LOW-LEVEL WASTE SHALLOW LAND BURIAL SOURCE TERM CONTAINER BREACH AND WASTE FORM LEACHING MODEL DEVELOPMENT

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ABSTRACT

A general computer model has been developed to predict the release and transport (i.e. source term) of radionuclides from shallow land burial facilities. This model predicts the processes of unsaturated water flow, metallic container degradation, leaching of radionuclides from the waste form, and their movement away from the waste form. This paper discusses model development work for the container degradation and leaching aspects of the source term model. Application of these models and the sensitivity of release rates to model parameters, e.g. diffusion coefficients, corrosion rates, etc., are also discussed.

INTRODUCTION

Currently, commercial low-level radioactive waste disposal is achieved using shallow land burial. Typically, waste containers are placed in an excavated trench and covered with soil. To accurately model this system requires the capability to model multiple containers and waste forms in an unsaturated porous media. This can be achieved using the computer codes FEMWATER(1) and BLT(2). A discussion on how these codes can be used to predict the source term can be found in Reference 2 and our companion paper(3).

FEMWATER is a two-dimensional finite element computer code used to predict water flow in heterogeneous soil systems above or below the water table. FEMWATER also predicts the moisture content of the soil.

BLT is also a two-dimensional finite element computer code. BLT predicts container Breach, waste form Leaching, and radionuclide Transport. BLT is a modification of the computer code FEMWASTE(4) which predicts transport in unsaturated/saturated porous soils. The major modifications to FEMWASTE include adding models to predict container breach and waste form leaching. The contaminant release rate from the waste form is input to the transport segment of the calculation as a source term. The Breach and Leach models are structured to allow any element of the finite element grid to be treated as a waste containing element. This allows modeling of different waste forms and containers within a single simulation. This paper will discuss the Breach and Leach models and present results from application of these models.

CONTAINER DEGRADATION (BREACH) MODEL

The purpose of the BREACH model is to compute the area of the waste container that is breached and the time at which it is first breached. The BREACH model considers both general and pitting corrosion of metallic containers through the use of semi-empirical correlations(5,6). The correlation coefficients are functions of the soil properties such as pH and degree of aeration. The correlation coefficients for pitting and general corrosion of carbon steel, a commonly used container material, have been obtained from regression analysis of data on corrosion in soils obtained by the National Bureau of Standards (NBS),(7) (currently the National Institute of Standards and Technology). General corrosion rates for 304L and 316L stainless steels have also been obtained from this data base.

A brief description of the pitting and general corrosion models follows. A more detailed discussion along with the compilation of the data can be found elsewhere(4,8).

Pitting Corrosion Model

The pitting model is an empirical correlation that is based on data obtained by the NBS. The maximum pit depth takes the form:

$$h = kt^n \, (A/372)^a \qquad \text{(Eq. 1)}$$

where h is the maximum pit depth in cm; k is the pitting parameter cm/yr^n ; t is the time in years; n is the pitting exponent which depends on soil properties; A is the surface area of the container in cm^2 ; the constant 372 cm^2 is a scaling factor that arises from the fact that the original test coupons which were used to obtain the data for determining k and n were 372 cm^2; and "a" is an experimentally derived correlation coefficient. Values for "a" depend on the material and soil. Extensive studies by Logan (9) indicated that for wrought irons and carbon steels "a" ranged from 0.08 to 0.32 with a mean value of 0.15(2,9).

The value of n can be determined through user specified input or, for carbon steels, it can be calculated through empirical expressions derived from the NBS data. When calculating the value of n, as a minimum, the degree of soil aeration must be known. If other information is not available, the value for n is selected as 0.26, 0.39, 0.44, or 0.59 for good, fair, poor, and very poor aeration, respectively. These values are the averages determined from the NBS study for their respective soil aeration.

If the clay content is known, n is calculated from:

$$n = n_o \, \theta \, (1\text{-CL})^{0.4} \qquad \text{(Eq. 2)}$$

where n_o = 1, 1.5, 2, or 2.5 for good, fair, poor, and very poor aeration, respectively; θ is the volumetric moisture content of the soil (calculated by FEMWATER) and θ CL is the clay fraction of the soil. An improved correlation for n as a linear function of and CL has recently been proposed(6), however, it has not been incorporated into the BLT code at this time.

The degree of soil aeration plays a central role in determining the pitting rate. In general, poor aeration correlates with deeper pits and higher long term pitting rates. However, it is not a well-defined property. General guidelines for determining aeration can be found in Romanoff(7). Aeration factors are those that influence the access of oxygen and moisture to the metal. In general, soils of coarse texture, such as sands, tend to have low water-holding capacity and are characterized by good drainage and aeration. Soils of fine texture and high water-holding capacity, such as clays, are usually characterized by poor drainage and aeration.

After calculating n, the pitting parameter k is determined. The value of k can be specified via input or, if the pH is specified, from the following relationships:

$$k = 0.0146\,(10 - \text{pH}) \qquad \text{pH} < 6.8 \qquad \text{(Eq. 3)}$$

$$k = 0.0457 \qquad 6.8 < \text{pH} < 7.3 \qquad \text{(Eq. 4)}$$

$$k = 0.026(\text{pH} - 5.13) \qquad 7.3 < \text{pH} \qquad \text{(Eq. 5)}$$

The maximum pit depth is calculated from Eq. (1) using the values of k and n obtained either from input or from the empirical expressions, Eqs. (2-5). If the pit depth does not exceed the container thickness, the container remains unbreached and water cannot access the waste form. When the calculated pit depth does exceed the metal thickness, the area breached is calculated using the following relationship:

$$A_b = N_p \pi \,(h^2 - MT^2) \qquad \text{(Eq. 6)}$$

where A_b is the area breached in cm^2; N_p is the number of penetrating pits per container, estimates for this value range from 1000 - 10000 for a surface area of a 55 gallon drum, 21,000 cm^2 (8); h is the maximum pit depth (cm) as described by Eq. (1) and MT is the thickness of the metal (cm). Equation (6) arises from the assumption that the pits are hemi-spherical in shape and continue to grow at the same rate once the metal has been penetrated. If the calculated breached area exceeds the actual surface area, the breached area is limited to the available surface area.

General Corrosion

The general corrosion of the metal is calculated assuming that the corrosion rate is constant and independent of time. This can be conservative because the NBS general corrosion data indicate that the rate decreases in time (7). The time dependence could be determined as a function of soil properties using linear correlation analysis similar to the approach used to calculate n in Eq. (2). For a constant corrosion rate, the thickness of metal corroded, d(cm), is:

$$d = gt \qquad \text{(Eq. 7)}$$

where g is the general corrosion rate in (cm/sec) and t is the time in seconds.

If the general corrosion thickness exceeds the container thickness, the entire surface area of the container is assumed to be corroded away. At this time, the container does not provide any barrier from water access to the waste form. No credit is taken for the corrosion products that are present.

RELEASE OF RADIONUCLIDES FROM THE WASTE FORM (LEACH)

In calculating releases from the waste form, BLT steps through all designated waste elements one by one. If the container for the element under consideration is not breached, the total release rate is zero. However, the mass available for release is decreased due to radioactive decay.

Once a breach has occurred, release is assumed to start immediately. If the first breach occurs in the middle of a time step, release is calculated to occur starting at the beginning of the time step and the time of first breach is recorded as the time at the beginning of the time step. This is conservative because it starts releasing material at an earlier time.

When the container is breached, BLT first calculates the average moisture content, the magnitude of the Darcy velocity, and the cross-sectional area of the element from information supplied by FEMWATER. With these variables plus input variables that define the release characteristics of the waste form, the LEACH subroutine calculates the releases from the waste form.

Three release mechanisms are considered:

(1) diffusion, which accounts for the diffusional transport of radionuclides through the waste form, for example through the pore waters in cement waste forms;

(2) dissolution, which frees the radionuclides from the bulk solid by dissolving the solid phase, this would be the major release mechanism for activated metal waste forms; and

(3) surface rinse, which releases all of the surface residing radionuclides using a mixing bath model.

The release properties of each waste type are input by the user. BLT has the capability of modeling all three release processes on a single waste form. The user can choose the relative contribution of each release mechanism. For example, for a cement waste form with surface contamination, 20% of the mass could be released via surface rinse and 80% via diffusion.

To account for the presence of partially breached containers, the total amount released from the waste form/container system is scaled by the breached ratio, (the breached area divided by the container area). Thus, if the breached ratio is 0.01, total release for transport is reduced by a factor of 100. The remaining 99% of the mass released is placed in the rinse model.

Diffusion Release

Conceptually, release from a porous solid is viewed as a diffusion process in which radionuclides move through the pore waters of the solid (8). Four different cases are modeled: a) the semi-infinite medium, in which release from the finite sized waste form occurs into an infinite half-space; b) the finite medium, in which release from the finite waste form occurs into a finite volume; c) the finite plane waste form with zero concentration at the outer boundary. In this model it is assumed that transport away from the waste form is fast enough to maintain essentially zero concentration at the edge of the waste form; and d) the finite cylinder waste form model with zero outer boundary concentration. This model is recommended for calculating diffusion coefficients from leach test data in ANS 16.1 (10), the American Nuclear Society Standard for conducting leach tests.

In all four cases, analytical solutions to the decay corrected diffusion equation are obtained. Initially, it is assumed that the concentration is uniform within the waste form and zero outside the waste form. The quantity of interest for calculating release is the flux of radionuclides out of the waste form. For plane geometry the flux is evaluated directly from the analytical solution. For cylindrical geometry, an analytical solution for the Cumulative Fractional Release, (CFR), is obtained. Knowing the CFR at two different times allows an estimate of the rate at which the contaminant leaves the waste form.

Semi-Infinite Medium

In this case, symmetry is used and only 1/2 the waste form is modeled. The modeled 1/2 of the waste form is located in the region $o < x < h$ and the porous medium contacting the waste form extends to infinity. This model represents a system in which transport processes are controlled by a single diffusion coefficient and best represents non-interacting waste forms. The boundary conditions are zero flux at the center of the waste form, $x = 0$, and zero concentration at $x = \infty$.

The flux at the edge of the waste form is(11):

$$J_D = \frac{1}{2} C_o \sqrt{\frac{D}{\pi t}} e^{-\lambda t} [1 - e^{-\frac{h^2}{Dt}}] \qquad \text{(Eq. 8)}$$

where D is the diffusion coefficient, t is the time, C_o is the initial concentration in the waste form, and λ is the decay constant.

Finite Plane - Zero Boundary Flux

In this case, symmetry again permits only 1/2 of the waste form to be modeled. The modeled 1/2 of the waste form is located in the region $o < x < h$ and the porous medium extends a distance $\ell - h$ from the waste form. Thus, the domain of interest is $0 < x < \ell$. The boundary conditions are zero flux at the center of the waste form, $x = 0$, and zero flux at the outer boundary of the system, $x = \ell$.

These boundary conditions specify that no mass leaves the system. Thus, this model would be most useful for approximating the case when two waste forms with similar release characteristics are aligned such that a plane of symmetry (i.e., zero flux) occurs between them.

Because we are no longer dealing with a semi-infinite medium, the solution to the diffusion equation becomes an infinite series. When LaPlace transforms are used to solve the diffusion equation, the infinite series consists of complementary error functions. This infinite series converges rapidly for small values of the dimensionless parameter called TMP ($TMP = Dt/\ell^2$). However, for large values of TMP the solution is slow to converge. When separation of variables is used to solve the diffusion equation the infinite series consists of trigonometric and exponential functions.

This trigonometric series converges rapidly for large values of TMP.

Therefore, to insure rapid convergence of the series the following expressions are used to calculate the flux (2,11).

If TMP < 1

$$J_D = \frac{1}{2} C_o \sqrt{\frac{D}{\pi t}}\, e^{-\lambda t} \quad \cdot$$

$$\sum_{n=-N}^{N} e^{-\frac{n^2\ell^2}{Dt}} - e^{-\frac{(h-n\ell)^2}{Dt}} \qquad \text{(Eq. 9)}$$

If 1 TMP

$$J_D = \frac{2D\ C_o\ e^{-\lambda t}}{\ell} \quad \cdot$$

$$\sum_{n=1}^{N} \sin^2 \left(\frac{n\pi h}{\ell}\right) \exp\left[-\frac{n^2\pi^2 Dt}{\ell^2}\right] \qquad \text{(Eq. 10)}$$

where the value of N is chosen to be large enough to evaluate the solution to the desired degree of accuracy. Solution (9) and (10) are equivalent for all values of TMP provided N = ∞.

Finite Plane - Zero Boundary Concentration

In this case, symmetry is assumed. The modeled 1/2 of the waste form is located in the region $0 < x < h$. At the boundary $x = h$, it is assumed that the concentration is zero. Thus, everything that reaches the boundary is swept away instantaneously. This insures that the concentration gradient is as large as possible. Therefore, the flux is maximized. At the $x = 0$ boundary zero flux is specified.

This model predicts the largest release rate that could occur from a plane, if diffusion is the release rate limiting process. This model is most appropriate when transport processes outside the waste form are much faster than within the waste form. This will often be the case for advection dominated flows expected at shallow land burial sites.

As before, the solution to this problem is expressed in an infinite series. Again, for computational ease, two different series solutions are used depending on the value of TMP $= Dt/h^2$.

The solutions used to evaluate the flux are (2,11):

If TMP 1

$$J_D = C_o \sqrt{\frac{D}{\pi t}}\ e^{-\lambda t} \quad \cdot$$

$$\sum_{n=0}^{N} (-1)^n \left(e^{-\left(\frac{n^2h^2}{Dt}\right)} - e^{-\left[\frac{(n+1)^2h^2}{Dt}\right]} \right) \qquad \text{(Eq. 11)}$$

If 1 TMP

$$J_D = 2D\ \frac{C_o}{h}\ e^{-\lambda t} \quad \cdot$$

$$\sum_{n=0}^{N} e^{-\left[\frac{\pi^2(2n+1)^2Dt}{4h^2}\right]} \qquad \text{(Eq. 12)}$$

The value of N is chosen to be large enough to insure the desired degree of accuracy. Equations (11) and (12) are equivalent for all values of TMP provided N = ∞.

Finite Cylinder - Zero Boundary Concentration

The waste form is a cylinder with radius, R, and height, H. At the edge of the waste form, the contaminant concentration is zero. As with the finite plane model, this boundary condition causes the maximum concentration gradient to form and leads to the highest release as compared to other possible boundary conditions.

The solution for the CFR is the product of two infinite series (12). To minimize the truncation error associated with taking a finite number of terms, the formulae developed by Pescatore (13) are used to evaluate the series.

$$\mathrm{CFR} = 1 - \frac{32 \quad \mathrm{Sp}\ \mathrm{Sc}}{\pi^2} \qquad \text{(Eq. 13)}$$

$$\mathrm{Sp} = \sum_{n=1}^{N-1} \exp \frac{[-(2n-1)^2\pi^2\ Dt/H^2]}{(2n-1)^2}$$

$$+ \frac{N}{(2N-1)^2} \exp\ [-(2N-1)^2\pi^2\ Dt/H^2]$$

$$- \frac{1}{2H} \sqrt{\pi\ Dt}\ \ \mathrm{erfc}\ [(2N-1)\ \pi\sqrt{Dt/H}] \qquad \text{(Eq. 14)}$$

$$\mathrm{Sc} = \sum_{m=1}^{M-1} \frac{1}{\beta_m^2} \exp\ [-\beta_m^2\ Dt/R^2]$$

$$+ [\frac{1}{\beta_M f_M} + \frac{1}{2\beta_M^2}]\ \exp\ [-\beta_M^2 Dt/R^2]$$

$$- \frac{1}{R f_M} \sqrt{\pi Dt}\ \ \mathrm{erfc}\ (\beta_M \sqrt{Dt/R}) \qquad \text{(Eq. 15)}$$

$$\text{where } f_M = \pi - \frac{1}{8\pi M^2}$$

where β_m are the zeroes of the zeroth order Bessel function. In BLT, the maximum number of terms in each series, N and M, are set to 10.

ANS 16.1 recommends use of the semi-infinite medium approach if the CFR is less than 0.2 and Eq. (13) if the CFR exceeds 0.2. This approach is not followed in BLT. The approach recommended in ANS 16.1 can lead to a numerical inconsistency when switching between the semi-infinite and finite medium models. In particular, the semi-infinite medium model always predicts greater release than the finite medium model. At the time step when the switch between models is made, this inconsistency leads to an underprediction of the release. To avoid this inconsistency, BLT calculates the CFR based on Eqn. (13) and the semi-infinite medium model and chooses the minimum of these two. This is done because for small values of CFR, (< 1%), the CFR predicted in Eqn. (13) exceeds that of the semi-infinite model due to truncation error.

After, calculating the flux by one of the four models, the amount released for transport is scaled by the breached ratio, the breached area divided by the total container area. The mass that has been predicted to leach out by diffusion but is not released for transport because the breached ratio is less than one is transferred to the rinse model.

Dissolution Release

The dissolution release model assumes that release occurs through congruent dissolution of the waste form. Thus, all species are released at the same rate. This rate is limited by solubility constraints. Also, as in the case of diffusion release, if there is a partially breached container, the amount released for transport is scaled by the breached ratio and the remaining mass is transferred to the rinse model.

The flux of material released is evaluated using the expression:

$$J_{dis} = U \cdot C_{wf}\ (1 - \frac{C_s}{C_{sat}}) \qquad \text{(Eq. 16)}$$

where U is the dissolution velocity in cm/s; C_{wf} is the concentration of the radionuclide in the waste form, C_{wf} is evaluated by taking the mass of the radionuclide in the waste form and dividing by the volume of the waste form, C_s is the concentration of the radionuclide in solution, this value is the average solution concentration in the finite element as calculated by the transport model at the beginning of the time step; and C_{sat} is the solubility limit for the radionuclide.

The mass release rate for transport is the flux multiplied by the surface area scaled by the breached ratio. The dissolved mass that is not immediately released for transport, i.e, (1 - breached ratio) multiplied by the flux and surface area, $_w$ is transferred to the rinse model.

The dissolution model is applicable to activated metals that release radionuclides due to general corrosion. As a first approximation, the mass released due to pitting corrosion or other localized corrosion processes is not considered because these releases are small when compared with general corrosion releases.

Rinse Release

The rinse release model is a mixing bath model. Release of material is a function of water flow rate and solubility constraints. The concentration within the waste form equals the total mass available for rinse release divided by the volume of water. The flux released is that concentration multiplied by the flow rate (Darcy velocity calculated by FEMWATER) multiplied by the ratio of the breached area divided by the total area. These relationships are expressed by:

$$C = AM \cdot \left(1 - \frac{C_s}{C_{sat}}\right) / (\theta \cdot V_{el}) \quad C < C_{sat} \quad \text{(Eq. 17)}$$

where C is the concentration in g/cm^3; AM is the available mass in g, this includes the mass originally assigned to the rinse model plus any mass that has been transferred to the rinse model because of having a partially breached container; C_s is the average concentration in solution in the finite element at the beginning of the time step; C_{sat} is the solubility limit (g/cm^3) for the contaminant under consideration; θ is the volumetric moisture content of the waste form; and V_{el} is the volume of the waste form. For consistency with the finite element approach, the volume of the waste form is assumed equal to the volume of the finite element.

Using the appropriate value for C, the flux of material released, J_R, is calculated as:

$$J_R = C \cdot V_D \cdot A_b / A_{wf} \quad \text{(Eq. 18)}$$

where V_D is the Darcy velocity (cm/s); A_b is the breached area (cm^2); and A_{wf} is the area of the waste form (cm^2).

At the end of each time step, the mass available for release is reduced to account for radioactive decay and mass released during the time step. Also, a mass balance is performed for the rinse release component of the waste form.

MODEL APPLICATIONS

The BLT code has been extensively tested to verify that the models work as intended and to examine sensitivity of release to different parameters (3,14). To examine the breach and leach models without unnecessary complications, a simple test problem was developed. This set of problems modeled a single soil with a uniform moisture content and Darcy velocity. Tests were conducted to determine the influence of pitting rate, diffusion coefficient, radioactive decay constant, solubility limits, choice of diffusion model, and waste form size on release. Detailed discussions of these tests can be found in Reference 14.

Figure 1 presents the results of a comparison between the three different diffusion models. In this case, pitting was suppressed to prevent penetration before general corrosion consumed the entire container. This occurred after 10 years and therefore, there was no release during this period. After breach, the three different plane diffusion models were used. In each case, the diffusion coefficient was 10^{-6} cm^2/s and the half-thickness of the waste form was 10 cm. For the finite-media model, the length of the water column outside the waste form was twice the length of the waste form. Therefore, the waste form occupied 1/3 the volume of the system.

The zero concentration at the outer boundary (ZCB) model releases mass at a much faster rate than the semi-infinite media or finite-media models. It can be shown that the initial release rate of the ZCB model is twice that of the other two models. The ZCB model releases all of its mass within a few years. The semi-infinite media model requires tens of years to release all of its mass. The difference between the two arises due to the buildup of concentration outside the waste form in the semi-infinite media model. The finite media model does not allow mass out of the system, therefore, the total release is limited by the ratio of the lengths of the water outside the waste form to the length of the entire system, which in this case is 2/3.

Figure 2 presents the effect of varying the pitting rate on total release. In this problem, the pitting rate was varied by changing the pitting parameter, k, in Eq. 1 from 0.02 - 0.15. The average value for k as deter-mined from the NBS data was 0.074. The general corrosion rate was chosen such that the 0.127 cm container wall thickness was consumed in 10 years. The pitting exponent n was set to 0.39, the average value for all soils. The area of the container was taken to be 21,000 cm^2, the area of a 55 gallon drum, and the area exponent "a" of Eq. (1) was taken as 0.2. In this problem, the waste form contained 1 gram available for rinse release and 1 gram available for diffusion release. The diffusion coefficient was 10^{-6} cm^2/s and the ZCB diffusion model was used.

In Fig. 2, at the lowest pitting rate, the container is not breached until 10 years. At this time, there is an extremely fast release due to the rinse mass and early stages of diffusion release. The entire rinse mass of 1 g is released within the first year of leaching. For all other values of the pitting parameter, the first breach occurs after 1 or 2 years. Although rinse release is a fast process, when only a small fraction of the container is breached, only a small fraction of the rinse mass is released. As indicated in Fig. 2, as the breached area increases the release rate increases. For the slower pitting rates, there is a pulse release at 10 years. This is caused by the great increase in flow through the containers after general corrosion has consumed the entire container. For example, in the k = 0.05 case approximately 8% of the container area is breached immediately prior to the entire breach due to general corrosion. Therefore, the rinse and diffusion mass that has been transferred to the rinse model are quickly washed away.

CONCLUSIONS

Models for container breach and waste form leaching have been developed. Container Breach models are applicable to metallic containers and consider pitting and general corrosion. Waste Form Leach models consider surface wash-off, diffusion, and dissolution. These new models

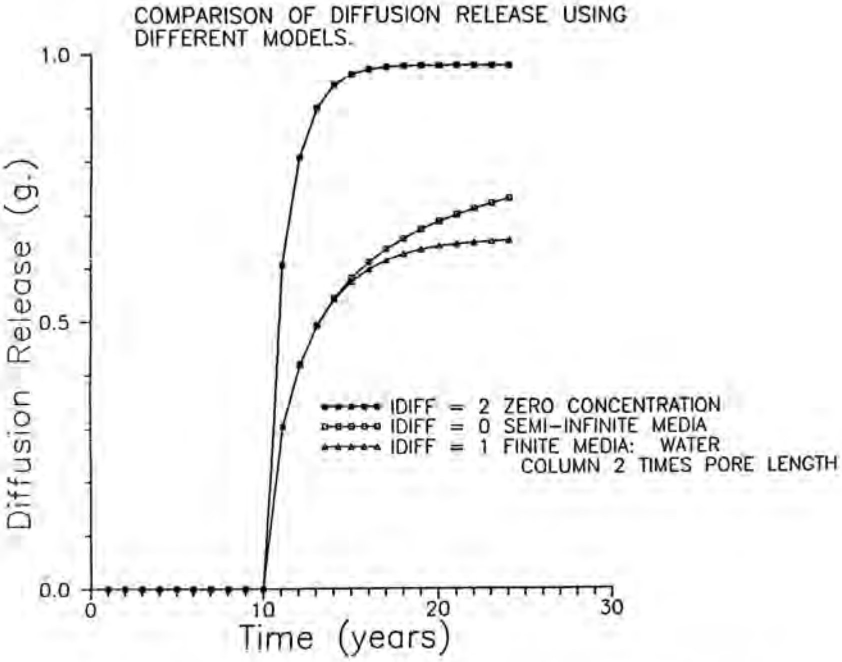

Fig. 1. Comparison of Contaminant Release Due to Diffusion Using Three Plane Diffusion Models.

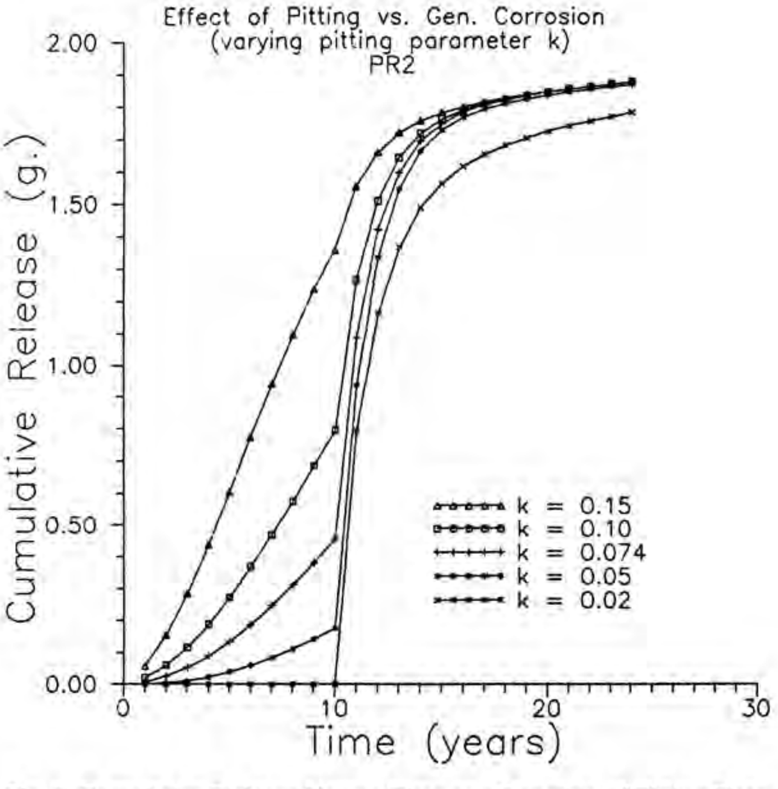

Fig. 2. Cumulative Release Versus Time as a Function of Pitting Rate.

have been incorporated into the BLT computer code which is part of the low-level shallow land burial source term model.

The Breach and Leach models have been verified and sensitivity analyses have been conducted for important model parameters such as diffusion coefficient, solubility limits, and pitting rate.

Current work with the source term model involves benchmarking of predicted results against lysimeter data and improving the diffusion release models such that release is coupled to solution concentration.

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