u>4.0</u>
TOTAL			16370	1.28×10^5	11.4

*Sample contains black glass flakes.

		High temperature (75°C)					Low temperature (25°C)		
		1 ml/day		10 ml/day	300 ml/day		10 ml/day	300 ml/day	
		Ch 65	Ch 69	Ch 39	Ch 42	Ch 43	Ch 7	Ch 8	Ch 10
Leachant flow ↑	D	12	9	13	3	2	11	10	1
	C	11	10	18	3	3	10	11	1
	B	22	28	21	2	9	15	15	2
	A	53	51	28	60	12	28	11	4

Fig. 10. Pu adsorption (%)
(Sandstone - Brine)

It is interesting to note that the total amounts of Pu leached and the individual leach rates are significantly greater in the 75°C rock present channels than in the comparable flow rock-absent channels although similar effects are not seen for Np. Because of the relatively small number of replicates involved, this could be an experimental or statistical artifact, but the possibility that interaction with the rock enhances the solution's ability to mobilize Pu is one which merits further investigation.

In the case of Np, adsorption on sandstone is not detectable at 25°C; all of the values in Table 14 are "less than" because the measurements were at or below the limit of detection of the alpha counting system. At 75°C, Np was easily detected in all samples and the data in Table 14 indicate that the fraction of the total Np leached that is retained on the rock varies from 10% at the slow flow (channels 65 and 69), to 3% at the medium flow (channel 39), to 0.5% at the fast flow (channel 43). These data suggest a flow rate dependence to a lesser extent compared to Pu. The distribution profile for Np adsorption throughout the entire rock column is shown in Fig. 11. With the exception of channel 42, which is suspect because of analytical problems, the profiles for the remaining four channels are similar; Np appears to be uniformly distributed from layer A to layer D. Both the total amounts sorbed and the distribution within the rock layers indicate the greater mobility of Np compared to Pu.

		High temperature (75°C)					Low temperature (25°C)		
		1 ml/day		10 ml/day	300 ml/day		10 ml/day	300 ml/day	
		Ch 65	Ch 69	Ch 39	Ch 42	Ch 43	Ch 7	Ch 8	Ch 10
Leachant flow ↑	D	2	4	0.6	0.1	0.1	ND	ND	ND
	C	3	3	0.9	0.2	0.1	ND	ND	ND
	B	2	3	0.9	0.1	0.1	ND	ND	ND
	A	2	2	0.7	0.9	0.2	ND	ND	ND

ND = not detected

Fig. 11. Np adsorption (%)

(Sandstone - Brine)

III. UO_2 WITH BRINE SOLUTION

This section deals with the leaching of UO_2 pellets with the brine described in the previous section.

UO_2 was the second "waste form" studied in the Bead Leach II experiment. The UO_2 was in the form of cylindrical pellets 0.86 cm in diameter and 0.89 cm in length. The pellets were fabricated at PNL⁵ from depleted uranium powder by precompacting the powder to about 5.0 g/cm^3 and then granulating and screening it through a 10-mesh Tyler screen. The granulated material was lubricated with 0.3 weight percent zinc stearate and pressed into pellets with a density of $5.85 \pm 0.05 \text{ g/cm}^3$. These were sintered at 1700°C for eight hours and later cleaned with alcohol before shipment.

As was the case with the glass beads, the UO_2 pellets were loaded into the leaching cells with and without crushed sandstone. Three pellets were loaded in each of fourteen cells and crushed sandstone was added to eight of them. Each group of three pellets weighed approximately 16 g and had a total geometric surface area of 10.68 cm^2 . The same four cells (25, 29, 57, and 59), which were run with rock only as blanks for the glass-sandstone samples, were also used here as blanks for the UO_2 -sandstone samples. Hereafter, this portion of the Bead Leach II experiment will be referred to as the " UO_2 -brine" channels in reference to the leaching cells and as the " UO_2 -brine" leachates in reference to the leachates collected from those channels.

The experimental matrix with information on the temperature and flow rate parameters is shown in Table 15.

TABLE 15. Experimental Matrix

UO₂-Brine Leachant

Channel No. (N°)	Temperature (°C)	Flow Rate (ml/d)	Leaching Cell Configuration		
			UO ₂	Rock	UO ₂ -Rock
13	25	10	X		
14	25	10	X		
16	25	300	X		
19	25	10			X
22	25	300			X
23	25	300			X
25	25	10		X	
29	25	300		X	
45	75	10	X		
48	75	300	X		
49	75	300	X		
51	75	10			X
52	75	10			X
54	75	300			X
57	75	10		X	
59	75	300		X	
66	75	1			X
70	75	1			X

Analytical Procedures

The procedure for the sampling and collection of the UO_2 -brine leachates was identical to that used with the glass leachates. Since uranium was the only element of interest in the UO_2 leachates, a rapid and convenient analytical procedure was developed to process the large number of samples involved. The analysis consisted of taking a large aliquot of the leachate (usually 5 ml or more in the case of the slow-flow channels and 100-200 ml in the case of the fast-flow channels) and separating the uranium by precipitation, using iron as a carrier. The concentrated uranium fraction was slurried with 300 mg of Avicel (cellulose powder) and dried under a heat lamp or at room temperature. The dried mixture was transferred to a 1-inch stainless steel die and pressed into a flat wafer with a Carver Press. This wafer was then counted in a x-ray fluorescence energy dispersive system and the uranium was quantified using a computer code. ICP analysis on the leachates was not possible because of the high concentration of brine and also because the uranium concentrations were at or below the limit of detection of 0.5 $\mu\text{g/ml}$.

Calculation

Leach rates (incremental and cumulative) were calculated using the uranium concentrations in the leachates. The algorithms used for these calculations were the same as described in the preceding section.

Results and Discussion

The weights of the UO_2 pellets in each cell, before and after leaching, are given in Table 16. These data indicate a gain in weight rather than a loss for most samples; only samples 14, 45, and 52 reflect a loss. It is difficult to state whether these small changes in weight

TABLE 16.
UO₂ Weights (Before and After Leaching)

Channel (N°)	Temperature (°C)	Flow Rate (ml/d)	Original Weight (g)	Final Weight (g)	ΔW (g)	ΔW (%)
13	25	10	16.3373	16.3376	+0.0003	0.0018
14	25	10	16.3055	16.3049	-0.0006	0.0037
16	25	300	16.3757	16.3784	+0.0027	0.0165
19	25	10	16.4159	16.4460	+0.0301	0.1834
22	25	300	16.1874	16.1891	+0.0017	0.0105
23	25	300	16.3812	16.3835	+0.0023	0.0140
45	75	10	16.3283	16.2364	-0.0919	0.5628
48	75	300	16.6844	16.7526	+0.0682	0.4088
49	75	300	16.2646	16.3045	+0.0399	0.2453
51	75	10	16.4785	16.4797	+0.0012	0.0073
52	75	10	16.4116	16.4114	-0.0002	0.0012
54	75	300	16.4717	16.4771	+0.0054	0.0328
66	75	1	16.2745	16.2750	+0.0005	0.0031
70	75	1	16.0839	16.1151	+0.0312	0.1940

are real or not and, consequently, equally difficult to make conclusions based on these data.

The concentrations of uranium ($\mu\text{g/ml}$) in the leachates are shown in Table 17. Many of these results are expressed as "less than" values, particularly in the case of the slow and medium flow channels during the first 20 days of the experiment. These "less than" values are the result of uranium concentrations in the leachates which were below the limit of detection of the x-ray fluorescence system, 0.1 μg . Subsequent to day 20, uranium was detected and measured in all samples. The uncertainty due to x-ray counting is 10-20%.

The uranium concentrations in Table 17 were used to calculate incremental leach rates for each of the sampling periods during the experiment. These results are shown in Table 18. The variables used in the calculation of leach rates are the volume of leachate collected per unit time, the weight of the UO_2 pellets, and the geometric surface area of the UO_2 pellets. All of these are included in the algorithm shown in the preceeding section of this report. Consequently, the uncertainty of the leach rate values depends on the combined effects of three variables plus the uncertainties in processing and counting the samples. An estimate of the overall uncertainty is difficult to make based on the available data; it is probably more than 20% for the samples in this section.

The leach rates for the UO_2 samples without sandstone (rock absent) have been plotted in Fig. 12 and the leach rates for the UO_2 samples with sandstone (rock present) in Fig. 13. Identification of channels is shown by using the channel number at the extreme right side of each

Table 17

BEADLEACH II

		TABLE OF URANIUM CONCENTRATION (UG/CM3) XRFA ANALYSIS										
CHAN TEMP FLOW SAMPLING DAYS		1	2	3	6	11	20	37	70	120	151	213
13	COLD MED	<1.00E-02	<1.00E-02	1.30E-02	1.20E-02	1.00E-02	3.40E-02	4.20E-02	0.	2.40E-01	0.	2.10E-02
14	COLD MED	<1.00E-02	<1.00E-02	3.40E-02	3.80E-02	2.00E-02	5.80E-02	5.60E-02	0.	2.10E-01	0.	1.80E-01
16	COLD FAST	1.20E-02	2.00E-03	0.	0.	1.00E-03	1.00E-03	1.00E-03	3.00E-03	1.50E-02	1.30E-02	3.00E-03
19	COLD MED	<1.00E-02	<1.00E-02	<1.00E-02	<1.00E-02	<1.00E-02	1.40E-02	1.80E-01	0.	1.60E-01	0.	6.70E-02
22	COLD FAST	5.00E-03	2.00E-03	0.	0.	1.00E-03	1.00E-03	2.00E-03	5.00E-03	1.30E-02	1.70E-02	4.00E-03
23	COLD FAST	3.00E-03	2.00E-03	0.	0.	2.00E-03	1.00E-03	5.00E-03	6.00E-03	2.10E-02	2.60E-02	5.00E-03
25	COLD MED	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.
29	COLD FAST	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.
45	HOT MED	<1.00E-02	<1.00E-02	7.10E-02	4.30E-02	3.70E-02	2.10E-02	2.00E-02	0.	5.80E-02	0.	8.00E-03
48	HOT FAST	1.30E-02	4.00E-03	0.	0.	4.00E-03	4.00E-03	2.00E-03	1.00E-03	9.00E-03	4.00E-03	2.00E-03
49	HOT FAST	9.00E-03	5.00E-03	0.	0.	3.00E-03	2.00E-03	2.00E-03	<1.00E-03	1.60E-02	1.70E-02	8.00E-03
51	HOT MED	<1.00E-02	<1.00E-02	<1.00E-02	<1.00E-02	<1.00E-02	0.	4.70E-02	0.	3.00E-02	0.	1.00E-03
52	HOT MED	<1.00E-02	<1.00E-02	<1.00E-02	<1.00E-02	<1.00E-02	5.00E-03	3.90E-02	0.	3.30E-02	0.	1.30E-02
54	HOT FAST	3.00E-03	3.00E-03	0.	0.	6.00E-03	4.00E-03	5.00E-03	2.00E-03	1.20E-02	1.10E-02	4.00E-03
57	HOT MED	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.
59	HOT FAST	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.
1- 10						11- 20	21- 30	31- 50	51- 80	81-120	121-150	163-213
66†	HOT SLOW	<1.00E-02	0.	0.	0.	<1.00E-02	<1.00E-02	1.80E-02	7.70E-02	0.	8.90E-02	5.00E-03
70†	HOT SLOW	<1.00E-02	0.	0.	0.	<1.00E-02	0.	1.70E-02	6.90E-02	3.10E-02	1.60E-02	3.20E-02

Note: † Solutions were collected over a cumulative period in order to obtain enough sample for analysis.

Table 17 (continued)

BEADLEACH II

		TABLE OF URANIUM CONCENTRATION (UG/CM3) XRFA ANALYSIS		
CHAN	TEMP FLOW SAMPLING DAYS	280	342	435
13	COLD MED	8.30E-02	9.40E-02	1.60E-01
14	COLD MED	1.60E-01	1.90E-01	2.10E-01
16	COLD FAST	3.00E-03	6.00E-03	7.00E-03
19	COLD MED	2.40E-01	7.00E-02	2.60E-01
22	COLD FAST	6.00E-03	5.00E-03	5.00E-03
23	COLD FAST	5.00E-03	5.00E-03	4.00E-03
25	COLD MED	0.	0.	0.
29	COLD FAST	0.	0.	0.
45	HOT MED	4.50E-02	3.70E-02	6.30E-02
48	HOT FAST	4.00E-03	5.00E-03	3.00E-03
49	HOT FAST	1.40E-02	1.30E-02	9.00E-03
51	HOT MED	5.00E-03	1.70E-02	3.00E-02
52	HOT MED	4.40E-02	5.60E-02	1.40E-01
54	HOT FAST	3.00E-03	6.00E-03	1.00E-03
57	HOT MED	0.	0.	0.
59	HOT FAST	0.	0.	0.
		246-280	310-342	408-435
66	HOT SLOW	2.00E-02	1.54E+00	4.40E+00
70	HOT SLOW	1.00E-02	4.00E-03	4.80E-02

Table 18

BEADLEACH II

URANIUM LEACH RATE (G/CM2.DAY)

CHAN TEMP FLOW SAMPLING DAYS		XRFA ANALYSIS										
		1	2	3	6	11	20	37	70	120	151	213
13	COLD MED	<1.46E-07	<1.20E-08	1.31E-08	1.28E-08	1.04E-08	3.45E-08	4.41E-08	0.	2.25E-07	0.	5.53E-08
14	COLD MED	<1.08E-08	<1.15E-08	3.79E-08	4.46E-08	2.18E-08	6.44E-08	6.17E-08	0.	2.30E-07	0.	1.81E-07
16	COLD FAST	3.99E-07	6.83E-08	0.	0.	3.23E-08	3.06E-09	3.26E-08	5.10E-08	4.86E-07	4.19E-07	9.72E-08
19	COLD MED	<1.15E-08	<1.19E-08	<1.17E-08	<1.18E-08	<1.13E-08	1.63E-08	2.08E-07	0.	1.80E-07	0.	7.27E-08
22	COLD FAST	1.71E-07	7.01E-08	0.	0.	3.41E-08	3.38E-08	6.68E-08	1.94E-07	5.98E-07	5.57E-07	1.33E-07
23	COLD FAST	1.58E-07	7.99E-08	0.	0.	6.62E-08	3.27E-08	1.67E-07	1.87E-07	6.81E-07	8.40E-07	1.95E-07
45	HOT MED	<7.02E-09	<1.05E-08	9.99E-08	6.50E-08	4.56E-08	2.69E-08	2.62E-08	0.	6.99E-08	0.	6.10E-09
48	HOT FAST	4.73E-07	1.45E-07	0.	0.	1.35E-07	1.34E-07	6.67E-08	3.24E-08	2.94E-07	1.31E-07	6.52E-08
49	HOT FAST	2.99E-07	1.70E-07	0.	0.	9.56E-08	6.38E-08	6.37E-08	<3.10E-08	5.06E-07	5.35E-07	2.56E-07
51	HOT MED	<1.34E-08	<1.24E-08	<4.08E-09	<1.51E-08	<1.53E-08	0.	5.94E-08	0.	3.41E-08	0.	1.46E-09
52	HOT MED	<1.12E-08	<1.15E-08	<1.11E-08	<1.06E-08	<1.09E-08	5.48E-09	4.16E-08	0.	4.90E-08	0.	1.32E-08
54	HOT FAST	1.04E-07	1.06E-07	0.	0.	1.99E-07	1.34E-07	1.66E-07	6.31E-08	3.92E-07	3.61E-07	1.27E-07
1- 10						11- 20	21- 30	31- 50	51- 80	81-120	121-150	153-213
66	HOT SLOW	<1.32E-09	0.	0.	0.	<1.30E-09	<1.33E-09	2.15E-09	9.67E-09	0.	2.23E-08	6.11E-10
70	HOT SLOW	<2.68E-09	0.	0.	0.	<1.37E-09	0.	1.92E-09	1.07E-08	2.07E-09	2.68E-08	6.55E-09

Table 18 (continued)

BEADLEACH II

URANIUM LEACH RATE (G/CM2.DAY)

CHAN TEMP FLOW SAMPLING DAYS			XRFA ANALYSIS		
		280	342	435	
13	COLD MED	1.16E-07	1.17E-07	1.93E-07	
14	COLD MED	2.34E-07	2.09E-07	2.16E-07	
16	COLD FAST	1.01E-07	2.04E-07	1.57E-07	
19	COLD MED	1.72E-07	1.13E-07	3.04E-07	
22	COLD FAST	1.85E-07	1.68E-07	1.59E-07	
23	COLD FAST	1.65E-07	1.60E-07	1.27E-07	
45	HOT MED	5.22E-08	4.00E-08	9.15E-08	
48	HOT FAST	1.34E-07	1.85E-07	9.72E-08	
49	HOT FAST	4.51E-07	4.67E-07	2.79E-07	
51	HOT MED	7.54E-09	1.63E-08	4.16E-08	
52	HOT MED	5.14E-08	6.05E-08	1.44E-07	
54	HOT FAST	9.98E-08	2.03E-07	3.26E-08	
		246-280	310-342	408-435	
66	HOT SLOW	2.02E-09	1.48E-07	5.80E-07	
70	HOT SLOW	2.24E-09	6.46E-10	7.68E-09

Fig. 13. UO₂-Brine Leachant

solid or dotted line in each graph. The leach rates in Fig. 12 vary from 10^{-8} g/cm²·d to about 10^{-6} g/cm²·d. There appears to be a flow rate effect at 75°C since the leach rates for the fast flow channels are higher than the medium flow channels. It is not clear whether this is true at 25°C. The leach rates for the channels with sandstone (rock present) in Fig. 13 exhibit the same pattern and values as those without rock. With the addition of the slow flow channels in the graph, the leach rates range from 10^{-9} g/cm²·d to 10^{-6} g/cm²·d. There is considerable scatter in the data and no conclusive evidence of a temperature effect.

Cumulative fractions leached were also calculated using the leach rate values. These cumulative fractions are shown in Table 19 and plotted in Figs. 14 and 15. With the exception of channels 16, 22 and 23 (fast flow at 25°C) and channels 48 and 54 (fast flow at 75°C), the cumulative fractions leached for the remaining channels at day 440 are expressed as "less than" values because the algorithm calculating cumulative fractions at any point during the experiment uses the upper limit of a "less than" value and then identifies the cumulative fraction from that point on as "less than". We recommend using these data cautiously in evaluating the total amounts of uranium leached.

A summary of the cumulative fractions leached at day 440 has been prepared in Table 20. The channels have been grouped according to temperature and flow rate. The results for the channels in which crushed sandstone was present reflect only the uranium found in the brine leachates. The sandstone itself was not analyzed to verify if any uranium was absorbed and, thus, we cannot make definite statements on adsorption of uranium on

sandstone. Based on the data and also assuming that uranium behaves similarly to neptunium as discussed in the previous section of this report, we would speculate that the adsorption of uranium is very small or negligible.

Table 19
BEADLEACH II

URANIUM CUMULATIVE FRACTION LEACHED(CFL)														
XRFA ANALYSIS														
CHAN	TEMP	FLOW	SAMPLING DAYS											
			1	2	3	6	11	20	37	70	120	151	213	
13	COLD	MED	<9.35E-08	<1.02E-07	<1.10E-07	<1.37E-07	<1.72E-07	<3.13E-07	<7.53E-07	<0.	<8.49E-06	<0.	<1.51E-05	
14	COLD	MED	<7.01E-09	<1.46E-08	<3.84E-08	<1.27E-07	<2.24E-07	<4.93E-07	<1.20E-08	<0.	<9.57E-06	<0.	<2.13E-05	
16	COLD	FAST	2.59E-07	3.04E-07	0.	0.	5.87E-07	7.00E-07	7.92E-07	1.70E-06	1.14E-05	2.05E-05	3.07E-05	
19	COLD	MED	<7.42E-09	<1.52E-08	<2.25E-08	<4.70E-08	<8.30E-08	<1.65E-07	<1.47E-08	<0.	<1.23E-05	<0.	<1.87E-05	
22	COLD	FAST	1.12E-07	1.58E-07	0.	0.	4.55E-07	6.57E-07	1.23E-06	4.46E-06	1.76E-05	2.94E-05	4.34E-05	
23	COLD	FAST	1.02E-07	1.54E-07	0.	0.	5.77E-07	8.57E-07	2.00E-06	5.85E-06	2.13E-05	3.68E-05	5.74E-05	
45	HOT	MED	<4.43E-09	<1.18E-08	<6.95E-08	<2.03E-07	<3.61E-07	<5.67E-07	<8.62E-07	<0.	<3.42E-06	<0.	<4.90E-06	
48	HOT	FAST	2.91E-07	3.91E-07	0.	0.	1.19E-06	1.96E-06	3.03E-06	4.07E-06	9.83E-06	1.39E-05	1.76E-05	
49	HOT	FAST	1.88E-07	3.09E-07	0.	0.	1.06E-06	1.52E-06	2.23E-06	<3.25E-06	<1.30E-05	<2.36E-05	<3.97E-05	
51	HOT	MED	<8.32E-09	<1.70E-08	<1.96E-08	<4.22E-08	<9.05E-08	<0.	<7.33E-07	<0.	<3.31E-06	<0.	<4.03E-06	
52	HOT	MED	<6.99E-09	<1.50E-08	<2.22E-08	<4.36E-08	<7.79E-08	<1.24E-07	<3.96E-07	<0.	<2.94E-06	<0.	<4.42E-06	
54	HOT	FAST	6.49E-08	1.39E-07	0.	0.	1.05E-06	2.00E-06	3.66E-06	6.08E-06	1.42E-05	2.17E-05	3.15E-05	
			1- 10					11- 20	21- 30	31- 50	51- 60	61-120	121-150	151-213
66	HOT	SLOW	<8.62E-09	<0.	<0.	<0.	<1.71E-08	<2.60E-08	<5.36E-08	<2.52E-07	<0.	<1.12E-06	<1.37E-06	
70	HOT	SLOW	<1.77E-08	<0.	<0.	<0.	<2.68E-08	<0.	<6.30E-08	<2.84E-07	<3.38E-07	<3.93E-07	<6.25E-07	

Table 19 (continued)

BEADLEACH II

		URANIUM CUMULATIVE FRACTION LEACHED(CFL)		
		XRFA ANALYSIS		
		SAMPLING DAYS	342	435
		280		
13	COLD MED	<1.93E-05	<2.41E-05	<3.42E-05
14	COLD MED	<3.07E-05	<3.96E-05	<5.26E-05
16	COLD FAST	3.50E-05	4.13E-05	5.23E-05
19	COLD MED	<2.48E-05	<3.00E-05	<4.46E-05
22	COLD FAST	5.03E-05	5.76E-05	6.76E-05
23	COLD FAST	6.50E-05	7.17E-05	8.04E-05
45	HOT MED	<6.60E-06	<8.36E-06	<1.28E-05
48	HOT FAST	2.20E-05	2.84E-05	3.67E-05
49	HOT FAST	<5.51E-05	<7.39E-05	<9.64E-05
51	HOT MED	<4.28E-06	<4.86E-06	<6.86E-06
52	HOT MED	<6.16E-06	<8.54E-06	<1.56E-05
54	HOT FAST	3.62E-05	4.25E-05	4.94E-05
		246-280	310-342	408-435
66	HOT SLOW	<1.44E-06	<7.36E-06	<3.74E-05
70	HOT SLOW	<7.68E-07	<8.17E-07	<1.18E-06

Fig. 14. UO_2 -Brine Leachant

CUMULATIVE FRACTION ELEMENTAL LEACH

Fig. 15. UO_2 -Brine Leachant

TABLE 20

Summary of Cumulative Fractions Leached, Day 440

Channel (No)	Temperature (°C)	Flow Rate (ml/d)	Fraction Leached (%)
13	25	10	$<3.4 \times 10^{-3}$
14	25	10	$<5.3 \times 10^{-3}$
19*	25	10	$<4.5 \times 10^{-3}$
16	25	300	5.2×10^{-3}
22*	25	300	6.8×10^{-3}
23*	25	300	8.0×10^{-3}
65*	75	1	$<3.7 \times 10^{-3}$
70*	75	1	$<1.2 \times 10^{-3}$
45	75	10	$<1.3 \times 10^{-3}$
51*	75	10	$<6.9 \times 10^{-4}$
52*	75	10	$<1.6 \times 10^{-3}$
48	75	300	3.7×10^{-3}
49	75	300	$<9.6 \times 10^{-3}$
54*	75	300	4.9×10^{-3}

* Crushed sandstone present

IV. UO₂ WITH SIMULATED BASALT GROUNDWATER

This section describes the leaching of UO₂ pellets with a simulated basalt groundwater. Of the 72 cells comprising the Bead Leach II experiment, 18 cells were assigned to this portion. Of these 18 cells, six contained only UO₂ pellets, eight contained both UO₂ pellets and crushed Umtanum Flow basalt, and the remaining four contained crushed basalt only; these last four were used as blanks to make background corrections on the cells with "rock present". The experimental matrix in terms of leaching cells, temperature, flow rate, and configuration is given in Table 21.

The leachant used here was a simulated basalt groundwater. It was prepared in the laboratory according to a recipe recommended by PNL. This recipe is given in Table 22. Unfortunately SiO₂ was omitted in the formulation of the basalt groundwater composition. At the beginning of this experiment, an approved basalt groundwater was not available from the Hanford site. The procedure for preparing this groundwater was given in Part I.

The procedure for preparing the UO₂ pellets, the crushed basalt, loading the leaching cells, and collection of the leachates have been given earlier in this report and in the Bead Leach II, Part I report. As described in the preceding section, uranium in the leachates was analyzed by x-ray fluorescence by taking large aliquots, 5 ml or more as available.

Results and Discussion

The weights of the UO₂ pellets in each cell before and after leaching, are given in Table 23. These data, as in the previous case of the UO₂

TABLE 21. Experimental Matrix
 UO₂ - Simulated Basalt Groundwater

Channel No. (N°)	Temperature (°C)	Flow Rate (ml/d)	Leaching UO ₂	Cell Configuration Rock	UO ₂ -Rock
15	25	10	X		
17	25	300	X		
18	25	300	X		
20	25	10			X
21	25	10			X
24	25	300			X
26	25	10		X	
30	25	300		X	
46	75	10	X		
47	75	10	X		
50	75	300	X		
53	75	10			X
55	75	300			X
56	75	300			X
58	75	10		X	
60	75	300		X	
68	75	1			X
72	75	1			X

TABLE 22

Simulated Basalt Groundwater Chemical Composition*

<u>Ion</u>	<u>mg/l</u>
Na ⁺	30
K ⁺	9
Ca ⁺²	6.5
Mg ⁺²	1
HCO ₃ ⁻	58
SO ₄ ⁻²	23
Cl ⁻	16
F ⁻	0.7
Analytical Reagents	
	mg/l
Na ₂ SO ₄	24.85
KCl	14.91
CaSO ₄	4.76
CaCl ₂	14.42
MgSO ₄ ·7H ₂ O	9.84
NaHCO ₃	79.8
KF·2H ₂ O	3.76

* Realyea, R. F. and Serne, R. J., Control Sample Program Publication No 2, "Interlaboratory Comparison of Batch KD Values", PNL 2872, June 1979.

TABLE 23.

UO₂ Weights (Before and After Leaching)

Channel (N°)	Temperature (°C)	Flow Rate (ml/d)	Original Weight (g)	Final Weight (g)	ΔW (g)	ΔW (%)
15	25	10	16.3684	16.4095	+0.0411	0.2511
17	25	300	16.2831	16.2800	-0.0031	0.0190
18	25	300	16.2532	16.2520	-0.0012	0.0074
20	25	10	16.0654	16.0628	-0.0026	0.0162
21	25	10	16.4523	16.4510	-0.0013	0.0079
24	25	300	16.1596	16.1592	-0.0004	0.0025
46	75	10	16.2527	16.2294	-0.0233	0.1434
47	75	10	16.4223	16.2795	-0.1428	0.8695
50	75	300	16.3249	16.3286	+0.0037	0.0227
53	75	10	16.3488	16.3433	-0.0055	0.0336
55	75	300	16.3310	16.3312	+0.0002	0.0012
56	75	300	16.3105	16.3123	+0.0018	0.0110
68	75	1	16.5704	16.5684	-0.0020	0.0121
72	75	1	16.2834	16.2833	-0.0001	0.0006

pellets leached with the brine solution, are not conclusive enough to point out real changes in weight at the end of the experiment. Of the fourteen values in Table 23, four reflect a weight gain and the rest show very small losses, on the order of 0.2% or less.

The concentrations of uranium in the leachates are shown in Table 24. Uranium was detected in all samples taken for analyses. The uncertainty attached to each value is on the order of 10-20% but this error includes the counting error only.

The uranium concentrations in Table 24 were used to calculate incremental leach rates. These are shown in Table 25 and plotted in Figs. 16 and 17. No statistical variation is shown for these values (refer to the previous section for a discussion of the overall uncertainty on leach rates). Fig. 16 shows the leach rates for the channels containing UO_2 only (rock absent); the numbers after the last collection at the end of each line in the graph identify the channel number. As can be seen in the graph, the leach rates range from about 5×10^{-8} g/m²·d to about 10^{-6} g/m²·d. It appears that the leach rates at 25°C for the medium and and fast flow channels are higher than those at 75°C except for some cross-over between day 250 and day 300 of the experiment.

There is a flow rate effect at 25°C, with fast flow leach rate > medium flow leach rate, but this effect is not obvious at 75°C, particularly during the second half of the experiment.

Fig. 17 shows leach rate data for the channels containing both UO_2 pellets and crushed basalt (rock present). The extra set of lines (dotted) in the graph belong to the slow flow channels (1 ml/d). Including

Table 24

BEADLEACH 11

		TABLE OF URANIUM CONCENTRATION (UG/CM3)											
		XRFA ANALYSIS											
CHAN TEMP FLOW SAMPLING DAYS		1	2	3	6	11	20	37	70	120	151	213	
15	COLD MED	1.14E+00	5.40E-01	2.30E-01	0.	1.90E-01	1.20E-01	1.10E-01	5.00E-02	2.00E-02	0.	9.00E-02	
17	COLD FAST	4.40E-02	2.00E-02	0.	0.	2.50E-02	9.00E-03	1.60E-02	5.00E-02	2.70E-02	3.00E-03	1.50E-02	
18	COLD FAST	4.80E-02	1.80E-02	0.	0.	1.90E-02	1.10E-02	1.90E-02	4.90E-02	1.80E-02	3.00E-03	1.30E-02	
20	COLD MED	4.00E-02	1.70E-01	2.00E-01	0.	1.90E-01	1.10E-01	8.00E-02	9.00E-02	2.00E-02	0.	1.80E-01	
21	COLD MED	0.	3.60E-01	1.30E-01	0.	2.50E-01	1.30E-01	1.00E-01	2.00E-01	7.00E-02	0.	8.00E-02	
24	COLD MED	4.10E-02	2.70E-02	0.	0.	1.80E-02	1.30E-02	6.00E-03	2.80E-02	1.40E-02	3.00E-03	9.00E-03	
26	COLD MED	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.	
30	COLD FAST	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.	
46	HOT MED	6.10E-01	2.70E-01	1.20E-01	0.	1.40E-01	6.00E-03	5.00E-03	5.00E-03	3.00E-03	0.	1.00E-02	
47	HOT MED	6.40E-01	3.20E-01	1.20E-01	0.	9.00E-02	8.00E-03	4.00E-03	0.	8.00E-03	0.	1.20E-02	
50	HOT FAST	5.60E-02	1.80E-02	1.40E-02	0.	1.90E-02	1.00E-02	1.50E-02	0.	1.50E-02	2.00E-03	3.00E-03	
53	HOT MED	5.00E-02	5.00E-02	5.00E-02	0.	8.00E-02	3.00E-02	2.00E-02	0.	8.00E-03	0.	3.00E-02	
55	HOT FAST	2.70E-02	1.40E-02	1.40E-02	0.	1.80E-02	1.30E-02	1.50E-02	0.	1.30E-02	5.00E-03	3.00E-03	
56	HOT FAST	3.00E-02	1.50E-02	1.80E-02	0.	2.20E-02	1.80E-02	1.30E-02	0.	9.00E-03	3.00E-03	3.00E-03	
58	HOT MED	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.	
60	HOT FAST	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.	
1- 10							11- 20	21- 30	31- 50	51- 80	81-120	121-150	183-213
68 [†]	HOT SLOW	3.00E-02	0.	0.	0.	0.	1.00E-01	1.00E-01	4.00E-02	0.	2.00E-02	2.10E-02	
72 [†]	HOT SLOW	4.00E-02	0.	0.	0.	5.00E-02	0.	5.20E-02	3.60E-02	0.	2.20E-02	0.	

Note: [†]Solutions were collected over a cumulative period in order to obtain enough sample for analysis.

Table 24 (continued)

BEADLEACH II

		TABLE OF URANIUM CONCENTRATION (UG/CM3) XRFA ANALYSIS		
CHAN	TEMP FLOW SAMPLING DAYS	280	342	435
15	COLD MED	7.00E-02	1.70E-01	1.60E-01
17	COLD FAST	4.00E-03	1.80E-02	2.00E-02
18	COLD FAST	4.00E-03	1.70E-02	1.70E-02
20	COLD MED	2.60E-01	1.60E-01	1.50E-01
21	COLD MED	1.00E-02	1.00E-02	7.00E-02
24	COLD MED	1.00E-03	1.60E-02	1.90E-02
26	COLD MED	0.	0.	0.
30	COLD FAST	0.	0.	0.
46	HOT MED	1.60E-01	4.40E-01	3.00E-02
47	HOT MED	1.20E-01	5.60E-01	7.00E-02
50	HOT FAST	2.00E-03	5.00E-03	8.00E-03
53	HOT MED	6.00E-02	9.00E-02	3.00E-02
55	HOT FAST	1.00E-03	2.00E-03	5.00E-03
56	HOT FAST	1.00E-03	3.00E-03	1.10E-02
58	HOT MED	0.	0.	0.
60	HOT FAST	0.	0.	0.
		246-280	310-342	408-435
68	HOT SLOW	5.30E-02	5.30E-02	3.50E-02
72	HOT SLOW	0.	3.20E-02	1.30E-02

Table 25
BEADLEACH II

URANIUM LEACH RATE (G/CM2.DAY)

CHAN TEMP: FLOW SAMPLING DAYS		XRFA ANALYSIS											
		1	2	3	6	11	20	37	70	120	151	213	
15	COLD: MED	1.26E-06	6.06E-07	2.59E-07	0.	2.05E-07	1.34E-07	1.22E-07	5.31E-08	1.87E-08	0.	9.09E-08	
17	COLD: FAST	1.44E-06	6.65E-07	0.	0.	8.03E-07	2.92E-07	5.17E-07	1.12E-06	8.69E-07	9.79E-08	5.07E-07	
18	COLD: FAST	1.54E-06	5.91E-07	0.	0.	6.06E-07	3.52E-07	5.99E-07	1.03E-06	6.04E-07	9.52E-08	4.13E-07	
20	COLD: MED	5.60E-07	5.23E-07	2.42E-07	0.	1.75E-07	1.12E-07	8.09E-08	8.14E-08	1.96E-08	0.	2.22E-07	
21	COLD: MED	0.	3.81E-07	1.31E-07	0.	3.23E-07	1.43E-07	1.04E-07	1.70E-07	6.60E-08	0.	7.03E-08	
24	COLD: MED	1.37E-06	9.22E-07	0.	0.	5.94E-07	4.41E-07	2.00E-07	8.72E-07	4.71E-07	1.01E-07	3.02E-07	
46	HOT: MED	7.70E-07	3.22E-07	1.52E-07	0.	1.85E-07	8.45E-09	6.22E-09	4.03E-09	2.74E-09	0.	1.19E-08	
47	HOT: MED	6.70E-07	3.19E-07	1.25E-07	0.	8.94E-08	8.05E-09	4.02E-09	0.	1.20E-08	0.	2.19E-08	
50	HOT: FAST	1.74E-06	5.68E-07	4.35E-07	0.	5.95E-07	3.13E-07	4.69E-07	0.	4.70E-07	5.75E-08	9.26E-08	
53	HOT: MED	5.58E-08	5.57E-08	5.23E-08	0.	6.61E-08	3.35E-08	1.95E-08	0.	7.94E-09	0.	3.31E-08	
55	HOT: FAST	8.83E-07	4.64E-07	4.59E-07	0.	5.94E-07	4.28E-07	4.83E-07	0.	4.12E-07	1.60E-07	9.81E-08	
56	HOT: FAST	9.54E-07	4.83E-07	5.67E-07	0.	6.90E-07	5.67E-07	2.95E-07	0.	2.80E-07	9.23E-08	9.32E-08	
1- 10							11- 20	21- 30	31- 50	51- 80	81-120	121-150	163-213
68	HOT: SLOW	3.18E-09	0.	0.	0.	0.	9.54E-09	1.08E-08	3.34E-09	0.	2.48E-09	2.78E-09	
72	HOT: SLOW	6.61E-09	0.	0.	0.	8.32E-09	0.	8.34E-09	5.43E-09	0.	1.85E-09	0.	

Table 25 (continued)

BEADLEACH II

URANIUM LEACH RATE (G/CM2.DAY)

CHAN TEMP: FLOW SAMPLING DAYS				XRFA ANALYSIS		
		280	342	435		
15	COLD: MED	8.77E-08	1.74E-07	1.86E-07		
17	COLD: FAST	1.40E-07	6.17E-07	6.35E-07		
18	COLD: FAST	1.38E-07	5.73E-07	5.31E-07		
20	COLD: MED	3.37E-07	2.12E-07	1.96E-07		
21	COLD: MED	1.05E-08	1.04E-08	7.32E-08		
24	COLD: MED	3.39E-08	5.45E-07	5.91E-07		
46	HOT : MED	2.17E-07	5.45E-07	5.35E-08		
47	HOT : MED	1.39E-07	5.96E-07	7.37E-08		
50	HOT : FAST	5.32E-08	1.72E-07	2.47E-07		
53	HOT : MED	9.83E-08	1.02E-07	3.38E-08		
55	HOT : FAST	2.92E-08	6.74E-08	1.51E-07		
56	HOT : FAST	3.18E-08	9.19E-08	3.62E-07		
		246-280	310-342	408-435		
68	HOT SLOW	8.12E-09	5.48E-09	4.67E-09		
72	HOT SLOW	0.	2.03E-09	1.85E-09

Fig. 16. UO₂-Basalt Groundwater

Fig. 17. UO₂-Basalt Groundwater

this set, the range of leach rates is from about 10^{-9} g/cm²·d to about 10^{-6} g/cm²·d. Again there is enough scatter in the data to make it difficult to evaluate actual differences and effects of temperature or flow rate.

The incremental leach rate values in Table 25 were used to calculate cumulative fractions leached. These are shown in Table 26 and in Figs. 18 and 19. In the "rock absent" graph, as well as in the "rock present" graph, the effect of flow rate is more evident than in the leach rate graphs. In Fig. 18, F>M, and in Fig. 19 F>M>S at both 25°C and 75°C. Also in both figures we see that the cumulative fractions for the fast channels at 25°C are higher than for the fast channels at 75°C. The pattern for the medium flow channels is similar except in the case of the "rock absent" channels where there is a cross-over at about day 350. From this point on the 75°C lines (dotted) show higher cumulative fractions than the 25°C channel. The cumulative fractions leached at day 440 for all channels have been tabulated in Table 27 and grouped according to temperature and flow rate. The data in this table seem to indicate that the "rock absent" cumulative fraction leached is greater than in the "rock present" channels by about 10% for the fast flow 75°C channels, about 25% for the fast flow 25°C channels, and about 300% for the medium flow 75°C channels. The medium flow, 25°C data are too scattered to point out a real difference between "rock absent" and "rock present". These cumulative fraction leached data suggest that some adsorption of uranium may occur on crushed basalt. Since the analysis of the crushed basalt was not performed to verify actual retention of uranium, we lack the necessary evidence to quantify adsorption of uranium in basalt.

Table 26

BEADLEACH II

URANIUM CUMULATIVE FRACTION LEACHED(CFL)
XRFA ANALYSIS

		SAMPLING DAYS										
		1	2	3	6	11	20	37	70	120	151	213
CHAN	TEMP FLOW											
15	COLD MED	8.20E-07	1.22E-06	1.38E-06	0.	2.58E-06	3.55E-06	4.97E-06	6.87E-06	8.05E-06	0.	1.20E-05
17	COLD FAST	9.36E-07	1.37E-06	0.	0.	5.74E-06	8.79E-06	1.34E-05	3.15E-05	6.57E-05	7.48E-05	8.77E-05
18	COLD FAST	1.00E-06	1.39E-06	0.	0.	4.93E-06	7.69E-06	1.31E-05	3.10E-05	5.88E-05	6.55E-05	7.63E-05
20	COLD MED	3.67E-07	7.20E-07	8.75E-07	0.	1.96E-06	2.79E-06	3.87E-06	5.72E-06	7.39E-06	0.	1.68E-05
21	COLD MED	0.	2.51E-07	3.32E-07	0.	1.58E-06	2.88E-06	4.23E-06	7.30E-06	1.12E-05	0.	1.52E-05
24	COLD FAST	8.96E-07	1.52E-06	0.	0.	5.90E-06	8.92E-06	1.25E-05	2.45E-05	4.75E-05	5.30E-05	6.16E-05
46	HOT MED	4.81E-07	7.09E-07	8.09E-07	0.	1.70E-06	2.20E-06	2.28E-06	2.40E-06	2.51E-06	0.	3.04E-06
47	HOT MED	4.09E-07	6.33E-07	7.15E-07	0.	1.26E-06	1.51E-06	1.58E-06	0.	2.03E-06	0.	3.13E-06
50	HOT FAST	1.09E-06	1.49E-06	1.77E-06	0.	4.50E-06	7.06E-06	1.15E-05	0.	3.79E-05	4.28E-05	4.59E-05
53	HOT MED	3.46E-08	7.39E-08	1.08E-07	0.	4.20E-07	7.00E-07	9.89E-07	0.	1.75E-06	0.	3.22E-06
55	HOT FAST	5.43E-07	8.71E-07	1.17E-06	0.	3.95E-06	6.88E-06	1.20E-05	0.	3.70E-05	4.26E-05	4.79E-05
56	HOT FAST	5.86E-07	9.28E-07	1.30E-06	0.	4.60E-06	8.25E-06	1.35E-05	0.	3.24E-05	3.60E-05	3.98E-05
		1- 10				11- 20	21- 30	31- 50	51- 80	81-120	121-150	183-213
68	HOT SLOW	2.04E-08	0.	0.	0.	0.	1.24E-07	2.61E-07	3.29E-07	0.	4.51E-07	5.61E-07
72	HOT SLOW	4.30E-08	0.	0.	0.	9.74E-08	0.	2.61E-07	3.72E-07	0.	5.03E-07	0.

Table 26 (continued)

BEADLEACH 11

URANIUM CUMULATIVE FRACTION LEACHED(CFL)

XRFA ANALYSIS

		SAMPLING DAYS		
		280	342	435
15	COLD MED	1.58E-05	2.21E-05	3.39E-05
17	COLD FAST	1.01E-04	1.17E-04	1.58E-04
18	COLD FAST	8.79E-05	1.03E-04	1.39E-04
20	COLD MED	3.06E-05	4.09E-05	5.44E-05
21	COLD MED	1.64E-05	1.68E-05	2.01E-05
24	COLD FAST	6.85E-05	8.10E-05	1.18E-04
46	HOT MED	9.95E-06	2.92E-05	4.34E-05
47	HOT MED	7.69E-06	2.75E-05	4.36E-05
50	HOT FAST	4.92E-05	5.42E-05	6.78E-05
53	HOT MED	6.66E-06	1.08E-05	1.45E-05
55	HOT FAST	5.05E-05	5.23E-05	5.97E-05
56	HOT FAST	4.24E-05	4.50E-05	6.00E-05
		246-280	310-312	408-435
68	HOT SLOW	8.47E-07	1.09E-06	1.42E-06
72	HOT SLOW	0.	7.47E-07	8.74E-07

Fig. 19. UO₂-Basalt Groundwater

TABLE 27

Summary of Cumulative Fractions Leached, Day 440

Channel (N°)	Temperature (°C)	Flow Rate (ml/d)	Fraction Leached (%)
15	25	10	3.4×10^{-3}
20*	25	10	5.3×10^{-3}
21*	25	10	2.0×10^{-3}
17	25	300	1.6×10^{-2}
18	25	300	1.4×10^{-2}
24*	25	300	1.2×10^{-2}
68*	75	1	1.4×10^{-4}
72*	75	1	0.87×10^{-4}
46	75	10	4.3×10^{-3}
47	75	10	4.4×10^{-3}
53*	75	10	1.5×10^{-3}
50	75	300	6.8×10^{-3}
55*	75	300	6.0×10^{-3}
56*	75	300	6.0×10^{-3}

* Crushed basalt present

V. UO₂ WITH BICARBONATE LEACHANT

UO₂ pellets were also leached with a 0.03 M sodium bicarbonate solution. Because of space limitations in the leaching system, only four of the total of 72 cells in the experiment were allocated for this purpose. No rock interaction was included in these four cells, but all of the remaining experimental parameters were kept consistent with the rest of the experiment. This was also true of the procedures pertaining to collection and analyses of the leachates.

Results and Discussion

The weights of the UO₂ pellets in the four cells before and after leaching, are shown in Table 28. Significant weight losses ranging from 0.3% to 4.5% are recorded for the four different experimental conditions. These data will be discussed further in conjunction with the cumulative fractions leached.

Leachate samples collected during the 440-day experiment were analyzed for uranium using the x-ray fluorescence method described earlier in this report. The results are shown in Table 29. Since the concentrations of uranium range from about 0.2 µg/ml to several µg/ml, the analyses were performed on small aliquots (1-2ml) of the total sample collected. The precision of the measurements is greatly improved at these concentration levels and, thus, the reliability of the data is improved. Based on counting only, the precision of the results is on the order of 5% or less.

Leach rates were calculated using the uranium concentrations in Table 29. These leach rate values are given in Table 30 and shown in Fig. 20. The pattern is essentially constant from beginning to end for all conditions

TABLE 28. Experimental Matrix and Weight Data

UO₂ - Sodium Bicarbonate Solution

Channel (N°)	Temperature (°C)	Flow Rate (ml/d)	UO ₂ Weights			
			Original Weight (g)	Final Weight (g)	ΔW (g)	ΔW (%)
27	25	10	16.2422	16.1888	0.0534	0.33
32	25	300	16.7813	16.7144	0.0669	0.40
61	75	10	15.9653	15.8089	0.1564	0.98
64	75	300	16.2786	15.5453	0.7333	4.51

Table 29

BEADLEACH II

		TABLE OF URANIUM CONCENTRATION (UG/CM3)										
		XRFA ANALYSIS										
CHAN	TEMP FLOW SAMPLING DAYS	1	2	3	6	11	20	37	70	120	151	213
27	COLD MED	5.80E+00	7.10E+00	8.10E+00	1.03E+01	1.25E+01	1.21E+01	1.48E+01	1.17E+01	1.76E+01	1.02E+01	9.50E+00
32	COLD FAST	2.40E-01	2.90E-01	3.10E-01	4.10E-01	3.00E-01	5.40E-01	4.60E-01	6.90E-01	4.40E-01	5.30E-01	4.90E-01
61	HOT MED	2.00E+01	3.80E+01	3.74E+01	0.	5.17E+01	3.03E+01	3.26E+01	1.26E+01	5.07E+01	2.95E+01	0.
64	HOT FAST	3.20E+00	4.60E+00	4.80E+00	7.20E+00	5.60E+00	4.30E+00	5.60E+00	4.40E+00	6.10E+00	4.40E+00	6.40E+00

Table 29 (continued)

BEADLEACH II

CHAN TEMP: FLOW SAMPLING DAYS		TABLE OF URANIUM CONCENTRATION (UG/CM3) XRFA ANALYSIS		
		280	342	435
27	COLD: MED	1.32E+01	0.	1.13E+01
32	COLD: FAST	5.70E-01	6.80E-01	4.10E-01
61	HOT : MED	2.48E+01	0.	3.91E+01
64	HOT : FAST	7.40E+00	9.30E+00	0.

Table 30

BEADLEACH II

URANIUM LEACH RATE (G/CM2.DAY)

CHAN TEMP FLOW SAMPLING DAYS			XRFA ANALYSIS										
			1	2	3	6	11	20	37	70	120	151	213
27	COLD	MED	6.25E-06	8.30E-06	9.30E-06	1.20E-05	1.41E-05	1.31E-05	1.57E-05	1.33E-05	1.95E-05	1.12E-05	1.05E-05
32	COLD	FAST	7.71E-06	9.48E-06	9.91E-06	1.31E-05	9.51E-06	1.71E-05	1.45E-05	2.16E-05	1.34E-05	1.62E-05	1.47E-05
61	HOT	MED	2.32E-05	4.40E-05	4.01E-05	0.	6.15E-05	3.82E-05	4.36E-05	1.48E-05	5.23E-05	3.14E-05	0.
64	HOT	FAST	1.00E-04	1.47E-04	1.49E-04	2.26E-04	1.76E-04	1.35E-04	1.75E-04	1.39E-04	1.84E-04	1.33E-04	1.99E-04

Table 30 (continued)

READLEACH II

URANIUM LEACH RATE (G/CM2.DAY)

CHAN TEMP FLOW SAMPLING DAYS

XRFA ANALYSIS

		280	342	435
27	COLD MED	1.47E-05	0.	1.30E-05
32	COLD FAST	1.86E-05	2.20E-05	1.26E-05
61	HOT MED	2.67E-05	0.	4.28E-05
64	HOT FAST	2.34E-04	3.09E-04	0.

Fig. 20. UO₂-Bicarbonate Leachant

and the leach rates range from 10^{-5} g/cm²·d for the 25°C samples to 10^{-4} g/cm²·d for the 75°C samples. Flow rate clearly affects the leach rate values at 75°C (F>M) but only mildly at 25°C; temperature is also a factor since leach rates at 75°C are greater than at 25°C, particularly in fast flow channel.

Cumulative fractions leached were also calculated. These are shown in Table 31 and in Fig. 21. At day 440, they are 0.40% and 0.48% for the 25°C medium and fast flow channels respectively and 1.0% and 6.2% for the 75°C medium and fast flow channels respectively. This latter value (6.2%) is an extrapolated calculation from the preceeding cumulative fraction (4.36% at day 342) assuming a constant leach rate from that day to the end of the experiment. The cumulative fractions leached can be compared to the weight loss results in Table 28 which are 0.33%, 0.40%, 0.98%, and 4.51% respectively. These data show good agreement between two independent sets of measurements, weight loss and uranium leached.

Table 31

BEADLEACH II

			URANIUM CUMULATIVE FRACTION LEACHED(CFL)										
			XRFA ANALYSIS										
SAMPLING DAYS			1	2	3	6	11	20	37	70	120	151	213
CHAN	TEMP	FLOW											
27	COLD	MED	4.07E-06	9.58E-06	1.55E-05	3.90E-05	8.12E-05	1.61E-04	3.24E-04	6.47E-04	1.21E-03	1.52E-03	1.96E-03
32	COLD	FAST	4.84E-06	1.09E-05	1.70E-05	4.14E-05	7.49E-05	1.54E-04	3.24E-04	7.09E-04	1.29E-03	1.58E-03	2.20E-03
61	HOT	MED	1.46E-05	4.63E-05	7.33E-05	0.	3.50E-04	6.41E-04	1.11E-03	1.75E-03	2.96E-03	3.80E-03	0.
64	HOT	FAST	6.15E-05	1.86E-04	2.64E-04	6.62E-04	1.29E-03	2.19E-03	3.93E-03	7.33E-03	1.30E-02	1.61E-02	2.30E-02

Table 31 (continued)

BEADLEACH II

URANIUM CUMULATIVE FRACTION LEACHED (CFL)
XRFA ANALYSIS

		SAMPLING DAYS 280	342	435
27	COLD MED	2.52E-03	0.	3.95E-03
32	COLD FAST	2.90E-03	3.71E-03	4.76E-03
61	HOT MED	6.18E-03	0.	1.00E-02
64	HOT FAST	3.24E-02	4.36E-02	0.

VI. PNL 76-68 GLASS BEADS WITH BICARBONATE SOLUTION

A portion of the Bead Leach II experiment was also set aside to obtain data that could be correlated with data from the Bead Leach I experiment. Four cells were loaded with glass beads, and these were leached with a solution of 0.03 M sodium bicarbonate using procedures already described in this report. The glass beads used here were from the same batch of glass beads used in the Bead Leach I experiment. Also, there was to be no rock interaction involved with these four cells as the purpose was to duplicate the experimental conditions of the Bead Leach I experiment in terms of flow rate, temperature, and type of leachant.

The leachates were analyzed for Pu and Np using procedures described in the Bead Leach II, Part I report.

Results and Discussion

The glass beads were weighed before and after leaching. The weight data in Table 32 indicate weight losses of 0.22% and 0.33% for the 25°C samples at 10 ml/d and 300 ml/d respectively whereas 3.44% and 20.3% losses were recorded for the 75°C samples at the same flow rates.

The bicarbonate leachates collected during the experiment were analyzed for Pu and Np. The concentrations in dpm/ml appear in Tables 33 and 34, respectively. Underneath each reported value is the percent uncertainty of the measurement based on counting statistics only.

Leach rate values and cumulative fractions leached were also calculated from the Pu and Np concentrations. The leach rate values appear

TABLE 32. Experimental Matrix

PNL 76-68 Glass Beads with Sodium Bicarbonate

Channel (N°)	Temperature (°C)	Flow Rate (ml/d)	Beads		Weights	
			Original Weight (g)	Final Weight (g)	ΔW (g)	ΔW (%)
28	25	10	2.2578	2.2529	0.0049	0.22
31	25	300	2.4517	2.4437	0.0080	0.33
62	75	10	1.9011	1.8252	0.0759	3.44
63	75	300	2.0722	1.6510	0.4212	20.3

Table 33

BEADLEACH II

PNL 76-68 Glass with Bicarbonate

TABLE OF PLUTONIUM CONCENTRATION (DPM/CM3)

CHAN TEMP FLOW SAMPLING DAYS

		1	2	3	6	11	20	37	70	120	151	213
28	COLD: MED	2.29E+01	3.29E+01	3.41E+01	3.01E+01	2.05E+01	1.61E+01	5.21E+00	8.81E+00	9.16E+00	7.83E+00	9.41E+00
		1.6E+00	1.3E+00	1.3E+00	1.5E+00	1.8E+00	1.4E+00	2.6E+00	2.1E+00	2.0E+00	2.3E+00	2.1E+00
31	COLD: FAST	5.29E+00	4.88E+00	3.96E+00	2.41E+00	1.65E+00	2.29E+00	1.51E+00	8.94E-01	7.64E-01	1.01E+00	1.10E+00
		4.0E+00	4.2E+00	4.6E+00	6.2E+00	7.3E+00	3.9E+00	4.6E+00	6.2E+00	7.2E+00	6.2E+00	5.3E+00
62	HOT: MED	2.90E-01	5.61E+00	2.16E+00	4.52E-01	5.30E-01	3.38E-01	5.72E-01	9.97E-01	6.63E-01	3.24E-01	4.30E-01
		1.0E+01	3.0E+00	5.1E+00	1.1E+01	7.9E+00	1.0E+01	7.1E+00	5.8E+00	7.7E+00	1.1E+01	9.2E+00
63	HOT: FAST	7.74E-01	3.10E-01	3.26E-01	6.32E-01	5.38E-01	9.65E-01	6.76E-01	1.10E+00	1.26E+00	8.90E-01	4.86E-01
		6.6E+00	1.5E+01	1.5E+01	1.0E+01	1.1E+01	6.3E+00	7.0E+00	5.8E+00	6.0E+00	7.1E+00	8.1E+00

Table 33 (continued)

BEADLEACH II

TABLE OF PLUTONIUM CONCENTRATION (DPM/CM3)

CHAN TEMP: FLOW SAMPLING DAYS

28	COLD-MED	0.	280	342	435
		0.	8.15E+00	9.06E+00	
31	COLD-FAST	0.	2.2E+00	1.9E+00	
		0.	7.97E-01	8.34E-01	
62	HOT-MED	0.	7.5E+00	6.8E+00	
		0.	3.26E-01	2.96E-01	
63	HOT-FAST	0.	9.9E+00	1.0E+01	
		0.	1.43E+00	1.85E+00	
		0.	5.2E+00	4.3E+00	

Table 34

BEADLEACH II

PNL 76-68 Glass with Bicarbonate

TABLE OF NEPTUNIUM CONCENTRATION (DPM/CM3)

CHAN TEMP FLOW SAMPLING DAYS

	1	2	3	6	11	20	37	70	120	151	213
28 COLD MED	1.23E+01	1.37E+01	1.31E+01	1.69E+01	1.55E+01	1.20E+01	7.58E+00	9.82E+00	9.14E+00	7.35E+00	8.63E+00
	2.1E+00	2.0E+00	2.1E+00	2.0E+00	2.1E+00	1.6E+00	2.2E+00	2.0E+00	2.0E+00	2.4E+00	2.2E+00
31 COLD FAST	1.47E+00	1.76E+00	1.19E+00	7.35E-01	8.32E-01	7.16E-01	8.21E-01	2.73E-01	4.07E-01	3.58E-01	5.28E-01
	7.2E+00	6.8E+00	8.0E+00	1.0E+01	9.5E+00	6.4E+00	5.8E+00	9.2E+00	8.6E+00	8.8E+00	6.8E+00
62 HOT MED	4.73E+01	1.63E+02	1.51E+02	1.46E+02	2.14E+01	5.02E+01	1.47E+02	1.35E+02	1.71E+02	1.52E+02	1.54E+02
	8.6E-01	5.6E-01	6.2E-01	6.3E-01	1.3E+00	8.9E-01	4.6E-01	5.1E-01	5.0E-01	5.3E-01	5.1E-01
63 HOT FAST	2.05E+01	1.46E+01	1.29E+01	1.07E+01	1.85E+01	2.02E+01	1.74E+01	2.49E+01	1.93E+01	2.54E+01	7.89E+00
	1.3E+00	2.3E+00	2.6E+00	1.9E+00	2.0E+00	1.4E+00	1.4E+00	1.3E+00	1.6E+00	1.4E+00	2.1E+00

Table 34 (continued)

BEADLEACH II

TABLE OF NEPTUNIUM CONCENTRATION (DPM/CM³)

CHAN TEMP: FLOW SAMPLING DAYS

			280	342	435
28	COLD MED	0.	8.06E+00	8.58E+00	
		0.	2.2E+00	1.9E+00	
31	COLD FAST	0.	1.08E+00	4.20E-01	
		0.	6.2E+00	8.3E+00	
62	HOT MED	0.	8.64E+01	7.95E+01	
		0.	6.5E-01	6.7E-01	
63	HOT FAST	0.	2.38E+01	2.34E+01	
		0.	1.3E+00	1.2E+00	

in Tables 35 and 36, respectively, and the cumulative fractions leached in Tables 37 and 38, respectively. Using these data and data from the Bead Leach I report, we have prepared three tables to compare the two experiments.

Table 39 contains the incremental leach rate data for Pu for eleven leachate sampling periods. These results indicate that Bead Leach I and Bead Leach II results are equivalent to varying degrees depending on temperature and flow rate. Some results agree well with each other (10%) and others differ by factors of 5, 10, or more. This type of variation is not surprising since Pu tends to adsorb on surfaces readily and may also reprecipitate on the beads during leaching thus introducing anomalies in the results. Consequently, a comparison of the two experiments based on Pu may be risky at best.

A comparison of the Np data is presented in Table 40. A total of eleven incremental leach rates spaced over the length of the experiments are shown and the agreement between the two sets of data is generally satisfactory. Many of the results agree within 10% but some are only good by factors of 2-3 which can be considered beyond the uncertainties of each individual value. The reader is referred to the Bead Leach I report⁴ for a more extensive statistical treatment of the reliability of individual data points and calculated values derived from the experimental data.

Finally, Table 41 shows a comparison of the Pu and Np cumulative fractions leached at the end of the two experiments. The Pu data shows remarkable agreement (10-20%) at 25°C and reasonable agreement (30-70%) at 75°C despite the unpredictable leaching behavior of Pu. The Np comparison is generally good; for each experimental condition the agreement is 10-30% which is within the uncertainties of the individual values.

Table 35

BEADLEACH II

PLUTONIUM LEACH RATE (G/CM2.DAY)

CHAN TEMP FLOW SAMPLING DAYS

		1	2	3	6	11	20	37	70	120	151	213
28	COLD MED	5.55E-07 1.6E+00	7.54E-07 1.3E+00	9.16E-07 1.3E+00	7.14E-07 1.5E+00	5.05E-07 1.9E+00	3.86E-07 1.4E+00	1.37E-07 2.6E+00	2.10E-07 2.1E+00	2.11E-07 2.0E+00	2.23E-07 2.3E+00	1.85E-07 2.1E+00
31	COLD FAST	2.92E-06 4.0E+00	2.71E-06 4.2E+00	2.18E-06 4.6E+00	1.32E-06 6.2E+00	9.13E-07 7.3E+00	1.25E-06 3.9E+00	8.18E-07 4.6E+00	4.79E-07 6.2E+00	4.11E-07 7.2E+00	5.48E-07 6.2E+00	5.83E-07 5.3E+00
62	HOT MED	9.09E-09 1.0E+01	1.69E-07 3.0E+00	6.70E-08 5.1E+00	1.35E-03 1.1E+01	1.52E-03 7.9E+00	9.82E-09 1.0E+01	1.67E-08 7.1E+00	2.83E-08 5.8E+00	1.87E-08 7.7E+00	7.22E-09 1.1E+01	1.03E-08 9.2E+00
63	HOT FAST	4.89E-07 6.6E+00	2.00E-07 1.5E+01	2.03E-07 1.5E+01	3.97E-07 1.0E+01	3.38E-07 1.1E+01	5.90E-07 6.3E+00	4.18E-07 7.0E+00	6.74E-07 5.8E+00	7.76E-07 6.0E+00	5.61E-07 7.1E+00	3.02E-07 8.1E+00

Table 35 (continued)

BEADLEACH II

PLUTONIUM LEACH RATE (G/CM2.DAY)

CHAN TEMP FLOW SAMPLING DAYS				280	342	435
28	COLD MED	0.	0.	1.63E-07	1.62E-07	
				2.2E+00	1.9E+00	
31	COLD FAST	0.	0.	4.01E-07	4.38E-07	
				7.5E+00	6.8E+00	
62	HOT MED	0.	0.	5.04E-09	7.81E-09	
				9.9E+00	1.0E+01	
63	HOT FAST	0.	0.	9.16E-07	1.06E-06	
				5.2E+00	4.3E+00	

Table 36

BEADLEACH II

NEPTUNIUM LEACH RATE (G/CN2.DAY)

CHAN TEMP FLOW SAMPLING DAYS

		1	2	3	6	11	20	37	70	120	151	213
28	COLD MED	2.61E-06 2.1E+00	2.74E-06 2.0E+00	3.07E-06 2.1E+00	3.51E-06 2.0E+00	3.33E-06 2.1E+00	2.52E-06 1.6E+00	1.74E-06 2.2E+00	2.05E-06 2.0E+00	1.84E-06 2.0E+00	1.83E-06 2.4E+00	1.49E-06 2.2E+00
31	COLD FAST	7.07E-06 7.2E+00	5.51E-06 6.8E+00	5.69E-06 8.0E+00	3.52E-06 1.0E+01	4.02E-06 9.5E+00	3.41E-06 6.4E+00	3.89E-06 5.8E+00	1.28E-06 9.2E+00	1.91E-06 8.6E+00	1.70E-06 8.8E+00	2.45E-06 6.8E+00
62	HOT MED	1.30E-05 8.6E-01	4.30E-05 5.6E-01	4.09E-05 6.2E-01	3.61E-05 6.3E-01	5.37E-06 1.3E+00	1.27E-05 8.9E-01	3.75E-05 4.6E-01	3.35E-05 5.1E-01	4.21E-05 5.0E-01	2.95E-05 5.3E-01	3.21E-05 5.1E-01
63	HOT FAST	1.13E-04 1.3E+00	8.04E-05 2.8E+00	7.02E-05 2.6E+00	1.03E-04 1.9E+00	1.01E-04 2.0E+00	1.08E-04 1.4E+00	9.40E-05 1.4E+00	1.33E-04 1.3E+00	1.03E-04 1.6E+00	1.40E-04 1.4E+00	4.28E-05 2.1E+00

Table 36 (continued)

BEADLEACH II
NEPTUNIUM LEACH RATE (G/CM².DAY)

CHAN TEMP FLOW SAMPLING DAYS

			280	342	435
28	COLD	MED	0. 0.	1.41E-06 2.2E+00	1.51E-06 1.9E+00
31	COLD	FAST	0. 0.	4.61E-06 6.2E+00	1.88E-06 8.3E+00
62	HOT	MED	0. 0.	1.17E-05 6.5E-01	1.03E-05 6.7E-01
63	HOT	FAST	0. 0.	1.33E-04 1.3E+00	1.18E-04 1.2E+00

Table 37

BEADLEACH II

			PLUTONIUM CUMULATIVE FRACTION LEACHED(CFL)										
CHAN TEMP FLOW SAMPLING DAYS			1	2	3	6	11	20	37	70	120	151	210
28	COLD	MED	2.38E-06	5.36E-06	8.97E-06	1.84E-05	3.26E-05	4.66E-05	6.37E-05	8.89E-05	1.12E-04	1.39E-04	1.90E-04
31	COLD	FAST	1.24E-05	2.30E-05	3.15E-05	5.07E-05	7.66E-05	1.11E-04	1.80E-04	2.72E-04	3.19E-04	3.79E-04	5.21E-04
62	HOT	MED	3.89E-08	7.10E-07	9.75E-07	1.35E-06	1.71E-06	2.09E-06	3.01E-06	6.29E-06	8.78E-06	1.03E-05	1.26E-05
63	HOT	FAST	2.14E-06	2.96E-06	3.78E-06	7.90E-06	1.69E-05	3.26E-05	6.74E-05	1.49E-04	2.30E-04	3.14E-04	4.25E-04

Table 37 (continued)

BEADLEACH 11

PLUTONIUM CUMULATIVE FRACTION LEACHED(CFL)					
CHAN TEMP FLOW SAMPLING DAYS					
			280	342	435
28	COLD	MED	0.	2.75E-04	3.43E-04
31	COLD	FAST	0.	7.69E-04	9.20E-04
62	HOT	MED	0.	1.61E-05	1.87E-05
63	HOT	FAST	0.	7.50E-04	1.14E-03

Table 38

BEADLEACH II

			NEPTUNIUM CUMULATIVE FRACTION LEACHED(CFL)										
CHAN TEMP FLOW SAMPLING DAYS			1	2	3	6	11	20	37	70	120	151	213
28	COLD	MED	1.12E-05	2.20E-05	3.41E-05	7.46E-05	1.57E-04	2.48E-04	3.91E-04	6.64E-04	8.72E-04	1.10E-03	1.52E-03
31	COLD	FAST	3.01E-05	6.34E-05	8.57E-05	1.36E-04	2.27E-04	3.43E-04	5.90E-04	9.54E-04	1.13E-03	1.35E-03	1.87E-03
62	HOT	MED	5.54E-05	2.26E-04	3.88E-04	8.60E-04	1.32E-03	1.62E-03	3.38E-03	8.52E-03	1.27E-02	1.71E-02	2.49E-02
63	HOT	FAST	4.96E-04	8.24E-04	1.11E-03	2.25E-03	4.78E-03	8.21E-03	1.52E-02	3.22E-02	4.51E-02	6.09E-02	8.41E-02

Table 38 (continued)

BEADLEACH II

NEPTUNIUM CUMULATIVE FRACTION LEACHED(CFL)				
CIAN TEMP FLOW SAMPLING DAYS				
		280	342	435
28	COLD MED	0.	2.23E-03	2.80E-03
31	COLD FAST	0.	3.68E-03	4.90E-03
62	HOT MED	0.	3.45E-02	4.07E-02
63	HOT FAST	0.	1.31E-01	1.81E-01

TABLE 39.

Comparison of Bead Leach II vs. Bead Leach I

Pu Leach Rate: $\text{g/cm}^2 \cdot \text{d}$

Day	25°C - 10 ml/d		25°C - 300 ml/d		75°C - 10 ml/d		75°C - 300 ml/d	
	B.L.II (Ch 28)	B.L.I (Ch 17)	B.L.II (Ch 31)	B.L.I (Ch 15)	B.L.II (Ch 62)	B.L.I (Ch 18)	B.L.II (Ch 63)	B.L.I (Ch 16)
	$\times 10^{-7}$		$\times 10^{-6}$		$\times 10^{-8}$		$\times 10^{-7}$	
1	5.6	12.8	2.9	6.3	0.9	21.3	4.9	27.1
2	7.5	12.2	2.7	2.0	16.9	3.3	2.0	2.9
3	9.2	7.7	2.2	1.4	6.7	2.0	2.0	3.1
6	7.1	9.5	1.3	2.1	1.4	1.5	4.0	7.1
11	5.1	5.7	0.9	1.0	1.5	2.6	3.4	5.8
20	3.9	3.6	1.3	1.3	1.0	3.0	5.9	6.3
37	1.4	2.8	0.8	0.5	1.7	0.6	4.2	4.8
70	2.1	1.3	0.5	0.5	2.8	0.7	6.7	4.3
120	2.1	0.9	0.4	0.5	1.9	1.0	7.8	5.5
213	1.9	1.4	0.6	0.6	1.0	0.6	3.0	5.6
435	1.8	1.5	0.4	0.2	0.8	0.02	10.6	4.1

TABLE 40.

Comparison of Bead Leach II vs. Bead Leach I

Day	Np Leach Rate: g/cm ² ·d							
	25°C - 10 ml/d		25°C - 300 ml/d		75°C - 10 ml/d		75°C - 300 ml/d	
	B.L.II (Ch 28)	B.L.I (Ch 17)	B.L.II (Ch 31)	B.L.I (Ch 15)	B.L.II (Ch 62)	B.L.I (Ch 18)	B.L.II (Ch 63)	B.L.I (Ch 16)
	x 10 ⁻⁶		x 10 ⁻⁶		x 10 ⁻⁵		x 10 ⁻⁴	
1	2.6	3.6	7.1	12.1	1.3	5.8	1.1	1.4
2	2.7	3.3	8.5	4.2	4.3	4.2	0.8	1.2
3	3.1	2.8	5.7	3.5	4.1	3.9	0.7	1.2
6	3.5	3.9	3.5	4.5	3.8	5.0	1.0	1.4
11	3.3	2.5	4.0	2.4	5.4	5.3	1.0	1.3
20	2.5	1.8	3.4	3.6	1.3	5.4	1.1	1.5
37	1.7	2.2	3.9	1.5	3.8	3.1	0.9	1.2
70	2.1	2.0	1.3	3.0	3.4	2.8	1.3	1.2
120	1.8	1.3	1.9	2.7	4.2	2.8	1.0	1.3
213	1.5	2.5	2.5	2.5	3.2	0.2	0.4	1.3
435	1.5	1.4	1.9	0.7	1.8	2.0	1.2	1.0

TABLE 41

Comparison of Bead Leach II vs Bead Leach I
Cumulative Fractions Leached

Temperature (°C)	Flow Rate (ml/d)	Experiment	P e r c e n t	
			Pu	Np
25	10	B.L. II	0.034	0.28
25	10	B.L. I	0.028	0.32
25	300	B.L. II	0.093	0.49
25	300	B.L. I	0.089	0.37
75	10	B.L. II	0.0019	4.1
75	10	B.L. I	0.0011	3.0
75	300	B.L. II	0.11	18.1
75	300	B.L. I	0.083	19.3

VII. SUMMARY

Based on both this report and its predecessor¹ covering the Bead Leach II experiment, the following results from this study are listed:

1. PNL 76-68 Glass Beads

PNL 76-68 glass beads were leached with three leachants: simulated basalt groundwater, brine solution, and bicarbonate solution. In the case of the basalt groundwater, the beads were leached with and without crushed basalt; in the case of the brine solution, they were leached with and without crushed sandstone; no rock was present in the case of the bicarbonate. All were leached at 25°C and 75°C and at 1, 10, and 300 ml/d except in the bicarbonate case where only 10 and 300 ml/d were used.

a) The leachates and the crushed rock were analyzed for Pu and Np. The Pu leach rates range from 10^{-10} g/cm²·d to 10^{-6} g/cm²·d at 25°C and 75°C. The leaching behavior of Pu is affected by both temperature and flow rate but not in a consistent manner. *Because of its tendency to be sorbed on surfaces,* the apparent Pu leach rates are much lower than would be expected on the basis of Np leach rates and bead weight loss. Pu leaching shows no clear difference between basalt and brine leachants in this study.

b) The Np leach rates range from 10^{-8} g/cm²·d to 10^{-6} g/cm²·d at 25°C and from 10^{-5} g/cm²·d to 10^{-4} g/cm²·d at 75°C. These

leach rates show a consistent temperature and flow rate dependency, with higher leach rates at higher temperatures and at higher flow rates. There is no obvious solution effect, both the simulated basalt groundwater and the brine solution give about the same leach rates.

c) Cumulative fractions leached were calculated for Pu and Np for the three types of leachants-basalt groundwater, brine and bicarbonate solution. These values, summarized in Table 42, are average values of three channels for each set of experimental conditions except for the slow channels (1 ml/d) of which there were only two channels. The uncertainties shown are one standard deviation from the mean value. There are no uncertainties shown for the bicarbonate values because only single channels were run at each temperature and flow rate. The cumulative fraction values in Table 42, include the amount of the leached Pu and Np that was adsorbed on the rock in those channels with both glass beads and crushed basalt or sandstone.

As can be seen from the data, there are temperature and flow rate effects for each type of leachant. The cumulative fractions leached increase with temperature and flow rate although the Pu values at 75°C carry large uncertainties. Because of these, the effect of solution type is confusing at 75°C. In the case of Np, however, the effect of solution type is clear and consistent. The Np data suggest that PNL 76-68

TABLE 42

Summary of Cumulative Fraction Leached, Day 440

PNL 76-68 Glass

Temp.	Flow Rate	Percent Pu			Percent Np		
		Basalt	Brine	Bicarb	Basalt	Brine	Bicarb
		$\times 10^{-3}$	$\times 10^{-3}$	$\times 10^{-3}$			
25	10	1.20 $\pm$ 0.02	3.5 $\pm$ 0.7	34.3	0.031 $\pm$ 0.007	0.046 $\pm$ 0.006	0.28
25	300	10.7 $\pm$ 2.7	6.5 $\pm$ 0.4	92.8	0.17 $\pm$ 0.03	0.075 $\pm$ 0.007	0.49
75	1	1.2 $\pm$ 0.3	2.5 $\pm$ 0.01	--	0.67 $\pm$ 0.20	0.97 $\pm$ 0.19	--
75	10	7.7 $\pm$ 7.4	5.8 $\pm$ 4.5	1.9	1.2 $\pm$ 0.1	2.6 $\pm$ 0.3	4.1
75	300	114 $\pm$ 145	200 $\pm$ 180	114	7.9 $\pm$ 1.0	10.5 $\pm$ 1.7	18.1

glass is more resistant to leaching with simulated basalt ground-water, less with brine solution, and least resistant to bicarbonate solution leaching. The agreement between Np cumulative fraction leached and bead weight loss indicates that congruent dissolution is the dominant leaching mechanism for Np.

d) The main purpose of the Bead Leach II experiment was to study the adsorption of Pu and Np on crushed basalt and sandstone. In the glass beads - Umtanum basalt - simulated basalt groundwater scenario¹, 69 to 99% of the leached Pu is adsorbed on the crushed basalt with the major portion of it sticking to the rock immediately adjacent to the "waste-form" regardless of temperature and flow rate. In the glass beads - Nugget sandstone - brine solution scenario, Pu is adsorbed from 8% at 25°C and 300 ml/d to 98% at 75°C and 1 ml/d. We have also determined that Np is not significantly adsorbed (not quantifiable) on either basalt or sandstone at 25°C. Of the total Np leached at 75°C in the case of basalt, 68% is adsorbed at 1 ml/d, 61% at 10 ml/d, and 19% at 300 ml/d while only 11%, 3.1%, and 0.5% is adsorbed at the corresponding flow rates in the case of sandstone. In both cases the retardation of Np is uniform in the rock column above the glass beads. These Np data clearly illustrate the effect of flow rate on adsorption of Np by either basalt or sandstone. The summary of these data is shown in Table 43. The sandstone values represent an average of two channels whenever applicable except in the case of channels 42 and 43 (75°C - 300 ml/day). Here, only the values from channel 43 are shown because of an anomalous value (sample A) in channel 42. The basalt values in Table 43 are also represented in the same manner and the reader is advised to refer to a previous report¹ for more detail.

TABLE 43

Summary of Adsorption
Plutonium (%)

	25°C-10 ml/d		25°C-300 ml/d			75°C-1 ml/d		75°C-10 ml/d		75°C-300 ml/d	
Layer	Basalt*	Sandstone	Basalt*	Sandstone		Basalt*	Sandstone	Basalt*	Sandstone	Basalt*	Sandstone
D	8	10	11	1		3	10	3	13	4	2
C	7	11	9	1		4	11	4	18	9	3
B	28	15	19	2		17	25	10	21	18	9
A	47	20	30	4		74	52	77	28	68	12
TOTAL	90	56	69	8		98	98	94	80	99	26

Neptunium (%)

	75°C - 1 ml/d		75°C - 10 ml/d		75°C - 300 ml/d	
Layer	Basalt*	Sandstone	Basalt*	Sandstone	Basalt*	Sandstone
D	14	3	12	0.6	4	0.1
C	20	3	16	0.9	4	0.1
B	19	3	19	0.9	6	0.1
A	15	2	14	0.7	5	0.2
TOTAL	68	11	61	3.1	19	0.5

* See Reference 1 for the detailed report.

e) Adsorption of other leached species was also studied but only in the case of basalt. Based on our findings, as described previously¹, U, Cs, and Mo behave similarly to Np at 25°C; that is, adsorption is not measurable. Based on one channel alone, B partitions equally between rock and leachate at 25°C (the cumulative fraction leached for B at day 440, however, is only 6.5×10^{-4} .) At 75°C, Mo adsorption is not detectable. Thus, we conclude that Mo is not adsorbed on basalt regardless of temperature and flow rate conditions. U is adsorbed at 75°C, from 38% at 1 ml/d, to 15% at 10 ml/d, to 13% at 300 ml/d. Of the total B leached at 75°C, only 2% appears to be adsorbed, and 40 and 80% of the leached Cs is adsorbed at 10 ml/d and 1 ml/d respectively.

2. UO₂ Pellets

a) UO₂ pellets were leached with three leachants: brine solution, simulated basalt groundwater, and bicarbonate solution. In the case of the brine solution, they were leached with and without crushed sandstone; in the case of the basalt groundwater, they were leached with and without crushed basalt; and in the case of the bicarbonate solution they were leached without any rock interaction. All were leached at 25°C and 75°C and at 1, 10, and 300 ml/day, except in the bicarbonate case where only 10 and 300 ml/day were used. Calculated leach rates range from 10^{-8} g/cm²·d to 10^{-6} g/cm²·d for samples leached at medium (10 ml/d) and fast flow rates (300 ml/d) regardless of temperature. Samples leached at slow flow rates (1 ml/d) appear to give leach rates on the order of 10^{-9} g/cm²·d. In bicarbonate, UO₂ leaches

at rates of 10^{-5} g/cm²·d to 10^{-4} g/cm²·d depending on flow rate and temperature. Thus, solution type may strongly influence leach rates although the difference between basalt groundwater and brine is not clear. The presence of crushed rock in the system may or may not diminish leach rates; the data are not conclusive enough to say that uranium is adsorbed on either basalt or sandstone.

b) Cumulative fractions leached were calculated. A summary of these results at day 440 of the experiment is shown in Table 44. We have tabulated the results differentiating samples leached with and without crushed rock. It must be recognized that the cumulative fractions leached for the samples leached with crushed rock in the system may be low because of the adsorbed uranium (if any) on the rock which was not quantified. Moreover, some of the results in Table 44 are shown with an uncertainty which is the standard deviation from the mean of two values indicating that the same experimental conditions were reproduced in duplicate. The results reported in Table 44 without an associated uncertainty indicate that only a single sample was leached. In general, the bicarbonate cumulative fractions are higher than either the basalt or the brine and the basalt cumulative fractions leached appear to be higher than the brine. The higher concentration of HCO_3^- in the bicarbonate leachant (about 30 times higher) may be the reason for the higher cumulative fractions leached. Leaching may be enhanced by $\text{CO}_3^{=}$ complexation. Temperature, flow rate, and solution type effects are all more evident in the cumulative fraction leached data than in the leach rate data.

TABLE 44
Summary of Cumulative Fraction Leached, Day 440
UO₂ Pellets

Experimental Conditions			Percent Uranium		
Temp (°C)	Flow Rate (ml/d)	Cell Configuration	Brine	Basalt	Bicarb
25	10	A	<0.0043	0.0034	0.40
		P	<0.0045	0.0037+0.0023	--
25	300	A	0.0052	0.015+0.001	0.48
		P	0.0074+0.0008	0.012	--
75	1	P	<0.0037	0.00011+0.00004	--
75	10	A	<0.0013	0.0044+0.0007	1.0
		P	<0.0016	0.0015	--
75	300	A	0.0037	0.0068	6.2
		P	0.0049	0.0060+0.0001	--

A - UO₂ only

P - UO₂ and crushed basalt or sandstone

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REFERENCES

1. Bazan, F., Rego, J., Failor, R., and Coles, D.G., "A Waste Form/Rock Interaction Leaching Study Using PNL 76-68 Glass Beads and Umtanum Basalt" Part I, LLNL Report UCID 19764, March 1983.
2. Shultz-McKinley, S., Garrison, J. McLean, S., and Coles, D. G., "Chemical, Physical, and Mineralogical Characterization of Fifteen Sandstones for Radionuclide Sorption Studies, LLNL Report UCID 19034, April 1981.
3. Molecke, M. A., "A Comparison of Brines Relevant to Nuclear Waste Experimentation," Sandia Report SAND 83-0516, May 1983.
4. Coles, D. G., Mensing, R. W., Rego, J., Weed, H. C., and Buddemeier, R. W., "A Leaching Study of PNL 76-68 Glass Beads Using the LLNL Continuous - Flow Method and the PNL Modified IAEA Method: A Final Report," LLNL Report UCID 19492, Revision 1, August 1982.
5. Davis, N., "Fabrication of UO_2 Pellets - Work Order A 53914" PNL Memo, June 1980.