OAK RIDGE NATIONAL LABORATORY
OPERATED BY
UNION CARBIDE CORPORATION
NUCLEAR DIVISION

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May 17, 1983

Dr. D. J. Brooks
HLW Technical Development Branch
Office of Nuclear Material Safety and Safeguards
U.S. Nuclear Regulatory Commission
449-SS
Washington, D.C. 20555

Dear Dave:

Enclosed is the progress report for the month of April 1983 for B0290, "Laboratory Evaluation of DOE Radionuclide Solubility Data and Other Geochemical Parameters, Experimental Strategies, Laboratory Techniques and Procedures."

Sincerely,

Allen

Allen G. Croff, Manager
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Chemical Technology Division

AGC:il

Enclosure

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MONTHLY PROGRESS REPORT FOR APRIL 1983

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

PROJECT MANAGER: A. G. Croff

ACTIVITY NUMBER: ORNL #41 88 54 92 6 (189 #B0290) / NRC #50 19 03 1

TECHNICAL HIGHLIGHTS

Neptunium Studies:

Preparation of the Alpha Laboratory facilities for the anoxic-condition tests with neptunium have continued during the month. Nearly all of the laboratory supplies and new equipment ordered has arrived. Transfer of the controlled-atmosphere alpha-containment glove boxes from the decommissioned plutonium facility to the Alpha Laboratory is expected to be accomplished next month. In preparation for the move, necessary ancillary craft work is under way. The computer-controlled counting equipment and data collection and assimilation programs are being prepared for the ^{235}Np and ^{237}Np analyses.

Technetium Studies:

A series of experiments have been carried out with crushed basalt under oxic conditions to confirm the technetium distribution coefficients reported by BWIP. [In the Site Characterization Report (SCR), BWIP recommended a conservative best-estimate value of 0 mL/g for technetium sorption on basalt under oxic conditions]. The range of test parameters investigated were:

Solid phase - Crushed Sentinel Gap basalt was used. Three particle sizes were tested: 20-70 mesh, 70-235 mesh, and <235 mesh. Some samples were ultrasonically washed to remove any fine particles not removed by the screening operation.

Solution - Synthetic groundwaters GR-1 and GR-2 were prepared according to BWIP procedures. The solutions were traced with ^{95m}Tc at 1E-12 M Tc(VII), and with ^{99}Tc at 1E-10, 1E-8, and 1E-6 M Tc(VII).

The tests were carried out at ambient temperature in glass tubes capped with PVC to minimize contact with air, i.e., oxygen, during the contact times of up to 3 days. After test completion and solution separation, selected solutions were examined by solvent extraction analytical techniques to measure technetium reduction; none was observed. Additional 3-day contact tests were carried out at 55°C in an air thermostat to see if the kinetics of technetium reduction by basalt would be accelerated at the elevated temperature; again no significant reduction was noted.

The results of the tests this month showed no significant change in the solution concentration (±10%) of added Tc(VII) in contact with basalt under these conditions. Thus, a calculated technetium sorption rate of 0 mL/g was obtained. This confirms the value recommended by BWIP in the SCR and is consistent with earlier published observations (Palmer and Meyer, J. Inorg. Nucl. Chem. 43, 2979 [1981]). The results also suggest that Tc(VII) is not rapidly reduced by contact with crushed basalt under these test conditions. The effect of oxygen from air dissolved in the synthetic groundwaters or in the space in the glass tube may be important here. The anticipated basalt reduction reaction with technetium may have been obscured or prevented by the oxygen in the test system. Information on this point will be gained in future anoxic condition tests.

Secondary Mineral Acquisition:

Suitable well-identified minerals are being acquired for future tests to evaluate BWIP basalt secondary mineral sorption and solubility data. This is necessary, since significant quantities of secondary minerals may not be available directly from BWIP until after completion of the exploratory shaft to the candidate repository level(s). A large quantity of western U.S. clinoptilolite is available. This material will be characterized by x-ray diffraction, inductively coupled plasma spectrometry, neutron activation, particle size, surface area, etc., analytical methods. A source of nontronite from Spokane, Washington has been identified and a supply of that mineral will be obtained and characterized. A source(s) of smectite clay(s) representative of probable BWIP secondary mineralization will be identified. Availability of these characterized minerals will allow initial sorption/solubility tests with secondary minerals to begin; these are among the more abundant minerals expected in the basalt flowtops. Additional less-abundant minerals will also be acquired and evaluated.

Calculational Activities:

The small thermodynamic data base provided with PHREEQ has been supplemented with some additional data. Preliminary test runs for speciation of uranium in surface water and groundwater systems have been made. Results indicate the need to add thermodynamic data for fluoride and phosphate complexes. In addition, GEOCHEM has been prepared for treatment of the same calculations. WATEQ2 may be less useful than the other models because new data cannot be conveniently added to it.

General Aspects:

A meeting between the ORNL and NRC staff members was held at ORNL to discuss the work plan and to tour the facilities to be employed in the experimental studies. In general, the work proposed in the plan was found to be acceptable and will provide the basis for the project's activities over the next several months. The work plan is currently being revised to accommodate a variety of comments provided by the NRC staff.

MEETINGS AND TRIPS:

A meeting was held at ORNL on April 12-13 to discuss the work to be performed under this contract and to review the first Draft Work Plan.

As a part of a trip conducted for another purpose, a brief meeting was held with G. Vandegrift et al. (ANL) to discuss procedures and view laboratory apparatus for the determination of radionuclide sorption measurements. Their flow-through leaching/sorption apparatus has many features of interest to this project because of its integrated, confirmatory nature. One of the most significant results of the early ANL experiments with the flow-through system is that hydrothermally-aged basalts apparently do not sorb as well as "fresh" basalt, contrary to expectations. The explanation for this was not apparent.

REPORTS AND PUBLICATIONS:

A trip report summarizing J. H. Kessler's trip to the Seattle ACS Meeting, PNL, and LBL was issued during this report period.

A meeting report concerning the visit of the NRC staff to ORNL was issued on April 15.

PROBLEM AREAS:

None

COST/BUDGET REPORT:

Expenditures for the month of April were $39.3K and expenditures to date are $166.3K. A more detailed cost/budget history is given in the attached exhibit. FY 1983 funding for the project in the amount of $400K was received during this report period.