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U.S. Nuclear Regulatory Commission
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Subject: Transmittal of Draft Conceptual and Mathematical Models of Potential Fast and Bypassing Pathways Associated with *In-Situ* Tank Closures (Intermediate Milestone 06004.01.007.210)

Dear Mr. Whited:

This letter transmits the subject deliverable which is identified in the Task 7 portion of the Operations Plan for Technical Assistance in Evaluating Non-High-Level Waste Determinations for the U.S. Department of Energy in South Carolina and Idaho.

This report describes conceptual and mathematical models that can be used to evaluate the potential influence of fast pathways and bypassing pathways on engineered barrier performance for radioactive waste reprocessing tanks that are closed in place. The report identifies and classifies various potential fast and bypassing pathways and describes the movement of fluids through such pathways. It also describes mass transport through microcracks, macrocracks, and conduits and presents observations about potential resulting effects on radionuclide releases to the environment.

If you have any questions about this report, please do not hesitate to contact me (210.522.2139) or Dr. Gary Walter (210.522.3805).

Sincerely,

David Turner, Ph.D.
Assistant Director
Non-Repository Programs

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DRAFT
CONCEPTUAL AND MATHEMATICAL MODELS OF POTENTIAL FAST AND BYPASSING PATHWAYS ASSOCIATED WITH *IN-SITU* TANK CLOSURES

Prepared for

U.S. Nuclear Regulatory Commission
Contract NRC–02–02–012

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QUALITY OF DATA, ANALYSES, AND CODE DEVELOPMENT

DATA: No original data were generated from the analyses presented in this report. Sources for data cited in this report should be consulted for determining the level of quality of those data.

ANALYSES AND CODES: No codes were used in the analyses contained in this report.

1 INTRODUCTION

This report describes conceptual and mathematical models that can be used to evaluate the potential influence of fast pathways and bypassing pathways in engineered barriers on radionuclide releases from radioactive waste processing tanks that are closed in place. For the purposes of this report, fast pathways refer to features that allow fluids (i.e., water or gas) and associated radionuclides and chemical constituents to flow at rates significantly faster than those estimated to occur through the bulk engineered barrier. Bypassing pathways refer to features for fluid or gas flow that may (i) allow dissolved radionuclides to bypass chemical barriers or (ii) allow meteoric water or environmental gases to bypass engineered barriers and either contact the waste form or accelerate degradation of engineered barriers. Bypassing pathways could also divert environmental fluids, particularly water, around the waste form.

2 BACKGROUND ON IN-PLACE TANK CLOSURES

The U.S. Department of Energy (DOE) has proposed *in-situ* disposal of residual radioactive materials contained in liquid storage tanks formerly used to process radioactive materials at the Savannah River Site (SRS)[1] and the Idaho National Laboratory pursuant to Section 3116 of the Ronald W. Reagan National Defense Authorization Act for Fiscal Year 2005. An important component of the disposal plan at these sites is the creation of an engineered barrier to radionuclide migration (in addition to preexisting barriers that were part of the original tank design) by filling the tanks with a cement-based grout and sealing any external tank penetrations (such as fill lines). Tanks also exist at DOE's Hanford, Washington, site that may be closed in a similar manner. Once filled with grout, an engineered cover will be placed over the tanks to act as an additional engineered barrier to limit infiltration of meteoric water and restrict human and biological intrusion into contaminated materials. Although this report is not specific to any tank closure plan currently proposed by DOE, specific tank designs and closure plans presented by DOE will be considered to develop classifications and conceptual models of potential fast pathways and bypassing pathways that may affect the performance of *in-situ* tank closures in general.

3 IDENTIFICATION AND CLASSIFICATION OF POTENTIAL FAST AND BYPASSING PATHWAYS

Figures 1 and 2 show typical as-built tank constructions at the Idaho and Savannah River sites, respectively. Common characteristics of the tanks at each site are a metal tank with penetrations through the domed roof for fluid transfer piping and equipment access (such as the center riser in Figure 2). The interior of the tanks contains various metal pipes, cooling coils, and metal support structures. The metal walls of the tanks are constructed with either carbon steel or stainless steel. A significant difference between the tanks shown in Figures 1 and 2 is that the tank at the Idaho site is a freestanding metal structure housed within a reinforced concrete vault, whereas the SRS

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[1]Savannah River Site is referenced frequently throughout this document. The abbreviation SRS will be used.

tank has a metal liner encased in reinforced concrete for structural support. Another difference between the two tanks is that the Idaho tank sits on a base of bedding sand connected to an overfill collection sump, whereas the Savannah River tank sits on a concrete base with drainage channels connected to a collection sump.

The generic plan for in-place closure of these tanks is to fill the tanks with a cementitious grout having a composition suitable for immobilizing or encapsulating residual radioactive materials. At SRS, the tanks might be filled with two types of grout: a reducing grout to chemically immobilize the waste and a strong grout to act as an intruder barrier, as illustrated in Figure 3. At the Idaho National Laboratory site, both the tank and the vault may be filled with grout, as illustrated conceptually in Figure 4. Although the grout placed in the tanks may mix with the residual waste, complete mixing may not occur, leaving some of the waste as discrete layers or lenses "… trapped between grout layers, and some encapsulated between the tank structure and the grout." in DOE (2006).

Although the tank construction and specific in-place closure procedure may vary from site to site, Figures 1–4 adequately identify the types of features that could act as fast or bypassing pathways. These features can be generally categorized as cracks and conduits. Cracks are two-dimensional features that form due to mechanical failure of a material, such as grout or concrete structures. As used here, conduits will refer to discrete fluid pathways that may be present or develop along specific engineered features (e.g., annular spaces between grout and a corroded pipe wall, corroded rebar in a concrete vault wall). Specific pathways within each of these categories are listed in Table 1 and discussed in detail in the following sections.

3.1 Cracks in Grout and Concrete Structures

For the purposes of this report, cracks in grout used to fill tanks, associated piping and risers, and concrete structures associated with the tanks (such as external concrete support, footings, and vault walls) can be classified as microcracks and macrocracks. Microcracks are microscopic cracks disseminated through a concrete matrix that probably exist in all concrete structures due to plastic shrinkage, heat of hydration drying shrinkage, and mechanical stress (Young, 1988; Hearn, 1999). Macrocracks are discrete fractures in a concrete material that are often visible to the naked eye.

3.1.1 Microcracks

Various types of microcracks are illustrated in Figure 5. These microcracks have been described as bond cracks and mortar cracks. As illustrated in Figure 5(A), bond cracks are due to separation of the mortar from the aggregate during drying. Thus, simple bond cracks are not likely to form interconnected pathways through the grout. Mortar cracks form in the mortar between the aggregate and may form interconnected networks through the concrete matrix as well as interconnecting bond cracks [Figure 5(B)]. Hearn (1999) also identified drying-shrinkage cracks, illustrated in Figure 5(C), that can be described as a combination of bond and mortar cracks.

Table 1. Categories and Features That May Act as Fast or Bypassing Pathways Associated With In-Place Tank Closures	
Category	**Feature**
Cracks	Microcracks in grout monolith
	Macrocracks in grout monolith
	Microcracks in external grout
	Macrocracks in external grout
	Cracked and corroded seams/joints in metal tank walls
	Macrocracks in concrete tank walls
Conduits	Annular spaces between grout and pipes, coiling coils
	Annular spaces due to metal pipe corrosion
	Discontinuities in grout pours
	Corroded rebar in concrete tank walls and footings
	Annular spaces between grout and external pipes and risers
	Liquid collection systems external to tanks

Due to their ubiquitous nature in concrete, microcracks affect the bulk permeability and diffusion properties of apparently intact concrete materials. For this reason, the influence of microcracks on fluid permeation and chemical diffusion is likely to be included with uncracked mortar properties in laboratory measurements of concrete permeability and diffusion coefficients (e.g., Young, 1988; Boulfiza, et al., 2003; Hearn, et al., 1994). Viewed on a macroscopic scale, microcracks would not be described as fast or bypassing pathways in calculations of bulk water, gas, or chemical transport through a grout monolith.[2] Combining the influence of microcracks with the matrix may

[2]As used in this report, monolith refers to an intact, continuous concrete structure, such as a continuous pour within a tank or a continuous section of grout within a pipe.

not be appropriate, however, for modeling the release of radionuclides from waste that is actually incorporated into the grout for the following reasons:

- Transport of dissolved radionuclides from the uncracked mortar to fluids in the microcracks may be diffusion limited, and radionuclide concentrations in fluids flowing through the microcracks would be ovestimated.

- Radionuclides in microcracks may not be in chemical equilibrium with reactants in the mortar or aggregate so that radionuclides traveling through the microcracks would not be immobilized.

- Transport of environmental fluids and reactants (water, dissolved salts, and atmospheric gases) from the microcracks to the uncracked mortar or aggregate may be diffusion limited so that degradation of the mortar or aggregate would be overestimated.

3.1.2 Macrocracks in Grout and Other Concrete Structures

Macrocracks in grout and concrete generally form after the cementitious material has hardened and are usually the result of mechanical stress (Wang, et al., 1996) or the mechanical stress in large concrete monoliths (e.g., Copen, 1974; Zhang, et al., 2003). Macrocracks can also form by expansion of preexisting microcracks due to mechanical stress, water freezing, and salt precipitation and hydration. By their nature, macrocracks are more widely spaced and have much larger apertures than microcracks. The spacing and aperture for macrocracks can vary significantly depending on the composition of the grout or concrete, size and shape of the structure, loading, and environmental exposure.

Laboratory tests on concrete core samples with mechanically induced macrocracks indicate that single or simple branching cracks can increase the average water permeability of the test specimen by many orders of magnitude. For example, tests results reported by Wang, et al. (1996) indicate hydraulic conductivities on the order of 10^{-16} meters per second (m/s) [3×10^{-16} ft/s] for concrete samples without macrocracks. Freedman (1974) reported values of hydraulic conductivity based on aged 46 × 46-cm [18 × 18-in] concrete specimens from various concrete dams ranging from approximately 4×10^{-13} m/s [1×10^{-12} ft/s] to 3×10^{-11} m/s [1×10^{-10} ft/s]. Test on 10-cm [4-in]-diameter cores with macrocracks with apertures of approximately 0.05 cm [0.02 in] had hydraulic conductivities as high as 10^{-4} m/s [3×10^{-2} ft/s] (Wang, et al., 1996). Aldea, et al. (1999) reported similar findings. For comparison, the hydraulic conductivity of the concrete without macrocracks is equivalent to an intrinsic permeability of approximately 10^{-16} m^2 [10^{-15} ft^2], which is about two orders of magnitude higher than the lower range of matrix permeabilities estimated for welded tuffs at Yucca Mountain, Nevada (Bechtel SAIC Company, LLC, 2004). The hydraulic conductivities reported for the cracked concrete by Wang, et al. (1996) and Aldea, et al. (1999) represent the hydraulic conductivity of the crack weighted by the surface area of the concrete core and are only meaningful if converted to the hydraulic conductivity of the crack itself. Assuming a single crack in the concrete cores with an aperture of 0.05 cm [0.02 in] and a 10-cm [4-in]-diameter core, the hydraulic conductivity of the macrocrack would be approximately 2×10^{-2} m/s [7×10^{-2} ft/s].

Due to the much higher permeability of macrocracks than the grout or concrete matrix,[3] most fluid flow through the grout monolith and external concrete structures will occur through macrocracks, if present, rather than through the grout or concrete matrix. Macrocracks in the grout monolith could act as fast pathways for environmental fluids (infiltrating water and air) to contact the grout matrix and any radionuclides incorporated in the matrix. If radionuclides are present as segregated layers, lenses, or pockets in the grout monolith, the macrocracks could act as fast pathways for radionuclide migration through the grout monolith and as bypassing pathways limiting contact of the radionuclides with constituents in the grout matrix intended to reduce the mobility of the radionuclides. Alternatively, macrocracks in the grout monolith where radionuclides are incorporated in the grout matrix will act as bypassing pathways, limiting the contact of percolating water with the waste.

3.2 Conduits

Conduits are distinguished from cracks in this report because they may occur as discrete pathways that are initially present or may develop along specific engineered features, rather than as disseminated pathways throughout a grout or concrete monolith, as is the case for cracks. Various types of potential conduits are listed in Table 1 and are illustrated in Figure 6. The conduits illustrated in Figure 6 can act as pathways for fast movement of environmental fluids (water and air) into the waste form (such as the corroded pipe pathway in Figure 7), as fast pathways from the waste form to the environment (such as the corroded tank pathway out of waste in Figure 7), or as bypassing pathways that divert environmental fluids away from the waste form (e.g., the tank annulus pathway past waste in Figure 7).

Conduits formed around preexisting engineering structures (e.g., pipes, tank walls) can be described as annular spaces. Figure 8 illustrates two types of annular spaces that could exist around a pipe sealed with grout. In Figure 8(A), the annulus is open (i.e., a gap exists between the pipe and the grout). Such a gap could result from shrinkage of the grout from the pipe wall during curing or corrosive degradation of the pipe. Figure 8(B) illustrates the case where the annulus is filled with a porous material, such as the oxidized residue of a metal pipe. In either case, the processes for fluid and mass exchange between the conduit and the matrix are fundamentally the same as for a crack in the grout (Figure 9). Liquid exchange between the conduit and the matrix is controlled by the capillary pressure gradient between the matrix and wetted surface of the annulus (Walton and Seitz, 1991; Kapoor, 1994; Tokunaga and Wan, 1997; Tokunaga, et al., 2000). Mass transport between the fluid in the conduit and matrix is controlled by diffusion in the matrix.

As indicated in Figure 6, other types of conduits could exist that would not be described as annular spaces, such as corroded reinforcing bars (rebars) in concrete vault walls and footings and liquid collection systems beneath the tanks and within the vaults. These types of conduits would generally be exterior to the waste form in the tank, although the sand pad at one tank at the Idaho site is known to be contaminated with

[3]The grout or concrete matrix refers to the grout or concrete mass between macrocracks that consists of mortar and aggregate and that may contain microcracks.

radioactive materials, and would potentially affect radionuclide releases either by acting as fast pathways from the waste to the environment or fast pathways for environmental fluids into the waste form. In some cases, however, conduits could act as bypassing pathways that divert percolating water around the waste form, as illustrated in Figure 10.

Finally, interfaces between grout pour layers or lenses of segregated residue waste could also act as conduits, as illustrated in Figure 11. Liquid flow and mass transport processes in such a conduit would be similar to those for macrocracks. Whether or not such layering or waste segregation is present in the grout monolith will depend on the properties and manner of grout emplacement.

4 MOVEMENT OF FLUIDS THROUGH POTENTIAL FAST AND BYPASSING PATHWAYS

Cracks and conduits are likely to act as the primary pathways by which environmental fluids, such as infiltrating water and atmospheric gases, can contact the waste form. These environmental fluids can affect radionuclide releases by acting as a carrier for radionuclides and by conveying chemical reactants that (i) physically degrade the grout or engineered structures or (ii) change the chemical properties of the grout and chemical environment of the waste form thus increasing radionuclide mobility. This section discusses the physical structure and properties that determine the mathematical description of fluid flow through the pathways described in Section 3 and the way such flow would be modeled. The mathematical description of mass transport processes is discussed in Section 5.

4.1 Fluid Flow in Cracks

Although macrocracks, by definition, have larger apertures than microcracks, the same fluid dynamic principles are usually applied in calculating fluid flow through both macrocracks and microcracks. Assuming that the macrocrack is fully saturated, the velocity distribution within the macrocrack will be the same as those illustrated for microcracks in Figure 12.

As discussed in Section 3.1, microcracks in grout and concrete form during the curing process and are an intrinsic part of the grout or concrete matrix. As such, the effect of microcracks on water and gas flow through the monolith is generally incorporated into estimates of the bulk permeability of the matrix (e.g., Young, 1988; Boulfiza, et al., 2003; Hearn, et al., 1994), and bulk flow through the monolith is computed using Darcy's Law. Viewed in this way, microcracks would not be treated as fast or bypassing pathways. On the other hand, fluids in microcracks would not necessarily be in hydraulic or chemical equilibrium with the mortar or aggregate.

If the interaction of fluids in the microcracks with the mortar or aggregate is important to the mobility of radionuclides, then microcracks should be treated as pathways bypassing the mortar and aggregate, and flow through the microcracks would need to be modeled to evaluate performance. In the simplest case, flow through microcracks can be considered as capillary flow between parallel plates, as illustrated in Figure 12(A), where the fluid velocity has a parabolic distribution perpendicular to the crack walls. This simple case assumes that there is no fluid flow between the crack and the matrix, although chemical mass transfer can occur between the crack and the matrix. The effect of this simple type of mass transfer between the crack and matrix has been studied extensively in the chemical engineering literature (e.g., Bird, et al., 1960), and various solutions are available for estimating the chemical interaction between the fluid in the crack and the matrix. In reality, crack surfaces are rough, and the crack walls are not parallel [Figure 12(B)]. In this case, the velocity distribution in the crack needs to be computed numerically using the Navier-Stokes equation (e.g., Thompson, 1991) if the microscopic variations in velocity are important for evaluating mass transport. Mass transport in microcracks is discussed in Section 5.1.

Fluid flow though saturated macrocracks and microcracks with relatively small apertures is usually described by capillary flow equations in which the permeability of a crack is related to the cube of the fracture aperture (e.g., Bird, et al., 1960). The so-called cubic law relationship is derived from viscous flow equations for closely spaced parallel plates. In reality, crack surfaces are rough, and opposing crack surfaces are not strictly parallel, which complicates the relationship between crack permeability and crack aperture. Nevertheless, research has indicated that a permeability can be assigned to a crack that is proportional to the cube of the crack aperture (e.g., Walton and Seitz, 1991; Oron and Berkowitz, 1998) so that flow through the crack described by Darcy's Law

$$K_c = \alpha \frac{\rho g}{12\mu} b^3 \tag{1}$$

and

$$q_c = -K_c \frac{dH}{ds} \tag{2}$$

where

K_c	—	saturated hydraulic conductivity of the crack [L/t]
α	—	factor that accounts for the crack roughness [dimensionless]
ρ	—	fluid density [M/L^3]
g	—	gravity [L/t^2]
μ	—	fluid dynamic viscosity [M/L-t]
b	—	crack aperture [L]
q_c	—	average fluid velocity in the crack [L/t]
dH/ds	—	is the hydraulic gradient along the crack [L/L]

Using Darcy's Law to describe fluid flow in fully saturated cracks is a common approach when the detailed velocity distribution in the crack is not important to the analysis (e.g., Walton and Seitz, 1991; Boulfiza, et al., 2003; National Council on Radiation Protection and Measurements, 2005). The application of Darcy's Law to flow in cracks that are not fully saturated is more complex, however, because the effective hydraulic conductivity will vary with the degree of saturation, as it does in porous media. Boulfiza, et al. (2003) developed an analytical relationship between crack saturation and relative hydraulic conductivity assuming a lognormal distribution of crack apertures. Their analysis indicated that single macrocracks or widely spaced macrocracks would be more conductive than the concrete matrix until the crack saturation dropped below about 10 percent. At that point, the matrix would become more conductive. The hydraulic properties of partially saturated cracks can also be described using relationships between saturation, capillary pressure, and relative permeability that have been developed for partially saturated porous media. These relationships will be described in the later discussion of dual continuum models for fractured materials.

Fluid flow in large aperture cracks could also occur as film flow, as illustrated in Figure 8 for a large aperture conduit. Cook, et al. (2005) considered film flow through macroracks in a performance assessment for the saltstone vaults at SRS. Film flow will be discussed further in Section 4.3 with respect to flow in conduits.

Even though macrocracks may be more conductive than the matrix, in general, the volumetric flow of either water or gas through a grout or concrete monolith depends on the surface area and water saturation of the macrocracks and matrix. Because macrocracks constitute a very small fraction of the surface area and volume of a concrete structure [generally less than 1 percent (Walton, et al., 1990)], their contribution to flow through the structure will be less than is implied by the permeability ratio between the cracks and the matrix. In addition, the water content and saturation of the grout or concrete matrix will generally be higher than that of the macrocracks except when water is actively being applied to the monolith surface. This is because the macrocracks drain more rapidly than the matrix, and the matrix has a much higher water retention capacity due to its small pore size than the macrocracks. As a result, the relative proportions of flow through macrocracks and the matrix will vary with time, as will fluid exchange between the cracks and the matrix (Boulfiza, et al., 2003). These conclusions about flow through macrocracks may not apply to microcracks because microcracks have much smaller apertures and are more prevalent than macrocracks. Thus, the water saturation of microcracks will generally be high (i.e., macrocracks at a given bulk water content of the monolith).

The application of Darcy's Law to flow through cracks can be extended to multiple cracks disseminated through the monolith using either discrete fracture or dual continuum models. Both of these approaches have been used extensively to model fluid flow and mass transport, including that of radionuclides, through naturally fractured rocks (e.g., Andersson and Thunvik, 1986; Huyakorn, et al., 1983).

In discrete fracture models, flow and transport in individual fractures is explicitly represented in the model along with fluid exchange with the matrix, as illustrated in Figure 13. Flow in the fractures is usually computed using Darcy's Law, and adjustments are made for partially saturated conditions, if necessary. Examples of discrete fracture models that include matrix interactions are SOLFRAC (Bodin, et al., 2007), TOUGH2 using specially constructed discrete fracture grids (Ito and Seol, 2003), and PORFLOW (Analytical & Computational Research, Inc., 2004). SOLFRAC uses a random walk method to simulate flow and transport through a fracture system. Although TOUGH2 (Pruess, et al., 1999) is usually used to perform dual continuum simulations of fracture flow and transport, Ito and Seol (2003) developed a special mesh generator (FRACMESH) that generates discrete fracture grids for use with TOUGH2. PORFLOW is a single continuum flow and transport model based on the finite element method that can reportedly represent discrete two-dimensional features such as fractures (Analytical & Computational Research, Inc., 2004). Other discrete fracture simulation models have been reported in the technical literature that vary from simply simulating fluid flow to simulating mass transport with matrix interactions. These have generally been specialized codes developed for specific research objectives and will not be discussed here.

The concept behind dual continuum models is illustrated in Figure 14. In dual continuum models, individual fractures are not explicitly represented. Rather, the hydraulic and mass transport properties of the fracture network are averaged over the volume represented by a model grid node (or cell) so that fracture network is treated as a continuum. The actual physical properties of the individual fractures can be represented in the fracture continuum by assigning appropriate values of hydraulic conductivity based on Eq. (1) and by using statistical representations of the fracture network to deduce equivalent hydraulic conductivities. The effects of partial saturation of the fracture network are accounted for using constitutive relationships between fracture saturation, fracture capillary pressure, and relative permeability. Exchange of fluid between the fracture continuum and the matrix continuum is computed using a linear transfer function. For example, water flow between the fracture and the matrix is computed by

$$q_{f-m} = A_{f-m}(H_f - H_m) \qquad (3)$$

where

q_{f-m} — water flux between the crack and the matrix [L/t]

A_{f-m} — hydraulic conductance term [1/t]

H_f — hydraulic head in the crack [L]

H_m — hydraulic head in the matrix [L]

TOUGH2 and MULTIFLO (Painter, et al., 2001) are examples of numerical models that have been used extensively to perform dual continuum simulations of fractured rocks. Either of these models could be used to simulate flow through macrocracks in a partially saturated grout or concrete monolith.

The hydraulic properties of a partially saturated fracture continuum are generally described using semiempirical relationships between water saturation, capillary pressure, and hydraulic conductivity or permeability.

One popular approach to describing the partially saturated fracture hydraulic properties is to use relationships developed by van Genuchten (1980), which relate capillary pressure, matrix saturation, and hydraulic conductivity. The relationship between saturation and capillary pressure in the fracture continuum is

$$P_{cf} = \frac{1}{\alpha_p m}\left[S_{ef}^{1/2} - 1\right]^{1/m} \qquad (4)$$

and

$$m = 1 - 1/\lambda_f \tag{5}$$

where

P_{cf} — capillary pressure in the fracture continuum (taken as positive)

S_{ef} — effective saturation of the matrix continuum

α_p and λ_f — fitting parameters

The effective saturation is defined as

$$S_{ef} = \frac{S_f - S_{rf}}{1 - S_{rf}} \tag{6}$$

where

S_f — actual fracture continuum saturation

S_{rf} — irreducible fracture saturation

The effective permeability (k_{ef}) for water flow in the fracture continuum is then

$$k_{ef} = k_{rf} k_f \tag{7}$$

where k_f is the permeability of the fracture continuum and k_{rf} is the relative permeability, which is assumed to be a function of the matrix saturation. The water relative permeability of the fracture continuum is described using van Genuchten's (1980) model

$$k_{rf} = S_{ef}^{1/2}\left[1 - \left(1 - S_{ef}^{1/\lambda}\right)\right]^2 \tag{8}$$

The partially saturated hydraulic properties of the matrix continuum are described by similar relationships.

The hydraulic properties of the fracture continuum can also be described using the active fracture model of Liu, et al. (1998). The active fracture conceptual model was developed to represent phenomena inferred to occur in fractured rock where the number of fractures actually conducting water varies with the saturation of the fracture network. This concept is also applicable to networks of cracks, not all of which are conducting water at any particular time. According the active fracture model, the capillary pressure/saturation relationship is given by

$$P_{cf} = \frac{1}{\alpha_f}\left[S_{ef}^{(\gamma-1)/\lambda_f} - 1\right]^{1/m} \tag{9}$$

and the relative permeability/saturation relationship by

$$k_{rf} = S_{ef}^{(1+\gamma)/2}\left[1-\left(1-S_{ef}^{(1-\gamma)/\lambda_f}\right)^{\lambda_f}\right]^2 \tag{10}$$

where γ is the activity factor that varies from zero to 1. When γ is zero, the model devolves into the conventional fracture model. Increasing values of γ cause the relative permeability of the fracture continuum to be higher at a given saturation. The constitutive relationship given by Eqs. (6) and (7) are still based on the van Genuchten (1980) porous media models where k_f is the permeability of the fracture continuum and k_{rf} is the relative permeability, which is assumed to be a function of the fracture saturation. The relative permeability for the fracture–matrix conductance is then

$$k_{rf-m} = S_{ef}^{(1+\gamma)} k_{rf} \tag{11}$$

where $k_{rf\text{-}m}$ is the fracture–matrix relative permeability. Eqs. (4)–(11) can also be applied to flow through discrete macrocracks if it is assumed that the interior of the fracture behaves more or less like a two-dimensional porous medium. This assumption is likely to be valid for cracks with rough surfaces (e.g., Boulfiza, et al., 2003). It may not be appropriate for cracks with smooth surfaces in which an analytical approach to computing the rate of flow through the crack should be used.

In practice, the parameters required to apply Eqs. (3)–(11) to simulate flow through cracks using a dual continuum model need to be determined empirically because they cannot be determined from first principles. The literature review conducted for this report did not reveal any studies of crack properties in large grout monoliths that were representative of grout-filled tanks.

The advective flow of gases into and out of partially saturated cracked grout or concrete can be modeled using the same basic principles as those described above for water flow. For example, the relative gas permeability of the cracks and matrix is related to the relative water permeability at a given water saturation by (Bear, 1972)

$$k_{rg} = 1 - k_{rw} \tag{12}$$

where k_{rg} is the relative gas permeability and k_{rw} is the relative water permeability, such as computed using Eqs. (7) or (9). The gas permeability is then

$$k_g = k_{rg} k \tag{13}$$

where k is the intrinsic permeability of the crack or matrix [L^2].

Some analytical and numerical models developed for the flow of liquids through discrete fractures or fractured continua can be applied to the flow of gases with an appropriate substitution of fluid properties (density and viscosity). Other models developed specifically for water flow may incorporate assumptions that are not appropriate for simulating the flow of gases. TOUGH2 and MULTIFLO are examples of numerical models specifically developed to handle the flow of both water and gas through dual porosity materials. With respect to the movement of gases through concrete materials, most research has focused on diffusion. Gas phase diffusive transport through cracks and conduits will be discussed in Section 5.

4.2 Flow Through Conduits

Although fluid flow through conduits is not fundamentally different than flow through cracks, the discrete nature and geometry of many types of conduits could require the use of different mathematical models than those used to simulate flow through cracks. Flow in annular-type conduits in which the liquid completely bridges the annular space or flows through an annular space filled with porous material, as in Figure 8(B), would be similar to Darcian flow through a discrete crack. However, models derived for flow through planar cracks in a porous matrix would need to be modified to account for radial flow into the matrix to be applied to an annulus around a pipe.

In the case of an annular space of large aperture, liquid flow could occur as isolated drops or as films [Figure 8(A)]. Film flow and mass transfer has received considerable attention in the chemical engineering literature because of its importance to various chemical processing technologies. Bird, et al. (1960) provides the following simple expression for the laminar flow of a film on a smooth, inclined surface

$$q = \frac{\rho g \delta^3 \cos\theta}{3\mu} \tag{14}$$

where

q — liquid flux [L/t]

ρ — liquid density [M/L^3]

g — gravitational acceleration [L/t^2]

δ — film thickness [L]

θ — angle from the vertical

μ — fluid viscosity [M/L-t]

Equation (14) can be applied to curved annular spaces if the liquid does not completely bridge the space and there is no imbibition into the matrix. Equation (14) also applies to macrocracks with large apertures. Or and Tuller (2000) developed an approach for calculating film flow on a rough porous surface and for calculating an equivalent unsaturated hydraulic conductivity as a function of roughness and matric suction of the porous medians. Cook, et al. (2005) used the Or and Tuller (2000) approach to estimate the influence of macrocracks on fluid flow through the saltstone vaults at SRS. Kapoor (1994), Tokunaga and Wan (1997), and Tokunaga, et al. (2000) developed models for laminar film flow in fractures and rough surface in porous materials that can be used to estimate film flow rates through macrocracks and conduits with large apertures. Their solutions would require some modification to flow through annuli around pipes, but would be appropriate for flow through annuli between the metal wall of a tank and the grout matrix where the radius of curvature of the tank is large.

The mathematical models described above apply only to laminar flow of liquids. Conduits, as well as macrocracks, can act as pathways for movement of atmospheric and soil gas into a grout monolith and for movement of gas phase radionuclides out of the waste form. With respect to movement of atmospheric gases into concrete, most research has focused on carbon dioxide transport by diffusion and its effect on concrete carbonation (e.g., Odeh, et al., 2006; Meier, et al., 2007; Monlouis-Bonnaire, et al., 2004). Thunvik and Braesler (1990) modeled advective-diffusive flow of subsurface gases through fractured rock to investigate gas flow and hydrogen transport from submarine geologic radioactive waste repositories. Although no studies specifically related to advective transport of gas through conduits in concrete materials were identified in the technical literature, the physics of gas flow through pipes and other open spaces are well understood. The potential for gas and associated contaminants to migrate through conduits in concrete building slabs and foundations is also well known (e.g., New York State Department of Health, 2006). Simple models have been developed for estimating the flow of gas and contaminant vapor through cracks in building slabs (e.g., Johnson and Ettinger,1991); these models could estimate gas flow through discrete, planar conduits into a grout monolith or waste form. Similar approaches could be used to estimate gas flow through annular conduits, either from the soil surrounding a tank or vault or from the atmosphere. Although rarely studied, the effects of atmospheric barometric pressure changes on gas flow into and out of the monolith through conduits needs to be considered to evaluate the effect of atmospheric and soil gases on concrete degradation and the migration of gas phase radionuclides out of the waste form. An approach similar to that used by Rossabi and Falta (2002) could be used to estimate barometrically induced gas flow through conduits into a grout monolith.

5 MASS TRANSPORT PROCESSES RELATED TO FAST AND BYPASSING PATHWAYS

Chapter 4 focused on the physics and models of fluid flow (water and gas) through cracks and conduits in the grout monolith and associated concrete structures (e.g., vault walls). This chapter considers mass transport processes associated with these flows that may affect the release of radionuclides to the environment including processes affecting degradation of physical and chemical barriers. The mass transport processes important to evaluating the influence of cracks and conduits on radionuclide releases from in-place closure of waste tanks are

- Transfer of radionuclides between the grout matrix and fluids in the cracks and conduits

- Advective, diffusive, and dispersive transport of radionuclides through the cracks and conduits

- Advective, diffusive, and dispersive transport of environmental reactants through the cracks and conduits

- Sorption of radionuclides within the cracks and conduits

- Decay of radionuclides and ingrowth within the cracks and conduits

5.1 Mass Transport in Microcracks

As discussed in Section 3.1.1, microcracks are ubiquitous in cementitious structures, and their influence on mass transport is often treated as part of the bulk properties of the concrete matrix (e.g., Young, 1988; Boulfiza, et al., 2003; Hearn, et al., 1994). Lumping the effects of microcracks with the bulk properties of the matrix may not be appropriate if the fluids in the microcracks are not in chemical equilibrium with the grout mortar and aggregate. If this is the case, the microcracks can be viewed as bypassing pathways even if they do not behave as fast pathways. Figure 15 illustrates such a scenario where the fluid in a microcrack is not in chemical equilibrium with reductants from blast furnace slag added to reduce the mobility of Tc-99. This scenario is similar to that considered by Esh, et al. (2006).

The fluid flow and mass transport processes in a grout or concrete monolith containing microcracks are similar to those that have been studied in multiporosity geologic materials, such as soil and rock containing macro- and microporosity. Most analytical and numerical models of dual porosity media use linear functions to represent fluid flow and mass transfer between a high permeability medium, such as a crack, and a low permeability medium, such as a grout matrix as illustrated in Figure 14. For example, water flow at each location is represented by Eq. (2) and diffusive mass transfer by equations of the form

$$\dot{m} = D_{f-m}(C_f - C_m) \tag{15}$$

where

$\dot{m}$ — mass flux $[M/L^2\text{-}t]$

D_{f-m} — fracture–matrix diffusion conductance term [L/t]

C_f — concentration in the crack $[M/L^3]$

C_m — concentration in the matrix $[M/L^3]$

Such formulations do not consider pressure or concentration gradients in the matrix; rather, these values in the matrix at a given location are assumed to be uniform or an average within the matrix. This approach may not be appropriate for simulating transient fluid flow over short time periods, or, more importantly, sharp concentration gradients due to sorption or chemical reactions within the matrix.

Transport through dual porosity materials has been studied extensively, and both analytical and numerical models have been developed for simulating the fluid flow and mass transport processes in such materials on a macroscopic scale (e.g., van Genuchten and Wierenga, 1976; Rasmussen and Neretnieks, 1981; Narasimhan, 1982; Corapcioglu and Wang, 1999; Huang and Hu, 2001). Multiporosity fluid flow and mass transport calculations are also included in numerical models such as HYDRUS (Simunek, et al., 1999), MULTIFLO (Painter, et al., 2001), and the TOUGH family of codes (Pruess, et al., 1999). For example, HYDRUS can simulate aqueous advective dispersion of a six-member decay chain with nonlinear sorption through a dual porosity medium in which water is treated as immobile in the matrix.

The TOUGH2 family of codes includes capabilities to simulate advective dispersion with sorption and chemical reaction in multiporosity media. Moridis, et al. (1999) published a module for TOUGH2 that simulates transport with chain decay of an n – 1 daughter product sequence. However, TOUGH2 is a complex set of process modules, and not all processes of interest to radionuclide releases from cementitious materials can be simulated simultaneously. For example, radioactive chain decay cannot be simulated with multicomponent chemical reactions. An interesting capability of TOUGH2 is its ability to simulate MINC[4] (Pruess, et al., 1999). The concept behind MINC is illustrated in Figure 16. MINC allows the matrix to be represented as multiple, overlapping continua in which fluid pressures and concentration gradients can be simulated. This approach could be useful for simulating chemical reaction zones in a cementitious matrix surrounding microcracks or dispersed macrocracks. The MINC approach might also be very well suited for simulations involving chemical reactions between the mortar matrix and aggregate within the mortar.

MULTIFLO (Painter, et al., 2001) can simulate simultaneous flow of water and gas with chemical reactions in dual porosity media and could be used to simulate flow and transport through a grout monolith with microcracks or dispersed macrocracks. MULTIFLO does not, however, have the capability to simulate radioactive chain decay or MINC grid structures. Painter, et al. (2006) recently developed a fluid particle-based methodology for simulating radionuclide transport through discrete fracture networks in porous media. Their methodology, termed Particle on Random Streamline Segment (PORSS), includes the effects of matrix diffusion and chain decay. PORSS is potentially applicable for simulating radionuclide transport through networks of micro and macrocracks, as well as through conduits.

5.2 Mass Transport in Macrocracks

Mass transport through a cementitious monolith containing macrocracks can be modeled using either the dual porosity approach or a discrete fracture pathway approach. The decision as to which approach is most appropriate depends on how widely the macrocracks are dispersed through the monolith and the purpose of the calculation. The dual porosity approach would be appropriate if the macrocracks are relatively closely spaced and well connected and steep concentration gradients are not expected in the matrix. Discrete fracture models would be better suited for simulation of flow through widely spaced cracks and transport with steep concentration gradients in the matrix. Because the dual porosity models discussed in Section 5.1 for microcracks are equally applicable to macrocracks, the dual porosity approach will not be discussed further.

Nearly all of the fluid flow and mass transport processes associated with the influence of macrocracks on radionuclide release and migration from in-place tank closure have

[4]Multiple Interacting Continua is referenced frequently throughout this document. The abbreviation MINC will be used.

been investigated in the context of matrix diffusion in porous geologic materials containing discrete fractures (e.g., Grisak and Pickens, 1980; Sudicky and Frind, 1982; Zhang and Woodbury, 2000; Sun and Buscheck, 2003; Neretnieks, 2006; Houseworth, 2006). Walton and Seitz (1991) and Walton (1992) applied some of these concepts to evaluating the performance of cracked concrete barriers for radioactive waste isolation. Mainguy, et al. (2001) applied similar matrix diffusion concepts to analyze concrete degradation due to leaching of calcium by water flowing through macrocracks. Roden and Scheibe (2005) used a one-dimensional modeling approach applicable to a discrete crack to study reductive immobilization of uranium(VI) in fractured soil.

Except for the work of Mainguy, et al. (2001) and Roden and Sheibe (2005), matrix diffusion mass transport studies have focused on attenuation of contaminant concentrations, particularly radionuclides, due to diffusion from the fracture into the rock or soil matrix. Some of these models consider decay of only a single constituent (e.g., Grisak and Pickens, 1980) while others consider chain decay (e.g., Sun and Buscheck, 2003; Neretnieks, 2006). Although the analytical models developed from such studies are applicable to the transport of radionuclides through microcracks in initially uncontaminated grout or concrete, they do not directly represent leaching of radionuclides from grout or concrete. Some of the simpler analytical models can be applied to leaching of contaminated grout or concrete with a relatively simple change in variables. For example, an analytic solution for a single constituent that is linear in terms of the concentration in the fluid flowing into the fracture, C_o, and the initial concentration in the matrix, C_1, is applicable to either the case where the fluid entering the fracture is contaminated, $C_o > 0$, and the matrix is uncontaminated, $C_1 = 0$, or vice versa. The more complex models, such as those that include chain decay (e.g., Sun and Buscheck, 2003; Neretnieks, 2006), may not be easily adapted to the leaching problem, although the mathematical methods on which these models are based can be applied to leaching.

5.3 Mass Transport in Conduits

Mass transport in conduits involves the same fundamental processes as mass transport through macrocracks. Analytical and numerical models developed for flow and transport through discrete cracks can also be used to model flow and transport through planar conduits and, to a good approximation, flow and transport through annular spaces with large radii of curvature, such as along tank walls. However, models derived for mass transport through planar cracks in a porous matrix would need to be modified to account for radial flow into the matrix to be applied to an annulus around a pipe. The difference in transport geometry between a planar crack and annulus around a pipe is illustrated in Figure 17.

Numerical models developed for fractured, dual porosity media (e.g., TOUGH2 and MULTIFLO) can also be used to simulate transport processes between conduits and the matrix if the fluid flow in the conduit can be assumed to be laminar or steady. If flow through cracks in the matrix surrounding a conduit is not important, single continuum numerical flow and transport models can also be used to simulate conduits. For example, Kaplan, et al. (2005) used PORFLOW (Analytical & Computational Research, Inc., 2004) to simulate water flow and oxygen transport through large aperture, symmetric cracks in a tank grout monolith and oxygen transport from the cracks to the grout. Because PORFLOW assumes a single, porous continuum, the cracks were treated as discrete elements and were assumed to be filled with a porous material so that Darcy's Law applied, even though the cracks were assumed to have apertures ranging from 1.5 to 3 cm [0.6 to 1.2 in].

6 SUMMARY AND RECOMMENDATIONS

As discussed in Section 3, microcracks, macrocracks, and conduits can

- Act as pathways for relatively rapid movement of environmental fluids (water, atmospheric gases, and soil gases) into the waste form and surrounding cementitious materials (grout and concrete structures)

- Act as fast pathways for radionuclide transport from these materials to the environment

- Act as bypassing pathways that either divert environmental fluids away from the waste form or create a condition of chemical disequilibrium with the waste form

Although the effect of these features on radionuclide releases to the environment will be site specific, some general observations about their potential effects can be made from basic principles of mass transport in fractured and porous media and on studies of cementitious materials in general.

Because microcracks form primarily during the curing process of a concrete monolith, they can be expected to be present in the grout used to fill a former waste tank or annular spaces around a tank. Most engineering studies of concrete materials, including studies of intrusion of water and chemicals that can degrade the concrete, have included the effect of microcracks in the properties of the matrix. These studies have shown that the permeability of the matrix is very low (saturated hydraulic conductivity less than 10^{-10} m/s [10^{-9} ft/s] in intake concrete structures. Thus, microcracks are not likely to act as fast pathways for bulk fluid flow through or from a grout or concrete matrix. Likewise, studies have shown that mass transport through a concrete matrix is diffusion dominated in the absence of macrocracks. Microcracks could, however, contain fluids and radionuclides that are not in chemical equilibrium with the mortar or aggregate. Chemical reactions between fluids and radionuclides in the microcracks are important for immobilization of radionuclides, such as in the maintenance of reducing conditions to limit the mobility of Tc-99. The extent to which such chemical equilibrium can be assumed depends on residence time and can be estimated using existing analytical and numerical models for diffusive and reactive transport through fractured, porous media.

Macrocracks that develop in a concrete monolith after it hardens are the features most likely to act as fast pathways between the waste form and the environment, although they could also act as bypassing pathways that limit contact of environmental fluids with the waste form and other engineered barriers. Fluid flow and mass transport through macrocracks in concrete structures is closely analogous to flow and transport through fractured geologic materials. Many of the same modeling methods used to simulate contaminant transport through fractured geologic materials can and have been used to simulate the effects of environmental chemicals (chloride and carbon dioxide) on the degradation of concrete structures. With the exception of relatively simple discrete fracture diffusion models (e.g., Walton, 1992), these modeling approaches have not been applied to study radionuclide releases from in-place tank closures or the effect of environmental fluids on long-term grout integrity. In addition, fluids conveying radionuclides in macrocracks may not be in chemical equilibrium with the chemical environment of the matrix and aggregate, and this disequilibrium may affect radionuclide mobility in the macrocrack.

Fluid flow and mass transport through a grout monolith or concrete structure containing macrocracks can be modeled using either dual porosity or discrete fracture models. The application of these modeling approaches to a specific in-place tank closure depends on the purpose of the study and the nature of the macrocracks. For example, many processes affecting radionuclide releases from a grout monolith with many well-connected macrocracks per unit volume might be adequately modeled using analytical or numerical dual porosity models. In the absence of a well-connected crack network, discrete fracture models would be necessary. Despite extensive research over the past 30 years on mass transport through fractured, porous geologic media, no single model or modeling approach was identified that could simulate all processes of interest in evaluating potential radionuclide releases from a fracture grout monolith. Existing models or slightly modified models would be capable of simulating many of the important processes relevant to the effect of microcracks on radionuclide release from in-place tank closures. The results of these process-level models could then be abstracted and integrated into a system-level modeling platform such as GoldSim (Goldsim Technology Group, 2004) (e.g., Esh, et al., 2006).

Conduits were distinguished from cracks as a separate type of pathway because they represent discrete fluid pathways that may be present or develop along specific engineered features with known locations, such as annular spaces between grout and a corroded pipe wall or corroded rebar in a concrete vault wall. Fluid flow and mass transport through conduits is fundamentally similar to that through discrete macrocracks, and many of the same modeling approaches would be applicable to conduit flow and transport. The flow geometry around small conduits, such as annular spaces around pipes, is different than that for cracks, which are roughly planar features, and this difference in geometry needs to be considered in model selection.

Although models and modeling approaches exist for simulating most of the flow and transport processes associated with fast and bypassing pathways, these models are of limited value without estimates of the nature, extent, and properties of the pathways. Information on cracks in concrete structures is available in the technical literature. However, most studies have focused on reinforced concrete structures of interest in civil engineering projects. These studies may be of limited value in estimating the nature, extent, and properties of cracks in a grout monolith inside a former process tank. Even less information may be available to estimate the properties of conduits. With respect to cracks in the monolith, mechanical models could be used to estimate the extent of cracking for performance assessment (e.g., DeBorst, et al., 2004) and chemical weathering models (e.g., Boddy, et al., 1999; Mainguy, et al., 2001; Yoon, et al., 2000; Boulfiza, et al., 2003) could be used to extrapolate how these cracks might evolve and affect the integrity of the grout.

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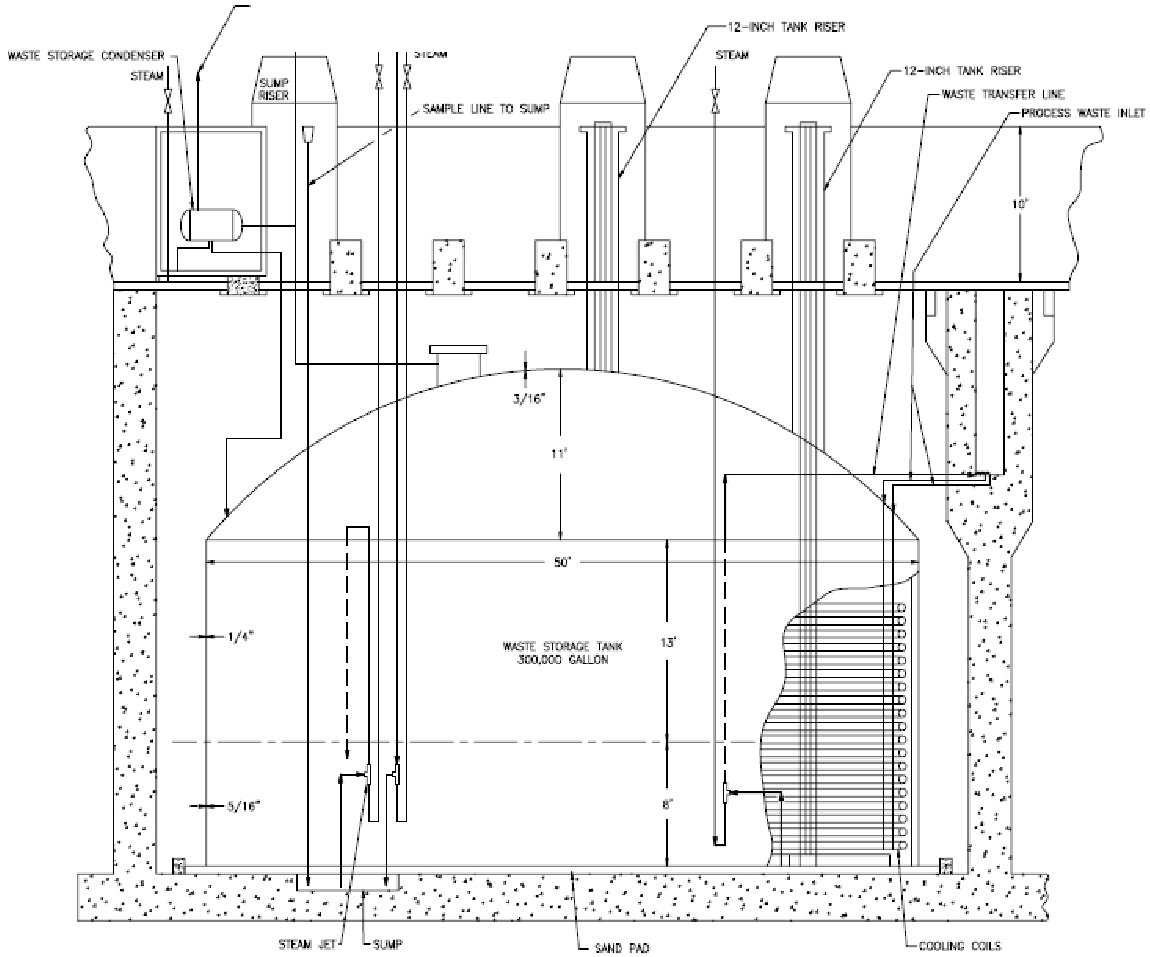


Figure 1. Cross Section of a Typical Tank With Cooling Coils at Idaho National Laboratory (From DOE, 2006)

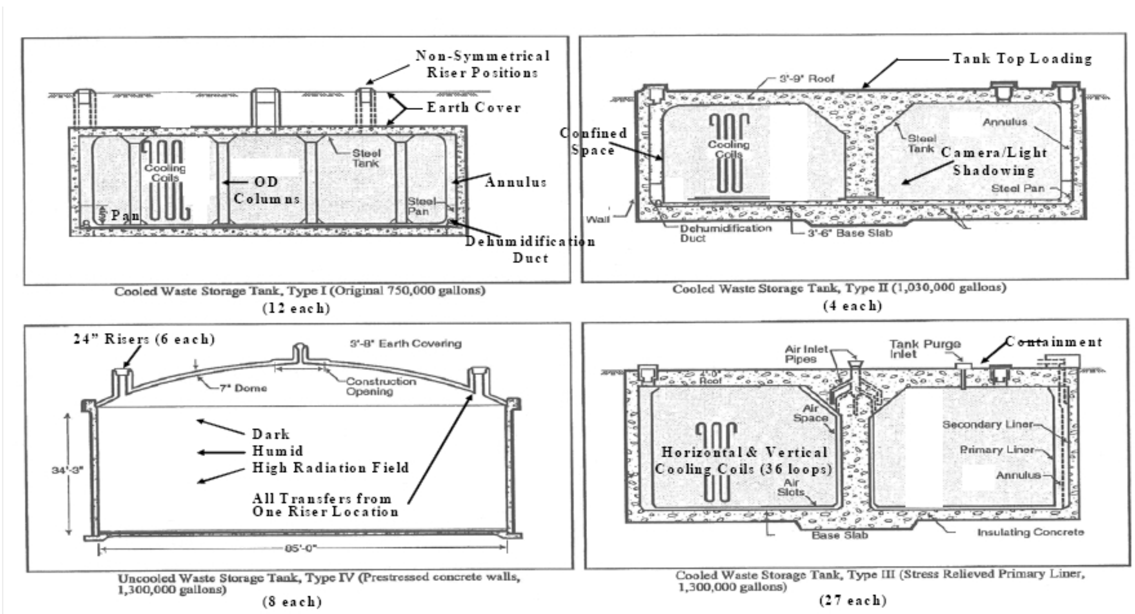


Figure 2. Diagrams of Tank Types at the Savannah River Site (From DOE, 2005)

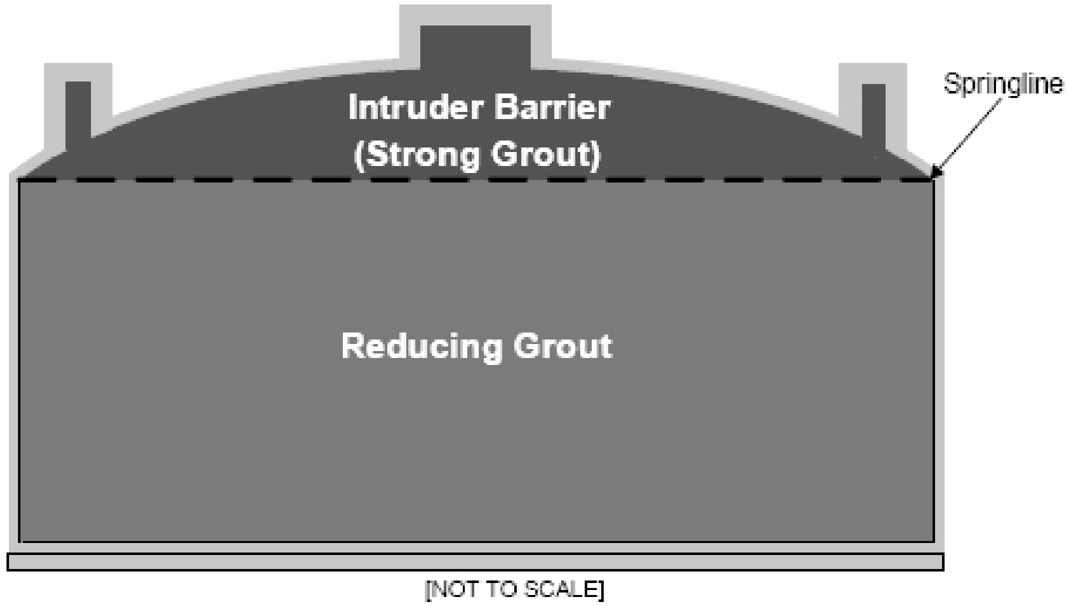
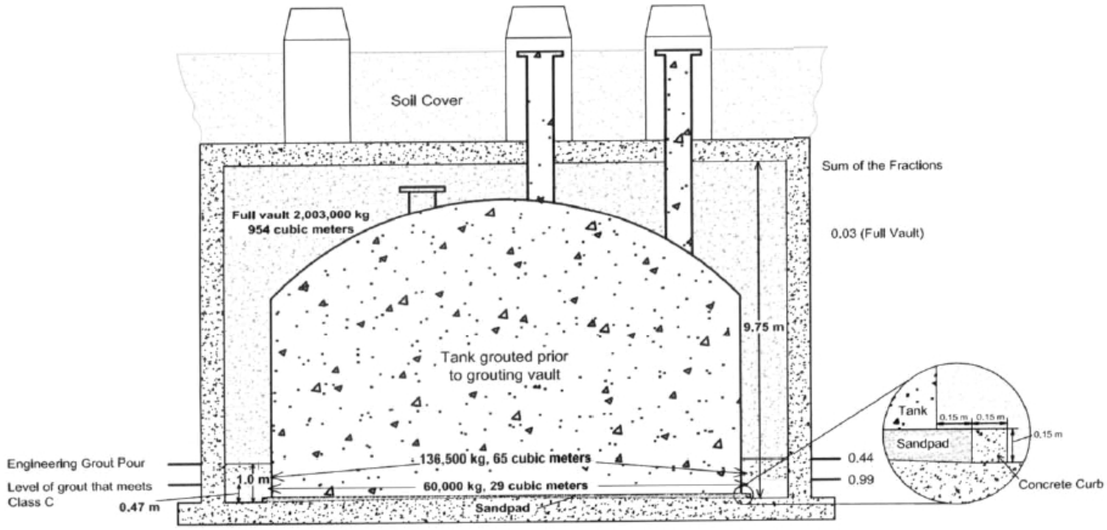


Figure 3. Conceptual Illustration of Grouted Tank at Savannah River Site (From DOE, 2005)



Cross-sectional view of typical tank and vault. Calculations are based on dimensions of the smallest tank vault and the highest detected amount of radioactivity (from Tank WM-182).

Figure 4. Illustration of Grouted Tank and Vault at Idaho Nuclear Technology and Engineering Center Tank Farm (From Lockie, et al., 2005)

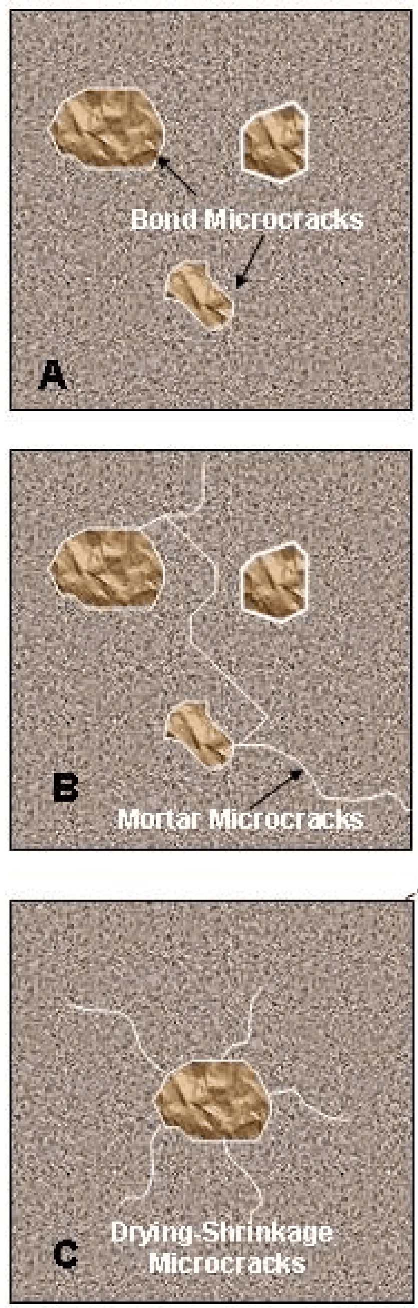


Figure 5. Types of Cracks in a Concrete Matrix. A: Illustration of Bond Microcrack, B: Illustration of Mortar Microcracks, and C: Illustration of Drying-Shrinkage Microcracks (Hearn, 1999)

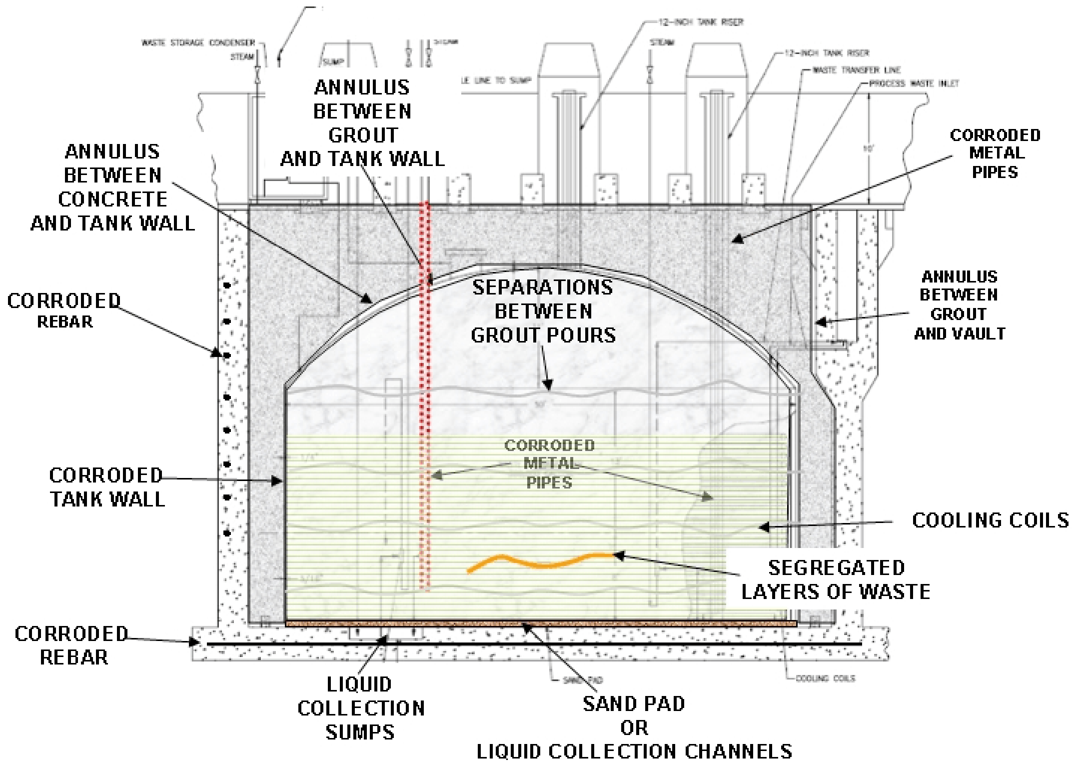


Figure 6. Cross Section of a Typical Tank With Cooling Coils at Idaho National Laboratory and Features That May Act as Conduits Superimposed on Tank Cross Section From DOE (2006)

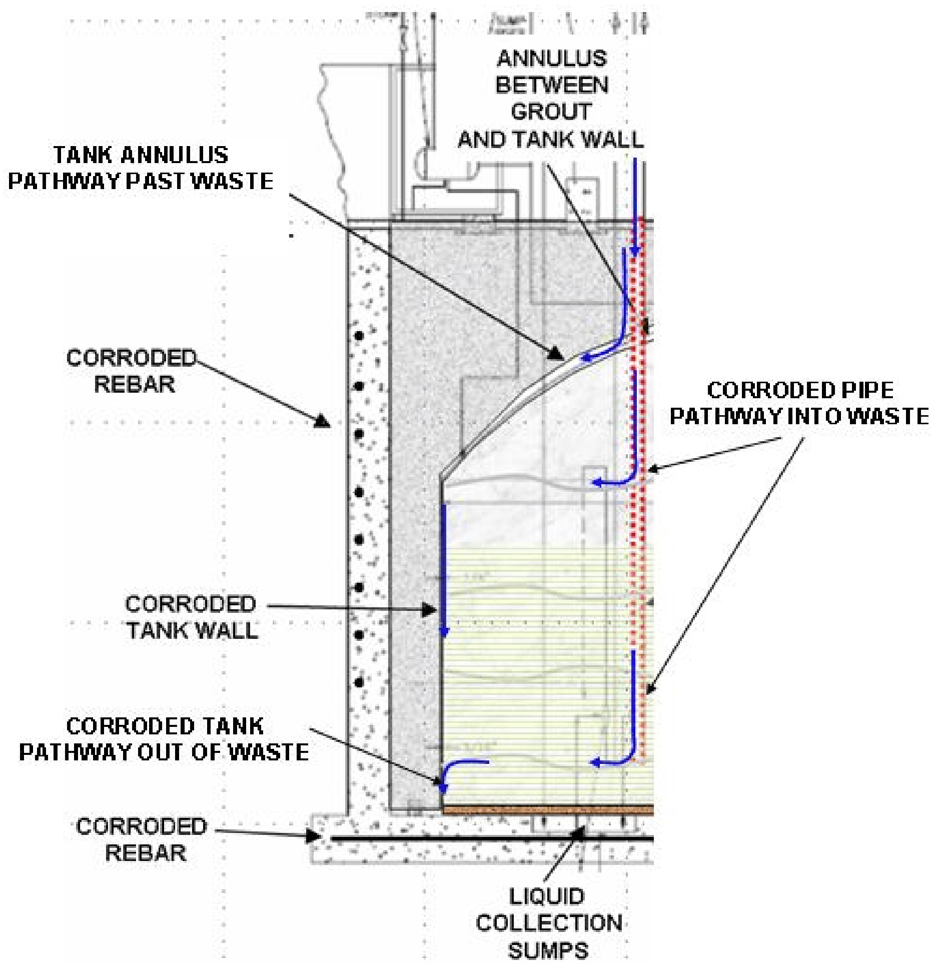


Figure 7. Illustration of Fast and Bypassing Conduit Pathways. Corroded Pipe May Act as Fast Pathway to the Waste Form. Corroded Tank Wall May Act as Either a Pathway Bypassing the Waste Form or a Fast Pathway From the Waste Form to the Environment (Based on Tank Cross Section From DOE, 2006).

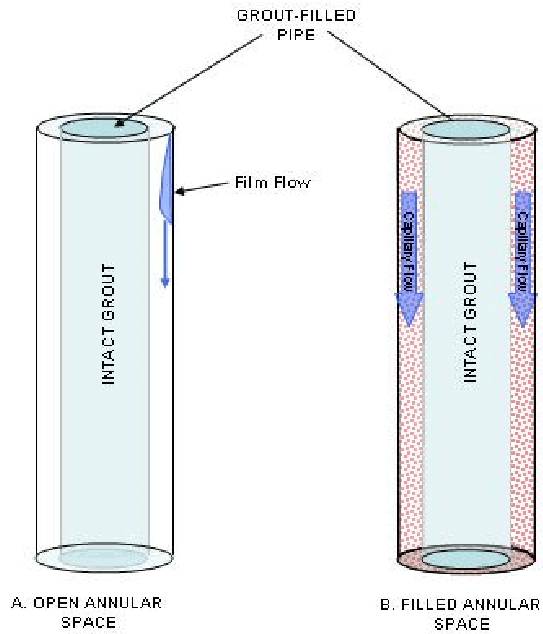


Figure 8. Liquid Flow Regimes in an Annular Space Around a Grouted Pipe. A: Film Flow in an Open Annular Space With Large Aperture; B: Capillary Flow in an Annular Space Filled With Corrosion Products or With Small Aperture.

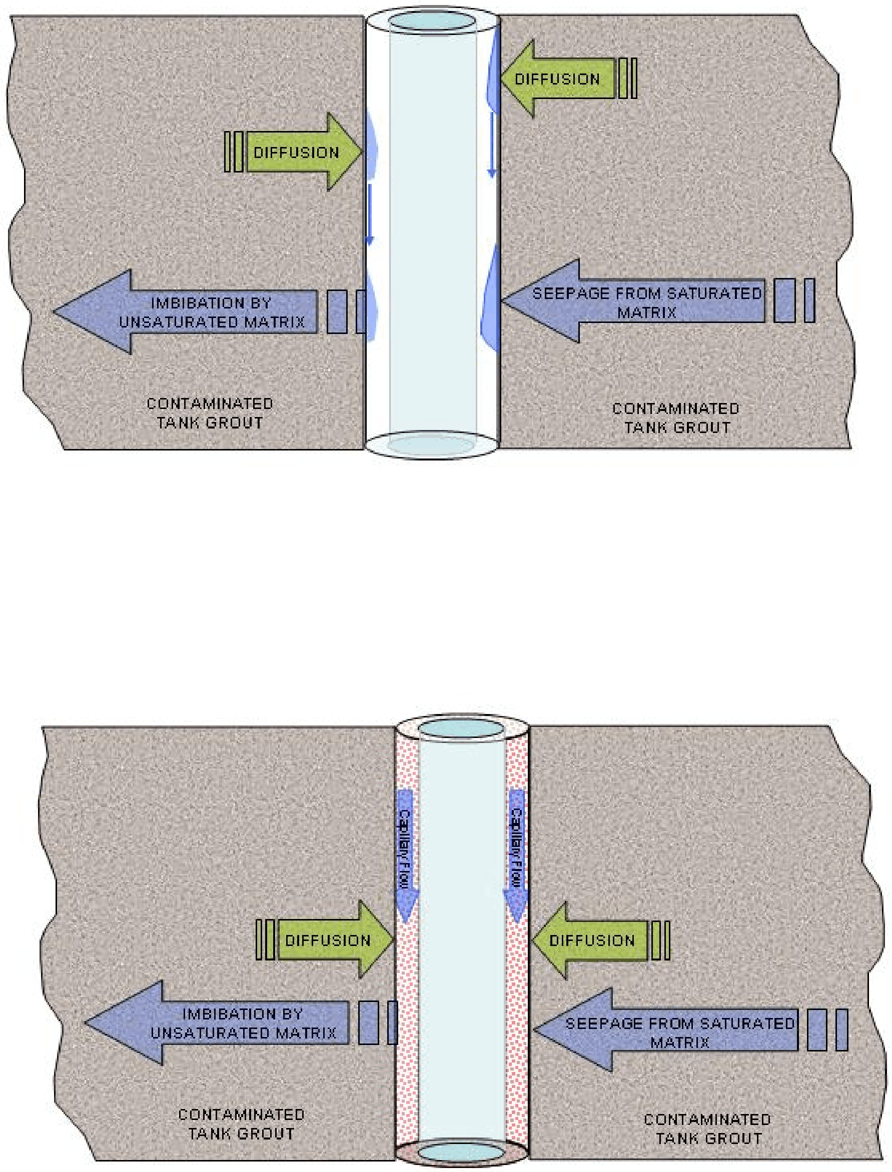


Figure 9. Fluid and Mass Transport Processes in Annular Spaces. A: Open Annular Space, B: Filled Annular Space.

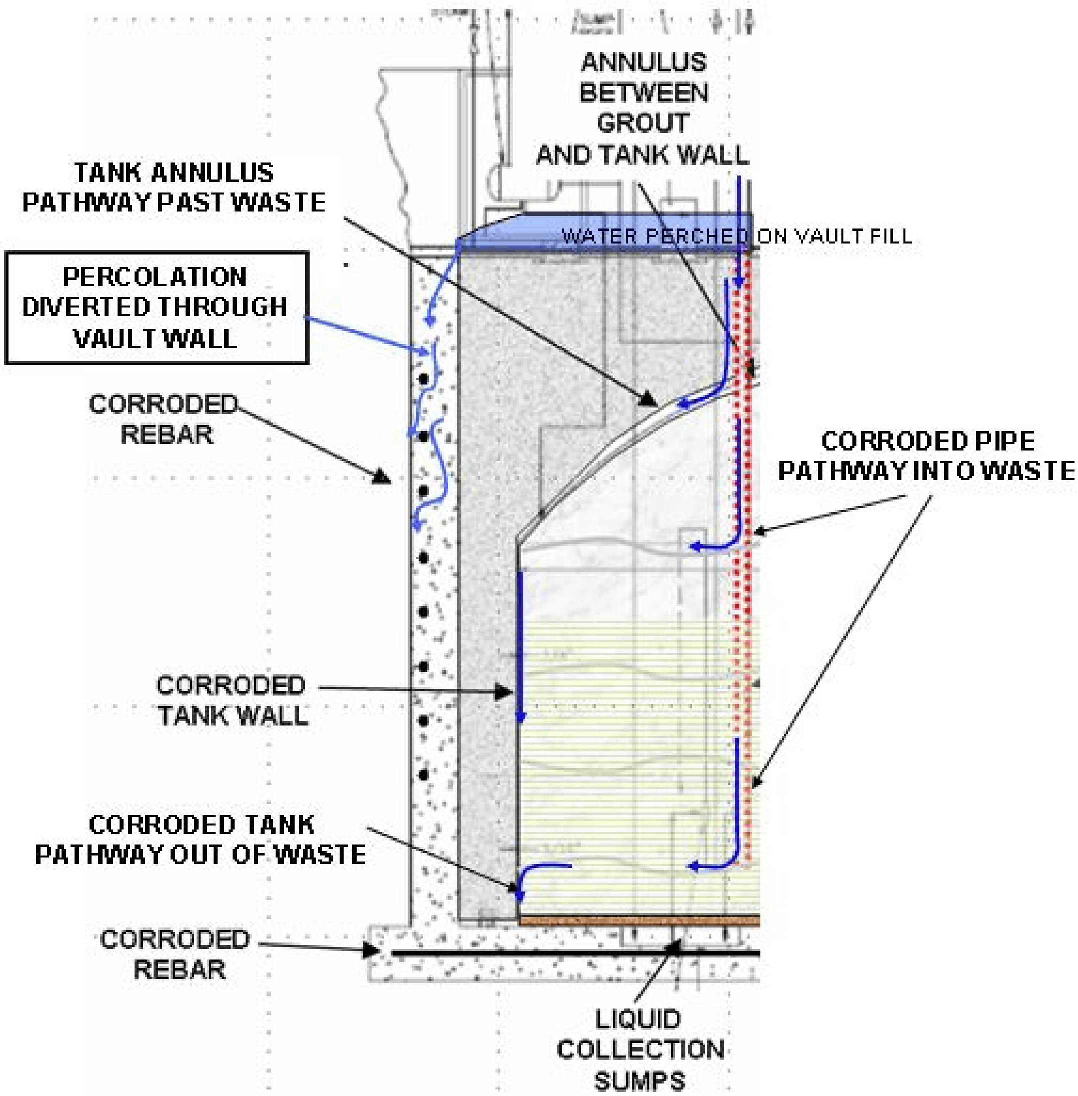


Figure 10. Illustration of Percolation Diverted Away From Waste Form Through Cracks and Conduits in Vault Wall (Based on Tank Cross Section From DOE, 2006)

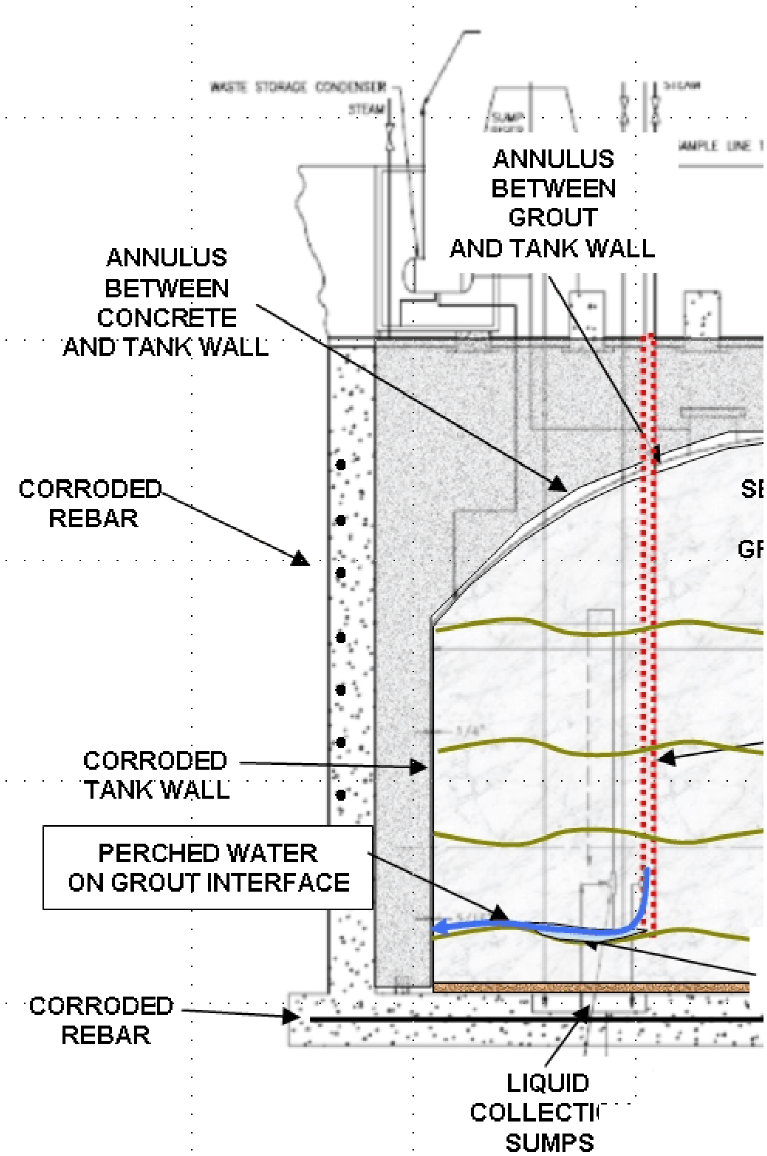


Figure 11. Conduit Flow Along a Grout Pour Interface Due to Perching on a Grout Layer (Based on Tank Cross Section From DOE, 2006)

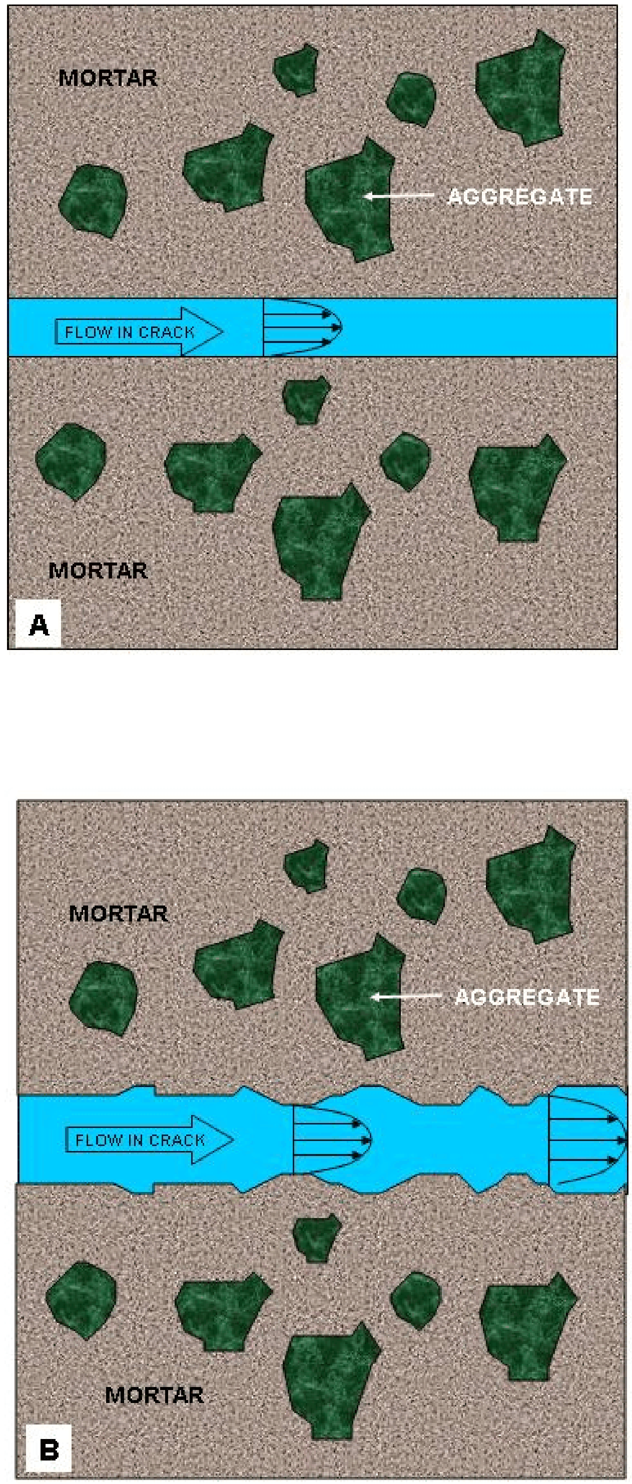


Figure 12. Illustration of Capillary Flow Through a Microcrack in a Porous Matrix. A: Smooth Crack Walls, B: Rough Crack Walls.

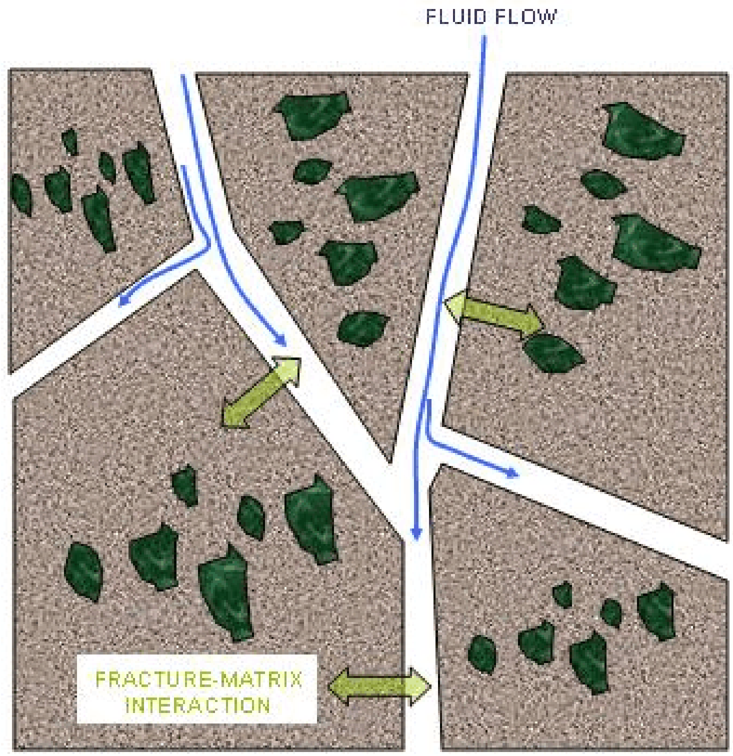


Figure13. Illustration of Discrete Fracture and Transport Model

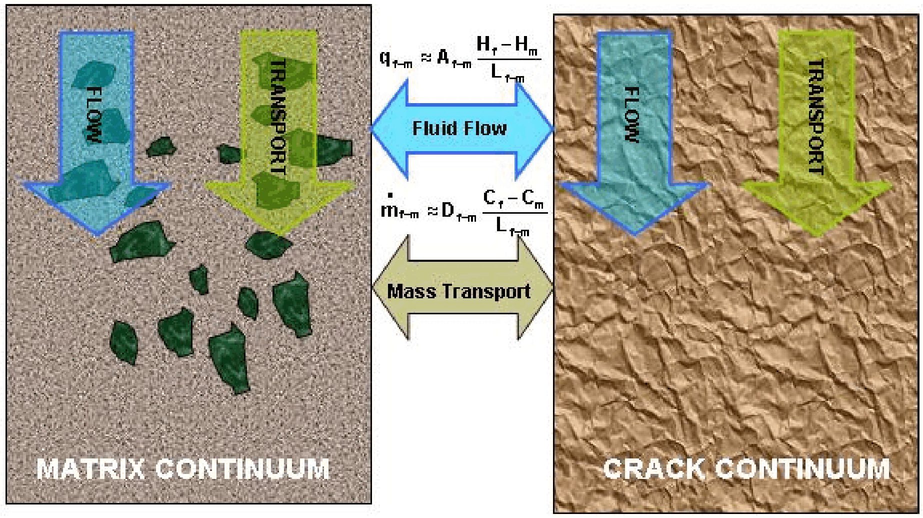


Figure 14. Conceptual Illustration of Flow and Transport Coupling in a Dual Porosity Continuum Model [Symbols Defined in Eqs. (3) and (16)]

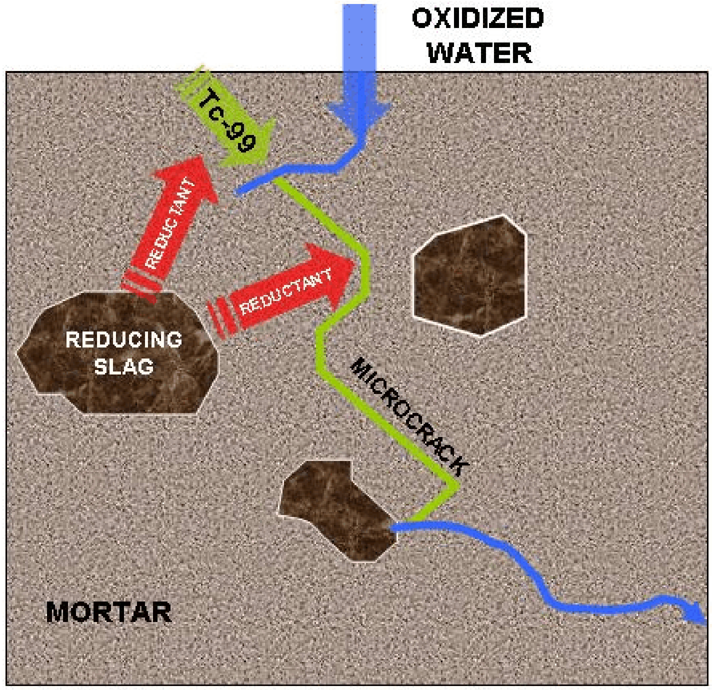


Figure 15. Conceptual Illustration of Chemical Disequilibrium Between Fluid in a Microcrack and a Reducing Aggregate

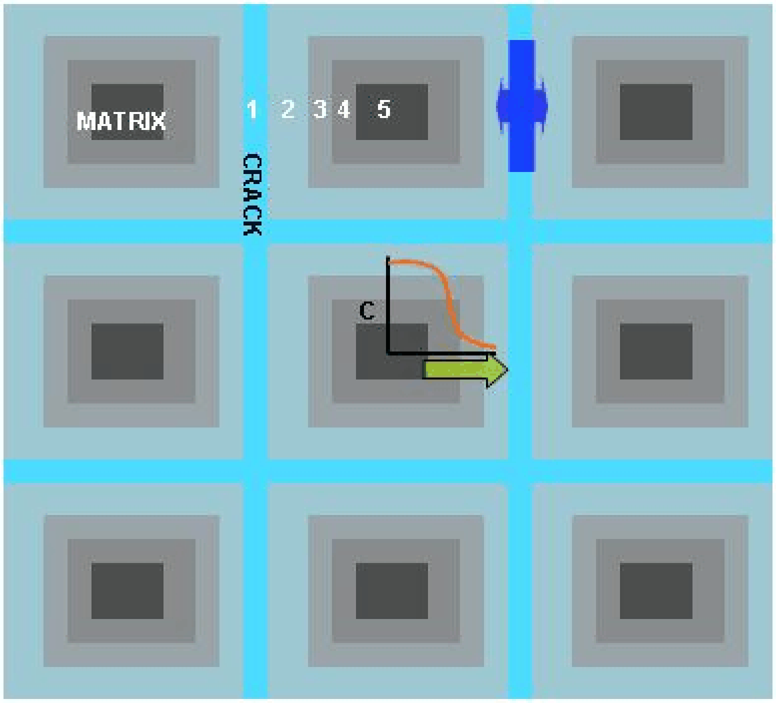


Figure 16. Conceptual Illustration of the Multiple Interacting Continua (MINC) Modeling Approach

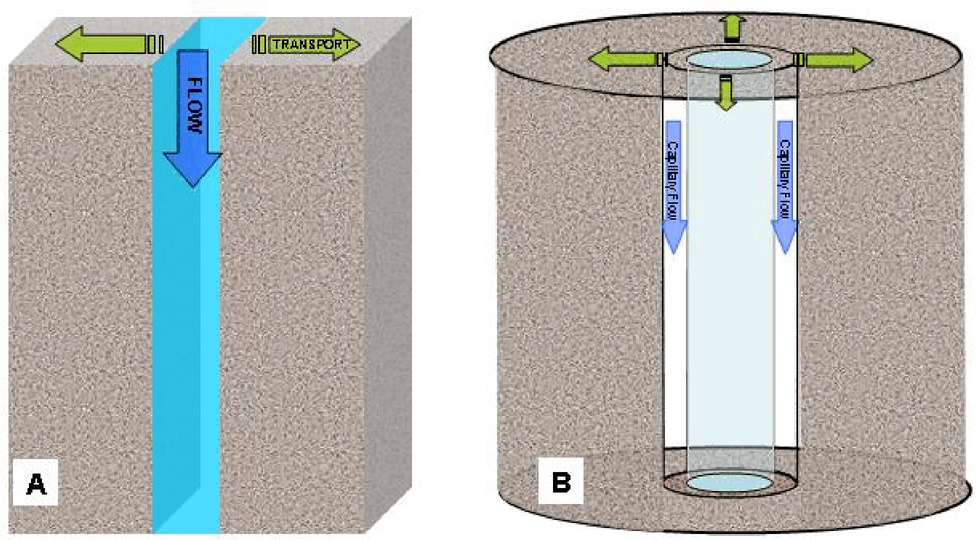


Figure 17. Illustration of Difference in Flow and Transport Geometry Between a Fracture and Small Conduit. A: Linear Flow and Transport from a Fracture, B: Radial Flow and Transport Away From Cylindrical Conduit.