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Authors

Kim, C.L.

Chambre, P.L.

Lee, W.W.L.

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February 1988

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**VARIABLE TEMPERATURE EFFECTS ON
RELEASE RATES OF READILY SOLUBLE NUCLIDES**

C.-L. Kim, P. L. Chambré, W. W.-L. Lee, and T. H. Pigford

Department of Nuclear Engineering
University of California

and

Earth Sciences Division
Lawrence Berkeley Laboratory
1 Cyclotron Road
Berkeley, California 94720

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Variable Temperature Effects on Release Rates of Readily Soluble Nuclides

C.-L. Kim
Korea Advanced Energy Research Institute
Daejeon, Korea
042 820-2871

W. B. Light, W. W.-L. Lee, P. L. Chambré & T. H. Pigford
Lawrence Berkeley Laboratory & Department of Nuclear Engineering
University of California, Berkeley, CA 94720-0001, USA
415-642-6469

1. Introduction

Radionuclides in waste packages in nuclear waste repositories can be released in two ways. The release of low-solubility species is controlled by exterior field diffusion-advection.¹ Readily soluble species such as cesium and iodine in fuel cladding gaps are known to dissolve rapidly when exposed to ground water.² However, the rate of release of these soluble species from the waste package will be limited by the rate of mass transfer through the surrounding porous media. Near waste packages in geologic repositories of nuclear waste, the temperature is likely to be elevated for hundreds to thousands of years. If dissolution and migration of radionuclides should occur, the higher temperature will influence their respective rates. In our previous analyses^{3,4} of solubility-limited diffusive dissolution with time-temperature-dependent solubility and diffusion coefficient, the dissolution rates at higher temperatures were orders of magnitude greater than at ambient temperature.

In this paper we study the effect of temperature on the release rate of readily soluble nuclides,⁵ as affected by a time-temperature-dependent diffusion coefficient. In this analysis ground water fills the voids in the waste package at $t = 0$ and one percent of the inventories of cesium and iodine are immediately dissolved into the void water. Mass transfer resistance of partly failed container and cladding is conservatively neglected. The nuclides move through the void space into the surrounding rock under a concentration gradient. We use an analytic solution to compute the nuclide concentration in the gap or void, and the mass flux rate into the porous rock.

2. Analysis

We assume, in the time scale of interest, that ground water suddenly fills the void space of volume V between the waste and the emplacement hole and a certain amount of the readily soluble species is released instantaneously into the void water, to an initial concentration N^0 . The void space volume is constant with time. The mass transfer resistance of partly failed containers and backfill have been conservatively neglected. Figure 1 shows the planar geometry used in this analysis. The position and time-dependent concentration of the soluble species in the water-saturated porous rock $N(x, t)$ is determined by the diffusive transport equation with sorption and radioactive decay

$$K \frac{\partial N(x, t)}{\partial t} = D(t) \frac{\partial^2 N(x, t)}{\partial x^2} - \lambda K N(x, t), \quad x > a, \quad t > 0 \quad (1)$$

where λ is the decay constant [T^{-1}], $D(t)$ is the species diffusion coefficient [L^2/T] and K is the species retardation coefficient.

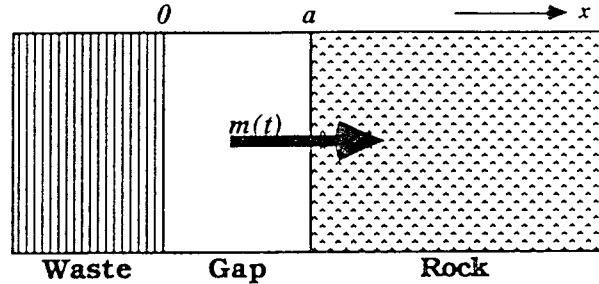


Fig. 1. Migration of readily soluble species from a gap into rock

We assume that the time-dependent temperature at the waste package surface is known,⁴ and that the same temperature exists everywhere in the porous rock. With the temperature-dependence of the diffusion coefficient given by the Nernst-Einstein equation

$$D\mu/T = \text{constant}$$

where μ is the absolute viscosity and T the temperature, and the time-dependence of temperature, we obtain the time dependence of the diffusion coefficient as

$$D(t) = D_0 g(t)$$

where D_0 is a constant and $g(t)$ is dimensionless.

The initial and boundary conditions are

$$N(x, 0) = 0, \quad x > a \quad (2)$$

$$N(a, t) = N(t), \quad t \geq 0 \quad (3)$$

$$N(\infty, t) = 0, \quad t \geq 0 \quad (4)$$

Here $N(t)$ is the time-dependent concentration of the soluble species in the water in the void. Because this void space is small, we assume that $N(t)$ is not position-dependent. To solve for $N(t)$, the mass balance in the void is

$$V \frac{dN(t)}{dt} = -\dot{m}(t) - \lambda V N(t), \quad t > 0 \quad (5)$$

where $\dot{m}(t)$ is the rate of migration of the species from the void into the rock. To solve (5), we use the initial condition

$$N(0) = N^0$$

Chambré has obtained the solution of equation system (1) through (5) for a constant diffusion coefficient,⁶ and Kim modified the solution for application to the problem with a time-dependent diffusion coefficient⁷ adapting the transformations given in Chambré *et al.*⁶ The species concentration in the void is

$$N(t) = N^0 e^{-\lambda t} \exp\left\{\beta^2 \int_0^t g(t') dt'\right\} \times \operatorname{erfc}\sqrt{\beta^2 \int_0^t g(t') dt'}, \quad t > 0 \quad (6)$$

where

$$\beta \equiv \sqrt{D_0 K \epsilon^2 / a^2}$$

The concentration of the nuclide in the rock is

$$N(x, t) = N^0 e^{-\lambda t} e^{K\epsilon(x-a)/a} \exp\left\{\beta^2 \int_0^t g(t') dt'\right\} \times \operatorname{erfc}\left\{\sqrt{\beta^2 \int_0^t g(t') dt'} + \frac{K\epsilon(x-a)}{2a} \frac{1}{\sqrt{\beta^2 \int_0^t g(t') dt'}}\right\}, \quad x \geq a, t > 0 \quad (7)$$

The mass flux of the species into the rock is

$$\dot{m}(t) = -SD(t)\epsilon \frac{\partial N(a, t)}{\partial x}, \quad t > 0 \quad (8)$$

where S is the surface area of the interface between the void space and the rock ($S \equiv V/a$). Using (7) the solution to (8) is

$$\dot{m}(t) = N^0 \beta V g(t) e^{-\lambda t} \times \left\{ \frac{1}{\sqrt{\beta^2 \int_0^t g(t') dt'}} - \beta e^{\beta^2 \int_0^t g(t') dt'} \operatorname{erfc}\sqrt{\beta^2 \int_0^t g(t') dt'} \right\}, \quad t > 0 \quad (9)$$

Eq. (6) and (9) are used for the numerical illustration.

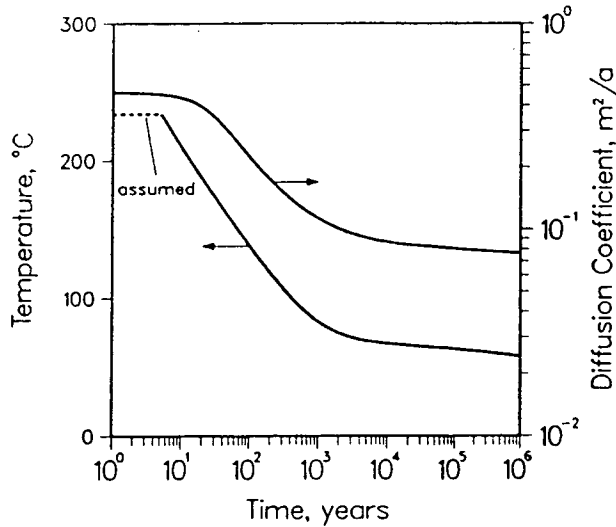


Fig. 2. Assumed temperature at waste package surface and time-temperature-dependent diffusion coefficient

3. Numerical Illustration

The Nernst-Einstein temperature dependence of the diffusion coefficient used is⁴

T (°C)		20	50	100
D (cm²/s)		1×10^{-6}	2×10^{-5}	4.5×10^{-5}
T (°C)		150	200	250
D (cm²/s)		7.9×10^{-5}	1.2×10^{-4}	1.6×10^{-4}

We assume a waste package temperature history as shown in Figure 2. Using the data on the temperature dependence of the diffusion coefficient and the temperature history of Figure 2, a spline function is constructed for the time-dependence of the diffusion coefficient, with the result also shown in Figure 2.

We use a void space of 0.45 m^3 , representing a gap thickness of 10 cm. At $t = 0$, one per cent of the total inventory of the soluble species, such as ^{135}Cs or ^{137}Cs , is released into the gap space. This results in an initial concentration of ^{135}Cs in the gap of

$$N^0 = 19.6 \text{ g/m}^3$$

The concentration gradient then drives the nuclides into the surrounding rock.

The results of the numerical calculations are shown in Figures 3 and 4 for ^{135}Cs . In both Figures 3 and 4 the results obtained using the time-temperature-dependent diffusion coefficient are compared with the results using a constant diffusion coefficient evaluated at 58°C , the ambient temperature that is reached only after thousands of years near a repository. Figure 3 shows the ^{135}Cs concentration in the gap predicted using a time-temperature-dependent diffusion coefficient is always lower than that predicted using a constant ambient-temperature diffusion coefficient, except at $t = 0$ and at large times

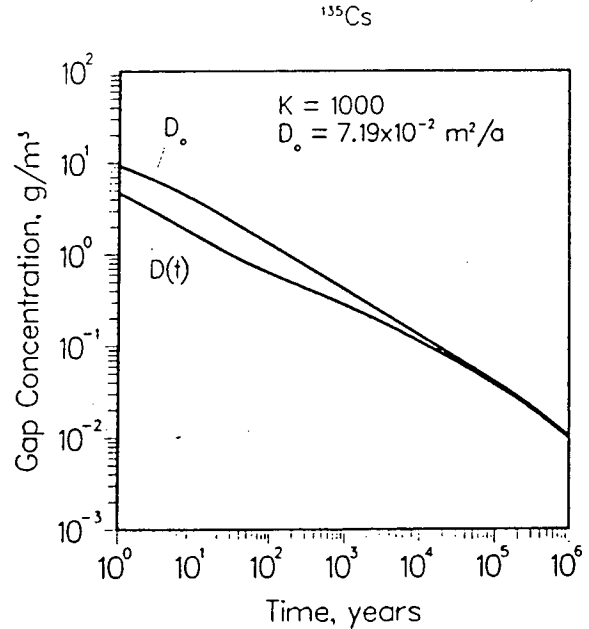


Fig. 3. Concentration of ^{135}Cs in the void space

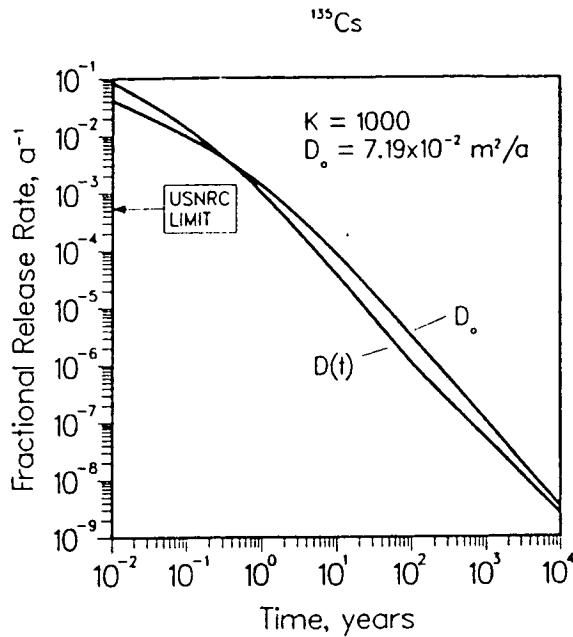


Fig. 4. Fractional release rate of ¹³⁵Cs into rock, normalized to initial inventory

when the two predictions converge. The reason for the lower gap concentration predicted by the time-temperature-dependent diffusion coefficient is shown in Figure 4. At very early times the higher diffusion coefficient causes higher mass transfer rate from the gap into the porous rock, depleting the concentration in the gap. Figure 4 shows that after one year the release rate predicted using a time-temperature-dependent diffusion coefficient becomes lower than that using a constant diffusion coefficient for ambient temperature. For most practical time scales of interest, the release rate predicted by using a constant ambient-temperature diffusion coefficient is therefore conservatively higher.

4. Conclusion

A method has been developed for incorporating the effect of temperature in predicting the release rate of readily soluble radionuclides in water-saturated porous rock. The analytic solution has been illustrated with typical nuclear waste repository conditions. The results help to justify the use of constant-diffusion-coefficient models and give a quantitative measure of the error involved in doing so. The effect of a space-dependent diffusion coefficient is yet to be investigated.

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LAWRENCE BERKELEY LABORATORY
TECHNICAL INFORMATION DEPARTMENT
UNIVERSITY OF CALIFORNIA
BERKELEY, CALIFORNIA 94720