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THE BACKFILL AS AN ENGINEERED BARRIER FOR NUCLEAR
 WASTE MANAGEMENT*

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ABSTRACT

This paper presents results from an experimental backfill barrier development program. The swelling, plastic flow, and relative impermeability of bentonite and hectorite were observed and measured after wetting with concentrated brines. Measurements of stable values of pH > 6.5 for the interstitial brines in wetted bentonite and hectorite confirmed conditions favorable for precipitation and sorption of transuranics. Values of $K_d > 2000$ ml/g were measured for Pu and Am. Calculated estimates of the effectiveness of a one-foot-thick backfill barrier are presented. They show that the breakthrough of Pu and other transuranics ($K_d = 2000$ ml/g) can be delayed for 10^4 to 10^5 years. The breakthrough of most fission products ($K_d = 200$ ml/g) can be delayed for 10^3 to 10^4 years, sufficient time for them to decay to very low concentrations. A backfill barrier can contribute significantly to a radioactive waste isolation system.

INTRODUCTION

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 concept for radioactive waste isolation. Barr and O'Brien (1) proposed that radioactive waste containers can be surrounded by radionuclide adsorbers or "getters" and that they could be emplaced as mixtures with backfilled solids,

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as powders, or as monolithic hole liners. Jacobson and Pusch (2) 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. (3) also recognized the potential effectiveness of a backfill as a physical and sorptive barrier to radionuclide migration.

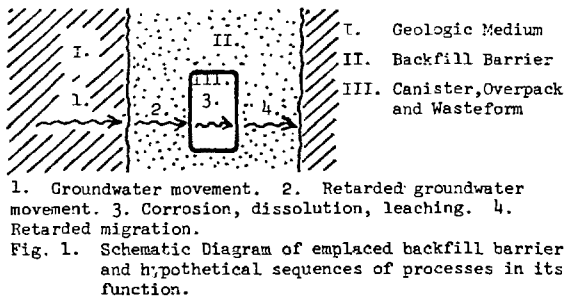
Radionuclide sorption and retention measurements on clays and soils have been underway for many years, and a ponderous body of literature has been generated. However, those data are not readily generalizable to specific backfill barrier applications. Comparatively little has been reported on measurements and calculations specific to the conditions of deep geologic isolation (4,5,6,7,8). It is to the latter area that this paper is directed.

A schematic representation of a backfill barrier is shown in Fig. 1. Groundwater, including brine which may be present in bedded salt, is the vehicle for potential waste migration. Capacity for sorption of waste species, low hydraulic permeability, adequate thermal conductivity, adequate plasticity to resist fracture, and sufficient support strength for waste containers are among the important properties which contribute to the effectiveness of a backfill as a barrier to radioactive waste migration. The backfill may also be designed to modify the groundwater chemically for decreased corrosion of the overpack and canister and decreased leaching of the wasteform. A more detailed description of this system and migration variables is given elsewhere (9).

A backfill barrier is best designed and analyzed as a part of a specific radioactive waste isolation system. The maximum migration velocity of radioactive waste species in a backfill barrier is a measure of its effectiveness. The migration velocity is determined by the sorption and mass transport properties of the backfill and the groundwater. Interactions among the groundwater, wasteform, canister, overpack, backfill and host geologic formation under the thermal, radiation, and lithostatic pressure conditions in a specific repository system can determine those pertinent sorption and transport properties.

SORPTION MEASUREMENTS

The capacity to sorb waste species is a crucial property of getters or sorbers for use in backfill barriers. Measurements of the batch sorption of ^{152}Eu by clays, soils, and a zeolite have been made. ^{152}Eu dissolved in nearly saturated brines was used to simulate transuranics for the proposed Waste Isolation Pilot Plant (WIPP) near Carlsbad, New Mexico. Brine A is representative of groundwater in contact with potash-containing layers above the



proposed waste horizon, and brine B represents groundwater in contact with the halite of the proposed waste horizon itself. Nominal compositions of brines A and B are given in Table I.

Sorption measurements were made by contacting brine containing ^{152}Eu and a solid sorbent in a continuously agitated vessel. Values of pH were controlled by additions of aqueous HCl or NaOH. Control samples of Eu-containing brine alone (sorbent-free) were included for each set of experimental conditions to correct the results for Eu losses not attributable to the sorbent. All samples, including the controls, were filtered through 0.8 μm pore size filter media before gamma-counting. Some of the getter materials were heated in air at 300°C for 6 hours and cooled prior to the room temperature sorption measurements. This heat treatment was done to test for the effect of a dry thermal cycle on clay properties.

Screening results are given in Table II as empirical distribution coefficients, K_d . It was shown previously (10) that the liquid phase precipitation of Eu-containing solids from brines A and B is unlikely at $\text{pH} \leq 5.5$. At $\text{pH} = 5.5$, the values of K_d for all of the materials were nearly the same except for montmorillonite in brine B. At $\text{pH} = 6.5$, there was greater variability among values of K_d , possibly because small changes near $\text{pH} = 6.5$ caused large changes in precipitation. Nevertheless, the values of K_d were consistently larger at $\text{pH} = 6.5$ than at $\text{pH} = 5.5$, and no large decreases in K_d resulted from heating the getters at 300°C. These results show that K_d values in the approximate range of 10^3 to 10^4 are achievable for all of the materials tested and that exposure to a dry thermal cycle from high level waste would not adversely

Table I. Representative Brine Compositions

| | Major Constituents, Molarity | | | | | | | |
|---------|------------------------------|----------------|------------------|------------------|-----------------|------------------------------|-------------------------------|--------------------------------|
| | Na ⁺ | K ⁺ | Mg ⁺⁺ | Ca ⁺⁺ | Cl ⁻ | SO ₄ ⁻ | HCO ₃ ⁻ | BO ₃ ⁻⁻⁻ |
| Brine A | 1.8 | 0.8 | 1.4 | 0.02 | 5.4 | 0.04 | 0.01 | 0.02 |
| Brine B | 5.0 | --- | --- | 0.03 | 5.0 | 0.04 | --- | --- |

affect their sorption properties.

Although increased K_d with increased pH is consistent with sorption by cation exchange, other evidence strongly contradicts that mechanism. Table III presents such evidence for sorption at pH = 5.5 in dilute and concentrated brines. There was little change in K_d for montmorillonite and hectorite with a two-orders-of-magnitude change in the competing ion concentration (virtually all Na⁺ for brine B). It was concluded that the sorption mechanism for montmorillonite and hectorite is more complex than simple ion exchange. Neretnieks (6) reported a similar conclusion for natural zeolites. The same conclusion was reached for the soils in these tests. However, the results for Zeolon are consistent with a simple ion exchange mechanism at pH = 5.5.

SELECTION AND PROPERTIES OF BENTONITE AND HECTORITE

The smectite swelling clays (11) montmorillonite and hectorite have physical properties which make them good candidates for backfill barrier components. When wetted, they swell and flow plastically to fill and seal voids, they are essentially impermeable, and they have adequate thermal conductivity and support strength when mixed with sand (12). Swelling to nearly double their initial volume and subsequent impermeability were verified experimentally for brines A and B at atmospheric pressure. Measurements of pH \approx 6.5 for the interstitial brine confirmed conditions favorable for precipitation and large values of K_d . Finally, initial measurements yielded \sim 0.5 W/mK for the thermal conductivities of dry clay-sand mixtures at atmospheric pressure.

Because of their favorable properties, commercially available bentonite (containing montmorillonite) and hectorite mixed with sand were chosen as basic backfill materials for further investigation. These materials establish physical properties for the backfill and a pH range for the interstitial brine or groundwater. They also act as getters for transuranic nuclides. Some preliminary batch sorption measurements with 10 wt% bentonite, 90 wt% sand mixtures have yielded K_d >2000 ml/g for Pu in brine B. Other

Table II. Batch Equilibration Results for $^{152}\text{Eu}^{3+}$ Sorbate at Room Temperature

$$C_0 = \sim 2 \times 10^{-7} \text{ M } \text{Eu}^{3+} \quad (\sim 0.1 \mu\text{Ci/ml})$$

K_d , ml/g

| Getter | Brine A | | | Brine B | | |
|-------------------------------|---------|--------|---------|---------|--------|---------|
| | pH=5.5 | pH=6.5 | | pH=5.5 | pH=6.5 | |
| | ----- | ----- | Heated* | ----- | ----- | Heated* |
| DLR(soil)** | 200 | 1600 | 1800 | 270 | 14000 | 7300 |
| Caliche | 140 | 8000 | 9000 | 220 | | 11000 |
| Tuff | 200 | 2500 | | 200 | 1400 | |
| Zeolon(Zeolite) | 50 | 690 | 270 | 60 | 6000 | 1400 |
| Montmorillonite (SWy-1)*** | 100 | 850 | 1100 | 6700 | 1300 | 3500 |
| Hectorite (SHCa)*** | | | 5500 | | | 7200 |
| Kaolin(DGa-1) | 60 | 1100 | | 200 | 1600 | |

*Heated 6 hrs in air at 300°C before sorption measurement at room temperature.

**Dewey Lake Redbeds, an outcropping in the Los Medanos area, Carlsbad, New Mexico.

***Samples from Source Clay Mineral Repository, University of Missouri, Columbia, MO

Table III. Sorption at Different Competing Ion Concentrations

$\text{pH} = 5.5, C_0 = \sim 2 \times 10^{-8} \frac{\text{M}}{\text{Eu}^{3+}} (\sim 0.1 \mu\text{Ci/ml})$

| | $K_d, \text{ml/g}$ | |
|-----------------|---------------------|--------------------------------------|
| | <u>100% Brine B</u> | <u>1% Brine B in Deionized Water</u> |
| Montmorillonite | 500 | 1000 |
| Hectorite | 3400 | 2600 |
| Zeolon | 12 | 25000 |

backfill components (13) may be required for getters of fission products in the presence of concentrated brines. Potential getters for pertechnetate, iodide, and iodate anions have been reported (14,15,16).

BACKFILL BARRIER PERFORMANCE ESTIMATES

Estimated breakthrough times for migrating waste species in backfill barriers free of cracks or channels were calculated. Breakthrough was defined as the appearance of a migrating species at ~1% of its initial concentration. A fixed bed model with sorption by linear equilibrium ion exchange was used as a first empirical approximation. Calculation techniques given by Vermeulen et al. (17) and Hefferich (18) were used for migration by convective transport. Relationships from Crank (19) were used for migration predominantly by diffusion. Details of these calculations are given elsewhere (9).

Values for K_d , the effective porosity ϵ , and the interstitial groundwater velocity v_g were chosen to bracket the appropriate ranges. A value of $K_d = \sim 2000 \text{ ml/g}$ has been shown to be achievable for Pu in concentrated brine, and it is realistic for other groundwaters (20). $K_d = 200 \text{ ml/g}$ is attainable for Sr in brine (13) and Cs in dilute aqueous solutions (20). Values of ϵ in the range of 0.01 to 0.1 are conservative estimates for clays (21). At interstitial groundwater velocities less than 0.1 ft/year, migration is predominantly by molecular diffusion and essentially independent of interstitial velocity. An upper bound of 1000 ft/year includes most of the interstitial velocities predicted or measured for generic or specific repository sites (20,21).

Calculated estimates for breakthrough times are given in Table IV. They are in the range of 10^4 to 10^5 years for Pu ($K_d = 2000 \text{ ml/g}$) and other transuranics. For $K_d = 200 \text{ ml/g}$, breakthrough times are in the range of 10^3 to 10^4 years, sufficient time for most of the fission products in high-level waste to have decayed to very low concentrations. The significant effects of interstitial groundwater velocity, K_d , and ϵ are also illustrated. These estimates show that backfill barriers are potentially effective contributions to the isolation of radioactive wastes in the presence of concentrated brines and other groundwaters.

Table IV. Times to Breakthrough for a One Foot Thick Backfill Barrier Having a Bulk Density of 2 g/cm³

| ϵ | K_d , ml/g | Interstitial Groundwater Velocity ft/year | Calculated Time to Breakthrough, years |
|------------|-----------------|---|--|
| 0.1 | 2000 | 0.1 | $\sim 10^4$ |
| | | 1.0 | $\sim 10^3$ |
| | | 10.0 | 2×10^3 |
| | | 100.0 | 4×10^2 |
| | | 1000.0 | 4×10^1 |
| 0.01 | 2000 | 0.1 | $\sim 10^5$ |
| | | 1.0 | $\sim 10^4$ |
| | | 10.0 | 2×10^4 |
| | | 100.0 | 4×10^3 |
| | | 1000.0 | 4×10^2 |
| 0.1 | 200 | 0.1 | $\sim 10^3$ |
| | | 1.0 | $\sim 10^2$ |
| | | 10.0 | 2×10^2 |
| | | 100.0 | 4×10^1 |
| | | 1000.0 | 4×10^0 |
| 0.01 | 200 | 0.1 | $\sim 10^4$ |
| | | 1.0 | $\sim 10^3$ |
| | | 10.0 | 2×10^3 |
| | | 100.0 | 4×10^2 |
| | | 1000.0 | 4×10^1 |

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