Hydrologic Nuclide Transport Models in Cyder, A Geologic Disposal Software Library - 13328

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ABSTRACT

Component level and system level abstraction of detailed computational geologic repository models have resulted in four rapid computational models of hydrologic radionuclide transport at varying levels of detail. Those models are described, as is their implementation in Cyder, a software library of interchangeable radionuclide transport models appropriate for representing natural and engineered barrier components of generic geology repository concepts. A proof of principle demonstration was also conducted in which these models were used to represent the natural and engineered barrier components of a repository concept in a reducing, homogenous, generic geology. This base case demonstrates integration of the Cyder open source library with the Cyclus computational fuel cycle systems analysis platform to facilitate calculation of repository performance metrics with respect to fuel cycle choices.

INTRODUCTION

Radionuclide containment behavior of a geologic repository is a function of spent fuel and high level waste composition, which varies among alternative fuel cycles. For this reason, a generic disposal model capable of integration with a systems analysis framework is necessary to illuminate performance distinctions of candidate repository geologies, designs, and engineering components in the context of fuel cycle options.

A generic repository model appropriate for systems analysis must emphasize modularity and speed while providing modeling options at various levels of detail. Sensitivity analyses and abstraction efforts conducted to develop the models described in this work sought to capture the dominant physics of detailed repository behaviors so that abstracted models can be robustly and flexibly implemented in the Cyder disposal environment library without sacrificing the simulation speed required by the Cyclus fuel cycle simulator.

MODEL INTERFACES

The interfaces between the models are essential to the understanding of the models themselves. The interfaces define boundary conditions in a number of forms based on information available internally to the component implementation.

In a saturated, reducing environment, contaminants are transported by dispersion and advection. 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 [1, 2, 3]:

$$
\begin{aligned}
J &= J_{dis} + J_{adv} \\
&= -\theta(D_{mdis} + \tau D_m)\nabla C + \theta v C \\
&= -\theta D \nabla C + \theta v C
\end{aligned}
$$

which, for uniform flow in $\hat{k}$, is

$$= \left(-\theta D_{xx}\frac{\partial C}{\partial x}\right)\hat{\imath} + \left(-\theta D_{yy}\frac{\partial C}{\partial y}\right)\hat{\jmath} + \left(-\theta D_{zz}\frac{\partial C}{\partial z} + \theta v_z C\right)\hat{k}, \qquad \text{(Eq. 1)}$$

where

$$\begin{aligned}
J_{dis} &= \text{Total Dispersive Mass Flux } [kg/m^2/s] \\
J_{adv} &= \text{Advective Mass Flux } [kg/m^2/s] \\
\tau &= \text{Toruosity } [-] \\
\theta &= \text{Porosity } [\%] \\
D_m &= \text{Molecular diffusion coefficient } [m^2/s] \\
D_{mdis} &= \text{Coefficient of mechanical dispersivity} [m^2/s] \\
D &= \text{Effective Dispersion Coefficient } [m^2/s] \\
C &= \text{Concentration } [kg/m^3] \\
v &= \text{Fluid Velocity in the medium } [m/s].
\end{aligned}$$

Solutions to this equation can be categorized by their boundary conditions and those boundary conditions serve as the interfaces between components in the Cyder library of nuclide transport models.
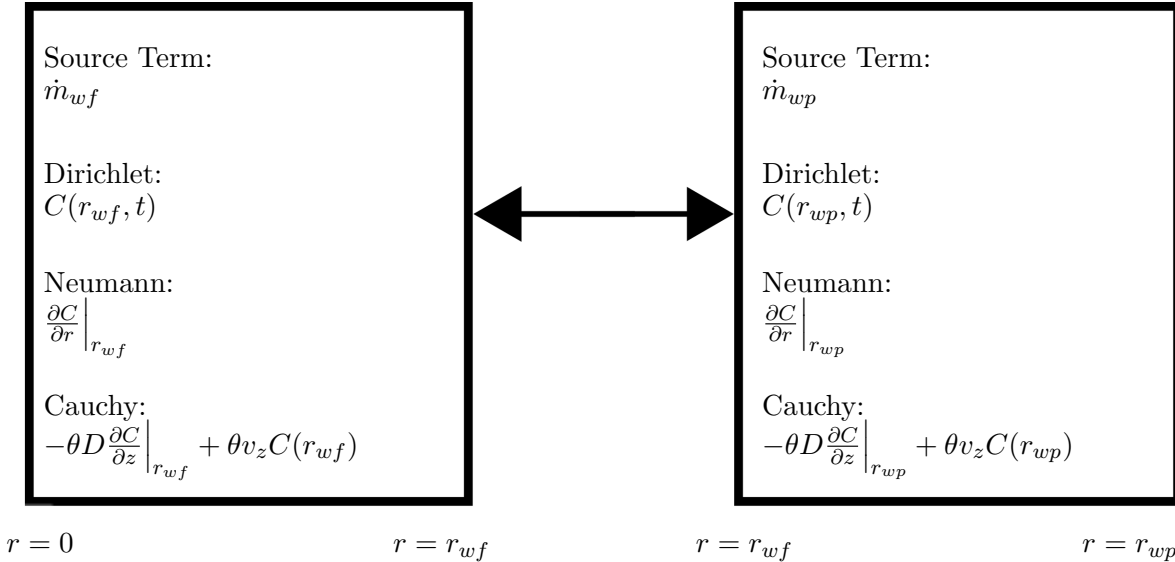

Figure 1: The boundaries between components (in this case, waste form (wf) and waste package (wp) components) are robust interfaces defined by Source Term, Dirichlet, Neumann, and Cauchy boundary conditions.

In addition to a specified source term (the zeroth type boundary condition, perhaps), the first, specified-head or Dirichlet type boundary conditions define a specified species concentration on some section of the boundary of the representative volume,

$$C(\vec{r}, t) = C_0(\vec{r}, t) \ \text{ for } \vec{r} \in \Gamma. \qquad \text{(Eq. 2)}$$

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

$$\frac{\partial C(\vec{r}, t)}{\partial r} = \theta D \vec{J}(t) \ \text{ for } \vec{r} \in \Gamma \tag{Eq. 3}$$

where

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

The third, head-dependent mixed boundary condition or Cauchy type, defines a solute flux along a boundary,

$$-D\frac{\partial C}{\partial z} + v_z C = v_z C(\vec{r}, t) \ \text{ for } \vec{r} \in \Gamma. \tag{Eq. 4}$$

The spatial concentration throughout the volume is sufficient to fully describe implementation of the following nuclide transport models within Cyder. This is supported by the implementation in which vertical advective velocity is uniform throughout the system and in which parameters such as the dispersion coefficient are known for each component. Since this is the case in Cyder, description of the Dirichlet condition is sufficient to fully define calculation of the Neumann and Cauchy type conditions.

DESCRIPTION OF THE MODELS

The results of this work consist of four models that are the product of an abstraction effort with more detailed tools. The analytic models modified by abstraction and implemented in Cyder include a degradation rate model, a mixed cell model, a response function model, and a one-dimensional solution (Leij et. al [4]) to the advection-dispersion equation.

Degradation Rate Radionuclide Transport Model

The degradation rate model, simulating the fractional degradation of the material containment properties, is the simplest of implemented models and is most appropriate for simplistic waste package failure modeling.

The materials that constitute the engineered barriers in a saturated repository environment degrade over time. The implemented model of this nuclide release behavior is based solely on a fractional degradation rate. This model incorporates the source term made available on the inner boundary into its available mass and defines the resulting boundary conditions at the outer boundary as solely a function of the degradation rate of that component.

This results in the following expression for the mass transfer, $m_{ij}(t)$, from cell i to cell j at time t :

$$\dot{m}_{ij}(t) = f_i(\cdots)m_i(t) \tag{Eq. 5}$$

where

$$
\begin{aligned}
\dot{m}_{ij} &= \text{ the rate of mass transfer from i to j } [kg/s] \\
f_i &= \text{ fractional degradation rate in cell i } [1/s] \\
m_i &= \text{ mass in cell i } [kg] \\
t &= \text{ time } [s].
\end{aligned}
$$

For a situation as in Cyder and CYCLUS, with discrete timesteps, the timesteps are assumed to be small enough to assume a constant rate m_{ij} over the course of the timestep. The mass transferred between discrete times t_{n-1} and t_n is thus a simple linear function of the transfer rate in (Eq. 5),

$$
\begin{aligned}
m_{ij}^n &= \int_{t_{n-1}}^{t_n} \dot{m}_{ij}(t')dt' \\
&= f_i(\cdots)m_i^{n-1}\left(t_n - t_{n-1}\right). \tag{Eq. 6}
\end{aligned}
$$

The concentration boundary condition must also be defined at the outer boundary to support parent components that utilize the Dirichlet boundary condition. For the degradation model, which incorporates no diffusion or advection, the concentration, C_{ij} at the boundary between cells i and j is the average concentration in the saturated pore volume,

$$
\begin{aligned}
C_{ij}^n &= \frac{m_i^n}{V_{vi}} \tag{Eq. 7} \\
&= \frac{\text{solute mass in cell i}}{\text{void volume in cell i}}.
\end{aligned}
$$

For the case in which all engineered barrier components are represented by degradation rate models, the source term at the outermost edge will be solely a function of the original central source and the degradation rates of the components.

To support parent components that utilize the Cauchy boundary condition, the degradation model assumes that the fluid velocity is constant across the cell as is the concentration. Thus,

$$-\theta_i D_{zz}\frac{\partial C}{\partial z}\hat{k} + \theta_i v_z C\hat{k} = \theta_{i0}v_0 C_0 \hat{k}$$

$$
\begin{aligned}
\theta_i &= \text{ porosity in cell i } [-] \\
D_{zz} &= \hat{k} \text{ component diffusion tensor component in } \hat{k} \text{ direction } [m^2/s] \\
C_i &= \text{ concentration in cell i } [kg/m^3] \\
v_z &= \text{ velocity in } \hat{k} \text{ direction } [m/s]
\end{aligned}
$$

reduces to

$$\theta_i^n v_z^n C_i^n = \theta_i^{n-1}v_z^{n-1}C_i^{n-1}. \tag{Eq. 8}$$

Mixed Cell Volume Radionuclide Transport Model

Slightly more complex and suited to representing waste form and buffer components, the mixed cell model incorporates solubility limited, congruent release under the influence of elemental solubility limits, sorption, diffusive behavior, and advective behavior. Abstraction results concerning the transition between primarily diffusive and primarily advective transport regimes were used for benchmarking and to iteratively improve accuracy in the development of this model.

A main nuclide transport component model used in this work is a mixed cell component module incorporating solubility and sorption effects as well as engineered material dissolution.

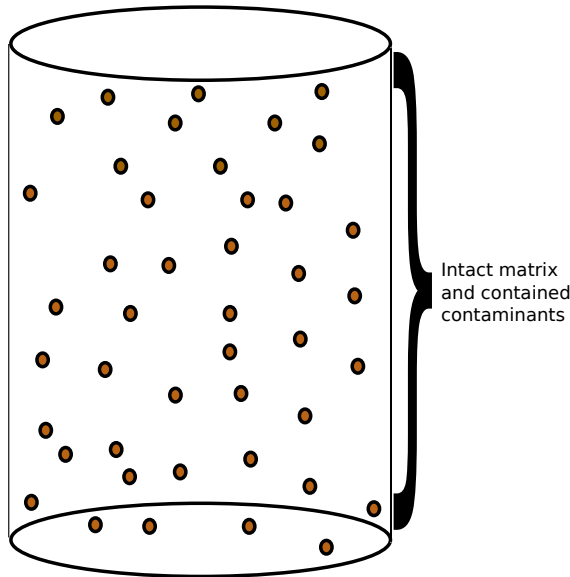A graphical representation of the mixed cell model is given in Figures 2 and 3.

Figure 2: The control volume contains an intact material matrix. Contaminants are unavailable to neighboring subcomponents until dissolution has begun.
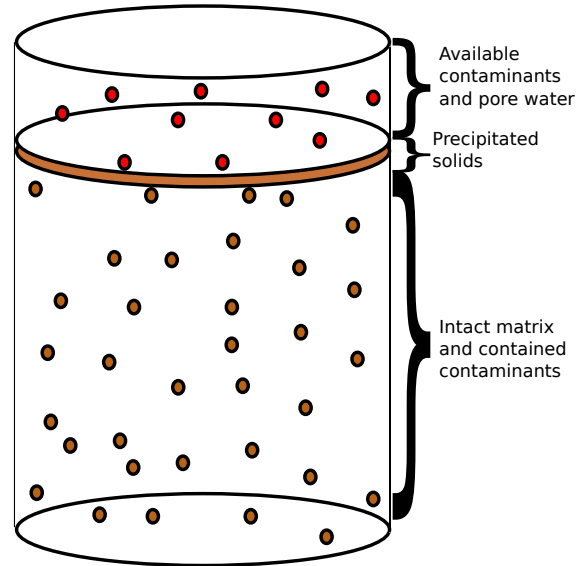
Figure 3: Once dissolution begins, the control volume contains a partially dissolved material matrix, contaminated pore water, and degraded and precipitated solids.

After some time degrading, the volume of free fluid can be expressed as

$$V_{ff}(t_n) = \theta V_T \int_{t_0}^{t_n} f(\cdots)dt. \tag{Eq. 9}$$

The volume of the intact matrix can be expressed as

$$V_{im}(t_n) = V_T - V_T \int_{t_0}^{t_n} f(\cdots)dt. \tag{Eq. 10}$$

Finally, the volume of the degraded and precipitated solids can be expressed as

$$V_{ps}(t_n) = (1 - \theta)V_T \int_{t_0}^{t_n} f(\cdots)dt. \tag{Eq. 11}$$

This model assumes that all net influx to the cell enters the free fluid rather than the intact matrix. The total volumetric contaminant concentration in the intact matrix, can be expressed as

$$C_{im}(t_n) = C_0 \qquad \text{(Eq. 12)}$$

$$= \frac{m_0}{V_{im}(t_0)} \qquad \text{(Eq. 13)}$$

where

$$m_0 = \text{ total initial mass.}$$

The resulting contaminant mass in the intact matrix at time t_n is

$$m_{im}(t_n) = C_0 V_{im}(t_n)$$

$$= C_0 V_T \left(1 - \int_{t_0}^{t_n} f(\cdots) dt \right). \qquad \text{(Eq. 14)}$$

The contaminant mass in the free fluid is just the initial pore water concentration times the free fluid volume plus the time integral of net influx to the cell such that

$$C_{ffT}(t_n) = \left[C_0 + \frac{\int_{t_0}^{t_n} \dot{m}_i(t')dt'}{V_{ff}(t_n)} \right] \qquad \text{(Eq. 15)}$$

and

$$m_{ffT}(t_n) = C_{ff}(t_n) V_{ff}(t_n)$$

$$= \left[C_0 + \frac{\int_{t_0}^{t_n} \dot{m}_i(t')dt'}{V_{ff}(t_n)} \right] V_{ff}(t_n)$$

$$= C_0 V_{ff}(t_n) + \int_{t_0}^{t_n} \dot{m}_i dt'. \qquad \text{(Eq. 16)}$$

It is limited, however, by both solubility limitation and sorption.

Sorption

The mass in both the free fluid and in the intact matrix exists in both sorbed and non-sorbed phases. The relationship between the sorbed mass concentration in the solid phase (e.g. the pore walls),

$$s = \frac{\text{mass of sorbed contaminant}}{\text{mass of total solid phase}} \qquad \text{(Eq. 17)}$$

and the dissolved liquid concentration,

$$c = \frac{\text{mass of dissolved contaminant}}{\text{volume of total liquid phase}} \qquad \text{(Eq. 18)}$$

can be expressed by a number of isotherm models.

In this model, sorption is taken into account throughout the volume. In the intact matrix, the contaminant mass is distributed between the pore walls and the pore fluid by sorption. So too, contaminant mass released from the intact matrix by degradation is distributed between dissolved mass in the free fluid and sorbed mass in the degraded and precipitated solids.

To solve for the boundary conditons in this model, the amount of non-sorbed contaminant mass in the free fluid must be found. This value, m_{ffl}, can be expressed in terms of the total degraded contaminant mass and the contaminant mass in the degraded and precipitated solid,

$$m_{ffl} = m_{ffT} - m_{psc}. \qquad \text{(Eq. 19)}$$

The mass of contaminant sorbed into the degraded and precipitated solids can be found using a linear isotherm model [1], characterized by the relationship

$$s_i = K_{di} c_i \qquad \text{(Eq. 20)}$$

where

$$
\begin{aligned}
s_i &= \text{ the solid concentration of isotope i } [kg/kg] \\
K_{di} &= \text{ the distribution coefficient of isotope i} [m^3/kg] \\
c_i &= \text{ the liquid concentration of isotope i } [kg/m^3].
\end{aligned}
$$

Thus, from (Eq. 17),

$$
\begin{aligned}
s_{i,ps} &= \frac{\text{contaminant mass in degraded and precipitated solids}}{\text{total mass of degraded and precipitated solids}} \\
&= \frac{m_{psc}}{m_{psT}} \\
&= \frac{m_{psc}}{m_{psm} + m_{psc}}
\end{aligned}
$$

where

$$
\begin{aligned}
m_{psm} &= \text{ noncontaminant mass in degraded and precipitated solids } [kg] \\
&= \rho_b V_{ps} \\
m_{psc} &= \text{ contaminant mass in degraded and precipitated solids } [kg] \\
\rho_b &= \text{ bulk (dry) density of the medium } [kg/m^3].
\end{aligned}
$$

$$\text{(Eq. 21)}$$

The following expression results, giving contaminant mass in the degraded and precipitated solids in terms of the sorption coefficient,

$$
\begin{aligned}
m_{psc} &= s_{ps} m_{psT} \\
&= K_d C_{ffl} m_{psT} \\
&= \frac{K_d m_{ffl} m_{psT}}{V_{ff}} \\
&= \frac{K_d}{V_{ff}}(m_{ffT} - m_{psc}) m_{psT} \\
&= \frac{K_d}{V_{ff}}(m_{ffT} - m_{psc})(m_{psm} + m_{psc}) \\
&= \frac{K_d}{V_{ff}}(m_{ffT} m_{psm} - m_{psc} m_{psm} + m_{ff} m_{psc} - m_{psc}^2) \\
&= \frac{K_d}{V_{ff}}(m_{ffT} m_{psm} + (m_{ffT} - m_{psm}) m_{psc} - m_{psc}^2)
\end{aligned}
$$

which, rearranged, becomes

$$
0 = m_{psc}^2 + \left(-m_{ffT} + m_{psm} + \frac{V_{ff}}{K_d}\right) m_{psc} - m_{ffT} m_{psm}
$$

and is solved using the quadratic formula, such that

$$
m_{psc} = \frac{m_{ffT} - m_{psm} - \frac{V_{ff}}{K_d}}{2} \pm \frac{\sqrt{\left(-m_{ffT} + m_{psm} + \frac{V_{ff}}{K_d}\right)^2 - 4 m_{ffT} m_{psm}}}{2}
$$

which, again rearranged, becomes

$$
\begin{aligned}
&= \frac{1}{2}\left(m_{ffT} - m_{psm} - \frac{V_{ff}}{K_d}\right) \\
&\pm \frac{1}{2}\sqrt{m_{ffT}^2 + 2 m_{ffT}\left(m_{psm} - \frac{V_{ff}}{K_d}\right) + \left(m_{psm} + \frac{V_{ff}}{K_d}\right)^2}.
\end{aligned}
\tag{Eq. 22}
$$

Plugging (Eq. 22) into (Eq. 19) results in the following expression for m_{ffl} in terms of known quantities

$$
\begin{aligned}
m_{ffl} &= m_{ffT} - \frac{1}{2}\left(m_{ffT} - m_{psm} - \frac{V_{ff}}{K_d}\right) \\
&\mp \frac{1}{2}\sqrt{m_{ffT}^2 + 2 m_{ffT}\left(m_{psm} - \frac{V_{ff}}{K_d}\right) + \left(m_{psm} + \frac{V_{ff}}{K_d}\right)^2}.
\end{aligned}
\tag{Eq. 23}
$$

Solubility

In addition to engineered barriers, contaminant transport is constrained by the solubility limit [5],

$$
m_{s,i} \leq V_w C_{sol,i},
\tag{Eq. 24}
$$

where

$$m_{s,i} = \text{ solubility limited mass of isotope i in volume } V_w [kg]$$
$$V_w = \text{ volume of the solution } [m^3]$$
$$C_{sol,i} = \text{ solubility limit, the maximum concentration of i } [kg/m^3].$$

The desired boundary conditions can be expressed in terms of m_{ffl}. First, the Dirichlet boundary condition is

$$C(x,y,z,t) = \frac{m_{ffl}(t)}{V_{ff}(t)} \forall (x,y,x) \in \Gamma. \quad \text{(Eq. 25)}$$

From this boundary condition in combination with global advective velocity data, all other boundary conditions can be found.

Lumped Parameter Radionuclide Transport Model

The response function model implemented interchangeable piston flow, exponential, and dispersion response functions [6]. For systems in which the flow is sufficiently slow to be assumed constant over a timestep, it is possible to model a system of volumes as a connected lumped parameter models (Figure 4).

$$C_{in0} \qquad C_{out0} = C_{in1} \qquad C_{out1} = C_{in2} \qquad C_{out2} = C_{in3} \qquad C_{out3}$$
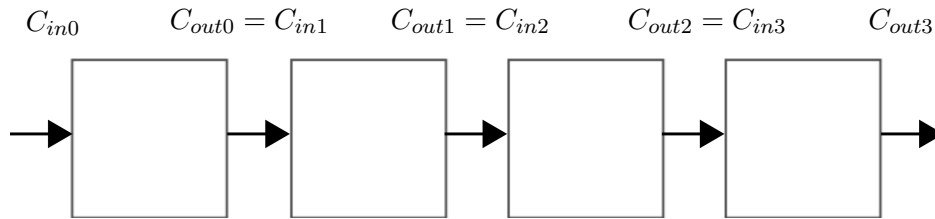
Figure 4: A system of volumes can be modeled as lumped parameter models in series.

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, $C_{out}(t)$,

$$C_{out}(t) = \int_{-\infty}^{t} C_{in}(t')g(t-t')e^{-\lambda(t-t')}dt' \quad \text{(Eq. 26)}$$

equivalently

$$C_{out}(t) = \int_{0}^{\infty} C_{in}(t-t')g(t')e^{-\lambda t'}dt' \quad \text{(Eq. 27)}$$

where

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

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 [6] include the Piston Flow Model (PFM),

$$g(t') = \delta(t' - t_t))$$
(Eq. 28)

the Exponential Model (EM)

$$g(t') = \frac{1}{t_t} e^{-\frac{t'}{t_t}}$$
(Eq. 29)

and the Dispersion Model (DM),

$$g(t') = \left(\frac{Pe \, t_t}{4\pi t'}\right)^{\frac{1}{2}} \frac{1}{t'} e^{-\frac{Pe \, t_t \left(1 - \frac{t'}{t_t}\right)^2}{4t'}},$$
(Eq. 30)

where

$$
\begin{aligned}
Pe &= \text{ Peclet number for mass diffusion } [-] \\
t_t &= \text{ mean tracer age } [s] \\
&= t_w \text{ if there are no stagnant areas} \\
t_w &= \text{ mean residence time of water } [s] \\
&= \frac{V_m}{Q} \\
&= \frac{z}{v_z} \\
&= \frac{z\theta_e}{q}
\end{aligned}
$$

in which

$$
\begin{aligned}
V_m &= \text{ mobile water volume } [m^3] \\
Q &= \text{ volumetric flow rate } [m^3/s] \\
z &= \text{ average travel distance in flow direction } [m] \\
v_z &= \text{ mean water velocity} [m/s] \\
q &= \text{ Darcy Flux } [m/s] \\
\theta_e &= \text{ effective (connected) porosity } [\%].
\end{aligned}
$$

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 are given in [6],

$$
C(t) = \begin{cases}
PFM & C_0 e^{\lambda t_t} \\
EM & \frac{C_0}{1 + \lambda t_t} \\
DM & C_0 e^{\frac{Pe}{2}\left(1 - \sqrt{1 + \frac{4\lambda t_t}{Pe}}\right)}.
\end{cases}
$$
(Eq. 31)

One Dimensional Permeable Porous Medium Radionuclide Transport Model

Finally, abstraction results informed modifications to the implementation of an analytic solution to the one dimensional advection-dispersion equation with finite domain and Cauchy and Neumann boundary conditions at the inner and outer boundaries, respectively.

Various solutions to the advection dispersion equation (Eq. 1) 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.

The conceptual model in Figure 5 represents solute transport in one dimension with unidirectional flow upward (a conservative assumption) and a semi-infinite boundary condition in the positive flow direction. The solution is given (Leij et. al, [4]) and described below.
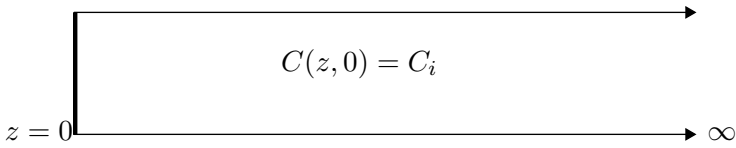
$$-D\frac{\partial C}{\partial z}\big|_{z=0} + v_z C = \begin{cases} v_z C_0 & t < t_0 \\ 0 & t > t_0 \end{cases} \qquad\qquad \frac{\partial C}{\partial z}\big|_\infty = 0$$

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

Figure 5: A one dimensional, semi-infinite model, unidirectional flow, solution with Cauchy and Neumann boundary conditions

For the boundary conditions,

$$-D\frac{\partial C}{\partial z}\big|_{z=0} + v_z c = \begin{cases} v_z C_0 & (0 < t < t_0) \\ 0 & (t > t_0) \end{cases}, \tag{Eq. 32}$$

$$\frac{\partial C}{\partial z}\big|_{z=\infty} = 0 \tag{Eq. 33}$$

and the initial condition,

$$C(z,0) = C_i, \tag{Eq. 34}$$

the solution is given as

$$C(z,t) = \frac{C_0}{2}\left[\mathrm{erfc}\left[\frac{L - v_z t}{2\sqrt{D_L t}}\right] + \frac{1}{2}\left(\frac{v_z^2 t}{\pi D_L}\right)^{1/2} e^{\frac{-(L - v_z t)^2}{4 D_L t}} \right.$$
$$\left. - \frac{1}{2}\left(1 + \frac{v_z L}{D_L} + \frac{v_z^2 t}{D_L}\right) e^{\frac{v_z L}{D_L}} \mathrm{erfc}\left[\frac{L - V_z t}{2\sqrt{D_L t}}\right] \right]. \tag{Eq. 35}$$

DISCUSSION OF THE BASE CASE DEMONSTRATION

A base case simulation was conducted as a proof of principle demonstration of the modularity and interchangeability of these models. The simplest of the contaminant transport models was used to represent the natural and engineered barrier components of a repository concept. This

concept consisted of a saturated clay environment. In this demonstration, the Cyder open source library integrates with the Cyclus computational fuel cycle systems analysis platform in order to calculate repository performance metrics with respect to candidate fuel cycle options. Thus, the demonstration illuminates the suitability of Cyder's interface for linking to other tools as well as for use as a stand-alone radionuclide transport calculation engine.

The demonstration case is an empty software architecture in which to implement the physical models. This demonstration has built and tested component module loading of models and data, information passing between components represented by degradation rate nuclide transport models, and database writing.

Results of unit tests and benchmarking efforts were positive as was a proof of principle base case demonstration of the interface between these models. The base case demonstration has used the degradation rate to represent nuclide transport through waste form, waste package, and buffer components in a generic, isotropic, permeable porous geological medium with reducing geochemistry as well as the near field. Expected degradation behavior and congruent release was observed in unit testing.
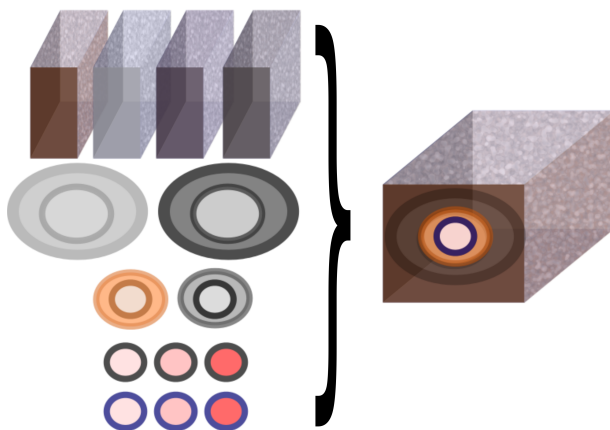

Figure 6: Waste form, waste package, buffer, and far field components posess transport behavior selected from available transport models, are parameterized by user data, and are loaded modularly into a cohesive framework.

CONCLUDING REMARKS

The Cyder source code in which these models are implemented as well as associated documentation are freely available to interested researchers and potential model developers. The application programming interface to this software library is intentionally general, facilitating the incorporation of the models presented here within external software tools in need of a multicomponent repository model.

Furthermore, this work contributes to an expanding ecosystem of computational models available for use with the Cyclus fuel cycle simulator. This hydrologic nuclide transport library, by virtue of its capability to modularly integrate with the Cyclus fuel cycle simulator has laid the foundation for integrated disposal option analysis in the context of fuel cycle options.

ACKNOWLEDGEMENTS

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