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Mr. Walton Kelly
U.S. Nuclear Regulatory Commission
Mail Stop 623-SS
Washington, DC 20555

Dear Mr. Kelly:

Enclosed is the trip report for the geochemistry project coordination meeting held at Oak Ridge National Laboratory on August 29, 1984. The meeting was authorized under FIN A-1756.

Please feel free to contact me if you have any questions or comments.

Sincerely,

Malcolm

Malcolm D. Siegel
Waste Management Systems
Division 6431

MDS:6431:jm

Enclosure

Copy to:
Office of the Director, NMSS
Attn: Program Support
Robert Browning, Director
Division of Waste Management
Malcolm R. Knapp
Division of Waste Management
John Starmer
Division of Waste Management
Office of Research, NRC
Document Control Center,
Division of Waste Management
6431 R. M. Cranwell
6431 M. S. Chu
6431 M. D. Siegel

# TRIP REPORT FOR PROJECT COORDINATION MEETING

# AT OAK RIDGE NATIONAL LABORATORY

August 29, 1984

FIN A-1756

Malcolm Siegel

September 13, 1984

Sandia National Laboratories
Waste Management Systems Division
Albuquerque, NM 87185

SUMMARY

Malcolm Siegel met with staff members of Oak Ridge National Laboratory on August 29, 1984 in Oak Ridge, Tennessee. The purpose of this meeting was to acquaint ORNL staff with the objectives and scope of the SNLA Geochemical Sensitivity Analysis Project (A-1756) and to familiarize SNLA staff with the ORNL geochemistry projects B-0287 (Technical Assistance in Geochemistry) and B-0290 (Laboratory Evaluation of DOE...Procedures). The following topics were discussed: (1) thermochemical data base development, (2) sorption data base development, (3) criteria for experimental design, and (4) theoretical calculations and experimental studies carried out at ORNL. It was agreed at this meeting that exchange of ideas and progress reports and any cooperation between SNLA and ORNL approved by the NRC would be beneficial to both geochemistry programs.

PARTICIPANTS

| | |
|---|---|
| J. G. Blencoe | ORNL/Chem. |
| G. K. Jacobs | ORNL/Environ. |
| A. D. Kelmers | ORNL/Chem. Tech. |
| D. C. Kocher | ORNL/Health and Safety |
| S. Y. Lee | ORNL/Environ. |
| R. E. Meyer | ORNL/Chem. |
| S. K. Whatley | ORNL/Chem. Tech. |
| M. D. Siegel | SNLA/6431 |

ITINERARY: AUGUST 29, 1984

8:30 A.M. Presentation and discussion of SNLA Geochemical Sensitivity Analysis Project (A-1756).

1:00 P.M. Discussion of ORNL geochemistry projects B-0290 and B-0287.

SIGNIFICANT FINDINGS

1. Thermochemical Data Base Development

Under FIN A-1756, SNLA will produce a critically-assessed state-of-the-art thermochemical data base and evaluate uncertainties associated with solubility/speciation calculations. During the meeting, the possible uses and contributions of ORNL to the data base were discussed. According to ORNL staff, theoretical calculations of radionuclide solubility, speciation, and sorption are not a

major task of the ORNL projects. ORNL, however, has calculated radionuclide solubilities in basaltic ground waters using the PHREEQE geochemistry code and the thermodynamic data base of Early et al. (1982). This type of calculation was also planned for FIN A-1756. The ORNL results should be examined to determine if the proposed SNLA calculations could provide any new information. ORNL staff are using MINTEQ to carry out solubility and saturation calculations, and they hope to install EQ3/6 onto their computer system in the near future. In FY85, the use of PHREEQE will be reduced and ORNL will rely on MINTEQ and EQ3/6, the same codes that DOE staff will probably use in their work. ORNL has modeled synthetic BWIP waters at 25°C and 60°C and calculated mineral saturation indices. Similar calculations may be carried out for ground waters at other sites. It was suggested that SNLA prepare a list of planned geochemical modeling calculations to identify possible areas of overlap between the ORNL and SNLA programs. This will be done if requested by the NRC.

During the meeting, a strategy was discussed to ensure that the SNLA efforts would be of maximum benefit to the ORNL project. In general, it was agreed that theoretical calculations of the solubilities and speciation of actinides have limited utility as a predictive tool due to the lack of relevant data. Their greatest uses are: (1) as a tool in performance assessment sensitivity studies, and (2) in conjunction with solubility/speciation experiments. Gary Jacobs at ORNL expressed interest in serving as a member of the technical advisory committee that will review the format and contents of the thermochemical data base. His participation is subject to approval by NRC and ORNL management.

It was suggested by ORNL staff that the data base be formatted to allow its use with the PHREEQE, EQ3/6, and MINTEQ geochemistry codes. With this approach, only thermochemical data ($\Delta G°_f$ $\Delta H°_f$, $C_p$) and empirical coefficients for high temperature (e.g., Clark-Glew) or high salinity (Pitzer) extrapolations would be included in the data base. Data preprocesser routines would be written to reformat the input file for each of the desired geochemistry codes. This strategy has several advantages: (1) it would facilitate quality control by standardizing the data bases used by different research groups; (2) errors due to misreading data during compilation of separate data bases would be greatly reduced; (3) new data would be readily available to all groups using the central "source" data base. This approach would require considerably more work than the compilation of a data base which is formatted for a single code. Some of this additional work, however, may have been carried out under other programs. Several preprocessors to convert data from one format to another have already been written. The code EQTL creates data bases formatted for EQ3/6 from a data file of thermodynamic

data. A companion code, MCRT, is a data base building program which calculates stability constants as a function of temperature from thermochemical data. (Isherwood and Wolery, 1984.) The code PHLT (INTERA, 1983) was written to transform the EQ3/6 data base into a format readable by the PHREEQE code. At Pacific Northwest Laboratories, a program is underway to make the MINTEQ and EQ3/6 codes more compatible. This topic will be discussed in detail at the first meeting of the technical advisory committee for the thermochemical data base. Specific recommendations on the approach to take in developing the data base will be sent to the NRC after this meeting.

2. Sorption Data Base Development

Under Task 1B of FIN A-1756, SNLA will compile available sorption data and examine the uncertainty in integrated discharge related to the assumption of linear equilibrium sorption in model calculations. We have previously proposed that this computerized data base be made accessible to the NRC and NRC contractors. Potential uses of a data base management system by ORNL were discussed at this meeting. These include the use of the system as a tool in evaluation of DOE data and as a source of information for the interpretation of new experimental data produced by ORNL under FIN B-0290. The data management system could also provide the NRC, SNLA, and other NRC contractors with access to the data produced under B-0290 in a timely fashion. Any coordination of efforts by SNLA and ORNL in this way must be first approved by the NRC. An accurate estimate of the resources required for any cooperative efforts and an assessment of the utility of this data base to ORNL will not be possible until more information on the format and capabilities of the data base management system is available. The two systems currently under investigation are ISIRS and System T2000. These data bases are discussed in the August monthly report for FIN A-1756. Detailed descriptions of the data base systems will be sent to the NRC and to ORNL (D. Kelmers) as they become available.

3. Criteria for Design of Sorption Experiments

Under Subtask 1C of FIN A-1756, SNLA will provide advice on the criteria for the design of experiments carried out to quantitatively observe the effects of important speciation reactions. Such effects may be important to both column and batch sorption experiments. ORNL staff expressed interest in the geochemical scenario development planned under this task and in the results of calculations which will describe the duration of sorption experiments that are required to ensure that important speciation effects are not overlooked. It was agreed that, for certain scenarios, the experimental run times that are required by available data may not be attainable. In such situations, the calculations carried out under Subtask 1C can identify the need for non-geochemical barriers to radionuclide migration.

4. Description of Geochemistry Projects at Oak Ridge

S. Whatley, G. Jacobs, and D. Kelmers described the results of recent work performed under FIN B-0290. Of particular interest were the results of apparent concentration limit measurements and sorption experiments for Np, Tc, and U in basalt ground-water systems. The results of anoxic ($10^{-10}$m $O_2$, Eh~-0.2 V) sorption experiments suggest that many of the sorption data obtained by BWIP staff under reducing (hydrazine) conditions are questionable and should not be included in the proposed SNLA sorption data management system. The sorption behavior of Tc under anoxic conditions was found to be very complicated and characterized by slow kinetics. It is very sensitive to ground-water composition and is considered by ORNL staff to be too complex to be unravelled under the ORNL project. A solution composition dependence was also found for Np sorption. Pentavalent Np was the predominant valence in solution while tetravalent Np was removable from the rock. The results of theoretical solubility calculations for Np, U, and Tc do not agree well with radioelement apparent concentration limits measured in experimental studies. The sources of these discrepancies, however, have not yet been identified. ORNL staff suggested that, in some cases, a detailed knowledge of the speciation and site-specific solubilities of many radionuclides may not be attainable before a HLW repository is licensed. Similar concerns were expressed by NNWSI contractors at the July NNWSI/NRC Geochemistry Workshop. This topic will be discussed at the October 1 meeting of the technical advisory committee for the thermochemical data base (to be developed under Task 1 of FIN A-1756). The lack of knowledge of radionuclide speciation will introduce uncertainty into calculations of integrated radionuclide discharge. The parametric calculations to be carried in Task 1 of FIN A-1756 will assess the magnitude of the associated error.

REFERENCES

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INTERA Environmental Consultants, 1983. PHREEQE: A Geochemical Speciation and Mass Transfer Code Suitable for Nuclear Waste Performance Assessment, ONWI-435. Battelle Memorial Institute, Columbus, Ohio.