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MATHEMATICAL MODELING AND EVALUATION
OF RADIONUCLIDE TRANSPORT PARAMETERS
FROM THE ANL LABORATORY ANALOG PROGRAM

by

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ABSTRACT

Computer model simulation is required to evaluate the performance of proposed or future high-level radioactive waste geological repositories. However, the accuracy of a model in predicting the real situation depends on how well the values of the transport properties are prescribed as input parameters. Knowledge of transport parameters is therefore essential.

We have modeled ANL's Experiment Analog Program which was designed to simulate long-term radwaste migration process by groundwater flowing through a high-level radioactive waste repository. Using this model and experimental measurements, we have evaluated neptunium (actinide) deposition velocity and analyzed the complex phenomena of simultaneous deposition, erosion, and reentrainment of bentonite when groundwater is flowing through a narrow crack in a basalt rock. The present modeling demonstrates that we can obtain the values of transport parameters, as added information without any additional cost, from the available measurements of laboratory analog experiments.

1. INTRODUCTION

1.1 MODELING

The U.S. Nuclear Regulatory Commission (NRC) is responsible for evaluating proposed geological repositories for the containment of high-level radioactive wastes (HLW). Because the problem is complex, computer model simulation is a major part of the evaluation process. Accurate assessment of a proposed repository requires that the mathematical models used in the analysis account for all physical processes, and the inputs of transport and other related parameters that describe the processes are reliable.

All modeling related to repositories can be classified into two broad categories:

- Far-field modeling: Large-scale modeling where interest is mainly in the far-away effects from a repository.
- Near-field modeling: Detailed modeling of all regions in the close vicinity of a repository.

Far-field modeling is easier because the number of processes to be modeled are fewer, and simplifying assumptions and approximations can be used without

grossly affecting the results of analysis (e.g., one-dimensional analysis can be performed assuming either a point source or a line source). Consequently, several computer models have been developed that are used extensively for analyzing far-field behavior.

Near-field modeling is more complex and not all phenomena are completely understood.

1.2 COMPUTER MODELING

There are more than 183 computer codes currently in existence that are designed for the analysis of one or more of the following phenomena.

- Saturated flow
- Unsaturated flow
- Surface water flow
- Solute transport
- Heat transport
- Combined solute and heat transport
- Geochemical processes
- Geophysical processes

Some of the codes are applicable to porous media, others apply to fractured media, and some are limited to analyzing only far-field phenomena, with others for near-field phenomena.

All the computer codes solve a set of one or more differential equations together with a number of submodels that describe various transport phenomena. To obtain a solution of the differential equations, all computer codes require values of transport and related parameters as inputs. Therefore, how well a computer model predicts the real situation depends on how well we prescribe the values of the input parameters.

1.3 TRANSPORT PARAMETERS

The magnitude of transport parameters (input needed in computer modeling) and their effects on the behavior of a repository are not fully known. Accordingly, grossly estimated or guessed values are commonly prescribed as input in computer modeling. As an example, the following is a list of some of the transport parameters required as input by the SWIFT computer program:

Hydraulic conductivity. The property of a structure (host rock) that determines the amount of fluid that will flow through under a given pressure drop. The magnitude largely depends on the porosity of a structure, and varies continuously with the deposition and swelling of deposits.

Solubility. Depends on temperature, pH, and other suspended material in a fluid.

Thermal diffusivity. Not generally known. Its magnitude changes with humidity, cracks, geometrical configuration, etc.

Nuclide deposition-adsorption. The rates of nuclide deposition and adsorption depend largely on the mechanism (process), e.g., whether nuclide is

first adsorbed by backfill (bentonite), which then deposits on a host rock surface, or adheres directly on the surfaces of a host rock.

Diffusivity. Generally known for simple component and dilute solutions, but not well understood and not known for mixtures of varying concentration of different components.

Without proper knowledge of the transport parameters, such as those described above, we cannot use a computer code to properly estimate the radwaste migration from a repository through a groundwater system. The error in computing concentrations of radioactive spectra at a given location can be as high as several orders of magnitude.

1.4 APPROACH

There are two approaches to determining and evaluating the magnitude of transport and related parameters:

- Develop and perform in-situ field measurements and/or laboratory experiments specifically to determine the magnitude of desired parameters.
- Obtain indirectly from other experimental measurements designed to measure integrated effect (e.g. modeling and evaluation of transport parameters as presented here).

The procedure of obtaining transport parameters in the second approach is as follows:

- Model the experiment.
- Determine fundamental transport phenomena.
- Derive the basic transport equations.
- Obtain the solution (approximate or numerical) of transport equations.
- Integrate the solution.
- Compare the integrated solution with experimental measurements.
- Evaluate the magnitude of the transport parameters from the comparison of the integrated effects.

The procedure outlined above leads further to better understanding of transport phenomena.

The major benefits of the second approach are the following:

- The existing experimental facility and data are used so that there is no additional experimentation cost.

- Information related to transport parameters is obtained as additional basic information, over and above the design goals of the experiments.
- More thorough understanding of fundamental transport processes (e.g., repetitive erosions and depositions) is reached.
- Important nonlinear interactions are determined that generally do not show up in simpler experiments.
- The approach leads to further guidance in the design of future experiments.

1.5 PRESENT WORK

In the present work, we have modeled ANL's Experimental Laboratory Analog Program, which is designed to simulate long-term radwaste migration process by ground water flowing through a high level radioactive waste repository. The experimental layout of ANL's Laboratory Analog Program is described in Sec. 2. The modeling of neptunium (actinide) deposition phenomenon and the evaluation of deposition velocity are discussed in Sec. 3. In Sec. 4, we present the modeling of complex phenomena of simultaneous deposition, erosion, and reentrainment of bentonite when groundwater is flowing through a narrow crack in a basalt rock. The governing equations relating to transport of actinides and backfill material are presented in Sec. 5. The modeling of the resaturation process is described in Sec. 6.

2. LABORATORY ANALOG PROGRAM AT ANL*

2.1 LAYOUT OF THE EXPERIMENT

The experimental layout of the Laboratory Analog Program at ANL is shown in Fig. 2.1. In the experiment, synthetic groundwater is pumped through basalt chips and backfill (bentonite), contacts solid nuclear waste, passes through more backfill, then traverses the fissure in the split rock (basalt) core and exits the apparatus. Groundwater solutions exiting the apparatus, as well as samples tapped at intermediate points within the apparatus, are obtained during the experiment. The experiment is preceded by a conditioning period in which the water flows through the backfill and rock before the waste form is added. The water is pumped at a low rate (~ 0.5 mL/hr) that produces a linear velocity of ~ 32 m/year through the bentonite and over the waste form. After the experiment has run for several months, the flow of groundwater is stopped and the repository materials (backfill, waste glass, and rock core) are removed for detailed analyses of physical and chemical changes.

* The Laboratory Analog Program was sponsored by the U.S. Nuclear Regulatory Commission under contract A-2230. One of the authors (Dr. Martin Seitz, Principal Investigator of the Laboratory Analog Program) provided the descriptions and all experimental data.

Fig. 2.1. Schematic of Apparatus for Analog Experiment. Groundwater passes through the basalt core after traversing backfill (bentonite) that brackets the radioactive waste glass. Solutions exiting the oven are collected in sealed containers.

The solid nuclear waste used in these experiments is composed of borosilicate glass frit (PNL 76-68) to which has been added 3% uranium as oxide, and the seven radionuclides ^{85}Sr , ^{133}Ba , ^{141}Ce , ^{152}Eu , ^{237}Np , ^{239}Pu , and ^{241}Am . A wafer of the waste solid ~0.40 g with a surface area of 2.6 cm² is used for each experiment.

Table 2.1 is a list of experiments that were performed in this program. In two of the experiments (Nos. 4 and 5), the backfill and rock samples were preconditioned to simulate 1000/2000-year aging due to radwaste heat and groundwater pressure. To determine the effect of groundwater on the canisters, two different materials (Hastelloy or Monel-400) were used for the casings and in one experiment (No. 3), the complete apparatus was placed in gamma field. In these analog experiments, only the integrated effects of all transport interactions that occur in the components of repository are being measured.

Table 2.1. Laboratory Analog Experiments

Analog Experiment No.	Condition of Waste Form, Backfill, and Rock	Apparatus and Experiment Conditions
1	Unaltered	Hastelloy
2	Unaltered	Monel-400
3	Unaltered	Hastelloy, gamma field
4	Thermo-, hydro- alteration (1000 y)	Hastelloy
5	Thermo-, hydro- alteration (2000 y)	Hastelloy

2.2 TRANSPORT PROCESSES

● Backfill (Bentonite)

In the backfill, erosion and deposition of bentonite occur as groundwater flows through the backfill. The groundwater carries bentonite in the form of suspended particles as it leaves the system.

● Radioactive Waste

Leaching occurs as groundwater comes in contact with radioactive waste. The concentration of dissolved radioactive material in groundwater depends on the rate of leaching on the surfaces of radioactive waste.

- Basalt Rock

As groundwater flows through the fissure of the basalt rock, the radioactive material in the groundwater is deposited on the surfaces of the rock. Simultaneously, the deposition and erosion of suspended bentonite in groundwater occur on the surfaces of basalt rock.

2.3 MEASUREMENTS

In the laboratory analog program, the following measurements are made.

During the Experiment

- Groundwater is tapped at the exit and at several intermediate points, and detailed analyses are performed to determine the physical and chemical changes of the groundwater.
- Pressure is adjusted continuously to maintain constant flow.
- Pressure drop and flowrates are measured.

After the Experiment

- The repository materials are removed for detailed analysis of physical and chemical changes.
- The distribution of radioactive materials retained on the rock surfaces are measured.

Figure 2.2 shows the ^{237}Np distribution on the surfaces of split basalt rock from three analog experiments. Some important parameters of the first three experiments are presented in Table 2.2.

Table 2.2. Summary of Important Parameters of the First Three Experiments

	1	2	3
Day's run	120	150	90
Avg. flow, mL/hr	0.58	0.89	1.0
ΔP , kPa (beginning)	70	70	70
ΔP , kPa (end)	1200	410	1370

Fig. 2.2 Neptunium Distribution on Surface of Split Basalt Cores from Three Analog Experiments

3. DEPOSITION OF NEPTUNIUM

3.1 MODELING ASSUMPTIONS

In the analog experiment, the two halves of basalt are separated by a 0.025-cm thick gold gasket to form a basalt fissure (gap). When a water solution of pH = 9.5 flows through this gap, $\text{NpO}_2^+(\text{V})$ in water solution adsorbs on the basalt surface in the form of $\text{NpO}_2^+(\text{IV})$.

Although the surface of either half of the basalt core is irregular, the two surfaces mate together extremely well. We can therefore assume that, to a first approximation, the flow through the basalt is a flow through a rectangular duct. Because the flow velocities are small at the mm/s level, we further assume a fully developed laminar flow through the crevice, in spite of the roughness of the surface and irregular blockages.

3.2 PERMEABILITY OF THE BASALT SPECIMEN

With the assumption of fully developed laminar flow through a rectangular duct, we can calculate the mean flow velocity of groundwater in basalt rock.

$$u_m = \frac{1}{12\mu} w^2 \frac{\Delta P}{L}, \quad (3.1)$$

where w is the mean gap thickness,

μ is the fluid viscosity,

ΔP is the pressure drop, and

L is the mean length of the passage.

The volumetric flow rate $\dot{Q}$ is given by

$$\dot{Q} = bwu_m, \quad (3.2)$$

where b is the width of the gap.

Solving Eqs. 3.1 and 3.2 for the mean gap thickness, we get

$$w^3 = \frac{12\mu\dot{Q}}{\Delta P} \frac{L}{b}. \quad (3.3)$$

From Darcy's law for flow in a porous media, we get

$$u_m = \frac{k_p}{\mu} \frac{\Delta P}{L}. \quad (3.4)$$

Here, k_p is the permeability of basalt rock. Comparison of Eqs. 3.1 and 3.4 gives us the permeability in terms of gap thickness.

$$k_p = \frac{w^2}{12}. \quad (3.5)$$

Knowing the flowrate and pressure drop in the experimental setup, we can calculate the mean gap thickness from Eq. 3.3, and the permeability from Eq. 3.5. The calculated values of w and k_p for three experiments are given in Table 3.1.

Table 3.1. Summary of Analysis on Neptunium Deposition

	1	2	3
Mean permeability, m^2 (beg.)	5.52×10^{-13}	7.17×10^{-13}	7.75×10^{-13}
Mean permeability, m^2 (end)	0.083×10^{-13}	2.21×10^{-13}	1.07×10^{-13}
Mean velocity, mm/s (beg.)	0.92	1.20	1.29
Mean velocity, mm/s (end)	2.37	2.16	3.48
Mean $2v_d/wu_m$, cm^{-1}	0.73	0.77	0.78
Mean v_d , m/s	0.86×10^{-7}	1.36×10^{-7}	1.53×10^{-7}
z_0 , cm	2.4	4.8	4.8
Count at z_0	60	60	32
Maximum count	70	(99 at transverse crack)	

3.3 DEPOSITION OF NEPTUNIUM

For one-dimensional uniform flow through a rectangular duct, we can write the governing conservation equation for neptunium as

$$\frac{\partial}{\partial t} (\rho c) = - \frac{2\rho c v_d}{w} - u_m \frac{\partial}{\partial z} (\rho c) + \frac{\partial}{\partial z} \rho D \frac{\partial c}{\partial z} \quad (3.6)$$

Here, ρ is mixture density,

c is concentration of neptunium,

v_d is deposition velocity (adsorption force of molecules per unit mass times relaxation time),

D is diffusivity, and

z is axial distance from the inlet.

The multiplier 2 in the deposition term is to account for the two surfaces of a channel.

If we assume a steady state and negligible diffusion, Eq. 3.6 simplifies to

$$u_m \frac{\partial c}{\partial z} + \frac{2v_d}{w} c = 0. \quad (3.7)$$

The solution of Eq. 3.7 is

$$c(z) = c_0 \exp[-2 v_d (z - z_0)/w u_m], \quad (3.8)$$

where z_0 is reference axial location and

c_0 is reference count (concentration) at position z_0 .

We can see from Fig. 2.2 that for $z > z_0$, the experiment results are well represented by Eq. 3.8. The reference location z_0 is, of course, slightly different for each experiment.

By matching the experimental observation and Eq. 3.8, we can calculate the parameters c_0 , z_0 , $2v_d/wu_m$, and v_d . The results of these calculations are presented in Table 3.1.

3.4 ANALYSIS

The spatial variation in the amount of deposition results in a variation of flow cross-sectional area. Therefore, we have a larger velocity (smaller flow area) at the inlet and a smaller velocity (larger flow area) at the outlet than the mean velocity (flow area). This variation can easily account for the departure from the distribution predicted by Eq. 3.8 for $z < z_0$, as evidenced by the experiment results. Examination of Eq. 3.8 indicates that a larger ($u_m w$) product would result in a lower count rate (concentration). This is because the product $u_m w$ did not remain constant as suggested by Eq. 3.2. Instead, the patched pattern of deposition produced a variation in the passage width b . Other possible explanations are that the maximum count may be limited by the concentration of $\text{NpO}_2^{+}(\text{V})$ or that different rock samples may have a different number of sites for adsorption.

4. DEPOSITION OF BENTONITE

4.1 DEPOSITION PHENOMENON

Careful examination of inlet pressure vs. time (Fig. 4.1) recorded in the ANL-analog experiments provide a clue for understanding the deposition mechanism of suspended bentonite particles in groundwater flow. At a constant flowrate, the pressure drop fluctuates, but the small-time averaged pressure

Fig. 4.1. Comparison of Approximate Solutions with Experimental Results

drop across the fissure of the rock core increases in magnitude with an increase in time. This phenomenon suggests deposition of bentonite in the passage formed by the crack in the basalt specimen (Fig. 4.2a), thus narrowing the passage flow area. The large fluctuation in pressure drop at constant flowrate is due to the following phenomena, in succession:

The deposition of bentonite particles (Fig. 4.2b) causes narrowing of the passage and an increase in pressure and flow velocity.

These processes in turn lead to:

erosion,

removal of the blockage,

reentrainment of the bentonite (to be deposited downstream), and

reduction of overall pressure drop (Fig. 4.2c).

These phenomena repeat as the amount of deposits increase until erosion and deposition equalize. Thereafter, the processes repeat and pressure drop continues to fluctuate but the average pressure drop no longer increases and no net deposition in the passage occurs over the time period (Fig. 4.2d). The phenomena are sketched conceptually in the ANL-Analog experimental measurements.

4.2 MODELING

The deposition phenomenon described in Sec. 4.1 is analyzed by a one-dimensional transient model.

For a dilute suspension of bentonite particles in water, a constant displacement pump gives a constant volumetric flow rate, Q . Thus,

$$Q = bw u_m = \text{constant}, \quad (4.1)$$

where b is the width, w is the height of the passage, and u_m is the mean flow velocity. Conservation of bentonite mass gives, for a negligible rate of diffusion,

$$\frac{\partial \rho_\beta}{\partial t} + u_m \frac{\partial \rho_\beta}{\partial z} = - \frac{2\rho_\beta v_d}{w} + \frac{E}{w}, \quad (4.2)$$

where t is time, z is distance from the entrance to basalt rock in the axial flow direction, ρ_β is the density of suspended bentonite, v_d is deposition velocity, and E/w is the increase in ρ_β per unit time by erosion and entrainment. The height of the passage changes according to

$$\frac{dw}{dt} = - \frac{2\rho_\beta}{\bar{\rho}_\beta} v_d + \frac{E}{\bar{\rho}_\beta}. \quad (4.3)$$

$\bar{\rho}_\beta$ is the density of bentonite in a deposit layer.

Fig. 4.2. Stage of Deposition, Erosion, Reentrainment and Deposition Downstream of Suspended Bentonite

The mean flow velocity, on the assumption of a fully developed laminar flow in a rectangular duct, is given by

$$u_m = \frac{w^2}{12\mu} \left(- \frac{dp}{dz} \right). \quad (4.4)$$

The erosion rate E (in mass/area-time) is a function of local mean velocity u_m and pressure gradient (dp/dz) to push away and disperse the deposit, and can be determined only experimentally. The primary proportionality relation is for E to be a finite value when a critical velocity u^* is reached. The postulated values of v_d and E can be checked with the overall pressure drop across the passage as it changes with time, or

$$\Delta P(t) = 12\mu \int_0^L \frac{u_m}{w^2} dz = \frac{12\mu Q}{b} \int_0^L \frac{dz}{[w(z,t)]^3}. \quad (4.5)$$

For a given initial passage height $w_0(z)$ and concentration of bentonite at the inlet $\rho_{\beta 0}$, and

$$E = E(u^*), \quad (4.6)$$

(that is, the erosion at a local velocity u^* or a local pressure gradient above a certain value), a lump of the deposit will break off. The average values of v_d and E can be determined from the pressure vs. time recorded from the experiments. Conversely, for known values of v_d and $E(u^*)$ relation, Eqs. 4.1-4.4 give $\rho_{\beta}(z,t)$, $w(z,t)$, $P(z,t)$, and $u_m(z,t)$ for given $\rho_{\beta 0}$ and $w_0(z)$. The general situation is illustrated in Fig. 4.2.

4.3 SIMPLIFIED COMPUTATION

It is desirable to perform a preliminary computation to gain an understanding of the problem and to provide a means of computer code check.

For this purpose, we consider a simplified situation for ANL-Laboratory Analog Experiment 2. The experimental data for this case are summarized as follows:

$$b = 0.0683 \text{ m},$$

$$L = 0.146 \text{ m},$$

$$Q = 0.89 \text{ mL/hr} = 2.472 \times 10^{-10} \text{ m}^3/\text{s}, \text{ and}$$

$$\mu = 2.90 \times 10^{-4} \text{ kg/ms at } 90^\circ\text{C}.$$

From the recorded pressure drop vs. time data we choose the initial time $t_0 = 1750 \text{ hr}$; then $\Delta p = 70 \text{ kPa}$. Equation 4.5 gives the average uniform passage height

$$\langle w^3 \rangle = \frac{12\mu Q}{b} \left(\frac{\Delta P}{L} \right)^{-1}, \quad (4.7)$$

or $w_0 = 2.98 \times 10^{-6}$ m. Equation 4.1 gives $u_0 = 1.215 \times 10^{-3}$ m/s. At the final time of $t = 3000$ hr, average $\Delta P = 410$ kPa, we have $w_f = 1.655 \times 10^{-6}$ m and $u_f = 2.187 \times 10^{-3}$ m/s. The concentration of the suspended bentonite at the inlet is 0.036 kg/m³, assuming $\rho_\beta = 1000$ kg/m³ (due to swelling and packing).

For the simplified computation, we will determine the average change in pressure drop due to gradual deposition, starting from the inlet at the aforementioned t_0 (Fig. 4.3a). We will assume that the erosion and deposition will balance each other when the wide passage is narrowed to 1.655×10^{-6} m (Fig. 4.3d). Because of small flow velocity, we can treat the system as quasistatic. Integration of Eq. 4.2 gives, for z_0 as defined in Fig. 4.3b (i.e., the length of deposit over which no further deposition occurs), the density of bentonite as

$$\begin{aligned} \rho_\beta(z) &= \rho_{\beta 0} \exp \left\{ -\frac{2 v_d (z - z_0) b}{\dot{Q}} \right\}, \quad \text{for } z \geq z_0, \\ \rho_\beta(z) &= \rho_{\beta 0} \quad \text{for } z < z_0, \end{aligned} \quad (4.8)$$

in which we assume $\frac{\partial}{\partial t} = 0$ and $E = 0$. The width of passage changes according to

$$\begin{aligned} w(z) &= w_0 - \frac{2 \rho_\beta(z)}{\rho_\beta} v_d (t - t_0), \quad \text{for } z \geq z_0, \\ w(z) &= w_f, \quad \text{for } z < z_0, \end{aligned} \quad (4.9)$$

by integrating Eq. 4.3 under the same simplification.

Time Δt when the deposited-layer distance z_0 extends by distance Δz is given by

$$\begin{aligned} b(w_0 - w_f) \Delta z &= \frac{2 v_d}{\rho_\beta} b \Delta t \int_{z_0}^{z_0 + \Delta z} \rho_\beta(z) dz \\ &= \Delta t \frac{\rho_{\beta 0}}{\rho_\beta} \dot{Q} \left[1 - \exp \left(-\frac{2 v_d b \Delta z}{\dot{Q}} \right) \right] \end{aligned} \quad (4.10)$$

via averaging ρ_β over Δz , thus determining the time interval Δt for deposition to cover Δz to narrow the passage to w_f .

The pressure drop is given by

$$-dp = \frac{12 \dot{Q} \Delta z}{b w^3}. \quad (4.11)$$

Fig. 4.3. Approximate Solutions

Integrating Eq. 4.11 for the case shown by $t = t_1$ in Fig. 4.3b, we get

$$-\Delta p(t) = \frac{12\mu \dot{Q} z_o}{b w_f^3} + \frac{12\mu \dot{Q}}{b} \int_{z_o}^L \frac{dz}{w^3}. \quad (4.12)$$

We define

$$y = \exp \left[- \frac{2v_d b (z - z_o)}{\dot{Q}} \right], \quad (4.13)$$

therefore,

$$dy = - 2 \frac{v_d}{\dot{Q}} b y dz, \quad (4.14)$$

or

$$dz = - \frac{\dot{Q}}{2v_d b} \frac{dy}{y}. \quad (4.15)$$

We rewrite Eq. 4.9 in the form

$$w = w_o(1 - \alpha y), \quad \text{for } z > z_o, \quad (4.16)$$

where

$$\alpha = 2\rho_\beta(z) v_d(t - t_o) / \bar{\rho}_\beta. \quad (4.17)$$

Substituting Eqs. 4.15 and 4.16 in Eq. 4.12, we get

$$-\Delta p = \frac{12\mu \dot{Q} z_o}{b w_f^3} + \frac{6\mu}{v_d} \frac{\dot{Q}^2}{b} \frac{1}{w_o^3} \int_{y_L}^1 \frac{y^{-1} dy}{(1 - \alpha y)^3}, \quad (4.18)$$

where the limits of integration have changed because

$$\text{at } z = z_o, \quad y_o = 1, \quad (4.19)$$

and

$$\text{at } z = L, \quad y_L = \exp \left[- \frac{2 v_d b (L - z_o)}{\dot{Q}} \right]. \quad (4.20)$$

Although α is a function of z , we approximate it to have the same value as at $z = z_o$. Thus,

$$\alpha = \frac{2 \rho_{\beta o} v_d \Delta t}{\bar{\rho}_\beta w_o} = 1 - \frac{w_f}{w_o}. \quad (4.21)$$

The integral in Eq. 4.18 is given by

$$I(\alpha, y) = -\text{Log}_e(1 - \alpha) + \frac{2\alpha}{1 - \alpha} + \frac{\alpha^2 y_L^2}{2(1 - \alpha)^2} \\ + \text{Log}_e \left| \frac{1 - \alpha y_L}{y_L} \right| - \frac{2\alpha y_L}{1 - \alpha y_L} - \frac{\alpha^2 y_L^2}{2(1 - \alpha y_L)^2} . \quad (4.22)$$

A compatible solution for v_d is obtained to satisfy both Eqs. 4.9 and 4.10, with $\Delta t = 117$ hr to cover $\Delta z = 0.0146$ m, and for $t = 1262$ hr to reach $w = w_f$. These conditions give $v_d = 0.55 \times 10^{-7}$ m/s for $\rho_{\beta 0} = 0.036$ kg/m³ and $\rho_\beta = 1000$ kg/m³. For the present computation, we took $\alpha = 0.4446$. Figure 4.1 shows the result of computation with average $-\Delta P$ given by Eqs. 4.18 and 4.22 for ANL-Laboratory Analog Experiment 2.

4.4 CONCLUSIONS

In a real situation, Fig. (4.3d) is the idealized averaged configuration of repeated buildup and reentrainment, as in Fig. 4.2(d).

If we use the values $v_d = 0.55 \times 10^{-7}$ m/s and $\rho_{\beta 0} = 0.032$ kg/m³, we get a total time of 1410 hr to reach the asymptotic condition at $t = 3160$ hr, indicated by a triangle (Δ) in Fig. 4.1. This will give a better fit at later times.

The estimated value of 0.032 kg/m³ in a bentonite concentration is judged to be reasonable compared with the measured value of 0.026 kg/m³* taken at the inlet of the core near the end of the experiment with a 0.4 μ m filter and dried sample weighed. Those particles less than 0.4 μ m in size not being collected and weighed may contribute to the discrepancy.

Another possible input to obtain a better fit to the recorded pressure data is to assume that the passage width increased as the pumping pressure increased beyond 200 kPa, causing an increase in flow passage width within the confine of the test specimen. In doing so, we found that the pressure drop decreased at 3200 hr as a slug of deposit was pushed downstream. At large pressure drops, large slugs of deposit were pushed away and settled downstream or dispersed and redeposited. Finally, the experiment discontinued at 3800 hr and the pressure drop decreased and fluctuated around its background value. Another possible explanation to account for the discrepancy between the simplified computation and the recorded data is that the suspended bentonite concentration changed with time through the experiment.

*Communication with D. Bowers, Chemical Technology, Argonne National Laboratory

5. TRANSPORT OF ACTINIDES AND BACKFILL MATERIAL

5.1 MODELING

The groundwater passing through a repository may carry with it dissolved actinides and backfill material (say bentonite) into rock strata of certain porosity and directional permeability; subsequent deposition of bentonite and adsorption of actinide may occur in the rock strata.

Groundwater carrying suspended bentonite particles will have heat J_{qc} and mass flux J_{mc} , given by

$$J_{mc} = \rho_c (U_c - U_w) = -D_{cm} \nabla \rho_c - D_{cm} S_T^i \rho_c \nabla T, \quad (5.1)$$

where D_{cm} is the mass diffusivity of bentonite (c for clay), ρ_c is the density of suspended materials, and S_T^i is the Soret coefficient[†], accounting for an effect that may become significant at large temperature gradients. Simultaneously,

$$J_{qc} = -D_{cm} S_T^i \rho_c T \mu_{cc} \nabla \rho_c - K \nabla T, \quad (5.2)$$

where μ_{cc} is a chemical potential, K is thermal conductivity, and the first term on the RHS accounts for the Dufour effect, which generally is small. Deposition of bentonite is given by

$$\frac{\partial \rho_c}{\partial t} + U_w \cdot \nabla \rho_c = -\frac{2\rho_c v_{dc}}{w} + \frac{2\rho_c D (U_w - U_w^*) C_e}{w}, \quad (5.3)$$

where w is gap width, v_{dc} is deposition velocity, U_w^* is a characteristic velocity for reentrainment, and C_e is a constant for the probability of reentrainment. The change in average gap width w , then, is

$$\frac{\partial w}{\partial t} = -\frac{2\rho_c}{\bar{\rho}_c} v_{dc} + \frac{2\rho_c D}{\bar{\rho}_c} (U_w - U_w^*) C_e \quad (5.4)$$

for deposition on both walls of the gap.

The concentration of actinide carried by groundwater can be represented, before equilibrium is reached, by

$$\frac{\partial \rho_A}{\partial t} + U_w \cdot \nabla \rho_A = -\frac{2\rho_A v_{dA}}{w}, \quad (5.5)$$

[†] $S_T^i \sim 2.95 \times 10^{-3} \text{ K}^{-1}$, $D_{cm} \sim 4.57 \times 10^{-5} \text{ cm}^2/\text{s}$.

where v_{dA} is the adsorption velocity. Because of the low speed of groundwater flow, leaching (if any occurs) and adsorption on rock can be treated as occurring at equilibrium condition for the given pH value of groundwater and its subsequent changes.

For a rock mass with cracks of average width w and n_1, n_2, n_3 cracks per unit length in the principal directions x_1, x_2 , and x_3 (which may be aligned in any orientation with the position coordinates of the problem), the porosity and permeability can be described accordingly:

$$\gamma = w(n_1 + n_2 + n_3) = w \sum_{1,2,3} n_i \quad (5.6)$$

$$k_1 \cong \frac{1}{20} w^3 n_1 \quad (\text{for } w \ll n^{-1}) \quad (5.7)$$

$$k_2 \cong \frac{1}{20} w^3 n_2 \quad (5.8)$$

$$k_3 \cong \frac{1}{20} w^3 n_3 \quad (5.9)$$

The coefficient $1/20$ includes tortuosity and shape factors. Equations (5.6) - (5.9) can be readily applied to compute the mass flux in the coordinates chosen for the problem.

The effect of actinides carried forth by groundwater and their adsorption affects the temperature distribution in the rocks according to

$$c_B \rho_B \frac{\partial T_B}{\partial t} = K_B \nabla^2 T_B + n_A \left(\sum n_i \right) J_E \frac{c_B \rho_B (1 - \gamma_B)}{c_B \rho_B (1 - \gamma_B) + c_w R \gamma_B},$$

where n_A is the number of g-atom actinide per unit area of rock surface (which is less than the number of sites for chemisorption of actinide). J_E is the heat released by radioactivity per g-atom actinide. K_B, c_B, ρ_B are the thermal conductivity, specific heat, and density of the rock.

5.2 CONCLUSIONS

The feasibility of formulating the complete problem is readily seen. The groundwater characteristics need to be given the inlet pH value. Solubility of the rock, the bentonite, and the actinide, and how the pH value alters while coming into equilibrium with various surfaces also need to be known. The adsorptivity of actinide on various rocks (e.g., basalt) and various backfill materials (e.g., bentonite) needs to be determined. We have determined some of them by using experimental results of the laboratory analog problem.

6. FLOW AND RESATURATION OF GROUNDWATER THROUGH BACKFILL

6.1 RESATURATION PROCESS

The initial condition after backfilling a repository may be idealized according to Fig. 6.1, which shows that an excavation in basalt rock (B) is backfilled with bentonite (C for clay) which has been air-dried with porosity γ_{cd} after packing; the pores are filled with air. The repository (R) may be treated as a body with a distribution heat flux around its surface $J_{qR}(r_R, t)$, where r_R is the position coordinate around the surface of repository and t is the time. Right after backfilling, the bentonite is at temperature T_0 , and the water (W) at temperature T_{w0} is ready to seep in from the rock (B) of given porosity γ_B . The pressure distribution in various parts of the system is initially at $P_0(r)$ at $t = 0$.

After the whole system is in place for some time, but before evaporation of water occurs (it might never occur, depending on the pressure-temperature relation), the situation will be as depicted in Fig. 6.2. At time t , part of the backfill becomes saturated with degree of saturation s^* equal to 1.

$$s^* = \frac{s, \text{ mass of water absorbed per unit mass of backfill}}{s_0, s \text{ at saturation}}.$$

Behind this front of $s^* = 1$, the porosity is γ_{cs} , including the effect of swelling and permeability k_{cs} . Because of the relative rates of saturation and porous body flow, we expect a thickness between $s^* = 1$ and $s^* = 0$, where the groundwater at velocity U_w reaches at time t . Ahead of this front, the trapped air in the backfill may be displaced by the water ($U_a = U_w$ at interface S of $s^* = 0$), dissolved in the water, or trapped behind as water bypasses certain points. In the meantime, because of heating by the repository, both the dry and wet backfill materials and the fluids are being heated, flow occurs in the pores, and pressure P and temperature T_{cd} and T_{cs} increase. Temperature gradient ∇T and pressure gradient $\nabla P'$ appear in the backfill.

$$\nabla P' = \nabla(P + P_c) - \rho_m g, \quad (6.1)$$

where P_c is the capillary pressure and ρ_m is the density of materials constituting the hydraulic pressure under gravitational acceleration g .

When evaporation occurs, another domain will evolve.

6.2 APPROXIMATE FORMULATION

Although a general set of equations for detailed computation is readily formulated, the nature of the problem is best illustrated with a set of approximate formulations.

As indicated above, a first-order approximation is to assume that the water seeps forward as it saturates the backfill. In this case, part of the

Fig. 6.1 Idealized Initial Condition for Flow and Resaturation of Backfill

Fig. 6.2 Resaturation at $t > 0$ showing the Moisture Front with Thickness between $S^* = 0$ and $S^* = 1$

mass flux of groundwater is to saturate the backfill as it moves forward. Extending the Darcy formulation to include the effect of temperature gradient gives

$$\underline{J}_{mw} = \rho_w \underline{U}_w = -\rho_w \frac{k_{cs}}{\mu_w} \frac{\alpha_w}{\beta_w} \nabla T - \rho_w \frac{k_{cs}}{\mu_w} \nabla P' , \quad (6.2)$$

while the rate of saturation per unit volume is $s_o \rho_c (1 - \gamma_{cd}) \frac{\partial U_w}{\partial r}$, where ρ_w is the density of water, γ_{cd} is the porosity of the backfill, μ_w is the viscosity of water, k_{cs} is the permeability of saturated backfill, and

$$\alpha_w = -\rho_w^{-1} (\partial \rho_w / \partial T)_P , \text{ and} \quad (6.3)$$

$$\beta_w = \rho_w^{-1} (\partial \rho_w / \partial P)_T \quad (6.4)$$

are the coefficients of thermal expansion and of compressibility of water. P_c depends on the geometry and surface of the pores. The continuity condition gives

$$\nabla \cdot \underline{U}_w = 0 . \quad (6.5)$$

The energy equation is now

$$c_c \rho_c + s_o \rho_c (1 - \gamma_{cs}) c_w \frac{\partial T}{\partial t} = K_{cs} \nabla^2 T , \quad (6.6)$$

where we have neglected the convection effect of w because of slow motion. In Eq. 6.6, K_{cs} is the thermal conductivity of the saturated backfill, and c_c and c_w are specific heat of backfill and water. The heat transferred by the water is small unless evaporation occurs. When local P is lower than the saturation pressure at the local temperature T , the front surface of water seepage is given by

$$S(r, t) = \int_0^t \underline{U}_w dt . \quad (6.7)$$

Ahead of this surface S , we have the region of dry backfill and the repository. In the dry backfill, the mass flux of air is given by

$$\underline{J}_{ma} = \rho_a \underline{U}_a = -\rho_a \frac{k_{cd}}{\mu_a} \frac{P}{T} \nabla T - \rho_a \frac{k_{cd}}{\mu_a} \nabla P'' , \quad (6.8)$$

where subscript a denotes air, $\alpha_a / \beta_a = P/T$, $\rho_a = P/R_a T$, and R_a is the gas constant,

$$\nabla P'' = \nabla P - \rho_m \underline{g} . \quad (6.9)$$

The continuity condition gives

$$\gamma_{cd} \frac{\partial \rho_a}{\partial t} = - \nabla \cdot \underline{J}_{ma} \quad (6.10)$$

and the energy equation is given by

$$c_c \rho_{cd} \frac{\partial T}{\partial t} = K_{cd} \nabla^2 T. \quad (6.11)$$

With given properties, the above equations can be solved simultaneously with the following boundary conditions:

At $S(\underline{r}, t)$, P , T , and U are continuous (T would not be continuous if a phase change of water occurred; $U_a = 0$ if air dissolves in water),

At $\underline{r} = \underline{r}_B$, P , T_w are given,

At $\underline{r} = \underline{r}_R$, $\underline{J}_{qR} = -K_{cd} \nabla T$ (K_{cd} will be replaced by $-K_{cs}$ when surface S reaches $\underline{r}_R$), and

At the top surface of the backfill, P and T are given; the ground surface may be assumed impervious and insulated.

The initial conditions are given in Fig. 6.1.

6.3 CONCLUSIONS

The general cases need not be symmetric, as suggested by Figs. 6.1 and 6.2. Figure 6.3 illustrate possible cases of groundwater seepage past a repository. Case a illustrates the saturation where available temperature and pressure gradients keep the groundwater from seeping into the repository. Case b illustrates complete resaturation of backfill; leaching of repository material may occur. Water cannot reach the repository where

$$(\alpha_w / \beta_w) \nabla T = - \nabla P',$$

but instability and "fingers" need to be explored.

7. CONCLUDING REMARKS

Reliable prediction of high-level-waste (HLW) repository performance is necessary for safe and permanent isolation of radioactive waste. However, due to lack of available data, the values of transport and related parameters are being estimated or guessed in current computer modeling efforts. The information about the transport parameters is needed and essential for true and realistic prediction of HLW repository performance. The understanding of transport processes also is useful for extrapolation of full-scale repository performance from small-scale laboratory analog measurements.

Fig. 6.3 Possible Cases of Groundwater Flow Past Repository

The present analysis describes how we can obtain transport parameters without additional experimental costs. The information about transport parameters are obtained as additional useful information from the existing analog experiments, thus maximizing the gain from a given experiment. Furthermore, detailed analysis of this kind increases our understanding of various transport processes that occur simultaneously in the far and near fields of a HLW repository.

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