IMPROVED MODELING OF ENGINEERED BARRIERS FOR LOW-LEVEL WASTE DISPOSAL

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ABSTRACT

Interest in the use of engineered concrete barriers in LLW disposal facilities has preceded the development of a comprehensive understanding of the long-term performance of these barriers. With this in mind, Rogers and Associates Engineering Corporation has developed a new disposal facility assessment code, called RAESTRICT. RAESTRICT adopts a more complete, mechanistic approach to modeling concrete degradation, groundwater infiltration, leaching, and radiological risks.

INTRODUCTION

The Low-Level Radioactive Waste Policy Act of 1980 charges states and compacts with the responsibility of providing for the disposal of all low-level radioactive waste (LLW) generated within their boundaries. While shallow land disposal has been used for LLW disposal to date, a number of states and compacts have shown interest in disposal by alternative means. A number of Department of Energy sites are developing or considering alternative disposal technologies as well.

The majority of these alternative technologies employ multiple engineered barriers to improve waste containment or isolation from the environment. While a number of materials are used in barrier construction, the most prevalent is concrete.

Interest in the use of engineered concrete barriers in LLW disposal facilities has preceded the development of a comprehensive understanding of the long term performance and degradation characteristics of these barriers. With this in mind, Rogers and Associates Engineering Corporation, with the backing of the Electric Power Research Institute, developed the BARRIER code (1).

Given the infancy of assessing the performance of alternative disposal technologies involving concrete, the BARRIER code was recognized as a research code. As such, it was one of the first attempts at modeling long-term concrete performance as applied in low-level waste disposal. It was anticipated that the subsequent availability of more data and information on modes of concrete failure would enable improved codes to be developed. Rogers and Associates Engineering Corporation has since undertaken development of a new disposal facility performance assessment code called RAESTRICT (RAdiological Evaluation of the STRuctural Integrity of Concrete Technologies).

The sequence of events comprising a performance assessment using RAESTRICT is illustrated in Fig. 1. The simulation begins with modeling of the unsaturated flow of water from infiltration. Concrete degradation due to chemical and physical attack mechanisms is considered next, followed by a structural analysis of the facility. If the integrity of the structure is compromised, hydrologic properties of the structure are modified and the unsaturated flow conditions are recalculated. When the water is able to flow into the facility, the rates at which nuclides are leached from the waste are calculated, and contaminant release and transport from the facility are modeled. Finally, doses and risks resulting from exposures to the waste and radionuclides leached from the waste are calculated.

RAESTRICT differs from the BARRIER methodology in several aspects. The code adopts a different, more complete, mechanistic approach to modeling concrete degradation, with particular emphasis on the corrosion of steel reinforcement, the loss of calcium hydroxide through leaching, and sulfate attack. The structural analysis capabilities include flexural, shear, tensile and compressive crack development and permit consideration of a broader array of design features and technologies, and loading conditions. In addition, the unsaturated groundwater transport routines are analytically based for ease of use. Finally, a comprehensive set of transport and exposure pathways are included for dose and risk assessments. These new modeling capabilities are discussed below.

CONCRETE DEGRADATION

Corrosion of Steel Reinforcement

Damage due to corrosion of steel reinforcement manifests itself in the form of expansion, cracking, and spalling of the cover. The reinforced-concrete member may sustain structural damage due to the loss of bond between the steel reinforcement and the concrete, and loss of rebar cross-sectional area. This structural damage may occur to such an extent that structural failure is inevitable.

Normally, the highly alkaline environment found in Portland cement concrete protects steel reinforcement from corrosion. In this environment, where the pH may be 12.5 or greater, a tightly adhering iron oxide film forms over the steel, effectively isolating the reinforcement from corrosive agents.

Corrosion may occur if the passive film surrounding the steel reinforcement is destroyed or penetrated. This de-passivation process generally occurs via two primary mechanisms. Carbonation may result in de-passivation of the steel reinforcement due to a lowering in the pH of the concrete. Penetration of chloride ions to the reinforcement
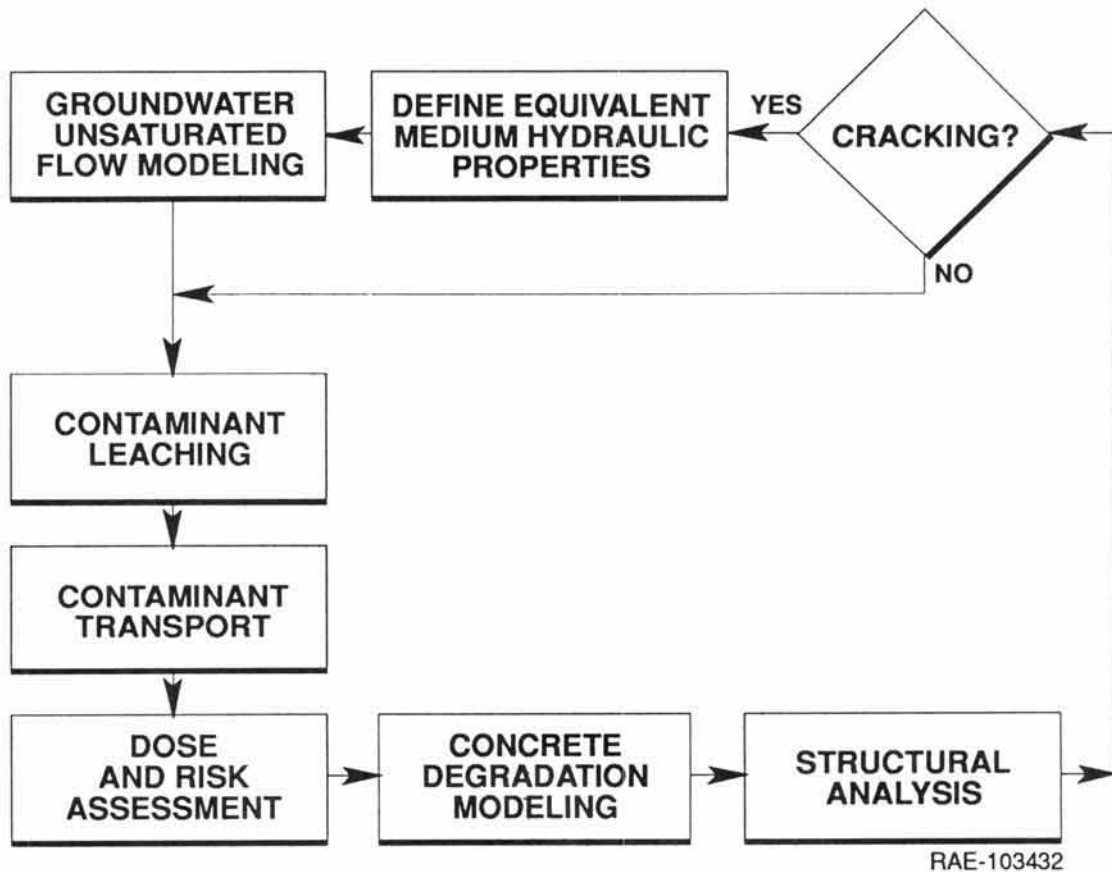
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Fig. 1. Sequence of events in RAESTRICT analysis.

may also disrupt the passive layer through chemical reaction.

RAESTRICT considers the competing effects of chloride ion penetration and carbonation in modeling the initiation phase of corrosion. Carbonation of the concrete will occur as CO_2 diffuses into the concrete. In general terms, the position of the carbonation front in the reinforced concrete is given by

$$X = k \cdot t^{0.5} \qquad \text{Eq. (1)}$$

where

X = depth of carbonation

k = carbonation coefficient

t = time.

The carbonation coefficient is calculated using the work of Tuutti (2) as a basis. Simplifying for assumed boundary conditions, the coefficient is given by

$$k = [(CO_2e/CO_2b-1) \cdot 4 \cdot D_{CO_2} / \pi^{0.5}]^{0.5} \qquad \text{Eq. (2)}$$

where

CO_2e = external concentration of CO_2 (mole/l)

CO_2b = concentration of CO_2 bound in concrete (mole/l)

D_{CO_2} = diffusion coefficient of CO_2 in concrete (m2/s).

The CO_2 diffusing inward that is bound by constituents in the concrete is effectively removed from penetrating to the steel reinforcement and plays no role in de-passivation. The amount of CO_2 bound in the concrete is set equal to quantity of hydrated lime in the concrete (2), calculated as the product of the CaO content in the concrete and the degree of hydration. The degree of hydration is calculated for Portland cements as a function of the water-to-cement ratio (2).

The carbonation front, and the associated pH front, is assumed to be discontinuous in this modeling approach. When the front has penetrated to the depth of the steel reinforcement, de-passivation of the steel is assumed to be complete.

De-passivation of the steel reinforcement due to the penetration of chloride ions in the groundwater and chloride incorporated in the concrete at the time of construction, will occur when a threshold concentration of ions has been achieved at the steel. The chloride concentration at the steel is calculated using a standard solution to Fick's First Law of Diffusion:

$$CL_s = CL_{gw} \cdot [1 - erf(C_d/(4 \cdot D_{cl} \cdot t)^{0.5})] +$$

$$CL_i erf(C_d/(4 \cdot D_{cl} \cdot t)^{0.5}) \qquad \text{Eq. (3)}$$

where

CL_s = chloride ion concentration at the steel reinforment (mole/l)

CL_{gw} = groundwater chloride ion concentration (mole/l)

C_d = concrete cover depth (cm)

D_{cl} = effective diffusivity of chloride ions in concrete (cm2/s)

CL_i = initial chloride ion concentration in concrete (mole/l)

t = time (s).

The threshold concentration of chloride ions required for de-passivation of the steel has been considered by numerous investigators. Hausmann (3) found that a chloride ion to hydroxide ion concentration ratio of 0.61 was sufficient to de-passivate the steel. The hydroxide ion concentration used in this ratio will be a function of the pH of the concrete, itself a function of the alkali and calcium hydroxide content of the concrete.

De-passivation of the steel reinforcement, through either carbonation or chloride ion penetration, marks the beginning of the propagation phase. The rate at which corrosion proceeds will be a function of numerous factors, among them the electrical resistivity of the concrete and the supply of oxygen at the steel reinforcement.

The resistivity of the concrete is determined by a variety of factors, including the moisture and chloride ion content of the concrete, and temperature (4,5). For modeling purposes, data from Ref. 5 were regressed on moisture content and chloride ion concentration to predict concrete resistivity. When the calculated resistance is less than the threshold resistance, supplied by the user, corrosion may occur. Threshold resistivities are generally 5,000 to 10,000 ohm cm, but can be as high as 60,000 ohm cm (5,6).

The corrosion reaction requires the presence of oxygen at the reinforcement. The diffusive flux of oxygen to the steel can be calculated using Fick's First Law of Diffusion:

$$J_o = D_o A\, P_c\, d \left[\frac{O_2}{dx}\right] \qquad \text{Eq. (4)}$$

where

J_o = oxygen flux (g/cm^2-s)

D_o = effective diffusivity of O_2 through cement paste (cm^2/s)

A = cross-sectional area of steel reinforcement (cm^2)

P_c = porosity of concrete

$d \left[\dfrac{O_2}{dx}\right]$ = dissolved oxygen concentration gradient (g/cm^4).

This approach assumes that (1) the growth of the reaction product layer around the reinforcement is slow compared to the rate of O_2 diffusion through said layer, and (2) the rate of O_2 diffusion through the product layer is fast relative to diffusion rates through the cement paste. If it is also assumed that the rate of oxygen consumption by the corrosion reaction is greater than the rate of diffusion of oxygen to the reaction interface, the corrosion rate is seen to be limited by the flux of oxygen.

Corrosion of steel reinforcement may be curtailed through the use of epoxy-coated reinforcement. RAESTRICT models the long-term performance of the epoxy-coating by assuming a linear rate of failure and calculates a fraction of the coating which has failed. This fraction is used to modify the area of steel reinforcement available for corrosion.

Calcium Hydroxide Leaching

Leaching of calcium hydroxide ($Ca(OH)_2$) from concrete results in a reduction in concrete strength and a decline in pH. The loss in strength will affect the concrete structure's ability to withstand the loads placed upon it. A decline in the pH of the concrete may favor de-passivation of the reinforcement and the initiation of steel corrosion.

$Ca(OH)_2$ will leach from the concrete due to both diffusion and groundwater, or advective, mechanisms. The potential for groundwater leaching of $Ca(OH)_2$ will depend on the level of groundwater saturation with respect to calcium carbonate. If the groundwater is saturated with calcium carbonate no dissolution of $Ca(OH)_2$ will occur. Percolating groundwater which is not saturated may leach $Ca(OH)_2$ from the concrete.

The degree of calcium carbonate saturation of groundwater is characterized using the calcium carbonate saturation, or Langelier index (7). This index accounts for the effects of temperature, total dissolved solids, total alkalinity, pH, and calcium content on the saturation characteristics of the groundwater.

The Langelier index is predicted in RAESTRICT using a regression of the investigator's experimental data (7) on groundwater temperature and total dissolved solids. If the index is negative, indicating that the groundwater is not saturated with calcium carbonate, leaching of $Ca(OH)_2$ from the disposal structure is calculated using

$$L_g = P/C_{thk} \cdot C_p/C_c \qquad \text{Eq. (5)}$$

where

L_g = Ca(OH)$_2$ groundwater leach rate (yr^{-1})

P = percolation rate through repository (m/yr)

C_{thk} = thickness of concrete structural component (m)

C_p = Ca(OH)$_2$ concentration in concrete pore solution (mole/l)

C_c = Ca(OH)$_2$ concentration in concrete (mole/l).

Rates of Ca(OH)$_2$ leaching will increase if the groundwater contains constituents which react with Ca(OH)$_2$. Atkinson (8) notes that magnesium and carbonate are likely to have the greatest effect on Ca(OH)$_2$ losses. The effect of these species is modeled using Eq. (5), increasing the pore solution concentration of Ca(OH)$_2$ by the sum of the groundwater concentrations of magnesium and carbonate.

Loss of Ca(OH)$_2$ through diffusion is given by (9)

$$Q = C_p/1000 \cdot (4./3.14 \cdot D_c \cdot 3.15E7)^{0.5} \qquad \text{Eq. (6)}$$

where

Q = annual loss of Ca(OH)$_2$ (mole/m^2)

C_p = Ca(OH)$_2$ concentration in concrete pore solution (mole/l)

D_c = Ca(OH)$_2$ pore diffusion coefficient (m^2/s).

Groundwater leach rates and diffusive fluxes of Ca(OH)$_2$ are converted to total annual losses. The greater of these losses is used in subsequent calculations of reductions in pH and concrete strength.

The pH of the concrete is maintained at levels greater than approximately 12.5 by the presence of alkalis, NaOH and KOH. As these highly soluble species are lost the pH declines until reaching 12.5, at which point the pH of the concrete is controlled primarily by the Ca(OH)$_2$ content (8).

Modeling of pH changes in the concrete takes these different species into account. The pH is assumed to decline linearly from the initial pH to 12.5 in proportion to the loss of NaOH and KOH. Groundwater leaching of alkalis is modeled using Eq. (5), while diffusive losses are calculated using Eq. (6). The larger of these annual losses are used in pH determinations.

Changes in concrete pH following the removal of NaOH and KOH from the concrete is based on work by Greenberg and Chang (10). All Ca(OH)$_2$ losses are assumed to deplete calcium in the calcium-silicate-hydrate (C-S-H) system of the concrete. Changes in this ratio are used to estimate concrete pH.

The loss in concrete strength due to the loss of Ca(OH)$_2$ is calculated using the approach of Atkinson and Hearne (9). An equivalent depth of concrete from which all Ca(OH)$_2$ has been lost is calculated, and used to determine the fractional concrete strength, given by

$$C_{str} = 1. - X_{ca}/C_{thk}^{.625} \qquad \text{Eq. (7)}$$

where

C_{str} = fraction of original concrete strength

X_{ca} = equivalent depth of concrete from which all Ca(OH)$_2$ has been lost (m)

C_{thk} = thickness of structural member (m).

Sulfate Attack

Sulfate reacts with the aluminate phases of Portland cement to form ettringite and, at high external concentrations of sulfate, gypsum. These reactions yield a greater volume of solids in the concrete, a portion of which appears as bulk volume expansion in the concrete. The stress created in the concrete as a result of this expansion leads to mechanical rupture and loss of concrete by spallation.

Modeling of the spallation of concrete from surfaces of the disposal structure due to sulfate attack is based on the mechanistic model developed by Atkinson and Hearne (11). The magnitude of said attack is characterized in terms of the rate at which spallation occurs, and is given by

$$R = E\,B^2\,C_s\,C_E D_I/(ag(1-v)) \qquad \text{Eq. (8)}$$

where

R = degradation rate (m/s)

E = Young's modulus (kg/m s^2)

B = linear strain caused by one mole of sulfate reacted in 1 m^3 (m^3/mole)

C_s = groundwater sulfate concentration (mole/m^3)

C_E = concentration of reacted sulfate as ettringite (mole/m^3)

D_I = intrinsic diffusion coefficient (m^2/s)

a = roughness factor for fracture path

g = fracture surface energy of concrete (J/m^2)

v = Poisson's ratio.

STRUCTURAL ANALYSIS

Concrete has a high compressive strength and a very low tensile strength. In reinforced concrete, the steel provides tensile strength and can also resist compression forces. Reinforced concrete structures are subject to loads which develop the internal bending moment, shear and axial tension, or compression in the sections of structural elements. Cracking of the concrete may result due to those forces. The RAESTRICT code calculates the cracking moment, cracking shear, cracking tension, and normal axial compressive strength as a function of the time-varying concrete properties.

Flexural cracking is determined by comparing the maximum bending moments with the cracking moment

determined from the modulus of rupture. The cracking shear is determined from standard American Concrete Institute expressions. Different expressions are used depending on whether shear forces occur in the presence of flexural, tension or compressive forces. The cracking shear is then compared to the maximum shear force from the loads on each concrete member. Cracking from tension or compression is determined by comparing the forces with the tensile or compressive strength of the concrete.

The methodology allows consideration of a wide variety of disposal configurations including vaults and canisters. At the time cracks begin to develop, the code calculates the crack widths, spacings and crack depths for each type of cracking. It also tracks subsequent crack propagation with time.

The RAESTRICT code also calculates the ultimate strengths of reinforced concrete sections. Once the force at the section reaches the ultimate strength, the reinforced concrete section will fail or form a new joint which may cause redistribution of the forces in the structural analysis.

EXPOSURE PATHWAYS ANALYSIS

In order to project the health impacts from the disposal of LLW, it is necessary to determine rates of contaminant release from the disposal facility, modes and rates of transport of contaminants from the facility to locations accessible to humans, and rates of contaminant intake. The RAESTRICT methodology considers all of these aspects of pathways analysis, allowing a comprehensive analysis of radiological doses and risks.

The rates at which contaminants are leached from the waste and migrate from the facility will depend, in large part, on the amount of water percolating through the disposal units. Percolation rates for the intact disposal facility and the facility following failure of the engineered barriers may be specified by the user, or may be modeled as part of the simulation.

Modeling of groundwater percolation in the vicinity of the disposal facility is performed using the HELP computer code methodology (12). HELP was developed to facilitate rapid, economical estimates of surface runoff, evaporation, and subsurface drainage that may be expected to result from the operation of a wide variety of landfill designs. The program models the effects of a wide variety of hydrologic processes, including precipitation, surface storage, runoff, percolation, evapotranspiration, soil moisture storage, and lateral drainage using a quasi-two-dimensional approach.

Waste contaminants migrating to the aquifer may be accessed directly by humans, or indirectly if the aquifer outcrops to a surface stream. Exposures to radionuclides may also occur during disposal operations and through disruption of disposal units following closure. Doses and

risks from these potential exposure routes are modeled in RAESTRICT using the PATHRAE computer code methodology (13).

PATHRAE was developed by the Environmental Protection Agency for use in establishing standards for the disposal of LLW. The code models onsite and offsite pathways through which humans may come in contact with waste contaminants. Offsite pathways include the ground and surface water pathways, surface erosion, and atmospheric transport. The onsite pathways of concern arise principally from worker doses during operations and from post-closure site reclamation (intruder) activities.

Exposures to radionuclides may result from the ingestion of contaminants, inhalation of suspended contamination, and direct radiation. Modeling of the ingestion pathway considers the use of contaminated river or well water as a primary source of drinking water and the consumption of contaminated vegetables, grains, and animal products. Food crops may become contaminated through the irrigation of crops with contaminated water or through root uptake of radionuclides from the soil. Animals may assimilate radioactivity in drinking water and forage irrigated with contaminated water, resulting in contaminated milk and meat.

Radioactive contamination suspended during disposal operations may be inhaled by a worker at the disposal facility, or may be transported to an individual or population situated downwind from the site. Individuals may also be exposed to airborne contaminants as a result of a fire during disposal operations. Following closure, individuals intruding onto the site may inhale waste contaminants exposed during excavatory activities.

Modeling of external exposures considers doses received from undisturbed buried waste and doses from surface contamination. Contaminants may be brought to the surface during site intruder activities, such as site excavation during home construction. Radionuclides assimilated by plants growing over the site may be deposited on the surface following death and decay of the plant matter.

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