CONF-781121--6

LA-UR -78-2746

MASTER

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SUBMITTED TO: Symposium on Science Underlying Radioactive Waste Management, Boston, MA, Nov. 28 - Dec. 1, 1978

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Form No. 836 R2
St. No. 2629
1/78

DEPARTMENT OF ENERGY
CONTRACT W-7405-ENG. 36

SORPTION AND MIGRATION OF RADIONUCLIDES IN GEOLOGIC MEDIA*

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ABSTRACT

The interactions of a quartz monzonite, an argillite, an alluvium, and several tuffs with various radionuclides in selected phreatic waters has been studied. The sorption-desorption behavior of Sr, Tc(VII), Cs, Ba, Ce, Eu, U(VI), Pu, and Am under ambient and 70°C temperature conditions has been measured.

EXECUTIVE SUMMARY

A major requirement for the evaluation of the long term safety of nuclear waste in a deep geologic storage environment is a thorough understanding of the mechanisms and phenomenology of the sorption-desorption behavior of the various radionuclides that are biologically hazardous. This knowledge will aid in the prediction of the fate of the radionuclides during the length of time required for radioactive decay to reduce the waste to safe levels.

The Los Alamos Scientific Laboratory has begun a significant effort for the study of the partition of various radionuclides between different rock types and natural or synthetic ground waters. The geologic materials studied include a quartz monzo-

*This work was supported by the U.S. Department of Energy.

nite porphyry (Climax Stock), a kaolinite-montmorillonite argillite (Eleana), a tuffaceous alluvium (Frenchman Flats), and several lithologic varieties of tuffs (Jackass Flats; pyroclastic rocks formed from fragmental products of explosive volcanic eruptions). These materials are all from the Nevada Test Site (NTS).

Various particle sizes ranging from 106 to 850 μm, and both ambient and elevated (70°C) temperatures were used with a batch technique. Changes in the sorption-desorption behavior with time were examined for intervals of 1, 2, 4, and 8 weeks. The elements studied were Sr, Tc(VII), Cs, Ba, Ce(III), Eu(III), U(VI), Pu, and Am. The measurements were performed under atmospheric oxygen conditions and all materials were pre-equilibrated prior to use. Details of the experimental methods used and of the initial results of these studies are given in Refs. 1-4.

The table summarizes the results obtained (sorption ratio R_d = activity per g of solid/activity per ml of water) using only the sorption data. The sorption ratio is generally high for the alluvium, the argillite, and for most of the tuffs. The presence of alteration minerals such as clays and zeolites generally results in high sorption ratios. A similar relationship also exists between the presence of glass phases and high sorption ratios for Cs, Sr, and Ba. Sorption increases slowly with time. Increased sorption is observed for Sr and Ba as the temperature is increased. The reverse trend seems to be valid for Ce and Eu, while Cs is not affected by temperature. Desorption ratios are significantly greater than the sorption ratios.

A microautoradiographic technique has been developed (5) in order to identify the individual mineral components in the rock that are responsible for the sorption. This procedure has been used for a study of U(VI) and Am sorption. In the quartz monzonite, most of the sorbed U(VI) and Am was contained in secondary clay-rich alteration bands in the feldspars. The argillite samples indicated preferential sorption of U(VI) and Am on the clay matrix, with insignificant amounts sorbed onto the detrital quartz and secondary calcite. In the tuffaceous alluvium, most of the Am and U(VI) was sorbed on the glass phase and on clay minerals. In the tuff specimens, most of the radionuclides were localized on the secondary zeolite minerals.

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AVERAGE SORPTION RATIOS, R_d (ml/g)

Material	Element	22°C	70°C
Quartz Monzonite	Sr	20	40
	Tc(VII)	< 80	< 16
	Cs	440	1440
	Ba	160	730
	Ce	740	470
	Eu	960	540
	U(VI)	9	
	Pu	1300	3600
	Am	2600	6600
Argillite	Sr	130	290
	Tc(VII)	< 40	< 3
	Cs	2500	1900
	Ba	4200	18000
	Ce	> 40000	13000
	Eu	> 50000	22000
Alluvium	Sr	200	
	Cs	7000	
	Ba	5000	
	Ce	> 20000	
	Eu	> 20000	
	U(VI)	10	
	Pu	> 1000	
Vitric tuff	Sr	13000	14000
	Cs	15000	18000
	Ba	5000	50000
	Ce	40	40
	Eu	30	80
	Pu	170	
	Am	170	220
De-vitrified tuff	Sr	60	110
	Cs	120	100
	Ba	400	1000
	Ce	80	80
	Eu	90	200
	Pu	110	
	Am	110	100
Zeolitized tuff	Sr	240	1000
	Cs	600	1400
	Ba	750	3700
	Eu	6000	4000
	Pu	280	
	Am	590	700