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MASS TRANSFER IN A GEOLOGIC ENVIRONMENT

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ABSTRACT

A new analytical solution is presented that predicts the rate of dissolution of species from a waste package surrounded by a wet porous medium. By equating the rate of diffusive mass transfer into the porous rock to the rate of liquid-surface chemical reaction, an analytical solution for the time-dependent dissolution rate and the time-dependent concentration of dissolved species at the waste surface is obtained. From these results it is shown that for most of the important species in a package of radioactive waste the surface liquid quickly reaches near-saturation concentrations and the dissolution rate can be predicted by the simpler theory that assumes saturation concentrations in the surface liquid.

INTRODUCTION

We have earlier presented analytical (nonempirical) equations that predict the rate at which radionuclides in liquid at the surface of a waste package can be transported into groundwater in the surrounding porous medium [1-4]. The expected groundwater velocities in some repositories are so low that the best estimate of the mass-transfer rate from the surface can be obtained from Chambre's diffusive-transport solutions that neglect groundwater motion near the waste surface. Once the dissolved species have diffused into groundwater in the surrounding rock, convection remains important to the further transport of dissolved species into the near and far fields.

The rate of mass transfer by diffusion from the surface liquid is found to be the phenomenon that controls the net rate at which most species in a waste solid dissolve in a geologic environment. This was demonstrated [4] by conservatively assuming saturation concentration in the surface liquid to compute diffusive mass-transfer rates and by comparing these rates with the much larger rates deduced from laboratory experiments where solid-liquid reactions are controlling. The effect of the low rate of diffusion into the surrounding porous or fractured medium in a geologic environment is to cause a rapid build-up of the concentration of dissolved species in the surface liquid. As saturation of each species is approached the net rate of solid-liquid reaction decreases to match the slow rate that the dissolved species can diffuse from the surface liquid.

The diffusion process analyzed here is not diffusion through surface layers of precipitates and alteration products, as inferred by some [5] from laboratory leach experiments; we are analyzing the molecular diffusion of dissolved species through the groundwater continuum in tortuous pathways formed by pores and fractures in the surrounding rock. Chambre's first analytical solutions applied to a waste solid in contact with porous rock. Later solutions included a backfill layer of crushed rock or clay between the waste solid and rock, but such layers are neglected in the new theory presented here.

Extensive data are available on the rate of dissolution of waste-solid samples in laboratory experiments, where dissolution rate is controlled by the rate of solid-liquid reactions. Techniques have been proposed to extrapolate the laboratory leach-rate data to predict dissolution rates in a repository environment, with the unjustified assumption that solid-liquid reactions will control the dissolution rate in a repository [6,7]. Here we make no

assumptions about which process is controlling. We solve the governing equations for the time-dependent mass-transfer (dissolution) rate from the surface of a waste solid into a surrounding saturated porous medium, using the experimentally determined surface-liquid reaction rate as a boundary condition to predict the actual concentration of the dissolved species in the exterior field and at the waste surface. This results in a more realistic estimate of the dissolution rate than has been calculated heretofore by assuming saturation concentration in the surface liquid. The resulting analytical solutions can be used to determine the conditions under which either chemical reaction rate or exterior-field diffusion controls mass transfer (dissolution).

#### DERIVATION OF THE THEORY

We consider a spherical waste solid surrounded by porous rock containing ground water, with water pore velocity low enough that mass transfer from the surface liquid is controlled by molecular diffusion of dissolved species through the tortuous liquid pathways in the rock [4]. Neglecting radioactive decay, the governing equation for diffusive mass transfer is:

$$K \frac{\partial C}{\partial t} = D \frac{1}{r^2} \frac{\partial}{\partial r} \left( r^2 \frac{\partial C}{\partial r} \right), \quad t > 0, \quad r_0 < r < \infty \quad (1)$$

with the initial condition:

$$C(r, 0) = 0, \quad r_0 < r < \infty \quad (2)$$

where  $C(r, t)$  is the concentration of dissolved species in ground water,  $K$  is the retardation coefficient,  $D$  is the diffusion coefficient in the liquid,  $r$  is the radial distance from the waste center,  $r_0$  is the radius of the waste form, and  $t$  is the time.

The continuity boundary condition at the waste-liquid interface specifies that the diffusive current of dissolved species at the waste outer surface equal the net rate of dissolution by solid-liquid reaction:

$$-\epsilon D \frac{\partial C(r_0, t)}{\partial r} = j_0 \left[ 1 - \frac{C(r_0, t)}{C_s} \right], \quad t' > 0 \quad (3)$$

Here  $\epsilon$  is the porosity of the surrounding medium,  $C_s$  is the saturation concentration of the dissolved species, and  $j_0$  is the experimental forward reaction rate of that species, per unit external surface area. The forward reaction rate is measured when the surface is in contact with a well-mixed liquid that contains none of the dissolved species being considered in Equation (1). The form of the right hand side of Equation (3), which meets the expected condition of zero dissolution rate when the solution is saturated, is suggested by experiments on dissolution of quartz [8-9] and implies a first-order solid-liquid reaction. It is a good approximation for the concentration-dependent dissolution of silica from borosilicate glass [12-14], and it is assumed to apply to other species in the waste.

The remaining boundary condition is:

$$C(\infty, t) = 0, \quad t > 0 \quad (4)$$

From the solution to the above set of equations we obtain the time-dependent concentration  $C(r_0, \tau)$  in the liquid adjacent to the outer surface of the waste:

$$C(r_0, \tau) = C_s \frac{R(1 - e^{-\tau} \operatorname{erfc}\sqrt{\tau})}{1 + R} \quad (5)$$

and the time-dependent dissolution rate  $j_0(r, \tau)$  at  $r_0$ :

$$j_0(r_0, \tau) = j_0 \frac{1 + R e^{-\tau} \operatorname{erfc}\sqrt{\tau}}{1 + R} \quad (6)$$

where the dimensionless time  $\tau$  is defined as:

$$\tau \equiv \frac{(1+R)^2 D t}{K r_o^2} \quad (7)$$

The dimensionless "flux ratio"  $R$  is defined as:

$$R = \frac{j_o r_o}{\epsilon D C_s} \quad (8)$$

and can be interpreted as:

$$R = \frac{\text{forward reaction rate per unit area at } r_o}{\text{steady-state diffusive mass transfer rate at } r_o} \quad (9)$$

We can interpret the forward reaction rate  $j_o$  in terms of a reaction-rate constant  $k$ :

$$j_o = k C_s \quad (10)$$

which results in:

$$R = \frac{k r_o}{\epsilon D} \quad (11)$$

Thus  $R$  is the square of a modified Thiele modulus.

At steady state the concentration and mass-transfer rate at the surface are:

$$C(r_o, \infty) = C_s \frac{R}{1+R} \quad (12)$$

and

$$j(r_o, \infty) = \frac{j_o}{1+R} \quad (13)$$

Equations (12) and (13) show that when the flux ratio  $R$  is large  $C(r_o, \infty) = C_s$  and  $j(r_o, \infty) = \epsilon D C_s / r_o$ . Under these conditions the net dissolution rate is controlled by molecular diffusion in the exterior field. When  $R \ll 1$  the surface concentration  $C(r_o, \infty)$  is much less than the saturation concentration, and the net mass-transfer rate is controlled by the solid-liquid reaction and is equal to  $j_o$ . For intermediate values of  $R$  one must use Equations (12) and (13).

Another quantity of interest is the time  $t_s$  necessary for the mass flux at the surface to reach within five percent of the steady-state value. From Equation (6) it can be determined that:

$$t_s = \begin{cases} 0 & \text{for } R \leq 0.01 \\ \frac{4 \times 10^2 K r_o^2}{\pi D} & \text{for } R \gg 1 \end{cases} \quad (14)$$

Chambre<sup>7</sup> has shown [15] that if radioactive decay is included in Equation (1) the above conclusions are affected by both decay and retardation. The analysis of the surface concentration and dissolution (mass-transfer) rate, including the effects of species decay within the waste form, has also been treated.

The normalized dissolution rate  $j(r_o, \tau)/j_o$  and the normalized surface concentration  $C(r_o, \tau)/C_s$  are shown in Figure 1 as a function of the dimensionless time  $\tau$  and the dimensionless parameter  $R$ .

#### APPLICATION TO SILICA AND CESIUM IN BOROSILICATE GLASS

To illustrate, we assume a waste glass cylinder of radius 0.15 m and length 2.4 m, resulting in an equivalent sphere of radius 0.44 m. From the

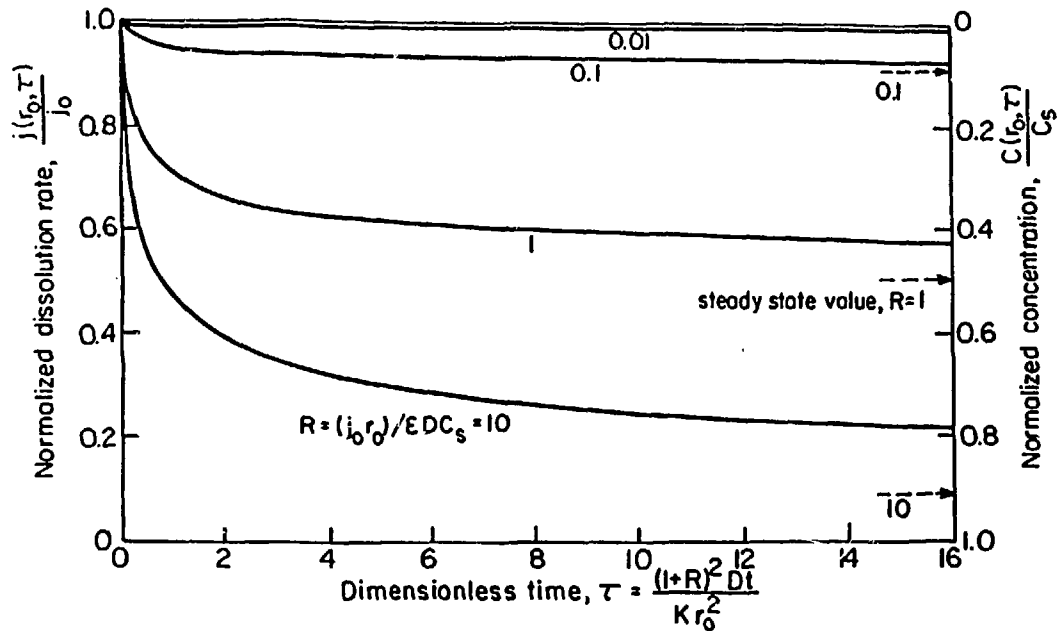


Fig. 1. Normalized dissolution rate and surface-liquid concentration as a function of dimensionless time for various values of the flux ratio  $R$ .

laboratory leach data of Pederson et al. [12] for PNL 76-68 borosilicate glass we derive the following effective values for silica at 90°C:

$$j_0 = 1.18 \text{ g SiO}_2/\text{m}^2 \text{ day}$$

$$C_s = 200 \text{ g SiO}_2/\text{m}^3$$

The room-temperature diffusion coefficient of silicic acid in water [9,16] is corrected for temperature [17] to obtain  $D = 7.7 \times 10^{-2} \text{ m}^2/\text{yr}$  at 90°C.

Reduction of  $D$  because of tortuosity in the rock is conservatively neglected. Assuming a rock porosity of 0.01, we estimate  $R_{\text{SiO}_2} = 1240$ . Assuming no sorption of silica on the rock ( $R = 1$ ), the estimated time to steady state is 320 years. From Equation (12)  $C(r_0, \infty) = 0.999 C_s$  for silica, and the steady-state mass transfer will be controlled by exterior-field diffusion.

Figure 2 shows the normalized surface mass flux of silica and the normalized surface concentration as a function of the dimensionless time  $\tau$  and chronological time  $t$ . Also shown are the results predicted by assuming that saturation concentration always exists in the surface liquid, as in our earlier conservative estimates of mass-transfer rate. For assumed constant saturation the early mass-transfer rate is infinite, a result not physically reasonable. It is at these early times that the solid-liquid reaction rate is important for the dissolution of silica. The actual surface concentration is initially zero, and it grows with time at a finite rate determined by the solid-liquid reaction rate. Within about seven minutes after bare glass waste has been placed in contact with saturated rock ( $\epsilon = 0.01$ ) the concentration of silica in the surface liquid will have reached 82 percent of saturation and the mass-transfer rate will be within 5 percent of that predicted for constant saturation at the surface, assuming no silica sorption in the rock. If silica sorbs with a retardation coefficient of 100, this time is increased to 700 minutes.

The time to reach near-saturation concentration will be longer if there is stagnant liquid between the waste surface and the exterior porous medium. This time delay is easily added to the theory because the intervening liquid is likely to be well mixed.

From high-flow leaching experiments [14] we estimate the forward reaction rate of cesium dissolving from PNL 76-68 glass at 90°C as  $j_0 = 2.0 \times 10^{-2} \text{ g/m}^2 \text{ day}$ . From [18] the effective saturation concentration of cesium in equilibrium with solid phases from glass dissolution is  $C_s = 5.97 \text{ g/m}^3$ . From [19] we adopt  $K_{C_s} = 1000$ . For a typical liquid diffusion coefficient  $D = 1.2 \times 10^{-1} \text{ m}^2/\text{yr}$  at 90°C, conservatively neglecting tortuosity, the calculated concentration of cesium in the surface liquid as a function of time is shown in Figure 3. After 100 days the surface concentration is 90 percent of saturation. Steady-state mass transfer is achieved only after  $2 \times 10^3$  years.

#### STEADY-STATE CONCENTRATIONS AND MASS-TRANSFER RATES FOR OTHER SPECIES

Table I presents values of the steady-state concentration ratio  $C(r_0, \infty)/C_s$  and the flux ratio  $R$  for silica and several radioelements, calculated from Equations (12) and (13), together with the values of  $j_0$  and  $C_s$  selected for these calculations. In all cases the steady-state surface concentration is within a fraction of a percent of saturation, the ratio  $R$  is large, and except for early times the dissolution rate is controlled by exterior-field mass transfer. The mass-transfer rates can be estimated accurately by assuming saturation concentration in the surface liquid, except during the first few days after the waste solid is first exposed to liquid.

#### COMPARISON WITH DATA ON NATURAL MINERALS

To illustrate the importance that solid-liquid reaction rate can have on

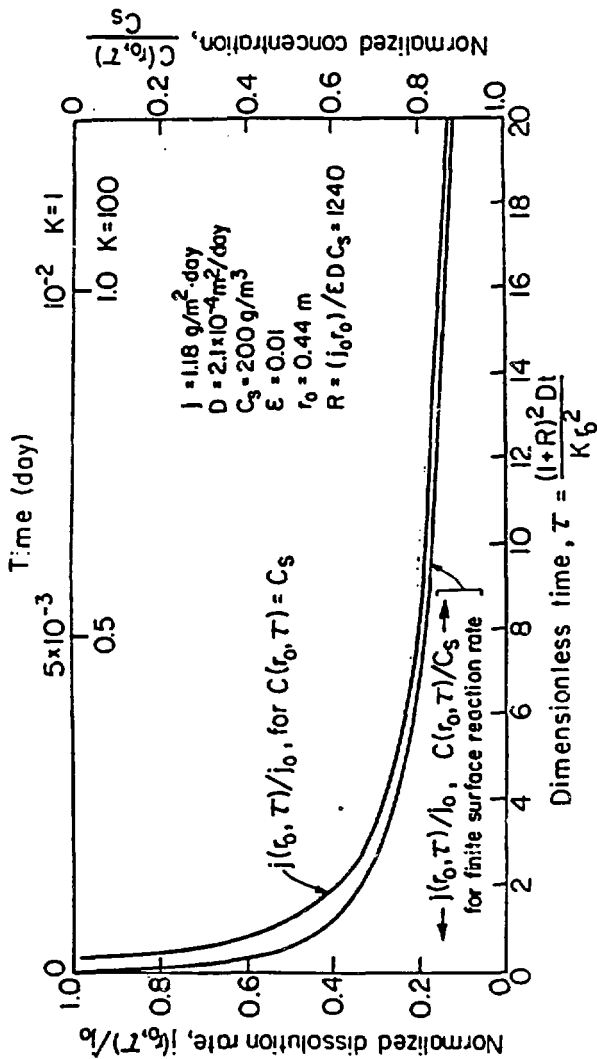


Fig. 2. Normalized dissolution rate and surface-liquid concentration of silica from borosilicate glass as a function of time, compared with dissolution rate for constant saturation concentration at the surface.



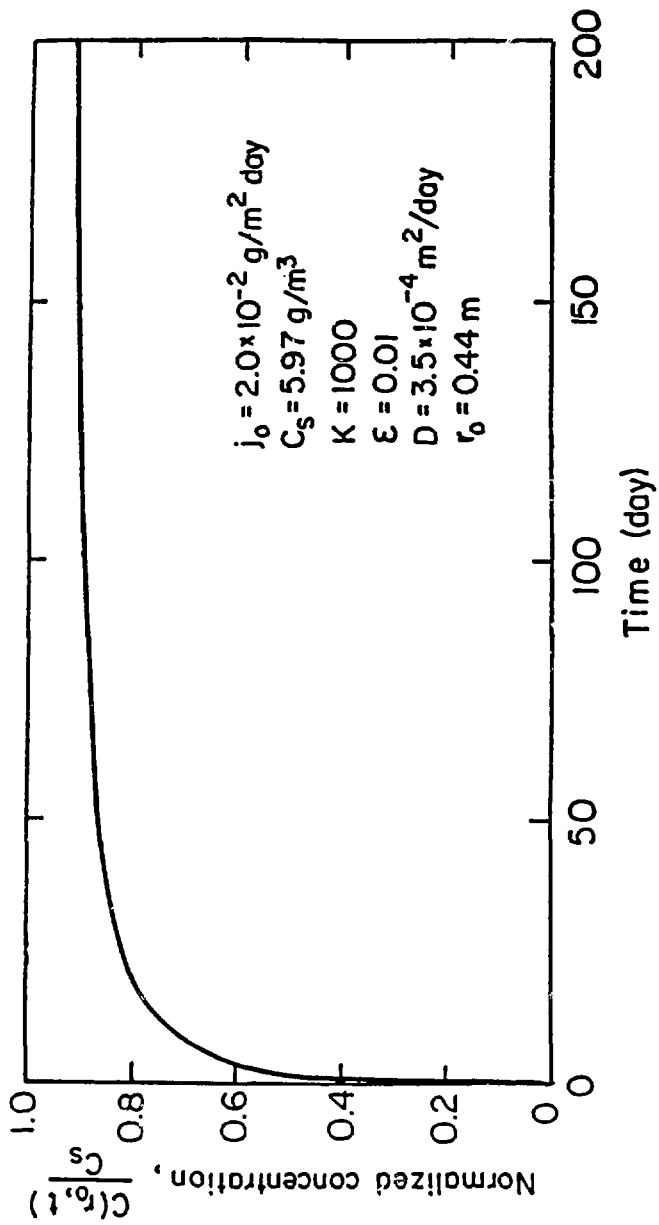


Fig. 3. Normalized surface-liquid concentration of cesium from borosilicate glass as a function of time.

Table I. Normalized surface concentration and flux ratio for borosilicate-glass constituents at steady state ( $r_0 = 0.44 \text{ m}$ ,  $\epsilon = 0.01$ )

Species	$J_0, \text{ g/m}^2 \text{ day}$	Solubility, $\text{g/m}^3$	Concentration ratio	Flux Ratio
			$C(r_0, \infty)/C_s$	R
$\text{SiO}_2$	1.18	$2.0 \times 10^2$	0.999	$1.2 \times 10^3$
U	$9.84 \times 10^{-5} \text{ a/}$	$9.52 \times 10^{-2} \text{ c/}$	0.998	$5.3 \times 10^2$
Np	$3.02 \times 10^{-4} \text{ a/}$	$2.37 \times 10^{-2} \text{ c/}$	0.999	$6.5 \times 10^3$
Pu	$1.20 \times 10^{-5} \text{ a/}$	$1.43 \times 10^{-5} \text{ c/}$	0.999	$4.3 \times 10^5$
Am	$6.29 \times 10^{-5} \text{ a/}$	$1.0 \times 10^{-4} \text{ d/}$	0.999	$3.2 \times 10^5$
Tc	$2.02 \times 10^{-1} \text{ a/}$	$1.0 \times 10^{-3} \text{ d/}$	0.999	$1.0 \times 10^8$
Cm	$2.57 \times 10^{-7} \text{ a/}$	$1.0 \times 10^{-3} \text{ d, e/}$	0.992	$1.3 \times 10^2$
Cs	$2.0 \times 10^{-2} \text{ b/}$	$5.97 \text{ f/}$	0.998	$4.3 \times 10^2$

a/ at  $22^\circ\text{C}$ , IAEA test, first-day leach data[20].

b/ at  $90^\circ\text{C}$ , data of Strachan and Exarhos, as given by Chick and Turcotte[14].

c/ at  $25^\circ\text{C}$ , concentration after 341 days in desaturated static leach experiment[21].

d/ Estimates by Krauskopf[17].

e/ Data subject to uncertainty.

f/ at  $90^\circ\text{C}$ , static leaching of borosilicate glass[18], concentration in deionized water after 1 year, possible compound formation.

dissolution rate, even for a dissolving solid surrounded by saturated rock, we consider the dissolution of natural quartz. The solubility of alpha silica is quoted to be  $44 \text{ g/m}^3$  [22], over fourfold less than the apparent saturation concentration of silica in borosilicate glass. From the data of van Lier et al. [9],  $j_0 = 3.1 \times 10^{-5} \text{ g/m}^2\text{day}$ , over four orders of magnitude less than that deduced for silica in borosilicate glass. Thus, even for the same size of dissolving material the flux ratio R for quartz would be about four orders of magnitude less than that for silica in glass, and dissolution of quartz would be limited by solid-liquid reaction rates. Correcting further for the small size of quartz samples that have been studied as natural analogs, the flux ratio R is further decreased, and exterior-field diffusion is expected to be unimportant for quartz dissolution, consistent with the results of Wood and Walther [23].

Berner [24] has measured dissolution rates of various small grains of minerals in water-saturated rocks, solids, and sediments and observes that substances with solubilities greater than  $10^{-4}$  moles/liter dissolve at a rate controlled by exterior-field diffusion, whereas substances with solubilities less than  $10^{-5}$  moles/liter dissolve at rates controlled by surface reactions. Whether Berner's observations are consistent with the theory presented here is unclear. In the form of Equation (11), the flux ratio R is not dependent upon solubility, unless there is some connection between the reaction-rate constant k and  $C_0$ . However, Berner observes that the solid-liquid reaction is usually complex, and a higher-order reaction could result in the observed ranking. Without data on grain size  $r_0$ , exterior-field porosity  $\epsilon$ , and  $j_0$  or k for Berner's experiments, we cannot test his observations with the theory presented herein.

Berner's observations with small grains cannot be generalized to the much larger sizes of solid wastes in a repository. The above theory shows that if the size of each separate dissolving grain is small enough (small  $r_0$ ) a small flux ratio R can result and solid-liquid reaction rate can control dissolution, whereas for large  $r_0$  exterior-field diffusion can control.

Although no generalization can be made on solubility alone, data in Table I show that for borosilicate glass waste the constituents with the lowest solubilities, i.e., plutonium and americium, have the largest values of R, and their dissolution rate is controlled not by surface reaction rate but by exterior field mass transfer, assuming first-order solid-liquid reactions. This conclusion is consistent with that arrived at by comparing maximum exterior-field dissolution rates with those determined in laboratory leach experiments [19].

#### EFFECT OF INTERNAL CRACKS IN THE WASTE SOLID

When exterior-field mass transfer is controlling the dissolution rate and the flow of groundwater through the waste solid is negligible, the exterior surface area of the waste solid is the proper surface area on which to base the calculation of release rate, as has been done in the above analysis. If solid-liquid reaction rate is controlling, the availability of internal surface in cracks and fissures can increase the effective surface available for solid-liquid reaction, and a highly fragmented solid will increase the value of  $j_0$  used in the above analysis. The values of  $j_0$  used in the numerical illustrations herein are based on solid samples with a reported effective surface area for solid-liquid reaction about fivefold greater than the outer geometric surface area of the sample. If the equivalent waste solid in the repository were to have no equivalent internal surface area for solid-liquid reaction, the effective  $j_0$  would decrease by about fivefold, and R could decrease by this amount. If the other data are correct, the R values would still be large and our conclusions would remain the same.

#### EFFECT OF TORTUOSITY

The above numerical illustrations conservatively assume a diffusion coefficient appropriate for a liquid continuum. Measured coefficients for molecular diffusion in granite [25,26] are 100- to 1000-fold less than the values used, whereas values tenfold lower are typical for other rocks. The lower diffusion coefficients increase the value of  $R$ , they further reduce the importance of solid-liquid reaction rates, and they increase the time to steady-state.

## CONCLUSIONS

The theory for predicting waste-solid dissolution rate on the basis of mass-transfer rate in surrounding groundwater has been extended to include solid-liquid chemical reaction rate as a boundary condition. A quantitative criterion is developed to predict under what conditions either of the rate processes controls dissolution rate.

From the data deduced from laboratory experiments on PNL 76-63 borosilicate glass, and using Krauskopf's [19] solubility data for some species, the dissolution rates of silica, uranium, neptunium, plutonium, americium, technetium, curium and cesium are predicted to be controlled by exterior-field mass transfer, except at early times of the order of weeks or months after the beginning of exposure to ground water.

Based on data adopted herein, dissolution rates for these species can be adequately estimated by assuming saturation concentration in the liquid at the waste surface. This justifies the use of analyses developed earlier by Chambre' [1-4] that include the effects of backfill and radioactive decay and assume constant saturation concentration at the waste surface.

This theory is a mechanistic representation of processes expected to control or affect the rate of dissolution of waste solids in a saturated-rock repository. It does not require ad hoc assumptions of effective ratios of waste surface area to repository water volume and effective water-solid contact time as postulated by others [6,7] to predict waste dissolution in a repository.

The theory identifies the experimental data important for predicting waste performance in a repository: saturation concentrations, diffusion coefficients, effective porosity, and retardation coefficients. Forward-reaction rate data are useful to confirm the importance of exterior-field mass transfer or to predict dissolution rates if solid-liquid reaction rate is important. Such data are needed for spent-fuel constituents such as carbon, iodine, cesium, technetium.

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