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THE BACKFILL BARRIER AS A COMPONENT IN A
MULTIPLE BARRIER NUCLEAR WASTE ISOLATION
SYSTEM

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ABSTRACT

Quantitative estimates of the potential effectiveness of backfill barriers based on a linear sorption model are presented. Using getters such as clays (known sorbents), a backfill approximately 1-foot-thick can delay by 10^4 to 10^5 years the breakthrough of transuranics. A delay of 10^3 years is possible for major cationic fission products. These delays can be achieved provided that (1) the distribution coefficient (K_d , a measure of affinity for sorbed species) for the barrier material is equal to or greater than 2000 ml/g for transuranics and 200 ml/g for fission products; (2) the interstitial groundwater velocity through the barrier is limited to 1 ft/year or less; (3) the effective porosity of the barrier is equal to or less than 0.1; and (4) the physical integrity of the barrier is maintained (no channels or cracks). Mixtures containing expanding clays such as smectites and other getters are expected to satisfy these criteria.

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CONVERSION FROM CUSTOMARY TO SI UNITS

Customary units rather than SI units have been used where they enhance the communication of magnitudes and facilitate comparisons with previous results. Given below are the appropriate factors for conversions to SI units:

<u>To convert from</u>	<u>to</u>	<u>multiply by</u>
day (sidereal)	s	8.62×10^4
ft (foot)	m	3.05×10^{-1}
ft/day (sidereal)	m/s	3.54×10^{-6}
ft/year (sidereal)	m/s	9.66×10^{-9}
l (liter)	m ³	1.00×10^{-3}
micron	m	1.00×10^{-6}
ton (short)	kg	9.07×10^2
year (sidereal)	s	3.16×10^7

THE BACKFILL BARRIER AS A COMPONENT IN A MULTIPLE BARRIER NUCLEAR WASTE ISOLATION SYSTEM

INTRODUCTION

Backfill materials can be emplaced around a radioactive waste package, completely surrounding the wasteform and its containers (e.g., canister and overpack). It isolates them from the geologic medium of a deep burial repository as shown in the general schematic representation of Figure 1. It may also be packed into drifts and tunnels as shown by the schematic for a mined repository in the same figure. Some authors, for example Jacobson and Pusch (1977) have called engineered materials emplaced immediately around the waste containers "buffer substances."

Groundwater (or brine) is the liquid vehicle for potential leaching and migration of waste species from the wasteform to the geologic medium should the waste containers be breached. The backfill can be designed for low hydraulic permeability to delay the contact of the waste by groundwater or migrating brine, for chemical interactions to decrease the effectiveness of the liquid for leaching waste species from the wasteform, and for both low permeability and strong sorption to delay the breakthrough of any released waste species to the geologic medium. Thus, a backfill material emplaced and maintained as a continuous layer or blanket surrounding waste containers can be designed to be part of an interacting multiple barrier system for radioactive waste isolation.

The backfill barrier concept has a short history. Barr and O'Brien (1976) proposed that radioactive waste containers could be surrounded by radionuclide sorbers or "getters" and that they could be emplaced as mixtures with backfilled solids as powders, or as monolithic hole liners. Jacobson and Pusch (1977) gave their criteria for the properties of suitable "buffer substances" which could be emplaced around waste containers as elements in a Swedish (KBS) conceptual repository design. They suggested the use of bentonite-sand mixtures for that purpose. Guiffre et al. (1979) also recognized the potential effectiveness of a backfill as a physical and sorptive barrier to radionuclide migration.

FIGURE 1
TYPICAL BACKFILL CONFIGURATIONS

GENERAL SCHEMATIC

**SCHEMATIC FOR
MINED REPOSITORY**

Radionuclide sorption and retention measurements on clays and soils have been underway for many years, and a ponderous body of literature has been generated. Although those data are not all directly generalizable to specific backfill barrier applications, they are useful for basic background information. Comparatively little has been reported on measurements and calculations specific to the conditions of deep geologic isolation (Allard et al., 1977 and 1978; Neretnieks, 1977, 1978a and 1978b; Nowak, 1979). It is to the latter area that this paper is directed.

This paper presents calculated estimates of the effectiveness of backfill barriers for delaying the breakthrough of released transuranics and fission products. For this purpose, probable values or ranges of values for system parameters have been estimated from the literature. Breakthrough times were calculated using well-known computational methods for sorption from flowing fluids in packed beds. Considerations of finite mass transport rates as well as sorption equilibrium were included.

PHENOMENOLOGICAL AND MATHEMATICAL MODELS

Phenomenological Model

A one-dimensional model has been chosen for these calculations. At zero time ($t = 0$), a waste species in groundwater moves into the backfill along the direction x . It enters as a sharp step increase in its liquid phase concentration, C , from zero to a constant value, C_0 . As sorption takes place on the getter or sorber, the solid phase concentration of sorbing species, S , increases from zero to an equilibrium value, S^* . The groundwater velocity vector is constant with time and the same everywhere in the bed. That is, the groundwater moves through the bed as a solid plug might move longitudinally (along x) at constant velocity, v_g , through an empty tube. Local velocity and concentration perturbations or discontinuities due to the discreteness of the sorbing backfill particles are treated as sources of longitudinal mixing or dispersion which tend to broaden the initially sharp concentration steps into S-shaped curves. Molecular diffusion in the x -direction also tends to broaden the curves.

Mathematical Model

A material balance around a differential volume element in the bed can be expressed mathematically as follows:

$$\epsilon \frac{\partial C}{\partial t} + \frac{\partial S}{\partial t} + \epsilon v_g \frac{\partial C}{\partial x} - \epsilon D_L \frac{\partial^2 C}{\partial x^2} = 0 \quad (1)$$

- where
- C = liquid phase concentration, quantity of sorbing species per unit volume of liquid.
 - S = concentration of species sorbed on the solid phase, quantity of sorbed species per unit volume of bed (liquid plus solid volumes).
 - ϵ = effective porosity of bed (fraction of bed volume containing flowing liquid).
 - v_g = average interstitial velocity of flowing liquid.

- x = distance in bed along direction of flow and longitudinal diffusion.
 D_L = coefficient of longitudinal dispersion and diffusion combined.
 t = time

The first two terms are the rates of accumulation (or depletion) of sorbing species in the liquid and on the solid per unit volume of bed. The third term is the net transport of liquid phase species by convection. The fourth term is the net transport of liquid phase species by dispersion and diffusion.

The first two terms are related by rate expressions for local mass transport in the liquid and solid phases and an equilibrium sorption isotherm at the interface. It can be shown (Helfferich, 1962; Vermeulen, et al., 1973) that the application of linear sorption isotherm and "linear-driving-force" mass transport rate relation approximations yield the following simplified differential mass balance:

$$\left(1 + \frac{\rho_b K_d}{\epsilon}\right) \frac{\partial C}{\partial t} + v_g \frac{\partial C}{\partial x} - D_L^* \frac{\partial^2 C}{\partial x^2} = 0 \quad (2)$$

- where $\rho_b K_d$ = S^*/C , a linear sorption isotherm (S^* = S at equilibrium with C).
 ρ_b = the bulk packing density of solid sorbent, mass of solid per unit bed volume.
 K_d = the "distribution coefficient" for a linear sorption isotherm, the ratio of quantity of sorbed species per unit mass of solids to quantity of mobile species in the liquid phase per unit volume of liquid.
 and D_L^* = modified coefficient of longitudinal dispersion and diffusion which includes separate terms for local liquid and solid phase mass transport rate effects as well as for dispersion due to finite particle size and for molecular diffusion in the x -direction (Helfferich, 1962).

Solutions of equation 2 are available for boundary conditions that correspond to the chosen phenomenological model. They have been compiled in

tabular and graphical form for the rapid calculation of sorption in packed beds (Helfferich, 1962; Vermeulen et al., 1973).

Equation 2 will be applied to the calculation of breakthrough times for backfill barriers. First, a justifiable range of groundwater velocities will be chosen for the calculation of other significant variables. Then, the linear sorption equilibrium approximation will be justified, and reasonable values for K_d will be chosen. Finally, it will be shown that the effects of local mass transport are negligible for the backfill properties assumed here. That is, they do not contribute significantly to the value of D^*L .

Assumptions and Approximations

1. Groundwater Velocities

For backfills of consolidated or compacted swelling clays such as smectites (See Appendix A), the hydraulic conductivity of the barrier will be very low and will probably limit the flow rate to negligibly small values. In the case of a repository in salt, a disruptive event may cause fissures in the salt or at interfaces between salt and plugging materials which could conduct water or brine until the plastic flow of salt reseals them. In that case, while flow continues, the conductivity of the barrier would probably control the pertinent flow rate.

Alternatively, the barrier may be composed of a loosely packed material which does not swell upon contact with water, such as a kaolin or a soil. Then the flow rate through the barrier may be determined largely by the flow rate of groundwater through the surrounding geologic media or aquifers.

The minimum flow rate of significance is determined by the estimated rate of molecular diffusion in the aqueous phase. If the flow rate through the barrier is small enough, the molecular diffusion of radionuclides, rather than convective transport, will be the predominant mechanism for migration. Bulk diffusion is considered here, but surface diffusion cannot be ruled out as a contributing mechanism. There is a need for research on transport by surface diffusion in repository systems. Transport calculations to be presented later show that aqueous phase molecular diffusion is likely to

predominate (cause much faster migration than that due to convective flow) at interstitial fluid velocities less than 0.1 ft/year. Therefore, 0.1 ft/year was taken as the smallest interstitial average liquid velocity to be considered. Velocities of that order are expected (Kibbe and Boch, 1978) in granite at horizons more than 5000 ft below the surface.

An upper bound on the range of flow rates to be considered here was chosen to be equal to 1000 ft/year so that the range would include most of the flow rates expected or measured for generic or specific repository sites. For example, expected (Kibbe and Boch, 1978) interstitial velocities for groundwater are 0.05 to 50 ft/year in granites, 5 to 50 ft/year in shales, and 10 to 100 ft/year in basalts. Groundwater velocities calculated for NTS (Nevada Test Site) aquifers from on-site measurements (Borg, Stone, Levy and Ramspott, 1976) are in the range of 6 to 60,000 ft/year, with most estimates in the range of 6 to 1000 ft/year. Estimated (Geohydrology Associates, Inc., 1978) rates of groundwater movement through the aquifers of the Rustler Formation in the Los Medanos area near Carlsbad, N.M. (location of the Waste Isolation Pilot Plant) are within an order of magnitude of 1 ft/day. The Rustler Formation is located approximately 1000 ft above the planned waste storage horizon of the Waste Isolation Pilot Plant, and has been assumed as a potential source of intruding groundwater. The bedded salt itself is well known to be slowly self-healing and impermeable under the existing lithostatic pressure.

Groundwater velocities, then, are taken to be within the range of 0.1 to 1000 ft/year.

2. Linear Sorption Isotherm

The transuranics are expected to have extremely low solubilities. Therefore, they are unlikely to saturate the getter material in the backfill, and a linear sorption isotherm is a reasonable first model for these calculations. It is recognized, as stated later, that equilibrium sorption may be inadequate as a complete model for transuranics.

Cesium, strontium, and other fission products are more soluble. Their concentrations in the groundwater could be determined by their release rates and the groundwater flow rate.

Assuming that fission products would be released solely by dissolution of the wasteform (i.e., ignoring the contribution of diffusion in the wasteform), the total concentration of all fission products was estimated to be less than 10^{-4} M. That concentration was considered low enough to use a linear equilibrium model as a reasonable first approximation for the fission products. It was calculated using a crude geometric model and a cylindrical glass wasteform 3 m long and 0.3 m in diameter with 100 as a surface roughness factor. Some other chosen values are 10^{-7} g/cm² x day for an average dissolution rate in water at 25 C (Kibbe and Boch, 1978), a fission product oxide content of 11 wt% for high level waste glass from spent irradiated power reactor fuel reprocessing (Bradley, 1978), and 0.03 m/year for a minimum groundwater flow rate. The use of an engineered multiple barrier system could decrease the dissolution rate substantially. Therefore, 10^{-4} M is probably a maximum value.

A linear equilibrium relationship is characterized by a quantity, K_d , which is the constant ratio of the concentration in the solid phase (getter plus inerts) to the concentration in the liquid phase (flowing water) at equilibrium. It is called a distribution coefficient. There is no parameter in radionuclide migration studies which seems to have been the focus of more controversy, uncertainty, and misunderstanding. K_d is not a single equilibrium constant. Rather, it is a function of one or more equilibrium constants and the chemical potentials for some of the dissolved species. It is nearly constant only under special conditions and for single uptake mechanism and chemical reaction. The value calculated for K_d from a set of data for a single radionuclide-solid phase pair can be a function of the concentrations of all species in solution, including radiolysis products. It can be changed by changes in pH, Eh, the quantity of dissolved CO₂, total dissolved ions and their charge and molecular weights, extent of competing reactions, and trace impurity concentrations in both phases. Consequently, literature values span a wide range. A sample of the literature and literature values is given in Borg, et al. (1976, Part 2). Although values of K_d are given for a wide variety of systems, seldom is it firmly established that the uptake mechanism is ion exchange for which linear equilibrium is a reasonable approximation. There is often doubt as well about the

composition and chemistry of the sorption sites in the solid phase where uptake occurs. With these cautions in mind, values of K_d have been chosen for calculated estimates of getter performance. It must be recognized that, as it is used here, K_d is an empirical parameter rather than a basic chemical property of the system.

Values of K_d equal to 200 ml/g and 2000 ml/g have been chosen to bracket the range of values that are expected to be achievable for radioactive waste migration in backfill barriers. The larger value, 2000 ml/g, has been shown to be achievable with clays for the transuranic cations in concentrated brines and in ground waters. This value is well within the span of pertinent literature values and was established in getter studies for the WIPP using Eu as a simulant for the transuranics (Nowak, 1978). The lower value is probably achievable for Cs, Sr, and most of the other fission products. In the case of concentrated brines, the achievement of 200 ml/g for K_d may require further development of special materials which have shown promise in laboratory tests (Doch, 1978). In some specific cases, 2000 ml/g may be achievable also for the fission products. Potential getters for pertechnetate, iodide and iodate anions have also been found (Fried et al., 1978; Couture, Seitz and Steindler, 1979).

3. Fast Local Mass Transport

Helfferich (1962) and Vermeulen, et al. (1973) have given rapid methods to estimate the relative contributions to D_L^* from local mass transport rates as well as longitudinal dispersion processes. These methods require values for several system variables.

A practical backfill bed length or layer thickness in the direction of flow, h , was taken to be equal to 1 ft. The bed was assumed to be packed to a bulk density, ρ_b , of 2 g/ml with particles that could have an average diameter, d_p , in the range of 5×10^{-4} to 5×10^{-2} cm. The maximum value for this range corresponds to the approximate size of particles passing through 32 mesh or 500 micron screens and is anticipated to be a convenient particle size for a crushed and sized backfill material. The minimum value is small enough so that longitudinal dispersion would be negligible for that particle size. The fluid phase diffusivity, D_f , was taken to have an

average value equal to $10^{-5} \text{ cm}^2/\text{s}$. That is a well known order-of-magnitude estimate for the liquid phase diffusivities of dissolved ions in aqueous solutions at room temperature. It is approximately equal to the value of $2 \times 10^{-5} \text{ cm}^2/\text{s}$ used by Neretnieks (1978a) for cations in his clay barrier calculations for a Swedish repository. A value of $10^{-7} \text{ cm}^2/\text{s}$ was taken for the particle phase diffusivity, D_p , of the migrating and absorbing species. That value is representative of tabulated values for particle phase diffusivities in ion exchangers (Vermeulen, Klein, and Hiester, 1973). It is also comparable to the value measured by Neretnieks (1978a) for compacted montmorillonite clay.

All of the values assumed or estimated for the physical and chemical properties of the backfill are summarized in Table I.

TABLE I
Values of Assumed or Estimated Physical and
Chemical Properties for the Backfill

Distribution Coefficient	$K_d = 200 \text{ to } 2000 \text{ ml/g}$
Bulk Density	$\rho_b = 2 \text{ g/cm}^3$
Bed Length (backfill thickness)	$h = 1 \text{ ft or } 3 \text{ ft}$
Average Particle Size	$d_p = 5 \times 10^{-4} \text{ to } 5 \times 10^{-2} \text{ cm}$
Particle Phase Diffusivity	$D_p = 10^{-7} \text{ cm}^2/\text{s}$
Liquid Phase Diffusivity	$D_L = 10^{-5} \text{ cm}^2/\text{s}$

Following the relationships given by Vermeulen et al (1973) and using their Figure 16-17, it was determined that local mass transport to and through the sorbing particles should not contribute significantly to D^*_L , the modified coefficient of diffusion and dispersion.

Then,
$$D^*_L = D_L$$

Mathematical Description for Calculations

Convection, longitudinal molecular diffusion, and longitudinal dispersion remain as the significant processes which determine breakthrough for the chosen set of parameters. These processes are described by the following modification of equation 2:

$$\frac{\partial C}{\partial t} + \frac{v}{R_f} \frac{\partial C}{\partial x} - \frac{D_L}{R_f} \frac{\partial^2 C}{\partial x^2} = 0 \quad (3)$$

where $R_f = \left(1 + \frac{\rho_b K_d}{\epsilon}\right)$, the well-known retardation factor.

When the interstitial velocity, v , and the particle size, d_p , are small enough, convection and dispersion are small compared with molecular diffusion. Longitudinal transport is by diffusion alone. In that case, equation 3 becomes

$$\frac{\partial C}{\partial t} - \frac{1}{\sqrt{2}} \frac{D_f}{R_f} \frac{\partial^2 C}{\partial x^2} = 0 \quad (4)$$

where D_f = the liquid phase molecular diffusivity

and $\frac{1}{\sqrt{2}}$ = a tortuosity factor to account for the tortuous diffusion path through the porous bed.

The boundary condition is

$$C = C_0, \quad x = 0, \quad t > 0.$$

The initial condition is

$$C = 0, \quad x > 0, \quad t = 0.$$

Crank (1956) gives the following solution for equation 4:

$$\frac{C}{C_0} = 1 - \operatorname{erf} \left\{ \frac{x}{2 \left(\frac{D_f t}{\sqrt{2} R_f} \right)^{1/2}} \right\} \quad (5)$$

CALCULATED BREAKTHROUGH TIMES

The time to breakthrough is defined here as the value of time, t_b , for which $C/C_0 = 0.01$ at $x = h$, the length of the bed. That is, it is the time required for the concentration of waste species in the liquid at the outer surface of the backfill to reach one percent of the value of the concentration at the inner surface (at the interface with the waste package).

The solutions and methods of Helfferich (1962) and Vermeulen et al. (1973), Crank's (1956) equation 5, and the values in Table I, were used to calculate the breakthrough times given in Table II.

At interstitial groundwater velocities equal to 0.1 and 1.0 ft/year, transport is mostly by diffusion. The convection and dispersion terms are small. Therefore, the estimated breakthrough time, t_b , was calculated using equation 5 for diffusion alone. The breakthrough time would be approximately the same for any velocity less than 1.0 ft/year. These are the greatest breakthrough delays that the backfill can be expected to achieve for the chosen range of distribution coefficients and porosities.

At interstitial groundwater velocities equal to and greater than 10 ft/year, convection was found to be significant, and the methods given by Helfferich (1962) and by Vermeulen et al. (1973) were used. For $v_g = 10$ ft/year, both diffusion and dispersion contribute to D_L , that is, to broadening of the moving concentration step. The effect of this broadening is to cause breakthrough ($C/C_0 = .01$) in approximately half the time that would be predicted for an unbroadened sharp step. For $v_g \geq 100$ ft/year, dispersion is the predominant contribution to D_L , and the resulting broadening causes a decrease in the breakthrough time of only approximately 25 percent.

These results illustrate the importance of minimizing the interstitial liquid velocity. The backfill barrier can be highly effective, delaying the breakthrough of transuranics for 10^4 to 10^5 years, if the interstitial velocity is less than 1.0 ft/year. The backfill would be far less effective if it allowed an interstitial velocity equal to 1000 ft/year. In that case, breakthrough would be delayed by only a few tens or hundreds of years.

TABLE II

Times to Breakthrough from 1.0 ft (0.3 m) and 3.0 ft (0.9 m) Thick
Backfilled Getter Bed for Selected Values of ϵ , R_d ,
and Interstitial Groundwater Velocity and for Linear Sorption

$$h = 0.3 \text{ m}, \rho_b = 2 \text{ g/cm}^3, C/C_0 = 0.01$$

ϵ	R_d , ml/g	R_f	Interstitial Groundwater Velocity, v_g ft/year	<u>Times to Breakthrough, t_b</u>	
				1.0 ft Long (0.3m) Getter Bed, t_b , years	3.0 ft Long (0.9m) Getter Bed, t_b , years
0.1	2000	40,000	0.1	1×10^4	1×10^5
			1.0	1×10^4	2×10^4
			10.0	2×10^3	8×10^3
			100.0	3×10^2	1×10^3
			1000.0	3×10^1	1×10^2
0.01	2000	400,000	0.1	1×10^5	1×10^6
			1.0	1×10^5	2×10^5
			10.0	2×10^4	8×10^4
			100.0	3×10^3	1×10^4
			1000.0	3×10^2	1×10^3
0.1	200	4,000	0.1	1×10^3	1×10^4
			1.0	1×10^3	2×10^3
			10.0	2×10^2	8×10^2
			100.0	3×10^1	1×10^2
			1000.0	3×10^0	1×10^1
0.01	200	40,000	0.1	1×10^4	1×10^5
			1.0	1×10^4	2×10^4
			10.0	2×10^3	8×10^3
			100.0	3×10^2	1×10^3
			1000.0	3×10^1	1×10^2

There is evidence that the longest times to breakthrough in Table II are realistic estimates for a practical backfill barrier. Known materials such as the smectite group (Grim, 1968) of expanding clays (e.g. hectorite or montmorillonite in bentonite) can limit interstitial groundwater velocities to less than 0.1 ft/year when they are confined by the lithostatic pressure existing at the depth of the waste in the repository (Pusch, 1978). These materials also flow to conform to the voids that contain them, thus tending to heal cracks and maintain an integral barrier (Pusch, 1978). See Appendix A for a general description of the smectite group of minerals. Values of K_d equal to 2000 ml/g for the transuranics and 200 ml/g for Cs and Sr are considered achievable by these and other materials, as stated earlier. Potential getters for anions have also been found (Fried et al., 1978; Couture, Seitz and Steindler, 1979; Lynch and Dosch, 1979).

Then, a delay of 10^4 to 10^5 years is possible for the transuranics. That would be a large contribution to the overall effectiveness of a waste isolation system. A delay of approximately 10^3 years would be possible for major fission products. That is sufficient time for most of the fission products to decay to negligible concentrations. Neretnieks (1978) also estimated 10^3 years to breakthrough for Cs in a 1-ft-long bed of bentonite and sand. Containing appropriate combinations of getters and other reagents, backfill barriers can be effective and practical.

The effectiveness of a backfill barrier can be increased by increasing its thickness around the waste as also shown quantitatively in Table II. At $v_g = 0.1$ ft/year, diffusion predominates as before, and the breakthrough time is increased with the square of the thickness. At values of v_g from 1 to 100 ft/year, dispersion and diffusion broaden the wave moving by convection. At 1000 ft/year, only dispersion and convection are significant, and breakthrough time is increased approximately directly with the thickness.

CONCLUSIONS

A backfill barrier containing one or more getter materials and reagents can be an effective component in a multiple barrier approach to radioactive waste isolation. Using known getter materials, a backfill approximately one foot thick, physically unbreached, can delay by 10^4 to 10^5 years the breakthrough of transuranics which may be released from the wasteform should groundwater intrude, breach the canister, and leach the wasteform. A delay of approximately 10^3 years is possible for major cationic fission products. In concentrated brines such as those which have been assumed as intrusive fluids in safety assessments for the Waste Isolation Pilot Plant (WIPP), special getter materials (e.g., titanates) may also be desirable for the absorption of some of the low valence cationic fission products (e.g., strontium). Useful anion absorbers for iodine and technetium could be developed further from among known anion absorbing minerals for inclusion in the backfill.

The effectiveness of a backfill barrier depends on the distribution of interstitial aqueous flow rates within the backfill. Effectiveness can be high if the interstitial velocity is relatively uniform and less than 0.1 ft/year, or it can be virtually negligible if the interstitial velocity is greater than 1000 ft/year. Therefore, providing a nearly homogeneous fluid flow barrier can be an important backfill barrier function.

Smectite clays swell and have low liquid permeabilities when they are wetted by water or brine. Thus they are useful as components which can make the backfill an effective flow barrier. In addition, they are good sorbers of most waste species.

REFERENCES

Allard, B., Kipatsi, H., and Rydberg, J., Sorption of Long-Lived Radionuclides in Clay and Rock: Part 1, Determination of Distribution Coefficients, KBS Teknisk Rapport, 55, Stockholm, 1977.

Allard, B., Kipatsi, H., and Torstenfelt, B., Sorption av Langlivade Radionuclider i Lera och Berg: Part 2, KBS Teknisk Rapport, 98, Stockholm, 1978.

Barr, G. E., and O'Brien, P. D., Personal Communication and Invention Disclosure to Sandia Laboratories and the United States Department of Energy (DOE), March 1976.

Borg, I. Y., Stone, R., Levy, H. B., and Ramsdell, L. D., Information Pertinent to the Migration of Radionuclides in Ground Water at the Nevada Test Site, Part 1, Review and Analysis of Existing Information, UCRL-52076 Part 1, May 1976.

Bradley, D. J., Leaching of Fully Radioactive High-Level Waste Glass, PNL-2664/UC-70, September 1978.*

Couture, R. A., Seitz, M. G. and Steindler, M. J., Adsorption of Iodate by Hematite, Argonne National Laboratory, presented at the ANS Annual Meeting, Atlanta, Georgia, June 1979.

Crank, J., The Mathematics of Diffusion, pp. 18-19 and 121-122, Oxford Press, London, 1956.

Dosch, R. G., The Use of Titanates in Decontamination of Defense Waste, SAND78-0710, June 1978.*

Fried, S., Friedman, A. M., Cohen, D., Hines, J. J. and Strickert, R. G., The Migration of Long-Lived Radioactive Processing Wastes in Selected Rocks, Annual Report to the Office of Waste Handling Project AN0115A, ANL-78-46, Argonne National Laboratory, Argonne, Illinois, April 1978.

Geohydrology Associates, Inc., Ground-Water Study Related to Proposed Expansion of Potash Mining near Carlsbad, New Mexico, for Bureau of Land Management, Denver, CO, Contract No. YA-512-CT7-217, June 1978.*

Grim, R. E., Clay Mineralogy, Second Edition, McGraw-Hill, New York, 1968.

Guiffre, M. S., Koplick, C. M., Plum, R. L., and Talbot, R., Information Base for Waste Repository Design; Vol. 5: Decommissioning of Underground Facilities, NUREG/CR-0495, TR-1210-1, 1979.*

Welferich, F., Ion Exchange, pp. 447-480, McGraw-Hill, New York, 1962, available as an authorized facsimile of the original book, University Microfilms International, Ann Arbor, Michigan, 1977.

Jacobson, A., and Pusch, R., Deposition of High-level Radioactive Waste Products in Bore-Holes with Buffer Substance, KBS Teknisk Rapport 03, Stockholm, 1977.

Kibbe, R. K. and Boeh, A. L., Technical Support for GEIS: Radioactive Waste Isolation in Geologic Formations, Vol. 21, Ground Water Movement and Nuclide Transport, Y/OWI/TN-36/21, April 1978.*

Lynch, A. W. and Dorch, R. G., Interaction of Radionuclides with Geomedia from the Nevada Test Site, in; Scientific Basis for Nuclear Waste Management, 2, C. J. Northrup (Ed.), Plenum Press, 1980.

Neretnieks, I., Retardation of Escaping Nuclides from a Final Repository, KBS Teknisk Rapport 30, Stockholm, 1977.

Neretnieks, I., Transport of Oxidants and Radionuclides through a Clay Barrier, KBS Teknisk Rapport 79, Kungl Tekniska Hogskolan Stockholm, February 20, 1978a.

Neretnieks, I., and Skagius, C., Diffusivity Measurements in Wet Compacted Clay Na Lignosulfonate, Sr⁺⁺, Cs⁺, KBS Teknisk Rapport 87, Stockholm, 1978b.

Nowak, E. J., The Migration of Eu through Geologic Media, presented at the 1978 Fall Meeting of the American Geophysical Union, San Francisco, December 1978; and abstracted in Transactions, American Geophysical Union, Vol. 59, No. 12, p. 1224, December 1978.

Nowak, E. J., The Backfill as an Engineered Barrier for Nuclear Waste Management, in: Scientific Basis for Nuclear Waste Management, 2, C. J. Northrup (Ed.), Plenum Press, 1980.

Vermeulen, T., Klein, G., and Hiester, N. K., Adsorption and Ion Exchange, Section 16 in Chemical Engineers' Handbook, Fifth Edition, Perry, R. H., and Chilton, C. H., editors, McGraw-Hill, New York, 1973.

APPENDIX A

Brief Description of Smectite Clays

Following Grim (1968), we have used "smectite" as a classifying name for a group of clay minerals. There is a difference of opinion concerning the use of "montmorillonite-saponite" instead of "smectite," and one often hears "montmorillonites" also used for group classification.

Nevertheless, we used "smectite" to name a group of crystalline, three-layer, expanding lattice, aluminosilicate clay minerals. The outstanding feature of the smectite structure is that water and other polar molecules can enter between the unit layers, causing the lattice to expand. Exchangeable cations, such as Na^+ , Ca^{++} , H^+ occur between the silicate layers to restore the charge balance that is always disrupted in smectites by lattice substitutions such as Mg^{++} for Al^{3+} or Al^{3+} for Si^{4+} . Some members of the smectite group are montmorillonite, beidellite, nontronite, hectorite, and saponite.

Extensive information on the smectites can be found in Grim (1968).