Title

Mass Transport in Salt Repositories: Steady-state Transport through Interbeds

Authors

Hwang, Y
Lee, W W.-L.
Chambré, P L
et al.

Publication Date

1989-03-01

LBL-26704
UC-802

Lawrence Berkeley Laboratory
UNIVERSITY OF CALIFORNIA

EARTH SCIENCES DIVISION

**Mass Transport in Salt Repositories:
Steady-state Transport through Interbeds**

Y. Hwang, W.W.-L. Lee, P.L. Chambré,
and T.H. Pigford

March 1989

Prepared for the U.S. Department of Energy under Contract Number DE-AC03-76SF00098.

DISCLAIMER

LBL-26704
UCB-NE-4136
UC-802

Mass Transport in Salt Repositories:

Steady-State Transport Through Interbeds

Y. Hwang, W. W.-L. Lee, P. L. Chambré and T. H. Pigford

Department of Nuclear Engineering
University of California

and

Earth Sciences Division, Lawrence Berkeley Laboratory
1 Cyclotron Road
Berkeley, CA 94720

March 1989

This work was supported in part by the Director, Office of Civilian Radioactive Waste Management, Office of Systems Integration and Regulations, Licensing and Compliance Division, of the U.S. Department of Energy under Contract No. DE-AC03-76SF00098.

Contents

Figures

1. Introduction

Salt has long been a candidate for geologic disposal of nuclear waste.[1] Because salt is extremely soluble in water, the existence of rock salt in the ground atest to the long-term stability of the salt. Both bedded salt and salt domes have been considered for nuclear waste disposal in the United States and Europe.[2,3,4] While the salt is known to be quite pure in salt domes, bedded salt is interlaced with beds of sediments. Traditionally rock salt has not been considered water-conducting, but sediment layers would be classical porous media, capable of conducting water. Therefore there is interest in determining whether interbeds in bedded salt constitute a significant pathway for radionuclide migration.

In this report we consider steady-state migration of radionuclides from a single waste cylinder into a single interbed. Two approaches are used. In 1982 Neretnieks proposed an approach for calculating the steady-state transport of oxidants to a copper container.[5] We have adapted that approach for calculating steady-state radionuclide migration away from the waste package, as a first approximation. We have also analyzed the problem of time-dependent radionuclide diffusion from a container through a backfill layer into a fracture, and we used the steady-state solution from that problem for comparison.

Section 2 gives a brief summary of the geology of interbeds in bedded salt. Section 3 presents the mass transfer resistances approach of Neretnieks, summarizing the formulation and giving numerical illustrations of the steady-state two-dimensional diffusion analysis. Section 4 gives a brief statement of the steady-state result from a related analysis. Conclusions are stated in Section 5.

2. Geology of Interbeds in Bedded Salt

Using the Deaf Smith County site as an example, the following summary of the geology of interbeds is taken from the *Environmental Assessment*.[6]

The potential repository at the Deaf Smith site is to be in a thick salt layer known as Unit 4 of the Lower San Andres Formation. Because no exploratory wells have been drilled at the precise location of the site, the detailed geology is inferred from the stratigraphy of nearby wells and regional geology. The depth to the top of Unit 4 is about 700 meters and Unit 4 is about 76 meters thick. It consists of 2 meters of gray anhydritic mudstone and 25 meters of interbedded dolomite, anhydrite, and limestone overlain by approximately 49 meters of bedded salt. The host salt layer is composed predominantly of halite, anhydrite, and mudstone, with trace amounts of several other constituents such as dolomite and celestite.

In the host salt layer, about 8 per cent by volume is mudstone. About half of this mudstone is in discrete mudstone beds, and half is distributed within the salt as chaotic mudstone/salt mixtures. From 91 to 116 discrete mudstone beds have been identified in the cores of 3 exploration wells (Table I). Average mudstone bed thickness ranges from 1.5 to 3 cm and maximum mudstone bed thickness is 31 to 64 cm.

Mudstone beds are composed of halite, anhydrite and mudstone. Mudstone is a clastic material composed of silt- and clay-sized material, and consitutes about 65 per cent of the mudstone beds at the Deaf Smith site. In the mudstone beds, there is about 20 per cent halite occurring as coarse to euhedral crystals, cements and fracture fillings. The anhydrite, 15 per cent of the mudstone beds at Deaf Smith, occurs as nodules, blebs and cement.

Table I. Composition of the Lower San Andres Unit 4

Well Location	J. Friemel No. 1 4.8 km SE	G. Friemel No. 1 27 km SE	Detten No. 1 22 km SE
Thickness (m)	50.5	46.2	52.3
Composition (vol. %)			
Halite	87	89	87
Anhydrite	4	4	5
Mudstone	9	7	8
Anhydride Beds	86		
Mudstone Beds	91	116	112
Composition (vol. %)			
Halite	20		
Anhydrite	15		
Silt and Clay	65	63	75
Thickness			
Maximum (mm)	470	310	640
Average (mm)	30	15	19

It can be seen that interbeds are a significant part of bedded salt horizons.

3. The Mass Transfer Resistances Approach

3.1 Derivation

The situation studied is shown in Figure 1. A waste package is inserted into an emplacement hole in a salt repository and backfilled with crushed salt. The emplacement hole intersects an interbed. Given the large

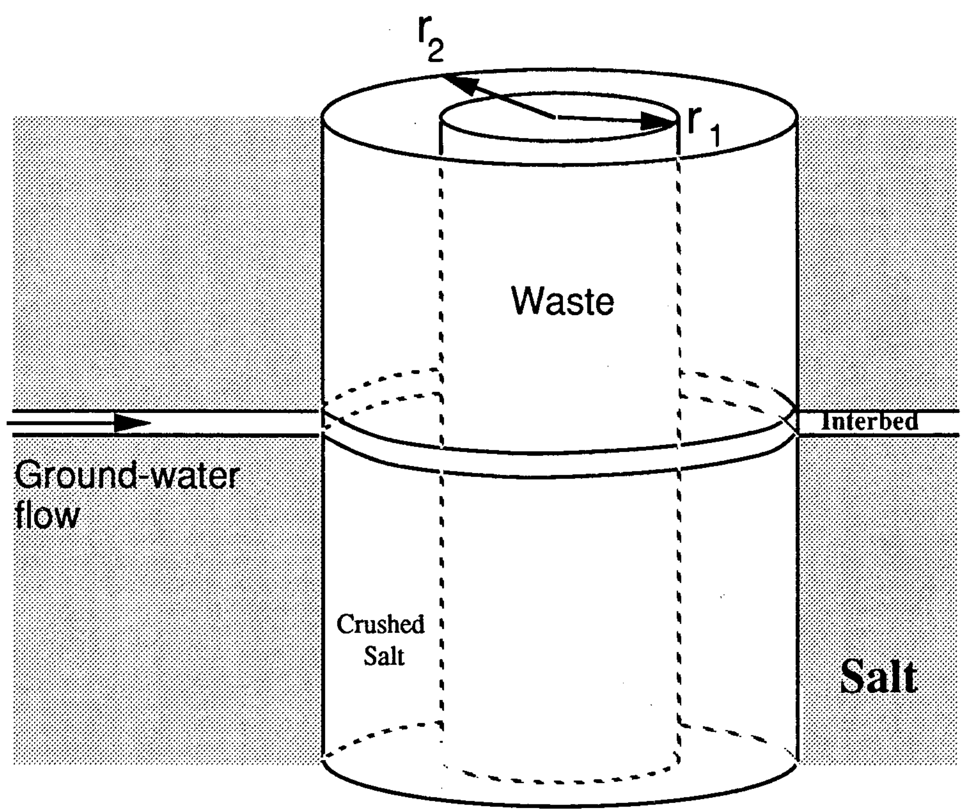

Figure 1 Waste package intersected by an interbed

number of interbeds observed in cores near the Deaf Smith site, this is probable. The following assumptions are used.

- The crushed salt has consolidated to the extent that there is no flow within the crushed salt region.
- There is ground water flow in the interbed and the velocity is constant.
- The interbeds are planar and perpendicular to the longitudinal axis of the waste cylinder.
- The spacing between interbeds is the same.
- The waste cylinder is infinitely long. That is, end effects are ignored.
- Temperature effects are accounted for by using constant values of parameters such as diffusion coefficients, but for the highest temperatures expected.
- The surrounding salt is impervious to water flow and radionuclide transport.
- Steady-state conditions prevail.

With these assumptions, the problem reduces to one of one-dimenional diffusion. For a nuclide to move from the waste cylinder where it is at a higher concentration to the flowing ground water in the interbed where it is at a lower concentration, the problem can be conceptualized as

$$\dot{m} = KA\Delta C \tag{1}$$

where $\dot{m}$ is the mass transport rate, C is the species concentration and ΔC is the concentration-gradient driving force, A the area over which mass transport is taking place and K a mass transport coefficient. In this case there are two resistances to mass transport, the crushed salt layer and the porous material in the interbed. If we designate the total mass transport resistance as R_Σ and define it as

$$R_\Sigma = \frac{\Delta C}{\dot{m}} \tag{2}$$

then we have individual mass transport resistances R_i and R_c in the interbed and the crushed salt respectively. Using the theory of additivity of mass transfer resistances[7]

$$R_\Sigma = R_i + R_c \tag{3}$$

From (1) and (2)

$$R = \frac{1}{KA}$$

and

$$\frac{1}{KA} = \frac{1}{k_i A_i} + \frac{1}{k_c A_c} \tag{4}$$

Using the contact area with the waste cylinder as the reference area of mass transport, setting $A = A_c$, we have

$$K = \frac{1}{\frac{A_c}{k_i A_i} + \frac{A_c}{k_c A_c}} \tag{5}$$

Thus the overall mass transport is

$$\dot{m} = KA_c\Delta C \tag{6}$$

$$= \frac{k_i k_c A_c A_i}{k_i A_i + k_c A_c}\Delta C \tag{7}$$

If we assume that the species concentration at the waste cylinder surface is saturation and the initial concentration in the interbed is zero, then ΔC is simply C_s, the saturation concentration of the species in ground water

$$\dot{m} = \frac{k_i k_c A_c A_i}{k_i A_i + k_c A_c} C_s \tag{8}$$

Now we need to determine the parameters in (8).

First we consider transfer from the waste cylinder. The contact between the waste cylinder and the crushed salt can be visualized as a cylindrical strip around the waste cylinder, with a thickness and a radius. For the thickness, Neretnieks suggested a logarithmic average thickness δ,

$$\delta = \frac{S - 2b}{\ln(S/2b)} \tag{9}$$

where S is the spacing between interbeds and $2b$ is the thickness of the interbed. The radius of the cylindrical strip can be approximated by a logarithmic average radius

$$r_m = \frac{r_2 - r_1}{\ln(r_2/r_1)} \tag{10}$$

where r_1 is the waste cylinder radius and r_2 is the emplacement hole radius. We then have

$$A_c = 2\pi\delta r_m$$

$$= 2\pi \frac{r_2 - r_1}{\ln(r_2/r_1)} \frac{S - 2b}{\ln(S/2b)} \tag{11}$$

The mass transport coefficient in the crushed salt region is the diffusion coefficient divided by the distance traversed, reduced appropriately by the porosity

$$k_c = \frac{\epsilon_c D_f}{r_2 - r_1} \tag{12}$$

where ϵ_c is the porosity of the crushed salt region and D_f is the molecular diffusion coefficient in the same region.

The contact area between the crushed salt region and the interbed is

$$A_i = 2\pi r_2 2b \tag{13}$$

Neretnieks suggests the following mass transfer coefficient in the interbed[5]

$$k_i = \sqrt{\frac{4D_f \epsilon_i}{\pi t_k}} \tag{14}$$

where ϵ_i is the porosity in the interbed and $t_k = \pi r_2/u_p$ is a contact time defined as "the time a 'liquid parcel' with a velocity of u_p remains at the interface." Substituting in the expression for t_k we have

$$k_i = \sqrt{\frac{4D_f \epsilon_i u_p}{\pi^2 r_2}} \tag{15}$$

Multiplying (13) and (14) we have

$$A_i k_i = 2\pi r_2 2b \sqrt{\frac{4D_f \epsilon_i u_p}{\pi^2 r_2}} \tag{16}$$

Rearranging (16) we get

$$A_i k_i = 8bD_f \epsilon_i \sqrt{\frac{u_p r_2}{D_f \epsilon_i}} \tag{17}$$

Recognizing $v = u_p/\epsilon_i$ as the pore velocity and vr_2/D_f as the Peclet number, we have

$$A_i k_i = 8bD_f \epsilon_i \sqrt{Pe} \tag{18}$$

to be substituted into (8). This completes the specification of parameters in (8).

Although the derivation given above assumes an infinite cylinder, we need to consider a cylinder of finite dimensions to obtain a fractional release rate. A long cylinder would intersect many interbeds. Each interbed would *see* only a portion of a waste cylinder. If the length of a waste cylinder is L and the 1000-year inventory of the species is $\bar{m}$, then the fractional release rate f can be defined as

$$f = \frac{L\dot{m}}{S\bar{m}} \tag{19}$$

Having defined all the parameters, we can now provide numerical illustrations of Eq. (8).

3.2 Numerical Illustrations

We calculate the fractional release rate for ^{238}U from a single waste cylinder through an interbed. Table II shows the values of parameters we used in numerical calculations. We use a waste cylinder diameter of 0.31 m, a crushed salt region thickness of 3 cm. Salt material properties are taken from studies by RE/SPEC Inc.[9] and McTigue.[10] We vary the porosity of the porous material in the interbed from 0.005 to 0.1. We consider three values of interbed separation, 0.5 m, 1.0 m and 2.0 m; and three values of interbed thickness, 0.1 cm, 1 cm and 5 cm. The fractional release rates are direct multiples of the species solubility and we use two estimates of uranium solubility, 0.001 g/m^3 from an earlier estimate for reducing conditions[11] and a more recent estimate for oxidizing conditions, 50 g/m^3 (P. Cloke, personal communication, 1987).

Table II. Input Data

Radius of waste cylinder	0.31 m
Length of waste cylinder	3.65 m
U-238 1000-year inventory	5.4×10^6 g
Crushed salt thickness	3 cm
Crushed salt porosity	0.001
Interbed thickness	0.1, 1.0, 5 cm
Interbed separation	0.5, 1.0, 2.0 m
Interbed porosity	0.005, 0.01, 0.1
Diffusion coefficient	10^{-7} cm^2/s
Uranium Solubility	0.001, 50 g/m^3

The fractional release rates of ^{238}U from a waste cylinder through a single interbed and as a function of the parameters in Table II are shown in Figures 2, 3 and 4. As might be expected, the release rates are well below the allowable U.S. Nuclear Regulatory Commission (USNRC) limit of 10^{-5} per year.

4. Time-dependent Diffusion Approach

We compare the results obtained above from the mass transfer resistances approach with a closely related approach.[12] In this model we consider a waste canister surrounded by a backfill layer consisting of bentonite and crushed rock in a borehole intersected by a fracture, in water-saturated rock. Radionuclides are released at a constant concentration at the waste surface into the backfill. Ground water flows in the fissure. We assume no ground-water flow in the backfill, so that radionuclide transport through the backfill is controlled by molecular diffusion. The rock matrix is assumed to be completely impervious, thus mass transport in the rock takes place in the fracture only. Here salt substitutes for the rock and an interbed for the rock fracture. The mass flux into the rock/salt is given by the mass transfer coefficient times the average nuclide concentration across the fissure mouth. For a small hole-to-canister radius ratio, cylindrical geometry can be simplified to planar geometry. This problem has been solved analytically over the entire time domain. The steady-state solution from that analysis is plotted in Figure 5 for comparison. This transient analysis is a more detailed analysis and it is not surprising that there is some difference between the results of the two approaches.

Figure 5 also shows the fractional release rates predicted by a modified mass transfer resistance approach.

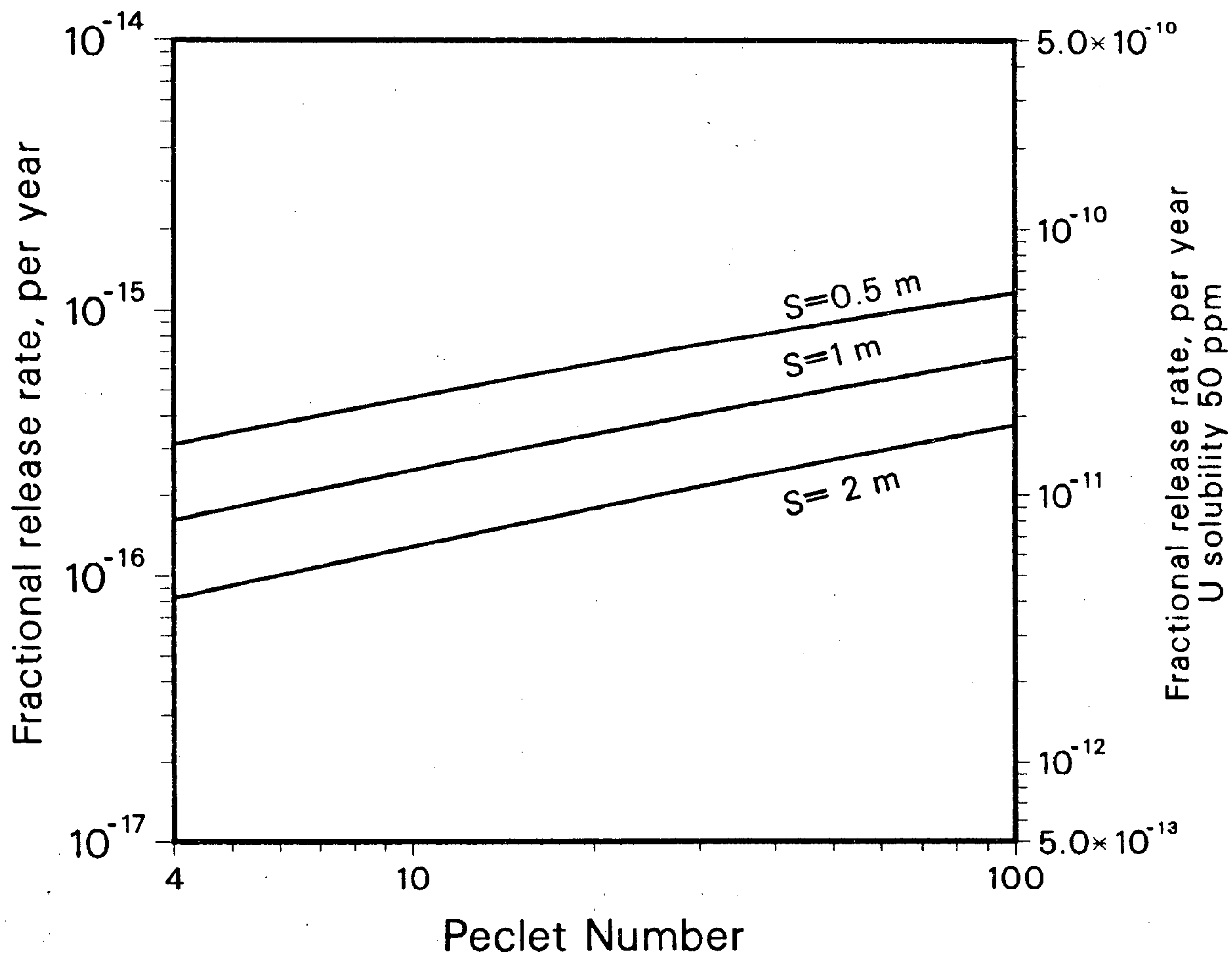

Figure 2. Fractional Release Rates with Different Interbed Separations

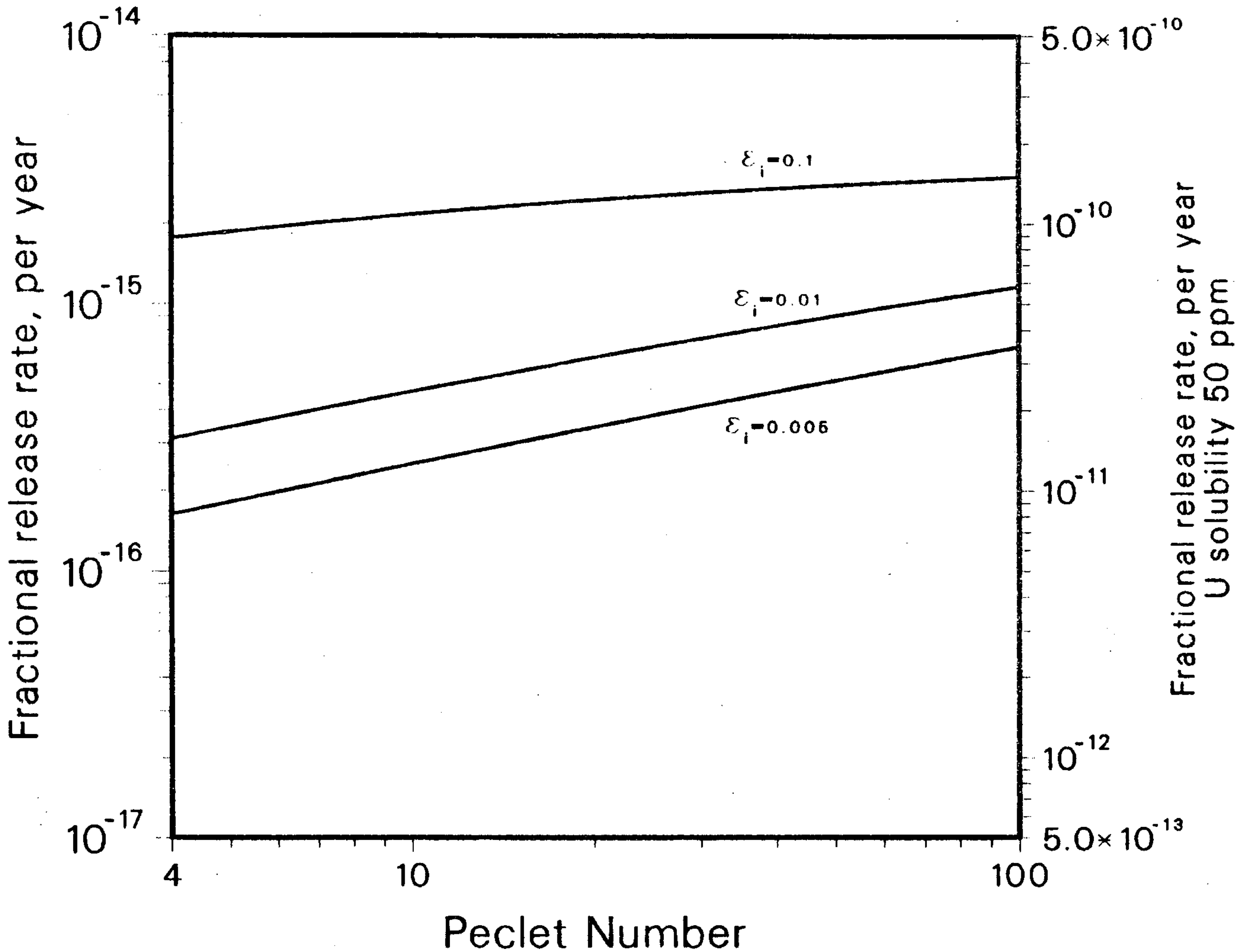

Figure 3. Fractional Release Rate as a Function of Interbed Porosity

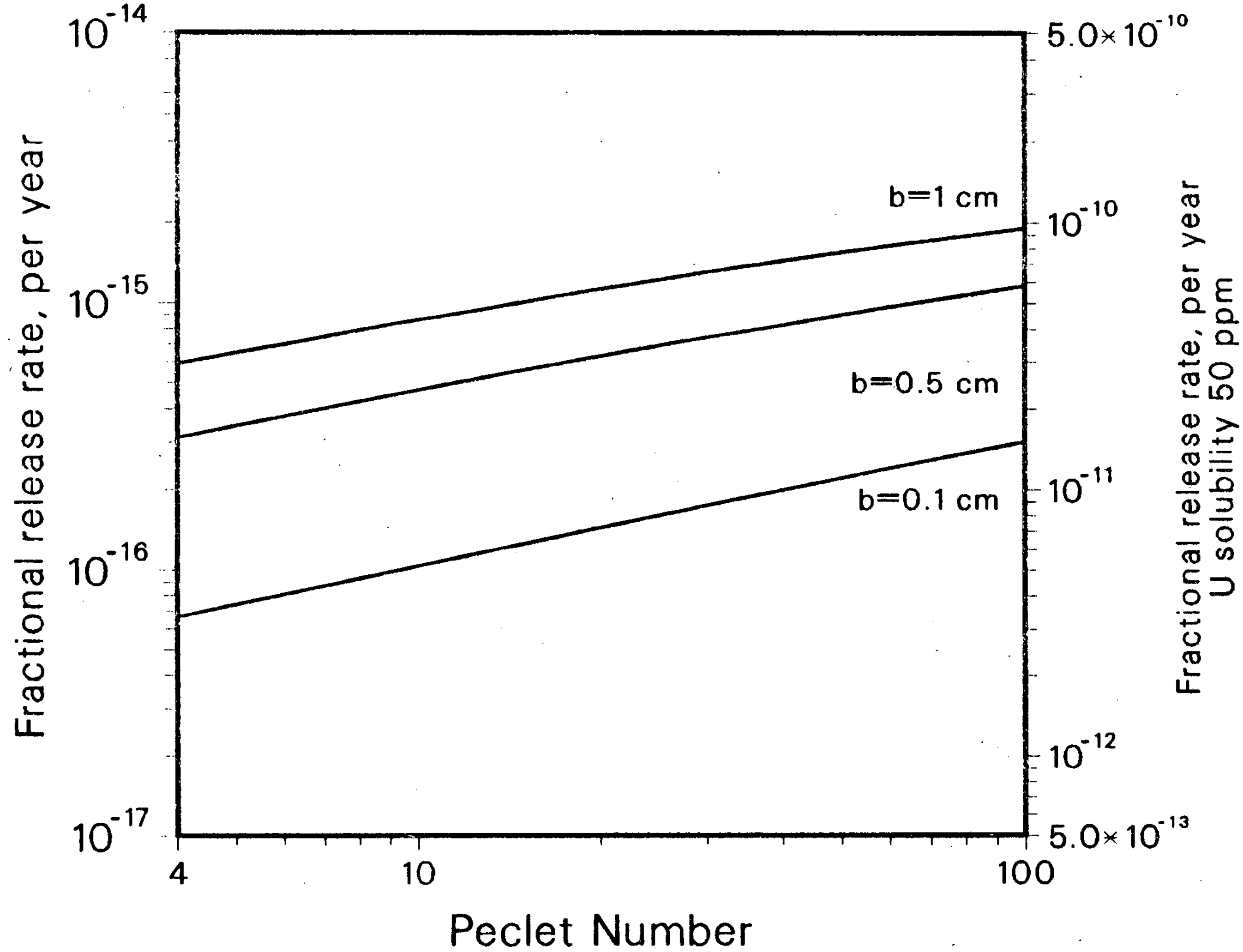

Figure 4. Comparison of Fractional Release Rates of U-238 for Different Interbed Thicknesses

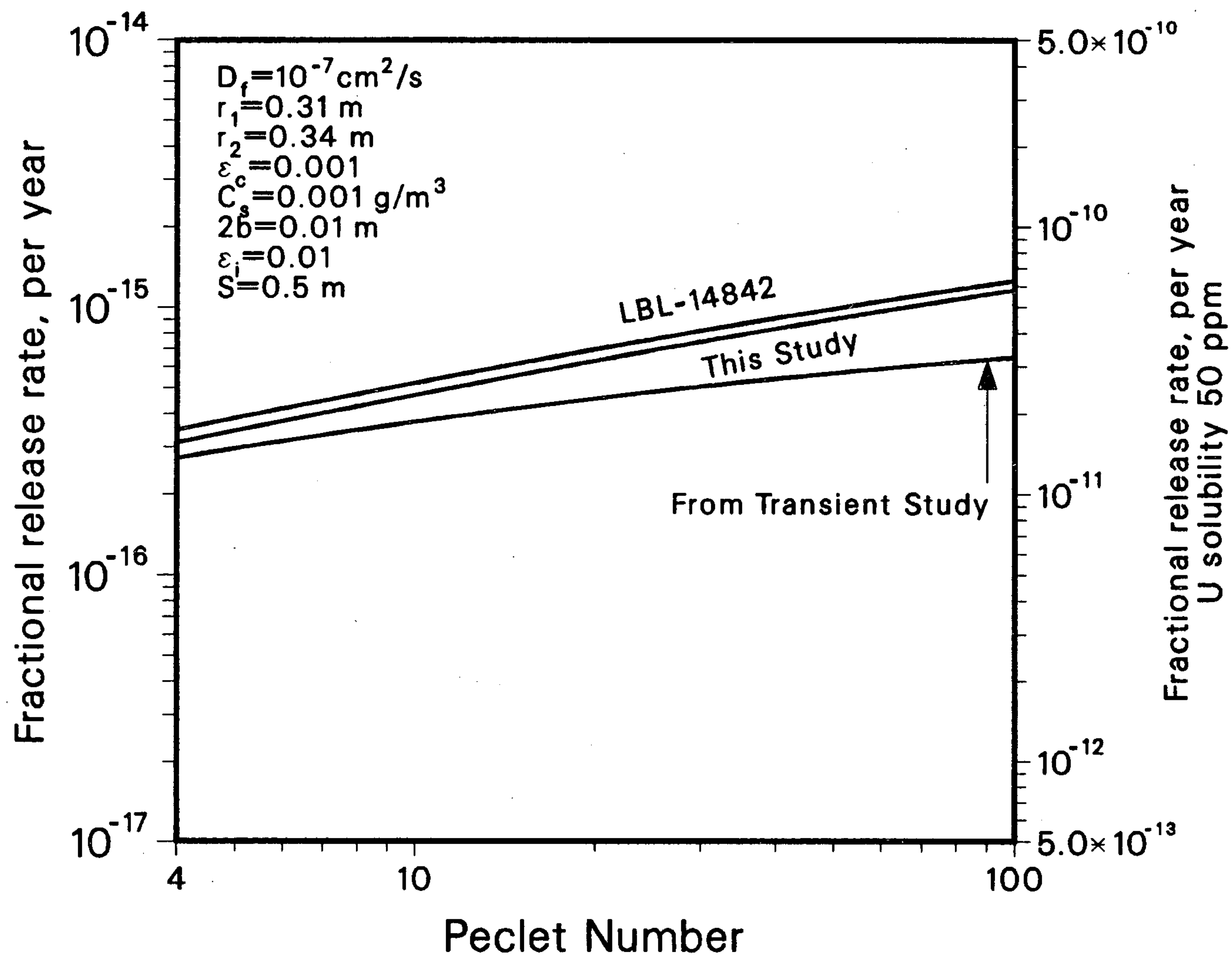

Figure 5. Comparison of Fractional Release Rates of U-238 for Three Different Approaches

In previous work,[13] we have used an equation for the mass transfer rate that is well known in heat and mass transfer and that is almost identical to (18)

$$A_i k_i = 16 b D_f \epsilon_i \sqrt{Pe/\pi} \tag{20}$$

Eq. (20) gives the steady-state mass transfer rate per unit length of an infinite cylinder and a typical result for an interbed of thickness $2b$ is the line indicated as "LBL-14842" in Figure 5.

5. Conclusions

This is a first-cut analysis of mass transport through interbeds in bedded-salt nuclear waste repositories. Some assumptions have been made, such as steady state conditions and flowing ground water in the interbeds. The results indicate that mass transport under such assumptions is small and for the parameter values used in the numerical illustration, radionuclide transport through interbeds is not a significant pathway.

References

1. National Academy of Sciences, 1957, *The Disposal of Radioactive Waste on Land,* Report of the Committee on Waste Disposal, Division of Earth Sciences, Publication 519, Washington, DC.

2. U. S. Department of Energy, 1985, *The Mission Plan for the Civilian Radioactive Waste Management Program,* DOE/RW-0005, Washington, DC.

3. K. D. Closs and K. Einfeld, 1986, "Overview of the FDG Activities on Spent Fuel Disposal," in Proceedings of the 2nd International Conference on Radioactive Waste Management, Winnipeg, Canada, 26.

4. H. D. K. Codee and J. Virjen, 1987, "Radwaste in the Netherlands," *Waste Management '87, 1,* 97.

5. G. Andersson, A. Rasmuson and I. Neretnieks, 1982, *Migration model for the near field, Final Report,* SKBF/KBS Report 82-24.

6. U. S. Department of Energy, 1986, *Environmental Assessment, Deaf Smith County Site, Texas,* DOE/RW-0069, Washington, DC.

7. T. K. Sherwood, R. L. Pigford and C. L. Wilke, 1975, *Mass Transfer,* New York: McGraw-Hill.

8. U. S. Nuclear Regulatory Commission, Disposal of High-Level Radioactive Wastes in Geologic Repositories, 10 *Code of Federal Regulations* 60.113(a)(1)(ii)(B).

9. T. Brandshaug, 1987, *Estimate of Consolidation of Crushed Salt Around a Spent Fuel Waste Package,*

RE/SPEC Report RSI-315.

10. D. F. McTigue, 1986, "Thermoelastic Response of Fluid-Saturated, Porous Rock," *J. Geophy. Res., 91, B9,* 9533.

11. T. H. Pigford, J. O. Blomeke, T. L. Brekke, G. A. Cowan, W. E. Falconer, N. J. Grant, J. R. Johnson, J. M. Matusek, R. R. Parizek, R. L. Pigford, and D. E. White, *A Study of the Isolation System for Geologic Disposal of Radioactive Wastes,* Washington: National Academy Press, 1983.

12. C. H. Kang, P. L. Chambré, W. W.-L. Lee and T. H. Pigford, "Time-Dependent Nuclide Transport Through Backfill into a Fracture," *Trans. Am. Nuc. Soc., 55,* 134, 1987.

13. P. L. Chambré, T. H. Pigford, Y. Sato, A. Fujita, H. Lung, S. Zavoshy, and R. Kobayashi, *Analytical Performance Models,* Report LBL-14842, 1982.

LAWRENCE BERKELEY LABORATORY
TECHNICAL INFORMATION DEPARTMENT
UNIVERSITY OF CALIFORNIA
BERKELEY, CALIFORNIA 94720