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ANALYTICAL METHODS FOR PREDICTING CONTAMINANT TRANSPORT

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ABSTRACT

This paper summarizes some of the previous and recent work at the University of California on analytical solutions for predicting contaminant transport in porous and fractured geologic media. Emphasis is given here to the theories for predicting near-field transport, needed to derive the time-dependent source term for predicting far-field transport and overall repository performance. New theories summarized include solubility-limited release rate with flow backfill in rock, near-field transport of radioactive decay chains, interactive transport of colloids and solute, transport of carbon-14 as carbon dioxide in unsaturated rock, and flow of gases out of and into a waste container through cracks and penetrations.

1. INTRODUCTION

An extensive body of analytical theory exists to predict the isolation performance of geologic repos-itories. Important contributions have been made by workers in Sweden, France, Japan, UK, Canada, Switzerland, U.S., and elsewhere. The theories are in the form of analytical solutions to the equations for diffusive-advective transport of contaminants in ground water or in gases. They provide means to predict the transport of radionuclide chains in porous and fractured media, cumulative releases to the environment, rates of release of dissolved species from buried waste solids into ground water, migration of radionuclides in salt, transport of gaseous contaminants released to unsaturated rock, corrosion rates of copper canisters, and transport of colloids in porous and fractured media.

An analytic solution is frequently the first mathematical formulation of a given transport problem. Although they may be limited to simple geometries, analytical solutions provide quantitative insight into the importance of the phenomena. They provide guidance for experiments to validate the theories and to obtain necessary parameters for their application. The formulation of the governing equations provides the mathematical framework for more detailed numerical calculations in more complicated geometries and for space-time-dependent properties.

In this paper we review the principal features of some of the analytical solutions that have been developed in our research at Berkeley, with illustrations of their application.

2. SOLUBILITY-LIMITED RELEASE INTO POROUS ROCK

Our first near-field analyses presented the analytic solutions for the time-dependent mass transfer of low-solubility radioactive species by diffusion and convection into surrounding rock [1,2]. For a conservative estimate, we assumed that each elemental species is at its solubility limit in ground water at the waste surface. The results are illustrated in Figure 1, where the mass release rate of a long-lived species normalized to its inventory is plotted as a function of the upstream pore velocity for flow around a cylindrical waste solid or, at low flow, around an equivalent sphere. Chambré's exact analytical solution for the time-dependent release rate is simplified to an asymptotic equation for the steady-state release of a stable species, if the Peclet number exceeds four. This is illustrated by the straight line in Figure 1. There are no arbitrary or adjustable parameters in the analytical results. Each parameter, such as the diffusion coefficient, porosity, upstream pore velocity, and retardation coefficient, can be measured by a separate and independent experiment. Others have used boundary-layer approximations to obtain similar approximate forms of this asymptotic solution, but such approximations do not yield the important time-dependent release, nor do they reveal the large deviations that must occur at low flow, illustrated by the dashed line in Figure 1.

A subsequent analysis by Chambré [3] gives an explicit equation for the steady-state mass transfer when backfill with negligible flow is interposed between waste and rock. The exact solution for the time-dependent mass transfer through backfill and into porous rock has been given [4]. This solution applies for negligible flow in backfill. A more recent solution [5] includes the effect of flow in backfill as well as in porous rock.

Temperature changes can affect several properties than influence mass transfer. For those repositories wherein the expected flow rates are so low that molecular diffusion controls near-field mass transfer, the important effect of heating is to change the solubility and to increase the diffusion coefficient. Chambré's equations have been reformulated [6] to allow for an arbitrary time-dependent temperature.

3. SOLUBILITY-LIMITED RELEASE INTO FRACTURED ROCK

The first analysis of mass transfer involving diffusive transport through porous backfill and diffusive-convective transfer into planar fractures in surrounding rock was done by Neretnieks. His analysis was limited to steady state. Diffusion directly from backfill into the porous rock matrix was neglected. From a boundary-layer approximation he obtained a steady-state estimate of the diffusive-convective mass transfer from backfill to ground water flowing in the fracture. Time-dependent mass transfer into fracture water can be predicted from Chambré's exact equations for the time-dependent mass transfer for flow around a cylinder [2].

Kang *et al.* [7] have analyzed the time-dependent mass transfer through backfill directly into rock fractures, assuming no flow in backfill. Their results are illustrated in Figure 2. For low-flow repositories, in which molecular diffusion controls the mass transfer into fracture water, Ahn has analyzed the time-dependent mass transfer from a cylindrical waste through backfill and into both the rock matrix and the fracture [8].

4. RELEASE LIMITED BY SOLID-LIQUID REACTION RATE

The analytical solutions discussed above for solubility-limited mass transfer have a clear meaning for a single-component waste solid that has a well-defined solubility appropriate to the chemical environment at the waste surface. Solubility is a conservative upper-limit boundary concentration, if effects of colloids can be neglected. The same equations can be applied to any waste constituent of low enough solubility so that the species will form a precipitate at the waste surface as the waste solid reacts with water. This analysis can be expected also to be realistic if the solid-liquid reaction rate is rapid enough relative to the rate of mass transfer in the exterior porous-fractured media.

Our more-detailed analyses adopt a reaction-rate boundary condition that assumes zero-order forward reaction and first-order back reaction between water and waste matrix. Our time-dependent analysis [9] assumes diffusion-controlled mass transfer in the surrounding porous rock. It shows that for both spent uranium fuel and borosilicate glass waste the concentration of dissolved matrix builds up to within a fraction of a percent of the effective solubility within a short time after the beginning of dissolution. This means that after a few days or months of waste contacting water, release rate is controlled by mass transfer through the rock and not by chemical reaction rate.

Hughes, Marples and Stoneham [10] have suggested that for rapid flow the mass transfer will be controlled by chemical reaction rate rather than by exterior field diffusion and convection. Chambré has developed an analytical solution for this problem at steady state, adopting the solid-liquid reaction-rate equation described above [11]. The steady-state release rate of silica from borosilicate glass and uranium from spent fuel will be affected by chemical reaction rate only at Peclet numbers well above 100, far beyond any flow rate reasonably expected in a repository. We do not expect chemical reaction rate to control release rate of the waste matrix and of low-solubility species in the repository environments usually considered.

However, the release rates of soluble species, such as Cs-135, I-129, and Tc-99, from the waste matrix are not well defined. It appears that for both spent fuel and borosilicate glass soluble species may be released congruently with the solid-phase transformation of the waste matrix. If so, we must use an experimental alteration rate as the boundary condition. Mass transfer analyses can then predict the transient releases from backfill into rock, similar to the mass-transfer analysis of readily soluble species discussed in the next section. Local conditions may result in solubility limits of Tc-99, if in a sufficiently reducing environment, and of Cs-135, to form pollucite in the presence of borosilicate glass. Experiments and theory to better define the alteration rates and congruency should receive high priority.

5. RELEASE RATES OF READILY SOLUBLE SPECIES

In a spent-fuel waste package the soluble cesium and iodine accumulated in fuel-cladding gaps, voids, and grain boundaries of spent fuel rods are expected to dissolve rapidly when groundwater penetrates the fuel cladding. Even though dissolution may be rapid, the rate of release of these soluble species from the waste package will be limited by the rate of mass transfer of the dissolved species into the surrounding porous media. Chambré has developed the analytic solution for the fractional release rate, assuming instantaneous dissolution of the soluble species into ground water that has penetrated in the waste package voids, and assuming that mass-transfer into surrounding porous rock is controlled by molecular diffusion [12].

The results are illustrated in Figure 3, assuming that ten percent of the total cesium, iodine and technetium is readily soluble "gap" activity and assuming void water equivalent to a 10-cm layer between the fuel rods and the surrounding rock. With these assumed parameters, the fractional release rates of the soluble activity are usually orders of magnitude greater than the fractional release rate of the waste matrix. Because of the assumed negligible sorption of iodine in the surrounding rock, the early fractional release rates of iodine-129 and technetium-99 are lower than the release rate of strongly sorbing cesium. Consequently, iodine and technetium-99 are not depleted so rapidly by release in early times, resulting in their greater

fractional release rate of iodine after about 20 years.

6.   RELEASE OF RADIOACTIVE DECAY CHAINS

The release rate of a species in a radioactive decay chain will be governed by chemical and diffusive properties of that species and of all its precursors. Chambré has developed the analytical solutions for the time-dependent diffusion of an n-member decay chain through backfill and the release of those species into surrounding porous rock. For the typical decay chain

$$^{234}U \rightarrow ^{230}Th \rightarrow ^{226}Ra \rightarrow$$

we assume that uranium is solubility-limited by uranium precipitate at the waste surface. Because thorium and radium are formed continuously in the precipitate, we expect that these daughters are released congruently with the release of uranium. Because of the shorter half lives of the daughters, we assume that their concentrations do not approach solubility. The analytical solution for the mass transfer of a decay chain of arbitrary length has been obtained. Results are illustrated in Figure 4.

7.   RELEASES IN UNSATURATED ROCK

7.1   Releases if there are no diffusion pathways

The U.S. Yucca Mountain project proposes to predict release rates from waste emplaced in unsaturated tuff by assuming that infiltration water drips on a waste package and permeates through penetrations in a failed container [13]. This is assumed to occur only after the thermal period. During the thermal period hot waste is expected to evaporate infiltration water at atmospheric pressure, resulting in no liquid pathways for transport of radionuclides from the waste package. Assuming that releases to surrounding rock occur only after water fills the container and overflows, we have developed analytical solutions for the rates of release by this "wet-drip" scenario [14]. For a low-solubility element, the release rate of an isotope is given by the product of the volumetric drip rate, the elemental solubility, and the time-dependent isotopic fraction.

In Figure 5 we show bulk-flow solubility-limited releases for plutonium isotopes in spent fuel and borosilicate glass. The mass release rate of each isotope is normalized to its 1000-year inventory. Solubilities are from Bruton's EQ3/6 calculations [15,16]. From data on void volumes within containers, and for an estimated Darcy velocity of 0.5 mm/yr [13], the container fill time is about 8,000 years for spent fuel and 1,400 years for borosilicate glass. Assuming first water penetration at 1,000 years, the first overflow release from the spent-fuel container is at 9000 years. The release rate of 6,580-yr Pu-240 decreases rapidly with time because of decay, allowing greater concentrations of the longer-lived Pu-239 and Pu-242 within the constraints of elemental solubility. As 24,400-yr Pu-239 decays, greater solubility-limited concentrations of 379,000-yr Pu-242 are possible and its release rate increases. As compared with spent fuel, the lower solubility predicted for plutonium in borosilicate glass waste overcomes the lower inventory, resulting in a three-fold lower fractional release rate than for spent fuel.

For this same scenario, Sadeghi et al. [14] have developed the analytical solutions for the time-dependent release rates of readily soluble species in the fuel-cladding gap, fuel plenum, and grain boundaries of spent fuel. The equations for alteration controlled releases have also been derived, assuming a constant rate of alteration of the spent-fuel matrix and congruent release of soluble species from the matrix. Numerical results have been presented elsewhere [17].

7.2   Effect of Diffusion Pathways

The above wet-drip scenario for unsaturated rock assumes that the porous rock will not contact the waste package or that the annular gap between container and rock will not fill with sediments. Otherwise, after the waste container has degraded, species dissolved in ground water at the waste surface can diffuse through porous corrosion products and through ground water in the partly saturated pores of the surrounding rock. Resulting transient diffusive mass-transfer rates can be as much as three orders of magnitude greater than the bulk-flow solubility-limited release rate at the low flow rates predicted for the tuff repository, depending on the magnitude of the diffusion coefficient. Therefore, key issues of validating waste-package release-rate predictions for the tuff repository include the long-term integrity of the air gap and the diffusion coefficient for dissolved species in unsaturated and saturated tuff [18].

8. TRANSPORT OF COLLOIDS AND SOLUTE

Applications of the solubility-limited near-field release concepts have assumed that the upper limit of concentration of a dissolved species is determined by its elemental solubility and the isotopic fraction of that species. Colloids can result in effective concentrations far greater than solubility. Previous studies [19,20] have discussed the formation and transport of colloids in porous media, including removal of colloids by filtration and sedimentation. Colloids can migrate faster than solute because of weaker sorption on stationary solids and because of hydrodynamic chromatography of colloidal particles in flow channels [19]. However, the migration of colloids and pseudocolloids can be retarded by their interaction with solute, and the migration of solute in local equilibrium with colloids can be more rapid than if colloids were not present.

Hwang et al. [21] have developed a new quantitative analysis to predict the interactive migration of pseudocolloids and solute in porous and fractured media. Solute is assumed to sorb reversibly on colloids to form pseudocolloids. One-dimensional transport in the fracture is assumed, with equilibrium sorption of both solute and colloids on fracture surfaces and sediments. Only solute is assumed to diffuse into the rock matrix. Figure 6 illustrates the predicted concentrations of solute and pseudocolloids as a function of distance from the fracture inlet, for various values of the solute-colloid equilibrium constant K_{d3}. For large K_{d3} the solute is strongly sorbed on the pseudocolloid. Strong interaction causes the solute to transport at a speed near that of the colloid. For a given value of K_{d3}, the concentrations of solute and pseudocolloid are in constant ratio. Solute-colloid interaction reduces the effective migration speed of solute species sorbed on the pseudocolloids.

We have also developed analytical solutions for the transport of colloids that are in local equilibrium with solute as their dissolved species.

9. RELEASE RATES IN A SALT REPOSITORY

High-level waste packages emplaced in natural salt will be surrounded by consolidated salt a few years after emplacement. Hot salt near a waste package expands against the waste package and surrounding salt. The local compressive stresses and the expansion of grain-boundary brine in salt cause transient migration of brine outward into cooler salt. To determine the extent to which advection by brine in grain boundaries is an important transport mechanism for released radionuclides, we have developed the analytical solution for the time-dependent migration of brine after salt consolidation. This provides the convective term for calculating diffusive-convective release of dissolved species from a waste solid in salt.

10. EFFECT OF A LOCALIZED PRECIPITATION FRONT

Garisto and Garisto [22] have pointed out the possibility of localized precipitation fronts in the near field of a waste solid. A precipitation front can result from a spatial variation in temperature, redox potential, or other parameters that can affect solubility. Neretnieks [23] has analyzed the moving redox front caused by diffusion and reaction of peroxide, formed from α-radiolysis. Our analytical solution [24] shows the effect of a nearby precipitation front on the diffusion-controlled release rate of a dissolved species from a waste solid. A nearby front can increase the concentration gradient for diffusive mass transfer. When sufficiently close to the waste surface it can cause release rates to be controlled by chemical reaction rate.

11. RELEASE OF GASEOUS CARBON-14

Spent-fuel waste has been shown to release gaseous carbon-14 from its cladding surfaces when heated. Escape of gaseous carbon-14, as carbon dioxide, through cracks and penetrations in waste containers is important for repositories in unsaturated media, where air in surrounding rock fractures can transport the carbon-14 to the surface. Transport of carbon-14 will be retarded by absorption of carbon dioxide in ground water in the porous matrix. Light et al. [25] have developed the analytic solution for the advective transport of carbon-14 in unsaturated rock, assuming bicarbonate absorption equilibrium at the fracture-matrix interface. Results for an assumed band release of 200 curies from a planar source are shown in Figure 7. The maximum temporal concentration of carbon-14 in air near the surface, 350 m up from the waste, is plotted as a function of the time duration of band-release. The insensitivity of the maximum concentration to the release rate has been noted and explained in our earlier studies [26, 27].

5

12. FLOW OF GAS THROUGH CONTAINER PENETRATIONS

In a repository in unsaturated rock, with atmospheric pressure at the outer surface of a waste container, cracks and penetrations will eventually appear in the container, allowing gas to flow out of the container. As the waste cools air can leak in, resulting in oxidation of Zircaloy cladding and uranium dioxide even before water can intrude into the failed container. Zwahlen *et al.* [28] has analyzed the time-dependent flow of gas into and out of a container, as affected by effective penetration size. For penetrations equivalent to a 20 micrometer hole or larger, the rate and amount of air flow into a container will be controlled by the rate of cooling of the waste and will be affected little by hole size.

13. SUMMARY

Analytical solutions for near-field mass transfer now provide the most comprehensive tools for predicting source terms and release rates from waste emplaced in a geologic repository. The analytical solutions define the parameters that are important and the sensitivity of release rates to these parameters. New and better experiments are needed to supply reliable values for these parameters. Many new analytical solutions have emerged, in response to new technological issues that must be quantified and assessed. Experiments should be devised to validate the mass-transfer theories that have emerged. The new analytical studies of interactive transport of colloids and solute are based on interaction parameters that need experimental confirmation.

Analytical solutions are usually limited to simple geometries and to linear relations. New numerical codes tailored to near-field transport are also needed.

13. ACKNOWLEDGMENT

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$R = 48.6$ cm
$\epsilon = 0.1$
$D = 315$ cm^2/yr
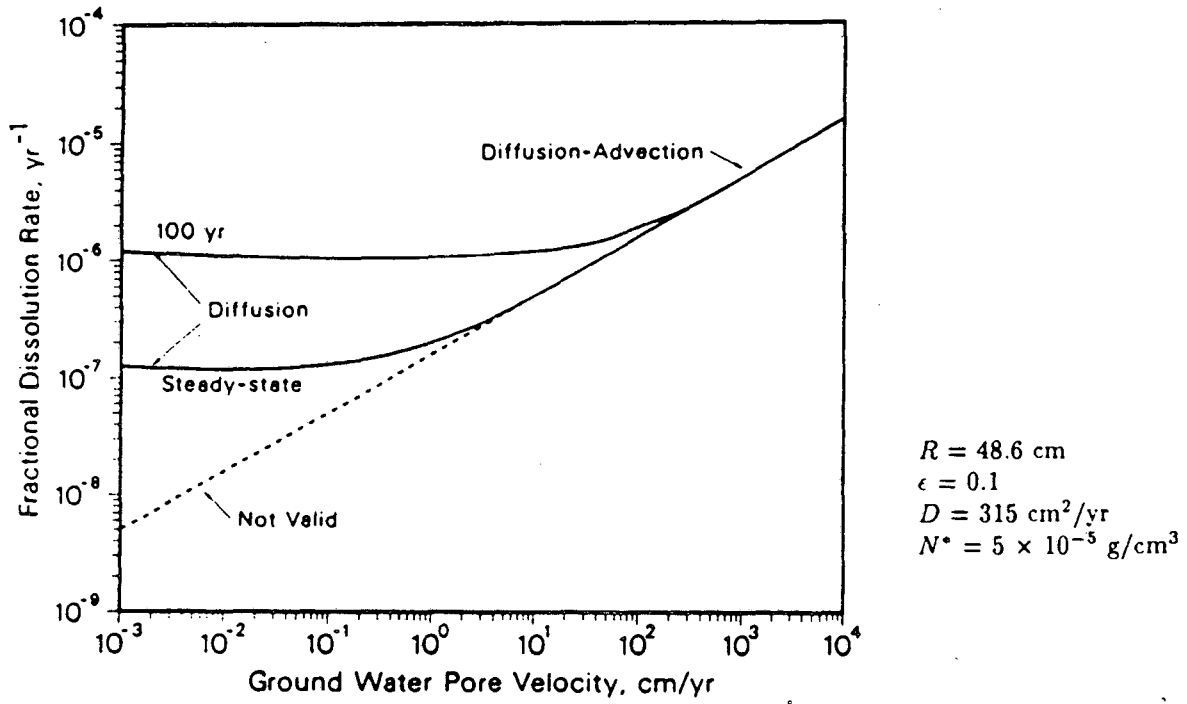$N^* = 5 \times 10^{-5}$ g/cm^3

Figure 1. Release Rates of Solubility-Limited Species Into Porous Rock, Normalized to Species Inventory.

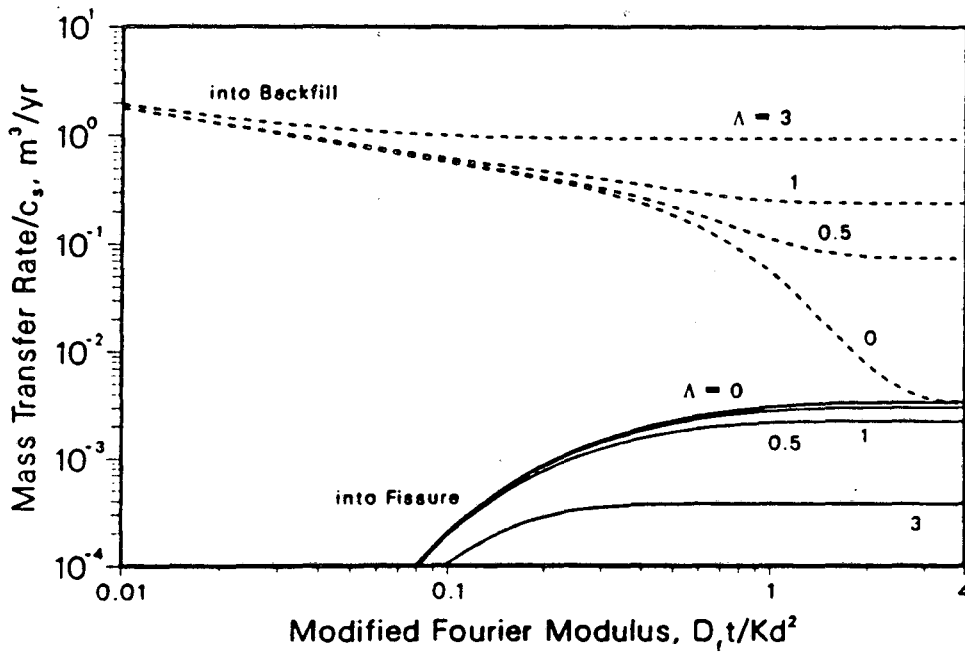

Figure 2. Normalized Mass Transfer Rate into the Backfill and into the Fissure, for Sh= $hd/D_f\epsilon$ = 3.2 and Various Values of $\Lambda = \sqrt{\lambda d^2 K/D_f}$. Symbols and Parameters are Defined in [7].
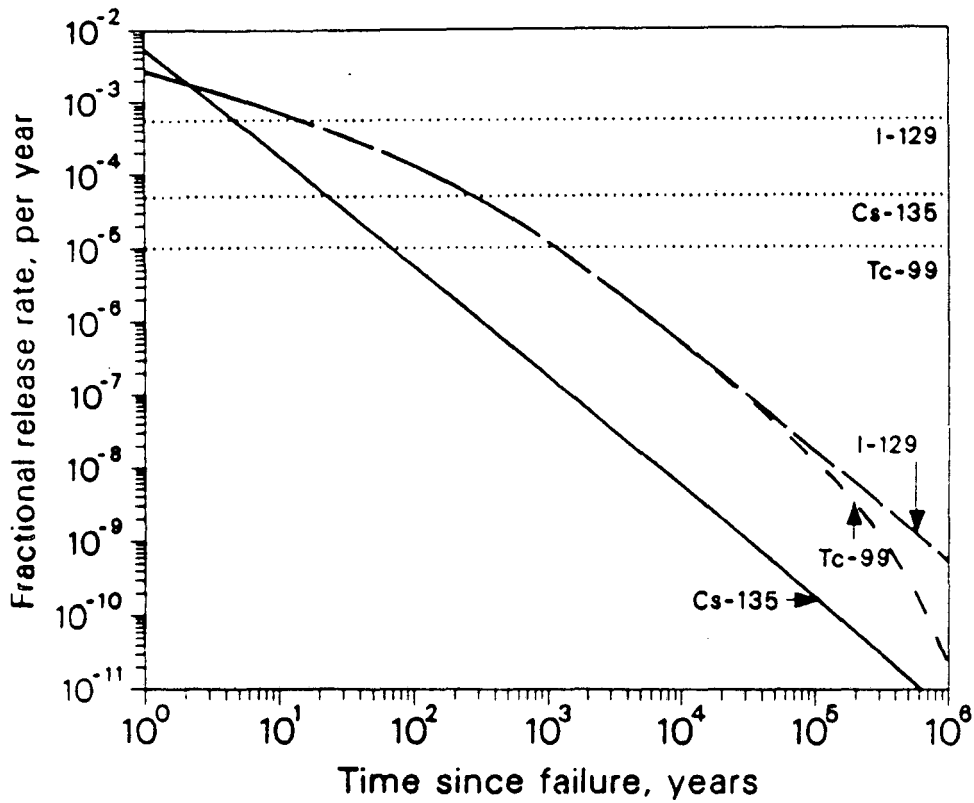
Figure 3. Release Rates of Readily Soluble Species Into Porous
Rock. Symbols and Parameters are Defined in [12].
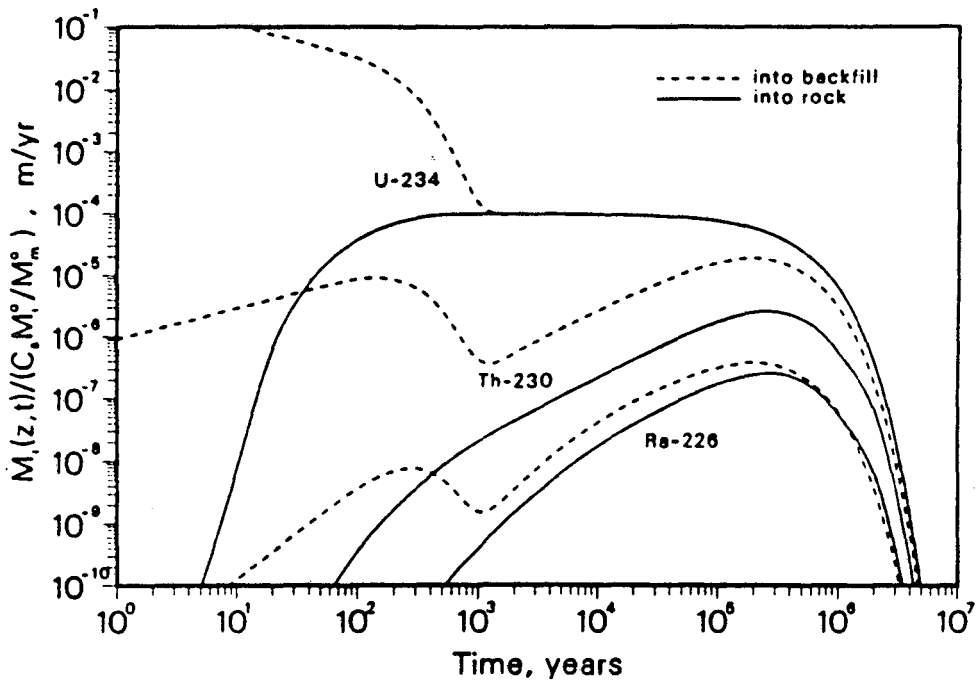
Figure 4. Mass Fluxes into the Backfill at $z = 0$ and into the Rock
at $z = L$, Normalized by the Initial Isotopic Fraction
of the Chain Member. Symbols and Parameters are
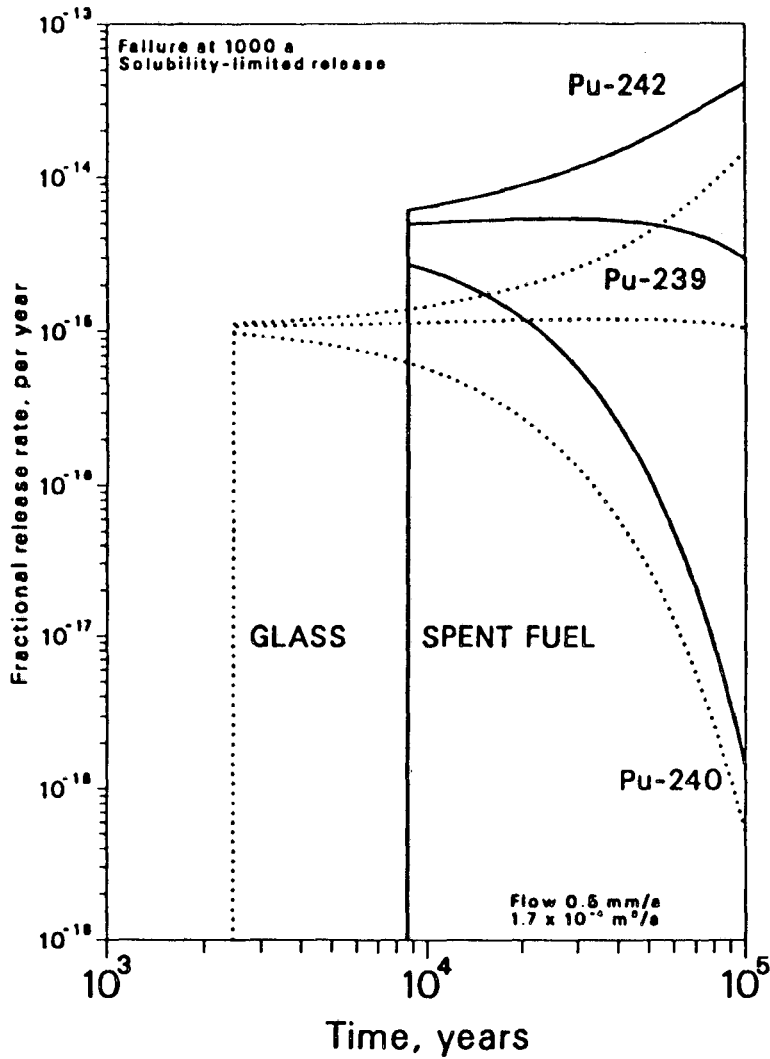Defined in [5].

Figure 5. Fractional Release Rate of Plutonium, Wet Drip Scenario, Solubility-Limited Release, Container Failure at 1,000 Years. Symbols and Parameters are Defined in [14].
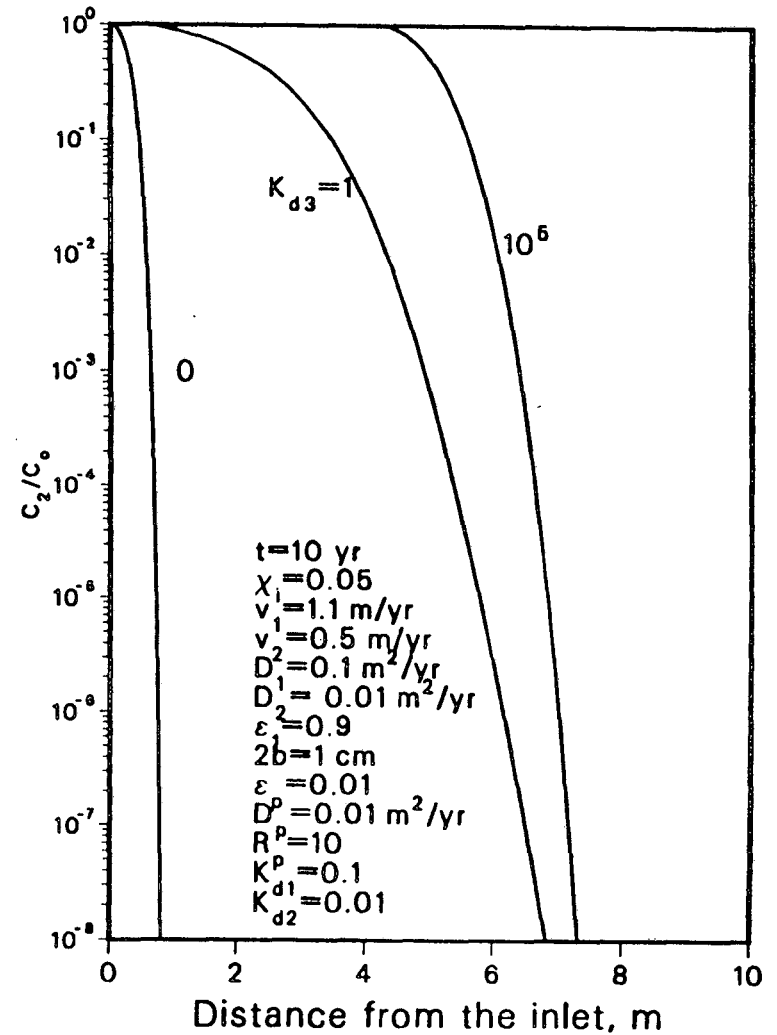

Figure 6. Concentration of Solute in Local Equilibrium With Pseudocolloid, as a Function of Solute-Pseudocolloid Distribution Coefficient K_{d3}. Symbols and Parameters are Defined in [21].
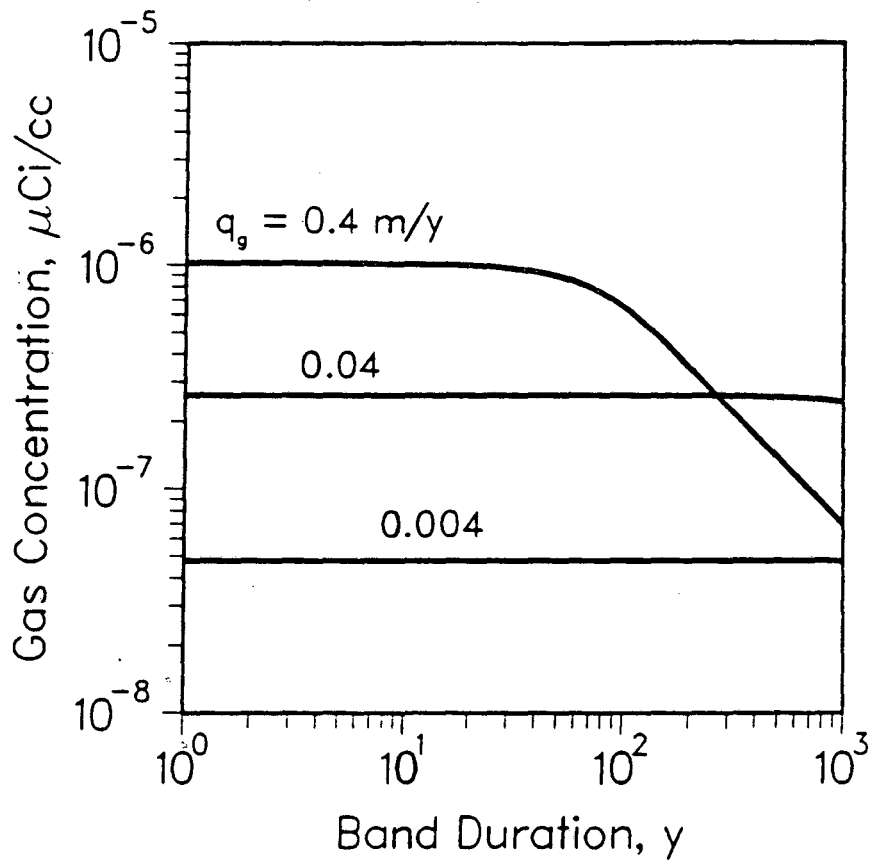
Figure 7. Peak Concentration of ^{14}C 350 m above a Repository treated as an Infinite Plane Source, as a Function of Duration of Release and Gas Darcy Velocity. Symbols and Parameters are Defined in [25].

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