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**PERFORMANCE MODELING OF CONCRETE/METAL BARRIERS
USED IN LOW-LEVEL WASTE DISPOSAL**

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ABSTRACT

Low-Level radioactive wastes generated in government and commercial operations involving nuclear materials need to be isolated from the environment almost in perpetuity (at least for 300 to 500 years or, in some cases, even longer). An increasing number of disposal sites are using concrete/metal barriers (so called "engineered" barriers) to isolate these wastes from the environment. Two major concerns hamper the use of engineered barriers; namely, the lack of ability to reliably predict the service life of these barriers and to

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estimate the confidence level of the service life predicted. Computer codes (SOURCE1 and SOURCE2) for estimating the long-term (centuries to millennia) service life of these barriers are presented. These codes use mathematical models (based on past observations, currently accepted data, and established theories) to predict behavior into the future. Processes modeled for concrete degradation include sulfate attack, calcium hydroxide leaching, and reinforcement corrosion. The loss of structural integrity due to cracking is also modeled. Mechanisms modeled for nuclide leaching include advection and diffusion. The coupled or linked effects of these models are addressed in the codes. Typical outputs from the codes are presented and analyzed. These results suggest that major efforts (including research) are needed to compare, evaluate, verify, and eventually validate codes (such as SOURCE1 and SOURCE2) for predicting the long-term behavior of engineered barriers.

Key Words

Advective leaching, concrete degradation, concrete barriers, concrete service life, diffusive leaching, low-level radioactive waste, low-level waste disposal, mass transport, mathematical models, mechanistic modeling, metal barriers, performance assessment, performance testing, source terms, waste disposal units, waste form leaching.

INTRODUCTION

Site-specific radiological performance assessments are required for the disposal of low-level radioactive waste. The purpose of these assessments is to demonstrate compliance, over time, with the performance objectives for low-level waste (LLW) disposal as set forth by the appropriate authorities (local, state, and/or federal). Silo, trench, tumulus, vault, and well disposal units are currently utilized at LLW facilities. These disposal units are constructed with a variety of engineered concrete and metal barriers that are designed to isolate the waste from the environment. An estimate of the release of radionuclides from these units over time (i.e., a source term) is needed to provide input information (e.g., ref. 1) for hydrogeological models that evaluate the migration of radionuclides within the environment.

The SOURCE1 and SOURCE2 computer codes (collectively called the SOURCE computer codes) were developed [2] to provide estimates of the rate at which radionuclides might be released from disposal units. It should be noted that the codes are equally applicable to stable nuclides and other contaminants. A description of the SOURCE codes methodology is presented. First, a general overview of these codes is given. Then, a description of the modeling of concrete

degradation is provided. This description is followed by an outline of the structural analysis and concrete-cracking models. Next, the numerical algorithms for determining radionuclide release rates caused by advection and diffusion are presented. Finally, results for a few sample cases are presented and compared to illustrate applications and responses of the codes to several key input parameters.

OVERVIEW OF THE CODES

The SOURCE computer codes are used to estimate the source term, or radionuclide release rate, from various types of disposal units. SOURCE1 simulates releases from a tumulus-type disposal unit (Fig. 1). SOURCE2 simulates releases from silo, well, well-in-silo, and trench disposal units. A typical silo-type disposal unit modeled in SOURCE2 is presented in Fig. 2. Figure 3 shows the logic flow of the SOURCE computer codes. This logic flow is common to both SOURCE1 and SOURCE2.

Radionuclide release rates from waste disposal units are a function of the integrity of the waste (or waste form) and the engineered barriers used in construction of the disposal unit. When intact, engineered barriers minimize the contact

Fig. 1. Representative tumulus-type disposal unit modeled by SOURCE1.

*SOURCE2 IS APPLICABLE TO WELLS, WELLS INSIDE SILOS, AND UNLINED TRENCHES.

Fig. 2. Representative silo-type disposal unit modeled by SOURCE2. (SOURCE2 is applicable to silo, well, well-in-silo, and unlined trench disposal technologies.)

Fig. 3. Logic flow in the SOURCE computer codes.

of water with the waste, thereby minimizing releases of radionuclides from a disposal unit. As the barriers deteriorate over time, water can more readily contact the waste and mobilize radionuclides, thereby accelerating releases to the environment.

The SOURCE codes predict the long-term performance of engineered barriers used in waste disposal units. Changes in the material properties of the barriers caused by chemical attack and physical stress are modeled. The projected material properties are then considered in structural and cracking analyses of the disposal units. These analyses are performed to assess the ability of the disposal units to bear the loads placed upon them. As the ability to bear design loads is compromised and the structures crack or fail, rates of percolation of water through the waste are adjusted.

The SOURCE computer codes consider two mechanisms through which waste radionuclides are released to the environment, advection (bulk flow driven by hydraulic pressure differences) and diffusion (nuclide movement driven by concentration differences). The calculated total release rate resulting from advection and diffusion is compared with the solubility limit of the nuclide in water. If this limit is exceeded, the release rate is adjusted to the value dictated by the solubility limit. As the disposal units

degrade, rates of percolation of water through the waste increases. Thus, except for cases constrained by solubility, advective releases will increase and, in general, dominate the total release.

The output of the SOURCE codes includes summaries of the barrier degradation and failure analyses, and contaminant release rates as a function of time. The generation of a source term with these codes requires more than 100 input parameters to describe the physical and chemical characteristics of the disposal unit and waste type under consideration. A summary of the input parameters required is presented in Table 1. Table 2 provides an abridged output listing for a SOURCE1 simulation for ^{137}Cs .

CONCRETE DEGRADATION

Concrete is widely used in waste storage and disposal; thus, computer codes to predict the long-term behavior of concrete under service conditions are highly desirable. A number of codes (e.g., refs. 1-4) have been and are being developed worldwide. The degree to which such codes mimic the real-world, long-term behavior of concrete is generally acknowledged to depend upon numerous factors. Mechanistic and/or empirical models for many of these factors are not currently understood sufficiently or available at all. Foremost among the factors that limit the confidence placed

TABLE 1--Sample input data for the SOURCE1 code

Simulation Length: 1000 years Output Edit Frequency: 50 years

Flux Entering Trench (cm/month):

Jan.	Feb.	Mar.	April	May	June	July	Aug.	Sept.	Oct.	Nov.	Dec.
9.58	8.56	8.30	5.77	7.43	0.67	0.20	0.25	0.16	0.10	0.07	6.86

Waste Trench Area 4.66E+02 m³
 Total Dissolved Solids 3.49E+02 ppm
 Groundwater Temp 1.50E+01 C
 Groundwater pH 6.75E+00

Saturated Hydraulic Conductivity
 Recharge 5.80E-07 cm/s
 Soil Backfill 3.50E-03 cm/s
 Concrete 1.00E-10 cm/s

Groundwater Constituent Concentrations

Ca ²⁺	2.10E-03 mol/L	Mg ²⁺	5.21E-04 mol/L
Cl ⁻	2.04E-04 mol/L	SO ₄ ²⁻	2.62E-04 mol/L
CO ₃	1.00E-03 mol/L	O ₂	1.63E-04 mol/L

Constituent Solubilities

Ca(OH) ₂	2.00E-02 mol/L
CO ₃ ²⁻	1.20E-03 mol/L
Mg ²⁺	1.20E-03 mol/L

Concrete Constituent Concentrations

Calcium Concentration in C-S-H System	1.75E+00 mol/L
Calcium Concentration in Pore Fluid	2.00E-02 mol/L
CaO Content in Cement	2.11E+00 mol/L
Free Cl ⁻	1.00E-02 mol/L
Silica Concentration in C-S-H System	7.10E-01 mol/L

Diffusion Coefficients in Concrete

NaOH, KOH	2.12E-11 m ² /s
Ca(OH) ₂	1.82E-11 m ² /s
Cl ⁻	5.08E-11 m ² /s
CO ₂	1.92E-10 m ² /s
O ₂	2.10E-10 m ² /s
SO ₄ ²⁻	1.06E-11 m ² /s

Concrete Design Specifications

Compressive Strength at 28 Days	3.52E+02 kg/cm ²	Concrete Water/Cement Ratio	4.00E-01
Poisson's Ratio of Concrete	1.50E-01	Concrete Density	2.40E+00 g/cm ³
Modulus of Elasticity of Steel	2.04E+06 kg/cm ²	Concrete Porosity	1.50E-01
Yield Strength of Steel	4.22E+03 kg/cm ²	Cement Content	3.85E+02 kg/m ³
Modulus of Subgrade Reaction	1.41E+02 kg/cm ²	Initial pH	1.25E+01
Young's Modulus of Elasticity	2.04E+05 kg/cm ²		

TABLE 1--(continued)

Tumulus Design Specifications			Cask Dimensions		Concrete Member Thickness	
Layers of Casks	2		Cask Width	1.52E+00 m	Roof	1.78E+01 cm
Number of Casks Wide	8		Cask Length	2.13E+00 m	Walls	1.78E+01 cm
Number of Casks Long	18		Cask Height	1.65E+00 m	Floor	1.78E+01 cm
Steel Reinforcement Radius			Spacing of Steel Reinforcement			
Roof	7.94E-01 cm		Roof			2.54E+01 cm
Walls	6.35E-01 cm		Walls			3.05E+01 cm
Floor	6.35E-01 cm		Floor			3.05E+01 cm
Concrete Cover Thickness on Tension Face			Soil and Waste Properties			
Roof:			Earthen Cover Thickness			1.83E+00 m
X-Direction	7.77E+00 cm		Earthen Cover Density			1.76E+00 g/cm ²
Y-Direction	9.37E+00 cm		Friction Angle of Waste Backfill			4.00E+01 degree
Walls:			Friction Angle of Soil Backfill			3.00E+01 degree
Horizontal Direction	8.26E+00 cm		Density of Waste Backfill			1.76E+00 g/cm ²
Vertical Direction	9.52E+00 cm		Density of Soil Backfill			1.76E+00 g/cm ³
Floor:			Waste Density			1.76E+00 g/cm ³
X-Direction	5.08E+00 cm		Average Moisture Content of Waste			9.90E-01 vol. frac.
Y-Direction	6.35E+00 cm					
Static Load			Concrete and Waste Package Failure Rates			
Cask Layer 1	3.65E-01 kg/cm ²		Waste Container			
Cask Layer 2	7.10E-01 kg/cm ²		Start of Failure			0.00E+00 year
			Time to Complete Failure			6.00E+01 year
			Epoxy Coating			
			Start of Failure			0.00E+00 year
			Time to Complete Failure			2.00E+01 year
Nuclide-Specific Parameters						
Nuclide	Half-life (year)	Solubility (mol/L)	Waste Kd (ml/g)	Waste (m ² /s)	Concrete (m ² /s)	Initial Inven. (g)
Cs-137	3.00E+01	1.60E+01	1.99E+01	6.80E-12	5.12E-13	5.81E-04

TABLE 2--Sample output data (abridged) for the SOURCE1 code

Nuclide: ¹³⁷Cs

Year	Vault inventory (g)		Leaching rate (g/year)		
	Intact	Cracked	Advection	Diffusion	Total
1	0.567730E-03	0.000000E+00	0.562810E-11	0.000000E+00	0.562810E-11
2	0.554763E-03	0.000000E+00	0.109991E-10	0.000000E+00	0.109991E-10
3	0.542092E-03	0.000000E+00	0.161218E-10	0.000000E+00	0.161218E-10
4	0.529711E-03	0.000000E+00	0.519010E-11	0.000000E+00	0.519010E-11
5	0.517612E-03	0.000000E+00	0.256563E-10	0.000000E+00	0.256563E-10
⋮					
100	0.575915E-04	0.000000E+00	0.342576E-10	0.368914E-08	0.372340E-08
101	0.562723E-04	0.000000E+00	0.334730E-10	0.383311E-08	0.386658E-08
102	0.274915E-04	0.268883E-04	0.607502E-06	0.397833E-08	0.611480E-06
103	0.265668E-04	0.259839E-04	0.587071E-06	0.407941E-08	0.591150E-06
104	0.256731E-04	0.251098E-04	0.567325E-06	0.417874E-08	0.571503E-06
105	0.248094E-04	0.242650E-04	0.548240E-06	0.427617E-08	0.552516E-06
⋮					
169	0.271749E-05	0.265781E-05	0.601015E-07	0.500926E-08	0.651108E-07
170	0.262379E-05	0.256617E-05	0.580305E-07	0.495572E-08	0.629862E-07
171	0.000000E+00	0.495526E-05	0.112056E-06	0.490143E-08	0.116958E-06
172	0.000000E+00	0.473106E-05	0.106989E-06	0.479262E-08	0.111781E-06
173	0.000000E+00	0.451689E-05	0.102148E-06	0.468501E-08	0.106833E-06
⋮					
200	0.000000E+00	0.127984E-05	0.289655E-07	0.232333E-08	0.312888E-07

TABLE 2--(continued)

CONCRETE DEGRADATION SUMMARY FOR YEAR 200

Concrete Member Thickness		Concrete Loss Due to Sulfate Attack		Fractional Loss Of Yield Strength Due to $\text{Ca}(\text{OH})_2$ Leaching	
Roof	7.57E+00 cm	Roof	1.02E+01 cm	Roof	8.52E-02
Walls	7.57E+00 cm	Walls	1.02E+01 cm	Walls	8.52E-02
Floor	7.57E+00 cm	Floor	1.02E+01 cm	Floor	8.52E-02

CORROSION RESULTS

Time to Onset of Corrosion		Corrosion Product Layer Thickness		Remaining Steel Reinforcement	
Roof	0 years	Roof	0.00E+00 cm	Roof	7.94E-01 cm
Walls	0 years	Walls	0.00E+00 cm	Walls	6.35E-01 cm
Floor	0 years	Floor	0.00E+00 cm	Floor	6.35E-01 cm

CONCRETE CRACKING ANALYSIS FOR YEAR 200

Cracking Due to Corrosion of Steel			Cracking Due to Loading and Shear	
Cask Roof	None	Cask Roof	Cracked	
Cask Walls	None	Cask Walls	None	
Cask Floor	None	Cask Floor	Cracked	

Crack Characteristics:

Upper Vault		Lower Vault	
Cask Roof	7.44E-04	Cask Roof	2.80E-02
Average Crack Width (cm)	1.72E-05	Average Crack Width (cm)	5.54E-04
Fractional Volume of Cracks		Fractional Volume of Cracks	
Cask Floor	0.00E+00	Cask Floor	2.09E-02
Average Crack Width (cm)	0.00E+00	Average Crack Width (cm)	4.95E-04
Fractional Volume of Cracks		Fractional Volume of Cracks	

in these predictive codes are the validity of the models used to extrapolate concrete degradation and to evaluate mass transport through the concrete. The completeness of the models and the correctness of the linkages utilized between models that influence other models are generally not well known. This can restrict further the confidence placed in these codes. Essentially all models currently available to predict the long-term behavior of concrete deal with effects caused by external factors (e.g., pH, Eh, and ionic composition of surrounding water). Very few models are available that deal with effects caused by internal factors (e.g., added inorganic and organic materials deleterious to concrete) and of those available, the majority are not generically applicable.

The numerous factors that influence the durability of concrete [3-6] can be grouped broadly as chemical attack, physical stress, and microbial action. Degradation caused by chemical attack includes the effects of sulfate attack, calcium hydroxide leaching, alkali-aggregate reaction, salt crystallization, and metal reinforcement corrosion. Degradation caused by physical stress includes the effects of freeze-thaw cycles, wet-dry cycles, and osmotic pressure. Degradation caused by microbial action includes the effects of sulfur-oxidizing and nitrifying bacteria and heterotrophic organisms.

The SOURCE codes include the chemical degradation mechanisms of sulfate attack, calcium hydroxide leaching , and steel reinforcement corrosion. The codes assume that initially flow through concrete barriers is described by the permeability of the concrete and that cracks are caused by subsequent degradation as previously described herein. Unfortunately, data on original cracks (i.e., those formed during curing and aging before a disposal unit is put into service) in concrete are, for the most part, not available. The latter observation can be extended to include data on the initial quality of the concrete used in construction of a disposal unit, on the skill with which the concrete is emplaced, and on the conditions to which the concrete is exposed after emplacement but before a performance assessment begins.

Sulfate Attack

Sulfate attack occurs when sulfate ions from the environment penetrate concrete and react expansively with the concrete. The resulting internal expansion causes stress, cracking, and exfoliation. The sulfate attack model is based on the work given in ref. 7. The rate of degradation can be expressed as:

$$R = \frac{E\beta^2 C_o C_e D_i}{\alpha\gamma(1-\mu_c)} , \quad \text{Eq. (1)}$$

where

R = degradation rate, m/s;

E = Young's modulus, Pa;

β = linear strain caused by a mole of sulfate reacted in a m^3 ;

c_o = groundwater sulfate concentration, mol/ m^3 ;

C_e = concentration of sulfate as ettringite, mol/ m^3 ;

D_i = intrinsic diffusion coefficient, m^2/s

α = roughness factor for fracture path, dimensionless;

γ = fracture surface energy of concrete, J/ m^2 ; and

μ_c = Poisson's ratio for concrete, dimensionless.

Calcium Hydroxide Leaching

As NaOH, KOH, and $Ca(OH)_2$ are leached from the concrete, the pH of the concrete is adjusted. The pH declines linearly in direct proportion to the reduction in NaOH and KOH. After NaOH and KOH are totally leached from the concrete, changes in pH are modeled as a polynomial function of the $Ca(OH)_2$ content. As the $Ca(OH)_2$ is leached, the strength of the concrete declines. The calcium-hydroxide-leaching model includes advective and diffusive flow and is based on the work described in refs. 8-11. If the groundwater is not saturated with calcium carbonate, advective leaching of $Ca(OH)_2$ is calculated using[9]

$$Ca_1 = I \frac{Ca_p}{C_t Ca_c}, \quad \text{Eq. (2)}$$

where

- Ca_1 = groundwater release rate of $Ca(OH)_2$, year^{-1} ;
 I = percolation rate through concrete, m/year ;
 Ca_p = $Ca(OH)_2$ concentration in concrete pore solution,
 mol/L ;
 C_t = concrete member thickness, m ; and
 Ca_c = $Ca(OH)_2$ concentration in concrete, mol/L .

Corrosion of Steel Reinforcement

Corrosion of steel reinforcement is initiated upon depassivation of the steel by carbonation or chloride penetration. The corrosion propagates at a rate determined by the rate of diffusion of oxygen to the steel. The corrosion-of-steel-reinforcement model is based on work reported in refs. 12 and 13. The flow rate of oxygen to the surface of the steel reinforcement is modeled using Fick's first law of diffusion

$$J_o = -D_o A \left(\frac{d[O_2]}{dx} \right), \quad \text{Eq. (3)}$$

where

- J_o = oxygen flow rate at the steel reinforcement,
 g/s ;

- D_0 = effective diffusion coefficient of oxygen through concrete, cm^2/s ;
- A = surface area over which oxygen diffuses to the reinforcement, cm^2 ; and
- $\frac{d[\text{O}_2]}{dx}$ = dissolved oxygen concentration gradient, g/cm^4 .

The rate of oxygen consumption by the corrosion reaction is assumed to be greater than the rate of oxygen diffusion to the reaction surface. Under these conditions, the corrosion rate is limited by the oxygen flow rate at the steel reinforcement.

Steel that is epoxy-coated may delay the onset of corrosion by providing isolation from aggressive ions and oxygen. The epoxy coating is not assumed to delay the time of depassivation of the reinforcement. However, upon depassivation, the assumption is made that corrosion is prevented as long as the coating remains intact. The epoxy coating is predicted to fail as a linear function of time.

Corrosion of Metal Barriers

Corrosion of components other than steel reinforcement also affects the long-term performance of a disposal unit. Specifically, metal boxes placed inside vaults, corrugated-

steel liners used in the construction of silos, and cast-iron pipes used in well construction will all fail eventually because of corrosion. The SOURCE codes consider corrosion of steel and iron barriers used in tumulus, silo, well, well-in-silo, and trench disposal technologies. Failure rates of these barriers are modeled using linear failure functions. The user specifies the time at which corrosion of the metal component begins and the number of years required, following this specified time, for the member to fail completely. Using these data, a failure fraction is calculated for each time period of the simulation.

CONCRETE STRUCTURAL AND CRACKING ANALYSES

Two distinct analyses are carried out to evaluate the long-term performance of the disposal units caused by chemical attack and physical stress. First, a structural analysis is carried out, and then a cracking analysis is performed. These analyses[2] are of such complexity and length that they can be described only generically in this paper.

Concrete Structural Analysis

The structural analysis (based upon the loads placed on the disposal unit) is used to determine shear, bending moments, axial tension, and compressive forces placed on the various

structural components. As these loads vary with the structural component under consideration, this analysis is carried out for the roof, wall(s), and floor of each disposal unit. The analysis for the roof is used to evaluate shear forces and bending moments caused by uniform loads. For the wall(s), the structural analysis is used to calculate shear forces caused by uniform and hydrostatic loads, bending moments caused by uniform and hydrostatic loads, compressive forces caused by roof reaction and wall weight, as well as axial and ring compressive forces for the silo wall. Finally, the floor structural analysis is utilized to determine shear forces and bending moments caused by concentrated loads.

Concrete-Cracking Analysis

The concrete-cracking analysis is used to monitor the structural integrity of the disposal unit by comparing shears, bending moments, axial tensions, and compressive forces to loads and forces at which structural or cracking failure will occur. Cracking caused by shear occurs if the shear force on a concrete component exceeds the cracking shear of the member. Cracking caused by bending occurs if bending moments for a concrete member exceed the cracking moment for the component. These cracks will penetrate the member if the bending moment exceeds the ultimate strength of the member. Cracking caused by compression occurs if the

compressive forces on the wall exceed the ultimate strength of the wall. Finally, cracking caused by corrosion of steel reinforcement occurs if the tension stress at the steel-concrete interface exceeds the tensile strength of the concrete.

Changes in concrete properties caused by chemical attack and physical stress must be taken into account in the cracking analysis. Therefore, it is closely coupled to the concrete degradation analysis throughout the simulation. This coupling is illustrated in the concrete degradation and cracking logic flow presented in Fig. 4.

CONTAMINANT LEACHING MODELS

Two mass transport mechanisms are modeled in the SOURCE codes, advection and diffusion. The total release by these mechanisms cannot exceed the solubility limit of a nuclide. Rates of release from disposal facilities which have not undergone significant structural failure will generally be low (dependent largely on the relative water saturation of the waste and concrete) and, for the most part, the result of diffusion. As the facility deteriorates and undergoes cracking, water may more easily percolate through the waste. Under these conditions, leaching of radionuclides caused by advection will be accelerated and tend to overshadow leaching caused by diffusion.

Fig. 4. Logic flow in concrete degradation and cracking subroutines for the SOURCE codes.

Leaching Caused by Advection

Leaching of radionuclides caused by advection will be directly proportional to the amount of water contacting the waste and inversely proportional to the degree to which radionuclides are sorbed by the waste matrix. Two different models are used to simulate these mechanisms, a zero-order model[2] and a first-order model.[14] The first-order model was incorporated into the codes for comparison with the zero-order model. Both models account for radioactive decay.

Zero-order advective model--The zero-order model considers the advective leach rate to be independent of time within a time step (hence, the term "zero-order"). The inventory of a nuclide is adjusted at the beginning of the subsequent time step. This model quantifies the radionuclide release rate in the following manner:[2]

$$\Delta Q = \left[\frac{Q_s}{h_w(H_w + K_d\rho_w)} \sum_{i=1}^{12} I_i \right] e^{-\lambda_a t_a}, \quad \text{Eq. (4)}$$

where

ΔQ = radionuclide release due to advection in year a ,
g/year;

- Q_0 = radionuclide inventory available for leaching at the beginning of year a, g;
 h_w = waste thickness, cm;
 H_w = relative saturation of waste (volume H_2O /volume waste);
 K_d = radionuclide distribution coefficient, mL/g;
 ρ_w = density of waste, g/cm³;
 I_i = water percolation rate through the waste during month i, cm/month;
 λ_a = decay constant ($\ln 2/t_{1/2}$), year⁻¹;
 $t_{1/2}$ = half-life of a radionuclide, year; and
 t_a = duration of leaching interval, one year.

First-order advective model--The first-order model introduces time dependence within a time step for the leach rate calculation. This model uses the following equation[14]:

$$q_i = \left[\lambda_{L,i} Q_0 e^{-\lambda_{L,i} t} \right] e^{-\lambda_a t}, \quad \text{Eq. (5)}$$

where

- q_i = radionuclide release rate caused by advection during month i, g/month;

Q_0 = original mass in the contaminated volume, g;

$\lambda_{L,i}$ = leach rate constant for month i , month⁻¹, =

$$\frac{I_i}{h_w(H_w + K_d\rho_w)}; \quad \text{Eq. (6)}$$

λ_d = decay constant, month⁻¹; and

t = time, month.

Leaching Caused by Diffusion

Leaching caused by diffusion is calculated in the SOURCE codes using the FLOTHRU computer program.[15] The FLOTHRU program is based on the model described below.

The waste-concrete system is depicted (Fig. 5) as a two-layer slab with an inner layer of half-thickness a , initially containing a contaminant with concentration C_0 and decay constant λ , and with an outer layer of thickness $b - a$, initially uncontaminated. If C_1 is the concentration in the inner layer, and C_2 is the concentration in the outer layer, the diffusion equations for the contaminant are:

Fig. 5. Representation of the system modeled by the FLOTHRU computer code.

$$D_1 \left(\frac{\partial^2 C_1}{\partial x^2} \right) - \lambda C_1 = \frac{\partial C_1}{\partial t} \quad \text{Eq. (7)}$$

and

$$D_2 \left(\frac{\partial^2 C_2}{\partial x^2} \right) - \lambda C_2 = \frac{\partial C_2}{\partial t} , \quad \text{Eq. (8)}$$

with initial conditions

$$C_1(x, 0) = C_0, \quad 0 \leq x < a \quad \text{Eq. (9)}$$

and

$$C_2(x, 0) = 0, \quad a \leq x < b , \quad \text{Eq. (10)}$$

and boundary conditions

$$C_2(b, t) = 0 , \quad \text{Eq. (11)}$$

$$\left(\frac{\partial C_1}{\partial x} \right)_{x=0} = 0 , \quad \text{Eq. (12)}$$

$$C_1(a, t) = C_2(a, t) , \quad \text{Eq. (13)}$$

and

$$D_1 \left(\frac{\partial C_1}{\partial x} \right)_{x=0} = D_2 \left(\frac{\partial C_2}{\partial x} \right)_{x=0} . \quad \text{Eq. (14)}$$

where

$$D = \text{retarded diffusion coefficient, m}^2/\text{s},$$

$$= \frac{D_s}{G(1 + K)H^{-1}} ; \quad \text{Eq. (15)}$$

D_s = self-diffusion coefficient, m^2/s ;

G = geometry factor, dimensionless;

K = partition coefficient, dimensionless;

H = fraction of pore capacity to hold liquid that is filled, dimensionless;

C = concentration, g/m^3 ;

λ = decay constant, s^{-1} ;

t = time, s; and

x = distance, m.

The mathematical techniques used to solve the above equations are complex and lengthy[15] and are not presented in this paper.

COMPARISON OF ADVECTIVE MODELS

Wastes containing ^{137}Cs ($t_{1/2} = 30$ years) and ^{135}Cs ($t_{1/2} = 3 \times 10^6$ years) were evaluated using the SOURCE1 code to illustrate differences between the two advective models for

different half-life radionuclides. Figures 6 and 7 present the predicted leach rate as a function of time for ^{137}Cs and ^{135}Cs , respectively. The cumulative amount released is taken as the summation of the annual leach rates. Cumulative amounts released are presented in Figs. 8 and 9 for ^{137}Cs and ^{135}Cs , respectively. In general, for relatively short-lived radionuclides (e.g., ^{137}Cs) the zero-order model predicts a higher leach rate and higher cumulative release than the first-order model. However, for relatively long-lived radionuclides (e.g., ^{135}Cs), the two models predict approximately the same release rate and cumulative release. For example, the difference in the cumulative release predicted by the two models for ^{135}Cs is about 5% at 105 years and 2% at 200 years as shown in Fig. 9.

Figure 10 presents a comparison of the zero-order and first-order advective models for two different distribution coefficient, K_d , values. The simulation is for ^{137}Cs with K_d values of 1.9 and 19. For each K_d value, the results show that the zero-order model cumulative amount released exceeds that of the first-order model. Also, as would be expected, both models show a lower cumulative amount released for $K_d = 19$ as compared to $K_d = 1.9$. More extensive simulations need to be carried out before generalizations can be made about differences between these models with respect to K_d .

Fig. 6. Comparison of ^{137}Cs leach rates from a tumulus (SOURCE1) using zero-order and first-order advection models.

Fig. 7. Comparison of ^{135}Cs leach rates from a tumulus (SOURCE1) using zero-order and first-order advection models.

Fig. 8. Comparison of the cumulative amount of ^{137}Cs leached from a tumulus (SOURCE1) using zero-order and first-order advection models.

Fig. 9. Comparison of the cumulative amount of ^{135}Cs leached from a tumulus (SOURCE1) using zero-order and first-order advection models.

Fig. 10. Comparison of the cumulative amounts of ^{137}Cs leached from a tunnel using zero-order and first-order advection models for two values of K_d . (SOURCE:)

For further comparison of the two advective models a sensitivity and uncertainty analysis was performed on the SOURCE1 code. The Latin Hypercube sampling method [16] was used to analyze the effects of input variables on the two advective models. The PRISM code [17] was used to implement the Latin Hypercube sampling technique. Of the slightly more than 100 input parameters required for a code simulation, 47 were selected for examination. The range and mean of these parameters were estimated for ^{137}Cs in a tumulus-type disposal unit. The sensitivity analysis revealed 13 sensitive parameters. Sensitive parameters were taken to be those which contributed at least 5% of the variability in the leach rate calculation. Table 3 summarizes the results of the sensitivity analysis for the two advective models.

TABLE 3--Summary of sensitive parameters determined by a sensitivity analysis of the SOURCE1 code^a

Sensitive parameters ^b	Advective leaching model	
	Zero-order	First-order
Density of earth cover, g/cm ³	X	X
Density of waste, g/cm ³	X	X
Moisture content of waste, vol. H ₂ O/vol. waste	X	X
Diffusion coefficient of SO ₄ ²⁻ in concrete, m ² /s	X	X
Time required for complete corrosion of waste containers, year	X	X
Time required for complete failure of epoxy coating, year	X	X
Saturated hydraulic conductivity of recharge component, cm/s	X	X
Saturated hydraulic conductivity of concrete, cm/s	X	X
Radionuclide distribution		

coefficient, mL/g	X	X
Radionuclide inventory, g	X	X
Concrete diffusion coefficient, m ² /s	X	X
Concentration of SO ₄ ²⁻ inside vault, mol/L		X
Concentration of SO ₄ ²⁻ outside vault, mol/L		X

^aRadionuclide leached is ¹³⁷Cs.

^bSensitive parameters are those which contribute at least 5% to the leach rate calculation.

The 13 most sensitive parameters were then utilized in the uncertainty analysis. A most probable value and a distribution were developed for each parameter and used to randomly generate 200 input data sets for the SOURCE1 code. Leach rates for each of these input data sets were then determined for each of the two advective models. A statistical summary of the results is presented as a frequency distribution in Fig. 11. This frequency distribution shows that, in general, the zero-order model will predict a higher leach rate than the first-order model for ¹³⁷Cs. Also, for this case, this distribution shows that the zero-order model is the more conservative of the two advective models.

CONCLUSIONS

The SOURCE computer codes provide a one-dimensional model for nuclide (stable and radioactive) transport that can be applied to several disposal technologies. The codes account for the degradation of concrete as well as the corrosion of

Fig. 11. Results of uncertainty analyses of SOURCE1 for ^{137}Cs depicting relative frequency γ for zero-order and first-order advection models.

metal waste containers and other metal barriers. These effects are closely coupled to concrete structural and concrete-cracking analyses. The results of these analyses are used to determine the amount of water passing through the disposal unit. Advective and diffusive transport are both modeled to evaluate the leaching of nuclides from a disposal unit. These leach rates, along with barrier degradation and failure analyses, are provided as outputs of the codes.

Future work on these codes should include verification against other codes and validation against field data, as available. The advective leaching models (zero-order and first-order) should be evaluated further to determine which model is more appropriate for a performance assessment. The diffusion model should be revised to allow changing the boundary conditions at each time step. In addition, consideration should probably be given to the treatment of the ingrowth of radioactive daughters. Finally, the need to modify additional concrete and waste properties (e.g., distribution coefficients and diffusion coefficients) as a function of changing conditions should be evaluated.

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