LBL-26852

Lawrence Berkeley Laboratory
UNIVERSITY OF CALIFORNIA

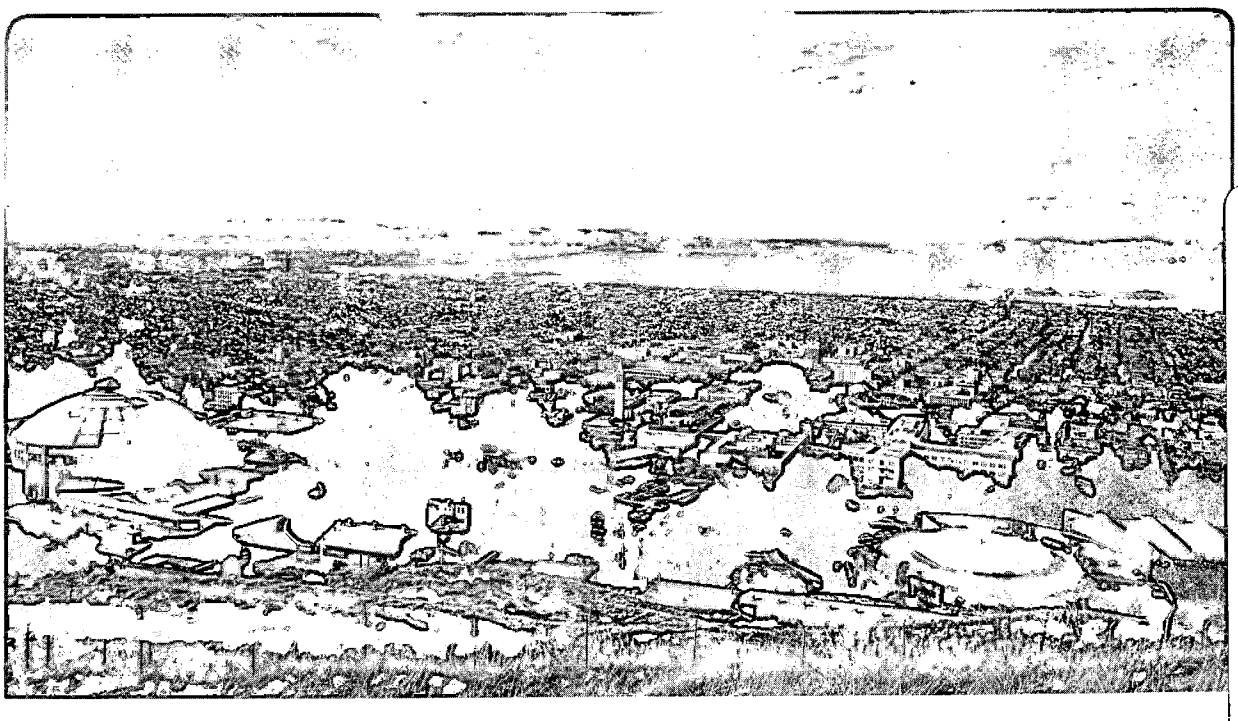
EARTH SCIENCES DIVISION

To be presented at the International Symposium on the Safety Assessment of Radioactive Waste Repositories, Paris, France, October 9–12, 1989, and to be published in the Proceedings

Mass Transfer and Transport in Salt Repositories

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

February 1989



LOAN COPY
Circulates
for 2 weeks
Bldg. 50 Library.
Copy 2
LBL-26852

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

DISCLAIMER

This document was prepared as an account of work sponsored by the United States Government. While this document is believed to contain correct information, neither the United States Government nor any agency thereof, nor the Regents of the University of California, nor any of their employees, makes any warranty, express or implied, or assumes any legal responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by its trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof, or the Regents of the University of California. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof or the Regents of the University of California.

LBL-26852

Mass Transfer and Transport in Salt Repositories

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

Earth Sciences Division
Lawrence Berkeley Laboratory
University of California
Berkeley, California 94720

February 1989

This work was supported 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.

UCB-NE-4140
LBL-26852

MASS TRANSFER AND TRANSPORT IN SALT REPOSITORIES

T. H. Pigford, P. L. Chambré & W. W.-L. Lee

**Department of Nuclear Engineering
University of California
and
Lawrence Berkeley Laboratory, University of California
Berkeley, CA 94720-0001 USA**

ABSTRACT

Salt is a unique rock for isolation of nuclear waste because it is "dry" and nearly impermeable. In this paper we summarize some mass-transfer and transport analyses of salt repositories. First we analyse brine migration. Heating by high-level waste can cause brine in grain boundaries to move due to pressure-gradients. We analyze brine migration treating salt as a thermoelastic solid and found that brine migration is transient and localized. We use previously developed techniques to estimate release rates from waste packages by diffusion. Interbeds exist in salt and may be conduits for radionuclide migration. We analyze steady-state migration due to brine flow in the interbed, as a function of the Peclet number. Then we analyze transient mass transfer, both into the interbed and directly to salt, due only to diffusion. Finally we compare mass transfer rates of a waste cylinder in granite facing a fracture and in salt facing an interbed. In all cases, numerical illustrations of the analytic solution are given.

1. Introduction

Salt is the proposed host rock for geologic repositories of nuclear waste in several nations because it is "dry" and probably "impermeable." Although experiments and experience at potential salt sites indicate that salt may contain brine, the low porosity and permeability of salt make it still a good choice for geologic isolation. In this paper we summarize some mass-transfer and transport analyses of salt repositories. Readers should consult our technical reports for details [1,2,3,4].

2. Bedded Salt: Brine Migration

Salt is a unique rock for isolation of nuclear waste because it is "dry" and nearly impermeable. The existence of salt through geologic time is *prima facie* evidence of such properties. If the hypothesis that salt is saturated with brine in grain boundaries [5] is correct, then it is possible to have brine flow due to pressure differences within the salt. Inward brine flow can affect the corrosion rate of nuclear waste containers and outward brine flow can affect transport of radionuclide from waste packages, thus it is paramount to know the magnitude of such flow.

Brine exists in natural salt as inclusions in salt crystals and in grain boundaries. Brine inclusions in crystals move to nearby grain boundaries when subjected to a temperature gradient, because of temperature-dependent solubility of salt. Brine in grain boundaries moves under the influence of a pressure gradient [5,6]. When salt is mined to create a waste repository, brine from grain boundaries will migrate into the rooms, tunnels and boreholes because these cavities are at atmospheric pressure. After a heat-emitting waste package is emplaced and backfilled, the heat will impose a temperature gradient in the surrounding salt that will cause inclusions in the nearby salt to migrate to grain boundaries within a few years, adding to the brine that was already present in the grain boundaries.

The lithostatic pressure of the surrounding salt, augmented by the compressive stresses of heating, causes salt to creep against the waste container. Creep of the salt will result in consolidated salt completely enclosing the waste container within a few years after emplacement of a high-level waste container.

After the consolidation of salt around the waste package, neglecting the consumption of brine by container corrosion, brine in grain boundaries near the waste package can only migrate outward into the surrounding salt, under the influence of pressure gradients caused by transient heating of the salt. Hot salt near the waste package expands against the waste package and surrounding salt, creating high compressive stresses near the waste package and resulting in pressure above the lithostatic pressure. Brine pressure further increases because grain-boundary brine expands more than does the salt and this increased pressure gradient causes brine to flow outward into the cooler salt. Outward flow of brine and salt creep relieve the pressure gradient on the fluid, which finally relaxes to near-lithostatic pressure. If the waste containers are failed by corrosion or cracking, this outward brine movement can become a mechanism for radionuclide transport. To determine the extent to which advection by brine in grain boundaries is an important transport mechanism for released radionuclides, it is necessary to estimate the time-dependent migration of brine after salt consolidation.

Using thermoelastic theory [1,6] we calculate brine migration velocities. We consider a spherical-equivalent spent-fuel waste package of 0.72-meter radius in an infinite salt medium, with initial heat flux at the waste form surface of 928 W/m^2. The parameter values used in the calculations are from McTigue, and shown in Table I.

Figure 1 shows the Darcy brine velocity near a waste package as a function of distance, at 0.1, 1 and 10 years. Pressure-induced brine migration within consolidated salt is a transient phenomenon. After about ten years the velocity is nearly zero. Even the highest velocity is of the order of *millimeters per year*, and occurs only within a meter or so of a waste package. At these Darcy velocities molecular diffusion may be the dominant transport process for radionuclides in salt.

Table I. Salt Properties		
Property	Value	Units
Conductivity	6.60	$W\ m^{-1}K^{-1}$
Heat Capacity	1.89×10^6	$J\ m^{-3}K^{-1}$
Drained Bulk Modulus	20.7	GPa
Fluid Bulk Modulus	2.0	GPa
Solid Bulk Moduli	23.5	GPa
Shear Modulus	12.4	GPa
Porosity	0.001	
Permeability	10^{-21}	m^2
Fluid Expansivity	3.0×10^{-4}	K^{-1}
Solid Expansivity	1.2×10^{-4}	K^{-1}
Fluid Viscosity	1.0×10^{-3}	Pa s
Poisson's Ratio	0.25	
Undrained Poisson's Ratio	0.27	
Fluid Diffusivity	0.16×10^{-6}	$m^2\ s^{-1}$
Thermal Diffusivity	3.5×10^{-6}	$m^2\ s^{-1}$
Source: [6]		

3. Bedded Salt: Fractional Release Rates by Diffusion

Observing that pressure-driven brine migration is transient and localized, we applied previously developed techniques for predicting dissolution rates for water-saturated porous media to repositories in salt. Figure 2 shows the release rates of some low-solubility species, calculated on a congruent release basis. It can be seen that for the parameter values used, the release rates are well below the USNRC limits. For a UO_2 matrix solubility under oxidizing conditions of 50 g/m^3, the release rates are proportionally higher, but still very much below USNRC limits. For readily soluble species we assume at $t = 0$ the metallic container fails and brine contacts the interior of spent fuel, with one percent of the cesium and iodine released instantaneously into brine that has filled void space within spent fuel waste packages. The nuclides then move by diffusion into the rock salt. We assume that the waste package and void geometry is planar and the void volume is 0.45 m^3 per package. Time-dependent fractional release rates, normalized to initial inventories, are shown in Figure 2 for a diffusion coefficient of 10^{-7} cm^2/s, for retardation coefficients of 10 and 1 for Cs and I respectively. The release rates for ^{135}Cs and ^{129}I are below the calculated USNRC limits for all times shown. For ^{137}Cs the calculated USNRC limit is exceeded until about 300 years when practically all of the ^{137}Cs has decayed. However, the presence of a container would in all likelihood prevent this excedent.

4. Interbeds: Steady-State Mass Transport with Flow

While the salt is known to be quite pure in salt domes, bedded salt is interlaced with beds of sediments. Salt may not be water-conducting, but sediment layers would be classical porous media, capable of conducting water. We need to determine whether interbeds in bedded salt constitute a significant pathway for radionuclide migration.

A waste package is in an emplacement hole intersecting an interbed. We study steady-state mass transport from this configuration using three approaches. Neretnieks [7] proposed an approach for calculating the steady-state transport of oxidants to a copper container. We used that approach for calculating steady-state radionuclide migration away from the waste package [3]. We also used a similar but different method of estimating the mass transfer coefficient [8]. Finally, we use the steady-state solution from a transient study [9]. In all cases, a solubility boundary condition is specified at the waste, and transport of the radioactive species into the interbed is caused by flow in the interbed. Figure 3 shows the fractional release rate at the emplacement hole/interbed interface as a function of Peclet of flow in the interbed from three approaches using the parameters of Table II.

Table II. Data for Interbed Calculations	
Radius of waste cylinder	0.31 m
Length of waste cylinder	3.65 m
U-238 inventory	5.4×10^6 g
Crushed salt thickness	3 cm
Crushed salt porosity	0.001
Interbed thickness	1.0 cm
Interbed separation	0.5 m
Interbed porosity	0.01

5. Interbeds: Transient Diffusion from a Waste Cylinder

A more realistic situation is when there is *no* ground-water flow in the interbeds, and diffusion is the only transport mechanism. Here the species is at solubility at the waste cylinder surface, and moves into the interbed and into salt across the entire surface of the waste cylinder by diffusion. We use analytic solutions previously developed [10] for transient diffusion of radioactive species from a waste cylinder intersecting a planar fracture to the problem of diffusion from a waste cylinder intersecting an interbed in a salt repository [4]. For a waste cylinder 0.31 m in radius and 3.65 m long, we calculate the mass flux into the fracture/interbed and directly into the rock/salt. Figure 4 shows the instantaneous mass fluxes of a stable species, from a waste cylinder into salt and granite using the data in Table III.

Table III. Salt and Granite Data used in Comparison

	Porosity	Diffusion coefficient cm^2/s	Retardation coefficient	Solubility g/m^3
Salt	0.001	10^{-7}	20	10^{-3}
Salt Interbed	0.01	10^{-7}	1	10^{-3}
Granite	0.01	10^{-5}	500	10^{-3}
Granite Fracture	1	10^{-5}	1	10^{-3}

Although porosities are higher in the fracture/interbed, the integrated mass transfer from the waste cylinder surface directly into the rock/salt is higher. A more likely scenario is that localized corrosion exposes only some of the waste to contact with the interbed/fracture, and the partly degraded container continues to cover the rest of the waste cylinder, making the mass transfer rate into the interbed/fracture more important. In terms of total mass transfer rates, the product of porosity and diffusion coefficient of salt is approximately

10^{-3} times less than that of granite, and the mass transfer rate to the surrounding salt is almost 10^{-3} times less than that of granite.

6. Conclusions

These examples show that mass transfer analyses are useful tools in safety assessment of salt repositories.

References

1. Hwang, Y., P. L. Chambré, T. H. Pigford and W. W.-L. Lee : *Pressure-driven Brine Migration in a Salt Repository,* Report LBL-25768, 1988.
2. Hwang, Y., W. W.-L. Lee, P. L. Chambré and T. H. Pigford : *Release Rates in Salt by Diffusion,* Report LBL-25767, 1988.
3. Hwang, Y., W. W.-L. Lee, P. L. Chambré and T. H. Pigford : *Mass Transport in Salt Repositories: Steady-State Transport Through Interbeds,* Report LBL-26704, 1989.
4. Hwang, Y., W. W.-L. Lee, P. L. Chambré and T. H. Pigford : *Mass Transport in Salt Repositories: Transient Diffusion into Interbeds,* Report LBL-26703, 1989.
5. Bredehoeft, J. D. : "Will Salt Repositories Be Dry?" *Eos, 69,* (1988), 121.
6. McTigue, D. F. : "Thermoelastic Response of Fluid-Saturated, Porous Rock," *J. Geophy. Res., 91, B9,* (1986), 9533.
7. Andersson, G., A. Rasmuson and I. Neretnieks : *Migration model for the near field, Final Report,* SKBF/KBS Report 82-24.
8. Chambré, P. L., T. H. Pigford, Y. Sato, A. Fujita, H. Lung, S. Zavoshy, and R. Kobayashi : *Analytical Performance Models,* Report LBL-14842, 1982.
9. Kang, C.-H., 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,* (1987), 134.
10. Ahn, J., P. L. Chambré and T. H. Pigford : "Transient Diffusion from a Cylindrical Waste Solid into Fractured Porous Rock," *Radioactive Waste Mangement and the Nuclear Fuel Cycle,* in press.

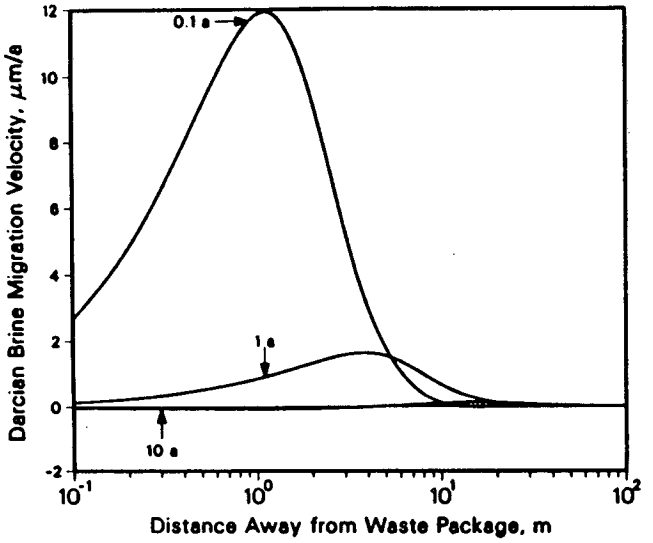


Figure 1. Brine Migration Velocity in Consolidated Salt

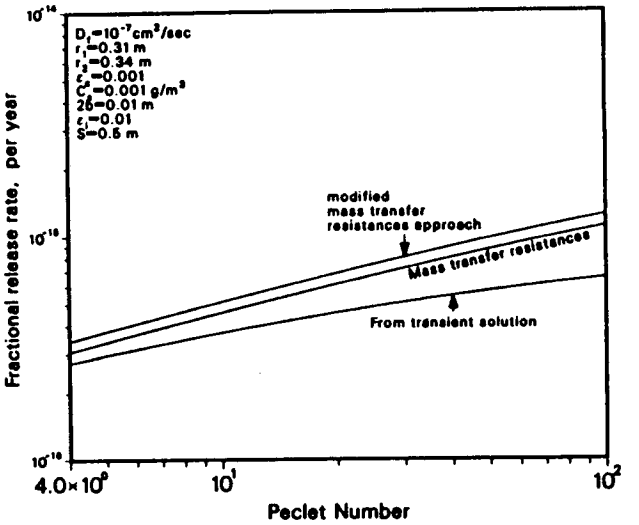


Figure 3. Fractional Release Rates of U-238 by Three Methods

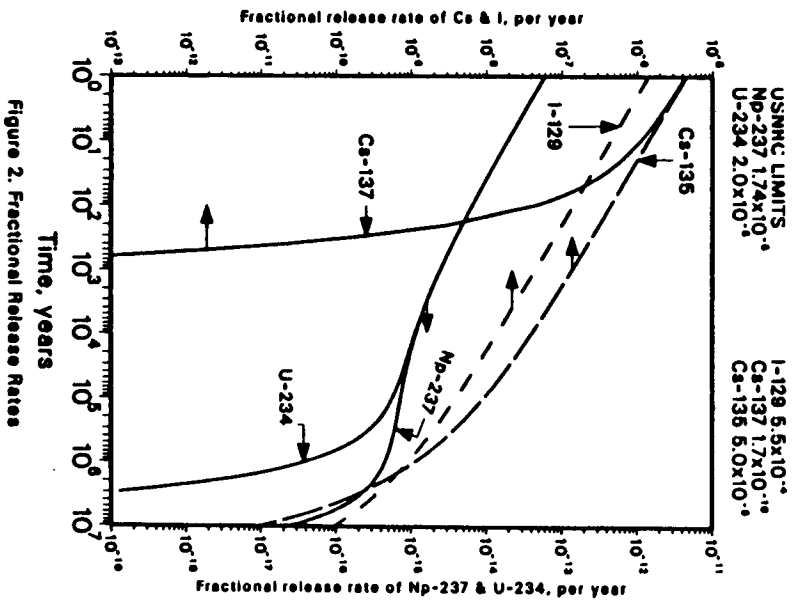


Figure 2. Fractional Release Rates

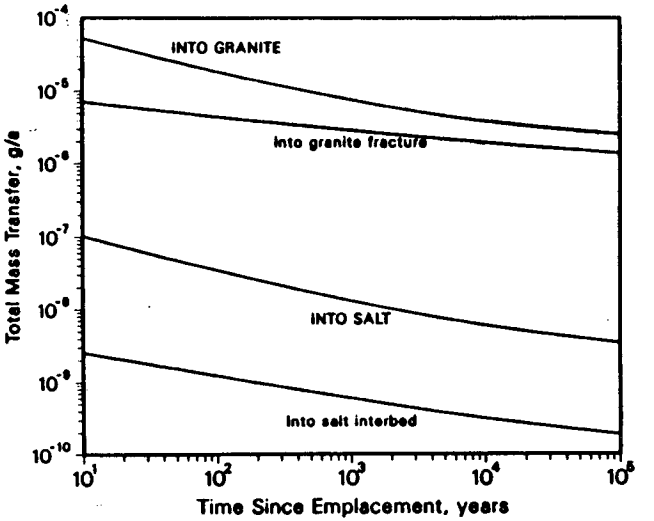


Figure 4. Total Mass Transfer of a Stable Nuclide from a 3.65-m Long Waste Cylinder in Salt and Granite

XBL 898-3131

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