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Sorption-Desorption Studies of Nevada Test Site

Alluvium and Leaching Studies of

Nuclear Test Debris

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SORPTION-DESORPTION STUDIES OF NEVADA TEST SITE

ALLUVIUM AND LEACHING STUDIES OF NUCLEAR

TEST DEBRIS

by

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ABSTRACT

Distribution coefficients were determined for sorption-desorption of radioactive tracers between Nevada Test Site alluvium and alluvial ground water. The order of increasing distribution coefficient by element is: Sb, Sr, U(VI), I, Ru, Nb, Ba, Cs, Co, Y, Ce, and Eu. Some distribution data were also obtained for bentonite. Volatile and refractory debris samples from nuclear tests in tuff, alluvium, and rhyolite were leached with ground water. Most leachable are the volatile species I, Mo, and Sb, which have condensed on chimney rubble. Comparison of the laboratory data with field measurements indicates that leaching is the main process responsible for the presence of radionuclides in water from the site of the underground nuclear test Cambric.

I. INTRODUCTION

This report presents results of laboratory investigations for the Radionuclide Migration Project[1] sponsored by the Nevada Operations Office, Department of Energy (DOE), which involves the study of possible migration of radionuclides from sites of underground nuclear explosions. Previous publications in support of this project are a review of existing pertinent information[2] and an investigation of radionuclide distribution in the environment of the Cambric nuclear explosion after ten years.[1] The purpose

of the laboratory studies is to obtain information that can be correlated with existing information from field studies and that can be used to predict migration behavior from sites for which no field information is available. The information gained may also be pertinent in describing migration of nuclides from nuclear waste repositories.

The phenomenology of underground nuclear explosions is reviewed in Ref. 2. Several points are pertinent to migration. Most of the refractory nuclides (those with boiling points high enough for condensation out of the vapor phase by the time of cavity collapse, e.g., ^{239}Pu, ^{95}Zr, ^{147}Nd) are retained in fused material (also called rock melt and puddle glass) which is found near the bottom of the cavity formed by the nuclear explosion and in the lower regions of the chimney of material that collapses into the cavity as it cools. The chimney may or may not propagate to the surface Nuclides which are volatile or which have volatile precursors (e.g., ^{90}Sr, ^{137}Cs, ^{91}Y, ^{131}I, and ^{125}Sb) are depleted in the fused material and are found in higher regions of the chimney where they have condensed. The distribution of ^{85}Kr and ^{3}H are discussed in Ref. 1. Migration with ground water, of course, is not possible unless at least part of the cavity is below the water table and it has been refilled with water. For migration to occur by solution mechanisms it is necessary for a nuclide to dissolve from the debris. This initial dissolving, be it from fused material or from chimney rubble, is called leaching. As a nuclide is transported by ground water it may sorb and desorb from solids. Possible mechanisms may be ion exchange, precipitation, and other phenomena. Transport of radionuclides on suspensions or colloids is not considered in this report.

Sorption-desorption ion-exchange reactions are the basis for chromatographic work. A commonly used parameter for describing such phenomena at equilibrium in ion-exchange work is the distribution coefficient, K_d (see, for example, Ref. 3), which is equal to the concentration of a species on a solid phase divided by its concentration in the liquid phase. Many investigators have adopted the same

parameter in modeling the transport of nuclides through an underground medium by ground water (see, for example, Refs. 4 and 5). This work will report on measurements of the distribution coefficients for a number of nuclides for sorption-desorption on alluvium from Areas 3 and 5 of the Nevada Test Site (NTS).

Most of the laboratory leaching experiments reported in Ref. 2 involve leaching of fused debris. The nuclides other than ^{3}H that were detected in the water pumped from the cavity and chimney of the Cambric event are ^{90}Sr, ^{137}Cs, ^{106}Ru, and ^{137}Cs. These are volatile or have volatile precursors and are depleted in the fused material. Leaching of nuclides from material on which volatiles have condensed, as well as from the fused material, is reported.

II. MATERIALS AND THEIR CHARACTERIZATION

These studies are performed with the actual type of media from which the nuclides would be leached and through which they would be transported and also with the ground water present in the vicinity.

A. Alluvium and Bentonite

Most experiments were conducted with material obtained in the Cambric investigation[1] from hole U5e (RNM-1) in Frenchman Flat from a vertical depth of 256 m, and some from 331 m. Material for some other experiments came from a preshot sample taken at a depth of 238 m from hole U3bv in Yucca Flat. All samples were taken below the water table. A geologic description[6] of the unsorted material from the Frenchman Flat samples follows:

Petrographic Description

Alluvium consists of mineral and tuff fragments in an iron-oxide-rich, fine-grained matrix, not strongly consolidated.

Major constituents large enough to be identified optically are quartz, plagioclase, alkali feldspar, calcite, glass shards, and opaques.

Minor constituents large enough to be identified optically are amphibole, biotite, orthopyroxene, and clinopyroxene.

X-ray Data:

Glass (broad hump), plagioclase, alkali feldspar, and minor quartz.

Microprobe Data:

Fresh glass, also fine-grained devitrification products (probable zeolites)

$Na_{0.76 \pm 0.11}K_{0.61 \pm 0.07}Fe_{0.05 \pm 0.02}Mg_{0.02 \pm 0.02}Al_{1.43 \pm 0.05}Si_{7.52 \pm 0.06}O_{18} \cdot nH_2O$

Plagioclase phenocrysts

$Na_{0.55 \pm 0.02}Ca_{0.46 \pm 0.04}K_{0.03 \pm 10.01}Al_{1.47 \pm 0.03}Si_{2.96 \pm 0.02}O_8$

Alkali feldspar

$Na_{0.43 \pm 0.03}Ca_{0.02 \pm 0.01}K_{0.58 \pm 0.03}Al_{1.04 \pm 0.02}O_8$

Calcite (one analysis)

$Ca_{0.99}Mg_{0.01}CO_3$

Clinopyroxene (one analysis)

$Ca_{0.85}Mg_{0.77}Fe_{0.32}Si_{2.0}O_6$

Orthopyroxene

$Ca_{0.04}Mg_{1.30}Fe_{0.72}Si_{1.95}O_6$

The alluvium was dried at 110°C, and portions of it were graded by sieving.

Bentonite is the material often used in drilling mud at NTS. It has been suggested that the clays or zeolites in this material may offer a much better absorbing medium for radionuclides than the geologic medium. The material was dried and used without grading.

Because we are interested in the behavior of the samples in their natural state, no attempt was made to purify the materials or change the ions already on the sorption sites. Cation exchange capacity was determined by shaking separate 100-mg portions of the solids with 20 ml of 0.5 $\underline{M}$ CsCl (pH = 8.2) or 0.5 $\underline{M}$ SrAc$_2$ (pH = 8.5) in deionized water, spiked with ^{137}Cs or ^{85}Sr, respectively, for 3 to 5 days. The pH of these solutions was not adjusted because it was sufficiently close to that of the ground waters. After washing the solids with water, they were counted on a NaI detector to determine the amount of Sr or Cs that exchanged. Portions of the samples were also sent to the Lawrence Livermore Laboratory (LLL) for determination of surface area by nitrogen absorption (which, we recognize, may not be applicable) and sieve analysis. The results of these characterizations are given in Table I.

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TABLE I

CATION EXCHANGE CAPACITY, SURFACE AREA, AND SIEVE ANALYSES

Material[c]	Mesh Size	Cation Exchange Capacity (meq/100 g) Sr	Cs	Surface Area[a] (m^2/g)	Sieve Analyses[a,b] (μm) 10%	50%	90%
Alluvium							
U5e, 256 m	100 - 200	14	10	14.9	70	105	155
U5e, 256 m[d]	100 - 200	16	13	12.4	29	90	175
U5e, 256 m	200 - 400		17	20.2	26	50	71
U5e, 256 m	+ 100	10		6.11	200	780	4000
U5e, 331 m	ungraded	16					
U3bv, 238 m	100 - 200	16	12	11.7	61	95	122
U3bv, 238 m	+ 100	16					
U3bv, 238 m	ungraded	22		9.2	87	270	2000
Bentonite	ungraded	56	72	33.8	21	35	85
Test Debris							
U7ajs[e]	100 - 200			15.5	55	105	142
U7ajs[g]	100 - 200			16.5	58	105	145
U20n[e,g]	100 - 200			11.6	30	63	135
U20N[e,g]	200 - 325[h]			6.7	42	83	130
U20n[f,g]	100 - 200			2.1	26	72	140
U20n[f]	200 - 325[h]			0.63	20	29	59
U3km[g]	ground			1.2	46	92	140
U3ki	ground			0.79	23	51	115

[a]Surface area and sieve analyses arranged for by D. G. Coles (LLL) and performed by A. H. Bierman (LLL).
[b]Particle size diameters corresponding to 10, 50, and 90 percentiles of the cumulative distribution.
[c]The material is labeled by hole designation and depth.
[d]Ground alluvium. All others are sievings of natural sizes.
[e]Not high-graded; refractory.
[f]Not high-graded; volatile.
[g]Bimodal distribution.
[h]Fractions were not used in further studies.

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Neither the cation exchange capacity nor the gas-absorption surface area varies more than about a factor of two with the sieve size, the inverse of the 50% diameter,or the source of alluvium. However, the relationship between the surface area and cation exchange capacity is also no better than a factor of two. The diameters of the particles as calculated from the gas-absorption measurements are in the 0.2-to 0.4-μm range, much smaller than the sieve sizes. These data, together with the cation exchange capacity measurements, indicate that internal surface area must play a major role in sorption for alluvium. The ungraded bentonite has a somewhat greater cation exchange capacity and surface area.

B. Test Debris

Debris samples were recovered by sidewall-core sampling of holes drilled into the chimneys and puddles of underground nuclear tests in several geologic media. In general, a leaching experiment was started within a few weeks of the test because of the increased sensitivity obtained from working with relatively short-lived nuclides. Debris from hole U20n in rhyolite did not become available until ≈180 days after the test, and the experiments with this material had lower sensitivity than those done at earlier times.

Refractory debris samples were obtained from three tests in alluvium in Yucca Flat. Material from holes U3km, U3ki, and U3kf was high-graded by picking out fused chunks having high gamma activity. This material was ground in a steel ball mill to a powder. Some of the material from hole U3kf was not ground after high-grading, and chunks having dimensions ≈2 to 5 mm were selected.

Samples from one core containing refractory products and another containing volatile products from hole U7ajs in tuff in Yucca Flat were dried under a heat lamp. Approximately 10% of the ^{131}I was lost in the heat treatment. The dried samples were sieved, and the 100- to 200-mesh fractions were retained for leaching. Refractory material from hole U7ap also in tuff was high-graded, and small 2- to 5-mm chunks were selected.

Dried debris from volatile and refractory samples from a shot in rhyolitic material--hole U20n on Pahute Mesa--was sieved without high-grading, and the 100- to 200-mesh fractions were used.

Surface area and sieve analyses for some of the shot debris are given in Table I. It is noteworthy that the high-graded, fused debris

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from holes U3km and U3ki was ground to dimensions corresponding to smaller than 100 mesh, but the surface area is less than 10% of that of alluvium of similar size, indicating that fusing of the geologic medium has decreased the internal area. The debris samples from hole U7ajs that were not high-graded have surface areas comparable to alluvium of similar size, indicating that they contain much unchanged material. The rhyolitic material from hole U20n probably has less internal surface area than alluvium and tuff.

The shot debris samples are also characterized by radionuclides in Table II.* In general, one observes a higher number of atoms of ^{95}Zr (a refractory nuclide) in the high-graded refractory samples than in the non-high-graded refractory samples. Nuclides of Sr, Mo, Ru, Sb, Te, I, and Cs are enriched relative to Zr in the volatile samples and depleted in the refractory samples. Although ^{89}Sr was found in the volatile sample from hole U7ajs, no ^{90}Sr was found in this sample; also, ^{140}Ba is depleted in this sample. These facts indicate that the location in the chimney where volatile species or their daughters condense is a function of both the half-lives of the nuclides in the chain and their volatilities.

C. Ground Water

The ground water for the sorption experiments and for the leaching experiments of debris from shots in alluvium and tuff was taken from well RNM-2S[1] in Frenchman Flat at a depth of 341 m in alluvium. The well is 90 m from hole RNM-1, from which most of the alluvium in this work came. The water represents a sampling of the well from November 13 to 25, 1975. Ground water for the leaching experiments of debris from a shot in rhyolitic material came from well Ue19c on June 23, 1976. The well samples water at 2587 m in a formation of rhyolitic composition.

The composition of the two ground waters is given in Table III. The composition of water from RNM-2S is similar to that of water from other wells sampling aquifers in alluvium (e.g., wells 5B and 5C in

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*For simplification, the convention that the entry aEn means a x 10^n is used in this report.

TABLE II
NUCLIDE DATA FOR SHOT DEBRIS

Nuclide	U7ajs[a,b] (volatile)	U7ajs[a,c]	U3km[c,d]	U3ki[c,d]	U7ap[c,e]	U7ap[c,e,f]	U20n[a,c]	U20n[a,g] (volatile)
			Atoms per Atom of ^{95}Zr at Zero Time					
^{59}Fe					3.5E-3	3.7E-2		
^{54}Mn					2E-2	2E-2	3.5E-2	3.4E-2
^{58}Co					3.4E-3	3.4E-3	4.2E-3	6.2E-3
^{60}Co							8.2E-3	8.5E-3
^{88}Y			1.5E-2	9E-3	8.8E-4	9.6E-4	1.4E-2	1.4E-2
^{89}Sr		7.91						
^{99}Mo	6.9E-1	13.7						
^{103}Ru	9E-2	16.8	4.9E-1	3.5E-1	1.6E-1	8.6E-2		
^{106}Ru			2.9E-1	1.1E-1			7.3E-2	4.4E-1
^{124}Sb			7.6E-3	1.9E-2	2.6E-3	1.5E-3		
^{125}Sb							2.7E-2	1.1E-1
^{126}Sb			5.8E-3	3E-3				
^{127}Sb	1.9E-2	12.7						
^{129}Te^{m}			7.3E-2	4.0E-2	9.1E-3	1.2E-2		
^{131}I	8.4E-2	10.7	2.7E-1	1.5E-1	9.5E-2	6.1E-2		
^{132}Te	8.6E-2	23.4						
^{136}Cs	4.2E-3	6.1E-1			5.5E-3	4.8E-3		
^{137}Cs			3.4E-1	1.3E-1			5.6E-2	2.56
^{140}Ba	3.5E-1	3.2E-1	1.26	6.4E-1	2.9E-1	3.7E-2		
^{141}Ce	5.5E-1	1.1E-1	1.32	8.5E-1	5.7E-1	6.4E-1	9.7E-1	8.9E-1
^{144}Ce	7.0E-1	<3.8E-1			7.9E-1		8.1E-1	7.3E-1
^{182}Ta			4.2E-2		4.9E-3	5.7E-3	1.1E-1	1.2E-1
			Atoms ^{95}Zr Per Gram					
	3.0E12	3.8E11	7.8E12	2.1E13	3.7E12	1.4E13	1.4E12	3.2E12

[a]Material was not high-graded.
[b]Material taken 47 m above puddle.
[c]Material taken in the vicinity of the puddle.
[d]Fused material was high-graded and ground.
[e]Fused chunks, 2-5 mm across.
[f]Material used for run 4760 in argon.
[g]Material taken 45 m above puddle.

TABLE III

COMPOSITION OF GROUND WATERS[a]

	Alluvial Water[b]	Rhyolitic Water[c]
	(mg/1)	
Calcium (Ca)	18	4.2
Magnesium (Mg)	5.5	0.0
Potassium (K)	10	0.8
Sodium (Na)	66	110
Lithium (Li)	0.030	0.120
Strontium (Sr)	0.110	0.030
Bicarbonate (HCO_3)	170	240
Carbonate (CO_3)	0	0
Chloride (Cl)	23	3.9
Sulfate (SO_4)	41	21
Fluoride (F)	0.6	9.9
Silica dissolved (SiO_2)	62	44
Alk. Tot. as $CaCO_3$	140	200
Residue, 180°C	314	308
Residue, Calc.	310	312
	Meq/1	
Total Cations	4.48	5.02
Total Anions	4.32	5.00
	pH	
	8.5	8.3

[a]Analyses provided by W. W. Dudley, Jr. (USGS).
[b]Pumped from well RNM-2, November 13-24, 1975.
[c]Pumped from well 19C3, June 23, 1976.

Frenchman Flat) and in tuff (e.g., Test Well A and Test Hole 7 in Yucca Flat and Well J-12 in Jackass Flats).[7] The resemblance of water from RNM-2S to the water from tuff formations is probably due to detrital tuff rather than detrital limestone or dolomite being a major constituent of the alluvium. However, note that the sum of the chloride and sulfate concentration in waters from alluvium is ≈60 ppm, whereas that of waters from tuff is ≈20 ppm.

The deionized water used has a Na concentration of <1 ppb and a resistivity of >18 MΩ-cm. The pH is 5.9-6.6.

III. SORPTION EXPERIMENTS
Experimental

The equilibrium distribution coefficient, K_d, for the distribution of activity is conventionally defined as:

$$K_d = \frac{\text{activity in solid phase per unit mass of solid}}{\text{activity in solution per unit volume of solution}}.$$

In most of these experiments, the activity was counted only in the solutions to retain identical counting geometry and, consequently, the fraction of the activity, f, remaining in the liquid phase after contact with a solid was measured. The equation for the distribution coefficient can then be expressed:

$$K_d = \left(\frac{1-f}{f}\right)\left(\frac{\text{ml of solution}}{\text{g of solid}}\right).$$

It must be noted that, under many conditions, it is not known whether equilibrium is achieved. However, the distribution of activities between phases is measured and the resulting value is called the distribution ratio, R_d, which is identical to K_d, but does not imply equilibrium.

Three types of traced solutions were prepared for the sorption studies. The first was prepared by evaporating a mixture of the following commercially available "carrier-free" nuclides to dryness in a polyethelyene tube: ^{144}Ce, ^{152}Eu, ^{133}Ba, ^{85}Sr, ^{95}Zr-^{95}Nb, ^{137}Cs, ^{85}Sr, ^{60}Co, and ^{124}Sb. Concentrated HCl was added, and the mixture was taken dry again to convert the salts to chlorides. Then ground water was

added, and, after stirring, the mixture was centrifuged for 1 h at 16 000 rpm. The supernate was centrifuged a second and a third time in fresh tubes and was then diluted with more ground water and stored in polyethylene. The ^{95}Zr was not soluble in the ground water. The pH of the spiked ground water remained at 8.5, and no activity was deposited on the polyethylene bottle or on the polyethylene tubes in which the experiments were performed. Similar spiked solutions of ^{88}Y and ^{237}U in both ground water and deionized water were prepared. The uranium was in the form of uranyl ion. A last type of radioactive solution was prepared by leaching a tuff sample containing volatile fission products from the underground nuclear explosion in hole U7ajs. The ground-water leachate contained mostly ^{131}I, and also measurable quantities of ^{99}Mo, ^{103}Ru, and $^{124, 126, 127}Sb$. Ascending paper chromatography revealed at least three iodine species having the following R_f values and per cent of total of iodine: 0.02, 10%; 0.25, 40%; 0.68, 50%. These species are probably IO_4^-, IO_3^-, and I^-, respectively.[8]

Batch sorption experiments were performed by shaking weighed quantities of a specified solid, which had been pre-equilibrated with ground water, with 20 ml of tagged water for a given time in stoppered polyethylene centrifuge tubes at room temperature (16°-24^oC) or in sealed polyethylene bottles at 70^oC. At the end of the shaking period, the aqueous phase was separated from solids for analysis by three centrifugings, each in a new polyethylene centrifuge tube, for 1 h at 16 000 rpm. Desorption experiments were performed by shaking the solids from the sorption experiments with 20 ml of ground water. Again, the aqueous phase was separated for analysis.

A 9- or 10-ml aliquot of the aqueous phase was acidified and counted on a calibrated, 95-cm^3, coaxial Ge(Li) detector. An aliquot of the original solution before contact with solids was counted for comparison to determine the fraction sorbed or desorbed. Intensities of individual peaks in the spectra were resolved, and atoms of nuclides present at zero time were determined with the GAMANAL code.[9] A limit for the number of atoms derived from the code represents 2σ uncertainty for the gamma-spectrum continuum in the region of a specified abundant gamma ray for a nuclide when that peak is not found.

The pH of the water was measured after many of the experiments. There was very little change in pH after alluvial ground water (initial pH 8.5) was contacted with alluvium or bentonite; final pH values ranged randomly between 8.3 and 8.7. The pH of the deionized water (initial pH ≈ 6) increased to values of 8 to 8.6 after contact with alluvium and to ≈ 9 with bentonite. The potential of a platinum half-cell immersed in the solutions was measured relative to a calomel half-cell (244 mV). All the ground waters measured in the laboratory gave values of 400 to 435 mV before or after contact with alluvium or bentonite. According to Garrels and Christ,[10] the dissolved oxygen from air provides a mild oxidizing effect, and one should expect Eh values between 300 and 350 mV at a pH of 8. The potential of water fresh from the RNM-2S well was measured in the field, before the water had contacted the atmosphere. A value of 330 mV for the platinum half-cell was obtained, still indicating a mild oxidizing effect. The pH of the water at that time was 8.0. We are aware of the difficulties in making good Eh measurements, of the problems with electrode measurements, and of problems with interpreting such measurements. The higher pH values observed in the laboratory are probably due to loss of CO_2 from the water.

The R_d values obtained from each experiment with alluvium are shown in Tables IV and V; those for bentonite in Tables VI and VII. The reader should carefully note the type of alluvium, temperature, and other conditions explained in the footnotes. Ratios obtained from desorption experiments follow those from sorption experiments.

In examining Tables IV-VII one can make some general observations as well as obtain K_d's which are R_d's at equilibrium or steady state. The scatter in the apparent equilibrium values is larger than the experimental uncertainties, which are usually $\leqslant 10\%$. Consequently, strictly identical samples or conditions were not sampled and the standard deviation of the population is given as a more realistic error rather than the standard deviation of the average as the error for a K_d. Species with low K_d values attain equilibrium by sorption in times of less than one week whereas those with large K_d values require three to four weeks.

TABLE IV

DISTRIBUTION RATIOS FOR ALLUVIUM

Experiment[a,g]	Mesh Size	Shaking Time (days)	Distribution Ratio, R_d (ml/g)						
			Sr	Ba	Cs	Co	Y	Eu(III)	Ce(III)
4870 (70°C)	100–200[d]	2.7	416	5620	5840	17500		28800	>28000
4870	100–200[d]	2.7	284	2910	3810	9710		>20000	>24000
4090	100–200	2.8	219	1680	3600	3440		>4800	>300
4100	100–200[d]	11.6	272	3360	8090	7800		>5270	>720
4800 (70°C)	100–200[d]	23.7	629	9100	6520	16800		22000	>20000
4860[b] (70°C)	200–400	23.7	48	973	2670	9560		>59000	>13000
4850[b] (70°C)	Ungraded	23.7	68	1100	7420	11200		7950	>13000
4810	200–400	23.7	433	7990	9140	14300		48500	>24000
4820	+100	23.7	569	8690	7880	21100		14100	>23000
4830[c]	Ungraded	23.7	151	4080	6510	14000		42000	>20000
4840[b]	Ungraded	23.7	109	2010	8100	44900		7640	>20000
4890 (70°C)	100–200[d]	23.9	563	8417	5730	19600		22000	>23000
4800	100–200[d]	23.9	266	4740	6300	14300		>90000	>20000
4110	100–200	27.6	205	4150	9150	12100			>500
4170[h]	100–200	27.8					13700		
4130[e,h]	100–200	27.8					24600		
4120	100–200	41.8	186	6160	10400	13400		>640	>1090
4140[e,h]	100–200	42.9					36100		
4131[e,f,h]	100–200	34.7					117000		
4093[f]	100–200	39.0	182	5420	10600	25000		>27000	>9100
4181[f,h]	100–200	49.0					17700		
4172[f,h]	100–200	53.8					12900		
4103[f]	100–200	144.1	165	3380	7310	21900		>39000	>2000
4112[f]	100–200	144.6	177	3570	6230	37700		12400	>2400

[a]Unless otherwise specified, samples are from U5e, 256 m; weight of alluvium was ∿1 g. Ground water (20 ml) from RNM-2S was used. Experiment involved sorption from solution.

[b]From U3bv, 238 m.

[c]From U5e, 331 m.

[d]Different sieving from rest of 100–200 mesh from same source.

[e]Deionized water was used.

[f]Experiment involved desorption from solid.

[g]Unless otherwise indicated, experiment conducted at room temperature (16° to 24°C).

[h]0.5 g of alluvium.

TABLE V

DISTRIBUTION RATIOS FOR ALLUVIUM

Experiment[a,g]	Size	(days)	Distribution Ratio, R_d (ml/g)					
			Sb	U(VI)	I	Mo	Nb	Ru
4870 (70°C)	100-200[d]	2.7	8.63					
4780	100-200[d]	2.7	3.90					
4430[i]	100-200	2.7	25.3		16.6	15.8		
4090	100-200	2.8	5.33				529	
4100	100-200	11.6	6.35				1520	
4440[h]	100-200	11.7	33.7		25.0	23.4		976
4450[i]	100-200	18.7	29.4		23.7	23.4		1020
4880 (70°C)	100-200	23.7	12.5					
4860 (70°C)	200-400	23.7	4.67					
4850[b] (70°C)	Ungraded	23.7	2.43					
4810	200-400	23.7	6.26					
4820	+100	23.7	16.3					
4830[c]	Ungraded	23.7	5.40					
4840[b]	Ungraded	23.7	1.84					
4890 (70°C)	100-200[d]	23.9	12.5					
4800	100-200[d]	23.9	8.43					
4110	100-200	27.6	6.35				2800	
4170[h]	100-200	27.8		7.45				
4130[e,h]	100-200	27.8		55.9				
4120	100-200	41.8	6.32				2720	
4180	100-200	42.9		5.75				
4140[e,h]	100-200	42.9		65.3				
4460[i]	100-200	60.7	36.4		24.7			1120
4431[f,h,i]	100-200	24.8	195		421			1630
4171[f,h]	100-200	34.8		8.9				
4093[f]	100-200	39.0	116				5100	
4441[f,i]	100-200	48.8	235		858			3390
4103[f]	100-200	144.1	71				2630	
4112[f]	100-200	144.6	58				2640	

[a-h] See footnotes in Table IV.

[i] Tagged solution from leaching of material from an underground nuclear test was used.

TABLE. VI

DISTRIBUTION RATIOS FOR BENTONITE

Distribution Ratio, R_d $(ml/g)^c$

Experiment	Weight (g)	Shaking Time (days)	Sr	Ba	Cs	Co	Y	Eu(III)	Ce(III)
4010	2	2.8	1380	1790	1580	637		>280	>190
4050	1	2.8	2640	2800	1780	1610		>5400	>360
4029	2	11.6	904	1660	1420	428		>1490	>330
4060	1	11.6	1130	1560	1570	504		1430	>540
4030	2	27.6	1110	2590	1480	460		1450	>280
4070	1	27.6	2040	3340	2080	990			>510
4230	0.5	27.9					3890		
4120[a]	0.5	27.9					5830		
4040	2	41.8	2490	14800	1890	2080		>4500	>300
4080	1	41.8	1140	6880	1940	2260			>660
4240	0.5	42.9					1580		
4220[a]	0.5	42.9					3840		
4231[b]	0.5	34.8					3880		
4211[a,b]	0.5	34.8					3490		
4013[b]	2	39.0	2510	7350	2190	1510		>5900	>740
4053[b]	1	39.0	2370	6920	2040	4800		>14000	>1520
4212[a,b]	0.5	53.8					44600		
4241[b]	0.5	65.7					5610		
4221[a,b]	0.5	65.7					16500		
4023[b]	2	141.1	2900	5380	1960	2600		>5740	>1060
4063[b]	1	141.1	2310	4730	2280	7610			>2200
4032[b]	2	144.6	2680	6100	1800	2950		>14000	>1470
4072[b]	1	144.6	2690	5450	2150	9410		24100	>2400

[a] Deionized water was used.

[b] Experiments involved desorption from solids.

[c] All experiments conducted at room temperature.

TABLE VII

DISTRIBUTION RATIOS FOR BENTONITE

Experiment	Weight (g)	Shaking Time (days)	Distribution Ratio, R_d (ml/g)[c]		
			Sb	U(VI)	Nb
4010	2	2.8	5.80		184
4050	1	2.8	6.90		940
4029	2	11.6	5.20		511
4060	1	11.6	8.51		505
4030	2	27.6	5.35		465
4070	1	27.6	6.57		843
4230	0.5	27.9		77.8	
4210[a]	0.5	27.9		334	
4040	2	41.8	4.97		465
4080	1	41.8	6.51		1690
4240	0.5	42.9		73.1	
4220[a]				214	
4231[b]	0.5	34.8		271	
4211[a,b]	0.5	34.8		168	
4013[b]	2	39.0	22.8		1490
4053[b]	1	39.0	58.7		2901
4023[b]	2	141.1	10.9		1490
4063[b]	1	141.1	53.6		
4032[b]	2	144.6	18.3		1400
4072[b]	1	144.6	44.1		

[a,b,c,]See footnotes of Table IV.

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In some cases apparent equilibrium values reached by desorption are different from those reached by sorption. It is speculated that these observations may be a consequence of the following phenomena. The solution from which sorption takes place may contain different species of the same element (for example, ions of different oxidation states, differently complexed ions, and various degrees of hydration or polymerization). If the exchange between such species is very slow and they exhibit different sorption characteristics, only one species may sorb strongly while the other remains in solution. The nonsorbing species is then absent in the desorption experiment.

The sorption properties for some ions are more sensitive to the type of alluvium, size distribution, and temperature than those for other ions. This may be due to various types of minerals or grains playing different roles in the sorption of various ions. This effect will be studied.

Such uncertainties lead to large errors in K_d, sometimes factors of two or more. However, it is hoped that the values will still be useful since some of the other parameters in modeling the transport of nuclides are only known with much larger uncertainties. Some conclusions for the alluvium studies are outlined in the following paragraphs:

Strontium: Equilibrium appears to be attained by sorption in fewer than 3 days. However, the K_d appears to be dependent on temperature and particle size. For the sorption experiments with the 100- to 200-mesh size, values for K_d of 239 ± 40 ml/g at room temperature and 536 ± 109 ml/g at 70°C are obtained. For desorption at room temperature and the same mesh size, a somewhat smaller value, 175 ± 9 ml/g, is obtained. The change may be due in part to changes in the solid during the sorption and desorption runs (perhaps dissolution of carbonates). A value of 217 ± 45 ml/g is recommended for the equilibrium value for sorption-desorption for 100- to 200-mesh alluvium at room temperature. The behavior of Sr as a function of alluvium source and particle size should be studied further.

Barium: It appears that the time for equilibrium is 12-24 days at room temperature, and more than 3 days at 70°C. As with Sr, the K_d is dependent on the source of the alluvium. One obtains a K_d value of 4400 ± 1100 ml/g at room temperature and 8750 ± 500 ml/g at 70°C from all values for the 100- to 200-mesh material after 12 days for both sorption and desorption. For all U5e samples at room temperature after 12 days, one gets 5200 ± 2000 ml/g. For Sr and Ba, a ratio of K_d's (Sr/Ba), 0.056 ± 0.015, is obtained for all experiments in spite of the large variations in the individual K_d's.

Cesium: Equilibrium appears to be established after ≈12 days at room temperature and ≈3 days at 70°C. For the 100- to 200-mesh alluvium, the K_d is 8300 ± 1800 ml/g at room temperature and 6000 ± 400 ml/g at 70°C. The difference between these values is not as great as those for Sr and Ba. Further, the values for other particle sizes and sources are similar to these numbers. We recommend using an overall average of 7700 ± 1600 ml/g.

Cobalt: Equilibrium for sorption seems to be attained after about 2 weeks for room temperature and 70°C. For sorption, the apparent K_d's are 13 300 ± 1200 ml/g and 18 000 ± 1500 ml/g for room temperature and 70°C. For desorption, the apparent K_d is significantly higher at room temperature, 28 000 ± 9000 ml/g. The difference may be due to the preparation of the tracer from an evaporated chloride residue. There may be some competition between ion exchange and chloride complex formation in the sorption experiments.

Yttrium, Cerium, Europium: In macro quantities the trivalent ions of these elements form insoluble hydroxides at pH 8 to 8.5. The measured K_d's are quite large. Any small values are probably due to incomplete centrifugation or the presence of colloids. It is believed that all the K_d's are greater than 20 000 ml/g.

Antimony: The complex chemistry of antimony, particularly in basic solution, is reflected in the spread of distribution ratios with different experimental conditions. Sorption experiments using the commercial tracer dried to the chloride and then hydrated, gave K_d's of 6.0 ± 1.5 ml/g and 12 ± 2 ml/g at room temperature and 70°, respec-

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tively, in periods as short as 3 days. The K_d for desorption is much greater than for sorption, $\approx$65 ml/g, assuming equilibrium was attained in 140 days. This difference may be due to chloride complex formation, or to almost irreversible changes on sorption. Also noteworthy is the fact that the alluvium from U3bv has a significantly lower K_d for Sb than the material from U5e. The Sb tracer obtained from leaching of shot debris where no extra chlorides are involved gave still different K_d's: 32 ± 5 ml/g for sorption and 215 ± 30 ml/g for desorption.

Uranium (uranyl): The contact times for experiments with ^{237}U were limited by its 7-day half-life. The K_d's for sorption and desorption from ground water are $\approx$7 and $\approx$9, respectively. From deionized water, the K_d for sorption is $\approx$60. The difference may be due to higher carbonate concentration in the ground water leading to competition between carbonate complexing and sorption of the uranyl ion.

Iodine: The three iodine species in the leach water have been mentioned above. For sorption, a relatively small K_d, 24.5 ± 0.7 ml/g is obtained; however, the K_d for desorption is 400-900 ml/g. Perhaps only one species absorbs strongly.

Molybdenum: The 66-h half-life of ^{99}Mo severely limited its usefulness. The K_d appears to be $\approx$23 ml/g.

Niobium: Equilibrium seems to be established in 3 weeks for sorption and somewhat longer periods for desorption. The K_d is 2700 ± 100.

Ruthenium: For sorption the K_d appears to be 1040 ± 80 ml/g and is attained in 2 weeks. As with I the K_d is larger for desorption, 2500 ± 1200 ml/g.

The results of the experiments with bentonite will not be interpreted extensively. The starting material had not been in equilibrium with alluvial ground water, and the pre-equilibration treatment may not have been sufficient. Consequently, macro chemical changes still may have been occurring during the sorption experiments. Further, the mixing for the 2 samples may have been poor, contributing to a worse overall scatter of values than for alluvium. There is not as large a difference in the K_d's for Sr and Ba for bentonite as there is for alluvium.

Table VIII lists recommended K_d values in order of increasing value for room temperature for alluvium and bentonite. K_d values for U(VI) and Mo are not listed because desorption experiments were not done. We do not see any large advantage in using bentonite as a barrier for migration.

IV. LEACHING EXPERIMENTS

Leaching experiments were performed in a manner somewhat similar to the sorption-desorption experiments. Weighed 1-g quantities of radioactive test debris were shaken for a specified length of time with 20-ml aliquots of ground water. After three centrifugings, an aliquot of the solution containing the leached radionuclides was acidified and counted on a Ge(Li) detector. A portion of the debris was counted in similar geometry. To determine ^{239}Pu and ^{89}Sr, the solutions and debris were analyzed by conventional radiochemical methods.[11] In several experiments, the debris was leached a second time with fresh ground water. The fraction of a nuclide leached for a given time was obtained from the measurements.

Tables IX through XII give the location from which the test debris was obtained (see IIB above), the shaking time, and the fraction of each nuclide leached.

Tables IX and X list the results for leaching of the refractory and volatile samples from hole U7ajs in tuff. These samples were not high-graded, and the refractory sample contained tuff that was not fused and that may have contained volatile nuclides. Except for ^{131}I, nuclides dissolved to a greater extent from the volatile sample than from the refractory sample.

Data for similar types of samples from both refractory and volatile debris from hole U20n in rhyolite are given in Table XI. The generally low leachability, the age of the material, and the quantity of material used allowed for the determination of only limits of leachability for most nuclides observed in the debris. Only ^{137}Cs and ^{181}W were seen in the leachate, and these, at levels of a few tenths of one per cent.

TABLE VIII

RECOMMENDED K_d VALUES

	Alluvium	Bentonite
Sb	215 ± 30[a]	35 ± 20[a,b]
Sr	217 ± 45	2000 ± 700
I	640 ± 300[a]	
Ru	2500 ± 1200[a]	
Nb	≈ 2600[a]	1800 ± 700[a]
Ba	5200 ± 2000	5400 ± 600[a]
Cs	7000 ± 1600	2100 ± 200[a]
Co	20000 ± 9000[a]	5000 ± 3000[a]
Y	>20000	>2000
Ce	>20000	>2000
Eu	>20000	>10000

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[a]Value taken from desorption experiments.
[b]Value from commercial tracer experiments.

TABLE IX

LEACHING DATA FOR U7ajs REFRACTORY DEBRIS

Experiment Shaking Time (days)	Fraction Leached								
	4370 3.9	4380 13.0	4390 19.9	4400 61.9	4371[a] 24.8	4401[b] 165.9	4410[c] 19.0	4420[c] 61.9	4411[c,d] 209.0
Nuclide									
^{95}Zr	<8E-6	<2E-5	<2E-6	<4E-6	<2E-5	4E-5	<9E-6	<2E-6	
^{99}Mo	1.2E-2	1.4E-2	1.6E-2			1.8E-2			
^{103}Ru	<6E-5	6.7E-5	9.8E-5	1.1E-4	1.1E-4	1.6E-3	3.2E-4	1.5E-4	<5E-5
^{106}Ru						<2E-3	2.5E-4		<2E-3
^{124}Sb			9.7E-3	6.5E-3	<5E-3	<1E-1	1.1E-2	1.3E-2	<1E-1
^{125}Sb						9.1E-3			<2E-3
^{126}Sb			1.1E-2						
^{127}Sb	7.6E-3								
^{131}I	2.4E-1	2.3E-1	3.2E-1	2.8E-1	4.4E-2				
^{132}Te	7.3E-4	4.4E-3					<3E-4		
^{137}Cs			<7E-3	<2E-2		<2E-2	<8E-3	<2E-2	<1E-2
^{140}Ba	<2E-5		<2E-5	<9E-5	<2E-5		<5E-6	<2E-4	
^{141}Ce	<3E-5	<9E-5	<9E-6	<5E-6	<2E-5	<9E-5	<1E-5	<3E-5	<2E-5
^{144}Ce						<2E-5			
$^{239,240}Pu$				<4E-5		<6E-5	<5E-6		<4E-5

[a] Second leach of material from 4370.

[b] Second leach of material from 4400.

[c] Deionized water was used.

[d] Second leach of material from 4410.

TABLE X

LEACHING DATA FOR U7aja VOLATILE DEBRIS

Experiment Shaking Time (days)	Fraction Leached									
	4310 3.9	4320 13.0	4330 19.9	4340 61.9	4311[a] 24.8	4321[b] 48.8	4341[c] 165.9	4350[d] 19.0	4360[d] 61.9	4351[d,e] 209.0
Nuclide										
^{54}Mn							<7E-3			<9E-3
^{88}Y							<8E-4			<8E-4
^{89}Sr			1.8E-4	1.8E-4						
^{95}Zr			<3E-4	<1E-4	<3E-4	<2E-4	<4E-4	<1E-4	<4E-4	<8E-4
^{99}Mo	3.6E-1	3.6E-1	3.8E-1					3.3E-1		
^{103}Ru	8.0E-4	9.1E-4	9.7E-4	1.1E-3	1.8E-3	2.1E-3	1.0E-2		5.6E-3	6.0E-3
^{106}Ru				1.0E-3	1.7E-3	2.0E-3	1.0E-2		5.6E-3	6.0E-3
^{124}Sb		1.3E-2	1.5E-2	1.5E-2	5.0E-3	5.0E-3	4.1E-2	1.7E-2	2.0E-2	1.1E-2
^{125}Sb							4.0E-2			1.0E-2
^{126}Sb	7.6E-3	1.3E-2	1.4E-2	1.5E-2	8.1E-3	4.9E-3			2.0E-2	
^{127}Sb	8.3E-3	1.1E-2						1.5E-2		
^{129}Te^{m}	8.0E-3	3.3E-3	3.9E-3	3.7E-3	6.5E-3	5.9E-3	3.0E-2	5.9E-3	7.8E-3	1.3E-2
^{132}Te	1.0E-2									
^{131}I	2.1E-1	2.1E-1	2.1E-1	2.1E-1	4.4E-2	5.1E-2		1.1E-2	2.3E-1	
^{137}Cs				<4E-4	<1E-3	<8E-4	<4E-4	<1E-4	<5E-4	<7E-4
^{140}Ba			<8E-5	<8E-4	<2E-3	<6E-4		<1E-4	<1E-3	
^{141}Ce			<3E-3	<6E-4	<8E-4	<6E-3		<4E-3	<9E-3	<7E-3

[a] Second leach of material from 4310.

[b] Second leach of material from 4320.

[c] Second leach of material from 4340.

[d] Deionized water was used.

[e] Second leach of material from 4350.

TABLE XI

LEACHING DATA FOR U20N DEBRIS

	Fraction Leached					
	Refractory Debris			Volatile Debris		
Experiment	4270	4280	4271[a]	4290	4300	4291[b]
Shaking Time (days)	102	277	175	102	277	175
Nuclide						
^{54}Mn	<8E-4	<7E-4	<6E-4	<5E-4	<5E-4	<5E-4
$^{58,60}Co$	<6E-3	<7E-3	<7E-3	<1E-2	<1E-2	<1E-2
^{88}Y	<3E-3	<4E-3	<4E-3	<1E-3	<1E-3	<1E-3
^{95}Zr	<1E-4	<3E-4	<3E-4	<2E-4	<1E-4	<1E-4
^{106}Ru	<2E-3	<5E-3	<5E-3	<2E-3	<4E-4	<4E-4
^{125}Sb		<7E-3	<8E-3	<2E-3	<1E-3	<4E-4
^{137}Cs	<5E-2	<1E-2	<1E-2	4.4E-3	1.4E-3	2.3E-3
^{144}Ce	<1E-4	<2E-4	<2E-4	<2E-4	<6E-5	
^{155}Eu		<8E-3	<8E-3		<2E-3	
^{181}W				1.5E-3		
$^{239,240}Pu$	<2E-4	<1E-4	<3E-4	<3E-4	<1E-4	<9E-5

[a]Second leach of material from 4270.
[b]Second leach of material from 4290.

TABLE XII

LEACHING DATA FOR HIGH-GRADED REFRACTORY DEBRIS

Fraction Leached

	Ground Material		Chunks	
Experiment	4530	4510	4750	4760[a]
Test Location	U3km	U3ki	U7ap	U7ap
Shaking Time (days)	23.6	23.6	35.3	35.3
Nuclide				
^{54}Mn			2.9E-4	9.0E-5
^{58}Co			4.4E-4	9.6E-5
^{88}Y	<8E-5	<6E-5	<8E-4	<2E-4
^{95}Zr	<8E-6	<2E-6	<4E-5	<5E-5
^{106}Ru	9.6E-5	3.1E-5	5.1E-5	1.3E-5
^{124}Sb	5.0E-3	2.6E-3	2.5E-4	4.4E-4
^{126}Sb	5.6E-3	2.9E-3		
^{129}Te^{m}	2.7E-3	1.8E-3	9.6E-3	1.5E-3
^{131}I	2.0E-2	1.4E-2	8.8E-3	7.8E-3
^{137}Cs	<2E-3	<2E-3		
^{140}Ba	6.5E-5	5.1E-5	2.2E-4	9.6E-5
^{141}Ce	<2E-6	<1E-6	<4E-6	<3E-5
^{181}W	1.1E-2	3.8E-3	1.2E-3	3.5E-4
^{188}W	7.7E-3	4.5E-3		
^{182}Ta	<3E-4	<2E-4		
^{237}U	2.5E-3	1.2E-3	6.1E-4	2.6E-4
^{239,240}Pu	<6E-6	<5E-6		

[a]Solution and container were purged with argon before leaching.

The leaching data for the high-graded fused material are given in Table XII. Although ^{131}I appears to be leached to the greatest extent, its leachability from these samples is much less than from the refractory material from hole U7ajs, which was not high-graded. Nuclides of Ru, Sb, Te, I, W, and U are leached somewhat better (1.5 to 3 times) from the ground debris from hole U3km than from that from hole U3ki. It is shown in Table II that the latter debris is somewhat more deleted in the volatiles than the former. In one of the experiments with chunks from hole U7ap, argon was bubbled through the water before sealing and shaking to purge air from the system. As with the ground debris, there were small differences in leachability, but these might also be attributed to differences in the samples indicated in Table II.

The difference in leachability between the ground material (diameters 0.05 to 0.09 mm) and the chunks (2 to 5 mm) is not as great as would be suggested by the difference in size if only surface area is important. From areas of spheres, one might expect that nuclides in the ground material would be of the order of 100 times more leachable; however, U, W, and Sb were only of the order of 10 times more leachable. The difference in leachability of I was only a factor of 2 to 4. The leachabilities of Ru were comparable.

Tables IX and X indicate that some nuclides have at least two leach rates. Nuclides of Mo, Ru, Sb, Te, and I were leached faster in the first three days than they were at later times. It cannot be shown from the data when or how fast they were leached during this initial period; the fraction that was leached so rapidly is called "prompt" in this context. The prompt leachability for the refractory and volatile samples from hole U7ajs and limits deduced for the high-graded fused material are summarized in Table XIII. It appears from this summary that prompt leaching occurs from nonfused material.

The data presented and the times spanned (the experiments are continuing to longer leach times) are limited, and conclusions about leach rates must be deemed as tentative. The observed leachabilities are too small for these times to come to conclusions about what sort of rate laws govern the leachings. In Table XIV we give the leach rates in fraction leached per day, after prompt leaching, calculated

TABLE XIII

PROMPT LEACHABILITY OF DEBRIS

| | Fraction Leached | | |
Nuclide	U7ajs[a] (Volatile)	U7ajs[b] (Refractory)	High-Graded Fused Refractories[c]
^{99}Mo	(3.7 ± 0.1)E-1	(1.4 ± 0.2)E-2	
^{103,106}Ru	(9.0 ± 0.9)E-4	(9 ± 2)E-5	<5E-5
^{124,127}Sb	(1.2 ± 0.3)E-2	(6.7 ± 2.0)E-3	<3E-3
^{129,132}Te	(5 ± 2)E-3	<4E-3	<2E-3
^{131}I	2.1E-1	2.7E-1	<2E-2

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[a]From data in Table X.
[b]From data in Table IX. Not high-graded.
[c]From data in Table XII.

TABLE XIV
SUMMARY OF LEACHING DATA

Location of Debris[b]	U7ajs[c] (volatile)	U7ajs[c]	U3km[d]	U3ki[d]	U7ap[e]	U7ap[e,f]	U20n[c]	U20n[c] (volatile)
Nuclide								
^{54}Mn					8.2E-6	2.6E-6	<3E-6	<2E-6
^{58,60}Co					1.2E-5	2.7E-6	<3E-5	<4E-5
^{88}Y	(6±3)E-6		<3E-6	<1E-6	<2E-5	<6E-6	<2E-5	<4E-6
^{95}Zr	<4E-6	<1E-7	<3E-7	<8E-8	<1E-6	<2E-6	<6E-7	<4E-7
^{103,106}Ru	(6±1)E-5	(7±4)E-6	4.1E-6	1.3E-6	1.4E-6	3.7E-7	<2E-5	<1E-6
$^{124-127}$Sb	(2.0±0.9)E-4	<6E-5	2.2E-4	1.1E-4	7.1E-6	1.2E-5	<3E-5	<2E-6
^{129,132}Te	(1.8±0.7)E-4		1.1E-4	7.6E-5	2.7E-4	4.3E-5		
^{131}I	(1.5±0.5)E-3	≈2E-3	8.5E-4	5.9E-4	2.5E-4	2.2E-4		
^{137}Cs	<3E-6	<1E-4	<8E-5	<8E-5			<4E-5	(2±2)E-5
^{140}Ba	<1E-5	<2E-6	2.7E-6	2.1E-6	6.2E-6	2.7E-6		
^{141,144}Ce	<1E-5	<1E-7	<8E-8	<4E-8	<1E-7	<9E-7	<7E-7	<2E-7
^{181,188}W			4.0E-4	1.8E-4	3.4E-5	1.0E-6		1.5E-5
^{182}Ta			<1E-5	<8E-6				
^{237}U			1.0E-4	5.1E-5	1.7E-5	7.4E-6		
^{239,240}Pu		<4E-7	<3E-7	<2E-7			<4E-7	<4E-7

Column header note: Fraction Leached per Day[a]

[a] For times of less than ≈1 yr. Does not include prompt leaching in Table XIII.

[b] Refractory unless otherwise stated. See Tables IX-XII and text.

[c] Not high-graded

[d] High-graded and ground.

[e] Refractory chunks.

[f] Purged with argon.

from our data. The data should not be extrapolated to long times. Nuclides of I, Sb, Te, U, W, and Ru have the highest leach rates. Table II indicates that these are volatile to some extent.

What occurs over geologic times, because of possible alteration of the fused materials or of some of the minerals of the media to zeolites, clays, or feldspars, is not considered in this report. Most likely solution processes are involved in such alterations, and radionuclides may migrate, be incorporated in, or sorb on the alteration products.

COMPARISON WITH FIELD RESULTS

Some comparison between data generated in this study and that obtained in the Cambric study[1] can be made. Ref. 1 defines and lists depletion factors for several nuclides relative to tritium in water samples from the Cambric cavity and chimney. Although unitless, the factors would be proportional to values of K_d if only sorption-desorption processes were involved for nuclides in the water. The depletion factors vary by as much as factors of 50 with sample location; however, there is some agreement within this variation between the values of K_d and E_d except for Ru.

A ratio of depletion factors, E_d, for a water sample is the inverse ratio of fractions in solution for two nuclides. Relative fractions in solution for leaching alone or for sorption alone can be calculated from K_d values or from leaching data, respectively. Table XV gives the ratios based on Ru for the laboratory and field studies for Sr, Sb, Cs, and Ru. The agreement of field data with the laboratory leaching data is fairly good, but that with sorption data is not. The laboratory ratios for sorption alone are too high. This probably indicates that leaching is the more important mechanism responsible for the presence of the nuclides in and near the test debris. Although there are no laboratory data, it would be expected that sorption on fused material is poorer than on alluvium. Once water transports nuclides away from a test locality, the nuclide concentrations should be governed by sorptive properties of the medium. Appropriate comparisons should be made if activities are ever detected at RNM-2 or other sources of water at some distance from test locations.

TABLE XV

COMPARISON OF LABORATORY AND FIELD DATA

	Ratio of Fractions in Solution			
	Sr/Ru	Sb/Ru	Cs/Ru	Pu/Ru
From K_d[a]	11.5	11.6	0.32	
From Leaching Data[b]	0.05	4.5	<0.05	<0.005
Water from U5e[c]				
B13	0.09	1.23	0.007	2E-5
C33	0.05	0.3	0.004	3E-6
D13	0.12	0.5	0.01	1E-5

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[a]Recommended K_d values or those for desorption were used.

[b]Calculated for a period of 1 yr from prompt leaching plus slower leach rate.

[c]Calculated from Table XIII of Ref. 1. The B-13 and C-33 samples were pumped from the lower cavity region; sample D-13 came from the upper cavity region.

The laboratory experiments reported here were done in air and, therefore, under slightly oxidizing conditions. Shot debris in the vicinity of the cavity is formed in a reducing atmosphere. The electrode measurements made on the water from RNM-2 (see section III.A) indicated that the ground water may be mildly oxidizing. Therefore, it is not certain whether it is proper to extrapolate the uranium leach rates observed in the laboratory to an underground environment. Likewise, the nature of the species of other products (e.g., antimony and ruthenium) depends on the E_h of the solution in contact with the debris.

ACKNOWLEDGMENTS

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