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Bentonite-Protected Waste Canister**

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STEADY RADIONUCLIDE RELEASE RATES FROM A BENTONITE-PROTECTED WASTE CANISTER

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ABSTRACT

A bentonite-based packing or engineered barrier is one ingredient of the multibarrier concept for underground storage of high-level nuclear waste. This paper investigates the possible effectiveness that such a smectite-laden barrier might have in controlling steady, solubility-limited release rates from a cylindrical waste canister.

Darcy's law and the convective-diffusion equation in two dimensions are solved numerically with and without an annulus of packing material. Reduced fractional release rates quantifying the role of the packing are studied over wide ranges of ground water velocities, packing amounts, and packing permeability and effective diffusivity. Release rates are shown to be strongly dependent on the physical properties of the packing, particularly the effective diffusivity.

Although the low permeability of a bentonite-rich packing negates convection next to the canister, calculated release rates of the packing-protected waste canister do not differ dramatically from the those for the unprotected canister, except at high ground water velocities. To afford more protection, consideration must be given to reducing the porosity of the packing material.

INTRODUCTION

Design of high-level nuclear waste repositories in geologic media relies on minimizing the rate at which hazardous radionuclides might be released into the accessible environment. To aid in reducing release rates, it is proposed that a bentonite-based packing or engineered barrier surround each waste canister.

At early times, during the transient regime, the smectite-rich packing inhibits water imbibition thereby delaying the onset of canister corrosion [1]. Also, once the canister is breached, the favorable ion-exchange properties of montmorillonite retard the migration of cations into the host rock [6]. Further discussion of transient cation diffusion in compacted montmorillonite is found in our companion paper [4].

In a mature repository approaching steady conditions, the low hydraulic permeability of

montmorillonite prevents convective transport near the canister surface and provides an additional mass-transfer resistance influencing radionuclide release rates. This paper addresses the quantitative calculation of steady, solubility-limited radionuclide release from a bentonite-protected waste package into an isotropic host porous medium. Such a calculation applies only after transient release has subsided. The time necessary to reach steady repository behavior under purely diffusive conditions can be estimated as

$$\epsilon_m D_m t / \alpha R_o^2 = O(1),$$

where ϵ_m is the geologic-medium porosity, D_m is the solute diffusion coefficient in the medium, t denotes time, α is the sorption retardation factor [12] (see Equation (8) of reference [4]), and R_o is the canister radius. With $\epsilon_m = 0.01$, $D_m = 10^{-5} \text{ cm}^2/\text{s}$, $\alpha = 100$, and $R_o = 15 \text{ cm}$, this results in a steady state after about 10^4 years. With additional convective transport this time will be considerably shortened [2]. Since repository life times must be orders of magnitude longer, a steady analysis of radionuclide release is useful [3,12].

Relyea and Wood [8] have performed an analysis of packing effectiveness with similar intent to ours. However, because they impose a one-dimensional flow field, steady state is not attained. More importantly, their one-dimensional hydrodynamics requires convective flow in the bentonite packing to be independent of absolute permeability. To relieve these restrictions, we analyze a two-dimensional geometry.

THEORY

Consider a unit length of an infinitely long, cylindrical fuel canister of radius R_o surrounded by packing out to radius R_p . The porosity and permeability of the packing are ϵ and K , respectively, while those of the geologic medium are ϵ_m and K_m . Ground water flows transverse to the isolated canister with a uniform approach superficial velocity designated by U . Fluid continuity, $\nabla \cdot \mathbf{v} = 0$, and Darcy's law combine to specify the pressure and velocity fields in the two concentric porous media encompassing the waste rod.

Let r be the radial coordinate measured from the center of the waste cylinder, and let θ be the angular coordinate gauged from the

forward stagnation point. The velocity components in the host rock are then

$$v_{r,m} = -U \left[1 - \frac{(1-\beta)}{(1+\beta)} \left(\frac{r}{R_p} \right)^{-2} \right] \cos \theta, \quad (1)$$

and

$$v_{\theta,m} = U \left[1 + \frac{(1-\beta)}{(1+\beta)} \left(\frac{r}{R_p} \right)^{-2} \right] \sin \theta, \quad (2)$$

where the parameter β is defined by

$$\beta \equiv \left(\frac{K_p}{K_m} \right) \frac{1 - (R_p/R_o)^{-2}}{1 + (R_p/R_o)^{-2}}. \quad (3)$$

Likewise, for the packing we find that

$$v_{r,p} = -U \left(\frac{K_p}{K_m} \right) \left[1 + \frac{(1-\beta)}{(1+\beta)} \right] \frac{1 - (r/R_o)^{-2}}{1 + (R_p/R_o)^{-2}} \cos \theta, \quad (4)$$

and

$$v_{\theta,p} = U \left(\frac{K_p}{K_m} \right) \left[1 + \frac{(1-\beta)}{(1+\beta)} \right] \frac{1 + (r/R_o)^{-2}}{1 + (R_p/R_o)^{-2}} \sin \theta. \quad (5)$$

Note the explicit dependence on the permeability of the packing material. To recover the case of no engineered barrier, one simply replaces R_p in Equations (1) - (3) with R_o .

The absolute permeability the packing is reported to vary from $10^{-8} \mu\text{m}^2$ for pure bentonite [9] to $10^{-4} \mu\text{m}^2$ for a mixture of bentonite and quartz [10]. A granite permeability of $1 \mu\text{m}^2$ [11] then yields a range for the permeability ratio, K_p/K_m , from 10^{-8} to 10^{-4} . Hence, Equations (4) and (5) demand velocities in the packing which are at most four orders of magnitude smaller than the ground water flow. A bentonite-impregnated packing material is very effective in preventing convection immediately adjacent to the canister. This contradicts the one-dimensional treatment where the superficial velocity in the packing is determined erroneously from mass balance to be U [8].

A dilute radionuclide in a supporting background electrolyte solution obeys, at steady state, the convective-diffusion equation:

$$v_n \cdot \nabla C_n = \epsilon_n D_n \nabla^2 C_n, \quad \begin{array}{ll} n = p & \text{for } R_o < r < R_p, \\ n = m & \text{for } r > R_p, \end{array} \quad (6)$$

where $v_n = v_{r,n} \hat{r} + v_{\theta,n} \hat{\theta}$.

Also, $D_n = D_m / \tau^2$ where D_m is the species molecular diffusion coefficient in water and τ^2 is the tortuosity of medium n . With this definition of D_n , we have neglected parallel and transverse dispersion. Equations (6) are combined with Equations (1) - (5) and solved subject to the following boundary conditions:

$$C_p(R_o, \theta) = C_s, \quad (7a)$$

$$C_m(R_p, \theta) = C_p(R_p, \theta), \quad (7b)$$

$$\epsilon_m D_m \frac{\partial C_m}{\partial r}(R_p, \theta) = \epsilon_p D_p \frac{\partial C_p}{\partial r}(R_p, \theta), \quad (7c)$$

$$\frac{\partial C_m}{\partial \theta}(r, 0 \text{ \& } \pi) = \frac{\partial C_p}{\partial \theta}(r, 0 \text{ \& } \pi) = 0, \quad (7d)$$

$$C_m(\infty, \theta) = 0. \quad (7e)$$

Restriction to solubility-limited canister dissolution is invoked with Equation (7a). Numerical solution of the linear Equations (6) is by Galerkin finite elements with bilinear basis functions. A critical test applied to assess the accuracy of the numerical procedure was matching of the integrated net radial flux at two arbitrary radial positions.

Following others [3,12] we ascertain the fractional release rate of a radionuclide, f . f is the angular-integrated species flux from the waste-canister surface divided by the amount of that nuclide originally in the canister [3,12]. To evaluate the role of packing, it proves convenient to report the reduced fractional release rate, f_p/f_o , or the ratio of the fractional release rates with and without packing:

$$f_p/f_o = \left(\frac{\epsilon_p D_p}{\epsilon_m D_m} \right) \frac{\int_0^\pi \left| \frac{\partial C_p}{\partial r}(R_o, \theta) \right| d\theta}{\int_0^\pi \left| \frac{\partial C_m}{\partial r}(R_o, \theta) \right| d\theta}, \quad (8)$$

where the integral in the denominator is evaluated with no packing present. In this manner we probe the effects of packing independent of any particular dissolving chemical species.

The reduced fractional release rate depends on the Peclet number

$$Pe \equiv UR_o / \epsilon_m D_m,$$

and on the effective diffusivity ratio $\epsilon_p D_p / \epsilon_m D_m$, the permeability ratio K_p/K_m , and the amount of packing or R_p/R_o . Superficial ground water velocities for candidate geologic media range from 10^{-6} to 0.1 m/yr [5,7]. For $R_o = 15 \text{ cm}$, $\epsilon_p = 0.01$ (i.e., granite [9]), and $D_m = D_o / \tau^2 = 10^{-5} \text{ cm}^2/\text{s}$, the Peclet numbers extend from $5(10^{-4})$ to 50. In this work Pe is varied from 10^{-4} to 10^3 .

The effect diffusivity in the packing, $\epsilon_p D_p$, is an important parameter. With compacted bentonite, $\epsilon_p D_p \sim 0.5$ to 0.75 . Knowledge of $D_p = D_o / \tau^2$ is sparse. Estimates might be from 10^{-7} to $10^{-5} \text{ cm}^2/\text{s}$. The latter high value of $10^{-5} \text{ cm}^2/\text{s}$ appears likely for cations and arises from surface migration [4]. We study the ratio $\epsilon_p D_p / \epsilon_m D_m$ from 0.1 to 10. Expected amounts of packing might be from one canister radius, $R_p/R_o = 1$, out to 5 or 6 canister radii.

As indicated earlier, permeabilities in bentonite and bentonite-sand mixtures are extremely small: $10^{-4} > K_p/K_m > 10^{-8}$. For K_p/K_m less than about 10^{-3} we find no influence whatsoever on f_p , independent of the Peclet number. This means that transport through bentonite packing is purely by diffusion [4]. For the calculations presented below, K_p/K_m is set at 10^{-5} .

RESULTS AND DISCUSSION

Figure 1 (all figures are at the end of the paper) gives reduced fractional release rates as a function of Pe for two different amounts of packing. If the packing were to have no influence, f_p/f_o would be unity for all Peclet numbers.

At low Pe ($< 5(10^{-2})$), release from the canister-packing system is by diffusion. Here, f_p/f_o is greater than unity because the diffusive resistance in the packing with $\epsilon = 0.75$ is less than that in the host medium with $\epsilon = 0.01$. At high Pe (> 50), nuclide transport is by diffusion in the packing and convection in the medium. In this large Pe regime, f_p approaches a constant asymptote characteristic of diffusion in the packing while f_o increases as $\sqrt{Pe}$ [2]. Accordingly, in Figure 1, f_p/f_o decreases as $1/\sqrt{Pe}$ in the high Pe range.

At intermediate Peclet numbers, a slight maximum appears. This is because the more porous packing, which in Figure 1 has a lower diffusive resistance than in the geologic medium, feels the onset of convection before the medium does. Hence, f_p rises more quickly with Pe than does f_o .

Figure 1 shows that unless high ground water flows exist in the repository, a packing with $\epsilon D_p/\epsilon D_m = 7.5$ has little benefit in the steady state. Additional packing does not mitigate this conclusion.

Figure 2 investigates the role of the effective diffusivity in the packing on the reduced fractional release rates for $R_p/R_o = 6$. As the effective diffusivity ratio is reduced, fractional release rates from the canister-packing assemblage are diminished significantly. Especially noteworthy is the lowering of the transition Peclet number between convective and diffusive transport. Figure 2 indicates that effort should be directed towards designing packing with ϵD_p small. Also, quantitative information on D_p is needed [4].

RAMIFICATIONS

Figures 1 and 2 ascertain that reduction in steady release rates with a bentonite-based packing is not dramatic except at very high Peclet numbers. The reasons are two-fold.

First, the porosity of host medium used in this work is very small: $\epsilon_m = 0.01$. To design a packing that exhibits a higher diffusive resistance than the host medium requires that ϵ_p be reduced. This can be accomplished by using large weight fractions of clay in conjunction with a finely divided matrix solid, such as crushed host rock. As long as all voids between the matrix solids are filled with the smectite clay, low permeability can be maintained while also reducing overall porosity [13]. Large grained or porous matrix particles would not be desirable.

Second, comparison of fractional release rates of the canister-packing-medium system to those of the canister-ideal medium system is unrealistic. After drilling, there will be a damaged zone of higher fracture density and porosity around the canister sites and a gap between the fuel canister and the damaged host rock. This gap requires backfilling with some material. Thus, comparison of bentonite-based packing should be made against other types of backfill and not against a uniform host medium that extends directly up to the canister surface.

We note that in our model calculations no account has been made for solute dispersion, for a fractured host medium, or for nonisothermal excursions. Our results hold only for isothermal, homogeneous porous media.

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Figure 1. Reduced fractional release rates: effect of packing amount.

Figure 2. Reduced fractional release rates: effect of packing diffusivity.

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