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THE EFFECTS OF SORPTION AND DECAY ON STEADY-STATE RADIONUCLIDE RELEASE RATES

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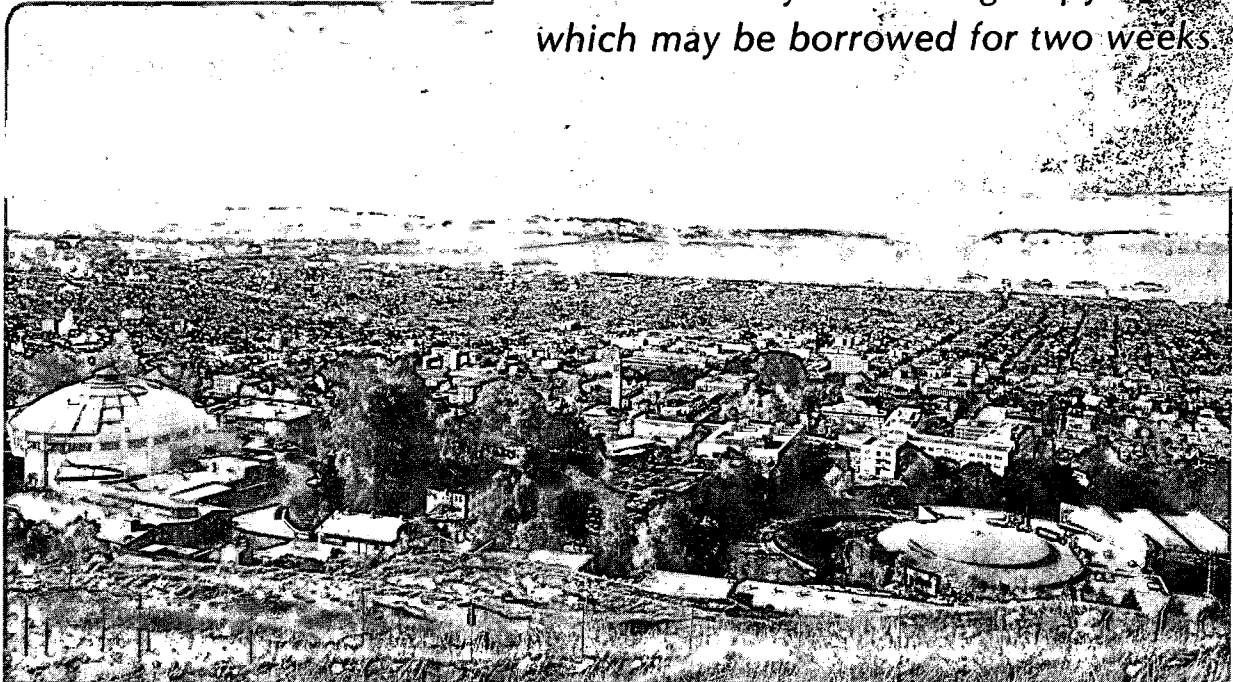
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RADIONUCLIDE RELEASE RATES

W.B. Light, W.W.-L. Lee, P.L. Chambré,  
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THE EFFECTS OF SORPTION AND DECAY ON STEADY-STATE  
RADIONUCLIDE RELEASE RATES

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In this paper we analyze the effects of equilibrium sorption and radioactive decay on steady-state mass-transfer rates from waste packages in nuclear waste repositories. Understanding these processes will contribute to meeting the Nuclear Regulatory Commission's subsystem performance requirement for the engineered barrier system<sup>1</sup> to the design of backfills for nuclear waste packages.

We considered steady-state dissolution of a nuclide, without precursors, from a spherical-equivalent waste form surrounded by backfill and porous rock. The backfill and porous rock are water saturated. We assume low enough flow of ground water so that diffusion is the dominant process of mass transfer<sup>2</sup>. Assuming local sorption equilibrium and constant near-saturation concentration of dissolved species at the waste-form surface, the governing equations for space-time-dependent nuclide concentrations were solved, and a parametric study was performed to investigate the effects of sorption and decay. Two quantities are of interest: nuclide concentration at the backfill/rock interface, divided by the nuclide concentration at the same location when backfill is replaced by rock, and the mass transfer rate at the backfill/rock interface, normalized to mass-transfer rate for a stable nuclide at that location.

Results shown in Figures 1 and 2 are in terms of a modified Thiele modulus,

$$\phi^2 = r_1^2 \lambda K_2 / D,$$

where  $r_1$  = radius of spherical-equivalent waste form [L]  
 $\lambda$  = decay constant [T<sup>-1</sup>]  
 $K_2$  = retardation coefficient of the nuclide in rock  
 $D$  = diffusion coefficient [L<sup>2</sup>/T]

Figure 1 shows the ratio of steady-state concentration  $N_2$  at the backfill/rock interface to the concentration  $N_{2e}$  that would occur there if the backfill is replaced by rock, as a function of the backfill-to-rock retardation ratio,  $K_1/K_2$ . The ordinate also represents the ratio of the mass-transfer rate at the backfill/rock interface to the mass-transfer rate there if backfill is replaced by rock. Because for a stable nuclide sorption cannot affect the steady-state solution ( $\lambda = 0, \phi = 0$ ), the result is independent of the retardation ratios; it exceeds unity because of the greater porosity of backfill. Increasing the decay constant steepens the steady-state concentration gradient and can increase the interface concentration ratio if backfill retards less than rock ( $K_1 < K_2$ ). For finite decay and greater backfill sorption ( $K_1 > K_2$ ), increased diffusive time in the backfill results in greater decay in the backfill and lower concentration at the backfill/rock interface.

Figure 2 shows the mass-transfer rate at the backfill/rock interface, normalized to the mass-transfer rate  $M^*$  at that location for a stable nuclide, with backfill replaced by rock, as a function of the retardation ratio  $K_1/K_2$ .

Increasing the modified Thiele modulus  $\phi$  increases the mass-transfer ratio for small  $K_1/K_2$  but decreases it for large  $K_1/K_2$ , as is expected on physical grounds:

#### References

1. U. S. Nuclear Regulation Commission, Title 10, Code of Federal Regulations, 60.113(a)(1)(11)(B), 1983.
2. T. H. Pigford, J. O. Blomeke, T. L. Brekke, G. A. Cowan, W. E. Falconer, N. J. Grant, J. R. Johnson, J. M. Matuzek, R. R. Parizek, R. L. Pigford, and D. E. White, A Study of the Isolation System for Geologic Disposal of Radioactive Wastes. Washington: National Academy Press. 1983.

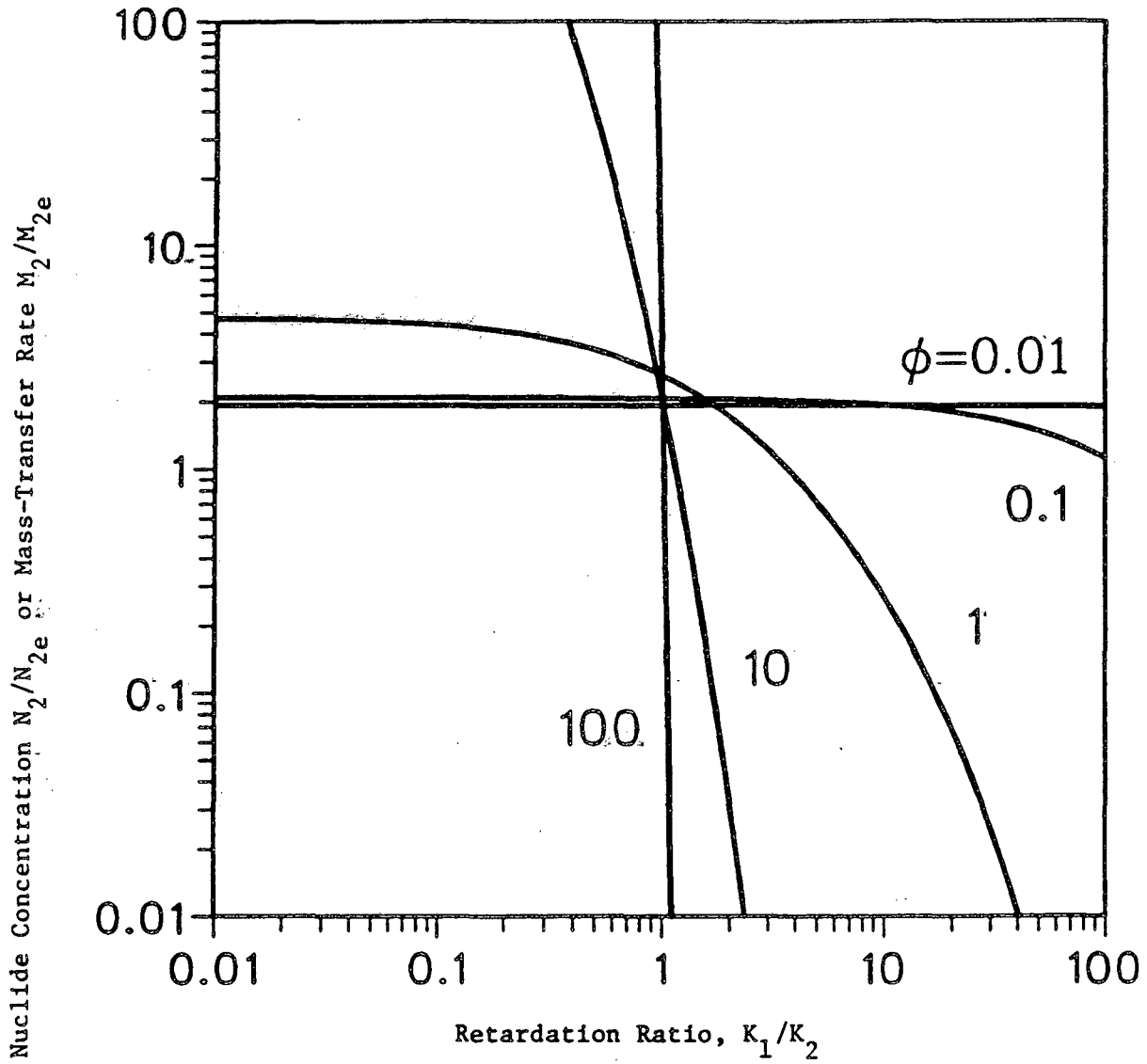


Figure 1. Steady-state concentration and mass-transfer rate at the backfill/rock interface, normalized to the case with no backfill present, as a function of the retardation ratio.

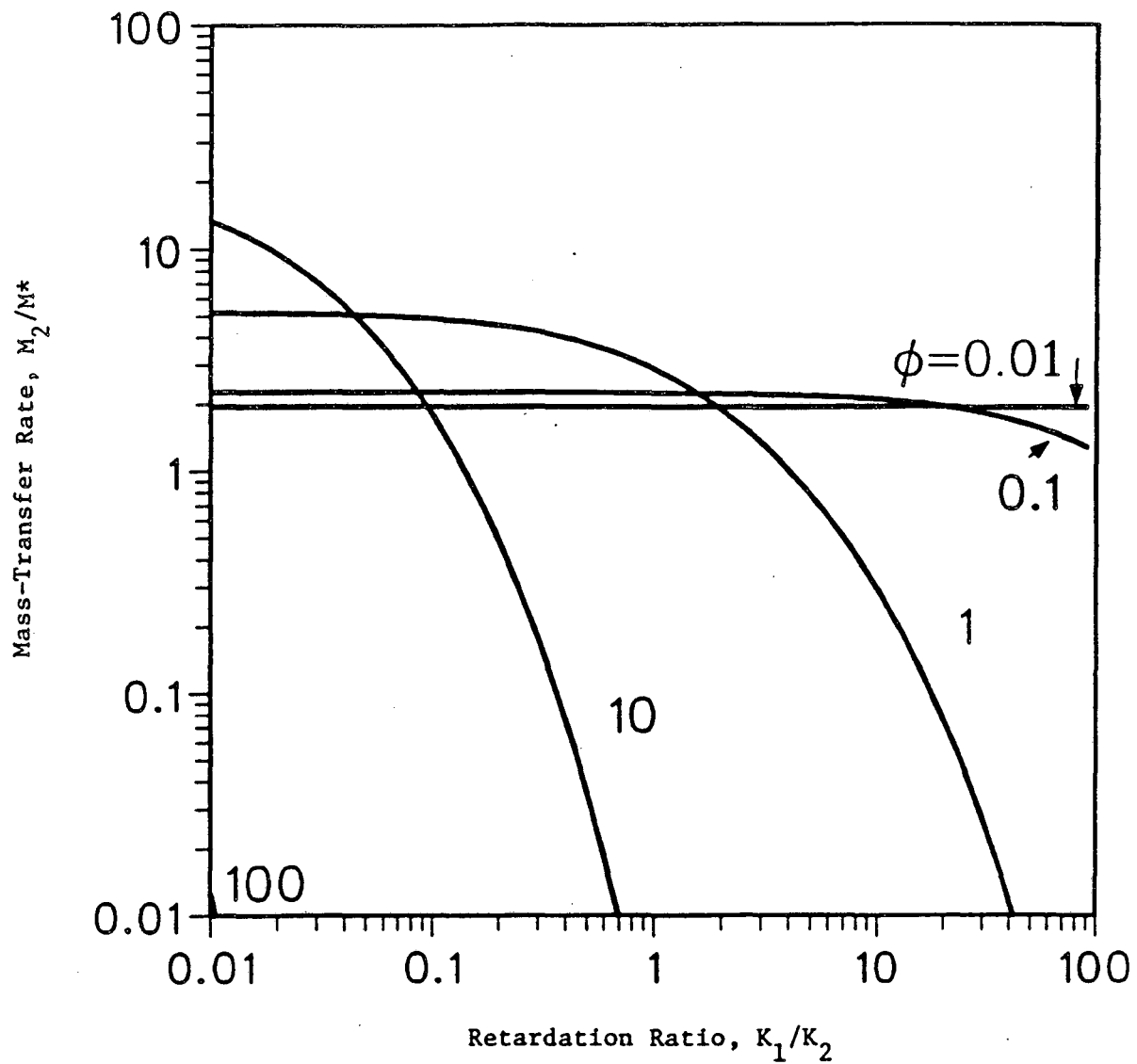


Figure 2. The steady-state mass-transfer rate at the backfill/rock interface normalized to the mass-transfer rate for a stable nuclide with no backfill present, as a function of the retardation ratio .



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