OAK RIDGE NATIONAL LABORATORY
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October 10, 1984

Dr. J. W. Bradbury
Geotechnical Branch
Office of Nuclear Material
Safety and Safeguards
U.S. Nuclear Regulatory Commission
Room 623-SS
Washington, D.C. 20555

Dear John:

Enclosed is the progress report for the month of September 1984 for B0290, "Laboratory Evaluation of DOE Radionuclide Solubility Data and Selected Retardation Parameters, Experimental Strategies, Laboratory Techniques and Procedures."

Sincerely,

Susan

Susan K. Whatley, Manager
Engineering Analysis And Planning
Chemical Technology Division

SKW:kk
Enclosure
cc: Office of the Director, NMSS (Attn: Program Support Branch)
Division Director, NMSS Division of Waste Management (2)
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R. J. Starmer, Geotechnical Branch
D. J. Brooks, Geotechnical Branch
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4. AUTHORS *(If more than three, name first author followed by "and others")*
A. D. Kelmers and others

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SKW
10/09/84

MONTHLY PROGRESS REPORT FOR SEPTEMBER 1984

PROJECT TITLE: Laboratory Evaluation of DOE Radionuclide Solubility Data and Selected Retardation Parameters, Experimental Strategies, Laboratory Techniques, and Procedures

PROJECT MANAGER: S. K. Whatley

TASK LEADER: A. D. Kelmers

SCIENTIFIC STAFF: W. D. Arnold, G. K. Jacobs, S. Y. Lee, R. E. Meyer, F. G. Seeley, and F. J. Smith

ACTIVITY NUMBER: ORNL #41 37 54 92 6 (FIN No. B0290)
NRC #50 19 03 1

HIGHLIGHTS:

Tests with McCoy Canyon basalt gave moderately high sorption ratio values for technetium under anoxic redox conditions at initial concentrations of 10^{-12} to 10^{-6} mol/L Tc(VII) in synthetic groundwater GR-2. Sorption was not observed in tests at 10^{-3} or 10^{-4} mol/L Tc(VII). Sorption increased by a factor of 2 to 3 in 50-d tests, as compared to parallel 14-d tests. The mechanism for technetium(VII) removal from solution by basalt continues to appear complex. Sorption is usually observed when air is carefully excluded from the test environment.

Hydrazine did not reduce neptunium(V) in synthetic groundwater GR-2 to neptunium(IV) under anoxic or oxic redox conditions in control experiments without basalt. This is further evidence that the addition of hydrazine to synthetic groundwaters does not simulate the expected redox condition for the basalt far field.

Sorption of uranium(VI) by McCoy Canyon basalt under anoxic redox conditions was similar to earlier results obtained under oxic conditions. Again, evidence for a uranium-containing precipitate was observed at the higher uranium concentrations. So far, there is no evidence which suggests that basalt is capable of reducing uranium to uranium(IV) in laboratory experiments.

TECHNICAL PROGRESS:

Technetium Studies

During September, two 50-d anoxic redox condition batch contact sorption test were completed with McCoy Canyon basalt and synthetic groundwater GR-2, one test at 27°C and one at 60°C. As in the 14-d tests reported in July, moderate sorption was observed from solutions with initial technetium concentrations of 10^{-12}, 10^{-8}, and 10^{-6} mol/L, but essentially no sorption was observed from solutions with initial concentrations of 10^{-4} and 10^{-3} mol/L. Average values of the sorption ratios for the three concentrations at which sorption was observed are given in Table 1.

Table 1. Sorption of technetium onto McCoy Canyon basalt under anoxic redox conditions[a]

Initial Tc(VII) (mol/L)	Time (d): Temp. (°C):	Average Rs (L/kg) 14 60	50 27	50 60
10^{-12}		3.5 ± 0.2	8.3 ± 0.1	11.1 ± 5.3
10^{-8}		5.5 ± 0.7	12.3 ± 1.0	9.9 ± 2.3
10^{-6}		4.0 ± 0.4	8.5 ± 0.2	3.6 ± 4.3

aBatch contact tests with synthetic groundwater GR-2.

The Rs values tended to increase by a factor of about 2 to 3 as the contact period was increased from 14 to 50 d. There appeared to be little effect on sorption of increased temperature.

Uranium Studies

During September, batch contact tests were completed for sorption of uranium(VI) onto McCoy Canyon basalt under anoxic conditions at 60°C. The contact period was 14 d, and the liquid/solid separation was effected by centrifugation for 30 min at 5000 rcf. For initial uranium concentrations of 10^{-7}, 10^{-6}, 10^{-5}, and 10^{-4} mol/L, the values of Rs were 3.9, 2.5, 2.0, and 35 L/kg, respectively. The high value of Rs at 10^{-4} mol/l initial concentration suggests that precipitation may have occurred. A white precipitate was clearly observed in a parallel blank test (no basalt present) at 10^{-4} mol/L initial uranium concentration. The observed values of Rs are virtually the same as those reported previously for oxic redox conditions at 60°C.

Neptunium Studies

We have previously reported [NUREG/CR-3581, Vol. 2] that the addition of hydrazine to neptunium(V)-traced groundwaters did not reduce the neptunium to Np(IV) under oxic experimental conditions, as expected by BWIP. This month we briefly explored the ability of hydrazine to reduce Np(V) under anoxic redox conditions and also checked our earlier results for oxic conditions. In blank tests (no basalt present) with 3×10^{-7} mol/L Np(V) in synthetic groundwater GR-2 containing 0.1 M hydrazine hydrate, most of the neptunium (80%) remained as Np(V) (Table 2). Standard solvent extraction methods were used to determine the neptunium valence. Since the initial Np(V) stock solution contained about 15-18 % Np(IV), it appeared that the addition of hydrazine had no effect on the neptunium valence under either oxic or anoxic redox conditions.

Table 2. Effect of hydrazine on neptunium valence at 25°C

Np Valence	Anoxic Conditions		Oxic Conditions	
	1 Day	1 Week	1 Day	1 Week
(IV)	14.5%	18.4%	19.2%	12.9%
(V)	82.1%	78.2%	81.9%	81.2%
(VI)	2.3%	3.5%	2.5%	2.8%
Material Balance	98.8%	100.1%	103.6%	96.9%

A series of batch contact tests are in progress to determine the neptunium(V) sorption rate onto Cohassett basalt from synthetic groundwater GR-4 under anoxic redox conditions in tests lasting from 1 to 60 d. The tests are still underway at this time.

Tests to explore neptunium(V) sorption onto Cohassett basalt from synthetic groundwater GR-4 under anoxic redox conditions at 60°C as a function of the basalt particle size or experimental surface area/volume ratio have been initiated; test data are incomplete at this time.

Chromatographic Studies

No progress to report this month.

Sample Acquisition

There was no contact this month from NNWSI staff relative to the samples of Yucca Mountain materials which we requested [Letter Report L-290-6, May 22, 1984].

Sample Characterization

Samples of both Cohassett and Umtanum basalts were mineralogically characterized using an electron microprobe with an energy dispersive x-ray analyzer. Like the McCoy Canyon basalt sample (described in the January-March 1984 progress report [NUREG/CR-3851, Vol. 2]), both of these basalt samples were composed of plagioclase, mesostasis, pyroxene, magnetite, apatite, and pyrite. The amounts of each mineral phase will be estimated from the relative areas present in a selected surface area. The quantitative mineralogical information for each basalt formation will be reported in the 1984 annual progress report.

Geochemical Calculations

No progress to report this month.

General Aspects

None

MEETINGS AND TRIPS:

None

REPORTS AND PUBLICATIONS:

The progress report for the period January-March 1984 has been typed in final form and is undergoing the ORNL document release process. It will be sent to the NRC Project Manager in October for issuance as NUREG/CR-3851, Vol. 2.

The draft progress report for April-June 1984 was sent to the NRC Project Manager on September 9. After receipt of the NRC review comments, it will be revised and prepared in final form.

PROBLEM AREAS:

None

COST/BUDGET REPORT:

Expenditures were $20.7K for the month of September and $583.2K for the fiscal year 1984. A detailed cost/budget report will be sent under separate cover.