

STRONTIUM-85 AND PLUTONIUM-239 SORPTION BY ROCKS FROM THE SEMIPALATINSK TEST SITE, KAZAKHSTAN

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ABSTRACT

The adsorption and desorption of strontium (Sr) and plutonium (Pu) were studied as a function both of rock samples and of simulated groundwaters from the Semipalatinsk Test Site (STS) where the Soviet Union tested nuclear weapons. Seven different rock types were obtained from the Balapan Region of the STS and were subjected to X-ray diffraction analyses. Two different groundwaters were simulated using data supplied by the National Nuclear Center. The results indicate the sorption and desorption of Sr is strongly dependent on the mineralogical characteristics of the rocks and on the total ionic strength of the groundwater whereas, in all cases, sorption of Pu was irreversible.

INTRODUCTION

The STS was used by the Soviet Union for testing nuclear devices for forty years. It includes about 20,000 km² of the vast steppe in eastern Kazakhstan with semiarid plains, scattered ridges and rounded hills. In the northeast the elevations are about 300 m above sea level with intermittent drainage to the Irtysh River via the Shagan River. The annual rainfall of 22 cm arrives as summer thunderstorms. The ground is frozen and covered with snow for 6 months of the year. Groundwater occurs as interstitial water in the permeable zones of unconsolidated deposits but is restricted to the weathered zones and fractures in the impermeable bedrock. Pore space in the hard dense rocks is low and not significant for groundwater movement. The water table in the unconsolidated materials occurs at depths of less than a meter to tens of meters and is hydraulically continuous with fracture water in the underlying rocks. The area around Shagan River is covered by alluvium and other unconsolidated sediments. No outcropping bedrock is visible (1,2).

Three areas which were used for nuclear tests are shown in Figure 1 as: the Experimental Area (for atmospheric tests, Degelen Mountain (for tunnel horizontal shots) and the Balapan Lack (for vertical shaft shots). No work has been reported on the subsurface movement of radionuclides although there are several surface studies (3,4). These surface studies show elevated surface radionuclide (Pu, Sr and Cs) contamination around all three testing areas. Subsurface movement of radionuclides is governed by many factors including rock structure, porosity, and ground water composition. The main rock types in the Shagan river area are sedimentary and volcanic including sandstone, shale, limestone, marl, welded tuff and lava flows.

The retardation of radionuclide migration is an important feature in assessing the transport from the STS into the Shagan River and nearby Irtysh River and from there, eventually, through sediment transport, contributing to contamination in the Arctic. We report here the sorption characteristics of Sr and Pu by seven core samples of the main rock types found in the Balapan Lake area of the test site where vertical underground shots were performed. Two waters which simulate the composition of two local groundwaters were used.

Studies of sorption of metals onto geologic materials has been conducted at other repository sites, in particular the Waste Isolation Pilot Plant (5) and the Yucca Mountain Project (6,7). Other studies have looked at potential migration and retardation of metals at contaminated sites as part of environmental restoration programs (8).

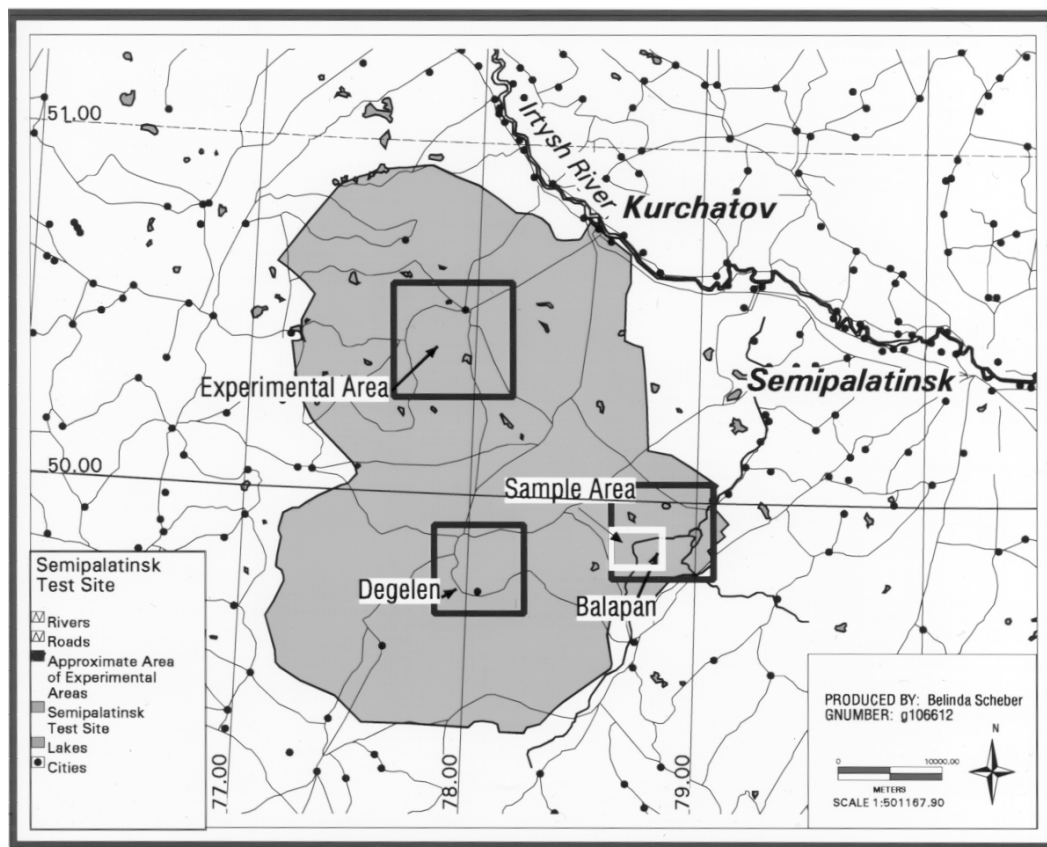


Figure 1.
Semipalatinsk Test Site, Kazakhstan showing the three test areas.

MATERIALS AND METHODS

Materials

Seven rock samples (Table I) were obtained from bore holes from the Balapan Lake area (Figure 1). The bore holes were drilled for the area geologic characterization analysis for nuclear weapons tests.

Table I.
Rock Samples supplied by National Nuclear Center, Republic of Kazakhstan

Sample Number	Bore Hole Number	Depth m	Description	Water Table Depth m
1	4076	91	tuff	37
2	4076	94	silt	37
3	4100	103.4	sand	13.6
4	4027	64.5	sandstone	?
5	4048	88.5	porphyrite	11.3
6	4041	76	sandstone	2
7	4035	92	tuff/silt	14.4

The bore hole locations are given in Figure 2. The samples obtained were all south of the Shagan river except for 4076 that is in a region adjacent to the river that seasonally becomes marsh. Geologic profiles of the six core samples are listed in Table II.

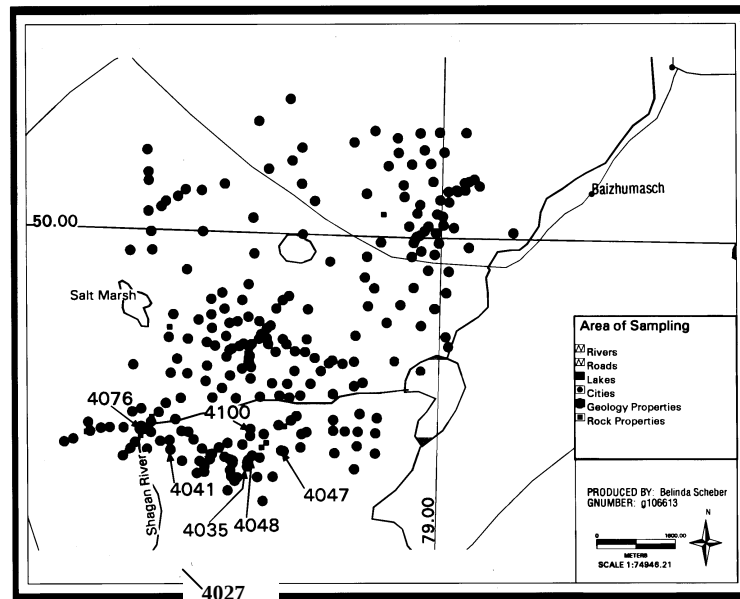


Figure 2.
Locations of Bore holes in the Balapan area.

Table II
Geologic profiles of the six cores used. The samples analyzed in this study are shown in bold.
(Data supplied by the National Nuclear Center, Kazakhstan)

4027		4035		4041		4048		4076		4100	
Start strata m	Geology	Start strata m	Geology	Start strata m	Geology	Start strata m	Geology	Start strata m	Geology	Start strata m	Geology
0	Sand	0	Sand	0	Sand Gravel	0	Sand Loam	0	Sand	0	Sand
6	Clay	2.3	Clay Sand	6	Tuff	5	Clay	3	Gravel	7.5	clay
17.5	Slate	3.5	Clay Sand	32	Silt	15.5	Sand-stone	7	Sand	56.4	granite
25	Slate	25	Clay Sand	36	Tuff	30.2	Basalt porphyrites	9.5	Clay	61.2	
32	Slate	36	Tuff/Sand-stone	47	Tuff	75	Basalt porphyrites	74.9	Sand-stone Silt	85.5	granite
47	Slate	60.7	Argillite	53	Tuff, silt Sand-stone	88	Basalt Porphyrites	80	Tuff	88	sand
68	Tuff, sand-stone	66	Tuff, Silt Sand-stone	63.5	Tuff, silt Sand-stone	100	Basalt porphyrites	81	Sand-stone/Silt	105.2	
		95	Tuff-breccia	80.6	Tuff,Silt Sand-stone	106.5	Basalt porphyrites	95	Tuff		

The chemical composition of the water from bore holes has been extensively analyzed. Table III shows the range of the major cations and anions found from many analyses varying from one (4076) to 56 (4035). The 4035 analyses were taken continuously over a period of five years.

Table III
Range of the major cations and anions found in round water
from the six bore holes.
(Data supplied by the National Nuclear Center, Kazakhstan)

Well Number	Depth m	pH	Na + K mg/l	Ca mg/l	Mg mg/l	NH ₄ mg/l	Cl mg/l	SO ₄ mg/l	HCO ₃ mg/l
4027	25-20	7.9-7.6	2135-872	410-267	279 - 167	6-0	3230 -740	2184-2040	159 -104
4035	40	8.3-6.6	1339-767	183-111	199 - 76	20-0	1749-1101	1225 - 622	221 -28
4041	20	8.4-6.6	1312-809	204-58	194 - 128	10-0	750-573	2256 - 1961	322 -43
4048	40-20	8.4-6.4	119-815	262-141	159 - 76	20-0	1446 - 817	1552 - 1187	197 - 101
4076	91-94	7.5	2524	494	532	5	4086	2691	439
4100	60	7.3-6.4	1470-58	277-29	221 - 6	15-0	1558 - 36	2996 - 107	651 - 92

Methods

The methods used for X-Ray Diffraction analyses are summarized in references 9,10, and 11.

All X-ray diffraction patterns (XRD) were obtained on a Siemens D500 X-ray powder diffractometer using CuK α radiation, incident- and diffracted-beam Soller slits, and a Kevex Si(Li) solid-state detector. X-ray diffraction runs were conducted from 2 to 50° 2 θ , with 0.02° steps and counting times of at least 4 seconds per step.

To prepare the samples, a small portion of each sample (~0.8 g) was mixed with 1.0- μ m corundum (Al₂O₃) internal standard in the ratio 80% sample to 20% corundum by weight. Each sample was then ground under acetone in an automatic Brinkmann Micro-Rapid mill (fitted with an agate mortar and pestle) for a time greater than 10 minutes. This produced a sample with an average particle size of less than 5 μ m and ensured thorough mixing of sample and internal standard. The fine particle size is necessary to ensure adequate particle statistics and to reduce primary extinction and other sample-related effects.

Quantitative XRD analyses for this report employed the internal standard or "matrix-flushing" method of Chung (12) using synthetic 1.0- μ m corundum as the internal standard. The method has been extensively modified, however, to utilize multiple reflections and intensity ratios (11).

Rocks

Each rock sample was ground to a powder. The powder was successively sieved through 500- μ m and 75- μ m US standard sieves. The 75 μ m to 500 μ m particle size range was

used for sorption experiments. The methods used to collect the core samples, unfortunately, are unavailable.

Solutions

Two synthetic groundwaters, referred to as 4076 (pH 7.5) and 4100 (pH 6.8), and two deionized waters, referred to as DI-4076 (pH 7.5) and DI-4100 (pH 6.7) were used. The 4076 synthetic groundwater consists of 7.2×10^{-3} M sodium bicarbonate (NaHCO_3), 2.19×10^{-2} M magnesium sulfate (MgSO_4), 6.33×10^{-3} M calcium chloride (CaCl_2), 0.103 M sodium chloride (NaCl), and 5.96×10^{-3} M calcium sulfate (CaSO_4). The ionic strength of the solution was 0.2405 M. The 4100 synthetic groundwater consists of 1.606×10^{-3} M NaHCO_3 , 2.467×10^{-4} M MgSO_4 , 1.015×10^{-3} M NaCl , 9.36×10^{-5} M sodium sulfate (Na_2SO_4) and 7.735×10^{-4} M (CaSO_4). The ionic strength of the solution was 6.861×10^{-3} M.

Tracer solutions

Plutonium-239 tracer solution. The $^{239}\text{Pu(V)}$ stock solution contained 7.65×10^{-4} M of Pu with 99.7% of Pu(V), and consists of 93.9% of ^{239}Pu . A $^{239}\text{Pu(V)}$ work solution was prepared by diluted 1.6 g of $^{239}\text{Pu(V)}$ stock solution with 998.4 g of nanopure deionized water resulting in a solution containing 1.143×10^{-7} M of Pu. The activity of the work solution was determined using a liquid scintillation analyzer. The α activity of $^{239}\text{Pu(V)}$ was 277,870 Bq kg^{-1} . The pH value of the work solution was 6.03.

Strontium-85 tracer solution. The ^{85}Sr stock solution contained 1.0 mCi of Sr in 0.1 M HCl, and consists of 99.98% of ^{85}Sr (half life $t_{1/2} = 64.8$ d). The ^{85}Sr activity of 1.0 mCi was measured on January 14, 1998 when ^{85}Sr stock solution was obtained. A ^{85}Sr work solution was prepared by diluted 0.15 g of ^{85}Sr stock solution with 999.85 g of nanopure deionized water resulting in a solution containing 0.165 mCi L^{-1} of ^{85}Sr . The activity of the work solution was determined using a gamma spectroscopy. The result was corrected by a fact of 0.28 (the efficiency of the gamma spectroscopy) and also by decay factor related to January 14, 1998, referred to as $t = 0$. The γ activity of the ^{85}Sr work solution was 632,555 counts per minute per milliliter (CPM ml^{-1}). The pH value of the work solution was 5.91.

Strontium-85 was used as a surrogate for ^{90}Sr (found at the STS). Strontium-85 is a gamma emitter and has the same chemical characteristics as ^{90}Sr but its measurement is faster and cheaper.

Sorption procedure

Sorption experiments were performed in duplicate at room temperature. One gram of rock powder sample, 20 ml of the desired water (e.g., 4076, 4100, DI-4076, or DI-4100) and 3 ml of either $^{239}\text{Pu(V)}$ or ^{85}Sr work solution were placed in a pre-weighed Oak Ridge centrifuge tube (50-ml), and were agitated on an orbital shaker at 150 rpm for 5 days. At the same time, blank samples were also set up using 20 ml of each water without any rock samples and 3 ml of $^{239}\text{Pu(V)}$ or ^{85}Sr work solution. At the end of sorption period, the

samples and blanks were centrifuged at 38,300 g for 30 minutes. The supernatant was filtered through a 0.2- μ m nylon filter membrane using a syringe. The adsorbed values of $^{239}\text{Pu(V)}$ or ^{85}Sr onto rocks were corrected with results from blank samples.

The α activities of the supernates collected from Pu sorption experiments were determined using the liquid scintillation analyzer. The γ activities of the supernates collected from Sr sorption experiments were measured as follows: one milliliter of the supernate collected from each rock sample was placed in a counting tube and counted for 20 minutes by gamma spectroscopy. The results were then converted to counts per minute per milliliter (CPM ml^{-1}) and corrected by a factor of 0.28 (the efficiency of the gamma spectrometer), followed by decay correction related to the start time, Jan.14, 1998, referred to as $t = 0$.

The sorption distribution coefficient (K_d , ml g^{-1}) of Pu or Sr on each rock sample is calculated using the following equation:

$$K_d = [(A_0 V_0 - A_i)/M]/A_j \quad [1]$$

where A_0 is the initial radioactivity of $^{239}\text{Pu(V)}$ or ^{85}Sr per milliliter solution, V_0 is the solution volume (ml) in contact with rock samples, A_i is the total radioactivity of $^{239}\text{Pu(V)}$ or ^{85}Sr remaining in the solution after sorption (CPM), A_j is the radioactivity remained in unit solution (CPM ml^{-1}), and M (g) is the mass of rock sample used in the sorption process.

Desorption procedure

Plutonium-239-loaded rock samples and ^{85}Sr -loaded rock samples were obtained from sorption process. After the wet weights of the rock samples were obtained, the samples were extracted successively with ^{239}Pu -free, or ^{85}Sr - free water of 4076, 4100, DI-4076, or DI-4100 and were agitated on an orbital shaker at 150 rpm. Desorption of ^{239}Pu from rock samples was allowed to proceed from 10 days to 70 days. Desorption of ^{85}Sr from rock samples proceeded for 15 days. For each sampling interval, the extracts were separated applying the same centrifuge and filter procedures used in the sorption experiments. The activity of ^{239}Pu in the extracts was measured using the liquid scintillation analyzer. The activity of ^{85}Sr in the extracts was measured using the same method applied in the sorption experiments. The results from ^{85}Sr desorption process were then converted to counts per minute per milliliter (CPM ml^{-1}) and corrected by a factor of 0.28 and by decay factor related to the initial time, $t = 0$.

RESULTS.

The X-ray diffraction analysis results of the rock samples are given in Table IV. The distribution coefficients (K_d) for sorption and desorption for Sr and Pu are given in Tables V- VIII. Figures 3 and 4 show the sorption and desorption data for Pu and Sr.

Table IV

Results of X-ray diffraction analyses.

Results are in weight percent. Errors are conservative 2-sigma values.

Trc. = Trace amount less than 0.5 weight percent. — = not detected.

Rock ID	Chlor-ite	Mica/Illite	Quartz	Feld-spar	Calcite	Dolomite/Ankerite	Hem-atite	Mag-netite	Total
1	13 ± 1	1 ± 1	13 ± 1	63 ± 9	6 ± 1	3 ± 1	1 ± 1	1 ± 1	101 ± 9
2	7 ± 1	3 ± 1	25 ± 2	38 ± 5	—	2 ± 1	1 ± 1	—	76 ± 6
3	2 ± 1	3 ± 1	30 ± 2	58 ± 8	—	1 ± 1	Trc.	1 ± 1	95 ± 8
4	7 ± 1	6 ± 2	16 ± 1	51 ± 7	—	Trc.	Trc.	—	80 ± 7
5	Trc.	11 ± 3	24 ± 2	32 ± 4	18 ± 1	1 ± 1	11 ± 1	1 ± 1	98 ± 6
6	9 ± 1	3 ± 1	29 ± 2	51 ± 7	2 ± 1	1 ± 1	Trc.	—	95 ± 8
7	5 ± 1	3 ± 1	33 ± 2	39 ± 5	2 ± 1	—	Trc.	—	82 ± 6

Table V.

Sorption Distribution Coefficient of Pu on Rocks (K_d , ml g⁻¹)
(5 days sorption)

Rock ID	4076 (pH 7.5)	DI-4076 (pH 7.5)	4100 (pH 6.8)	DI-4100 (pH 6.7)
1	5.57E+04	3.00E+04	3.31E+04	3.88E+04
2	1.25E+04	1.13E+04	1.42E+04	1.55E+04
3	4.15E+04	2.62E+03	1.78E+04	2.26E+04
4	3.09E+04	1.59E+03	9.08E+04	1.35E+03
5	1.88E+04	4.42E+03	4.77E+04	1.11E+04
6	3.89E+04	1.31E+04	2.62E+04	1.76E+04
7	2.96E+04	4.04E+04	2.27E+04	4.08E+03

Table VI.

Desorption Distribution Coefficient of Pu on Rocks (K_d , ml g⁻¹)
(15 days desorption)

Rock ID	4076 (pH 7.5)	DI-4076 (pH 7.5)	4100 (pH 6.8)	DI-4100 (pH 6.7)
1	7.34E+04	5.15E+04	8.53E+04	
2	7.33E+04	6.46E+04	6.01E+04	7.33E+04
3	1.50E+05	7.42E+03	3.88E+04	8.18E+03
4		1.04E+04	1.11E+06	1.57E+04
5	3.83E+04	3.23E+04		1.59E+05
6	4.25E+04	4.41E+04	4.70E+04	3.94E+04
7	1.30E+05	1.59E+04	5.03E+04	1.93E+04

Table VII.
Sorption Distribution Coefficient of Sr on Rocks ($K_{d,s,ml\ g^{-1}}$)
(5 days sorption)

Rock ID	4076 (pH 7.5)	DI-4076 (pH 7.5)	4100 (pH 6.8)	DI-4100 (pH 6.7)
1	0.6	55.0	13.8	53.0
2	1.6	126.0	30.4	138.0
3	0.7	73.4	10.8	74.8
4	1.6	845.0	59.0	918.0
5	1.8	285.0	59.3	294.0
6	2.2	27.0	11.3	27.6
7	1.0	205.0	28.2	204.0

Table VIII.
Desorption Distribution Coefficient of Pu on Rocks ($K_{d,d,ml\ g^{-1}}$)
(15 days desorption)

Rock ID	4076 (pH 7.5)	DI-4076 (pH 7.5)	4100 (pH 6.8)	DI-4100 (pH 6.7)
1	5.7	67.2	18.2	66.2
2	8.2	106.0	32.9	110.0
3	6.7	84.7	12.8	81.1
4	3.0	574.0	61.1	1040.0
5	3.8	223.0	55.3	197.0
6	21.6	29.6	13.6	31.3
7	5.7	153.0	30.4	145.0

Figure 3. Sorption and Desorption Data for $^{239}\text{Pu(V)}$

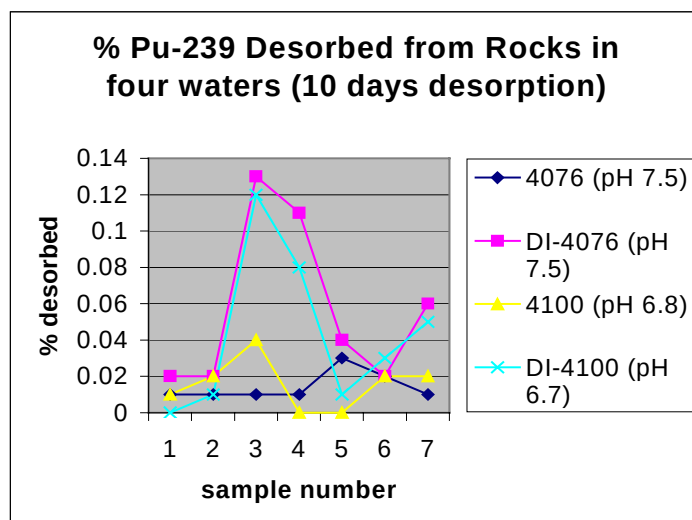
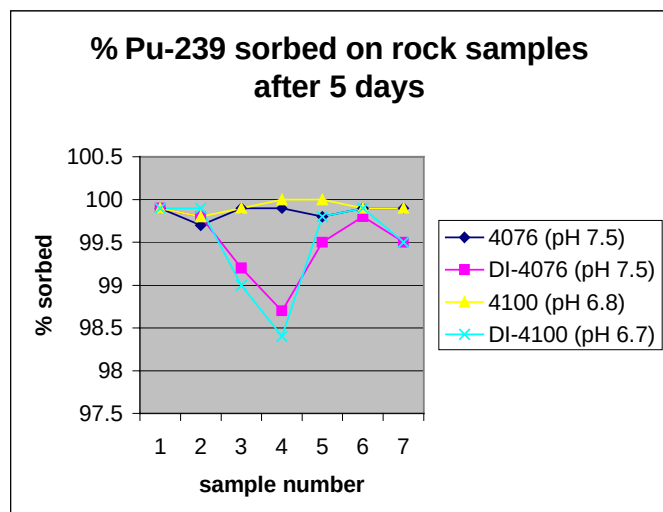
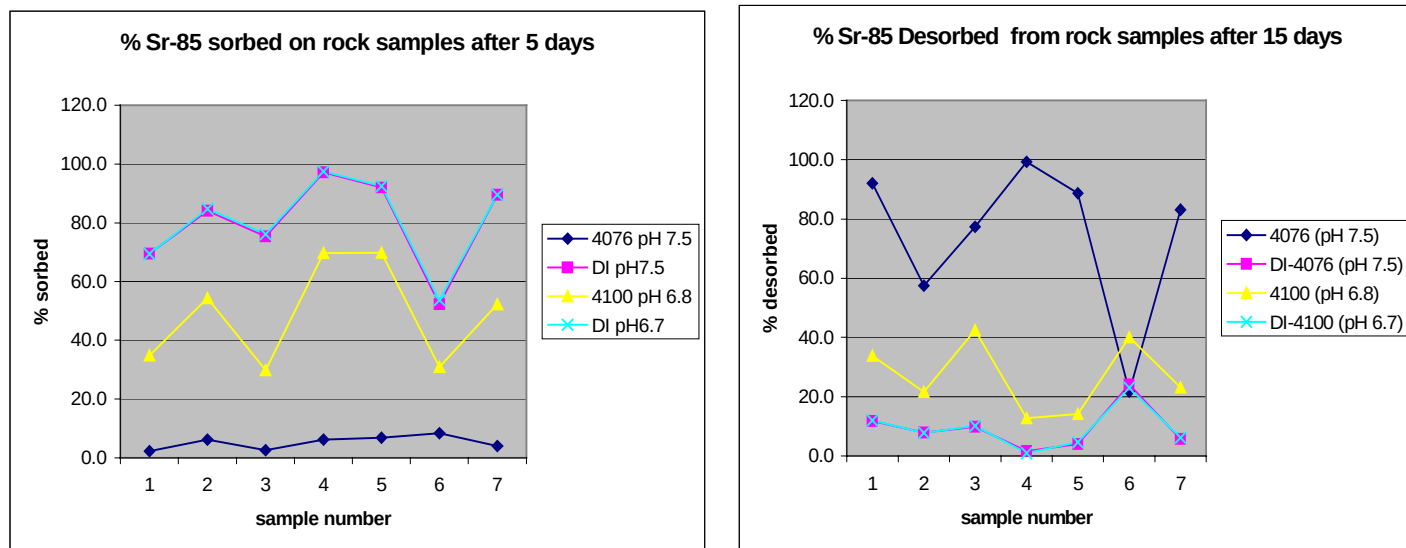


Figure 4. Sorption and Desorption Data for ^{85}Sr 

Discussion

The X-ray diffraction data shows that the major mineral components of the rocks are quartz and feldspars that are common in many rock types. Feldspar is an aluminum silicate MAlSi_3O_8 where $\text{M} = \text{Na}, \text{K}, \text{or Ca}$. Other minerals present include layer-silicates (mica, illite and chlorite), carbonates such as calcite, (calcium carbonate), dolomite (magnesium calcium carbonate), ankerite (iron calcium carbonate), and iron oxides (hematite and magnetite).

Plutonium may exist in the environment in several oxidation states, Pu(III), Pu(IV), Pu(V) and Pu(VI) (13). Under oxidizing conditions the most common state is Pu(V) as the cation PuO_2^+ , accounting for its use in these sorption studies. The lowest sorption for Pu found is 98.5% for rock sample 4. Since for sample 4, the total x-ray diffraction data only accounts for 80% of the total minerals present, it is likely that these changes in sorption are within experimental error and do not signify changes in sorption characteristics of the rocks.

Under the oxidizing experimental conditions used, Pu(V) is not reduced to Pu(IV) but there is the possibility of Pu(VI) species. The pH is too low to allow for the presence of carbonate species (< 8.3). The iron oxides present (magnetite and hematite) will strongly sorb all oxidation states of Pu, as has been found for colloidal hematite and goethite (14). Carbonates also strongly sorb Pu. The Pu is not easily desorbed. The K_d s found in these Pu sorption experiments is of the same order of magnitude as some samples obtained from Yucca Mountain (6) (zeolitic tuff $4 \times 10^{+2}$ and hematite $4 \times 10^{+3}$). The layer structure of mica allows easy sorption with a high cation exchange capacity and easily

exchangeable potassium, calcium and magnesium but is found not to be a strong sorber except for cesium and potassium. There is a negligible ionic strength effect, of the order of 0.1 % as all K_d values are high. The structure of feldspar, unlike that of mica, does not allow for easy sorption and so has a low K_d .

The samples come from between 64 -104 meters below the surface and below the water table where the environment may be partially reducing. Thus the experimental conditions may not accurately reflect the subsurface conditions.

Strontium, behaves very differently from Pu as it is an analog for calcium and is bioactive (readily absorbed by plants, soil microbes and animals). Strontium is readily exchanged for calcium because they are similar both electronically and in size (the dehydrated radius for Ca is 0.099 nm and for Sr 0.112 nm). Since Sr only exists in one valence state there are less opportunities for complex speciation and changes due to redox potential. The K_d values are significantly lower than for Pu due to the lower oxidation state. These lower values account for significant migration of Sr in solution. Values of K_d for Sr in a Canyon at Los Alamos are of the same order of magnitude ($K_d = 36 \text{ ml g}^{-1}$) (8). Sr is also readily desorbed

The ionic strength effect for Sr is much greater than for Pu due to the competition between different cations with the same charge and the lower K_d values. As with Pu, it is possible that Sr replaces calcium in the carbonates present. Independent experiments have shown that illite does not sorb Sr (15). Table III shows the range of measured major anions and cations from the wells taken over several years. The variation is large with resultant variation in total ionic strength due to extreme seasonal variations and the marshy characteristics adjacent to the Shagan River. Because of the elevation and distance from moderating oceans, the climate varies between extremes, by as much as 20° F over a 24 h time span.

In summary, it is unlikely that Pu will be transported by subsurface ground water over long distances due to the high K_d values for the local rock minerals. Sr is more likely to be transported in the subsurface. Since ^{90}Sr has a short half life compared to ^{239}Pu (30.2 compared to 24,100 years) over long time periods it is unlikely to be a major concern.

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