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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 September 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:bek

Enclosure:

Monthly Progress Report for September 1985, w/attachments

cc: Office of the Director, NMSS (Attn: Program Support Branch)
Division Director, NMSS Division of Waste Management (2)
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K. C. Jackson, Geotechnical Branch
D. J. Brooks, Geotechnical Branch
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	ORNL/NRC/LTR-85/31	3370-2110

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

AUTHOR(S) (List all authors and division codes; if not an ORNL employee, indicate institutional address)
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J. E. Meyer (04), and S. K. Whatley (03)

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NRC

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UCN-10369
(3 10-84)

GKJ
10/7/85
ORNL/NRC/LTR-85/31

MONTHLY PROGRESS REPORT FOR SEPTEMBER 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, and R. E. Meyer

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

PROGRESS HIGHLIGHTS:

HANFORD SITE INFORMATION EVALUATION

As reported earlier, we have begun sorption isotherm measurements for neptunium on basalt for the systems GR-2/McCoy Canyon basalt, GR-2/Umtanum basalt, and GR-4/Cohassett basalt. We had hoped to determine the concentrations of neptunium in the groundwaters before and after sorption tests by tracing the solutions with ^{235}Np, which has a half-life of 396 d and emits low energy x-rays that can easily be detected by well-type NaI scintillation detectors. However, at the higher concentrations of total neptunium (^{237}Np + ^{235}Np), there were complications in the determination of the concentrations of neptunium with a NaI detector as a result of the presence of x-rays from both ^{233}Pa (the decay product of ^{237}Np) and ^{237}Np itself. Other counting methods of determining ^{237}Np at the higher concentrations of the isotherm were investigated — it appears that high resolution counting with a germanium detector will be adequate for determination of ^{237}Np in the presence of ^{233}Pa. However, our Instrumentation and Controls personnel have inspected our germanium detector and have concluded that it is defective. We will pursue how to repair the detector, but our high-resolution counting capabilities are badly compromised by this loss.

YUCCA MOUNTAIN INFORMATION EVALUATION

Preparation of a controlled-atmosphere glove box for work with Yucca Mountain samples continues. Mixtures of air and CO_2 were passed through the box and the pH of synthetic J-13 groundwater was monitored as a function of the composition of the mixture. As reported last month, a mixture containing 1.5% CO_2 maintained the synthetic J-13 at a pH of 7. At present, this flow-through system of passing both air and CO_2 through the box is not used. Rather, pure CO_2 is added to the box until the partial pressure of CO_2 is 1.5% as measured by our

CO_2 monitor. The CO_2 concentration in the box decreases slowly with time, and a periodic manual introduction of CO_2 is adequate to maintain the CO_2 partial pressure, and thus, the pH of J-13 synthetic groundwater to the appropriate level.

Determination of sorption isotherms of strontium and cesium on crushed tuff were begun. Crushed samples of the Busted Butte outcrop were used in synthetic J-13 groundwater. The initial experiments were designed to compare the behavior of tuff used with and without pre-equilibration in synthetic J-13. Initial concentrations of cesium and strontium ranged from trace levels to 10^{-4} mol/L, for samples not pre-equilibrated, and trace levels to 10^{-8} mol/L for the pre-equilibrated samples. In these experiments, the samples are gently shaken in the 1.5% CO_2 atmosphere box, and the runs will be terminated after seven days of contact.

MEETINGS AND TRIPS:

J. G. Blencoe, G. K. Jacobs, A. D. Kelmers, and R. E. Meyer attended a Data Review at the Los Alamos National Laboratory on September 26. The purpose of the Data Review was to discuss sorption information for Yucca Mountain. A meeting report was issued under the B0287 project and a copy is included as attachment #1.

A meeting between ORNL and NRC staff (K. Jackson, D. Brooks, and J. Bradbury) was held at ORNL on September 11-12. The purpose of the meeting was to provide an overview of the B0287 and B0290 projects to K. Jackson and to allow him to tour the ORNL laboratory facilities. Plans for the detailed Program Review to be held in Silver Spring on October 16-17 were also discussed along with various technical aspects of the projects.

PROJECT MANAGEMENT:

Effective October 1, 1985 Gary K. Jacobs will be taking over as Project Manager for both the B0290 and B0287 projects. Please arrange to have all correspondence and inquiries directed through G. K. Jacobs at the following address:

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Oak Ridge, TN 37831
(615) 576-0567; FTS 626-0567

PROBLEM AREAS: None

COST/BUDGET REPORT:

Expenditures were $26.4K for the month of September and $462.7K for the fiscal year to date. A detailed cost/budget report will be sent under separate cover.

Attachment 1.

MR-287-5
10/7/85

MEETING REPORT

AUTHOR:	G. K. Jacobs
LOCATION:	Los Alamos National Laboratory, Los Alamos, New Mexico
DATE:	September 26, 1985
PURPOSE:	To participate in a Data Review of Sorption Information for Yucca Mountain
PROJECT TITLE:	Technical Assistance in Geochemistry
PROJECT MANAGER:	S. K. Whatley
ACTIVITY NUMBER:	ORNL #41 37 54 92 4 (189 #B0287) NRC #50 19 03 01

GENERAL COMMENTS

The Data Review provided an excellent opportunity to interact informally with the staff of Los Alamos National Laboratory (LANL) and to discuss our concerns relative to the sorption information that has been published to date. The meeting was organized around two presentations (given by A. D. Kelmers) that addressed our major concerns (see detailed discussion below). A tour of the laboratory facilities was taken after the informal discussions were completed.

The meeting was useful in that it provided an opportunity for frank and open interaction between ORNL and LANL staff. The NRC should be commended for arranging this meeting. Such interaction has not been possible in the more formal workshops that we have attended in the past at LANL. We strongly urge that similar meetings be held on a somewhat regular basis (e.g., every six months) for both the NNWSI and BWIP projects. To help minimize the number of persons involved, these meetings should be focused on specific aspects of geochemistry.

We feel that both ORNL and LANL staff benefited from the meeting and came away with a better understanding of the viewpoints and concerns of their counterparts. Clearly, we are now in a better position to formulate our experimental evaluation of sorption information for Yucca Mountain.

OBSERVATIONS RELATED TO THE PRESENTATIONS OF A. D. KELMERS

During the one day Data Review on radionuclide sorption information involving NRC/NMSS, ORNL, and DOE personnel from several facilities, which was held at Los Alamos National Laboratory (LANL) on September 26, 1985, A. D. Kelmers presented two informal talks. Extended and intensive discussion with LANL staff and others present at the meeting resulted from the subject matter of the talks; these interchanges are summarized below:

I. "Concerns Relative to the Applicability of the Yucca Mountain Sorption Information for Site Performance Assessment Purposes", A. D. Kelmers, ORNL.

Five concerns were identified in this talk; these problem areas were based on our Letter Report LR-287-7, July 15, 1985. The concerns and a brief summary of the discussion of each concern at the meeting are presented below:

1. Lack of Sorption Information Compilation and Synthesis

We felt that the extensive reporting of experimental sorption information obtained at LANL was primarily limited to descriptions of what was done and the numbers obtained, without accompanying compilation and explanation of how the information was to be used in site performance assessment. Thus, we were not able to assess the applicability, relevance, or completeness of the Yucca Mountain sorption information for NRC licensing purposes. LANL staff responded that some discussion had been included in earlier reports, particularly "Tuff 4" [LA-9328-MS (1982)], but agreed that more explanation and summarization would be useful. LANL staff stated that they are preparing a topical report on sorption; this may answer some of our questions. No expected completion date was given for this report.

2. Timeliness of Reporting

We noted that 12 to 18 months may pass from the time LANL performs an experiment until we see a published description of it in their quarterly progress reports. LANL staff correctly pointed out that our ORNL work is not available to them as NUREG/CR reports any more rapidly. We urged that some mechanism for more rapid and informal exchange and interaction between ORNL and LANL be considered, but there was no formal response to this suggestion from NNWSI management staff at the meeting. LANL staff indicated that the quarterly progress reporting had been discontinued and only topical reports will be issued in the future. Thus, it may be some time before we see any new sorption information for Yucca Mountain. This information hiatus may represent a significant problem for the NRC evaluation of DOE sorption information.

3. Absence of a Performance Assessment Strategy for Sorption Modeling

We indicated that available information does not allow one to determine the nature of the comprehensive strategy for performance assessment modeling of sorption at Yucca Mountain. We suggested that isotopes of Am, Pu, and Tc may be the key radionuclides for sorption modeling and that these elements are not well described in the published information. LANL staff was open and frank in stating that they had done considerable experimental work with Am and Pu which is not published because they could not understand the data; sorption ratios for these elements seemed to vary independent of test parameters, and the controlling sorption processes were not known. LANL staff has conducted sorption experiments with these elements for nearly seven years, but they seemed not to be able to predict when the behavior of these elements in the engineered facility or the far field at Yucca Mountain may be understood. This uncertainty is not surprising, however. The chemistry of actinides at

low concentrations in near-neutral solutions is extremely complex and difficult to study. There is a lack of general knowledge of the speciation and valence states - making it difficult to characterize even the starting solutions in sorption tests. This uncertainty in actinide chemistry represents a serious problem for the DOE site projects in attempting to develop a strategy for sorption/solubility tests, as well as for the NRC in developing a strategy for evaluating the data obtained by the DOE. There are experimental approaches that may provide some partial answers to the behavior of actinides in groundwaters of this type. We plan to consider this problem further and discuss with the NRC Project Manager some possible options for experimental efforts to pursue this problem. Also, we suggest that the NRC begin to consider what regulatory strategy may be necessary to ensure that sorption information on elements with complex chemical behavior will be acceptable for licensing purposes (e.g., detailed understanding or empirical relationships?).

We also discussed the apparent lack of definition of groundwater flow paths and mineralogical characteristics of these flow paths. Ideally, the flow paths and minerals present should be well-defined prior to starting sorption tests. Unfortunately, this ideal situation is not realized for the Yucca Mountain site. The approach that LANL has taken of testing a wide variety of tuff samples and going from "simple" radionuclides (Sr, Cs, Ba, Ra) to "complex" radionuclides (Am, Pu, Np) is understandable given their difficult situation of unknown flow paths and mineralogy. Unfortunately, they are just beginning to address the sensitivity of sorption results to parameters such pH, groundwater composition, ionic strength, etc. We encouraged LANL to pursue these sensitivity tests that may shed some light on the important parameters and processes that most affect sorption.

We expressed some concern over the potentially inappropriate modeling of sorption processes in the transport codes (i.e., simple Kd approach). A staff member from the performance assessment group at Sandia explained that a single Kd value will not be used in the models. Rather, a stochastic approach will be used where a mean value and associated distribution will be modeled. This approach may be acceptable, but the range of Kd values will have to be carefully evaluated to ensure that nonconservative results are not generated, especially considering the complexity of the geology and geochemistry at Yucca Mountain. This approach is a good example of using an empirical relationship rather than making extrapolations and predictions based on a sound qualitative understanding of the process. The potential complications in this approach to licensing are still of some concern to us.

4. Unevaluated Batch Contact Methodology Test Protocol and Parameters

We felt that the reported information did not explain how the methodology was optimized and that the information developed could, therefore, be biased or inaccurate. LANL staff vigorously defended their methodology. Some of the descriptive information was shown to be available in various progress reports, but LANL staff did agree that it was not summarized

in one place. The LANL assumption that freshly crushed rock surfaces correctly model in situ rock is of particular importance to the NRC evaluation (see below).

5. Groundwater Instability During Experiments

We expressed concern that, in much of the LANL work, the groundwater pH had risen by nearly two units due to loss of dissolved CO_2 to the atmosphere. LANL staff defended use of this information as being conservative, since it does not allow for carbonate complexation of actinides, and the fact that in recent LANL experiments where the pH was held stable (pH 7) by use of a CO_2-rich atmosphere, little (always in the direction of more favorable sorption) or no change had reportedly been seen in the sorption ratios for many elements. LANL stated that most experiments will be done outside of CO_2-atmosphere boxes and that only periodic checks on validity of this test method will be performed. We feel this approach may warrant additional attention by the NRC.

A second concern that we expressed involved the presence of microbiological growth in the J-13 groundwater solutions and the possible effects that this could have on the measured sorption ratios. The response of LANL to this concern was not entirely clear to us. The LANL project is beginning to develop tests to address this problem, but we want to emphasize that this issue merits future consideration. We were particulary interested in the interpretation of LANL to include any sorption onto biological particles as part of the "rock." This interpretation will require careful consideration when evaluating sorption modeling in performance assessments so that proper accounting of mitigating processes is included.

II. "Application of Radionuclide Sorption Information for Prediction of Retardation in Fracture-Flow Geologic Systems", A. D. Kelmers, ORNL.

In this talk, we reviewed the history of the development and the assumptions underlying the conventional use of batch contact sorption experimental methodology and of calculated retardation factors to predict radionuclide migration in geologic systems. Our conclusions were:

1. Reliance on equilibrium distribution coefficient (Kd) and retardation factor (Rf) concepts leads to inaccurate and nonconservative predictions of radionuclide releases to the environment for fracture-flow systems.

2. Use of freshly crushed rock is not representative of fracture-flow minerals.

3. Interesting modeling work is being done in Europe to take credit for matrix diffusion.

4. No migration model deals with nonequilibrium sorption reactions or multiple radionuclide species and forms; unfortunately, these conditions may predominate for key radionuclides.

While there was some interesting discussion following this presentation, the LANL staff indicated that the subject was not relevant to Yucca Mountain because the working hypothesis of the NNWSI Project is that the unsaturated

zone beneath Yucca Mountain behaves as a permeable medium with groundwater flow through the bulk matrix rather than as predominantly fracture-flow. This hydrologic issue will bear watching closely in the future, because if groundwater movement occurs principally by fracture flow, the sorption information for Yucca Mountian may be of limited applicability. Important to this issue is the consensus at the meeting that the saturated zone will have fracture flow as the dominant mechanism of flow.

III. Assumptions Underlying the LANL Sorption Approach

As a result of this meeting, we believe that we have developed a better understanding of some of the fundamental assumptions underlying the LANL approach to the development of sorption information for Yucca Mountain. Our interpretations of the LANL statements are listed below:

1. The current assumption is that, within the unsaturated zone, all tuff units at Yucca Mountain will exhibit porous flow. Therefore, (1) the bulk rock will be available for sorption, and use of sorption ratios measured in batch contact tests will model in situ behavior, (2) the bulk rock minerals will be the sorption medium rather than fracture-lining minerals, and (3) the use of freshly crushed rock samples will represent in situ sorptive minerals. This hydrologic issue warrants close monitoring in the future by the NRC geochemistry group. If the hydrology is not porous flow, then the LANL approach and resulting sorption information may be both inaccurate and nonconservative. LANL recognized that flow within the saturated portions of the site will be predominantly via fractures. Thus, the applicability of the crushed-rock, batch contact tests for these units remains somewhat questionable. We feel that some further consideration of this issue may be necessary.

2. Crushing drill core samples of various tuff units to obtain material for batch contact tests does not alter the mineral sorptive properties. LANL reached this conclusion because the crystal size of the minerals is stated to be smaller than the particles generated by crushing; care is taken by LANL to exclude any fines generated during crushing. It may be advisable to experimentally reexamine this assumption. If it should prove to be invalid, then all LANL sorption information could be suspect.

3. Work at 25°C is conservative for representing sorption at higher temperatures because sorption reactions would accelerate with temperature. Considering the reactions possible in these complex mineral/groundwater systems, this assumption may warrant reexamination. Some specific cases may exist where sorption decreases with temperature as a result of speciation changes, colloid formation, mineral surface reactions, etc.

4. Work without control of CO_2 partial pressure and change of solution pH by two units is conservative for modeling in situ sorption under constant pH. This LANL assumption seems particularly questionable and may require more than a casual check as was implied by LANL. Reactions that change mineral surfaces, radionuclide speciation, etc., could be

important and some cases may exist where the higher pH sorption values are nonconservative. We plan to explore this assumption in our initial experimental work.

MISCELLANEOUS DETAILED OBSERVATIONS

1. Our general impression is that their batch sorption work is carefully performed. There is no apparent need to be concerned with improper techniques, especially with regard to their radionuclide counting equipment. However, as discussed above, we are concerned with several aspects of the overall sorption methodology of LANL.

2. We are somewhat concerned with the column experiments utilizing long (1-2 m) columns of crushed tuff. Long, thin columns are sometimes prone to channeling. This area needs to be given further consideration.

3. Scott Sinnock (Sandia National Laboratory) discussed the relationship between percent groundwater saturation in tuff and matric potential. According to Sinnock, it is significant that numerous hydrologic measurements in the unsaturated zone at Yucca Mountain indicate groundwater saturation values of 60 to 70%. Sinnock stated that saturation values in this range imply extremely high matric potentials, which virtually preclude significant fracture flow in the deeper regions of the unsaturated zone. Furthermore, he stated that tuffaceous rocks that are 60 to 70% saturated have much higher matric potentials than tuffaceous rocks that are only slightly more saturated (e.g., the tuffs in G-tunnel at 90% saturation). The tuffs in G-tunnel, because they are closer to being saturated, have a much lower matric potential and are observed to have groundwater flowing through major fractures intersecting the tunnel. Therefore, Sinnock stated that the working hypothesis of NNWSI is that the relatively low saturation values (60 to 70%) preclude significant fracture flow in the deeper regions of Yucca Mountain, whereas saturation values near 90% (if they were observed) would permit such flow. This hypothesis is highly controversial and merits additional attention.

2. LANL has been unsuccessful in its attempts to use autoradiography to identify the principal minerals sorbing radionuclides from solution. The difficultly is that the amounts of radionuclides sorbed onto the minerals in polished thin sections at the completion of a sorption experiment are so small that they cannot easily be detected by conventional surface analytical techniques. One remedy for this problem is to increase the concentration of radionuclides in the solution used to contact the rocks during the sorption test. However, LANL correctly pointed out that this approach might be unsatisfactory because the speciation of radionuclides may be significantly different at higher concentrations, thereby (conceivably) influencing the sorptive behavior of the radionuclides and producing misleading results. To circumvent the problem of insufficient quantities of radionuclides sorbed onto the surfaces of the thin sections, it may be necessary to either increase water/rock ratios, lengthen contact time, or replenish radionuclide-containing solutions in low water/rock ratio tests.