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

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Joan-Jicket _____

Dear Mr. Kelly:

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

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

Sincerely,

Malcolm Siegel

Malcolm D. Siegel
Waste Management Systems
Division 6431

MDS:6431:jm

Enclosure

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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

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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 SEPTEMBER 1984

Task I Uncertainty in Integrated Radionuclide Discharge

Subtask 1A. Speciation Effects

The first meeting of the technical advisory committee for the thermochemical data base is set for October 1, 1984 at Oak Ridge National Laboratory. A revised agenda and a list of invitees are appended as attachments to this report. At this meeting, the committee members will be introduced to the objectives of FIN A-1756, the current data base and program plan will be reviewed, the charter of the committee and roles of the committee members will be defined, and a schedule for future meetings will be set.

Subtask 1B. Equilibrium Sorption Effects

Investigation of the capabilities of System 2000 and statistical software at Sandia was continued during September.

-Subtask IC. Kinetic Effects

Documentation of previous work was continued during September for preparation of the FY 84 progress report.

Subtask ID. Dynamic Effects

In NRC's reply to the July monthly report, several questions were raised concerning the equivalent porous medium approach described in Appendix C of NUREG/CR-3235 vol. 3. Many of these concerns can be addressed by considering the use of the porous medium approximation retardation factor with the codes LHS and NWFT/DVM. The specific issues raised by NRC are addressed below:

- (1) 1-D transport in joints and the matrix is assumed.

In NWFT/DVM, transport in the joints is assumed to be 1-D along the flow path, therefore, the approach is actually quasi-multi-dimensional. If the flow path is known or if a conservative flow path is assumed, this approach is an acceptable way to model the transport. As for transport in the matrix, it has been shown that for the purpose of modeling containment transport, the only significant flow in the matrix is perpendicular to the fracture and that flow parallel to the joints within the matrix is negligible [ref. 1].

- (2) A very large characteristic flow length (200 ft) is used.

The 200 ft flow length was used only in the analysis in NUREG/CR-3235. The criterion and error associated with the approximation can be evaluated for shorter lengths under this subtask.

- (3) Longitudinal dispersion in the joints is ignored.

In the equivalent porous medium approximation described in Appendix C of NUREG/CR-3235, vol. 3, an effective retardation factor was derived for the effect of matrix diffusion on transport in the bulk rock. With this retardation factor, a porous medium approach is then used to calculate flow and radionuclide discharge. Although longitudinal dispersion is ignored in the derivation of this effective retardation factor, the dispersion is taken into account in the calculation of fluid flow and radionuclide transport in the rock when NWFT/DVM is used.

- (4) Variability in fracture and matrix characteristics.

The variability in fracture and matrix properties are accounted for by assigning a range and a distribution to each input variable. The LHS sampling technique (Latin Hypercube Sample) is used to select values of each input variable from its range. Radionuclide discharge rates

are then calculated for each combination of selected input values. With this approach, the uncertainty and variability of input are reflected as a range of output (discharge) values.

(5) Only Saturated flow is considered.

The applicability of the equivalent porous medium approximation to unsaturated flow has not yet been considered in this project. Therefore, no response can be given at this point.

The derivation in Appendix C of NUREG/CR-3235 vol. 3 produced 1) a retardation factor for fractured porous media and 2) an expression for the numerical criterion which can identify conditions under which the porous medium approximation is valid. Under Subtask 1D of A-1756, we will assess the error associated with the porous medium approximation under different flow and chemical conditions. In addition, we will investigate other approximations that can be used to model transport for conditions which do not satisfy the criterion derived in NUREG/CR-3235.

[ref]: Nuttall, H. E. and Ray, A. K., "A combined Fracture/Porous Media Model for Contaminant Transport of Radioactive Ions," Scientific Basis for Nuclear Waste Management, vol. 3, Ed. John Moore, Plenum Press, N.Y., p. 577, 1981.

Task II Evaluation of Error Due to Organics and Colloids

Subtask IIA Organics

No activity during September

Subtask IIB Colloids

Two activities were completed during September, 1984 related to radioactive colloid modeling efforts. J. Catasca, a UNM student who was supported by the Associated Western Universities summer program, did a literature search for experimental data on nucleation and growth rate data for colloidal particles. His findings yielded nucleation and growth rate data as well as precipitate concentration-time and population-size-time data that might be used to estimate an upper bound for modeling the formation of radioactive colloidal particles in high-level waste repositories. A draft of a paper by E. J. Bonano and W. E. Beyeler on capture and transport of colloidal particles in single fractures was prepared. A copy of this paper which will be presented at the upcoming MRS meeting in November is attached (Attachment 3). The most significant results are: 1) the most critical factors affecting capture of the particles are the particle production rate and particle size and 2) the average velocity of the particle front for the conditions considered can be as much as 25% higher than the average fluid velocity.

In the NRC response to the July monthly report, several comments were made concerning an earlier letter report dealing with colloids (Bonano and Siegel, April, 1984). These comments are addressed below:

Comment 1.

There is ample description in the literature of the phenomena affecting colloid transport and capture. Not all of these phenomena have been explicitly monitored in experimental studies with radioactive colloids. However, those phenomena not explicitly accounted for in the experiments were still present implicitly, most likely, in the form of a lumped parameter. The objective of the proposed exercise is to determine whether the phenomena explicitly accounted for provide enough information to enable a model to adequately predict the experimental data.

Comment 2.

The models of Tang et al. (1981) and Sudicky and Frind (1982) can not be used to describe the transport of colloidal particles because they only consider dissolved species. However, theoretically, they can be coupled to the transport equations for colloids in order to monitor 1) the transfer of mass from the dissolved phase to the colloids and 2) the total release of radioactive matter as dissolved species and colloidal particles. The coupling arises through nucleation terms that are sinks in the transport equation for dissolved species and sources in the colloid conservation equation.

Task IV Short-term Technical Assistance

A draft document and a letter report dealing with geochemical data for candidate salt sites were received and are under review. A completion date of October 15, 1984 and an expenditure of \$1.5K were set for this task.

Allocation of Resources During July*

- Task I - 35%
- Task II - 50%
- Task IV - 15%

*Amounts are very approximate and should be used only for qualitative comparisons.

Anticipated Problems

Computer costs at Sandia have risen approximately 400% since the beginning of FY84. An additional \$60K may be required over the \$20K originally budgeted for computing costs (ADP) in FY85. The exact potential cost overrun in FY85 cannot be predicted at this time. We will keep the NRC informed on this subject.

AGENDA

**Technical Advisory Committee For Thermochemical
Database For High Level Waste Disposal**

**Oak Ridge National Laboratories
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 Laboratories for FY 85 - 87	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	K. Krupka/PNL
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	

Technical Advisory Committee for HLW Database
Preliminary List of Members and Observers

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*observer

Transport and Capture of Colloidal Particles in Single Fractures*

E. J. Bonano and W. E. Beyeler†
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Abstract

In this study, the transport and capture of colloidal particles were analyzed in a parallel-plate channel simulating a single fracture. The steady-state convective diffusion equation was solved with the particle velocity normal to the walls of the channel being the sum of the external forces acting on the particles. The forces considered were the gravitational, London-van der Waals and electric-double layer forces. The effects of parameters governing these forces and particle production mechanism on the rates of particle capture and transport are determined. The dynamic balance between particle production and capture has a significant effect on the concentration of particles leaving the fracture. The average particle velocity, though higher than the average fluid velocity seems to be insensitive to phenomena governing particle capture.

Introduction

Existing models for the transport of radioactive contaminants in geologic porous and/or fractured media only consider the migration of dissolved species. However, in recent years much attention has been given to the possibility of releasing radioactivity from nuclear waste repositories to accessible environments as colloidal particles[4]. Radioactive colloidal particles in a high-level waste repository can appear via three mechanisms[2]: (1) precipitation of dissolved radionuclides, (2) adsorption of dissolved radionuclides on the surface of foreign colloids suspended in the ground water, and (3) disintegration of the waste/waste package. To date, most studies concerned with radioactive colloids have been experimental[4]. The few theoretical analyses reported in the literature have invoked many simplifying assumptions that render them inadequate to establish the fate of radioactive colloids once these have formed[4]. These studies usually combined all phenomena affecting the transport of colloids in closed conduits in a lumped parameter. Thus, elucidation of the relative importance of these phenomena was not possible. In the present study, we treated all these phenomena as separate terms facilitating the assessment of their importance.

The fundamental principles in the conservation equation for the transport of colloidal particles is very similar to those for dissolved species[1,4]. Attenuation of the migration of both colloids and dissolved species is governed by physicochemical interactions with the walls

*This work supported by the United States Regulatory Commission and performed at Sandia National Laboratories which is operated for the U.S. Department of Energy under Contract Number DE-AC04-76P00789.

†OAO Corporation

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bounding the flow. The difference between colloids and dissolved species arises because of the different nature of these interactions. Interactions between dissolved species and a rock can be either chemical and/or physical (i.e., ion exchange reactions, diffusion), while interactions between colloids and solid surfaces are, generally, physical (electric double layer, London-van der Waals). The transport of colloids is also affected by hydrodynamic interactions between the particles, the flow, and the walls of the system[5]. These hydrodynamic interactions do not exist for dissolved species.

In this study, we solved the conservation equation for colloid transport in a parallel-plate channel simulating a single fracture. The objective was to elucidate the effects of production mechanisms and capture phenomena on the rate of colloid transport. Although the single-fracture geometry is ideal, nevertheless, it can be used to identify those phenomena significant to colloid transport. Furthermore, conclusions reached may be extended to porous media because the fundamental principles involved are identical for both types of geometries.

Model

We analyzed the transport of colloidal particles in the parallel-plates channel shown in Figure 1. We assumed that the laminar Poiseuille flow was not altered by the presence of a few small colloidal particles such that the fluid velocity profile was

$$u = 1.5z (2-z) \tag{1}$$

where u is the dimensionless fluid velocity normalized with respect to the mean velocity U and z is the distance from the bottom wall normalized with respect to the fracture halfwidth b . Thus, the steady-state two-dimensional conservation equation for the dimensionless number concentration, $n (= n^1/n_\infty)$, of colloids of radius a is

$$v_x \frac{\partial n}{\partial x} + \frac{\partial}{\partial z} (v_z n) - \frac{\partial^2 n}{\partial z^2} - f(x) = 0 \tag{2}$$

where n^1 is the actual concentration, n_∞ the maximum concentration, $x = x'/bPe$, Pe the Peclet number ($= bU/D$) and D the particle Brownian diffusivity. The dimensionless particle velocities, V_x and V_z are discussed below.

Equation (2) was obtained after assuming that (1) the channel was infinitely long in the y direction, and (2) the particle Peclet number was large[7]. The coordinate z in Equation (2) represents the vertical position of the center of the particle, however, in this analysis, it was more convenient to transform this variable to the dimensionless separation, h, of the particle from the bottom wall. Thus, we let $z = (h+1)a/b$ and Equation (2) became

$$v_x \frac{\partial n}{\partial x} + \left(\frac{b}{a}\right) \frac{\partial}{\partial h} (v_h n) - \left(\frac{b^2}{a^2}\right) \frac{\partial^2 n}{\partial h^2} - f(x) = 0 \quad (3)$$

The dimensionless particle velocity in the direction of flow was given by

$$v_x = 3(h+1)(a/b)F_2(h) - 1.5 (h+1)^2(a/b)^2 F_1(h) + 2a^2g(\rho_p - \rho)F_3(h)\sin\theta/9\mu U \quad (4)$$

where the first two terms accounted for the drag the fluid exerts on the particles and the last for the gravitational effect; μ being the fluid viscosity, g the gravitational acceleration constant, ρ_p the particle density, ρ the fluid density, and $F_1(h)$, $F_2(h)$, and $F_3(h)$ hydrodynamic resistance functions[1,5,6,8]. The component of the particle velocity normal to the walls was

$$v_h = F_{ext}b/6\pi a\mu Df_h(h) \quad (5)$$

where F_{ext} is the sum of external forces acting on the particle normal to the direction of flow. The function $f_h(h)$ was another hydrodynamic correction function accounting for lubrication effects as the particle approached the walls[1,5,6,8].

Three external forces were assumed to act on the particles normal to the walls: the gravitational, the London-van der Waals and the electric double layer forces. The exact forms of these forces are discussed elsewhere[1,3,6].

Equations (2) and (3) contain the source term $f(x)$ which was used to account for the production of particles in the channel. Three forms of $f(x)$ were used in this analysis:

$$f(x) = \begin{cases} 1 \\ e^{-x} \\ 1 - e^{-x} \end{cases} \quad (6)$$

when $f(x) = 1$, the production of particles is constant and uniform. The exponentially decaying and increasing forms were used to simulate production mechanisms that can vary within the channel such as precipitation of dissolved radionuclides and adsorption of dissolved

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species onto the surface of other colloids suspended in the groundwater. At present, there is no physical justification for these or any other expressions for the source term. Nevertheless, the expression chosen should establish the importance of the source term.

Equations (4) through (6) were substituted into (3) to give the final form of the conservation equation for transport of colloids to be solved subjected to the following inlet, and boundary conditions:

$$\begin{aligned} n(0, h) &= 0 \\ n(x, \delta) &= n(x, h_U) = 0 \end{aligned} \quad (7a, b)$$

The inlet condition ($x=0$) states that no particles come into the system. The boundary conditions in h reflect the effects that the surface interaction forces (London-van der Waals and electric double layer forces) have on the capture of particles. At a small distance from either wall, δ , the total potential of the surface interaction forces becomes infinitely negative, meaning that the potential is attractive. Particles at a separation δ from either wall are attached to the latter and removed from the suspension. A particle a distance δ from the upper wall has position $h = h_U = 2a/b - 2 - \delta$.

Under most circumstances of interest the gravitational force is negligible relative to the London-van der Waals and electric double layer forces due to the small size of the particles. Thus, it is reasonable to solve Equation (3) for only half of the fracture and substitute one of the boundary conditions in Equation (7b) with $\partial n / \partial h = 0$ at $h = b/a - 1$.

Equation (3) in its final form was highly nonlinear and did not allow an analytical solution. Therefore, it was solved numerically. The numerical scheme used is described in [3]. The solution of Equation (3) yielded concentrations as a function of x and h . However, for examining the effects of different parameters, it was more convenient to present the results in terms of the Sherwood number, Sh , and the average velocity of the particle front, \bar{U}_p .

The Sherwood number is a measure of the rate of particle capture and, for the lower wall, is given by

$$Sh(x) = \frac{1}{f_h(h)} \left. \frac{\partial n}{\partial h} \right|_{h=\delta} \quad (8)$$

The average velocity of the particle front is:

$$\bar{U}_p(x) = \frac{\int_{\delta}^{b/a-1} n(x, h) v_x(h) dh}{\int_{\delta}^{b/a-1} n(x, h) dh} \quad (9)$$

Since the dimensionless average fluid velocity is 1, then \bar{U}_p is also the dimensionless relative average particle velocity.

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Results and Discussion

We chose as the base case the following parameters: $a = 0.1 \mu\text{m}$, $U = 1 \text{ cm/s}$, $b = 15 \mu\text{m}$, $\delta = 0.01$, $\rho_p = 1.4 \text{ g/cm}^3$, $\rho = 1.0 \text{ g/cm}^3$, $\theta = 0^\circ$, $f(x) = e^{-x}$, and $\psi_1 = -15 \text{ mvolts}$, $\psi_2 = -25 \text{ mvolts}$, $\kappa = 1 \times 10^7 \text{ cm}^{-1}$. The particle surface charge (ψ_1), the channel surface charge (ψ_2) and the reciprocal thickness of the double layer (κ) are required in the electric double layer force. The values of some parameters are not necessarily representative of high-level waste repositories, however, they are typical of model systems for which similar calculations to the one of this study have been performed and are adequate for establishing the effects of the different phenomena governing particle capture and transport[1,6].

Figure 2 is a plot of concentration profiles for the base case at $x = 2$ and 4 . Near the inlet of the channel the concentrations increase sharply due to the source term. As the suspension moves downstream in the channel less particles are being produced ($f(x) = e^{-x}$) and, consequently, the concentration decreases due to capture. For $x \geq 5$, the concentration is negligible. In this case the electric double layer force is repulsive ($\psi = -15 \text{ mv}$, $\psi_2 = -25 \text{ mv}$). Nevertheless, because the double layer is collapsed ($\kappa=1 \times 10^7 \text{ cm}^{-1}$), capture is dominated by the attractive London-van der Waals force.

In Figure 3, the Sherwood number is plotted as a function of the dimensionless position along the fracture, x , for different source terms, $f(x) = 1$, e^{-x} and $1-e^{-x}$. The curves in this figure indicate that the rate of capture (Sherwood number) is a function of the concentration of particles in suspension. For $f(x) = 1$, a constant and uniform source of particles, capture was independent of x , hence, a constant value of $Sh = 0.080627$ was obtained. In this case, production and capture balance each other. For $f(x) = e^{-x}$ (base case), near the inlet of the channel where production is highest, the rate of capture is also the highest and decreases with x in the same fashion as $f(x)$. In real situations if particle production is a decreasing function of x , then the system need not be long for the particles to be captured. On the other hand, when production increases with x , then most capture should take place near the outlet of the system. For $f(x) = 1-e^{-x}$, near the inlet, capture (Sherwood number) is low, and as production approaches 1 asymptotically, the Sherwood number approaches 0.080627.

Another parameter that has a significant effect on capture is the ratio of the particle size to the system's cross section, a/b . Here we examined this effect by keeping b constant and varying a . In Figure 4, plots of Sherwood number versus x for $a = 0.1 \mu\text{m}$ (base case) and $a = 0.05 \mu\text{m}$ are presented. The effect of the relative particle size shown were expected [1,6]; the larger the particle, the higher the rate of capture. In our case, for a particle 50 percent smaller, the rate of capture can be at least an order of magnitude smaller.

In addition to the source term and particle size effects, we also examined the effects of average flow velocity on capture. In Figure 5 we present the Sherwood number versus x curves for $U = 1 \text{ cm/s}$ (base case), 0.5 cm/s , and 2.0 cm/s . The three curves are identical suggesting that fluid velocity has no significant effect on capture. In [6], average flow velocity was shown to have a slight effect on capture efficiency. However, in that study the system was a fibrous porous medium with a more complex flow field. The effect of average flow velocity on capture in [1] is not readily discernible because in that study this effect was lumped in a parameter that also included particle size effects. Thus, the average fluid velocity is a parameter the effects of which deserve more careful consideration in future studies.

Due to numerical complications in solving Equation (3), the effects of the particle and channel surface charges and the reciprocal double layer thickness could not be determined. Similar complications have been encountered in a previous study[1]. However, when solving the particle trajectory equation in porous media, these effects have been examined[6]. Results obtained under these circumstances should be expected to apply, at least qualitatively, to the system considered here. It was found in [6] that for collapsed double layers (large κ), changes in the surface charges (Ψ_1 and/or Ψ_2) do not affect capture. When the double layer is expanded (small κ), capture is very sensitive to the values and sign of Ψ_1 and Ψ_2 .

Finally, we examined the effect of the different phenomena on the rate of transport of the particles. We calculated the dimensionless relative particle average velocity with Equation (9). We found that \bar{U}_p was 1.2555 for all x 's and all values of the parameters. First, \bar{U}_p is larger than 1 because as shown in Figure 2, the majority of the particles travel near the center of the channel where the fluid velocity is highest. The higher number of particles at the center of the channel than near the walls have a larger contribution in the average velocity of the particle front. A constant value of \bar{U}_p was a surprising result. A careful observation of the concentration profiles in Figure 2 reveals the reason for a constant \bar{U}_p . The percentage change of the concentration for all values of h was constant as the suspension traveled from $x = 2$ to $x = 4$. Since the particle velocity $v_x(h)$ is independent of x , then the ratio of the two integrals in Equation (9) becomes constant.

In conclusion, the analysis of particle capture and transport in parallel-plate channels indicates that there is a direct relationship between the rate of particle capture and particle production, and that particle size is significant. These results suggest the need for careful experimental studies aimed at examining particle production under conditions typical of high-level waste repositories. Also, it was shown that since particles tend to travel in regions of higher fluid velocities, their average rate of transport may be higher than that of dissolved species.

References

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5. Goldman, A. J., Cox, R. G., and Brenner, H., "Slow Viscous Motion of a Sphere Parallel to a Plane Wall-I: Motion through Quiescent Fluid," Chem. Eng. Sci., 22, 637-651 (1967).
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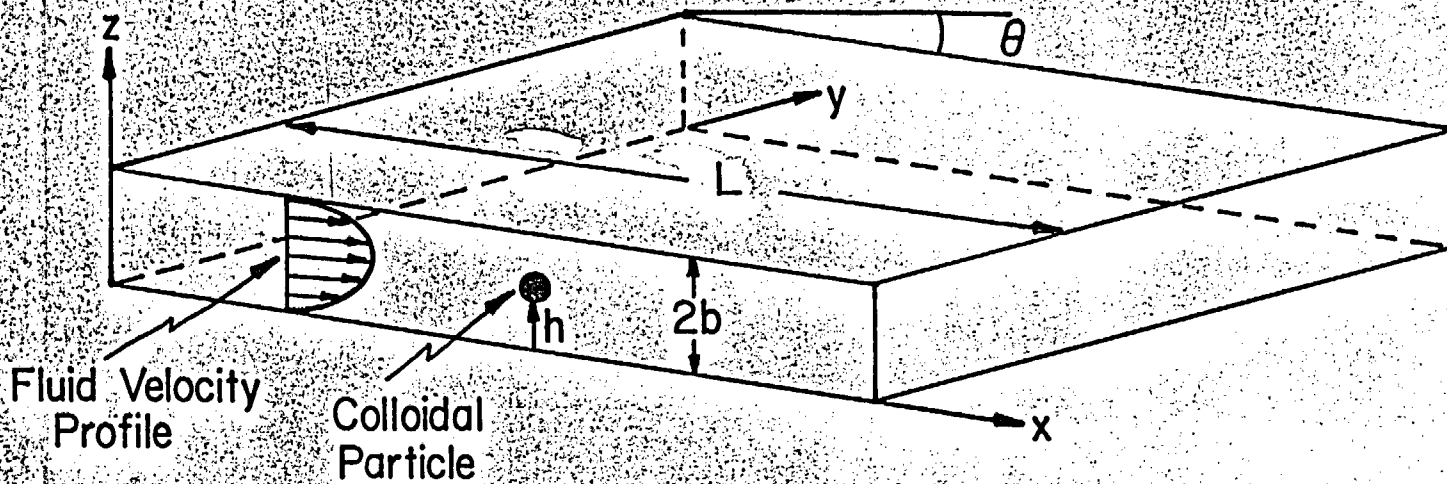
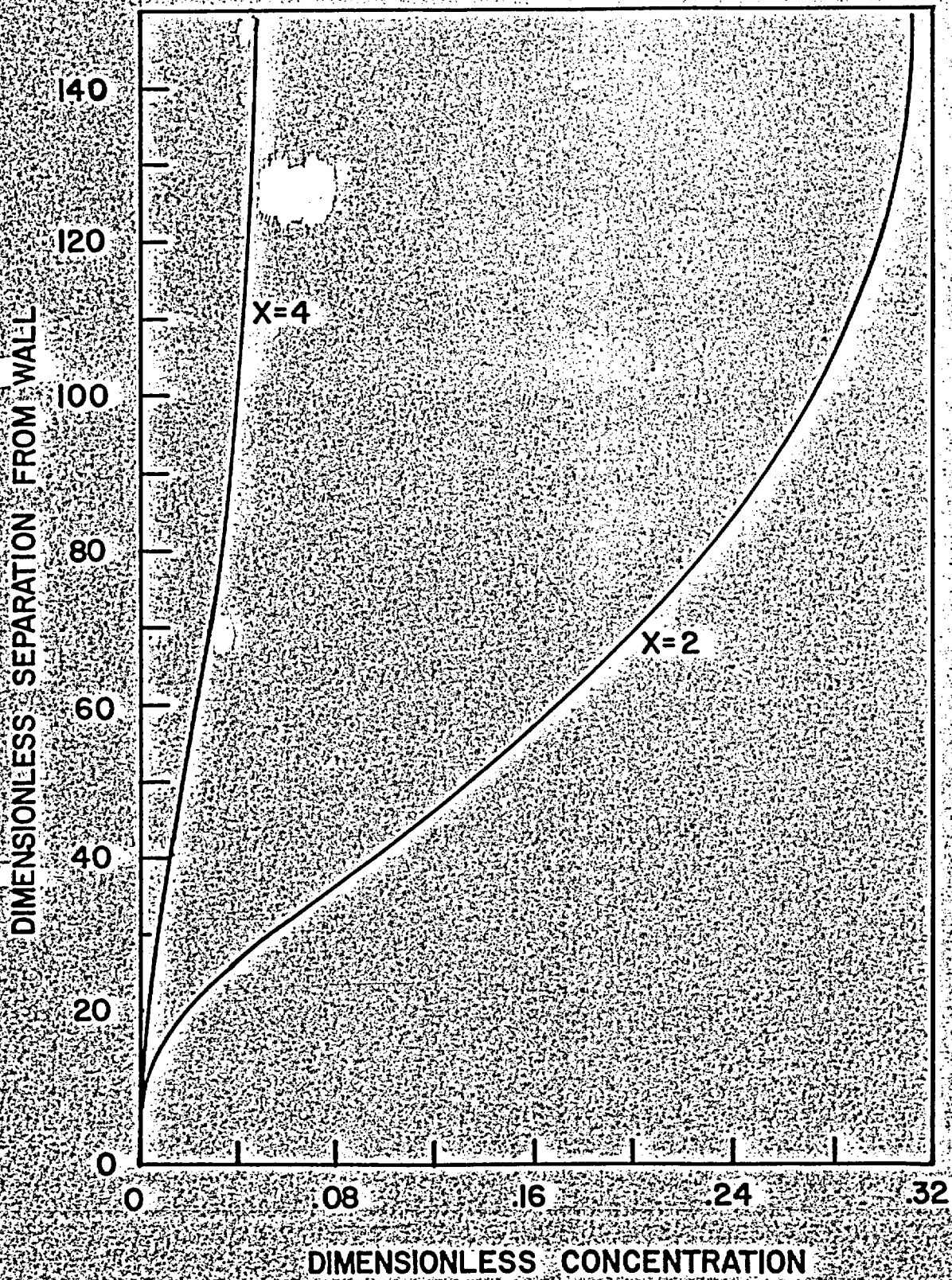
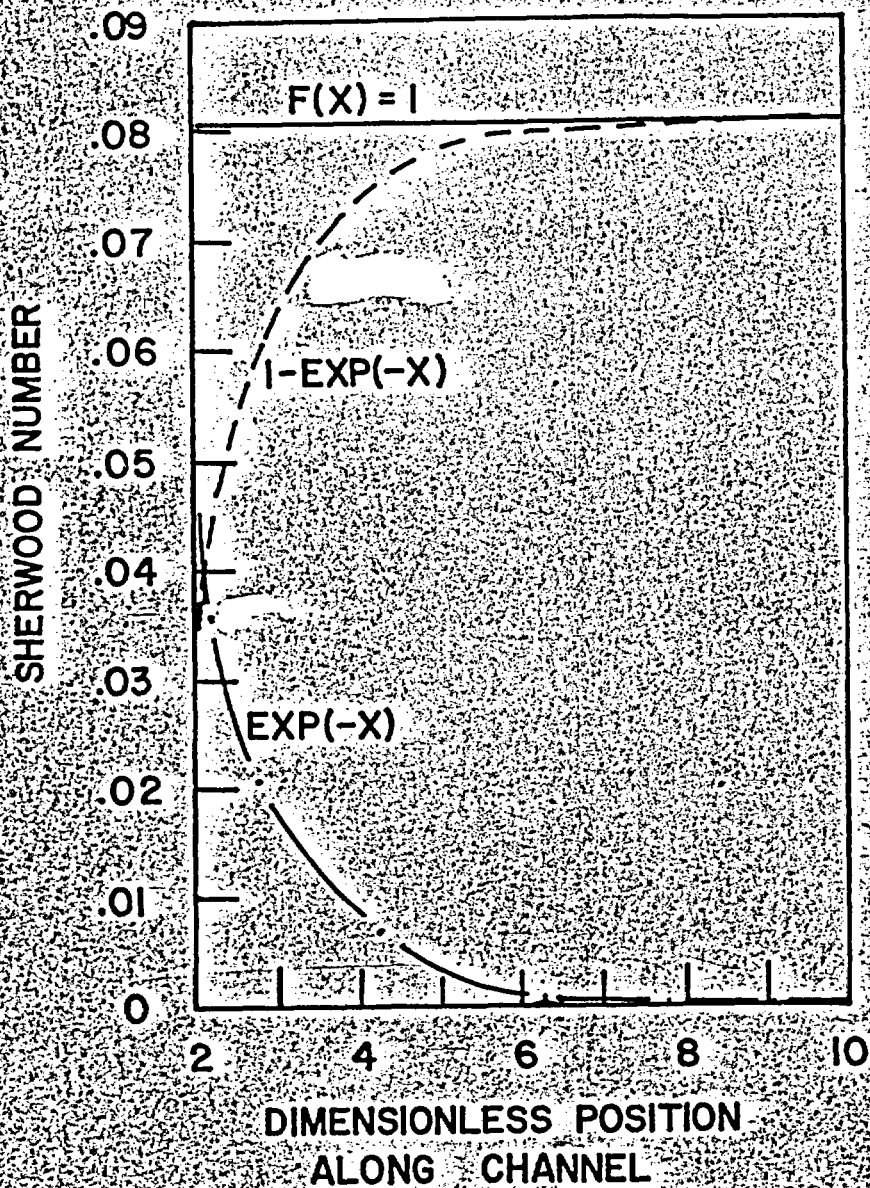


Fig. 1. System Considered.

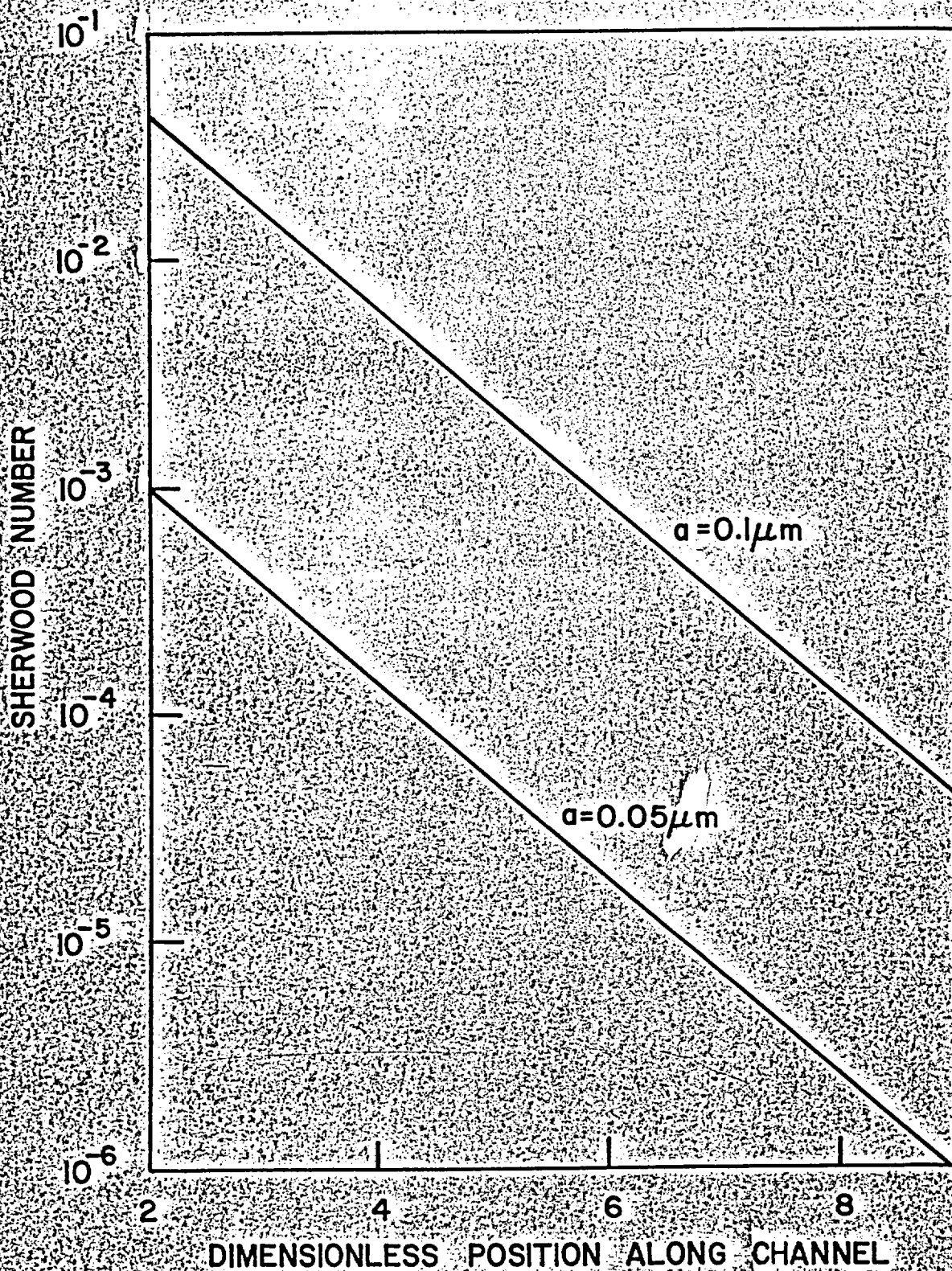
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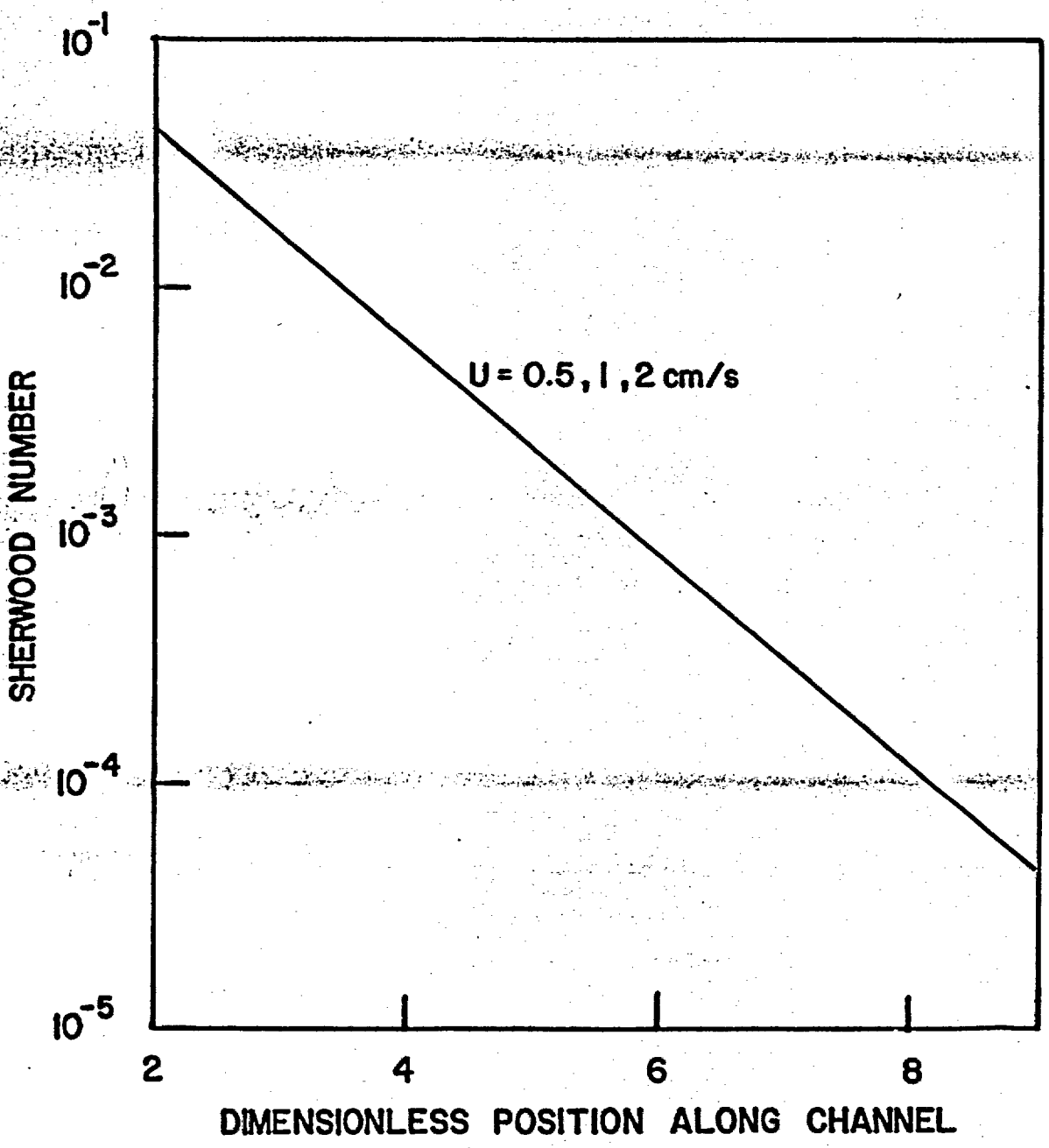
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October 10, 1984

Due to the end of our fiscal year, Sandia's Accounting Department will not make budget figures available to our organization until after October 16. We will forward budget sheets to you as soon as possible after that date.