Sandia National Laboratories
Albuquerque, New Mexico 87185

WM-RES

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September 11, 1984

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

Dear Mr. Kelly:

Enclosed is the monthly report for FIN A-1756, Geochemical Sensitivity Analysis for August 1984.

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:
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PROGRAM: Geochemical Sensitivity Analysis FIN#: A-1756

CONTRACTOR: Sandia National Laboratories BUDGET PERIOD: 4/20/84 - 9/30/84

DRA PROGRAM MANAGER: W. R. Kelly BUDGET AMOUNT: 200K

CONTRACT PROGRAM MANAGER: R. M. Cranwell FTS PHONE: 844-8368

PRINCIPAL INVESTIGATOR: M. D. Siegel FTS PHONE: 846-5448
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PROJECT OBJECTIVES

The objective of this project is to provide technical assistance to the NRC in determining the sensitivity of far-field performance assessment calculations to uncertainties in geochemical and hydrological input data and in the representation of geochemical processes in transport models. In Task I, the error in model calculations of integrated radionuclide discharge due to speciation, kinetic and sorption effects will be evaluated. In Task II, the potential importance of organic molecules and colloids will be examined. SNLA will assist the NRC in determining how geochemical processes should be represented in transport models under Task III. Short-term technical assistance will be carried out under Task IV.

ACTIVITIES DURING AUGUST 1984

Task I Uncertainty in Integrated Radionuclide Discharge

Subtask 1A. Speciation Effects

A meeting of the technical advisory committee for the compilation of the thermochemical data base has been scheduled for October 1, 1984, at Oak Ridge National Laboratory. An agenda and list of invitees have been appended as attachments to this monthly report. Attachment II is not to be interpreted as a finalized list of committee members; not all of the participants at this meeting will be members of the advisory committee. Proposed committee members are indicated on the list, but not all of the desired committee members can be present at the first meeting. The proposed membership of the committee will be formulated after the October meeting and a list of the members will be sent to the NRC for approval in the October monthly report.

We have requested all invitees to pay their own expenses. The committee meeting was scheduled to take advantage of the workshop on geochemical modeling sponsored by Oak Ridge

National Laboratory which will be held on October 2 to 5. All of the invited participants have been invited to the ORNL workshop. At this time, none of the invitees have indicated that they will require reimbursement for their travel expenses. The total costs for the meeting will be held to less than $2000. This figure includes only incremental costs for the committee meeting that are not associated with attendance at the ORNL geochemistry workshop.

The possibility that the thermochemical data base could be of use to the ORNL geochemistry programs sponsored by the NRC was discussed at a meeting between Sandia and ORNL personnel. Oak Ridge staff expressed interest in gaining access to the data base. A trip report describing these discussions will be forwarded to the NRC under a separate cover during September.

Subtask 1B. Equilibrium Sorption Effects

During August, the work under this subtask included investigation of the capabilities of two computerized sorption data managements that might be used in this project. These systems are the International Sorption Information Retrieval System, (ISIRS) installed at Pacific Northwest Laboratories, (PNL) and the System 2000 used by NNWSI which is maintained by DOE at Sandia Labs (Division 6312). Direct communication between Division 6431 SNLA and Division 6312 SNLA is prohibited by Sandia Laboratories policy. NRC-supported SNLA staff will be unable to access the DOE/NNWSI data base. The general capabilities of System 2000 at SNLA are known, however, to be similar to the data base system developed by Corstar for the NRC. This system can be used efficiently for data input and retrieval, but it has limited subsetting and statistics capabilities. The data base contains sorption data compiled from LA-9328-MS (Daniels et al., 1982) and some mineralogical and hydrogeologic data relevant to geochemistry. There are no subroutines for data manipulation or isotherm generation. This system, like the CorStar system, will probably not be useful for FIN A-1756.

Whereas System 2000 is a multi-use data management system, the ISIRS computerized information retrieval system has been developed specifically to facilitate the tabulation and synthesis of the multitudinous data needed to quantify nuclide retardation. The ISIRS has a number of capabilities which will be useful for performance assessment studies and geochemical sensitivity analysis in A-1756.

Of particular interest are the ISIRS data processing steps that allow means, ranges and frequency distributions to be produced for any element's distribution coefficient (Rd) for any set of specified conditions. The generation of frequency distributions is particularly useful for stochastic modeling of uncertainty/sensitivity. ISIRS also includes provisions to generate adsorption isotherms, to check for non-linearity in the isotherms and to identify the magnitude of irreversible

adsorption. When data are reported versus time, ISIRS can be used to investigate kinetics of adsorption-desorption.

The current ISIRS has very few data for basalt or tuff, 250 entries for sediments associated with salt, 450 entries for granite and 170 entries for clay minerals. The standard coding form used for data entry is very detailed and includes spaces for information on rock, ground-water, nuclide characteristics and experimental methodology. It is likely that values for many of the parameters listed in the form are not currently available in the literature. The completeness of the data form, however, ensures that the data base structure will not become obsolete if future reports of sorption studies contain more detailed descriptions of experimental procedures. Form entry and list applications subroutines are now available to facilitate data entry. These routines are detachable from the main ISIRS and can be used by anyone with a DEC VT52 (or equivalent) terminal. Data analysis and manipulation can be performed later on the central ISIRS host computer.

We do not feel that the applicability of the ISIRS to activities in FIN A-1756 can be evaluated without some "hands-on" experience with the system. In a future monthly report, we will submit a list of tasks to be carried out on the ISIRS. We propose to carry out these activities (plus any suggested by the NRC) on the ISIRS at PNL in order to assess the ease of operation of the system. A report of the activities, a discussion of the potential uses of ISIRS in the SNLA program and our recommendations would be sent to the NRC after the exercise.

Potential uses of a sorption data management system were discussed at the SNLA/ORNL meeting described above. ORNL staff expressed interest in use of the system, subject to NRC approval. This subject is discussed in more detail in the forthcoming trip report cited above.

Subtask IC. Kinetic Effects

No new activity during August.

Subtask ID. Dynamic Effects

Activities related to studies of matrix diffusion and equivalent porous medium approximations for fractured rock continued during August. The error associated with the use of a porous medium approximation instead of a semi-infinite approximation is under examination. Theoretically, the transport of radionuclides whose relative concentrations (C/C_0) are close to zero at the center of the matrix block bound by parallel fractures should be described by a semi-infinite medium approximation. It would be useful at this time to determine if development of such an approach is truly required for performance assessment calculations or if the porous medium approximation will be sufficiently accurate under site or medium-specific conditions.

During August, the concentration profiles and penetration depths of specific radionuclides in a semi-infinite basalt matrix were calculated. The concentrations of the radionuclides in the matrix adjacent to a fracture relative to the radionuclide concentration in the fracture (C/C_o) were calculated as a function of distance into the matrix at 10,000 years.

The equation describing the 1-D diffusion of a decaying radionuclide is

$$\partial C/\partial t = (D/R)\,\frac{\partial^2 C}{\partial x^2} - \lambda C$$

where D = effective diffusivity
R = retardation factor of radionuclide
λ = decay constant of radionuclide

For a constant concentration (C_o) boundary condition, the solution is given by

$$C(x,t) = \frac{C_o}{2}\left\{ e^{-x\sqrt{\frac{R\lambda}{D}}}\,\mathrm{erfc}\left[\frac{x}{2\sqrt{Dt/R}} - \sqrt{\lambda t}\right] + e^{x\sqrt{\frac{R\lambda}{D}}}\,\mathrm{erfc}\left[\frac{x}{2\sqrt{DT/R}} + \sqrt{\lambda t}\right]\right\}$$

The above equation was used to calculate $C(x,t)/C_o$ for the 21 radionuclides listed in the table below. The retardation factors used were taken from the WISP study (WISP, 1983). Data from the BWIP site (Long, 1978) indicate that spacing between fractures in basalt flows at Hanford ranges from 0.2m to 3m and is generally < 1m. The relative concentration C/C_o for each radionuclide at x = 0.5m (typical half-spacing between fractures at Hanford) was calculated. The C/C_o values listed in the table indicate the concentration level of each radionuclide at x = 0.5m, assuming that the matrix extends to infinity. However, if x = 0.5m were the true half-spacing location, C/C_o could be much higher than that shown in the table. Therefore, the results of this scoping calculation over emphasize the need for a semi-infinite approximation to model transport. The values of relative concentration listed in the table show that, except for the very short-lived (Pu241, Pu238, Am241) and the highly retarded (Th isotopes) radionuclides, the semi-infinite approximation is not required and, in fact, may be a poor representation for basaltic rock. A more sophisticated parametric examination of the behavior of the strongly retarded or short-lived nuclides will be carried out in the near future in order to determine if their behavior can be described by a modified equivalent porous medium approximation.

RESULTS OF SEMI-INFINITE MATRIX CALCULATIONS

Isotope	Half-life (Yr)	Retardation Factor	C/C_o +
Pu240	6.76E3	200	0.37
*Pu241	14.6	200	~0
Pu242	3.79E5	200	0.53
*Pu238	89.	200	~0
Pu239	2.44E4	200	0.48
U236	2.39E7	50	.75
U233	1.62E5	50	0.75
U238	4.51E9	50	0.75
U234	2.47E5	50	0.75
U235	7.1E8	50	0.75
*Th232	1.41E10	5000	1.6E-3
*Th229	7.3E3	5000	7.3E-4
*Th230	8.0E4	5000	1.5E-3
Cm245	8.27E3	500	0.21
Cm246	8.0E3	500	0.16
*Am241	433.	500	3.6E-3
Am243	7.65E3	500	0.20
Np237	2.14E6	100	0.66
Sn126	1.0E5	1000	0.15
Tc99	2.14E5	5	0.92
I129	1.6E7	1	0.96

*Nuclides for which semi-infinite approximation might be used and which will be examined in more detail in September and October.

+at $x = 0.5$ meter and $t = 10^4$ yrs

References

Long, P. E., "Characterization and Recognition of Interflow Structures, Grande Ronde basalt: Rockwell Hanford Operations," RHO-BWI-LD-10, 1978.

WISP, Waste Isolation Systems Panel, "A Study of the Isolation System for Geologic Disposal of Radioactive Wastes" National Research Council, National Academy Press, 1983.

Task II Evaluation of Error Due to Organics and Colloids

Subtask IIA Organics

No activity during August

Subtask IIB Colloids

An abstract submitted for of a technical paper describing the transport and capture colloids in single fractures has been accepted for presentation at the Materials Research Society meeting in November, 1984. The paper is currently being completed and emphasizes the effects of the rate of production of the colloids and changes in parameter values on the calculated rates of transport and capture of the particles. Average velocities are estimated for the particle front and compared to the average fluid velocity. The results of this work should indicate the relative importance of different phenomena governing colloid transport and capture and the degree of retardation of the migration of the colloids. A copy of the abstract is appended to this monthly report for NRC approval.

Task III Representation of Geochemical Processes in Models

No activity in August

Task IV Short-term Technical Assistance

No activity during August.

Travel During August 1984

Malcolm Siegel met with S. Whatley, G. Jacobs, D. Kelmers, and other scientists at Oak Ridge National Laboratories (ORNL) on August 29. The purpose of the meeting was to acquaint ORNL staff with objectives of FIN A-1756 and SNLA staff with the ORNL projects B-0290 and B-0287. A detailed description of the discussions will appear in a forthcoming trip report.

Allocation of Resources During August

Subtask 1A, 1B - 45%
Subtask 1D - 43%
Subtask IIB - 12%

A-1756
1646.010
August, 1984

THIS IS AN ESTIMATE ONLY AND MAY NOT MATCH THE INVOICES SENT TO NRC BY SANDIA'S ACCOUNTING DEPARTMENT.

	Month	Current Year-to-Date
I. Direct Manpower (man-months of charged effort)	2.6	9.3
II. Direct Loaded Labor Costs	28.0	89.0
Materials and Services	1.0	4.0
ADP Support (computer)	0.0	3.0
Subcontracts	2.0	11.0
Travel	0.0	4.0
Other	1.0	0.0
TOTAL COSTS	32.0	111.0

Other = rounding approximation by computer

III. Funding Status

Prior FY Carryover	FY84 Projected Funding Level	FY84 Funds Received to Date	FY84 Funding Balance Needed
NONE	200K	200K	NONE

ATTACHMENT I

AGENDA

Technical Advisory Committee for Thermochemical Database for High Waste Disposal

Oak Ridge National Laboratory
at Bldg. 4500-N. Chem. Tech. Conference Room

October 1, 1984

8:45 AM	Welcome and Overview of Charter of Committee	S. Phillips/LBL M. Siegel/SNLA
9:30 AM	Status of Current Database for HLW Disposal	S. Phillips/LBL
10:45 AM	Discussion of Generation of Thermochemical Data	Committee
12:00 PM	LUNCH	
1:30 PM	Workplan for Database Development at Lawrence Berkeley Laboratory for FY 85 - 89	S. Phillips/LBL
2:00 PM	Workplan for Database Development at Stanford University	V. Tripathi/SU
2:30 PM	Overview of Input Requirements for Geochemical Codes	To be announced
3:00 PM	BREAK	
3:15 PM	Discussion on Criteria for Selection of Data and Codes for NRC HLW Geochemical Studies	Committee
4:15 PM	General Discussion	
5:30 PM	ADJOURN	

ATTACHMENT II

INVITEES

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*Possible committee members

+Observers

TRANSPORT AND CAPTURE OF COLLOIDAL PARTICLES IN SINGLE FRACTURES*

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ABSTRACT

This paper describes a study of the transport and capture of colloidal particles in parallel-plate channels simulating single fractures, which represents a "worst" case with respect to the rate and concentration of colloids released from the system. The higher flow rates and porosity associated with the fractures compared to the porous matrix increase particle migration rate and decrease their capture. With this geometry an upper bound for the particle concentration leaving the system and a lower bound for their retardation are established. Particle retardation is determined from the average velocity of the particle front relative to that of the hydrodynamic front. The effects of inlet particle concentration conditions (different time-dependent inlet conditions are used to simulate different particle production mechanisms), particle size, fluid velocity and surface charges for the particles and the channel walls on particle capture and migration are elucidated.

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