OAK RIDGE NATIONAL LABORATORY
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December 10, 1985

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:

Please find enclosed the progress report for the month of November 1985 for B0290, "Laboratory Evaluation of DOE Radionuclide Solubility Data and Selected Retardation Parameters, Experimental Strategies, Laboratory Techniques and Procedures."

Sincerely,

Gary

Gary K. Jacobs
Environmental Sciences Division

GKJ/

Enclosure:

Monthly Progress Report for November 1985

cc: Office of the Director, NMSS (Attn: Program Support Branch)
Division Director, NMSS Division of Waste Management (2)
M. R. Knapp, Chief, Geotechnical Branch
K. C. Jackson, Geotechnical Branch
D. J. Brooks, Geotechnical Branch
Branch Chief, Waste Management Branch, RES
C. Hackbarth, Waste Management Branch, RES
A. D. Kelmers
A. P. Malinauskas
R. E. Meyer
S. K. Whatley
Lab Records (2)
GKJ File

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4. AUTHORS *(If more than three, name first author followed by "and others")*

G. K. Jacobs and others

5. NAME OF CONTRACTOR	MAILING ADDRESS *(Number and street, city, state and zip code)*	TELEPHONE NO.
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6. DATE MANUSCRIPT COMPLETED	7. NRC PROGRAM SPONSOR/TECHNICAL MONITOR	TELEPHONE NO.
12-85	J. W. Bradbury	427-4055

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a. NAME OF AUTHORIZED CONTRACTOR OFFICIAL OR NRC MONITOR *(Type or print)*: W. N. Drewery, Laboratory Records Supervisor

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ORNL/NRC/LTR-85/34

Contract Program Title:
Laboratory Evaluation of DOE Radionuclide Solubility Data and Selected Retardation Parameters, Experimental Strategies, Laboratory Techniques and Procedures

Subject of this Document:
Monthly Progress Report

Type of Document:
Technical Letter Report

Authors:
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Project Manager:
G. K. Jacobs

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Office of Nuclear Material Safety and Safeguards

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GKJ
12/05/85
ORNL/NRC/LTR-85/34

MONTHLY PROGRESS REPORT FOR NOVEMBER 1985

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

PROJECT MANAGER: G. K. Jacobs

SCIENTIFIC STAFF: W. D. Arnold, J. G. Blencoe, A. D. Kelmers, R. E. Meyer, and V. S. Tripathi

ACTIVITY NUMBER: ORNL #41 88 54 92 6 (FIN No. B0290)
NRC #50 19 03 1

PROGRESS HIGHLIGHTS:

Evaluation of Yucca Mountain Information

One of the questions regarding the methodology of obtaining sorption information for the tuffs of Yucca Mountain is whether or not it is necessary to impose strict control of the CO_2 atmosphere on the groundwater/tuff system during sorption tests. To maintain the pH of groundwater J-13 at ~7, it is necessary to maintain an atmosphere over the system that contains ~1.5% CO_2. The pH of the groundwater will drift upwards to higher values without control of the atmosphere. During this report period, tests were conducted without this atmosphere control for cesium and strontium sorption on crushed Busted Butte tuff to determine whether there would be a significant difference in sorption behavior (by comparison to previous test results using CO_2 atmosphere control). The results are shown in Table 1.

Table 1. Effects of CO_2 atmosphere on sorption of cesium and strontium onto Busted Butte tuff[a]

		With control		Without control	
Element	Initial conc. (mol/L)	Final pH	Rs (L/kg)	Final pH	Rs (L/kg)
Cs	2.0×10^{-10}	7.10	454±17	7.82	522±22
Cs	1.0×10^{-8}	7.13	448±58	7.67	450±101
Sr	2.0×10^{-12}	7.07	11.8±.5[b]	7.78	19.0±0.4
Sr	1.0×10^{-8}	7.05	11.8±.3	7.78	19.9±1.4

[a]Test conditions: Batch contact experiment with 0.4 g tuff in 4 mL synthetic groundwater J-13. Contacted 7 d at 25°C. Separation of solid by 40 min centrifugation at 25000 rcf. Uncertainty is based on triplicate samples.
[b]Initial concentration 2.0×10^{-9} mol/L.

The results in Table 1 show that there is a small increase in Rs for cesium sorption without atmosphere control in the tests with an initial concentration of 2.0 $x10^{-10}$ mol/L. An apparent increase also occurred in the tests with an initial concentration of 1.0 x 10^{-8} mol/L, but one anomalously low value in the triplicate samples reduced the average value and resulted in a larger than normal uncertainty. For strontium, the increase in sorption ratio is much larger, almost a factor of 2. These increases are not unexpected because it is quite common to observe increases in sorption with increased pH on natural minerals, especially on natural oxides. Thus, the increase in sorption ratios for these elements is probably a result of the increase in pH. The results of these experiments suggest that sorption data relevant to Yucca Mountain should always be taken with CO_2 atmosphere control. Otherwise the pH will increase, and values of Rs may be higher than they would be at the pH (~7) of actual J-13 well water.

Preliminary to the study of americium sorption, we have been investigating the behavior of Eu(III) as a stand-in for americium. Our previous experience with solutions containing europium was that significant losses of europium onto the tube walls were followed by further slow losses of europium with time. To investigate possible losses from J-13 water, a sample of synthetic J-13 water was prepared and adjusted to pH 5.7 by introduction of gaseous CO_2. Europium was added until the solution reached a europium concentration of 10^{-7} mol/L, and the solution was then traced with ^{155}Eu. The solution was contained in a 250 ml centrifuge tube, and 1 mL aliquots were periodically (approximately once a day) taken for counting after centrifugation for 30 min at 25000 rcf. After the initial centrifugation, there was an immediate 16% loss of europium. After subsequent centrifugations, losses ranged from 30 to 50% with no apparent pattern of behavior with time over a 7 d period. The solution was then adjusted to pH 6.3 by introduction of air. (Percent losses stated are relative to the original solution concentration [10^{-7} mol/L].) Centrifugation and sampling once a day were continued. For the pH 6.3 solution, the loss of europium after the initial centrifugation was 40% followed by increasing losses up to 54% after another 9 d. The pH was then increased to 6.7 and sampling continued. The initial loss of europium in this solution was over 60%. Europium losses from solution then slowly increased with time to 77% after another 19 d. A portion of this solution was transferred to a clean tube after 12 of the 19 d. The loss of europium increased to almost 85%. Possible causes of these losses include sorption on tube walls, sorption on colloidal impurities to form pseudo-colloids, and formation of true colloids of europium. Because europium does not begin hydrolyzing significantly until the pH is increased to ~8, it is probable that true colloids did not form in the systems that we studied. Probably most of the loss is a result of the formation of pseudocolloids and adsorption on tube walls. These effects will have to be taken into account when we begin adsorption experiments with europium and americium, and they will make the study of the sorption of europium and americium rather difficult and somewhat slow.

In preparation for studies with tuff from the core samples, sections from two Topopah Spring core samples were broken off, and the process of grinding and sieving them is in progress.

Geochemical Modeling

No activity this month.

MEETINGS AND TRIPS:

None.

REPORTS AND PUBLICATIONS:

We are currently working on a draft of the annual report for the NRC Project Manager's review.

PROJECT MANAGEMENT:

Nothing to report.

PROBLEM AREAS:

None.

COST/BUDGET REPORT:

Expenditures were $50.4K for the month of November and $95.2K for the year to date. A detailed cost/budget report will be sent under separate cover.