A Next-Generation Transport Simulator for the Waste Isolation Pilot Plant (WIPP) Performance Assessment

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INTRODUCTION

Waste Isolation Pilot Plant (WIPP) performance assessment (PA) calculations estimate the probability and consequence of potential radionuclide releases from the repository to the accessible environment for a regulatory period of 10,000 years after facility closure. The proposed replacement of waste panels in the WIPP challenges the modeling assumptions inherent in the two-dimensional (2-D) flared grid used in PA calculations of flow and transport in and around the repository. Therefore, development of a new three-dimensional (3-D) model for use in PA was warranted. At the core of this task is the development of a single, efficient, 3-D flow and transport simulator (PFLOTRAN [1]) that incorporates WIPP-specific process models to eventually replace a portion of the suite of software (i.e., BRAGFLO [2] and NUTS [3]) that is currently utilized for flow and transport in WIPP PA. The current work presents the development of the new transport mode in PFLOTRAN, named the Nuclear Waste Transport (NWT) Mode, and describes the improvements ~~over the currently utilized NUTS transport simulator~~for WIPP PA. Most notably, the NWT Mode is formulated in terms of the total bulk concentration, rather than the more common aqueous (dissolved) mass concentration, allowing it to accurately accommodate completely dry conditions without numerical difficulty.

GOVERNING EQUATIONS FOR TRANSPORT WITH EQUILIBRIUM CHEMISTRY

The Nuclear Waste Transport Mode in PFLOTRAN models the transport of a species as described by the governing equations for conservation of mass. NWT includes species advection (without diffusion), assuming equilibrium chemistry for precipitation/dissolution, and radioactive decay processes. The NWT Mode process model is executed sequentially at each time step after the full flow field solution is calculated. The primary dependent variable is the total bulk mass of species j, in phase α, denoted by M_j^α. The governing equation is given by,

$$\frac{\partial}{\partial t}\Sigma_\alpha\left(M_j^\alpha\right) + \nabla\cdot\Sigma_\alpha\left(\boldsymbol{u}^\alpha M_j^\alpha\right) = \Sigma_\alpha\left(\dot{Q}_j^\alpha\right) + \Sigma_\alpha\left(\dot{R}_j^\alpha\right) \quad (1)$$

where the total bulk mass of species j, in phase α, is given by,

$$M_j^\alpha = M_j^A + M_j^P \quad (2)$$

and is a sum of the aqueous (A) and precipitated (P) phases which are assumed to be in chemical equilibrium. The units of M_j^α are mol • m_{bulk}^{-3}. Furthermore, $\boldsymbol{u}^\alpha$ is the phase velocity in units of m • sec^{-1}. On the right-hand side, $\dot{Q}_j^\alpha$ is a generic source/sink term of species j in each phase (in units of mol • m_{bulk}^{-3} • sec^{-1}), and $\dot{R}_j^\alpha$ represents a source or sink of species j mass in each phase due to radioactive decay or ingrowth (in units of mol • m_{bulk}^{-3} • sec^{-1}).

Transport is assumed to occur only in the liquid fluid phase and not in the gas phase. Furthermore, the aqueous component of species j is the only mobile phase, while the precipitated phase is considered immobile (e.g., $\boldsymbol{u}^P = 0$). With these assumptions, the governing equation simplifies to,

$$\frac{\partial}{\partial t}\Sigma_\alpha\left(M_j^\alpha\right) + \nabla\cdot\left(\boldsymbol{u}^A M_j^A\right) = \Sigma_\alpha\left(\dot{Q}_j^\alpha\right) + \Sigma_\alpha\left(\dot{R}_j^\alpha\right). \quad (3)$$

The aqueous and precipitated phases of the total mass for each species j are defined as,

$$M_j^A = \emptyset S_{liq} C_j^A, \text{ and} \quad (4)$$

$$M_j^P = \emptyset S_{ppt} C_j^P = f\left(M_j^A\right) \quad (5)$$

where S_{liq} and S_{ppt} are the liquid and precipitant saturations (in units of m_{liq}^3 • m_{void}^{-3} and m_{ppt}^3 • m_{void}^{-3}, respectively) within the pore space $\emptyset$ (in units of m_{void}^3 • m_{bulk}^{-3}), and C_j^A and C_j^P are the aqueous and precipitated phase concentrations, respectively, of each species j (in units of mol_j • m_{liq}^{-3} and mol_j • m_{ppt}^{-3}, respectively).

The aqueous concentration for each species is calculated by first disbursing the total species mass within the pore space for each grid cell. Immediately after, the resulting aqueous concentration for each species is compared to the species solubility limit. If it is below the solubility limit, the aqueous concentration calculated remains unchanged. If instead it is above the solubility limit, the aqueous concentration is set to the species solubility limit and the

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amount of species mass in excess of the solubility limit is assigned as the precipitated mass.

With these definitions, the governing equation expands to,

$$\frac{\partial}{\partial t}\sum_{\alpha}\left(M_j^{\alpha}\right)+\nabla\cdot\left(\boldsymbol{q}^A S_{liq} C_j^A\right)=\sum_{\alpha}\left(\dot{Q}_j^{\alpha}\right)+\sum_{\alpha}\left(\dot{R}_j^{\alpha}\right) \quad (6)$$

where $\boldsymbol{q}^A$ is the liquid Darcy flux (in units of $m_{liq}^3 \bullet m_{bulk}^{-2} \bullet sec^{-1}$) and has the relationship $\boldsymbol{q}^A = \boldsymbol{u}^A \emptyset$.

The generic source/sink term, $\dot{Q}_j^{\alpha}$, is defined by,

$$\dot{Q}_j^{\alpha}=\frac{U_j^{\alpha}}{\forall}M_j^{\alpha}=\frac{U_j^{A}}{\forall}M_j^{A}=\frac{Q_j^{A}}{\forall}S_{liq}C_j^A \quad (7)$$

where U_j^{α} is a volumetric flow of species j in each phase α (in units of $m_{bulk}^3 \bullet sec^{-1}$). Because the only mobile phase is the aqueous phase, the volumetric flow of each species is defined entirely by U_j^A, the volumetric flow of each species in the aqueous phase. On the right-hand side of Eq. (7), we define the generic source/sink term using Q_j^A, the volumetric Darcy flux (in units of $m_{liq}^3 \bullet sec^{-1}$), and $\forall$, which is a volume. In this formulation, if the term in a source, C_j^A represents the aqueous phase concentration of species j in the fluid source. On the other hand, if the term is a sink, C_j^A represents the aqueous phase concentration of species j in the domain at the location of the sink.

The reaction term, $\dot{R}_j^{\alpha}$, consists of decay and ingrowth of radioactive isotope species. Radioactive decay and ingrowth is defined by the Bateman equations,

$$\dot{R}_j^{\alpha}=\frac{\partial M_j^{\alpha}}{\partial t}=-\lambda_j M_j^{\alpha}+\lambda_p M_p^{\alpha} \quad (8)$$

where the subscript j represents the radioactive isotope species, and the subscript p represents the radioactive parent of species j. The radioactive decay rate (in units of sec^{-1}) is given by λ. Decay and ingrowth is calculated for the total bulk mass, which is then distributed across the aqueous and precipitated phases after the Bateman equations are solved.

NUMERICAL METHODS FOR THE SOLUTION OF THE TRANSPORT GOVERNING EQUATIONS

The governing equation described by Eq. (1) is a set of equations in space and time. The set of equations is solved by first discretizing them using the finite volume method. The finite volume method uses a volume integral formulation of the set of equations with a finite partitioning set of volumes to discretize the equations in space and time. The expression in Eq. (6) becomes,

$$\int_V\left[\int_t\left[\frac{\partial}{\partial t}\sum_{\alpha}\left(M_j^{\alpha}\right)+\nabla\cdot\left(\boldsymbol{q}^A S_{liq} C_j^A\right)\right]dt\right]dV=$$

$$\int_V\left[\int_t\left[\sum_{\alpha}\left(\dot{Q}_j^{\alpha}\right)+\sum_{\alpha}\left(\dot{R}_j^{\alpha}\right)\right]dt\right]dV \quad (9)$$

which integrates the governing equation in space and time. Upon taking these integrals and applying the divergence theorem (noting several mathematical details omitted for brevity), the discretized equation becomes,

$$\partial\sum_{\alpha}\left(M_j^{\alpha}\right)\forall+\Delta t\sum_F\left(\boldsymbol{q}^A S_{liq} C_j^A\cdot\boldsymbol{n}A_F\right)=$$
$$\Delta t\forall\left[\sum_{\alpha}\left(\dot{Q}_j^{\alpha}\right)+\sum_{\alpha}\left(\dot{R}_j^{\alpha}\right)\right] \quad (10)$$

where Δt is the time step duration (in units of sec). The summation term, $\sum_F\left(\boldsymbol{q}^A S_{liq} C_j^A\cdot\boldsymbol{n}A_F\right)$, results from a surface integration on the discretized volume, $\forall$. This surface integral is discretized as a sum over a finite number of faces, where $\boldsymbol{n}$ is the unit normal vector for each face and A_F is the area of each face. In the case of a 3-D structured rectilinear grid, each volume represents a grid cell that has 6 faces.

The accumulation term (first term on the left-hand side of Eq. (10)) is discretized in time using a forward difference, and all other terms are taken at the new time level, which is described as an implicit backward Euler discretization method (as opposed to an explicit standard Euler discretization where all other terms are taken at the current time level). The implicit time discretization method has the advantage of numerical stability for any time step taken but requires more computational effort to solve than an explicit method.

The Newton-Krylov iteration method is used to solve the discretized governing equations in space and time. First, Eq. (10) is transformed into a residual equation by putting all terms on one side of the equation and setting their sum to zero, as expressed by,

$$R\left(M_j^{\alpha,t+1}\right)=0 \quad (11)$$

This method states that a generic system of non-linear equations, $f_i(x_1,x_2,\ldots,x_n)=0$, can be expressed as a new system of linear equations, described by,

$$f_i(x_1,x_2,\ldots,x_n)=$$
$$f_i\left(x_1^k,x_2^k,\ldots,x_n^k\right)+\sum_{j=1}^{n}\left(x_j^{k+1}-x_j^k\right)\frac{\partial f_i\left(x_1^k,x_2^k,\ldots,x_n^k\right)}{\partial x_j}=0 \quad (12)$$

of the form $Ax=b$. The matrix A is called the Jacobian and is the set of partial derivatives of the discretized residual equation (i.e., Eq. (10)) with respect to the primary variable, M_j^{α}. In the NWT Mode, analytical derivatives are defined and computed. The right-hand side, b, is the residual at the kth iteration. Finally, x is the solution vector update, thus giving,

$$\sum_{j=1}^{n}\left(\frac{\partial R_i\left(M_j^{\alpha,t+1}\right)^k}{\partial M_j^{\alpha}}\left(\delta M_j^{\alpha,t+1}\right)=-R\left(M_j^{\alpha,t+1}\right)^k\right) \quad (13)$$

The solution is then updated after each iteration according to,

$$\left(M_j^{\alpha,t+1}\right)^{k+1} = \left(M_j^{\alpha,t+1}\right)^{k} + \left(\delta M_j^{\alpha,t+1}\right) \qquad (14)$$

and the updated solution is used to evaluate the residual equation. This process is repeated until the residual value is zero. However, because the residual will never truly be zero, convergence is declared according to a variety of tolerances on $\delta M_j^{\alpha,t+1}$ and the residual value.

The specific convergence criteria used in NWT Mode include the infinity norm of the absolute residual value, the infinity norm of the scaled residual value, and the infinity norm of the relative solution update. The scaled residual value is the proportion of the residual relative to the accumulation term (i.e., first term on the left-hand side of Eq. (10)), described by

$$\frac{R\left(M_j^{\alpha,t+1}\right)}{\left(\sum_{\alpha}\left(M_j^{\alpha,t}\right)\forall\right)}. \qquad (15)$$

The relative solution update is the proportion of the solution update relative to the previous solution, described by

$$\delta M_j^{\alpha,t+1}/M_j^{\alpha,t}. \qquad (16)$$

Convergence is defined as meeting the absolute OR scaled residual criteria, AND the relative solution update criteria at each grid cell (e.g., infinity norm).

An uncommon and very advantageous feature in the NWT Mode of PFLOTRAN is the ability to define convergence criteria for each individual species. This is useful when there are large differences (e.g., several orders of magnitude) in the amount of mass between species, which may result from differences in the initial inventories, differences in solubility limits, or differences in the radioactive decay constants between species. Whether a result of any of these reasons, when large differences in the amount of mass between species exists, it may not be appropriate to assign a single value for the convergence criteria to all species. Having the capability to define species-specific convergence criteria values allows the numerical solution to converge with less error.

NWT MODE IMPROVEMENTS ~~OVER PREVIOUS SOFTWARE~~ FOR WIPP PA CALCULATIONS

Several NWT Mode features represent improvements for WIPP PA calculations. ~~over the previous software, NUTS.~~

First, the NWT Mode is formulated in terms of the total bulk concentration (moles per bulk cubic meter), allowing it to accurately accommodate completely dry conditions without numerical difficulty. Formulations in terms of the aqueous concentration, ~~(~~a more common formulation~~as is the case with NUTS)~~, cannot naturally handle completely dry conditions because the aqueous concentration becomes undefined, and the system of equations cannot be solved for the aqueous concentration. In such cases, modification to the governing equations is done to avoid mathematical singularity at dry-out conditions.

Second, NWT Mode uses finite volume discretization and solves the discretized equations with a fully implicit, backward Euler approach based on Newton-Krylov iteration. The finite volume discretization conserves mass, by definition~~, as opposed to the explicit, finite difference method that is used by NUTS~~. Furthermore, using a backward Euler approach based on Newton-Krylov iteration allows numerical stability for even large time steps, and can handle non-linearities in the problem formulation. The uncommon capability allowing species-specific assignment of convergence criteria in NWT Mode reduces numerical error when multiple species with disparate inventories are simulated and is a state-of-the-art feature that is lacking in many other transport simulators.

Third, in NWT Mode, the transport process model is solved sequentially after fluid flow at each time step. ~~This is an improvement over NUTS in PA calculations, which solves for transport after 55 years of the flow solution has been solved.~~ Solving for the transport solution at each flow time step ~~, as done by NWT Mode,~~ reduces operator-splitting error, producing a more accurate transport solution. Moreover, in NWT Mode, the transport process model is allowed to sub-step the flow solution in time in order to satisfy convergence criteria and tolerances. This additional feature reduces time truncation error in the numerical solution and is automatically activated when the transport solution cannot be accurately solved at as large of a time step duration as the flow solution was (due to complex flow regimes, for example).

~~Fourth, the NWT Mode can be sequentially coupled to a new wellbore model (currently under development by the authors) to additionally simulate transport in a wellbore emplaced within a larger reservoir. In the previous WIPP PA software architecture, BRAGFLO explicitly meshes the wellbore in the computational domain and uses a flared grid geometry to represent 3-D space on a 2-D grid. In the development of a new 3-D model for use in PA, explicitly meshing the borehole in the computational domain quickly becomes computational intractable due to the large number of grid cells required to do this with good grid quality. Using a wellbore model rather than explicitly meshing the wellbore, therefore, significantly reduces the computational expense. The flexibility of the NWT Mode to plug into updated features of the sequentially coupled flow mode, such the new wellbore model, significantly streamlines the software development process and allows PA calculations to remain practical in 3-D.~~

Finally, because it is part of PFLOTRAN, the NWT Mode inherits its massively parallel computing capability, which greatly speeds up 3-D simulations. In the previous PA, which~~NUTS~~ was limited to ~~serial~~ 2-D computations, serial computations were adequate. However, serial computations must move towards parallelization ~~practically inhibiting its use~~ for the new 3-D PA model.

CONCLUSIONS AND FUTURE WORK

Presented here is the development of the new 3-D transport mode in PFLOTRAN, named the Nuclear Waste Transport (NWT) Mode. Its development was undertaken as a result of the proposed replacement of waste panels in the WIPP, which challenges the modeling assumptions inherent in the 2-D flared grid used in PA calculations of flow and transport in and around the repository. The NWT Mode is part of a single, efficient, 3-D flow and transport simulator (PFLOTRAN) that incorporates WIPP-specific process models that will eventually replace a portion of the suite of software (i.e., BRAGFLO and NUTS) that is currently utilized for flow and transport in WIPP PA.

Besides its role in the updated WIPP PA calculations, the new NWT Mode in PFLOTRAN will also be broadly applicable for the PA community outside of WIPP and has already shown applicability in transport calculations which have been traditionally difficult to solve numerically, due to large non-linearities in the system of equations or complex partially fluid-saturated regimes.

NOMENCLATURE

α = phase index (aqueous or precipitated)
j = species index
p = species' parent index
M_j^α = total bulk mass of species j, in phase α
M_j^A = total bulk mass of species j, in aqueous phase
M_j^P = total bulk mass of species j, in precipitated phase
$\boldsymbol{q}^A$ = liquid Darcy flux
$\boldsymbol{u}^\alpha$ = phase velocity
$\boldsymbol{u}^A$ = aqueous phase velocity
$\boldsymbol{u}^P$ = precipitated phase velocity, assumed zero
U_j^α = volumetric flow of species j, in phase α
U_j^A = volumetric flow of species j, in aqueous phase
C_j^A = aqueous phase concentration of species j
C_j^P = precipitated phase concentration of species j
$\dot{Q}_j^\alpha$ = generic source/sink of species j, in phase α
$\dot{R}_j^\alpha$ = radioactive source/sink of species j, in phase α
$\emptyset$ = pore space
S_{liq} = liquid saturation
S_{ppt} = precipitant saturation
$\forall$ = discretized volume
A_F = area of each discretized volume face
n = normal vector of each discretized volume face
t, Δt = time, time step
m = meter
mol = mole
sec = second

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