

LOW-LEVEL WASTE SHALLOW LAND BURIAL SOURCE TERM WASTE FORM LEACHING MODEL DEVELOPMENT FOR THE BLT COMPUTER CODE

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ABSTRACT

A numerical model for leaching from solidified waste forms into a partially saturated porous medium has been developed. This model treats the waste form using the method of finite differences and the solution in the porous medium as a well stirred fluid (i.e., a uniform concentration mixing bath). The model simultaneously considers three release mechanisms: surface rinse, diffusion release, and dissolution release. The model has been verified through comparison to analytical solutions and used to successfully reproduce experimentally measured releases in wet/dry cycle leach tests. The model has been incorporated into the BLT computer code and is an improvement over previous models in the code because it treats all three release mechanisms simultaneously, allows solution concentration buildup in the mixing bath to influence diffusive releases, and allows partitioning of the contaminant between the waste form and the contacting solution.

INTRODUCTION

A general computer model has been developed to predict the release and transport (i.e., source term) of radionuclides from shallow land burial facilities (1). 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. The finite element computer code FEMWATER (2) predicts water flow and the code BLT (3,4), a modified version of the FEMWASTE code (5), predicts container Breach (6,7), waste form Leaching (7), and radionuclide Transport. This paper discusses model improvements for the leaching aspects of the source term model.

The waste form leaching model described previously considers three processes that cause release: surface wash off, diffusion, and dissolution (3,7). The surface wash off model is a mixing bath model that assumes radionuclides are readily washed off of the surface of the waste form constrained by solubility limits. It is most applicable to lab trash and other surface contaminated wastes such as dewatered resin beads. The diffusion model consists of analytical solutions to the diffusion equation for different boundary conditions and geometries (plane or cylindrical). It would be applicable to solidified wastes. A potential drawback of this model is that it does not allow for solution feedback effects to influence diffusive releases. The dissolution model assumes the radionuclide release rate is proportional to the dissolution rate and would be applicable to activated metals. Although, the leaching model allows the waste form to release radionuclides due to all three processes simultaneously, each of these processes is treated independently.

To remove the independence of the three release mechanisms and to allow solution feedback effects (e.g., concentration buildup outside of the waste form) to influence diffusive releases, the waste form and the surrounding en-

vironment have been modeled numerically in one-dimension (either plane or cylindrical geometry). The waste form is represented using the method of finite differences. Within the waste form, the processes of diffusion, dissolution, and surface wash-off are treated simultaneously. Release from the waste form is directly influenced by the solution concentration surrounding the waste form. The surrounding environment is treated as a mixing bath of uniform concentration. The value for the solution concentration in the mixing bath is calculated from a mass balance based on the rate of release from the waste form and the rate of transport away from the waste form due to advection. This approach is conceptually a substantial improvement over the previous model. In addition, the rinse model has been generalized to allow equilibrium partitioning between the waste form and contacting solution. Partitioning models have been widely used in predicting release from low-level waste disposal sites (8,9,10).

This paper presents the equations used to model the waste form and mixing bath, discusses verification test of the models, and compares model predictions to the experimental results of laboratory wet/dry cyclic leach tests (11).

GOVERNING EQUATIONS

For consistency with the earlier model (7), each release mechanism has a user-specified mass available for release. Thus, in general there are four sets of equations to be solved (one for each release mechanism: surface rinse, diffusion, dissolution, and one for the mixing bath solution). These equations are discussed in the following sections.

Surface Rinse Model

Surface rinse mass is allowed only on the waste form surface and is released directly into solution. Therefore, there is no need to calculate its distribution within the waste

form. Its release rate is treated as a source term in the equation for solution concentration and takes the form:

$$S_r = \frac{M_r (1 - \frac{C_s}{C_{sat}})}{\theta_b V_s \Delta t} \quad (\text{Eq. 1})$$

where S_r is the rinse mass release rate, M_r is the rinse mass available at the beginning of the time step, C_s is the concentration in solution, C_{sat} is the saturation concentration, θ is the moisture content of the surrounding media, V_s is the volume of the element, and Δt is the time step. The moisture content multiplied by the volume of the element gives the volume of water surrounding the waste form. Therefore, $M_r/\theta V_s$ is the concentration that would occur if all of the rinse mass was released into solution.

The mass available for release is decreased at the end of each time step to account for losses due to releases to the mixing bath solution and radioactive decay.

It is assumed that release is governed by an equilibrium between the solid waste form and liquid phases. As material is moved out of the mixing bath, more material is released to maintain the equilibrium. With the equilibrium assumption M_r of Eq. (1) becomes [12]:

$$M_r = \frac{(M_a(t) - (K_p/\beta)M_s(t))}{1 + K_p/\beta} \quad (\text{Eq. 2})$$

where $M_a(t)$ is the mass available for rinse release at the beginning of the time step, $M_s(t)$ is the mass in the mixing bath at time t , K_p is the partition coefficient and $\beta = \theta V_s/\rho V_{wf}$. Here ρ is the density, and V_{wf} is the volume of the waste form.

Examining Eq. (2) we see that as $K_p \rightarrow 0$, the mass released approaches that available for rinse release. That is, all of the mass is released instantly. In the other extreme, as $K_p \rightarrow \infty$, the mass released is negative and equal to the total mass in solution. That is all of the mass that enters the mixing bath is adsorbed on the solid.

Diffusion Model

For species that are free to diffuse through the waste form the diffusion equation takes the general form:

$$\theta_p \frac{\partial C}{\partial t} = \nabla \cdot \theta_p D \nabla C - \lambda \theta_p C \quad (\text{Eq. 3})$$

where:

- C is the concentration of mobile species within the waste form, (g/cm³);
- t is the time, (s);

θ_p is the moisture content within the waste form (for simplicity this is assumed to be the same as that calculated by FEMWATER for the finite element);

D is the effective diffusion coefficient for the waste form, (cm²/s); and

λ is the decay constant, (1/s).

To couple diffusion release to the contacting solution concentration, it is assumed that the concentration at the outer boundary of the waste form equals that of the contacting solution. For a porous waste form this assumption is equivalent to requiring continuity in concentration as you pass from the outer edge of the pore into the contacting solution.

Dissolution Model

If the waste form is dissolving, Eq. (3) is no longer appropriate. To account for dissolution, one could simply calculate the volume of the waste form that dissolved and multiply that by the concentration in the waste form to obtain the release due to dissolution. This is the approach used in the previous model (7). However, this approach does not account for changes in concentration due to diffusion processes. Also, as the waste form dissolves the distance over which diffusion must occur before mass is released from the waste form changes.

Physically, we view the dissolution process as one in which the outer surface of the waste form is continually removed, see Fig. 1. To model this with a straightforward finite difference approach is problematic. As the waste form dissolves, the region occupied by the waste form decreases. Therefore, the size of the finite difference mesh must decrease. This would require recalculating several variables at each time step to keep up with the changing size of the waste form.

To avoid this problem a change of variables is introduced. In particular, we use the transform:

$$y = x/L(t) \quad (\text{Eq. 4})$$

Using the transformation, Eq. (4), in the diffusion equation, Eq. (3), and evaluating the vector operators in one dimension gives:

$$\begin{aligned} \theta_p \frac{\partial C}{\partial t} = & \frac{\theta_p D}{L^2} \frac{\partial^2 C}{\partial y^2} + \frac{\theta_p u y}{L} \frac{\partial C}{\partial y} \\ & + C_{y1} \left(\frac{-\theta_p D}{y L^2} \frac{\partial C}{\partial y} \right) - \lambda \theta_p C \end{aligned} \quad (\text{Eq. 5})$$

Here $u(t)$ is the waste form dissolution velocity which represents the time rate of change in waste form lengths and takes the form:

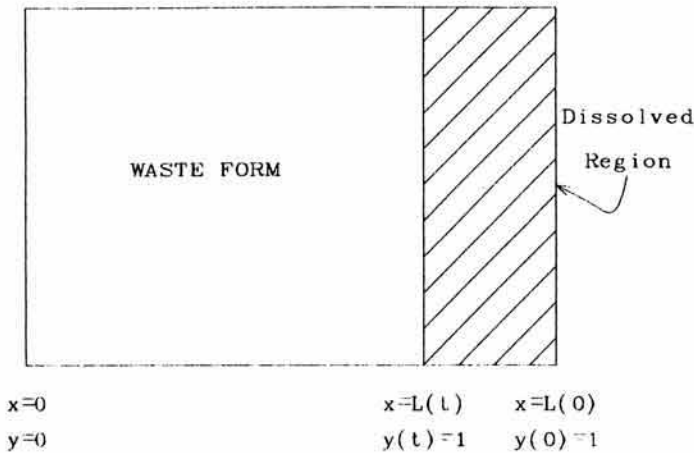


Fig. 1. One dimensional representation of a waste form that undergoes dissolution. x is the distance variable. y is a normalized distance variable that always has the dissolution front at $y=1$.

$$u(t) = \frac{dL}{dt} \quad (\text{Eq. 6})$$

In Eq. (5) C_{yi} takes the value of 0 for plane geometry and 1 for cylindrical geometry. Equation (5) forms the basis for the finite difference equations used to solve for the concentration of mobile species in the waste form.

Some species may be fixed in the structure of the waste form, for example, radionuclides contained by activated metals. These species are immobile and we take the effective diffusion coefficient, D , to be zero. In this case, Eq. (5) simplifies greatly leaving only the dissolution and decay terms.

Solution Concentration Model

The conceptual picture for the mixing bath model is displayed in Fig. 2. Within the finite element framework in BLT, there is a container and a waste form. After breach of the container, we view the environment surrounding the waste form but within the (partially) breached container as a mixing bath. That is, transport processes outside the waste form are fast enough to maintain a uniform concentration. The volumetric flowrate, (Q in Fig. 2) through the mixing bath occurs at a rate determined by the Darcy velocity calculated by FEMWATER and the breached area calculated in subroutine BREACH. Thus, the scaling of release of contaminant for transport by the breached area arises naturally out of the equations. Mass is delivered to the mixing bath at a rate determined by the flux out of the waste

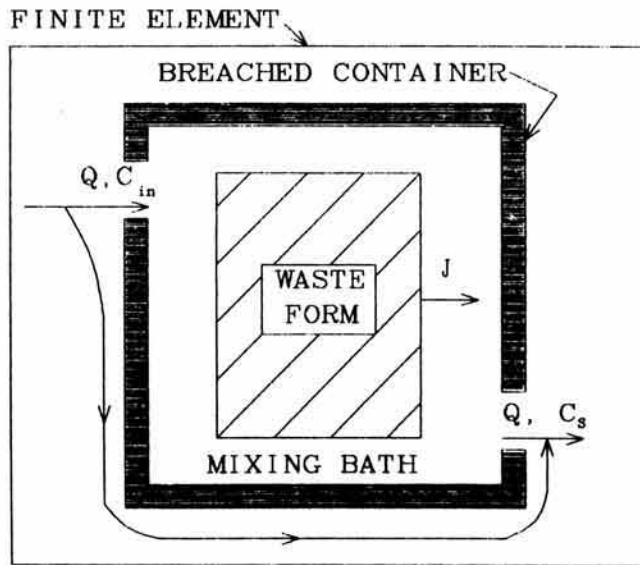


Fig. 2. Conceptual mixing bath model for leaching from partially breached containers.

form (J in Fig. 2) and removed by advection out of the breached container and radioactive decay.

Performing a mass balance within the mixing bath gives:

$$\theta_b \frac{\partial C_s}{\partial t} = \frac{SA}{V_{el}} (J) + \theta_b v_{wf} [C_i - C_s] - \lambda \theta_b C_s + S_r \quad (\text{Eq. 7})$$

where:

θ_b is the volumetric moisture content of the mixing bath. For consistency, θ_b is taken as the value calculated by FEMWATER for the finite element in which the waste form is located. Note, this is the same approximation as for moisture content in the waste form, θ_p .

C_s is the concentration in the mixing bath, (g/cm^3);

SA is the surface area of the waste form, (cm^2);

V_{el} is the volume of the mixing bath, (cm^3);

J is the flux of contaminant out of the waste form, ($\text{g}/\text{cm}^2/\text{s}$);

v_{wf} is the leachant renewal frequency, ($1/\text{s}$).

C_i is the concentration entering the mixing bath, (g/cm^3); and

S_r is the source term due to rinse mass, Eq. (1).

The term on the left of the equal sign represents the rate of accumulation of mass in the mixing bath. The first term to the right of the equal sign represents the rate of delivery of mass to the mixing bath from the waste form. The next term is an advective mass balance term where C_i represents the incoming concentration and C_s the outgoing concentration. The fourth term models radioactive decay and the last term models rinse mass release to solution.

The flux out of the waste form is evaluated with the following expression:

$$J = -\theta D \frac{\partial C}{\partial x} - \theta u C - (1 - \eta) u F \quad \text{Eq. (8)}$$

where η is the porosity of the waste form, F is the concentration of immobile species on the waste form and all other variables have been defined. The concentration and its gradient are calculated at the edge of the waste form. This expression directly couples waste form releases to the solution concentration as diagrammed in Fig. 2.

The leachant renewal frequency, ν_{wf} , is a measure of how fast the water within the mixing bath is replenished and is the ratio of the volumetric flow rate into the mixing bath divided by the volume of water in the mixing bath. It is calculated as:

$$\nu_{wf} = \frac{0.5V_d A_b}{\theta \cdot V_{el}} \quad \text{Eq. (9)}$$

where V_d is the Darcy velocity, A_b is the breached area of the container.

Equations (7) - (9) form the basis for calculating the concentration in solution and are implemented in the finite difference model. A complete discussion of the finite difference model is presented elsewhere (12).

Release for Transport

The release of contaminants from the mixing bath for transport in the BLT code is expressed as:

$$S_t = \nu_{wf}(C_s - C_i) \quad \text{Eq. (10)}$$

where S_t is the rate of material supplied for transport ($\text{g}/\text{cm}^3/\text{s}$).

VERIFICATION OF THE FINITE DIFFERENCE AND MIXING BATH MODELS

Test problems were developed to verify that the finite difference model was working as intended and could reproduce analytical solutions for appropriate conditions. Verification tests were done on the release from the waste form, J in Eq. (8), and release from the mixing bath, Eq. (10).

Analytical solutions of the release from the waste form and the mixing bath were calculated in plane and cylindrical geometry for the following cases:

- Diffusion
- Dissolution
- Diffusion and Dissolution
- Diffusion and Radioactive Decay
- Dissolution and Radioactive Decay
- Rinse with partitioning

The finite difference mixing bath model was able to reproduce the analytically predicted release within a few percent in all cases. Deviations were shown to be due to numerical error and could be reduced by taking smaller time steps or using a finer finite difference grid (12). A complete discussion of the analytical solutions and numerical results can be found elsewhere (12).

Along with verifying the models, a number of important observations were made on the relative importance of various parameters on release. These observations are best explained using dimensionless parameters that arise naturally out of the analytical solution to the problem of interest. These are presented in Table I.

MODELING WET/DRY CYCLIC LEACHING OF CESIUM IN CEMENT WASTE FORMS

A series of experiments were conducted at BNL to determine the effect on leach rates of varying wet and dry periods (11). A full description of these experiments can be found in the report referenced above. For the purposes of this discussion, a summary of this experiment follows.

Leaching was conducted in a column of polyethylene beads which surrounded a cylindrical cement waste form containing resins. The waste form had nominal dimensions of 5×10 cm. The beads were tested for adsorption of Cs by batch column tests and essentially no sorption was measured. The beads were approximately 2 mm in diameter and provided a 5 cm thick cylindrical layer around the waste form. Thus, the pore space around the beads was characterized by fairly large diameter channels as compared to natural soils. This is important because it allows quick drainage and thereby allows essentially no moisture to be retained around the cement during the 'dry' period. In an actual waste burial situation there will always be some moisture. Residual water contents in soils typically range from 5 - 20%.

In a given experiment, the length of the wet and dry periods was fixed. At the end of one wet/dry cycle, the cycle was repeated until the end of the experiment. A series of experiments were run with different wet and dry periods for total experimental times of greater than 200 days. Table II presents the range of wet and dry periods used in the experiments along with an experimental code used for identification. For comparison, one set of tests used a modified IAEA leach test (13).

TABLE I
Relative Importance of Release Parameters

<u>Release from the Waste Form</u>			
Dimensionless Parameter (β)	Result	Rate Controlling Parameter	Comments
$\beta = uL/D$	$\beta > 100$ $\beta < 0.01$	Dissolution Diffusion	$\lambda = 0$, $L = \text{length(radius)}$ of the waste form
$\beta = \lambda L/u$	$\beta > 100$ $\beta < 0.01$	Decay Dissolution	$D = 0$, plane or cylindrical geometry
$\beta = 4L^2/\pi^2 D$	$\beta > 100$ $\beta < 0.01$	Decay Diffusion	$u = 0$, plane geometry
<u>Maximum Release from the Mixing Bath</u>			
$\beta = v/(v+\lambda)$	$v > 100\lambda$ $v < 0.01\lambda$	Advection Decay	Decay unimportant. Mass released from the waste form decays in the mixing bath.

- Note: a) The maximum release from the mixing bath is the release from the waste form multiplied by $(v/(v+\lambda))$.
 b) If retardation occurs in the mixing bath, v is replaced by v/R in the expressions above. Here R is the retardation coefficient.
 c) All parameters in Table I have been previously defined in the text.

Selection of Wet/Dry Cycle Leaching Parameters

The finite difference mixing bath model for wet/dry cycle leaching assumed that diffusion was the only release mechanism. During the wet periods release was allowed into a mixing bath. During the dry periods, no release was allowed from the waste form. However, within the waste form diffusion was allowed. Therefore, during the dry periods mass moved from the center towards the outside of the waste form. Experimentally, after a dry period fresh leachant was placed around the waste form. This was simulated by setting the mixing bath solution concentration, C_s , in Eq. (7), to zero during the dry period, allowing diffusion in the waste form, while not allowing release from the waste form. During the wet periods, there was no drainage from the system, therefore, the leachant renewal frequency, ν_{wf} , was set to zero.

There is a consistency problem in modeling a 3-dimensional experiment with a 1-dimensional model. In particu-

TABLE II
List of Wet/Dry Cycle Experimental Conditions

Code	t-wet(d)	t-dry(days)
IAEA	continuous	never
B	1	1
C	3	1
D	1	2
L	1	4
M	1	6
N	1	20

lar, the 1-D model of a cylinder represents a slice of a cylinder and does not consider end effects. In this case, the surface area and the volume of the cylinder in the 1-D model do not correspond to the 3-D system. In the 1-D model, the effective "surface area" per unit height is $2\pi r$ and the effective "volume" per unit height is πr^2 . The volume to surface area ratio is $r/2$. In the experimental cylinder, the surface area is $2\pi r(r+h)$ and the volume is $\pi r^2 h$. The surface area to volume ratio is $(rh)/(2(r+h))$. As $h \rightarrow \infty$ this ratio reduces to the 1-D case.

For a wide range of waste forms, Cs release has been shown to follow a relationship of the form $\text{CFR}(\text{V/SA})$ is constant (11). This relationship is valid provided the CFR (cumulative fractional release) is less than 0.2 and the semi-infinite medium approximation for release can be used. Therefore, if the appropriate scaling is used, the 1-D and 3-D results can be compared directly. For this reason the results are presented in this format.

Whenever possible, the parameters for the 1-D finite difference model were selected to be consistent with the experiments. The diffusion coefficient, the waste form radius, the length of the wet and dry periods, and the volume of the mixing bath solution were all chosen to correspond to the experimental conditions described in (11).

The scaling is also important in determining the volume of water in the mixing bath. In the numerical modeling, the volume of water is taken to be 10 times the "surface area" of the 1-D cylinder ($2\pi r$). Table III lists the important input variables and their experimental values. For the volume of the leachant, Table III gives the total volume of water which equals the moisture content times the volume of the mixing bath.

Comparison of Experimental and Predicted CFR (V/SA)

Table IV presents the results of this investigation, it contains the length of the wet and dry periods, and three sets of $\text{CFR}(\text{V/SA})$ values: the experimentally measured values, the numerically predicted values, and the values one would obtain if one assumed that there was no diffusion within the waste form during the dry periods. This is just the release at the time corresponding to the number of days the waste form was immersed (i.e., for a 1 day wet and 1 day dry cycle, the release in this column corresponds to release after 1/2 of the experiment time). Results are presented at 150 days, the longest time at which all of the experiments were run.

As seen from Table IV the finite difference model does an excellent job of predicting releases under wet/dry cycle leaching conditions characterized by extended dry periods. Simply assuming that nothing happens during the dry period (Immersed $\text{CFR}(\text{V/SA})$) leads to a large underprediction of release. The finite difference model tends to slightly

overpredict the experimentally measured release rates. This is believed to be due to the slightly different V/SA ratios used in the finite difference model ($\text{V/SA} = 1.18$) and the actual experiment ($\text{V/SA} = 0.95$). At the experimentally measured CFR's at 150 days (0.3-0.5), the semi-infinite medium and therefore V/SA scaling do not exactly apply.

Comparing the analytical solutions for continuous immersion of the 3-D cylinder used in the experiment, $r = 2.35$, $H = 9.8$ cm, with a 1-D cylinder of $r = 2.35$ shows that at 150 days, the CFR of the 3-D cylinder is 46% and the $\text{CFR}(\text{V/SA})$ is 44%. In contrast, for the 1-D cylinder, the CFR is 39% and the $\text{CFR}(\text{V/SA})$ is 46%. Thus, the 1-D solution over predicts the 3-D solution for $\text{CFR}(\text{V/SA})$. Comparison of the 1-D and 3-D solutions for CFR values less than 20% show agreement in the $\text{CFR}(\text{V/SA})$ value to within 0.5%.

Although, the modeling of the wet dry cycle leaching experiments was successful, there are a number of areas that require future consideration. These include examining the behavior of Cs under periods of extended wet conditions. In this case, concentrations in the leachant may buildup and reduce releases; examining the behavior of Sr, Sr exhibited markedly different behavior which is not easily explained by diffusion (i.e., Sr showed a much greater reduction in release than Cs under the same conditions); and examining releases in partially saturated soils. As designed, these experiments fully drained the porous bead matrix around the waste form. In an actual situation residual moisture will exist under all conditions.

CONCLUSION

The model for leaching of solid waste forms in a partially saturated porous medium has been developed, verified against known analytical solutions, and compared to wet/dry cycle leaching experimental results. The model treats the waste form using the method of finite differences in plane or cylindrical geometry. The solution surrounding the waste form is treated using a mixing bath approach. The model has been incorporated into the BLT computer code. It improves upon previous models in the code as it treats all release processes simultaneously, allows solution concentration buildup to influence diffusive releases, and allows partitioning between the waste form and contacting solution.

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TABLE III
Input Variables Used to Predict Wet/Dry Cycle Releases

Parameter	Input Value	Experimental Value
Diffusion Coeff.	1.6E-8 cm ² /s	1.6E-8 cm ² /s
Radius	2.35 cm	2.35 cm
Surface Area	14.8 cm ²	179.4 cm ²
Volume (Waste Form)	17.4 cm ³	170.2 cm ³
V/SA	1.18 cm	0.95 cm
Volume (Leachant)	148 cm ³	1800 cm ³

TABLE IV
Comparison of Experimental and Predicted CFR(V/SA) at 150 Days

Exptl Code	t-wet (days)	t-dry (days)	Exptl ¹ CFR(V/SA)	Predicted CFR(V/SA)	Immersed CFR(V/SA)
IAEA	cont.	-	0.46	0.47	0.46
B	1	1	0.41	0.44	0.33
D	1	2	0.36	0.40	0.27
L	1	4	0.34	0.36	0.21
M	1	6	0.29	0.32	0.17
N	1	20	nr ²	0.18	0.10

¹ All experimental values are from Arora, 1986.

² nr - not reported.

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