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**Migration of radionuclides in fissured  
rock – The influence of micropore  
diffusion and longitudinal dispersion**

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Royal Institute of Technology, December 1979

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MIGRATION OF RADIONUCLIDES IN FISSURED ROCK — THE INFLUENCE  
OF MICROPORE DIFFUSION AND LONGITUDINAL DISPERSION

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## SUMMARY

The migration of radionuclides in the fissures in the bedrock surrounding a repository is discussed. A one-dimensional transport model is presented. It includes diffusion of the nuclides into the microfissures of the rock, and linear sorption and longitudinal dispersion in the bedrock. An analytical solution to the model is given in terms of an infinite integral. The integrand is a sometimes highly oscillatory function of the system parameters. A special integration method is developed to evaluate the infinite integral. The method utilizes the oscillatory behavior of the integrand.

The assessment of input parameters is discussed in some detail. Dimensionless breakthrough curves are given for the approximate range of variation of the input parameters. Calculations are made for a repository of spent fuel surrounded by fissured but fairly good rock ( $K_p = 10^{-9}$  m/s and fissure spacing  $S = 50$  m). Longitudinal dispersion may significantly affect the amount of radioactive material reaching the biosphere.

Radionuclides, which would decay completely without longitudinal dispersion, may arrive in non-negligible concentrations. Dispersion effects of the magnitude considered in this study can significantly diminish the retardation effects of matrix diffusion.

## INTRODUCTION

In a previous paper by Neretnieks (1980), the migration of radionuclides in the fissures in the bedrock surrounding a repository is discussed. The important transport mechanisms were shown to be flow of water and nuclides in the fissures, and transport of nuclides from the water in the fissures into the microfissures of the rock by diffusion. The main retarding mechanism is the diffusion of the nuclides into the microfissures of the rock and their sorption.

In this paper, a model is presented which extends the previous analysis by treating the finite block size of the rock as well as longitudinal dispersion in the bedrock.

## MATHEMATICAL MODEL

When water which contains radionuclides flows in fissured rock, the nuclides will migrate into the porous structure of the rock by diffusion. Many of the radionuclides will sorb on the walls of the microfissures. The concentration of the nuclide in the flowing water is thereby decreased, and the nuclide migration velocity will be slower than the water velocity. In the following analysis, the rock is regarded as consisting of porous blocks separated by fissures (a double-porosity medium). The water flow, however, is assumed to take place only in the fissures. The sorption is generally considered to occur in three distinct stages:

- diffusion of the component from the water in the fissures to the external surface of the blocks (external or film diffusion)
- diffusion through the porous network of the blocks (internal diffusion)
- the sorption process itself, when the component is bound to some sorption site in the pores or microfissures.

The dispersion in the bedrock affects the concentration of the radioactivity arriving at any time at a point of interest in two ways: a large dispersion tends to cause greater dilution, and a large dispersion will cause some radioactive material to arrive earlier than the major peak. The interplay between these effects is important in determining the amount of radioactivity which will escape to the biosphere.

In the mathematical development we make the following major assumptions:

- a) the block diameter is small in comparison with overall distance, and the rock is macroscopically uniform
- b) the dispersion of components in the bedrock is described by the "usual" (Fried and Combarous, 1971) one-dimensional differential equation for longitudinal dispersion
- c) the sorption equilibrium relationship is linear (trace amounts of radionuclides)
- d) local sorption equilibrium is reached instantaneously
- e) the movement of solute within the blocks can be described mathematically by a Fick's law diffusion equation, where the effective intraparticle diffusion coefficient is constant and independent of concentration
- f) the blocks may, for the description of internal diffusion, be regarded as spheres

For flow and sorption from the water in the fissures we have:

$$\frac{\partial C_f}{\partial t} + U_f \frac{\partial C_f}{\partial z} - D_L \frac{\partial^2 C_f}{\partial z^2} = - \frac{1-\epsilon_f}{\epsilon_f} \left( \frac{\partial q}{\partial t} \right) - \lambda C_f \quad (1)$$

The terms in this equation represent accumulation in the water in the fissures, convective transport, transport by axial dispersion, accumulation in the blocks, and radioactive decay.

Fick's law of diffusion for radial movement of solute in a spherical particle (including radioactive decay) is:

$$\frac{\partial q_i}{\partial t} = D_a \left( \frac{\partial^2 q_i}{\partial r^2} + \frac{2}{r} \frac{\partial q_i}{\partial r} \right) - \lambda_d q_i \quad (2)$$

Note that the definition of  $q_i$  as a local solute concentration includes solute both in the solid rock and in the water in the pores. We assume here that solid diffusion effects are negligible, and that the transport of solutes within the blocks may be effectively described by diffusion in the solution phase only. Thus, if it is assumed that the driving force for diffusion is the intrapore concentration gradient  $\partial C_p / \partial r$ , an alternate differential equation for intraparticle diffusion may be written:

$$\epsilon_p \frac{\partial C_p}{\partial t} + \frac{\partial C_s}{\partial t} = \epsilon_p D_p \left( \frac{\partial^2 C_p}{\partial r^2} + \frac{2}{r} \frac{\partial C_p}{\partial r} \right) - \lambda_d (\epsilon_p C_p + C_s) \quad (3)$$

The two terms on the left hand side give the accumulation in the pore fluid phase and in the solid phase, respectively. On the right hand side, the terms describe diffusion in the pore fluid phase, and radioactive decay of the nuclide in the pores and solid rock.

By definition :

$$q_i = \epsilon_p C_p + C_s \quad (4)$$

Further, using assumptions c) and d), we obtain :



$$q_i = K C_p \quad (5)$$

From equations (4) and (5) we obtain for  $C_s$ :

$$C_s = (K - \epsilon_p) C_p \quad (6)$$

Differentiating (6) to obtain  $\frac{\partial C_s}{\partial t}$ , and substituting into equation (3), we get:

$$\frac{\partial C_p}{\partial t} = \frac{\epsilon_p D_p}{K} \left( \frac{\partial^2 C_p}{\partial r^2} + \frac{2}{r} \frac{\partial C_p}{\partial r} \right) - \lambda C_p \quad (7)$$

If  $q_i/K$  is substituted for  $C_p$ , equation (7) becomes identical to equation (2) when  $D_a = \epsilon_p D_p / K$ .  $D_a$  is called the apparent diffusivity.

In a system which is initially free of nuclides, and in which the inlet ( $z=0$ ) nuclide concentration suddenly is increased to  $C_0$  at time zero, the initial and boundary conditions are:

$$C_f(0,t) = C_0 e^{-\lambda t} \quad (8)$$

$$C_f(\infty,t) = 0 \quad (9)$$

$$C_f(z,0) = 0 \quad (10)$$

$$C_p(0,z,t) \neq \infty \quad (11)$$

$$C_p(r_0,z,t) = C_p \Big|_{r=r_0} \quad \text{given by } \frac{\partial q}{\partial t} = \frac{3k_f}{r_0} (C_f - C_p \Big|_{r=r_0}) \quad (12)$$

$$C_p(r,z,0) = C_s(r,z,0) = 0 \quad (13)$$

Boundary condition (12) is the link between equations (1) and (7). It states mathematically that the mass entering or leaving the blocks must equal the flow of mass transported across a stagnant fluid film at the external surface. It is assumed that no mass accumulation occurs within the stagnant fluid film. Boundary condition (8) is used here because it describes a constant leach rate for a body containing a decaying nuclide. This is an important case for study.

#### SOLUTION OF THE EQUATIONS

An exact solution of equations (1) and (7), subject to the boundary conditions (8) - (13), is presented elsewhere (Rasmuson and Neretnieks, 1980). Here we summarize the steps in that solution.

First the solution for a stable species is obtained. Then, utilizing the properties of the Heaviside step function, the results for a finite release time are obtained by combination of two such solutions. Finally, the decay of the radionuclide is accounted for.

For a stable species ( $\lambda_d = 0$ ), the solution is:

$$\frac{C_f}{C_o} = \frac{1}{2} + \frac{2}{\pi} \int_0^{\infty} \exp\left(\frac{U_f z}{2D_L} - z \sqrt{\frac{\sqrt{x'(\lambda)^2 + y'(\lambda)^2} + x'(\lambda)}{2}}\right) \sin\left(\sigma \lambda^2 t - z \sqrt{\frac{\sqrt{x'(\lambda)^2 + y'(\lambda)^2} - x'(\lambda)}{2}}\right) \frac{d\lambda}{\lambda} \quad (14)$$

with:

$$x'(\lambda) = \frac{U_f^2}{4D_L^2} + \frac{\gamma}{mD_L} H_1 \quad (15)$$

$$y'(\lambda) = \frac{\sigma\lambda^2}{D_L} + \frac{\gamma}{mD_L} H_2 \quad (16)$$

where:

$$m = \frac{\epsilon_f}{1-\epsilon_f}$$

$$\gamma = \frac{3D_a K}{r_o^2}$$

$$\sigma = \frac{2D_a}{r_o^2}$$

$$R_F = \frac{r_o}{3k_f}$$

$H_1$  and  $H_2$  are complicated functions of  $\lambda$  and  $\nu = \gamma R_F$ :

$$H_1(\lambda, \nu) = \frac{H_{D_1} + \nu(H_{D_1}^2 + H_{D_2}^2)}{(1 + \nu H_{D_1})^2 + (\nu H_{D_2})^2} \quad (17)$$

$$H_2(\lambda, \nu) = \frac{H_{D_2}}{(1 + \nu H_{D_1})^2 + (\nu H_{D_2})^2} \quad (18)$$

where:

$$H_{D_1}(\lambda) = \lambda \left( \frac{\sinh 2\lambda + \sin 2\lambda}{\cosh 2\lambda - \cos 2\lambda} \right) - 1 \quad (19)$$

$$H_{D_2}(\lambda) = \lambda \left( \frac{\sinh 2\lambda - \sin 2\lambda}{\cosh 2\lambda - \cos 2\lambda} \right) \quad (20)$$

For small values of  $\lambda$  equations (19) and (20) simplify (for  $\lambda < 0.1$  the relative error is less than  $10^{-3}$ ) to:

$$H_{D_1} = \frac{4\lambda^4}{45} \quad (21)$$

$$H_{D_2} = \frac{2\lambda^2}{3} \quad (22)$$

It follows that  $\lim_{\lambda \rightarrow 0} H_{D_1} = 0$  and  $\lim_{\lambda \rightarrow 0} H_{D_2} = 0$ . For high values of  $\lambda$  equations (19) and (20) simplify (for  $\lambda > 5$  the relative error is less than  $10^{-3}$ ) to:

$$H_{D_1} = \lambda - 1 \quad (23)$$

$$H_{D_2} = \lambda \quad (24)$$

For the case of a finite step boundary condition (which implies that the inlet concentration is again decreased to 0 at time  $t_0$ ):

$$\begin{aligned} C_f'(0,t) &= C_0 & 0 \leq t \leq t_0 \\ C_f'(0,t) &= 0 & t > t_0 \end{aligned} \quad (25)$$

the Laplace transform of  $C_f'$  becomes:

$$\tilde{C}_f'(z,s) = (1 - e^{-t_0 s}) \tilde{C}_f(z,s) \quad (26)$$

and

$$C_f' = C_f(z,t) - C_f(z,t-t_0) H(t-t_0) \quad (27)$$

where  $H$  is Heaviside's step function.

The following dimensionless quantities are introduced:

$$\begin{aligned} \delta &= \frac{\gamma z}{m U_f} && \text{bed length parameter} \\ R &= \frac{K}{m} && \text{distribution ratio} \\ \text{Pe} &= \frac{z U_f}{D_L} && \text{Peclet number} \\ y &= \sigma t && \text{contact time parameter} \\ v &= \gamma R_F && \text{film resistance parameter} \end{aligned}$$

The bed length parameter may be thought of as a ratio of one time, specifying the moving fluid, to another time, specifying the diffusion in particles. The distribution ratio gives the relative magnitudes of the capacity of the particles and the capacity of the fluid in the fissures. The Peclet number is a measure of the amount of hydrodynamic dispersion. High values of the longitudinal dispersion coefficient yield low Peclet numbers. The film resistance parameter may be interpreted as a ratio of the external diffusion resistance to the internal diffusion resistance.

Equation (14) now becomes:

$$\frac{C_f}{C_o} = \frac{1}{2} + \frac{2}{\pi} \int_0^{\infty} \exp\left(\frac{1}{2} Pe - \sqrt{\frac{\sqrt{(z^2 x')^2 + (z^2 y')^2} + z^2 x'}{2}}\right) \sin\left(y \lambda^2 - \sqrt{\frac{\sqrt{(z^2 x')^2 + (z^2 y')^2} - z^2 x'}{2}}\right) \frac{d\lambda}{\lambda} \quad (28)$$

with:

$$z^2 x' = Pe \left(\frac{1}{4} Pe + \delta H_1\right) \quad (29)$$

$$z^2 y' = \delta Pe \left(\frac{2}{3} \frac{\lambda^2}{R} + H_2\right) \quad (30)$$

As  $Pe \rightarrow \infty$  (no dispersion), equation (28) becomes:

$$\frac{C_f}{C_o} = \frac{1}{2} + \frac{2}{\pi} \int_0^{\infty} \exp(-\delta H_1) \sin(\sigma \theta \lambda^2 - \delta H_2) \frac{d\lambda}{\lambda} \quad (31)$$

where  $\theta = t - \frac{z}{U_f}$ .

This solution was given by Rosen (1952). In the expression for  $\theta$ ,  $z/U_f$  is the water transport time.

For a decaying species ( $\lambda_d > 0$ ), the Laplace transform of  $C_f$ ,  $\tilde{C}_f$ , becomes a function of  $s + \lambda_d$  instead of  $s$ . Hence, due to the properties of the Laplace transform, the solution becomes:

$$\left(\frac{C_f}{C_o}\right)_{\lambda_d > 0} = e^{-\lambda_d t} \left(\frac{C_f}{C_o}\right)_{\lambda_d = 0} \quad (32)$$

For a decaying species with decay constant  $\lambda_d$ , released during time 0 to  $t_0$ , the concentration at any point along the z-axis is obtained by combining equations (14) (nondecaying species) and equation (27), giving the results for finite release. Finally, equation (32) gives the concentration for the decaying species.

Because of the complicated nature of the integral expression for  $C_f/C_0$ , numerical integration must be performed. Details of the integration are given in the Appendix.

## ASSESSMENT OF INPUT PARAMETERS

The environment which this study attempts to describe, is a moderately fissured crystalline rock such as the Swedish granite and gneiss investigated in the Swedish Nuclear Fuel Safety Project (KBS, 1978).

The characteristic time for molecular diffusion in the fissures is much less than that in the blocks. The characteristic time for diffusion is given by  $l^2/D$ , where  $l$  is a characteristic length, and  $D$  is a diffusion coefficient. Since the characteristic length ("radius") of the blocks is typically not less than  $10^3$  times that of the fissures, and since the apparent diffusion coefficient in the fissures is 10 to  $10^8$  times that in the blocks (Neretnieks, 1980), the characteristic time for diffusion in the fissures should be vanishingly small in comparison with that in the blocks. Thus, any radial concentration gradients in the fissures will be completely negligible. It follows that the film resistance  $v \sim 0$ . The expressions for  $H_1$  and  $H_2$  are accordingly simplified to:

$$H_1(\lambda, v) = H_{D_1}(\lambda) \quad (33)$$

$$H_2(\lambda, v) = H_{D_2}(\lambda) \quad (34)$$

The input parameters needed in the model are now:

$K$	volume equilibrium constant	$m^3/m^3$
$\lambda_d$	decay constant	$s^{-1}$
$D_a$	apparent diffusivity	$m^2/s$
$\epsilon_f$	average fracture porosity	$m^3/m^3$
$U_f$	average fracture velocity	$m/s$
$r_o$	"effective" block radius	$m$
$D_L$	dispersion coefficient	$m^2/s$



The equilibrium data was obtained from Allard et al. (1978) and Grundfelt (1978). The decay constants  $\lambda_d$  were obtained from Handbook of Chemistry and Physics (1977-1978). The apparent diffusivity is obtained from the expression above :

$$D_a = \frac{D \epsilon_p}{K} \quad (35)$$

$D_p$  is the diffusivity in the water in the pores, and is related to the diffusivity in pure water by:

$$D_p = D_v \frac{\delta_D}{\tau^2} \quad (36)$$

where  $\frac{\delta_D}{\tau^2} < 1$  is a geometric factor.

The diffusivity of a species of low molecular weight ( $M < 500$ ) in water at ambient temperature is  $D_v \approx 1 - 5 \cdot 10^{-9} \text{ m}^2/\text{s}$ . The values of  $\frac{\delta_D}{\tau^2}$  and  $\epsilon_p$  for rocks at different confining pressures were discussed by Neretnieks (1980). In his calculations, the values  $\delta_D/\tau^2 = 0.1$ ,  $\epsilon_p = 0.005$  and  $D_v = 2 \cdot 10^{-9} \text{ m}^2/\text{s}$  were used. For comparison, these values are also used in this paper.

Fracture spacing, the size of fracture openings, and fracture porosities are not directly observable, but the related property of permeability is commonly measured. The relation between permeability, fissure spacing, orientations, and fissure width is, however, not known in practice. Snow (1968) used a model in which fissures were assumed to be of equal width and to have equal spacing. Assuming a cubic system of orthogonal fractures, Snow obtained :

$$(2b)^3 = 1/2 \frac{12\mu}{\rho g} SK_p \quad (37)$$

and

$$\epsilon_f = 3 \left(\frac{2b}{S}\right) \quad (38)$$

The average fracture velocity is calculated from:

$$U_f \epsilon_f = K_p i \quad (39)$$

The cubic grid used to describe the fissure system implies that the blocks of rock are cubes. This geometry is awkward for modeling internal diffusion. Here, the cubes are approximated by spheres having the same surface-to-volume ratio as a cubic block. This means that the total surface area which the water contacts is the same for the spheres as it would be for the cubes. Further, the amount of solid in the bed is the same. The difference is that each individual sphere has somewhat less volume, but the number of spheres is greater, giving the same total volume of solid matter. This approximation gives exactly the same uptake for short times, and deviates only slightly for larger times (Neretnieks, 1972). The surface-to-volume ratio for a spherical particle is:

$$A/V = \frac{3}{r_o} \quad (40)$$

For the cubic blocks we get:

$$A/V = \frac{6}{S} \quad (41)$$

Accordingly:

$$r_o = 0.5 S \quad (42)$$

For high values of the distribution ratio  $R$ , the influence of this parameter can be shown to be negligible. This implies that the capacity of the fluid in the fissure is negligible in comparison to the capacity of the sorbing material. In equations (28)-(30),  $R$  appears only in the expression for  $z^2 y'$ . It is clear that if the following inequality is fulfilled over the entire interval of integration:

$$R \gg \frac{2}{3} \frac{\lambda^2}{H_{D_2}} \quad (43)$$

then the influence of  $R$  is negligible.

From the limiting expressions for  $H_{D_2}$  (equations (22) and (24)) we obtain:

$$R \gg 1 \quad \lambda < 0.1 \quad (44)$$

$$R \gg \frac{2}{3} \lambda \quad \lambda > 5.0 \quad (45)$$

The condition for the range  $0.1 < \lambda < 5.0$  may easily be estimated from the limiting expressions. The first condition is always satisfied for the cases of interest. To show that the second condition is satisfied for all  $\lambda$ , we need to know the rate of convergence of the integral for  $C_f/C_o$ , i.e.  $\lambda = \lambda_{\max}$  with a fixed truncation error.  $\lambda_{\max}$  may be estimated from the argument of the exponential function in  $C_f/C_o$  (Appendix) as :

$$\lambda_{\max} = \frac{2(\frac{1}{2} Pe + c)}{\delta Pe} \left[ \sqrt{2(\frac{1}{2} Pe + c)^2 - \frac{1}{4} Pe^2 + \delta Pe} - (\frac{1}{2} Pe + c) \right] \quad (46)$$

where the truncation error is considerably less than  $e^{-c}$ .  $\lambda_{\max} = \lambda_{\max}(\delta, Pe)$  for  $c = 20$  is given in Figure 1. For the sorbing nuclides considered here,  $R$  is larger than  $\sim 10^5$  (see next section). Thus,  $\lambda_{\max}$  has to be less than  $1.5 \cdot 10^5$ . From Figure 1, it may be seen that this is the case for higher values of  $\delta$  (e.g. large distances).

In these cases there is no need to determine  $U_f$  and  $\epsilon_f$  separately, since :

$$mU_f = \frac{\epsilon_f}{1-\epsilon_f} U_f \approx (\epsilon_f \ll 1) \epsilon_f U_f = K_p i$$

giving:

$$\delta = \frac{Y z}{K_p i} \quad (47)$$

For the reasons given,  $R$  has no influence, and it will be demonstrated in the next section that  $Pe$  is independent of  $U_f$ . Equations (37)-(39) are thus superfluous for the calculations of transport times. Furthermore, the only influence of the equilibrium constant is on  $\sigma$ . Thus, if the breakthrough curve (without radioactive decay) for one nuclide is calculated for certain values of the parameters, the breakthrough curves for all other nuclides (primed) may be obtained from the relation:

$$(C_f/C_o)' = (C_f/C_o) \quad \text{if } y' = y$$

giving:

$$t' = \frac{K'}{K} t \quad (48)$$

Dispersion is due to the combined action of both a purely mechanical phenomenon and of molecular diffusion. Most of the experimental studies of longitudinal dispersion have been performed in unconsolidated porous media (Fried and Combarous, 1971). From the results of these experiments, it may be inferred that there exist five different dispersion regimes with different relative importances of molecular diffusion and mechanical dispersion. In the cases of interest here, the contribution of mechanical dispersion is predominant. In unconsolidated porous media, the following relationship has been shown to hold (Fried and Combarous, 1971):

$$D_L = \beta d U_f \quad (49)$$

where  $d$  is a characteristic particle diameter. The experimental value of  $\beta$  is  $1.8 \pm 0.4$ . Note that the Peclet number is independent of velocity:

$$Pe = \frac{z}{\beta d} \quad (50)$$

Very few experiments have been performed for the case of consolidated porous media. It is expected that dispersion will be greater in consolidated than in unconsolidated media. The pore-size distribution is wider in consolidated than in unconsolidated media, and thus, the distribution of velocities is also wider. Raimondi et al. (1959) give values of  $\beta d$  as different as 0.001 (unconsolidated medium) and 0.2 (consolidated medium) for media with the same permeabilities. A series of tests conducted by Klotz and Moser (1974) show how dispersion increases with decreasing porosity, i.e. with greater compactness; the explanation is that growing compactness leads to a greater branching of flow paths.  $D_L$  corresponds more or less to  $\epsilon^{-3}$ ,  $\epsilon$  being the total porosity.

The studies of lateral dispersion are less numerous but more recent than those of longitudinal dispersion. Experiments in unconsolidated media have been performed for Peclet numbers ranging from  $10^{-2}$  to  $10^{+4}$ , and have shown the existence of four regimes of dispersion (Fried and Combarous, 1971). For these media, lateral dispersion is small in comparison with longitudinal dispersion. Burkholder et al. (1976) used this fact to simplify their model for nuclide transport and spreading. However, this assumption may not hold for heterogeneous media. Robertson (1974), in a study of radioactive pollution from a Test Reactor Site in southern Idaho, found higher values for the lateral than for the longitudinal dispersion coefficient. They attribute this observation in part to the fissured nature of the

aquifer. In the calculations performed in this paper, the lateral dispersion effect is assumed to be negligible. However, the importance of this parameter is currently under investigation by the authors.

There are some indications that the dispersion coefficient may increase with distance (Lallemand-Barrès and Peaudecerf, 1978). This may occur in a medium in which increasingly larger channels are found when larger rock volumes are considered. In such a case, there is no obvious particle size. If a particle size could be conceived, it would seem to increase with distance. In such a case, it would be better to assume that the Peclet number is constant (independent of distance) than to assume a constant dispersivity. The data of Lallemand-Barrès and Peaudecerf have Peclet numbers ranging from 0.5 to 50, with most of the data around 5.

#### DIMENSIONLESS BREAKTHROUGH CURVES

The range of variation of the dimensionless parameters  $\delta$ , R and Pe are given in Table 1.

Table 1: Estimated ranges of the dimensionless parameters  $\delta$ , R and Pe.

$\delta$	$4.8 \cdot 10^{-7}$	-	$1.2 \cdot 10^5$
R	$7.9 \cdot 10^4$	(9.1)*	- $4.6 \cdot 10^{10}$
Pe	0.5	-	$\infty$

\* Lower value of R within brackets is that for I-129 (K = 0.005). The other value is calculated from the nuclide with the second lowest K-value (Sr-90, K = 43).

These values are based on the following approximate ranges of the input parameters.

$$\begin{array}{rcccc}
 10^{-9} & < & K_p & < & 10^{-5} \\
 0.001 & < & i & < & 0.01 \\
 1 & < & S & < & 50 \\
 10 & < & z & < & 10\ 000 \\
 43(0.005) & < & K & < & 86\ 000
 \end{array}$$

The greatest permeability value given above corresponds to a very permeable rock such as can be found near the surface at depths less than 100 m.

However, this value is very large even for shallow rock. The low permeability value was observed in several boreholes in granite and gneiss in south Sweden (KBS, 1979). The values of  $\delta_D/\tau^2$ ,  $\epsilon_p$ , and  $D_v$  estimated by Neretnieks (1980) were used in the calculations, yielding an apparent diffusivity of  $D_a = 10^{-12}/K$ .

Dimensionless breakthrough curves (without decay) for  $R \rightarrow \infty$  are shown in Figures 2-7, with  $Pe$  as a parameter.  $\delta = 10^{-8}, 10^{-4}, 10^{-2}, 1, 10^2, 10^4$  and  $Pe = 0.5, 5.0, \infty$ . It may be seen that for low values of  $\delta$ , identical curves are obtained if the dimensionless concentrations  $C_f/C_o$  are functions of  $y/\delta^2$ ,  $R/\delta$ , and the Peclet number. This is not surprising, since only the outer surface of the blocks are utilized, due to the short contact times. The solution to the model then simplifies to error-functions of the form given in Neretnieks (1980).

For high values of  $\delta$ , the curves again nearly coincide, if plotted against  $y/\delta$  (Pe constant). This is the case for long contact times, when the material behind the front reaches equilibrium.

The effect of diffusion into the rock matrix (without longitudinal dispersion), as compared to retardation by surface reaction in the fissures only, was thoroughly treated by Neretnieks (1980). When longitudinal dispersion is included, it has a pronounced effect on the breakthrough curves. For  $Pe = 0.5$ , the time of first arrival is 100-1000 times earlier than for the same case with no dispersion. Consequently, radio-nuclides which otherwise would decay to insignificant levels may arrive at the site boundaries in nonnegligible concentrations.

In Figures 8-11, the influence of  $R$  is shown. It may be seen that, if  $\delta \geq 10^{-4}$ , the breakthrough curves for  $R = \infty$  and  $R = 10^5$  are practically identical. This simplifies the calculations of breakthrough curves. For I-129 ( $R=10$ ), this condition is fulfilled when  $\delta \geq 1$ .



## APPLICATION OF THE MODEL TO A FINAL REPOSITORY OF RADIOACTIVE WASTE

Table 2 shows the most important radionuclides in a repository for spent fuel (KBS, 1978), as well as K values for granite obtained by Allard et al. (1978) on material crushed to  $\sim 0.1$  mm. Contact times for the K measurements were up to 7 months. Reducing conditions prevail in granite which contains Fe(II) (Jacks, 1978; KBS, 1978; KBS TR 90, 1978). Calculations of breakthrough curves were made for all these nuclides, at distances  $z = 1, 3, 10, 30, 100, 300, 1000, 3000$  and  $10\ 000$  m from the repository. The input parameters were  $K_p = 10^{-9}$  m/s,  $i = 0.003$  m/m,  $S = 50$  m,  $D_a = \frac{10^{-12}}{K}$  m<sup>2</sup>/s and  $Pe = 0.5, 5.0$  and  $\infty$ . Figures 12-14 show the resulting curves at a distance of 1000 m (except for the very long-lived U-238). The figures apply to a case where every nuclide originates in the repository. No decay chains are accounted for.

If a nuclide never reaches the biosphere with a concentration greater than  $10^{-9}$  times that in the repository, it is considered to have decayed to insignificance (Neretnieks, 1980). This criterion strictly applies only to a nuclide originally present in the repository, but may be modified to approximately apply to migrating daughter nuclides as well. The criterion may be expressed in terms of the distance a nuclide must travel through rock before its maximum concentration is reduced to a level  $10^{-9} C_0$ . These distances cannot be explicitly calculated. However, approximate values were obtained from the breakthrough curves referred to above. The distances are given in the last columns of Table 2, for different values of the Peclet number. For  $Pe = \infty$ , the only three nuclides which do not decay "totally" within 300 m, are Cs-135, I-129, and U-238. However, when  $Pe = 0.5$ , the situation is

considerably worse. Now, after a travelling distance of 300 m, the concentrations of eight nuclides are higher than  $10^{-9} C_0$ : Pu-239, Pu-240, Pu-242, Tc-99, Np-237, Cs-135, I-129 and U-238. The combination of the value for fissure spacing used in the examples (and thus block size), and permeability  $K_p = 10^{-9}$  m/s, is probably conservative. A fissure spacing of 50 m allows much less contact surface than the 1 m spacing used in KBS Safety Analysis, 1977.

The decay chains of a series of nuclides may be assessed by approximate calculations. Member  $i+1$  is assumed to start its travel from the point where member  $i$  arrived with a maximum concentration of  $10^{-9} C_0$ . The distances which the daughters will travel, are in reality considerably smaller. Tables 3 and 4 show that if  $Pe = \infty$ , the two important chains, including Am-241, Np-237, Th-229 and Ra-226, will decay within a 150 m thick granite barrier of the quality assumed. At the highest dispersion level,  $Pe = 0.5$ , the upper limit of the barrier thickness is  $\sim 11,500$  m.

Using the model, simulations have also been conducted for other values of the input parameters. In particular, the effects of finite release time and time of canister breakdown have been investigated.

Table 2. Most important radionuclides in a repository for spent fuel and their migration in granitic rock

Nuclide	Initial*	half life years	K** m <sup>3</sup> /m <sup>3</sup>	Distance (m) in rock with K <sub>p</sub> = 10 <sup>-9</sup> m/s, S = 50 m i = 0.003 m/m for C=C <sub>0</sub> · 10 <sup>-9</sup>		
	Ci/ton			Pe = ∞	Pe = 5.0	Pe = 0.5
Cs-137	1.1 · 10 <sup>5</sup>	30.2	170	1-3	3-10	30-100
Sr-90	7.6 · 10 <sup>4</sup>	28.1	43	1-3	10-30	100-300
Am-241	7.8 · 10 <sup>2</sup> <sub>a</sub>	458	86 000	<1	1-3	3-10
Am-243	2.1 · 10 <sup>1</sup>	7 370	86 000	<1	3-10	30-100
Pu-239	3.2 · 10 <sup>2</sup>	24 400	810	10-30	30-100	300-1000
Pu-240	4.9 · 10 <sup>2</sup>	6 580	810	3-10	30-100	300-1000
Pu-241	1.1 · 10 <sup>5</sup>	13.2	810	<1	1-3	10-30
Pu-242	1.4 · 10 <sup>0</sup>	379 000	810	30-100	300-1000	1000-3000
Cm-244	2.0 · 10 <sup>3</sup>	17.6	43 000 <sub>f</sub>	<1	<1	1-3
Tc-99	1.4 · 10 <sup>1</sup>	212 000	135	100-300	300-1000	3000-10 <sup>4</sup>
Np-237	3.3 · 10 <sup>-1</sup> <sub>b</sub>	2.14 · 10 <sup>6</sup>	3 240	30-100	300-1000	3000-10 <sup>4</sup>
Cs-135	2.5 · 10 <sup>1</sup>	3 · 10 <sup>6</sup>	170	300-1000	1000-3000	10 <sup>4</sup> <
I-129	3.8 · 10 <sup>2</sup>	17 · 10 <sup>6</sup>	0.005 <sub>e</sub>	10 <sup>4</sup> <	10 <sup>4</sup> <	10 <sup>4</sup> <
Ra-226	1.1 · 10 <sup>0</sup> <sub>c</sub>	1 600	1 350	3-10	10-30	100-300
Th-229	8.5 · 10 <sup>1</sup> <sub>d</sub>	7 340	6 480	3-10	10-30	100-300
U-238		4.51 · 10 <sup>9</sup>	3 240	3000-10 <sup>4</sup>	10 <sup>4</sup> <	10 <sup>4</sup> <

\* Kjellbert, 1977

\*\* (Allard, Grundfelt, KBS, 1978) reducing conditions.

a builds up to 3.3 · 10<sup>0</sup> after 100 years from Pu-241

b builds up to 11 · 10<sup>0</sup> after 10<sup>5</sup> years from Am-241

c not initially there, builds up from Th-230, Max. conc. at 10<sup>6</sup> years

d not initially there, builds up from U-233, Max. conc. at 2 · 10<sup>5</sup> years

e K equal to porosity of granite matrix = 0.005

f assumed half that for Am

Table 3. The decay chain leading to Th-229 (Grundfelt, 1978)

Nuclide	Half life years	Distance (m) beyond which $C < C_0 \times 10^{-9}$		
		Pe = $\infty$	Pe = 5.0	Pe = 0.5
Cm-245	9300	1-3	3-10	30-100
Pu-241	13.2	<1	1-3	10-30
Am-241	458	<1	1-3	3-10
Np-237	$2.14 \cdot 10^6$	30-100	300-1000	$3000 \cdot 10^4$
U-233	$1.62 \cdot 10^5$	10-30	100-300	300-1000
Th-229	7340	3-10	10-30	100-300
	Total	44-145	415-1346	3443-11,440

Table 4. The decay chain leading to Ra-226. The chain via U-238 is ignored due to the very long half life of U-238:  $4.5 \cdot 10^9$  years and the small amount coming from this chain (KBS, 1978; Grundfelt, 1978)\*

Nuclide	Half life years	Distance (m) beyond which $C < C_0 \times 10^{-9}$		
		Pe = $\infty$	Pe = 5.0	Pe = 0.5
Am-242M	152	<1	<1	3-10
Cm-242	0.5	<1	<1	<1
Pu-238	86	1-3	3-10	30-100
U-234	$2.47 \cdot 10^5$	10-30	100-300	1000-3000
Th-230	$8.0 \cdot 10^4$	10-30	30-100	300-1000
Ra-226	1600	3-10	10-30	100-300
	Total	24-75	143-442	1433-4411

\* Uranium will be practically immobilized in a reducing water because of its low solubility.

## CONCLUSION

Longitudinal dispersion has a large impact on the early arrival of many radionuclides — distances travelled before decay may differ by orders of magnitude. If the dispersion effect is of the magnitude considered in this study, it significantly diminishes the strong retardation effect due to matrix diffusion.

The Peclet numbers used in the examples, were based on a recent compilation of about 50 field measurements (Lallemand-Barrès and Peaudecerf, 1978). The measurements are from widely different media, and the spread in the data is over several magnitudes. It is clear that dispersion effects in fissured consolidated media require better understanding.

## APPENDIX

METHOD OF INTEGRATION

Because of the complicated nature of the integral expression for  $C_f/C_o$ , numerical integration must be performed. Consider the integral:

$$I = \int_0^{\infty} f(\lambda) d\lambda \quad (1)$$

The straightforward numerical calculation of  $I$  is accomplished by first replacing the infinite integral  $I$  by the finite integral  $\int_0^{\lambda_{\max}} f(\lambda) d\lambda$ , and then replacing this finite integral by an approximating summation  $S$ .

Thus:

$$I = S + \epsilon_S + \epsilon_T \quad (2)$$

where  $S$  is the sum actually computed, and  $\epsilon_S$  and  $\epsilon_T$  are the summation and truncation errors, respectively.

However, due to the very rapid oscillation of  $f(\lambda)$  for certain parameter values, this straightforward method may fail. The function  $f(\lambda)$  is the product of an exponential decaying function and a periodic sine function. The total function is thus a decaying sine wave, in which both the period of oscillation and the degree of decay are functions of the system parameters. It may be seen that the decay is slow for low values of  $\delta$ , and further, that the period of the sine function is short for large values of  $y$ .

In some instances, the magnitude of the integrand is still considerable after a thousand oscillations of the sine wave. With ordinary integration methods, one must choose a step size which is small with respect to the wave length. Thus, some special integration method must be devised for the cases mentioned above. Different integration methods were used for rapidly and for slowly oscillating integrands. They are separately discussed below.

#### REPLACEMENT OF THE INTEGRAL BY AN ALTERNATING SERIES

Before going into a detailed discussion of the integration methods, we analyze  $I$  more thoroughly. It is clear that  $f(\lambda)$  changes sign for  $\lambda = \lambda_n$ , where  $\lambda_n$  are the roots of  $\sin h(\lambda) = 0$ . One could therefore subdivide the integration interval into subintervals  $(\lambda_n, \lambda_{n+1})$  and replace the integral by a sum of integrals with alternating signs:

$$I_n = \int_{\lambda_n}^{\lambda_{n+1}} \frac{\exp g(\lambda)}{\lambda} |\sin h(\lambda)| d\lambda \quad (3)$$

$$I = I_0 + \sum_{n=n_0}^{\infty} (-1)^n I_n \quad (4)$$

where  $\lambda_n$  are successive zeros of  $\sin h(\lambda)$ .

Since both  $\exp g(\lambda)/\lambda$  and the period of the sine function decrease with  $\lambda$ , this is an alternating series with monotonically decreasing terms. According to Leibnitz's convergence test, the series converges. The solutions of:

$$F(\lambda_n) = h(\lambda_n) - n\pi = 0 \quad (5)$$

were obtained by Newton-Raphson's method.

In this iterative method,  $\lambda^{i+1}$  (where  $i$  denotes iteration step) is defined as the abscissa of the point of intersection between the  $\lambda$ -axis and the tangent to the curve  $F(\lambda)$  in the point  $(\lambda^i, F(\lambda^i))$ . Thus, the iteration sequence is defined by:

$$\lambda^{i+1} = \lambda^i - \frac{F(\lambda^i)}{F'(\lambda^i)} \quad (6)$$

If the initial approximation  $\lambda^0$  is adequate, convergence will be rapid.

When  $D_L = 0$  and  $\lambda_n > 5$  (using the limiting expressions of  $H_{D_1}$  and  $H_{D_2}$ ) equation (5) is a second-order algebraic equation with the solution:

$$\lambda_n = \frac{\delta}{2\sigma\theta} (+) \sqrt{\left(\frac{\delta}{2\sigma\theta}\right)^2 + \frac{n\pi}{\sigma\theta}} \quad (7)$$

This value was used as the starting approximation, for  $\lambda_n > 5$ , in the cases where  $D_L > 0$ . The convergence of (6) was then very rapid. When  $\lambda_n < 5$ , the limiting expressions of  $H_{D_1}$  and  $H_{D_2}$  are no longer applicable, and equation (7) is not valid. Newton-Raphson's method was then employed with the initial approximation  $\lambda^0 = 1$ .



## INTEGRATION

f(λ) slowly oscillating

The integration for these cases were carried out with Newton-Cotes' formula. An adaptive Newton-Cotes algorithm (QNC7) from the Sandia Mathematical Program Library was used in the calculations.

The upper limit of the integral,  $\lambda_{\max}$ , was estimated as follows. We have :

$$\epsilon_T = \int_{\lambda_{\max}}^{\infty} \exp g(\lambda) \sinh h(\lambda) \frac{d\lambda}{\lambda} < \exp g(\lambda_{\max}) \quad (8)$$

Put :

$$g(\lambda_{\max}) = -c \quad (9)$$

If  $D_L = 0$  and  $\lambda_{\max} > 5$  we get simply:

$$\lambda_{\max} = \frac{c+\delta}{\delta} \quad (10)$$

If  $D_L > 0$ ,  $\lambda_{\max} > 5$  and furthermore making the approximation (this gives an overestimate of  $\lambda_{\max}$ ):

$$z^2 y' = \delta Pe \left( \frac{2}{3} \frac{\lambda^2}{R} + \lambda \right) \approx \delta Pe \lambda \quad (11)$$

equation (9) is simplified to a second-order algebraic equation with the positive solution:

$$\lambda_{\max} = \frac{2(\frac{1}{2} Pe + c)}{\delta Pe} \left[ \sqrt{2(\frac{1}{2} Pe + c)^2 - \frac{1}{4} Pe^2 + \delta Pe} - (\frac{1}{2} Pe + c) \right] \quad (12)$$

In the rare cases where  $\lambda_{\max} < 5$ , Newton-Raphson's iterative method was employed to solve equation (9). The initial approximations were taken from equations (10) and (12) respectively. Newton-Raphson's method was also used when  $K < 0.5$  and the approximation (11) becomes crude. The value of  $c$  was taken as 40 giving a truncation error less than  $10^{-17}$ . The summation error was less than  $10^{-10}$ .

### $f(\lambda)$ rapidly oscillating

In these cases the series form of  $I$ , equation (4), was used. Though the convergence of the alternating series is very slow, it can be accelerated. One way to do this is by repeated averaging of the partial sums. Let  $S_n$  be the sum of the first  $n+1$  terms. The columns to the right of the  $S_n$ -column in the example given below are formed by building averages: each number in such a column is the mean of the two numbers which stand to the upper left and lower left of the number itself.

$S_n$	$M_1$	$M_2$
.3029834E-01	.1647013E-01	.1591533E-01
.2641914E-02	.1536054E-01	.1583316E-01
.2807916E-01	.1630578E-01	.1589831E-01
.4532395E-02	.1549085E-01	.1584578E-01
.2644931E-01	.1620070E-01	.1588876E-01
.5952089E-02	.1557682E-01	.1585315E-01
.2520154E-01	.1612948E-01	.1588298E-01
.7057413E-02	.1563649E-01	.1585774E-01
.2421556E-01	.1607899E-01	.1587929E-01
.7942416E-02	.1567959E-01	.1586074E-01
.2341676E-01	.1604190E-01	.1587682E-01
.8667031E-02	.1571174E-01	.1586279E-01
.2275644E-01	.1601385E-01	.1587510E-01
.9271260E-02	.1573635E-01	.1586424E-01
.2220144E-01	.1599213E-01	.1587387E-01
.9782815E-02	.1575562E-01	.1586529E-01
.2172842E-01	.1597496E-01	.1587297E-01
.1022150E-01	.1577098E-01	.1586607E-01
.2132046E-01	.1596116E-01	
.1060186E-01		

Note that the values in each column oscillate. It can be shown in general (Dahlquist and Björck, 1974) that for alternating series, if the absolute value of the  $j$ :th term (considered as a function of  $j$ ) has a  $k$ :th derivative which approaches zero monotonically for  $j > n_0$ , then every other value in column  $M_k$  is larger than the sum, and every other value is smaller. This condition is clearly satisfied here. Twenty terms of the alternating series were generally used. The averaging procedure was continued until the accuracy  $10^{-8}$  was obtained. This occurred after at most  $k = 5$ .

## ACKNOWLEDGEMENT

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## NOTATION

b	half width of fissure	m
C	concentration in liquid	mol/m <sup>3</sup>
C <sub>f</sub>	concentration in liquid in fissures	mol/m <sup>3</sup>
C <sub>p</sub>	concentration in liquid in microfissures	mol/m <sup>3</sup>
C <sub>s</sub>	concentration in solid rock	mol/m <sup>3</sup>
C <sub>o</sub>	inlet concentration in the liquid	mol/m <sup>3</sup>
D <sub>a</sub>	apparent diffusivity in microfissures	m <sup>2</sup> /s
D <sub>L</sub>	longitudinal dispersion coefficient	m <sup>2</sup> /s
D <sub>p</sub>	diffusivity in water in pores	m <sup>2</sup> /s
D <sub>v</sub>	diffusivity in water	m <sup>2</sup> /s
g	gravitational constant	m/s <sup>2</sup>
H <sub>1</sub>	see equation (17)	
H <sub>2</sub>	see equation (18)	
H <sub>D1</sub>	see equation (19)	
H <sub>D2</sub>	see equation (20)	
i	hydraulic gradient	m/m
K	volume equilibrium constant	m <sup>3</sup> /m <sup>3</sup>
K <sub>p</sub>	hydraulic conductivity	m/s
k <sub>f</sub>	mass transfer coefficient	m/s
m	$= \frac{\epsilon_f}{1 - \epsilon_f}$	
Pe	$= \frac{zU_f}{D_L}$ , Peclet number	
$\frac{\Delta}{q}$	volume averaged concentration in blocks	mol/m <sup>3</sup>

$q_i$	internal concentration in blocks	$\text{mol/m}^3$
$q_s$	$= q_i(r_o, z, t)$	$\text{mol/m}^3$
$R$	$= \frac{K}{m}$ , distribution ratio	
$R_F$	$= \frac{r_o}{3k_f}$ , film resistance	s
$r$	radial distance from center of spherical particle	m
$r_o$	effective spherical radius	m
$S$	fissure spacing	m
$t$	time	s
$U_f$	average velocity of water in fissures	m/s
$x'$	see equation (15)	
$y$	$= \alpha t$ , contact time parameter	
$y'$	see equation (16)	
$z$	distance in flow direction	m

#### Greek letters

$\gamma$	$= \frac{3 D a K}{r_o^2}$	$s^{-1}$
$\delta$	$= \frac{\gamma z}{m U_f}$ , bed length parameter	
$\lambda_D$	constrictivity for diffusion	
$\epsilon_f$	porosity of fissures	
$\epsilon_p$	porosity of rock matrix	
$\lambda$	variable of integration	
$\lambda_d$	decay constant of radionuclide	$s^{-1}$
$\mu$	viscosity of water	$\text{Ns/m}^2$
$\nu$	$= \gamma R_F$	
$\rho$	density of water	$\text{kg/m}^3$
$\sigma$	$= \frac{2 D a}{r_o^2}$	$s^{-1}$
$\tau$	tortuosity	

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Figure captions

Figure 1 Upper limit of integral,  $\lambda_{\max} = \lambda_{\max}(\delta, Pe)$ , with the truncation error less than  $e^{-20}$  ( $c = 20$ ).

Figures 2-7 Dimensionless breakthrough curves (without decay) for  $R \rightarrow \infty$ .  
 $\delta = 10^{-8}, 10^{-4}, 10^{-2}, 1, 10^2, 10^4$  and  $Pe = 0.5, 5.0, \infty$ .

Figures 8-11 Dimensionless breakthrough curves (without decay). The influence of  $R$ .  $\delta = 10^{-8}, 10^{-4}$  and  $Pe = 0.5, \infty$ .

Figures 12-14 Breakthrough curves at the distance 1000 m and Peclet numbers 0.5, 5.0,  $\infty$  for the 15 most important radionuclides in spent fuel. The case depicted applies for the nuclides originally present in the repository.  $K_p = 10^{-9}$  m/s,  $i = 0.003$  m/m,  
 $D_p \epsilon_p = 10^{-12}$  m<sup>2</sup>/s,  $S = 50$  m.

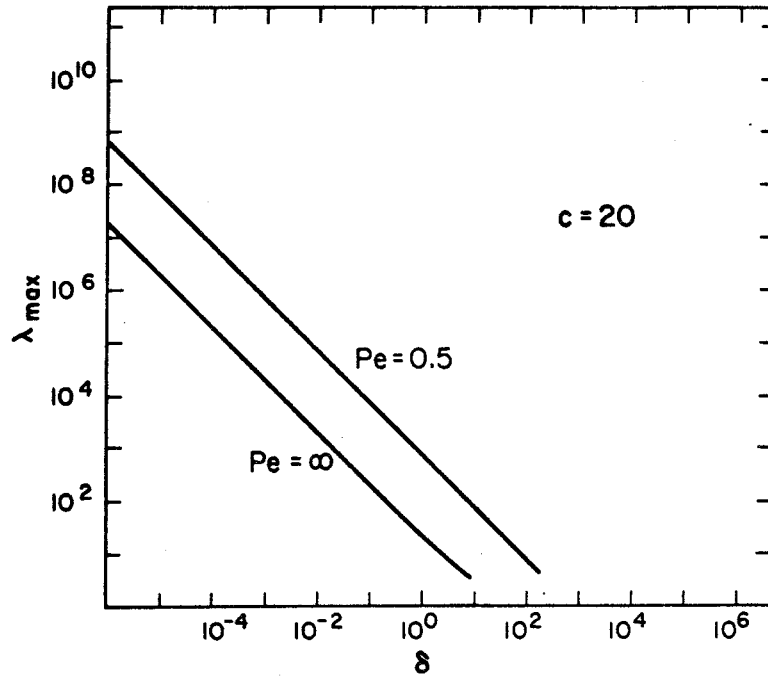


FIGURE 1

XBL 7912-13468

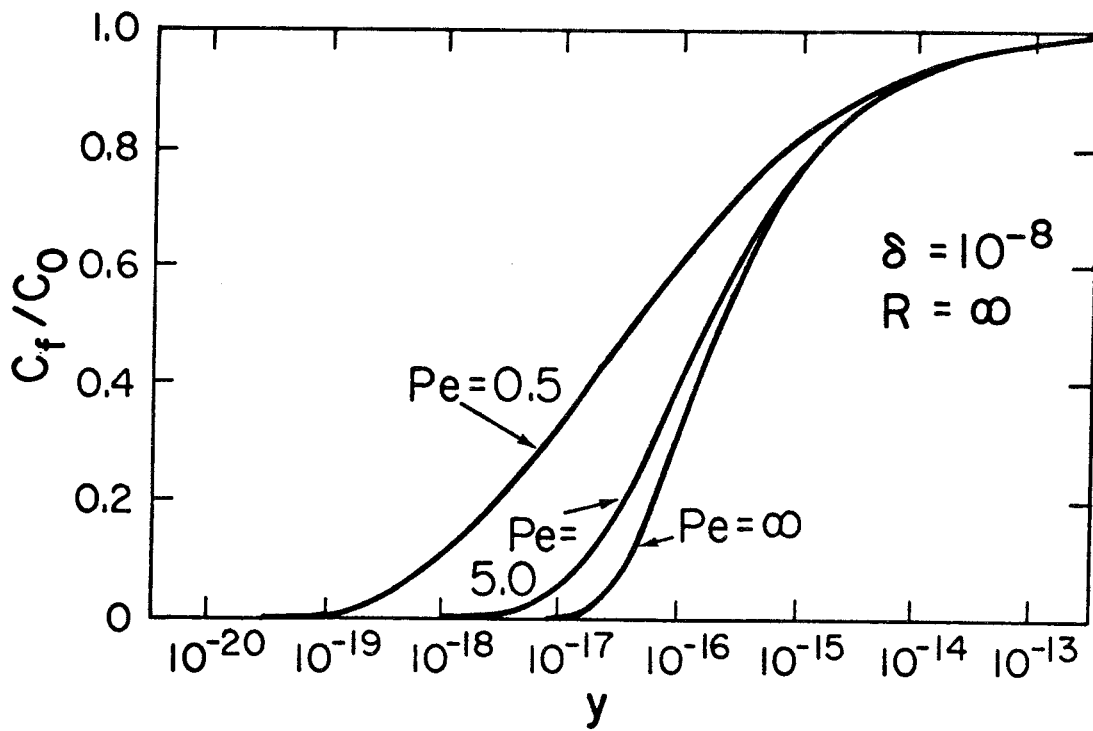


FIGURE 2

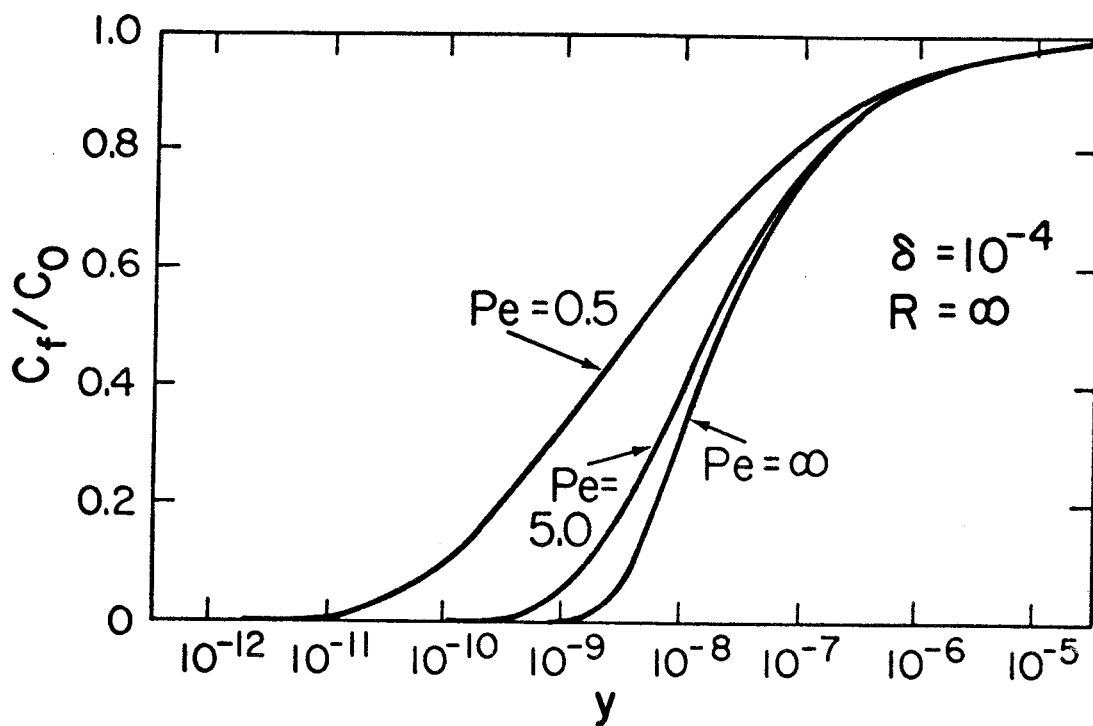


FIGURE 3

XBL 7911-13465

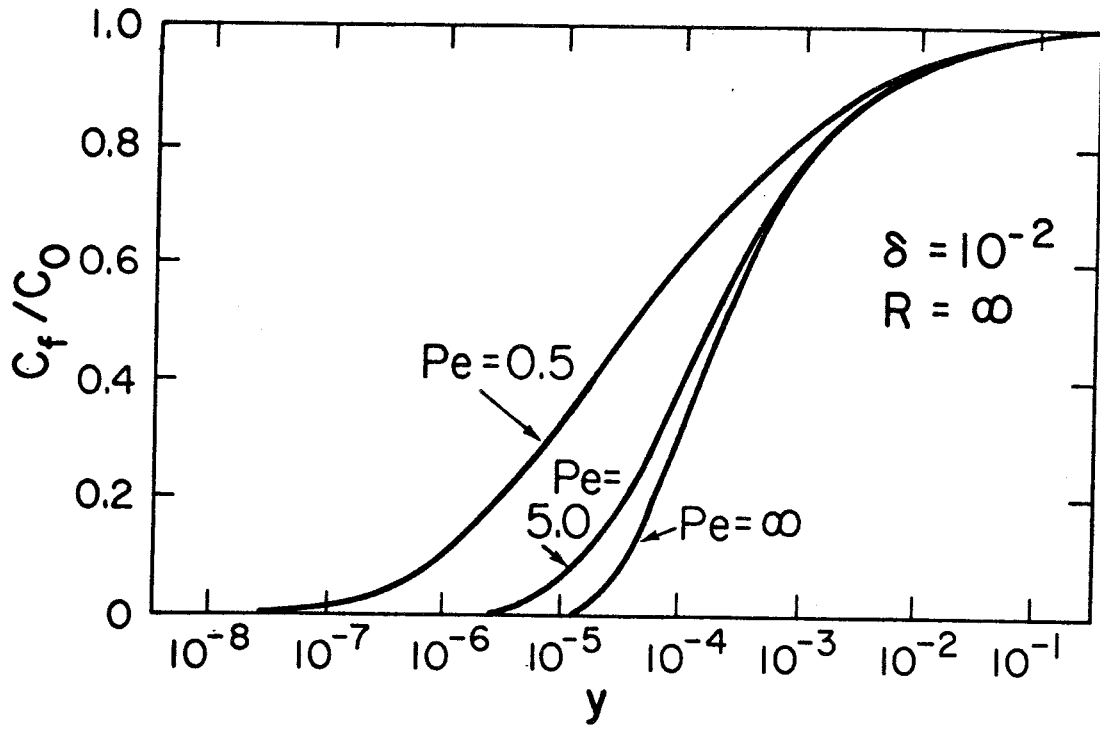


FIGURE 4

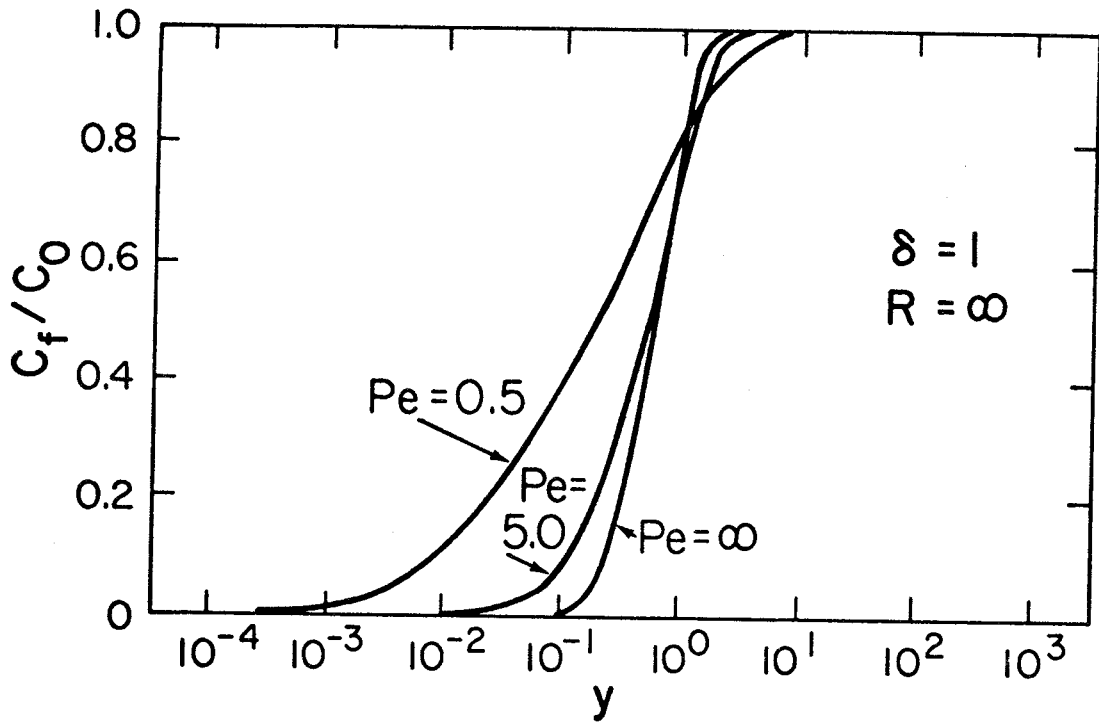


FIGURE 5

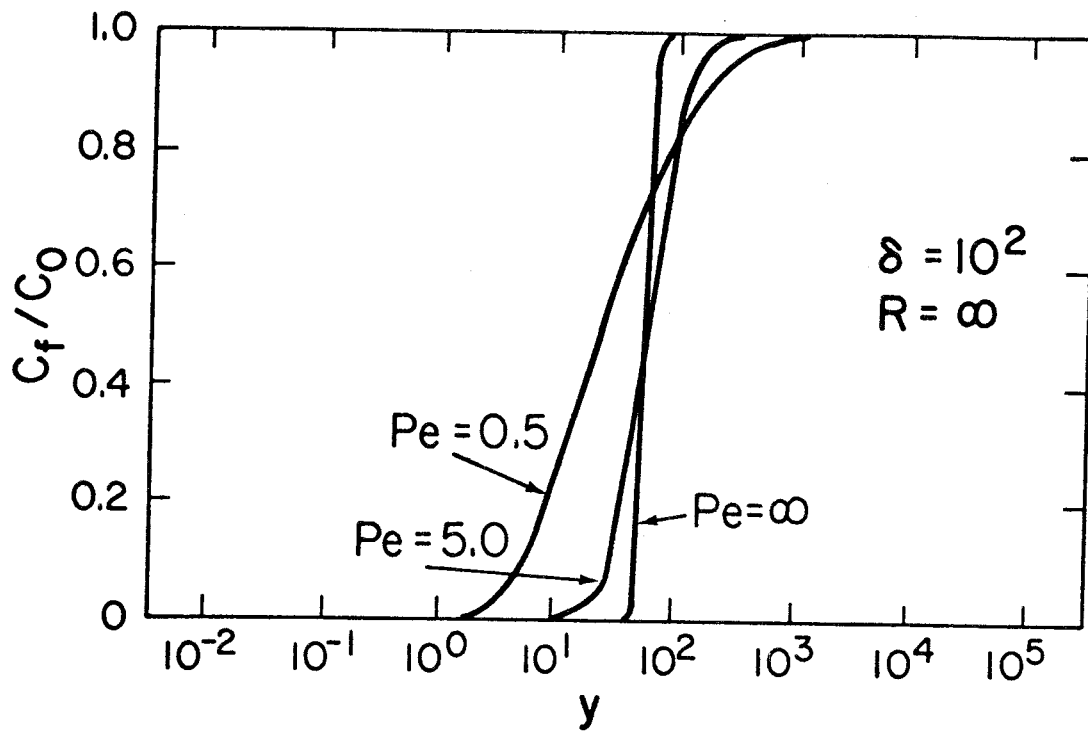


FIGURE 6

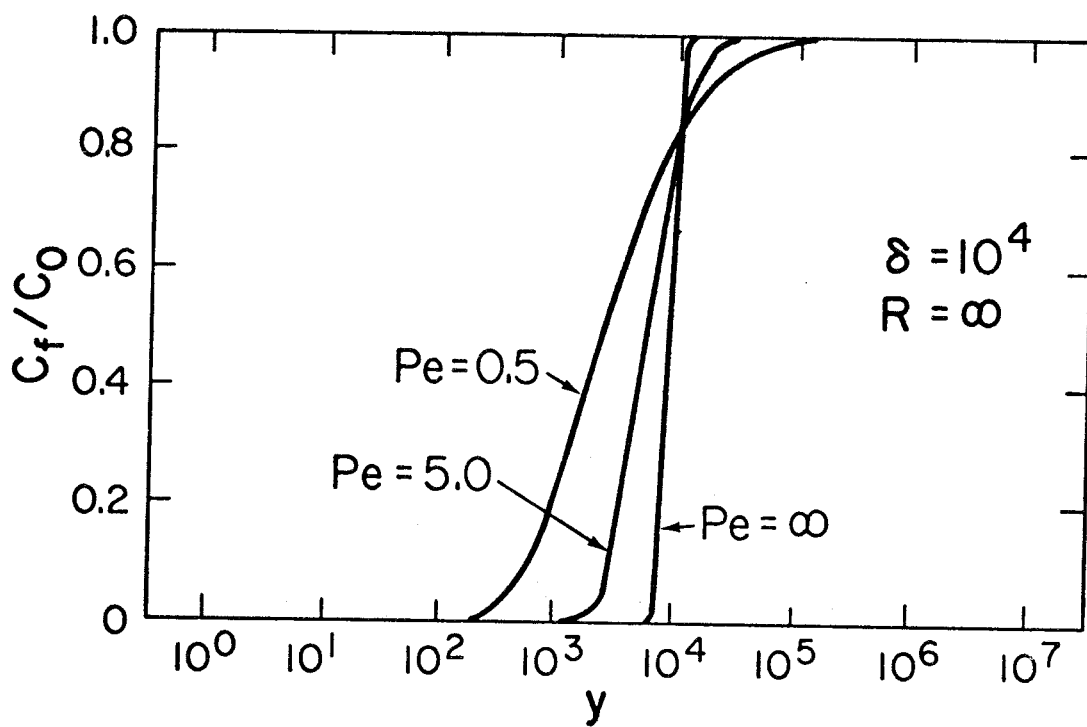


FIGURE 7

XBL 7912-13467

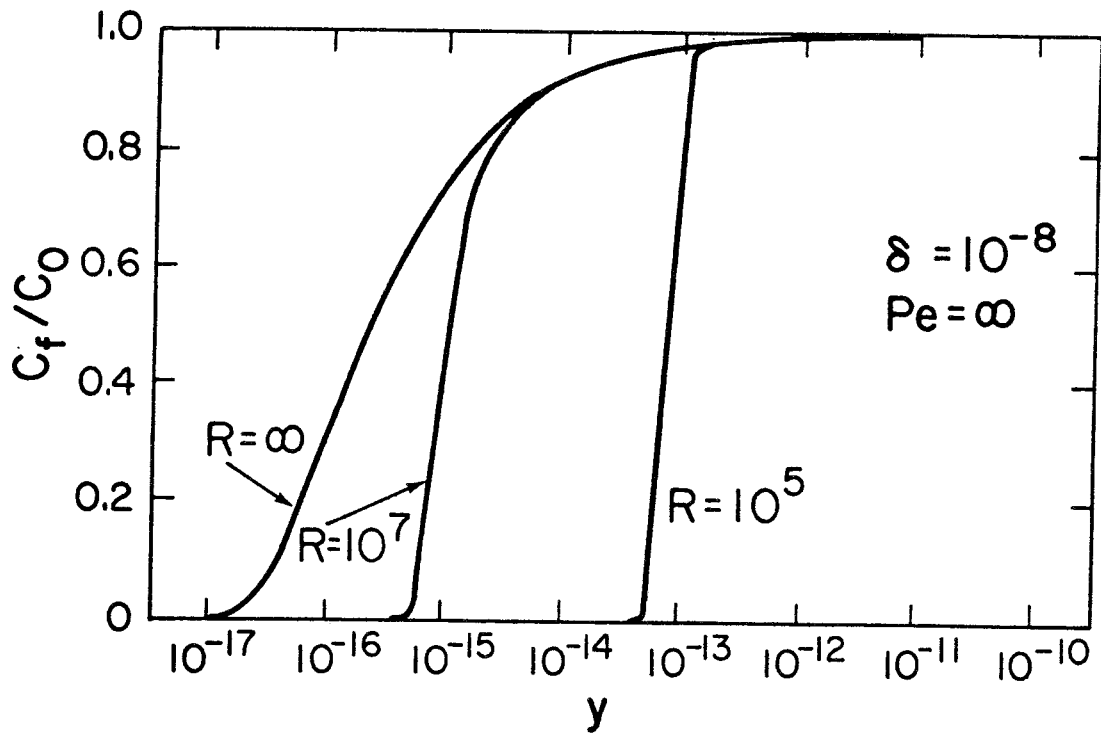


FIGURE 8

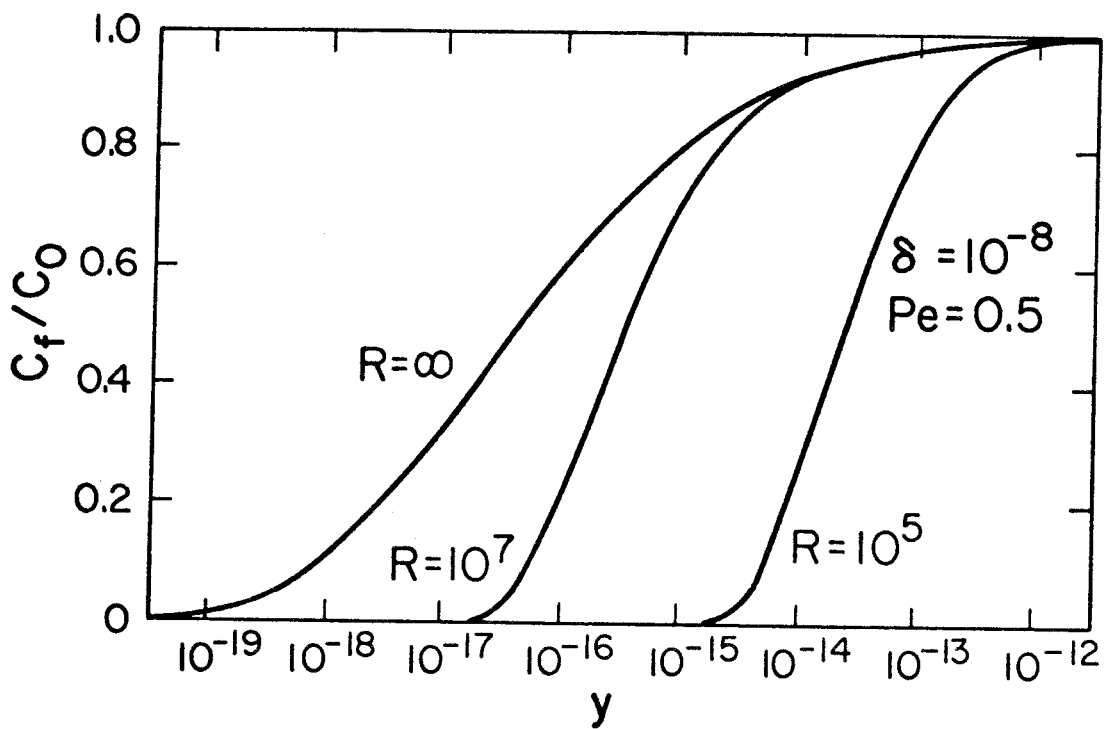


FIGURE 9

XBL 7911-13464

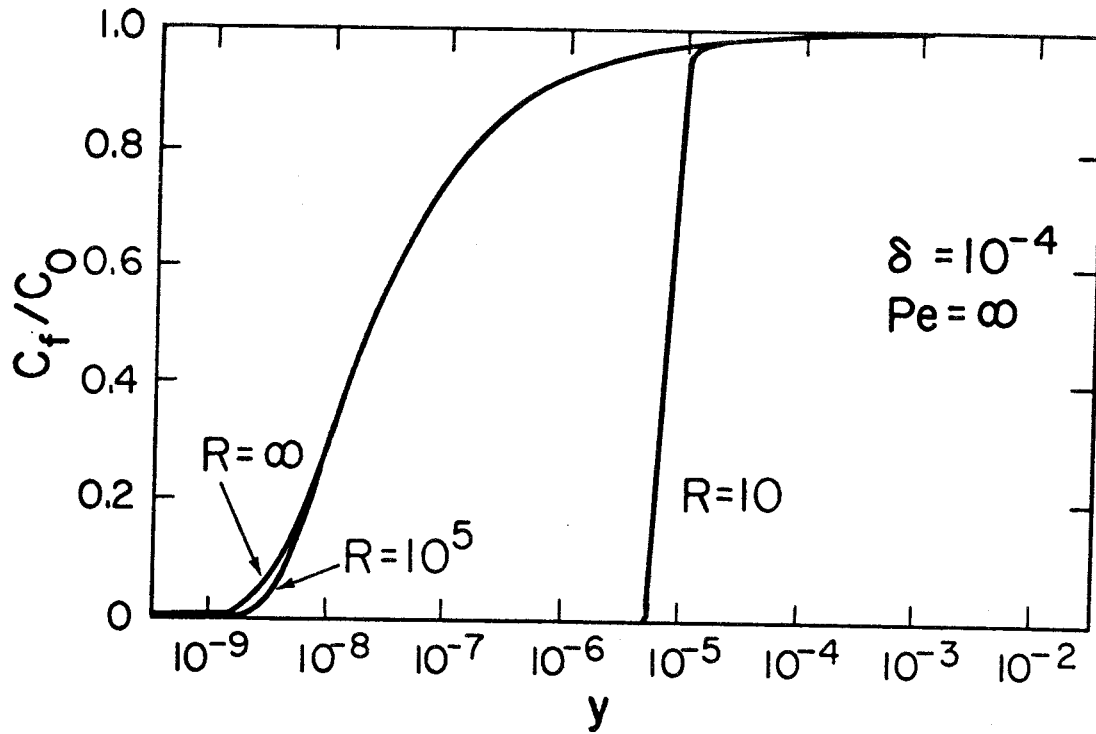


FIGURE 10

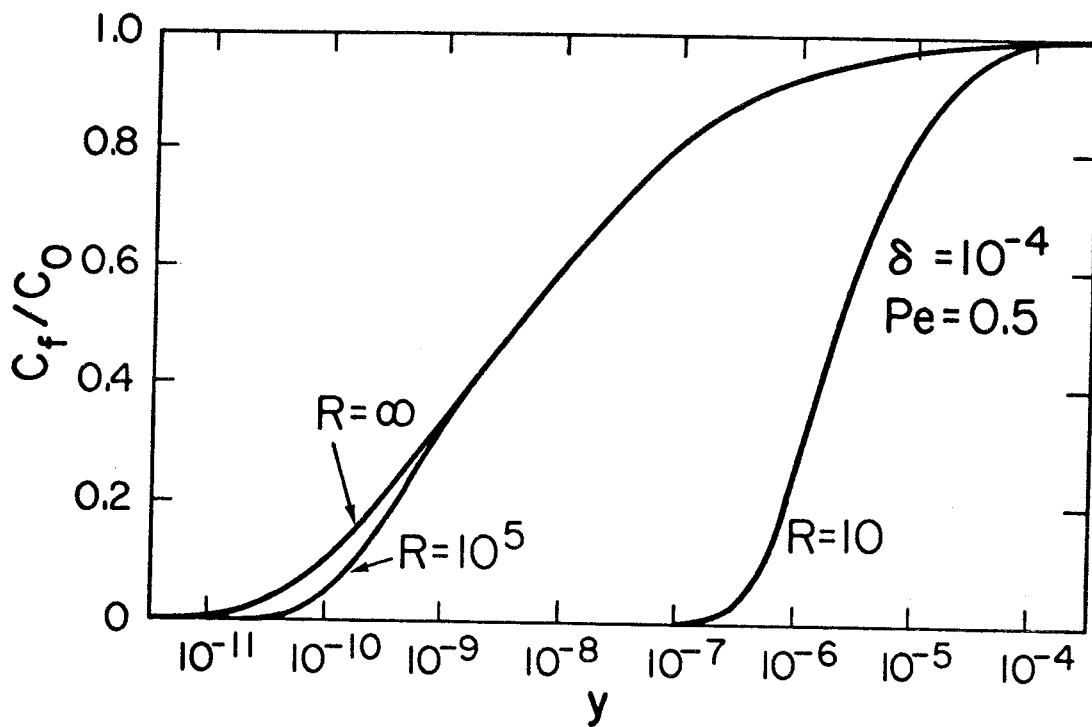


FIGURE 11



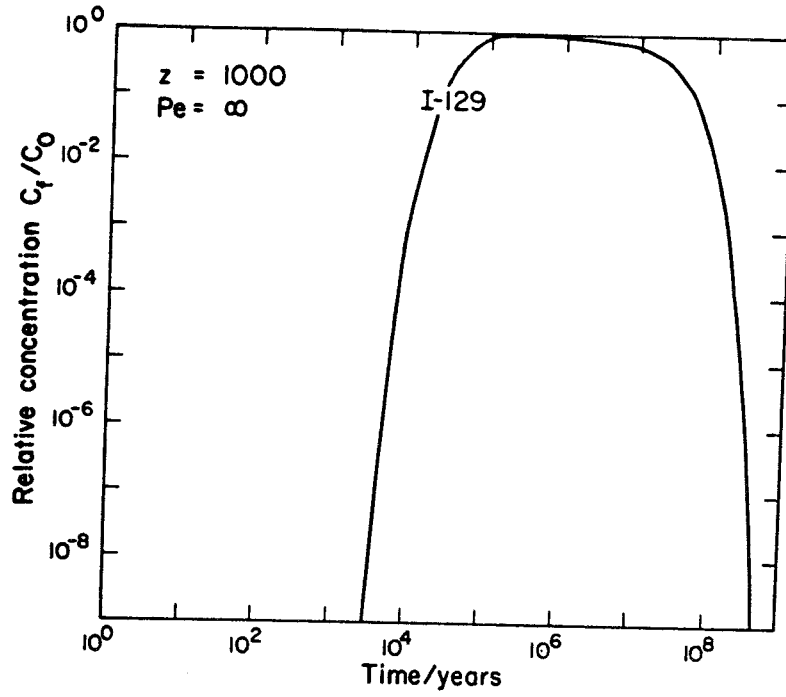


FIGURE 12

XBL 7912-13469

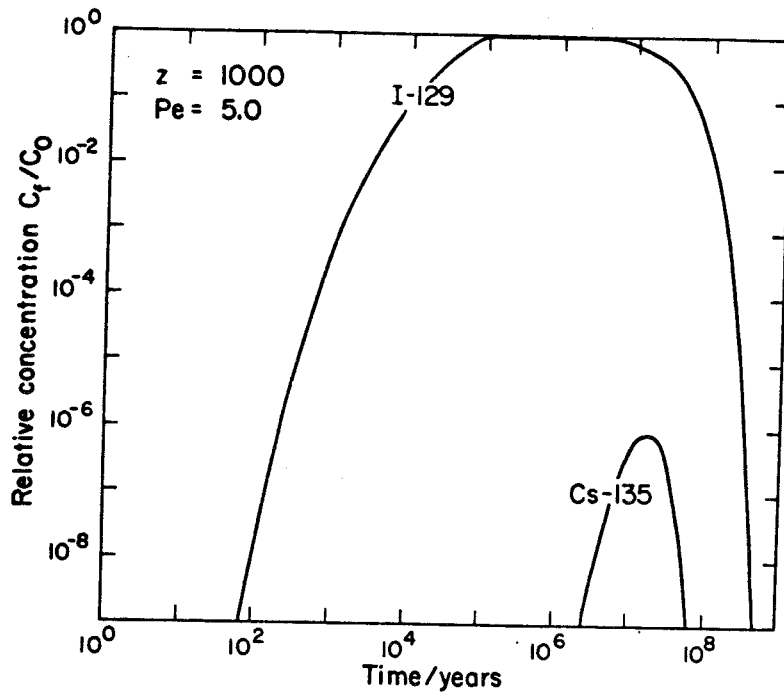


FIGURE 13

XBL 7912-13471

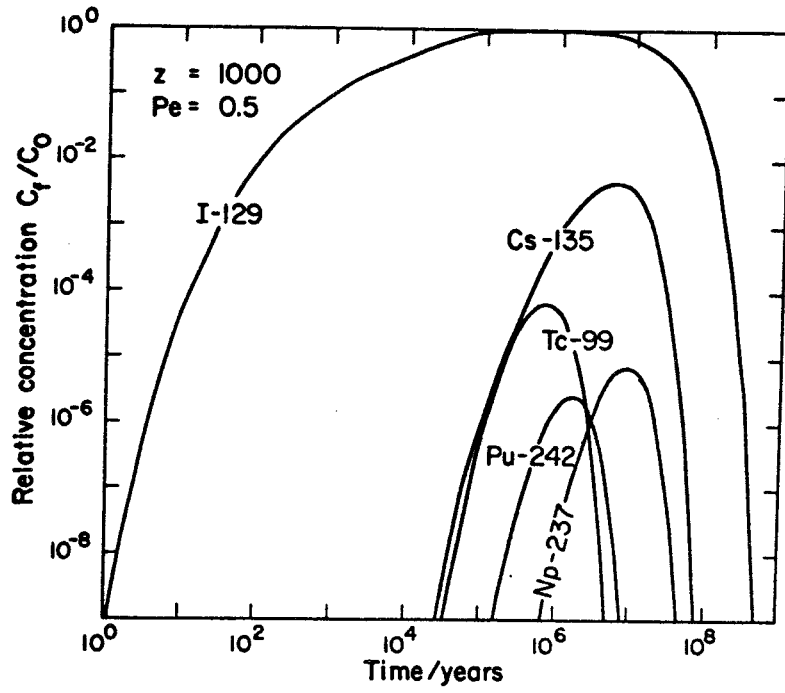


FIGURE 14

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