Development of a Performance Assessment Model: Lecture Notes

K. Huff

khuff@cae.wisc.edu

Contents

1 Groundwater Hydrology [1, 2]

Groundwater hydrology is the study of groundwater movement.

1.1 Hydraulic Head

Hydraulic head is the hydrologic indicator of potential energy. It has units of length, specifically height above a geological *datum*, the zero point relative to which heads are measured.

One way to derive this notion is according to Hubbart's derivation [2]. The work to raise the water to a pressure, P, and the work required to raise the water to an elevation, z, are additive.

The work required to raise a mass, m, to a pressure, P, is described by the integral

$$W_P = \frac{1}{m} \int_0^P V dP \tag{1}$$

where

$$m = \text{ mass of the water } [kg]$$
$$V = \text{ volume of the water } [m^3]$$
$$P = \text{ pressure } [kg/m/s^2].$$

In an incompressible fluid such as water, the work W_P required to raise a unit mass to some pressure, P, becomes

$$W_P = P/\rho_w \tag{2}$$

where

$$\rho_w = \text{ the density of water} [kg/m^3].$$

The work required to raise a unit mass to elevation z from elevation z_{ref} (the 'datum') is simply the work, W_g against gravity

$$W_g = g(z - z_{ref}) \tag{3}$$

where

$$g = \text{ acceleration of gravity } [m/s^2].$$

From these expressions for work the potential energy function, ϕ, of the two separate potentials, pressure and elevation, act additively on a unit mass of groundwater. Their combined effects can be written as the sum,

$$\phi = W_P + W_g$$
$$= \frac{P}{\rho_w} + g(z - z_{ref}.) \tag{4}$$

If

$$z_{ref} = 0$$

and the potential energy is directly proportional to gravity

$$\phi = gh$$

where

$$h = \text{Darcy's experimental head } [m],$$

then

$$
\begin{aligned}
h &= \frac{\phi}{g} \\
&= \frac{P}{\rho_w g} + z \\
&= \psi + z
\end{aligned}
\tag{5}
$$

where

$$z = \text{elevation head } [m]$$
$$\psi = \text{pressure head } [m].$$

Thus, the total hydraulic head, h, of a column of water is the vertical distance from the datum $(h = 0)$ to the height of the water surface. Elevation head, z, is the distance from the datum to the measuring point in the flow field. The pressure head, ψ, is the pressure, P in $[Pa]$, at that measurement point, divided by the density of the fluid, ρ_w, and the accelation of gravity, g. Pressure head, elevation head, and total head are shown in Figure 1.
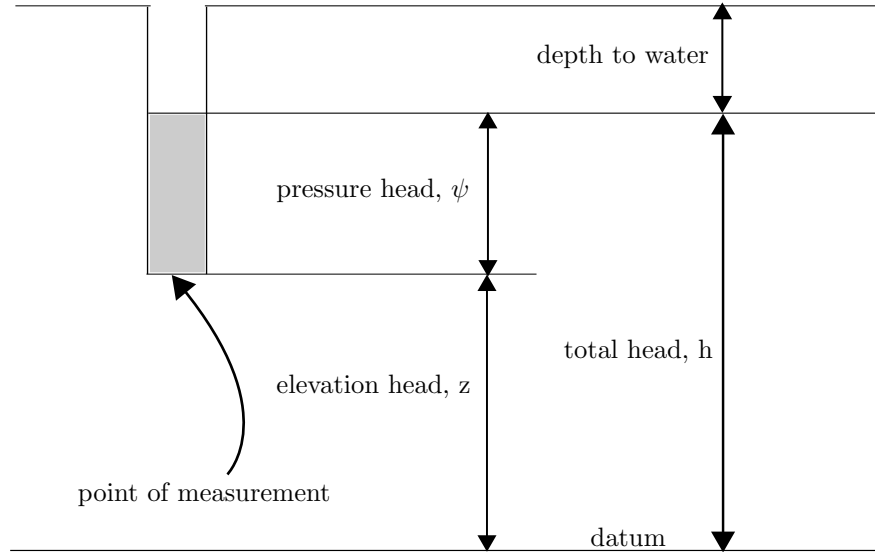
Figure 1: Various types of head value in a column of water in a well are measured with respect to the datum.

Different lithologies and candidate sites have different head gradients. Due to anisotropies in their hydraulic conductivity tensors (see section 1.4), some lithologies, such as clay, are less likely to have strong head gradients.

1.2   Porosity

Porosity is a hydraulic parameter which significantly contributes to the hydraulic conductivity of a medium.

The porosity of a medium is the ration of void volume, V_v, in the medium to the total volume, V_T of the medium. As shown in Figure 2, the volume of water sufficient to saturate a packed sand matrix is equivalent to the void volume.
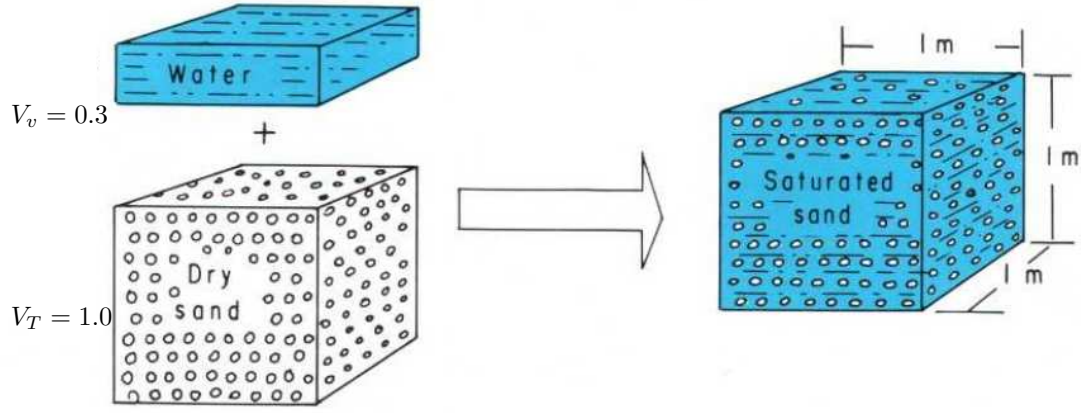
$$\text{Porosity(n)} = \frac{\text{Volume of Voids }_{(V_v)}}{\text{Total Volume }_{(V_T)}} = \frac{0.3m^3}{1.0m^3} = 0.3$$

Figure 2: The ratio of the volume of saturation water to the total packed sand volume gives the porosity, n, in [%] [3].

The porosity of a material is therefore mathematically defined as

$$
\begin{aligned}
n_T &= \frac{\text{void volume}}{\text{total volume}} \\
&= \frac{V_v}{V_T} \\
&= \frac{V_T - V_s}{V_T}
\end{aligned}
\tag{6}
$$

where

$$
\begin{aligned}
n_T &= \text{ total porosity } [\%] \\
V_v &= \text{ void volume } [m^3] \\
V_T &= \text{ total volume } [m^3] \\
V_s &= \text{ volume of solids } [m^3].
\end{aligned}
$$

The dry solid volume can be defined as

$$V_s = \frac{m_s}{\rho_s} \tag{7}$$

and the total dry volume can be defined as

$$V_T = \frac{m_s}{\rho_b}, \tag{8}$$

where

$$
\begin{aligned}
\rho_s &= \text{ grain density, or density of solids } [kg/m^3] \\
m_s &= \text{ mass of solids } [kg]
\end{aligned}
$$

4

and

$$\rho_b = \text{ bulk (dry) density } [kg/m^3],$$

equation (6) can be rewritten

$$n_T = 1 - \frac{\rho_b}{\rho_s}. \tag{9}$$

The total porosity is a combination of primary and secondary porosity, shown in Figure 3. Primary porosity is the macroscopically homogeneous void space present between grains in a geological matrix. This type of porosity includes empty space between grains of sand or consolidated rock characteristic of the level of packing or method of consolidation of the matrix. Secondary porosity is not intrinsic to the rock, but induced in it. Examples of secondary porosity include features such as fractures, lava tubes, and caverns.
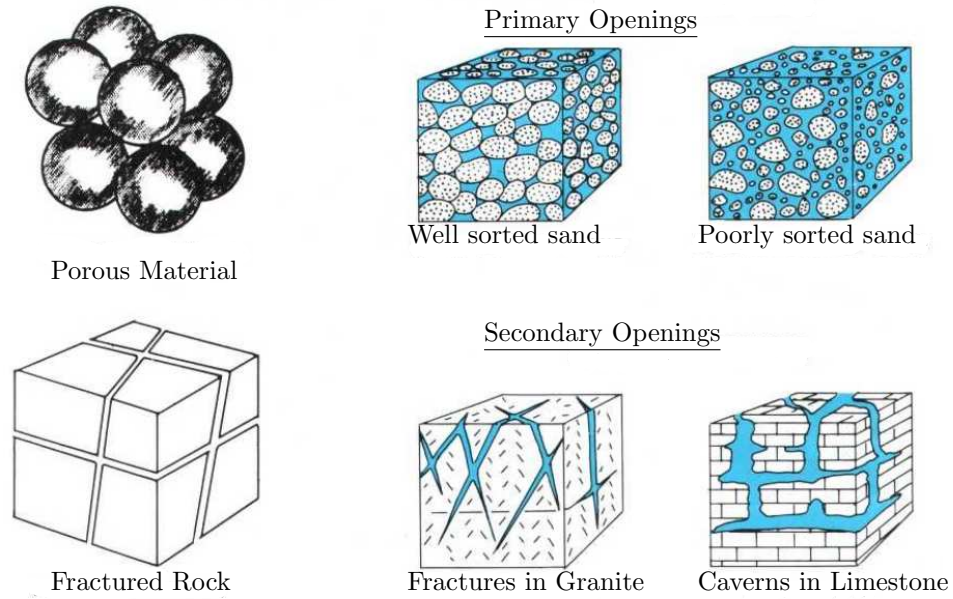
Figure 3: Primary porosity, a homogeneous characteristic of the rock matrix, and secondary porosity, such as discrete fractures, caverns, and lava tubes, both contribute to total porosity [3].

Equipped with a notion of the total porosity, it is possible to define the effective porosity shown in figure 4, which is the interconnected porosity through which fluid may flow,

$$n_{eff} = \frac{V_c}{V_T} n_T. \tag{10}$$

where

$$V_c = \text{ interconnected volume.}$$

Effective porosity is more influential on ground water flow in a porous medium than the total porosity, as it contributes to the permeability of the medium. In some media, the effective porosity is significantly different than the total porosity, resulting in much lower flow rates than the total porosity alone would indicate. Granite, for example, has an effective porosity that is nearly three magnitudes lower than its total porosity.

"poorly" connected pore volume

$n_{eff} \ll n_T$

$n_{eff} \sim n_T$

"well" connected pore volume
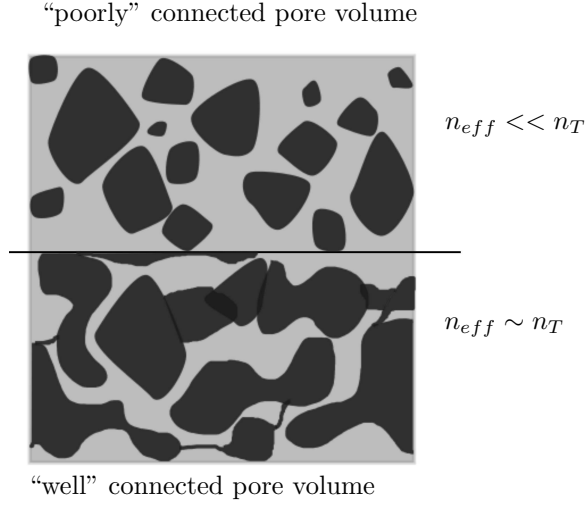
Figure 4: A medium with a poorly connected pore volume has low effective porosity, and vice versa.

1.3   Darcy's Law

Darcy's Law is analgous to Fourier's Law of heat conduction, and states that flow occurs as a function of a head gradient with a speed that obeys the hydraulic properties of the medium. Darcy's law for one dimensional flow is

$$\frac{Q}{A} = -K\frac{h_2 - h_1}{l_2 - l_1} \tag{11}$$

where

$$Q = \quad \text{volumetric flow rate } [m^3/s]$$

and

$$
\begin{aligned}
A &= \quad \text{cross sectional area of flow field } [m^2] \\
K &= \quad \text{hydraulic conductivity } [m/s]. \\
h_i &= \quad \text{hydraulic head measured at point i } [m] \\
l_i &= \quad \text{linear position of point i } [m]
\end{aligned}
$$

Using the the definition of specific discharge (also known as Darcy flux), $q = Q/A$, and expressing the law in its differential form gives

$$q = -K\frac{\partial h}{\partial l}. \tag{12}$$

The three dimensional form requires that the one dimensional form in equation (12) be true for each spatial component, such that

$$\vec{q} = -K\nabla h \tag{13}$$

where

$$
\begin{aligned}
\vec{q} &= \quad \text{specific discharge vector } [m^3/s] \\
&= q_x\hat{i} + q_y\hat{j} + q_z\hat{k}
\end{aligned}
$$

6

and

$$\nabla h = \frac{\partial h}{\partial x} + \frac{\partial h}{\partial y} + \frac{\partial h}{\partial z}.$$

In the case of anisotropic media, the hydraulic conductivity, K is given as a spatially heterogenous tensor, $\mathbf{K}$. The general, three dimensional, anisotropic Darcy equation is therefore

$$\vec{q} = -\mathbf{K}\nabla h \tag{14}$$

where

$$\mathbf{K} = \text{ hydraulic conductivity tensor } [m/s].$$

1.4 Hydraulic Conductivity

The hydraulic conductivity of a medium is a tensor that describes the ease with which water passes through the pore spaces of the medium in all directions. This tensor may be homogeneous or heterogeneous and isotropic or anisotropic.

1.4.1 Heterogeneity of the Hydraulic Conductivity

Heterogeneity of a hydrogeological unit describes the spatial variability of the hydraulic conductivity. If the hydraulic conductivity is the same when measured at all points in the hydrogeological medium,

$$\forall i, j \in V : K(x_i, y_i, z_i) = K(x_j, y_j, z_j), \tag{15}$$

then the medium is homogenous. If instead, the hydraulic conductivity is different at different locations, such that

$$\exists i \neq j \in V : K(x_i, y_i, z_i) \neq K(x_j, y_j, z_j), \tag{16}$$

then the medium is heterogeneous.

1.4.2 Anisotropy of the Hydraulic Conductivity

Anisotropy of a hydrogeological unit describes the local directional dependence of the hydraulic conductivity tensor. Specifically, at a single location, an isotropic medium will demonstrate the same hydraulic conductivity in all directions, while an anisotropic medium will not.

Mathematically speaking, if the conductivity is constant at all measurement angles, θ, such that

$$\forall \theta_i, \theta_j \in (0, 2\pi) : K(\theta_i, x, y, z) = K(\theta_j, x, y, z) \tag{17}$$

$$\tag{18}$$

then the medium is isotropic. If instead, the angular components of the hydraulic conductivity are nonconstant, such that

$$\exists \theta_i \neq \theta_j \in (0, 2\pi) : K(\theta_i, x, y, z) \neq K(\theta_j, x, y, z), \tag{19}$$

then the medium is anisotropic and the tensor is expressed most generally,

$$\mathbf{K} = \text{ hydraulic conductivity tensor } [m/s]$$
$$= \begin{bmatrix} K_{xx} & K_{xy} & K_{xz} \\ K_{yx} & K_{yy} & K_{yz} \\ K_{zx} & K_{zy} & K_{zz} \end{bmatrix} \tag{20}$$

where

$$K_{ij} = \text{jth component in the i direction.}$$

The general tensor in equation (20) is a second rank symmetric tensor and can always be simplified by aligning the tensor with the principal axis of anisotropy[1]. This principal direction is defined by the directions of maximum $(K_\parallel)$, minimum $(K_\perp)$, and intermediate $(K_\parallel \times K_\perp)$ hydraulic conductivity. That is, by aligning the tensor with the flow axis, it may be reduced to a diagonal matrix,

$$\mathbf{K} = \left[\begin{array}{ccc} K_{xx} & 0 & 0 \\ 0 & K_{yy} & 0 \\ 0 & 0 & K_{zz} \end{array} \right]. \tag{21}$$

2   Mechanisms of Solute Transport

Solutes in groundwater are transported due to various mechanisms.

2.1   Diffusion

Solutes will move from a location of high concentration to a location of low concentration due to the random thermal kinetic enery of the solute. Brownian motion is responsible for diffusion.
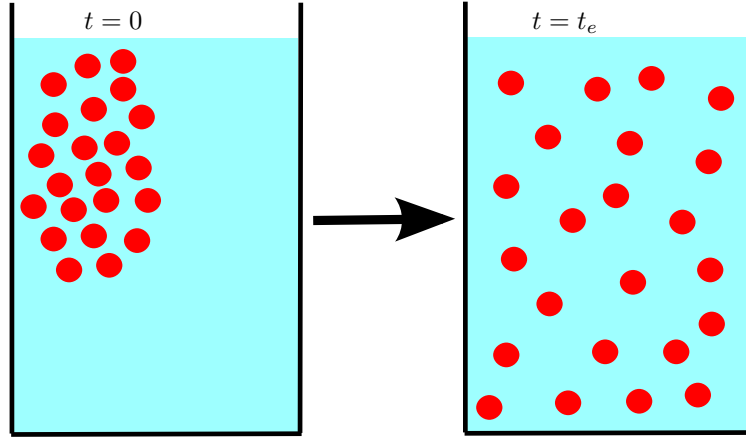
Figure 5: In diffusive transport, particles move according to random thermal motion down the concentration gradient [4].

A mathematical description of this phenomenon can be addressed by describing the particle's motion as a random walk and applying statistical analysis. Another derivation of this phenomenon describes the flux, J through a plane in a volume, and gives Fick's Law for porous media,

$$J_{xdif} = -D'\frac{dC}{dx}$$

$$\tag{22}$$

where

$$
\begin{aligned}
J_x &= \text{ Mass flux through the y-z plane } [kg/m^2/s]\\
D' &= \text{ Effective diffusion coefficient } [m^2/s]\\
C &= \text{ Concentration } [kg/m^3].
\end{aligned}
$$

(23)

The determination of the diffusion coefficient can be empirical or analytical. The Arrhenius equation gives a thermal dependence of the diffusion coefficient as does the Stokes-Einstein relationship.

The effecive diffusion coefficient for porous media is dependent on porosity and tortuosity, which affect the diffusion behavior of a solute. In a saturated porous medium, diffusion takes place only within the pore volume where liquid is present. To take this into account, the effective diffusion coefficient in a porous medium is reduced by both the absence of fluid in the matrix volume, and by the so-called "tortuosity" of the pathways.

$$
J_{xdif} = -n\tau D_m \frac{dC}{dx} \tag{24}
$$

(25)

where

$$
\begin{aligned}
J_{xdif} &= \text{ Mass flux through the y-z plane in a porous medium} [kg/m^2/s]\\
n &= \text{ porosity } [\%]\\
\tau &= \text{ tortuosity } [-]\\
D_m &= \text{ Molecular diffusion coefficient } [m^2/s]\\
C &= \text{ Concentration } [kg/m^3].
\end{aligned}
$$

The tortuosity of a saturated porous medium can be estimated from the porosity with an equation developed by Millington and Quirk (1961),

$$
\tau = \frac{n_w^{7/3}}{n^2} \tag{26}
$$

which for a saturated medium is estimated as

$$
\tau = n^{1/3}. \tag{27}
$$

(28)

2.2 Dispersion [1]

Solutes can be driven to move due to mechanical mixing, known as dispersion. Solutes acting under dispersion may travel further than they would due to advection alone. During fluid mixing in which a fluid with one solute concentration displaces a fluid with another concentration, dispersion causes a zone of mixing to develop around the advective front.

Dispersion in groundwater is caused by a combination of two phenomena. The first is mechanical dispersion, in which local variability of the fluid velocity around the mean due to mechanical heterogeneities of the rock at many scales. The second phenomenon is diffusion, discussed in section 2.1.

A mathematical description of this phenomenon will come generally from fluid dynamics. The total mechanical dispersive and diffusive phenomena can be combined as a total dispersive expression,

$$
D = D_{mdis} + \tau D_m \tag{29}
$$

9

where

$$D_{mdis} = \text{coefficient of mechanical dispersivity } [m^2/s]$$

$$= \alpha_L \frac{v_x^2}{|v|} + \alpha_{TH} \frac{v_y^2}{|v|} + \alpha_{TV} \frac{v_z^2}{|v|}, \tag{30}$$

$$\alpha_L = \text{longitudinal dispersivity } [m],$$

$$\alpha_{TH} = \text{horizontal transverse dispersivity } [m],$$

$$\alpha_{TV} = \text{vertical transverse dispersivity } [m],$$

$$\tau = \text{tortuosity } [-],$$

and

$$D_m = \text{coefficient of molecular diffusion } [m^2/s].$$

The dispersive mass flux can therefore be described, as in Burnett and Frind, 1987,

$$J_{dis} = J_{mdis} + J_{dif} \tag{31}$$

$$= -n\left(D_{mdis} + \tau D_m\right) \nabla C \tag{32}$$

$$= -nD\nabla C \tag{33}$$

where

$$D = \begin{bmatrix} D_{xx} & D_{xy} & D_{xz} \\ D_{yx} & D_{yy} & D_{yz} \\ D_{zx} & D_{zy} & D_{zz} \end{bmatrix},$$

$$D_{ij} = \begin{cases} \alpha_L \frac{v_x^2}{|v|} + \alpha_{TH} \frac{v_y^2}{|v|} + \alpha_{TV} \frac{v_z^2}{|v|} & i = j = x, \\ \alpha_L \frac{v_y^2}{|v|} + \alpha_{TH} \frac{v_x^2}{|v|} + \alpha_{TV} \frac{v_z^2}{|v|} + \tau D_m & i = j = y, \\ \alpha_L \frac{v_z^2}{|v|} + \alpha_{TV} \frac{v_y^2}{|v|} + \alpha_{TV} \frac{v_x^2}{|v|} + \tau D_m & i = j = z, \\ (\alpha_L - \alpha_{TH}) \frac{v_x v_y}{|v|} + \tau D_m & i = x, j = y, \\ (\alpha_L - \alpha_{TV}) \frac{v_x v_z}{|v|} & i = (x,z), j = (z,x), \\ (\alpha_L - \alpha_{TV}) \frac{v_y v_z}{|v|} & i = (y,z), j = (z,y), \end{cases} \tag{34}$$

and

$$|v| = \sqrt{v_x^2 + v_y^2 + v_x^2}. \tag{35}$$

For uniform flow, where $v = v_x$ and $v_y = v_z = 0$,

$$D_x = D_L$$

$$= \alpha_L v_x + \tau D_m \tag{36}$$

$$D_y = D_{TH}$$

$$= \alpha_{TH} v_x + \tau D_m \tag{37}$$

$$D_z = D_{TV}$$

$$= \alpha_{TV} v_x + \tau D_m. \tag{38}$$

2.3   Advection

Solutes are transported by advection when they move along with the water in which they are dissolved. The expression for mass flux due to dispersion can be added to the expression for mass flux due to advection to give total mass flux due to both,

$$J = J_{mdis} + J_{dif} + J_{adv} \tag{39}$$

$$= J_{dis} + J_{adv} \tag{40}$$

$$= -nD\nabla C + nvC \tag{41}$$

$$= \left(-nD_{xx}\frac{\partial C}{\partial x} - nD_{xy}\frac{\partial C}{\partial y} - nD_{xz}\frac{\partial C}{\partial z} + nv_xC\right)\hat{i}$$

$$+ \left(-nD_{yx}\frac{\partial C}{\partial x} - nD_{yy}\frac{\partial C}{\partial y} - nD_{yz}\frac{\partial C}{\partial z} + nv_yC\right)\hat{j}$$

$$+ \left(-nD_{zx}\frac{\partial C}{\partial x} - nD_{zy}\frac{\partial C}{\partial y} - nD_{zz}\frac{\partial C}{\partial z} + nv_zC\right)\hat{k}. \tag{42}$$

For unidirectional flow

$$J = \left(-nD_{xx}\frac{\partial C}{\partial x} + nv_xC\right)\hat{i} + \left(-nD_{yy}\frac{\partial C}{\partial y}\right)\hat{j} + \left(-nD_{zz}\frac{\partial C}{\partial z}\right)\hat{k}. \tag{43}$$

3 Geochemistry

Geochemical phenomena significantly affect solute transport. As such, the geochemical attributes of various environments contribute differently to repository performance.

3.1 Saturated and Unsaturated Environments

Saturated repository concepts are found below the water table. Water permeates the rock.

Unsaturated repository concepts are found above or near the top of the water table. Water does not permeate the rock.

Current investigations are focused primarily on saturated environments.

3.2 Reducing and Oxidizing Environments[5]

Elements exist in various oxidation states, a positive or negative number refering to the charge of the atom due to the number of electrons it posesses. Oxidation refers to the increase of the oxidation state number of an element, and reduction is the opposite. That is, while oxidation is the chemical loss of electrons by an atom, reduction is the gaining of electrons by an atom.

The redox potential is the tendency in potential energy for an element to give up its electron and become more oxidized. This potential energy is measured relative to the potential for hydrogen atoms at unit concentration to lose electrons and become oxidized, in volts,

$$H_2 \rightarrow 2H^+ + 2e. \tag{44}$$
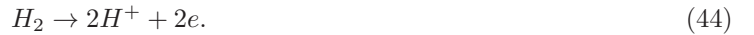
The potential for the redox equation in equation (44) is chosen to be 0 volts, such that for positive values of the redox potential, reactions tend to proceed in the oxidation direction. Accordingly, reactions proceed in the reduction direction for negative potentials.

Closed, saturated envrionments are reducing. Unsaturated environments and environments that are saturated and open are oxidizing.

The far field of most reducing environments are slightly oxidizing as they reach the zone near the overlying aquifer.

3.3 Salinity

Salinity affects sorption and solubility behaviors. It also expedites corrosion. Finally, salt is an important indicator of the historical flow at a site. Where should salinity information get incorporated in this model?

3.4 Sorption

Sorption is a factor that retards movement, in which solutes are incorporated into the surfaces of pores or fractures (adsorption) or incorporated into the rock matrix (absorption). More specificially, when water with some solute concentration, C_i, meets a porous medium or a granular solid, the reaction proceeds toward an equilibrium in which the solute mass has partitioned between the solution and the solid, such that,

$$S = \frac{V_w(C_i - C)}{m_s} \tag{45}$$

where

$$
\begin{aligned}
S &= \text{ mass sorbed on the surface } [kg/kg] \\
V_w &= \text{ volume of the solution } [m^3] \\
C_i &= \text{ initial concentration in the solution } [kg_m^3] \\
C &= \text{ equilibrium concentration in the solution } [kg/m^3] \\
m_s &= \text{ sediment mass } [kg].
\end{aligned}
$$

The function $S(C)$ is called a sorption isotherm. Two common forms for the sorption isotherm are

$$S = \begin{cases} \text{Freundlich :} & KC^n \\ \text{Langmuir :} & \frac{Q^0 KC}{1+KC} \end{cases} \tag{46}$$

where

$$
\begin{aligned}
K &= \text{ partition coefficient } [m^3/kg] \\
n &= \text{ shaping constant } [-]
\end{aligned}
$$

and

$$Q^0 = \text{ maximum sorptive capacity } [kg/kg].$$

If in the Freundlich isotherm,

$$n = 1, \tag{47}$$

then

$$S = K_d C \tag{48}$$

where

$$K_d = \text{ the distribution coefficient } [m^3/kg],$$

and the isotherm becomes very easy to incorporate into the mass transport equations, as in equation (62).

The simplest way to model sorption is with a linear isotherm and tabulated values. Sorption coefficients such as K_d are tabulated elementally because the character of the equilibrium reaction is element specific. For example, iodine has different sorption behavior than carbon. These coefficients are also host rock specific. That is, each element has different sorption behaviors in clay than in granite.

Geochemistry also plays a role, and sorption for some elements are higher in reducing environments. Sorption for some elements may be higher in oxidizing environments.

3.5 Solubility

The dissolution behavior of solutes in aqueous solutions is called its solubility. This behavior is limited by the solute's solubility limit, described by an equilibrium constant that depends upon temperature, water chemistry, and the properties of the element. The solubility constant for ordinary solutes, K_s gives units of concentration, $[kg/m^3]$, and can be determined algebraically by the law of mass action which gives the partitioning at equilibrium between reactants and products. For a reaction

$$cC + dD = yY + zZ, \tag{49}$$

where

$$
\begin{aligned}
c, d, y, z &= \text{ amount of respective constituent } [mol] \\
C, D &= \text{ reactants } [-] \\
Y, Z &= \text{ products } [-],
\end{aligned}
$$

the law of mass action gives

$$K = \frac{(Y)^y (Z)^z}{(C)^c (D)^d} \tag{50}$$

where

$$
\begin{aligned}
(X) &= \text{ the equilibrium molal concentration of X } [mol/m^3] \\
K &= \text{ the equilibrium constant } [-].
\end{aligned}
$$

The equillibrium constant for many reactions are known, and can be found in chemical tables. Thereafter, the solubility constraints of a solution at equilibrium can be found algebraically. In cases of salts that dissociate in aqueous solutions, this equilibrium constant is called the salt's solubility product K_{sp}.

This equillibrium model, however, is only appropriate for dilute situations, and nondilute solutions at partial equilibrium must be treated with an activity model by substituting the activities of the constituents for their molal concentrations,

$$[X] = \gamma_x (X) \tag{51}$$

where

$$
\begin{aligned}
[X] &= \text{ activity of X } [-] \\
\gamma_x &= \text{ activity coefficient of X} [-] \\
(X) &= \text{ molal concentration of X} [mol/m^3]
\end{aligned}
$$

such that

$$
\begin{aligned}
IAP &= \text{ Ion Activity Product } [-]. \\
&= \frac{[Y]^y [Z]^z}{[C]^c [D]^d} \tag{52}
\end{aligned}
$$

$$\tag{53}$$

The ration between the IAP and the equillibrium constant (IAP/K) quantifiesn the departure from equilibrium of a solution. This information is useful during the transient stage in which a solute is first introduced to a solution. When $IAP/K < 1$, the solution is undersaturated with respect to the products. When, conversely, $IAP/K > 1$, the solution is oversaturated and precipitation of solids in the volume will occur.

Two models of mass balance applicable to a control volume incorporating solubility limitation include the Ahn and Hedin models.

3.5.1 Ahn Solubility Limited Release Model

In the Ahn models, radionuclides with lower solubility coefficients are modeled with the solubility limited release model. Solubility values are assumed from TSPA for this model, and elements with a solubility of less than $5 \times 10^{-2} [mol/m^3]$ are taken to be 'low.' Elements in this 'low' category include Zr, Nb, Sn and some toxic actinides such as Th and Ra for an oxidizing, unsaturated environment similar to the Yucca Mountain Repository. It should be noted that in a reducing environment, the actinides are not as mobile, and the high and low solubility radionuclides will differ from this model. This model suggests that dissolution of radionuclides into the flowthrough water is dominated by diffusion, which is largely dependent upon the concentration gradient between the waste matrix and the water. The mass balance driving radionuclide release takes the form:

$$\dot{m}_i = 8\epsilon D_e S_i L \sqrt{\frac{Ur_0}{\pi D_e}} \tag{54}$$

where ϵ, U, r_0, and L are the geometric and hydrologic factors porosity, water velocity, waste package radius, and waste package length, respectively. D_e is the effective diffusion coefficient (m^2/yr) and S_i is the solubility (kg/m^3) of isotope i.

3.5.2 Hedin Solubility Limited Release Model

In the Hedin model of the waste matrix, the amount of solute available within the waste package is solved for, and for radionuclides with low solubility, the mass fraction released from the waste matrix is limited by a simplified description of their solubility. That is,

$$m_{1i}(t) \le v_{1i}(t) C_{sol} \tag{55}$$

where the mass m_{1i} in $[kg]$ of a radionuclide i dissolved into the waste package void volume v_1 in $[m^3]$, at a time t, is limited by the solubility limit, the maximum concentration, C_{sol} in $[kg/m^3]$ at which that radionuclide is soluble [6].

Various things affect solubility. Temperature, salinity, etc.

A good model for solubility limitation is at the release boundary of the volume, for mixed volumes.

4 Solid Dissolution

Dissolution occurs after some kind of alteration and degradation.

Alteration is when some chemical reaction changes the characteristics of a material.

Give glass example.

Show that there are physical dissolution models being generated.

Waste form dissolution results may take place at a constant rate, a time dependent rate, or a physical model dependent on both static and dynamic parameters such as temperature, water velocity, or pH.

4.1 Waste Package Failure

Waste package failure is described best by a probability distribution such that a few packages can fail discretely at any one time.

It's also possible that a good model would fail all of the waste packages fractionally at some rate. This will affect both the water going in and the water coming out, but it's not clear to me how much this will affect those things.

5 Solute Transport in Porous Media

5.1 Derivation [7, 8]

Here we shall derive the general equation for solute transport in heterogeneous porous media.

As discussed in previous sections, mass transport by advection is driven by bulk water velocity, diffusion is the result of random thermal motion and tends to drive solutes down concentration gradients, and hydraulic dispersion results from heterogeneities in the water velocity field and drives accelerated mixing at the head of advection fronts.

Fundamentally, the effect of these phenomena on mass transport is captured by the conceptual mass conservation expression

$$\text{In} - \text{Out} = \text{Change in Storage} \tag{56}$$

which describes the mass balance within a reference volume of porous media.

By rearranging equation 56 and defining incoming and outflowing fluxes in a control volume, solute transport in a permeable medium of homogeneous porosity can be written (as in Schwartz and Zhang [1])

$$\frac{\partial nC}{\partial t} = -\nabla \cdot (F_c + F_{dc} + F_d) + m \tag{57}$$

where

$$
\begin{aligned}
n = {}& \text{solute accessible porosity } [\%] \\
C = {}& \text{concentration } [kg \cdot m^{-3}] \\
t = {}& \text{time } [s] \\
F_c = {}& \text{advective flow } [kg \cdot m^{-2} \cdot s^{-1}] \\
= {}& nvC \\
F_{dc} = {}& \text{dispersive flow } [kg \cdot m^{-2} \cdot s^{-1}] \\
= {}& \alpha nv \nabla C \\
F_d = {}& \text{diffusive flow } [kg \cdot m^{-2} \cdot s^{-1}] \\
= {}& nD_e \nabla C \\
m = {}& \text{solute source } [kg \cdot m^{-3} \cdot s^{-1}].
\end{aligned}
$$

In the expressions above,

$$
\begin{aligned}
v = {}& \text{advective velocity } [m \cdot s^{-1}] \\
\alpha = {}& \text{dispersivity } [m] \\
D_e = {}& \text{effective diffusion coefficient } [m^2 \cdot s^{-1}]
\end{aligned}
$$

and

$$n \cdot v = \text{Darcy flux } [m \cdot s^{-1}]. \tag{58}$$

The method by which the dominant solute transport mode (diffusive or advective) is determined for a particular porous medium is by use of the dimensionless Peclet number,

$$
\begin{aligned}
Pe &= \frac{nvL}{\alpha nv + D_e}, \tag{59} \\
&= \frac{\text{advective rate}}{\text{diffusive rate}}
\end{aligned}
$$

where

$$L = \text{transport distance } [m].$$

For a high Pe number, advection is the dominant transport mode, while diffusive or dispersive transport dominates for a low Pe number. If one of these terms can be neglected, the solution is simplified.

Otherwise, the analytical expression in equation (57) will be the foundation of simplification by regression analyses for the radionuclide transport interface between components of the repository system model representing permeable porous media.

It is customary to define the combination of molecular diffusion and mechanical mixing as the dispersion tensor, D, such that the mass conservation equation becomes:

$$\nabla\left(nD\nabla C\right) - \nabla\left(nv\right) = \frac{\partial(nC)}{\partial t} \tag{60}$$

Adding sorption, by accounting for a change in mass storage,

$$\nabla\left(nD\nabla C\right) - \nabla\left(nv\right) = \frac{\partial(nC)}{\partial t} + \frac{\partial(s\rho_b)}{\partial t} \tag{61}$$

where

$$s = \text{sorbed phase concentration}$$
$$\rho_b = \text{bulk (dry) density } [kg/m^3].$$

If it is assumed that sorption can be approximated as a linear isotherm, reversible reaction,

$$\nabla\left(nD\nabla C\right) - \nabla\left(nv\right) = \frac{\partial(nC)}{\partial t} + \frac{\partial(s\rho_b)}{\partial t} \tag{62}$$

where

$$K_d = \text{species distribution coefficient.}$$
$$\tag{63}$$

This becomes

$$\nabla\left(nD\nabla C\right) - \nabla\left(nv\right) = \frac{\partial(nC)}{\partial t} + \frac{\partial(K_dC\rho_b)}{\partial t} \tag{64}$$

which, rearranged, gives

$$\nabla\left(nD\nabla C\right) - \nabla\left(nv\right) = \frac{\partial}{\partial t}\left(nC + K_dC\rho_b\right) \tag{65}$$

$$\nabla\left(nD\nabla C\right) - \nabla\left(nv\right) = \frac{\partial}{\partial t}\left(nC\left(1 + \frac{K_d\rho_b}{n}\right)\right). \tag{66}$$

In equations (66) it is clear that the storage term can be simplified with a retardation factor, such that if

$$R_f = \text{retardation factor} \tag{67}$$

$$= 1 + \frac{\rho_b K_d}{n} \tag{68}$$

then equation (**??**) can be written

$$\nabla\left(nD\nabla C\right) - \nabla\left(nv\right) = R_f\frac{\partial(nC)}{\partial t} \tag{69}$$

For uniform flow, the dispersion tensor, D, in equation 38 gives

$$D_x \frac{\partial^2 C}{\partial x^2} + D_y \frac{\partial^2 C}{\partial y^2} + D_z \frac{\partial^2 C}{\partial z^2} + v_x \frac{\partial C}{\partial x} = R_f \frac{\partial C}{\partial t}. \tag{70}$$

A special case of uniform flow, no flow, simplifies to the diffusion equation,

$$D_x \frac{\partial^2 C}{\partial x^2} + D_y \frac{\partial^2 C}{\partial y^2} + D_z \frac{\partial^2 C}{\partial z^2} = R_f \frac{\partial C}{\partial t}. \tag{71}$$

Solutions to these equations can be categorized by their boundary conditions. The first, or Dirichlet type boundary conditions define a specified species concentration on some section of the boundary of the representative volume,

$$C(x, y, z, t) = C_0(x, y, z, t) \ \text{ for } \ (x, y, z) \in \Gamma. \tag{72}$$

The second type or Neumann type boundary conditions describe a full set of concentration gradients at the boundary of the domain

$$\frac{\partial C(\vec{r}, t)}{\partial r} = nD\vec{J} \ \text{ for } \vec{r} \in \Gamma. \tag{73}$$

where

$$\vec{r} = \text{ position vector}$$
$$\Gamma = \text{ domain boundary}$$
$$\vec{J} = \text{ solute mass flux } [kg/m^2 \cdot s].$$

The third, Cauchy, defines a solute flux along a boundary,

$$vC(X, y, z, t) - D_x \frac{\partial C(x, y, z, t}{\partial x} = vg(x, y, z, t) \ \text{ for } \ (x, y, z) \in \Gamma \tag{74}$$

where

$$g(x, y, z, t) = \text{ arbirary flux profile} \tag{75}$$

this can also be written

$$-nD_{ij} \frac{\partial C}{\partial x_j} n_i + q_i n_i C = q_i n_i C_0. \tag{76}$$

5.2   Useful Known Solutions

Various solutions to the advection dispersion equation in Equation (70) have been published for both the first and third types of boundary conditions. The third, Cauchy type, is mass conservative, and will be the primary kind of boundary condition used at the source for this model.

5.2.1   One Dimensional Semi-Infinite Solution

The conceptual model in Figure 7 represents solute transport in one dimension with unidirectional flow and a semi-infinite boundary condition in the positive flow direction.

$$-D\frac{\partial C}{\partial x}\Big|_{x=0} + vC = \begin{cases} vC_0 & t < t_0 \\ 0 & t > t_0 \end{cases} \qquad\qquad \frac{\partial C}{\partial x}\Big|_\infty = 0$$

$$C(x,0) = C_i$$

$x = 0$ $\qquad\qquad\qquad\qquad\qquad\qquad\qquad\qquad\qquad \infty$

Figure 6: Case I, a one dimensional, semi-infinite model.

With the boundary conditions

$$-D\frac{\partial C}{\partial x}\Big|_{x=0} + v_x c = \begin{cases} vC_0 & (0 < t < t_0) \\ 0 & (t > t_0) \end{cases} \tag{77}$$

$$\frac{\partial C}{\partial x}\Big|_{x=\infty} = 0 \tag{78}$$

and the initial condition

$$C(x,0) = C_i, \tag{79}$$

the solution is given as

$$C(x,t) = \begin{cases} C_i + (C_0 - C_i)\, A(x,t) & 0 < t < t_0 \\ C_i + (C_0 - C_i)\, A(x,t) - C_0 A(x, t - t_0) & t > t_0 \end{cases} \tag{80}$$

where

$$A(x,t) = \frac{1}{2}\,\mathrm{erfc}\left[\frac{Rx - vt}{2\sqrt{DRt}}\right] + \sqrt{\frac{v^2 t}{\pi DR}}\, e^{-\frac{(Rx - vt)^2}{4DRt}}. \tag{81}$$

5.2.2 One Dimensional Semi-Infinite Solution with Discrete Source

The conceptual model in Figure 7 represents solute transport in one dimension with unidirectional flow and a semi-infinite boundary condition in the positive flow direction.
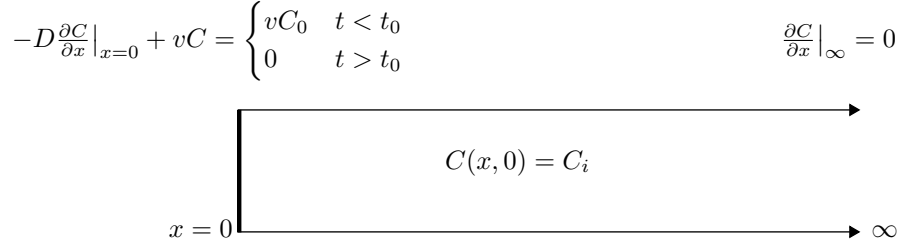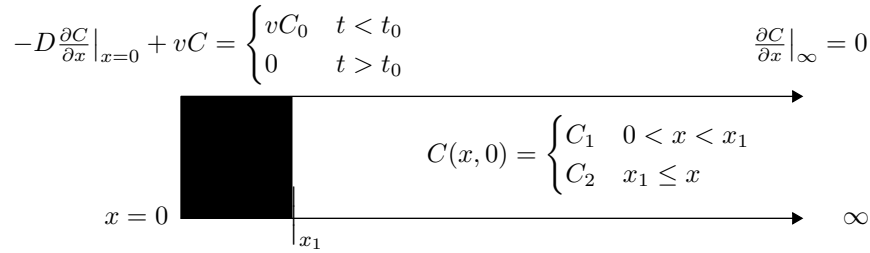
$$-D\frac{\partial C}{\partial x}\Big|_{x=0} + vC = \begin{cases} vC_0 & t < t_0 \\ 0 & t > t_0 \end{cases} \qquad\qquad \frac{\partial C}{\partial x}\Big|_\infty = 0$$

$$C(x,0) = \begin{cases} C_1 & 0 < x < x_1 \\ C_2 & x_1 \le x \end{cases}$$

$x = 0$ $\qquad\qquad\qquad\qquad\qquad\qquad\qquad\qquad\qquad \infty$

x_1

Figure 7: Case II, a one dimensional, semi-infinite model.

With the boundary conditions

$$-D\frac{\partial C}{\partial x}\Big|_{x=0} + v_x c = \begin{cases} vC_0 & (0 < t < t_0) \\ 0 & (t > t_0) \end{cases} \tag{82}$$

$$\frac{\partial C}{\partial x}\Big|_{x=\infty} = 0 \tag{83}$$

and the initial condition

$$C(x, 0) = C_i, \tag{84}$$

the solution is given as

$$C(x, t) = \begin{cases} C_i + (C_0 - C_i) A(x, t) & 0 < t < t_0 \\ C_i + (C_0 - C_i) A(x, t) - C_0 A(x, t - t_0) & t > t_0 \end{cases} \tag{85}$$

where

$$A(x, t) = \frac{1}{2} \operatorname{erfc} \left[\frac{Rx - vt}{2\sqrt{DRt}} \right] + \sqrt{\frac{v^2 t}{\pi DR}} e^{-\frac{(Rx - vt)^2}{4DRt}}. \tag{86}$$

5.2.3 One Dimensional Finite Solution

The conceptual model in Figure 8 represents solute transport in one dimension with unidirectional flow and a finite boundary condition in the positive flow direction.
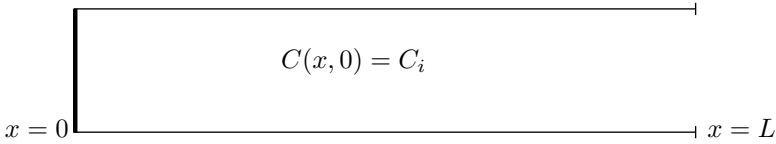
$$-D\frac{\partial C}{\partial x}\Big|_{x=0} + vC = \begin{cases} vC_0 & t < t_0 \\ 0 & t > t_0 \end{cases} \qquad\qquad \frac{\partial C}{\partial x}\Big|_{L} = 0$$

$$C(x, 0) = C_i$$

$$x = 0 \qquad\qquad\qquad\qquad\qquad\qquad x = L$$

Figure 8: Case III, a one dimensional, finite model.

With the boundary conditions

$$-D\frac{\partial C}{\partial x}\Big|_{x=0} + v_x c = \begin{cases} vC_0 & (0 < t < t_0) \\ 0 & (t > t_0) \end{cases} \tag{87}$$

$$\frac{\partial C}{\partial x}\Big|_{x=L} = 0 \tag{88}$$

and the initial condition

$$C(x, 0) = C_i, \tag{89}$$

the solution is given as

$$C(x, t) = \begin{cases} C_2 + (C_1 - C_2) A(x, t) + (C_0 - C_1) B(x, t) & 0 < t < t_0 \\ C_2 + (C_1 - C_2) A(x, t) + (C_0 - C_1) B(x, t) - C_0 B(x, t - t_0) & 0 < t < t_0 \end{cases} \tag{90}$$

where

$$A(x,t) = \frac{1}{2}\operatorname{erfc}\left[\frac{R(x-x_1)-vt}{2\sqrt{DRt}}\right] + \sqrt{\frac{v^2t}{\pi DR}}e^{\left[\frac{vx}{D}-\frac{R}{4Dt}(x+x_1+\frac{vt}{R})^2\right]}$$
$$-\frac{1}{2}\left[1+\frac{v(x+x_1)}{D}+\frac{v^2t}{DR}\right]e^{\frac{vx}{D}}\operatorname{erfc}\left[\frac{R(x+x_1)+vt}{2\sqrt{DRt}}\right] \tag{91}$$

$$B(x,t) = \frac{1}{2}\operatorname{erfc}\left[\frac{Rx-vt}{2\sqrt{DRt}}\right] + \sqrt{\frac{v^2t}{\pi DR}}e^{-\left[\frac{(Rx-vt)^2}{4DRt}\right]}$$
$$-\frac{1}{2}\left[1+\frac{vx}{D}+\frac{v^2t}{DR}\right]e^{\frac{vx}{D}}\operatorname{erfc}\left[\frac{Rx+vt}{2\sqrt{DRt}}\right]. \tag{92}$$

5.2.4 Three Dimensional Semi-Infinite Solution

The conceptual model in Figure 9 represents solute transport in three dimensions with unidirectional flow and a semi-infinite boundary condition in the positive flow direction.
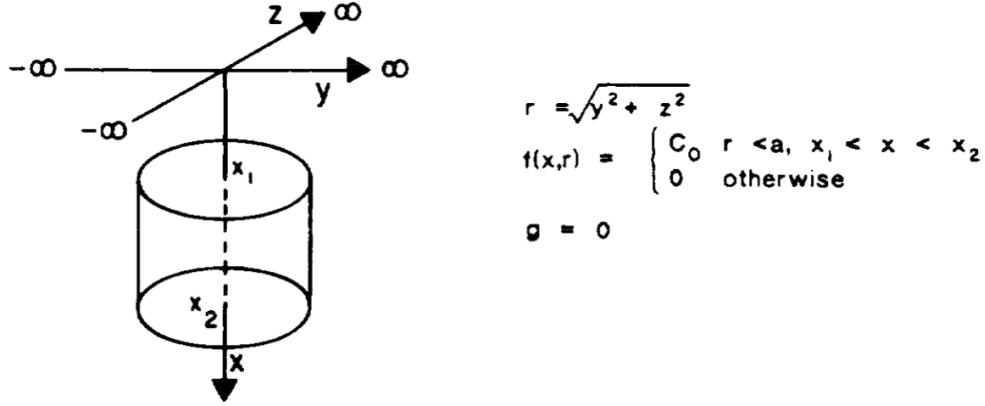

Figure 9: Case IV, a three dimensional, semi-infinite model.

With the boundary conditions

$$-D\frac{\partial C}{\partial x}\Big|_{x=0} + v_x c = \begin{cases} vC_0 & (0 < t < t_0) \\ 0 & (t > t_0) \end{cases} \tag{93}$$

$$\frac{\partial C}{\partial x}\Big|_{x=L} = 0 \tag{94}$$

and the initial condition

$$C(x,0) = C_i, \tag{95}$$

the solution is given as

$$C(x,t) = C_0 \int_0^a \Lambda_6(t)\Xi(\rho,t)d\rho + \frac{\lambda}{2R}\int_0^t\int_0^\infty \Xi(\rho,\tau)\Lambda_4(\tau)d\rho d\tau \tag{96}$$

where

$$\Xi(\rho, \tau) = \frac{\rho R}{4 D_r \tau} e^{\left(-\frac{R(r^2 + \rho^2)}{4 D_r \tau}\right)} I_0\left(\frac{R r \rho}{2 D_r \tau}\right), \tag{97}$$

$$I_0(x) = \sum_{m=0}^{\infty} \frac{1}{(m!)^2} \left(\frac{x}{2}\right)^{2m} \tag{98}$$

$$\Lambda_4(\tau) = \mathrm{erfc}\left[\frac{v\tau - Rx}{\sqrt{4 R D_x t}}\right] + \left(1 + \frac{v}{D_x}(x + v\tau/R)\right) e^{\left(\frac{vx}{D_x}\right)} \mathrm{erfc}\left[\frac{Rx + v\tau}{\sqrt{4 R D_x t}}\right]$$
$$- \sqrt{\frac{4 v^2 \tau}{\pi R D_x}} e^{-\left(\frac{(Rx - v\tau)^2}{\sqrt{4 R D_x \tau}}\right)}, \tag{99}$$

and

$$\Lambda_6(t) = e^{\frac{vx}{D_x}} \left[\left(1 + \frac{v}{D_x}(x + x_1 + vt/R)\right) \mathrm{erfc}\left[\frac{R(x + x_1) + vt}{\sqrt{4 R D_x t}}\right] \right.$$
$$- \left(1 + \frac{v}{D_x}(x_1 + x_2) + vt/R\right) \mathrm{erfc}\left[\frac{R(x + x_2) + vt}{\sqrt{4 R D_x t}}\right]\right]$$
$$+ \mathrm{erfc}\left[\frac{R(x - x_2) - vt}{\sqrt{4 R D_x t}}\right] i - \mathrm{erfc}\left[\frac{R(x - x_1) - vt}{\sqrt{4 R D_x t}}\right]$$
$$+ \sqrt{\frac{4 v^2 t}{\pi R D_x}} e^{\left(\frac{vx}{D_x}\right)} \left[e^{-\left(\frac{[R(x + x_2) + vt]^2}{4 R D_x t}\right)} - e^{-\left(\frac{[R(x + x_1) + vt]^2}{4 R D_x t}\right)}\right]. \tag{100}$$

Clearly, this functional form of the solution could be onerous to code. Thus, other solutions are being considered, including the lumped parameter model of radionuclide transport.

5.2.5 Lumped Parameter Model

For systems in which the flow can be assumed constant, it is possible to model a system of volumes as a connected lumped paramter models (Figure 10).

$$C_{in0} \qquad C_{out0} = C_{in1} \qquad C_{out1} = C_{in2} \qquad C_{out2} = C_{in3} \qquad C_{out3}$$
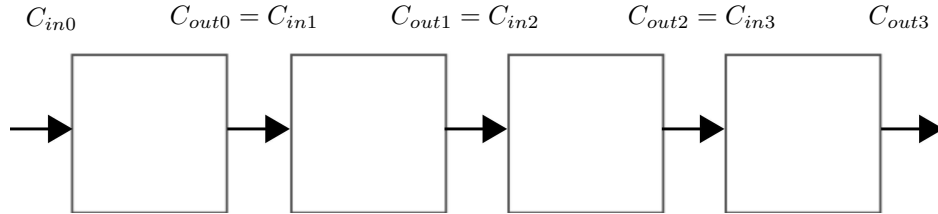
Figure 10: A system of volumes can be modeled as lumped parameter models in series.

The method by which each lumped parameter component is modeled is according to a relationship between the incoming concentration, $C_{in}(t)$, and the outgoing concentration, $C_{out}(t)$,

$$C_{out}(t) = \int_{-\infty}^{t} C_{in}(t') g(t - t') e^{-\lambda(t - t')} dt' \tag{101}$$

equivalently

$$C_{out}(t) = \int_{0}^{\infty} C_{in}(t - t') g(t') e^{-\lambda t'} dt' \tag{102}$$

where

$$
\begin{aligned}
t' &= \text{ time of entry } [s] \\
t - t' &= \text{ transit time } [s] \\
g(t - t') &= \text{ response function, a.k.a. transit time distribution}[-]] \\
\lambda &= \text{ radioactive decay constant, 1 to neglect}[s^{-1}]
\end{aligned}
\tag{103}
$$

Selection of the response function is usually based on experimental tracer results in the medium at hand. However, some functions used commonly in chemical engineering applications include the Piston Flow Model (PFM),

$$
g(t') = \delta(t' - t_t))
\tag{104}
$$

the Exponential Model (EM)

$$
g(t') = \frac{1}{t_t} e^{-\frac{t'}{t_t}}
\tag{105}
$$

and the Dispersion Model (DM),

$$
g(t') = \frac{\left[\left(\frac{t \pi t'}{t_t Pe} \right) \left(\frac{1}{t'} \right) e^{-\left(1 - \frac{t'}{t_t}\right)^2} \right]}{\frac{4t'}{t_t Pe}},
\tag{106}
$$

where

$$
\begin{aligned}
Pe &= \text{ Peclet number } [-] \\
t_t &= \text{ mean tracer age } [s] \\
&= t_w \text{ if there are no stagnant areas} \\
t_w &= \text{ mean residence time of water}[s] \\
&= \frac{V_m}{Q} \\
&= \frac{x}{v_w} \\
&= \frac{x n_e}{v_f}
\end{aligned}
$$

in which

$$
\begin{aligned}
V_m &= \text{ mobile water volume}[m^3] \\
Q &= t_w \text{ volumetric flow rate } [m^3/s \\
v_w &= \text{ mean water velocity}[m/s] \\
v_f &= \text{ Darcy Flux}[m/s]
\end{aligned}
$$

and

$$
n_e = \text{ effective porosity}[\%].
$$

The latter of these, the Dispersion Model satisfies the one dimensional advection-dispersion equation, and is therefore the most physically relevant for this application.

The solutions to these for constant concentration at the source boundary give

$$
C(t) = \begin{cases}
PFM & C_0 e^{\lambda t_t} \\
EM & \frac{C_0}{1 + \lambda t_t} \\
DM & \frac{C_0 e^{\frac{Pe}{\sqrt{\left(1 - (1 + \frac{4\lambda t_t}{Pe})\right)}}}}{2}
\end{cases}
\tag{107}
$$

5.3   Buffer Model

The buffer component is a long, but finite, cylindrical geometry with a series of cylindrical waste packages within it. These waste packages can be modeled as a single line source, a series of cylinders, or a series of points. If they are modeled as a single line source, the flux emitted by the line boundary will be expressed, in one dimension, as

$$vC_0 = v \sum_{p=1}^{p=P} C_p \tag{108}$$

where

$$
\begin{aligned}
vC_0 &= \text{ constant solute flux during the timestep } [kg/m^3/s < +check+ >] \\
C_p &= \text{ available concentration at waste package p } [kg/m^3] \\
P &= \text{ number of waste packages within the buffer cylinder } [\#].
\end{aligned}
$$

$$\tag{109}$$

If the line source is chosen, the one dimensional models in cases I, II, or III can be used, as can the lumped parameter model. In these cases, the flux at the inner buffer radius is described by the cylindrical source waste form. The concentration gradient can be defined as zero either at the outer buffer radius, or at infinity. These approximate solutions may not give significantly different results for many situations, but each will be investigated.

If alternatively, the waste packages are modeled as discrete cylinders, the buffer must also be modeled as a discrete cylinder within the broader, far field semi-infinite medium.

5.4   Far Field Solution

The three dimensional solution with a cylindrical source of constant flux is the most appropriate treatment for the boundary conditions of the far field component. With the length of the tunnels and outer radial dimension of the buffer, the far field can be modeled as a semi-infinite medium and the flux at the location of the perceived aquifer can be queried. In case IV, the flux escaping the cylindrical buffer will be an output of the buffer model. The concentration gradient is zero at infinity.

The far field may also be modeled as a lumped parameter system. It may either be modeled as a single lumped parameter component or as a series of smaller dimension lumped parameter components. For a homogeneous medium, the difference in accuracy between these two approaches will be zero, though for heterogeneous media for which local parameters are known, accuracy can be increased by decreased lumped parameter component length.

A corresponding one dimensional solution will be shown and its applicability will be justified.

The sides and bottoms of a two dimensional far field model are typically taken to be no flow boundaries. This garuntees that the only sources and sinks of water occur at the top surface, the aquifer.

5.5   Modeling Strategy

Comments on how to model this solution numerically will be discussed.

6   Analytical Models of Heat Transport

Heat transfer in saturated repository concepts will be dominated by conductive heat transfer. In a saturated closed system, very few air gaps will exist. For this reason, heat transfer by radiation is likely to be negligible. Similarly, since water velocities are comparatively low, heat transfer by mass transfer or by convection will be small relative to conduction.

A discussion of analytical models of heat transport follows that quantifies these modes of heat transfer and addresses their applicability to the model at hand.

6.1 Conduction

Conductive heat transfer occurs as a result of a temperature gradient. Heat flows diffusively from the hotter material to the cooler material over time and steadily approaches thermal equilibrium. The general form of the conduction equation can be expressed

$$\nabla^2 T + \frac{q'''}{k} = \frac{1}{\alpha}\frac{\partial T}{\partial t}, \tag{110}$$

which with no heat source becomes the transient Fourier equation,

$$\nabla^2 T = \frac{1}{\alpha}\frac{\partial T}{\partial t}, \tag{111}$$

or to the Laplace equation in steady state,

$$\nabla^2 T = 0. \tag{112}$$

Replacing the source gives the steady state Poisson equation,

$$\nabla^2 T + \frac{q'''}{k} = 0. \tag{113}$$

An areal heat flux, $q''[W/m^2]$ can be derived from an integration of Poisson's equation (113) and expressed in terms of the thermal conductivity of the material, $k[W/m \cdot^\circ K]$, and the temperature gradient $\nabla T[K/m]$ by the expression

$$q'' = -k\nabla T. \tag{114}$$

For the one dimensional case, equation 114 can be reduced using a finite difference approximation. For a body at x_1 with temperature T_1 and a body with temperature T_2 at position x_2,

$$q''_x = -k_x \frac{dT}{dx} \tag{115}$$

$$= -k_x \frac{(T_1 - T_2)}{x_1 - x_2}. \tag{116}$$

6.2 Convection

Convective heat transfer occurs advectively in accordance with fluid movement. Convection can be expressed as

$$\dot{q} = -hA\Delta T \tag{117}$$

where

$$\dot{q} = \text{ Thermal energy } [W]$$
$$h = \text{ the heat transfer coefficient } [W \cdot m^{-2}]$$
$$A = \text{ the surface area of heat transfer } [m^2]$$

6.3 Radiation

Heat transfer by radiation is the result of the emission of electromagnetic waves. Planck black body radiation is analytically described, using σ, the Stefan-Boltzmann constant as

$$\dot{q} = \sigma A_1 F_{1\rightarrow 2}(T_1^4 - T_2^4) \tag{118}$$

where

$$\sigma = 5.670373 \times 10^8 \, [W/m^2 K^4]$$

and

$$F_{1\rightarrow 2} = \begin{cases} \epsilon_1 & \text{for a point source,} \\ \frac{1}{\frac{1}{\epsilon_1} + \frac{1}{\epsilon_2} - 1} & \text{for parallel plates,} \\ \frac{2\pi r_1 L}{\frac{1}{\epsilon_1} + \frac{1}{\epsilon_2} - 1} \frac{r_1}{r_2} & \text{for concentric cylinders} \end{cases} \tag{119}$$

where

$$\epsilon_i = \text{emissivity of surface i } [-]$$
$$r_i = \text{radius of cylinder i } [m]$$
$$L = \text{cylinder length } [m].$$

6.4 Mass Transfer

Heat transfer by mass transfer is straightforward, resulting in the change in temperature in adjacent volumes as a result of matter movement. If the specific heat capacity of the transferred mass can be expressed as c_p, then the heat transfer is simply,

$$\dot{q} = \dot{m} c_p \left(T_j - T_i \right) \tag{120}$$

where

$$c_p = \text{ specific heat capacity } [J/kg^\circ K].$$

7 Thermal Effects on Repository Performance

7.1 Decay Heat

The radioactive decay of isotopes in spent nuclear fuel produces heat. The activity of a radioactive sample is equal to the sum of the activities of each of its constituent isotopes. In Bequerels, or disintegrations per second, radioactivity is expressed in terms of the decay constant,

$$\begin{aligned} A_{sample} &= \sum_i^n A_i \\ &= \sum_i^n \lambda_i N_i \end{aligned} \tag{121}$$

where

$$
\begin{aligned}
A_{sample} &= \text{ disintegrations per second in the sample } [Bq] \\
A_i &= \text{ disintegrations per second due to isotope i } [Bq] \\
n &= \text{ the number of isotopes species in the sample } [-] \\
\lambda_i &= \text{ the decay constant of isotope i } [Bq] \\
N_i &= \text{ the number of atoms of isotope i in the sample } [-].
\end{aligned}
$$

The specific activity of a sample, defined as the activity per unit mass, is then

$$
\begin{aligned}
SA &= \sum_i^n SA_i \\
&= \sum_i^n \frac{\lambda_i N_{Av}}{M_i}
\end{aligned}
\tag{122}
$$

where

$$
\begin{aligned}
N_{Av} &= \text{ Avogadro's number } 6.022 \times 10^{-23} [mol^{-1}] \\
M_i &= \text{ Atomic weight of isotope i } [g \cdot mol^{-1}].
\end{aligned}
$$

The specific power, in $MeV \cdot s^{-1} \cdot g^{-1}$ generated by that sample at that second employs an isotope specific energy per disintegration factor,

$$
p = \sum_i^n \frac{E_i \lambda_i N_{Av}}{M_i} [MeV \cdot s^{-1} \cdot g^{-1}]
\tag{123}
$$

where

$$
E_i = \text{ Average energy release per disintegration of isotope i } [MeV].
$$

Energy in MeV can be converted to Watts by the conversion factor,

$$
1[MeV \cdot s^{-1}] = 1.6 \times 10^{-13} [W],
\tag{124}
$$

such that the total heat of a sample can be expressed as

$$
Q = \sum_i^n 1.6 \times 10^{-13} \frac{I_i E_i \lambda_i N_{Av}}{M_i} [W]
\tag{125}
$$

where

$$
I_i = \text{ the inventory of isotope i in the sample } [g].
$$

This gives only the instantaneous decay heat production of a sample. To arrive at long term decay heat curves, decay heat contributions from the chain of daughter products must be calculated as well. Typically, the latter calculation is done by detailed depletion codes.

7.2 Capacity Determination for Arbitrary Waste Form

Modeling heat will require a transient model capable of arriving at a heat based capacity quickly for an arbitrary waste stream.

Superposition of known, mass normalized, decay heat evolution curves will result in a heat source term curve for the waste form for future times,

$$Q(t) = \sum_{i=0}^{n} I_i q_i(t) \tag{126}$$

where

$$
\begin{aligned}
Q(t) &= \text{Total waste form heat at time t } [W] \\
i &= \text{Index of the isotope species } [-] \\
n &= \text{Number of isotopes in the waste form } [-] \\
I_i &= \text{Inventory of isotope i in the waste form } [g] \\
q_i(t) &= \text{Heat due to one gram of i after time t } [W \cdot g^{-1}].
\end{aligned}
$$

Determination of $q_i(t)$ must come from tabulated and mass normalized results of depletion calculations, or otherwise available decay heat curves for heat generating nuclides and their daughters.

With this algebraic calculation of $Q(t)$, the heat evolution of the waste form, there are a number of approximations that may assist in estimating repository capacity for that waste stream while holding the package spacing constant.

7.2.1 Areal Power Density Approximation

A uniform areal power density can be used to determine appropriate repository loading for arbitrary waste streams.
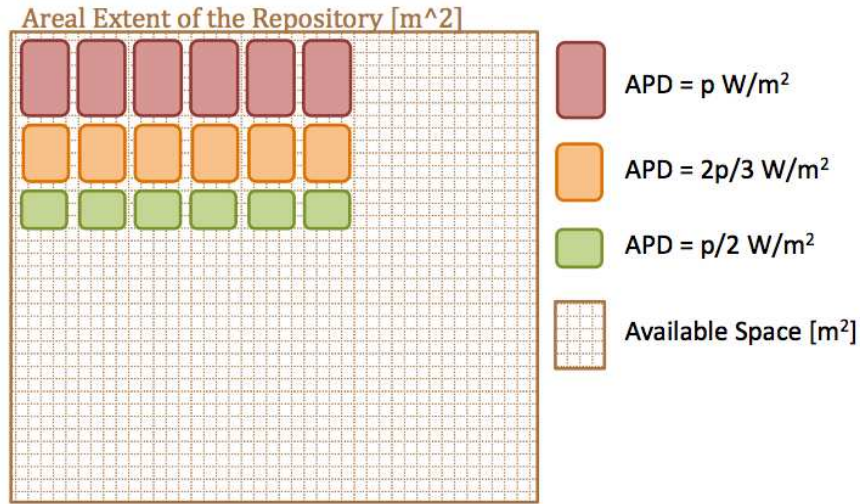

Figure 11: The areal power density loading scheme pictured above is not constrained by uniform spacing and takes advantage of denser spacing made possible by cooler packages (green).

In the areal power density loading schem, loading is subject to the constraints,

$$P_{tot} \leq P_{max} \tag{127}$$
$$APD_i \leq APD_{max} \tag{128}$$

where

$$P_{max} = A \cdot APD_{max} \tag{129}$$

$$P_{tot} = \sum_{i=0}^{N} n_i P_i \tag{130}$$

$$P_i = \text{power of package i}$$
$$n_i = \text{ith package}$$
$$N = \text{number of packages}$$
$$APD_{max} = \text{max areal power density}$$

7.2.2 Peak Heat Approximation

One conservative estimation could assume that $Q(t)$ is a constant heat flux, Q_{max} equal to the peak of calculated waste form heat evolution, $max(Q(t))$. Assuming $Q(t) = Q_{max} \forall t$, it would be possible to directly refer to previously calculated repository evolution results based on constant heat and spacing.
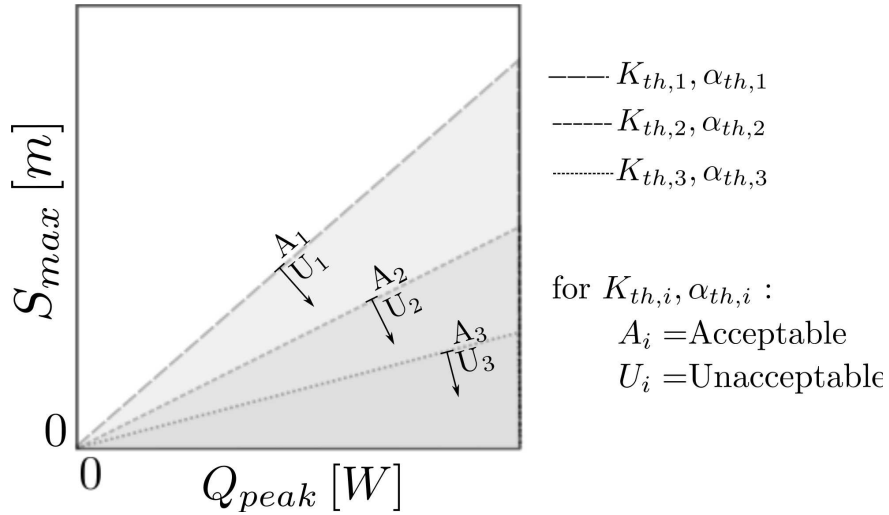

Figure 12: Above the empirically determined acceptance threshold for a certain K_{th}, α_{th} combination, the arbitrary waste stream is likely to be acceptable. Below it, the waste stream is likely to be unacceptable.

7.2.3 Heat Integral Approximation

Another approximation could develop the heat and spacing thresholds based on the integral of heat over time. Then, the time evolution of $Q(t)$ could be captured in the comparison.

One method in the literature not dissimilar to this is the Specific Temperature Integral by Jun Li[9].

A specific temperature integral may model the thermal source as linear along the emplacement paths, analagously to the areal power density method. However, a temperate integral takes account of heat transfer behavior in the rock, includes the effects of myriad SNF compositions, and gives the thermal integration over time for any specific location within the rock. Man-Sung Yim calls this the Specific Temperature Increase method[9].

In the same way that $Q(t)$, the heat flux from the waste streams, can be expressed as the superposition of the linear heat flux contributions of all the radionuclides in the waste, he temperature change expressed as a superposition of the temperature change contributions due to each radionuclide. Each radionuclide contributes in proportion to its decay heat generation and its weight fraction of the SNF. With information about isotopic composition of the SNF, the specific temperature increase can determine the maximum thermal
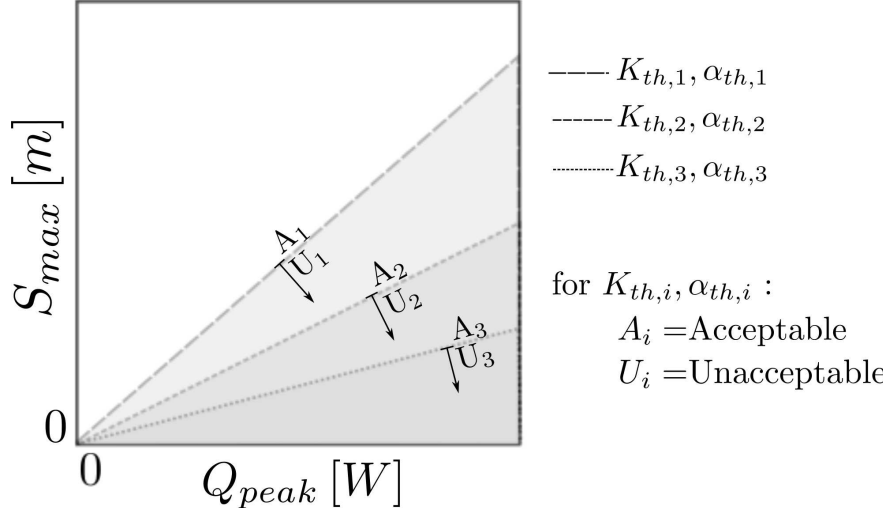
Figure 13: Above the empirically determined acceptance threshold for a certain K_{th}, α_{th} combination, the arbitrary waste stream is likely to be acceptable. Below it, the waste stream is likely to be unacceptable.

capacity of the repository in terms of tonnes/m. The length based accounting in $\frac{t}{m}$ is converted to $\frac{t}{Repository}$ by multiplication with the total emplacement tunnel length of the repository.

7.2.4 Effective Mean Decay Power

Introduced by Radel, Wilson et. al., the Specific Temperature Change method uses a linear approximation to arrive at the thermal loading density limit.

The mean decay constant of the radionuclide constituents in the waste form was compared to the thermal time constant in the rock.

When the thermal time constant of the rock is much shorter than the waste form decay package, the change in package wall temperature can be described by

$$q(t_0)\rho_{limit}C' = \Delta T_1 \tag{131}$$

where

$$\rho_{limit} = \frac{C_1}{Q_1}$$
$$C' = \text{Thermal constant } [-]$$
$$\Delta T = \text{Constant difference between } T_{lim} \text{ and } T_{amb}[°C]$$
$$T_{lim} = \text{Temperature limit } [°C]$$
$$T_{amb} = \text{Ambient rock temperature } [°C]$$

7.2.5 Thermal Half-Life Approximation

There is some notion of a thermal halflife in the literature.

7.3 Waste Stream Loading Density in Waste Forms

Either the user inputs the waste form loading density or the repository must determine how best to load the waste forms with waste .

7.4 Waste Package Loading Strategy in Repository

Either the user inputs the tunnel spacing and waste package spacing, or the repository must determine how best to space the tunnels and packages.

7.5 Solving for the Heat Evolution

This model should be capable of calculating the temperature field through the repository over time for a certain waste package layout.

References

[1] F. W. Schwartz and H. Zhang, *Fundamentals of ground water*. 2003.

[2] H. Wang and M. P. Anderson, *Introduction to groundwater modeling*. Freeman, 1982.

[3] R. C. Heath, *Basic ground-water hydrology*, vol. 2220. US Geological Survey, 1983.

[4] JrPol, "Diffusion.svg." http://upload.wikimedia.org/wikipedia/commons/1/12/Diffusion.svg, Sept. 2011.

[5] B. Mason, "Oxidation and reduction in geochemistry," *The Journal of Geology*, vol. 57, pp. 62–72, Jan. 1949.

[6] A. Hedin, "Integrated analytic radionuclide transport model for a spent nuclear fuel repository in saturated fractured rock," *Nuclear Technology*, vol. 138, no. 2, 2002.

[7] M. T. Van Genuchten, *Analytical solutions of the one-dimensional convective-dispersive solute transport equation*. No. 1661, US Dept. of Agriculture, Agricultural Research Service, 1982.

[8] F. J. Leij, T. H. Skaggs, and M. T. Van Genuchten, "Analytical solutions for solute transport in three-dimensional semi-infinite porous media," *Water resources research*, vol. 27, no. 10, p. 27192733, 1991.

[9] J. Li, M. S. Yim, and D. McNelis, "The specific temperature increase method for repository thermal analysis," *Transactions of the American Nuclear Society*, vol. 99, pp. 216 — 218, 2008.