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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 April 1985 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
Repository Licensing Analysis
and Support
Chemical Technology Division

SKW:bek

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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U.S. DEPARTMENT OF ENERGY

OMB NO. 038-R0190

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- [X] a. Scientific and technical report
- [ ] b. Conference paper: Title of conference _____

Date of conference _____

Exact location of conference _____ Sponsoring organization _____

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Nancy V. Taylor, Supervisor, Laboratory Records Dept.

Organization

Oak Ridge National Laboratory

Signature Nancy V. Taylor

Date May 7, 1985

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5/8/85
ORNL/NRC/LTR-85/17

MONTHLY PROGRESS REPORT FOR APRIL 1985

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, J. G. Blencoe, G. K. Jacobs, R. E. Meyer, and F. G. Seeley

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

PROGRESS HIGHLIGHTS:

Hanford Site Information Evaluation

Materials and Methods:

The materials and methods used in the evaluation of Hanford Site information have all been described in annual and quarterly progress reports. No additional activity is planned under this subtask.

Radionuclide Sorption/Solubility Values:

The BWIP seems to have taken the position that significant geochemical differences do not exist between basalt flows relative to radionuclide sorption or solubility, and that radionuclide retardation data obtained for a given basalt can be generalized for application to any or all basalt/groundwater systems along the release pathway to the environment. Therefore, we plan to measure neptunium sorption isotherms for three basalts in order to compare the effectiveness of different basalt/groundwater systems in retarding neptunium migration. The neptunium-235 which was ordered last September from ANL has finally been received and neptunium(V) solutions are now being prepared (the ^{235}Np is used to trace the ^{237}Np solutions) to allow determination of neptunium sorption isotherms for three basalts (Cohassett, Umtanum, and McCoy Canyon) under anoxic redox conditions at 60°C. The appropriate groundwaters will be used for each basalt. These sorption isotherms are the final neptunium activity planned relative to the Hanford Site.

All planned experiments to evaluate sorption of technetium onto basalt have been completed and the final results were reported last month. No further work with technetium is currently planned for this subtask. A summary of our conclusions concerning technetium sorption and solubility information for the Hanford Site will be given in the January-March quarterly progress report.

We have also completed essentially all planned experiments concerning sorption of uranium onto basalt. However, we still have underway a 50-day sorption isotherm for McCoy Canyon basalt at 60°C under anoxic conditions with synthetic groundwater GR-2. Prior experiments under these conditions had to be aborted due to defective seals between the caps and tubes. A new batch of caps and tubes are being used in the current experiment. In previous experiments with this McCoy Canyon/GR-2 system, the sorption ratios showed little difference between 14-day and 50-day experiments under oxic conditions at 60°C or under anoxic conditions at 27°C. The values of the sorption ratio were fairly low; in the range of 2 to 5 L/kg under either anoxic or oxic conditions. For our latest tests with the Cohassett/GR-4 system, we observed a significant difference between the results of 14- and 50-day experiments. The Rs values were in the range of several hundred L/kg in anoxic condition experiments after 50 days at 60°C. However, we used much more careful oxygen-removal techniques for these more recent Cohassett/GR-4 experiments than in the previous runs with McCoy Canyon basalt. These final uranium experiments underway with basalt are, therefore, designed to explore the possible effect of this improvement in methodology on the sorption behavior observed with McCoy Canyon basalt. The anoxic condition, 50-day, 60°C experiments now underway with McCoy Canyon basalt in GR-2 should tell us whether these more stringent oxygen-removal techniques will allow the uranium sorption ratios to exceed the 2 to 5 L/kg range which we observed in all previous experiments with this system.

Geochemical Modeling:

No additional activity is currently planned for this subtask. All our geochemical modeling work relative to the Hanford Site has been reported in annual and quarterly progress reports.

Yucca Mountain Information Evaluation

Materials and Methods:

The stability of the J-13 well water during storage is being evaluated. The NNWSI has previously reported microbiological growth in samples exposed to light. If we can not store J-13 well water in a stable manner, then we will have to consider use of synthetic J-13 water. Concern as to the stability and presence/absence of biological material in the water used in the sorption tests published by the NNWSI will also be evaluated. [The NNWSI has suggested that some anomalous plutonium sorption data may be due to scavenging of plutonium by microorganisms in the well water (LA-10032-PR)]. We have observed biological activity in samples of J-13 well water which have been exposed to daylight; colonies of microorganisms developed in approximately seven days. No growth was observed in other samples exposed only to fluorescent lights. We plan to evaluate what effect this biological activity might have on cation/anion concentrations in the well water, as well as its possible effect on radionuclide sorption.

As discussed last month, the pH of J-13 well water is very sensitive to the CO_2 content of the test atmosphere. We now plan to do all sorption

and solubility experiments under a CO_2/air atmosphere so that the solution pH is maintained near the natural pH value of 6.7 to 7.0. Most of the sorption work reported by the NNWSI was conducted in air and the pH of the well water rose to much higher values [The final pH values were 8 to 9 for many experiments (LA-984-PR, LA-10006-PR)], presumably due to loss of dissolved CO_2. We plan to devote considerable attention to consideration of the effect of this change in pH on the sorption data gathered by the NNWSI. We are now preparing the first controlled-atmosphere glove box for use with a CO_2-rich atmosphere (probably between 4 and 5% CO_2 in air). A PIR-2000 Process Gas Analyzer, which is capable of monitoring carbon dioxide in the concentration range of 1-10% in the atmosphere of one or more glove boxes, has been ordered from Horiba Instruments, Inc. This instrument, coupled with a small recycle pump, will allow us to monitor and control the CO_2 partial pressure in two glove boxes during sorption tests so that the solution pH can be maintained as desired.

We are comparing the Topopah Spring core and outcrop samples to determine if they appear to be sufficiently similar to permit use of the outcrop material in sorption tests to evaluate NNWSI data, or, conversely, to see if they appear sufficiently dissimilar so that use of the outcrop must be limited to methdology evaluation tests only. We will also characterize the Bullfrog and Calico Hills core samples because they are obviously so different in mineralogy and texture from Topopah Spring.

The NNWSI has not reported thin section analytical data for the rocks used in sorption experiments. Instead, they have pulverized their tuff samples and then tried to "back-out" mineralogical information from the powder by x-ray diffraction techniques. We are concerned that with rocks containing such large-scale heterogeniety (and prominent of lithic fragments in some samples), that pulverization of the tuff may obscure many important features and examination by classical petrographic techniques may offer a better opportunity to correlate the resulting sorption information with mineralogical and textural features of the rocks. The first thin sections obtained for the tuff samples were unsatisfactory, so the samples will be submitted to another vendor.

Radionuclide Sorption/Solubility Values:

Work under this subtask will be initiated once the concerns relative to the materials and methods, discussed above, are sufficiently resolved to permit the design of meaningful experiments.

When preparations for initiation of sorption studies with tuff are complete, we plan to begin by performing experiments with technetium. Because we do not anticipate significant sorption of technetium under the oxic environment of the tuff/groundwater system, we feel that relatively few experiments will be necessary to evaluate the published information. These experiments will utilize the existing supply of ^{95m}Tc, the gamma-emitting isotope which we use to trace our solutions of $^{99}TcO_4^-$. Its half-life is 65 days, and our present supply will decay to the point of uselessness in a few months. A usable amount of this isotope, which is cyclotron-produced, costs from $4000 to $5000. Technetium is usually identified as one of the key radionuclides which may be released from a repository, thus early evaluation of its behavior for the Yucca Mountain site is appropriate.

Geochemical Modeling:

The first priority for geochemical modeling relative to the Yucca Mountain site is to get EQ3/6 "on-line", because EQ3/6 is the major code in use by the staff of the NNWSI project. Once the code is debugged and some successful test cases have been run, the code will be used to simulate some of the calculations previously reported by the NNWSI project. This exercise will serve to help ORNL personnel become familiar with the code as well as provide several benchmarks for our version of EQ3/6.

A sensitivity study of uranium solubility/speciation is planned using MINTEQ and/or EQ3/6. The purpose of the study is to identify critical parameters that may significantly affect the results of sorption tests performed under various environmental conditions relevant to the Yucca Mountain site.

A few system-dependent inconsistencies were corrected in EQ3/6 this month in preparation for some test cases to be run next month. These logic errors were not part of the syntax errors corrected earlier (we previously reported that EQ3/6 was free of syntax errors), but were related to data disposition and device initialization and, thus, are specific to a given computer system. A few other errors are likely to be encountered during the running of test cases, however, it is anticipated that the errors will be small in number.

General Aspects:

All sorption information reported by the NNWSI for Yucca Mountain has been in an extended series of quarterly and summary progress reports by LANL. No topical report describing and collating sorption data and conclusions has been issued. The most recent progress report published by LANL is for October-December 1983 (LA-10032-PR). This represents a substantial delay in our learning of their more recent work and conclusions. It would be highly desirable to establish some mechanism for technical-level interaction between ORNL and LANL which would allow us more timely access to their research results. The present situation places us in the position of evaluating nearly two-year-old data, at best.

MEETINGS AND TRIPS:

None.

REPORTS AND PUBLICATIONS:

The annual report for FY 1984 is being prepared on mats and camera-ready copy will be send to the NRC Project Manager soon. The quarterly progress report for October-December 1984 was completed in draft form and sent to the NRC Project Manager for review and comment. The quarterly progress report for January-March 1985 is being compiled.

PROBLEM AREAS:

None

COST/BUDGET REPORT:

Expenditures were $39.2K for the month of April and $303.4K for the fiscal year to date. A detailed cost/budget report will be sent under separate cover.