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Isotopic Effects on Solubility-Limited Mass Transfer

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In our previous theoretical analyses, solubility-limited mass transfer from waste solids applies if a species is at constant elemental concentration in liquid at the waste surface.¹ For an element with isotopes that decay appreciably during the time of interest, a solubility boundary condition results in a time-dependent boundary concentration of each isotope. Here we present new mass-transfer equations that include the effects of isotopic decay boundary conditions.

Let there be n isotopes of the element, and γ_i the respective isotopic fraction in the waste solid so that $\sum_{i=1}^n \gamma_i = 1$. Because the n isotopes decay at different rates, $\gamma_i = \gamma_i(t)$. However, for simplicity the usual assumption has been^{2,3}

$$\gamma_i(t) = \gamma_i, \quad \forall t \quad (1)$$

With this assumption, the fractional release rate⁴ $f_i(t)$ for an isotope of elemental solubility C_{se} , released from the surface of a waste sphere of radius r_{sp} by diffusive mass transfer into surrounding saturated porous rock of porosity ϵ , based on the initial inventory M_i^0 , is⁵

$$f_i(t) = \frac{4\pi r_{sp} C_{se} \gamma_i D \epsilon}{M_i^0} \left\{ 1 + \sqrt{\frac{K_i \lambda_i r_{sp}^2}{D}} \operatorname{erf} \sqrt{\lambda_i t} + \sqrt{\frac{K_i r_{sp}^2}{\pi D t}} e^{-\lambda_i t} \right\}, \quad t > 0 \quad (2)$$

where D is the species diffusion coefficient,

K is the retardation coefficient,

λ is the decay coefficient.

Eq. (2) is a suitable approximation for most radionuclides important in high-level waste. It can be used conservatively for species that decay in the time of interest by adopting γ_i as the maximum value for that isotope during the interval.

A more accurate approach is to solve the mass-transfer equations for time-dependent $\gamma_i(t)$. For an element with two isotopes, the time-dependent inventories $M_i(t)$ and $M_s(t)$ for the radioactive and stable isotopes in the waste solid are

$$\frac{dM_i(t)}{dt} = -\lambda_i M_i(t) - 4\pi r_{sp}^2 \left(-\epsilon D \frac{\partial N_i(r_{sp}, t)}{\partial r} \right), \quad t > 0 \quad (3)$$

and

$$\frac{dM_s(t)}{dt} = -4\pi r_{sp}^2 \left(-\epsilon D \frac{\partial N_s(r_{sp}, t)}{\partial r} \right), \quad t > 0 \quad (4)$$

where $N(r, t)$ is the nuclide concentration in pore liquid. For short-half-life nuclides the assumption can be made that

$$4\pi r_{sp}^2 \left(-\epsilon D \frac{\partial N_i(r_{sp}, t)}{\partial r} \right) \ll \lambda_i M_i(t) \quad (5)$$

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and there is considerable simplification of the system of equations to be solved. (3) reduces to

$$\frac{dM_i(t)}{dt} = -\lambda_i M_i(t), \quad t > 0 \quad (3')$$

and (4) to

$$\frac{dM_s(t)}{dt} = 0, \quad t > 0 \quad (4')$$

and the fractional release rate is

$$f_i(t) = 4\pi r_{sp} C_{se} D \epsilon \left(\frac{e^{-\lambda_i t}}{M_i^o e^{-\lambda_i t} + M_i^o} + \sqrt{\frac{K_i r_{sp}^2}{\pi D}} \frac{e^{-\lambda_i t}}{M_i^o + M_i^o} \frac{1}{\sqrt{t}} \right. \\ \left. + \sqrt{\frac{K_i \lambda_i r_{sp}^2}{D}} \frac{2}{\sqrt{\pi}} \int_0^{\sqrt{\lambda_i t}} \frac{M_i^o e^{-2\lambda_i t - \eta^2}}{(M_i^o e^{-\lambda_i t - \eta^2} + M_i^o)^2} d\eta \right), \quad r > r_{sp}, \quad t > 0 \quad (6)$$

Table I. Data used for Illustration

Property	Units	⁸⁸ Sr	⁹⁰ Sr	Ref.
Decay Constant (λ)	per year	0	0.024	6
Isotopic Fraction (γ)		0.4	0.6	6
Initial Inventory (M)	g/MTHM	349	519	7
Retardation Coefficient (K)		200		8
Diffusion Coefficient (D)	m ² /a	3×10^{-3}		8
Elemental Solubility (C_{se})	g/m ³	0.01		8

To illustrate, we consider the release of solubility-limited strontium, consisting of radioactive ⁹⁰Sr and stable ⁸⁹Sr, from spent fuel waste surrounded by porous rock. An equivalent-sphere radius of 1 m is assumed. The waste container and fuel cladding are assumed not to be present, and the waste is assumed to contact ground water shortly after emplacement. Using assumed properties listed in Table I, calculated fractional release rates of ⁹⁰Sr for constant and time-dependent boundary concentrations are shown in Figure 1. Because ⁹⁰Sr is the only decaying species, the fractional release rate considering decay is much less than the more conservative value that neglects decay. Fractional release rates based on the 1000-year inventory, as required by federal regulations,⁴ can be obtained by multiplying the values in Figure 1 by $e^{1000\lambda}$.

Isayama⁹ provides additional illustration of these equations, in both near-field and far-field applications.

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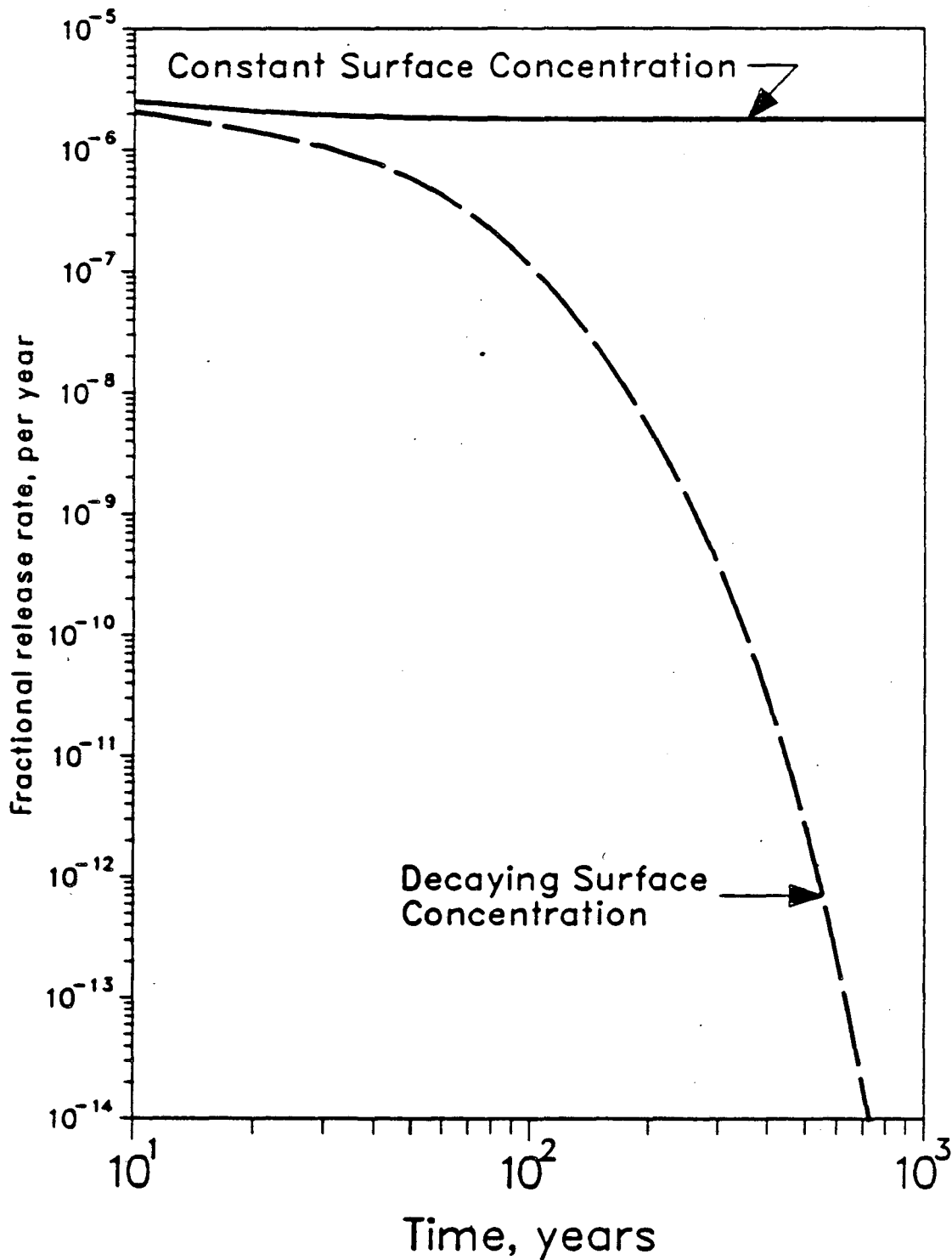


Figure 1. Solubility-Limited Fractional Release Rate of Sr-90 (based on initial inventory)

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