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INTEROFFICE MEMORANDUM

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To: K. H. Rosenberger, 705-1C

From: B. H. Lester, 705-1C 8/20/2018

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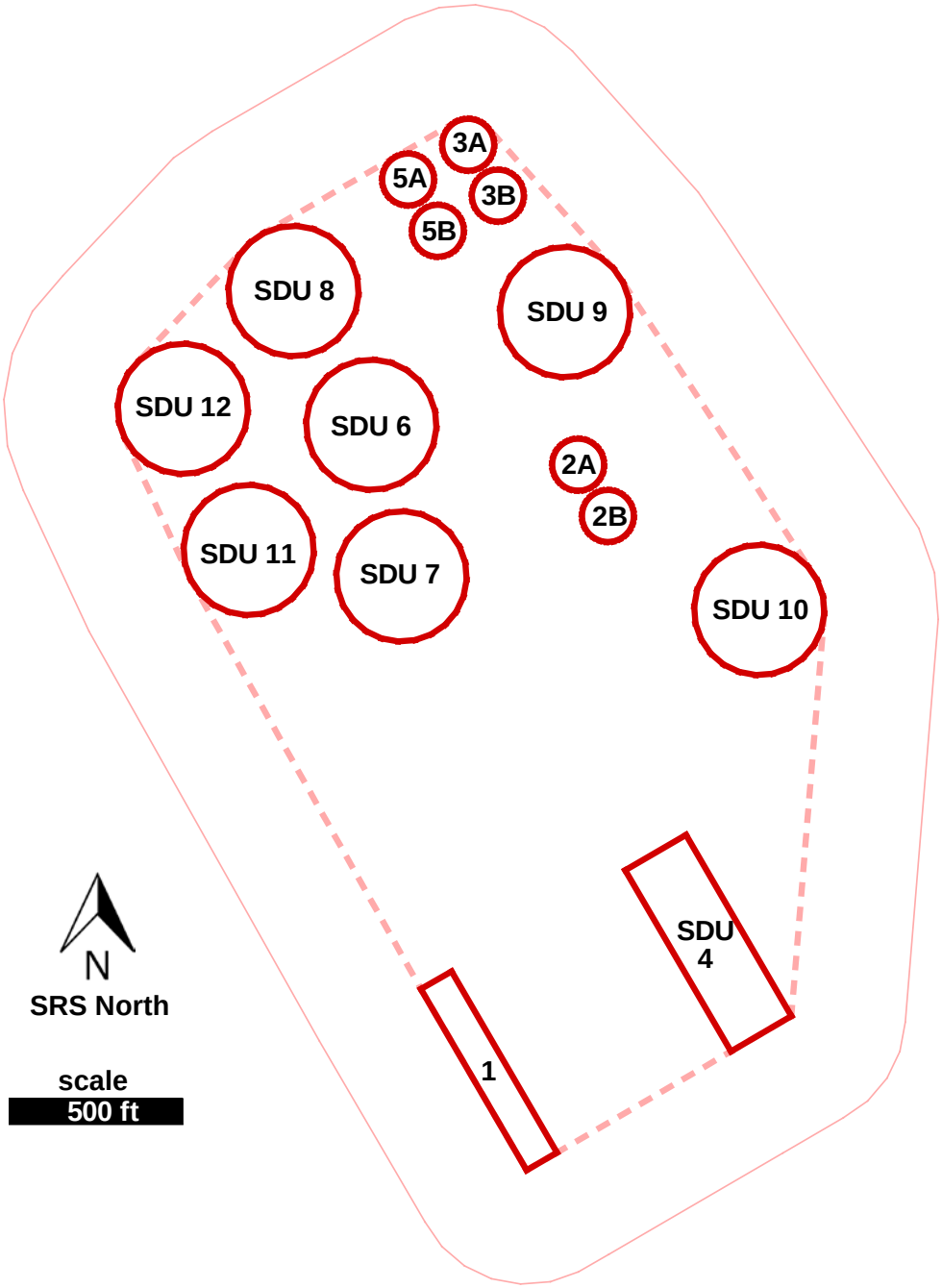
Technetium Solubility Limits for Simulating Tc-99 Releases from Saltstone SDUs

Waste Disposal Authority (WDA) is updating to the Saltstone Disposal Facility (SDF) Performance Assessment (PA) (SRR-CWDA-2009-00017). The update to the SDF PA will address changes in construction details, size, and locations of existing and planned-future Saltstone Disposal Units (SDUs) (i.e., concrete vaults used for containing low activity waste). The updated SDF PA shall also incorporate new data developed through ongoing research activities and apply updated disposal inventories based on disposal operations. As part of the PA effort, models shall be developed to evaluate the release of radionuclides from SDUs, and contaminant transport through groundwater pathways and through air pathways. Finally, resulting concentrations from transport models will be used to evaluate the potential risks imposed on members of the public (MOPs).

This memo describes the development and application of a GoldSim-based optimization model designed to assist in selecting appropriate values from actual experimental results for the technetium solubility limits to be used for simulating the release and transport of dissolved Tc-99 within the SDF waste form (i.e., saltstone) and surrounding cementitious materials (i.e., SDUs) located at the SDF depicted in Figure 1.

The optimization model was developed using GoldSim simulation software (GTG-2017a), an object oriented, probabilistic modeling software designed to evaluate parameter sensitivity and the influence of parameter uncertainty. GoldSim simulation software also includes a contaminant transport module that allows the user to simulate contaminant migration through the use of either; 1) Pipe pathway elements which use a Laplace transform approach to provide a set of analytical solutions to a broad range of advection-dominated advective-dispersive transport problems; and/or 2) cell pathway elements (CPEs) which use a numerical mixing-cell approach to solving advective, diffusive, or advective-diffusive transport problems for any number of transport and storage media. In addition, GoldSim provides the capability of performing optimization simulations allowing the user to calibrate input parameters to "best" minimize (or maximize) an objective function while meeting user-specified constraints.

Figure 1: Locations of Existing and Planned-Future Saltstone Disposal Units (SDUs)



Developed based on G-AES-S-00004.

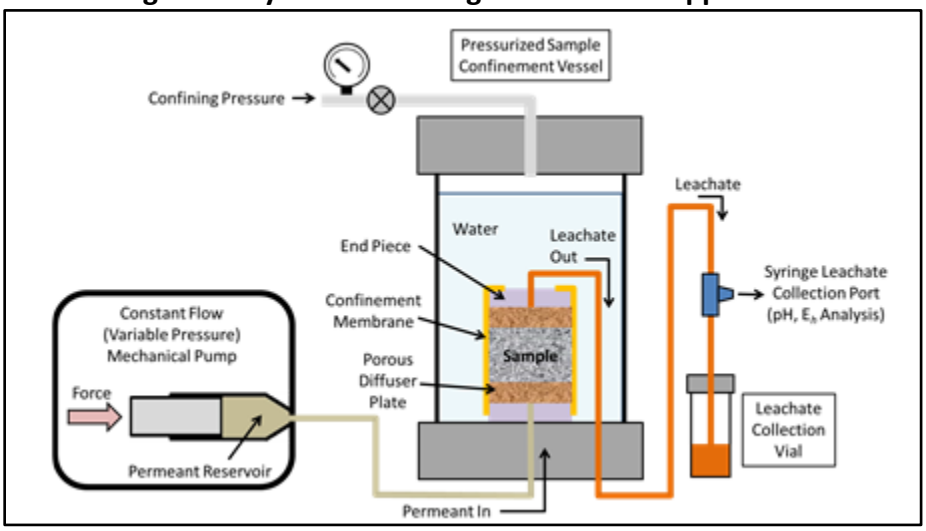
Using the GoldSim DLM Optimization Model to Simulate Tc-99 Release from DLM Testing

DLM Test Abstraction

This section describes the details of the Dynamic Leaching Method (DLM) simulant core study (SREL-R-15-0003, SREL-R-16-0003, SREL-R-17-0005) that serves as the basis for the GoldSim-based Tc-99 DLM Optimization Model. The model utilizes GoldSim's cell pathway elements to provide a mixing-cell

approach to simulating the release of Tc-99 from a simulated DLM sample (see Figure 2). The simulated release results are used in conjunction with GoldSim's optimization capabilities to develop a best-fit match to the Tc-99 release records from an on-going DLM study being conducted by the Savannah River Ecology Laboratory (SREL) (SREL-R-15-0003, SREL-R-16-0003, SREL-R-17-0005). Specifically, the optimization is designed to approximate the Tc-99 releases from a saltstone simulant sample identified as 45-45-10_LeHigh and two actual saltstone core samples taken from SDU-2A (Core A and Core B). The 45-45-10_LeHigh sample is being studied to better understand what impact changes to the blast furnace slag (i.e., SRR switching from Holcim Grade 100 to LeHigh Grade 120 blast furnace slag) will have on Tc-99 release behavior.

Figure 2: Dynamic Leaching Method Test Apparatus



From SRR-CWDA-2018-00033.

Conceptually, the Tc-99 inventory in the cementitious sample is initially assumed to be fully mixed in the pore water and if concentrations reach solubility limits, the excess mass precipitates. In the simplified abstraction, the quantity of precipitate is assumed to be low enough that there is little change in the pore structure imparting negligible change on the flow field. The GoldSim model used for this analysis was extracted from a lysimeter model, so it has the capability to evaluate the influence of radial diffusion, but since this option is not considered in this analysis, the conceptual model assumed here is that of a cylindrical (disc-like) sample subject to one-dimensional flow parallel to the axis of a cylinder. Migration of Tc-99 through the saltstone simulant sample can be approximated by the governing equation for one-dimensional advective-dispersive transport through a homogeneous porous medium as shown in Equation (1):

$$\varphi R(t)\frac{\partial c}{\partial t} = -v_{darcy}(t)\frac{\partial c}{\partial x} + D_L\frac{\partial^2 c}{\partial x^2} - \varphi R(t)\lambda c \quad (1)$$

where:

φ = saltstone simulant porosity [-];
$R(t)$ = time-dependent radionuclide retardation coefficient [-];
v_{darcy} = time-dependent Darcy velocity (volumetric water flux rate $Q(t)$ per unit area) through the DLM sample [L/T];
$D_L = \alpha\, v_{darcy} + D_{eff}$ = longitudinal dispersion coefficient of the DLM sample [L^2/T];

D_{eff} = effective diffusion coefficient of the DLM sample [L^2/T];
c = DLM sample dissolved radionuclide concentration [M/ L^3];
λ = radionuclide decay coefficient [1/T]; and
t = time [T].

Based on mass balance criteria, the upgradient (x=0) and downgradient (x=l) boundary conditions for Equation (1) can be defined as:

$$\left[v_{darcy}(t)c - \frac{\partial c}{\partial x}\right]_{x=0^+} = \left[v_{darcy}(t)c\right]_{x=0^-} = 0, \quad (2)$$

where $c_{x=l^-} = 0$ for the inlet upgradient boundary condition, and

$$\left[v_{darcy}(t)c - \frac{\partial c}{\partial x}\right]_{x=l^-} = \left[v_{darcy}(t)c\right]_{x=l^+} \quad (3a)$$

for the downgradient boundary condition, which assuming $c_{x=l^-} = c_{x=l^+}$, reduces to

$$\left[\frac{\partial c}{\partial x}\right]_{x=l^-} = 0 \quad (3b)$$

at the downgradient boundary (see Massabo, et.al., 2011).

The retardation coefficient presented in Equation (1), is defined as:

$$R(t) = 1 + \frac{\rho_b K_d(t)}{\varphi} \quad (4)$$

where:
ρ_b = bulk density of the DLM sample [M/L^3];
K_d = saltstone simulant radionuclide sorption coefficient [L^3/M].

The time-dependency in the retardation coefficient and associated K_d found in Equations (1) and (4), reflects a potential change in the sorption coefficient due to a change in the chemical environment (SRNL-STI-2009-00473). This assumed change in the sorption coefficient reflects changes in Tc-99 mobility that is expected when the chemical environment changes. For technetium, changes in K_d are expected to occur when the saltstone (or other cementitious materials) transitions from a Reduced Moderately-Aged cementitious stage to an Oxidized Moderately-Aged stage. Changes in the K_d are also expected as Oxidized Moderately-Aged material continues to age (SRNL-STI-2009-00473). While cementitious material is in the Reduced Moderately-Aged stage, the mobility of technetium is expected to be controlled by precipitation of Tc-99 species unless concentrations (defined as c=m/V_{pore} where m is the dissolved mass of Tc-99 and V_{pore} is the pore-volume of the porous medium) fall below solubility limits. In the GoldSim-based DLM Optimization model, all mass in a mixing cell remains dissolved or portioned between the dissolved phase and a sorbed phase until the saturation capacity, S_{cap} (Equation (5)) is reached after which any increase of mass in the cell is precipitated.

$$S_{cap} = S_{limit}\varphi R(t) = S_{limit}[\varphi + \rho_b K_d(t)] \quad (5)$$

Because SREL's DLM testing is still in an early stage of the leaching process, it only reflects processes controlling Tc-99 mobility in the Reduced Young-Aged to Reduced Moderately-Aged stages of saltstone chemistry. Also note that the influence of the third term on the right-hand side (RHS) of Equation (1), which represent the influence of radionuclide decay, will be negligible for this study due to the short time frame of the DLM testing relative the long half-life of Tc-99 (2.1E+05 years).

Four DLM tests for Tc-99 releases were simulated using a GoldSim-based DLM Optimization Model which is based on a cylindrical (disc shaped) GoldSim cell-pathway network comprised of 20 layers of mixing cells (see Figure 3), with each layer being comprised of 10 concentric cylindrical cells. Each cell pathway in the bottom layer is linked to a sink element by an advective mass-flux link. Although as designed, the transport of radionuclides in the DLM simulant sample will be advection dominated, the release of Tc-99 from the sample will also be influenced by mechanical dispersion. For the versions of the DLM Optimization Model discussed herein, the process of mechanical dispersion can be approximated, in an ad hoc manner, by controlling the number of mixing cells that are linked in series in the flow direction, in the cell-pathway network. In the flow direction, the numerical dispersion inherent to the GoldSim model becomes an analogue for mechanical dispersion. The equivalent numerical dispersivity associated with this system of *N* cells and a DLM simulant core of L_{core} in length is based on the following equation (GTG-2017b).

$$\propto \approx \frac{L_{core}}{2N} \quad (7)$$

where:

α = numerical dispersivity [L];
L_{core} = DLM simulant core length [L].
N = number of core cells [-].

Based on Equation 7, the influence of numerical dispersion in the Tc-99 DLM model would approximate the effect mechanical dispersion and produce a longitudinal dispersivity of 0.125 cm in the 2.5 cm long sample model with 20 layers of mixing cells (or a Peclet number of 20). Note that for the simplified one-dimensional flow and transport system simulated in this study, the Peclet number, *Pe*, which reflects the degree of dominance of advection over dispersion, can be defined as:

$$Pe = \frac{Advective\ Transport\ Rate}{Dispersive\ Transport\ Rate} = \frac{L_{core}}{\propto} \quad (8)$$

The logic used in the model to control the dissolution of Tc-99 from the solid phase is based on the surrogate solubility model described in SRNL-STI-2014-00083. This solubility model utilizes a surrogate K_d value to approximate the processes of precipitation and dissolution allowing for a reduction-capacity dependent function to control the dissolution of technetium in the solid phase at a rate dependent on the amount of slag remaining in individual mixing cells making up DLM sample model. With this solubility

model the Tc-99 is release from the solid phase to the liquid phase uniformly over the period of cell oxidation (SRNL-STI-2014-00083). The surrogate K_d used to approximate the precipitation/dissolution process can be defined as:

$$K_d = {x_{Re}}^p {K_d}^{solubility} + (1 - {x_{Re}}^p){K_d}^{redox} \qquad (8)$$

where:

$${K_d}^{redox} = max\left[\frac{c_{slag,0}}{c_{Ox}} x_{Re} - \frac{Sn}{\rho_b}, K_{d,Ox}\right] (9),$$

$${K_d}^{solubility} = max\left[\frac{c_T - nSc_{sol}}{\rho_b C_{sol}}, K_{d,Re}\right] \qquad (10),$$

where:

$c_{slag,0}$ = initial slag concentration [meq e^-/g];
c_T = total bulk concentration [m/L^3] = m_{Tc}/V (the total mass of technetium in a cell divided by total volume of the cell);
c_{Ox} = dissolve oxygen concentration [meq e^-/g];
c_{sol} = solubility limit [mol/L^3];
X_{Re} = reduced fraction of slag [-] = (1- X_{Ox}) where X_{Ox} is the oxidized fraction of slag [-];
$K_{d,Ox}$ = sorption coefficient under oxidized conditions [mL/g];
$K_{d,Re}$ = a minimum value for $K_d^{solubility}$ [mL/g];
s = saturation [-];
n = porosity [-].

Optimization

A review of the leachate releases from the DLM test samples showed a resultant pH range of between 12.8 to 10.0. The DLM test sample releases were compared with GWB generated solubility versus pH curves for $TcO2{\cdot}1.6H_2O$(solid) at Eh values of -0.1V, -0.2V, -0.3V, and -0.4V and similar curves for $TcO2{\cdot}2.0H_2O$(solid) (Seaman and Coutelot, 2018). The comparison of experimental data with the Geochemists Workbench (Bethke and Yeakel 2018) generated curves showed negligible correlation between the GWB data and the DLM test releases (see Appendix A Figures A-1 through A-4). Figures A-5 and A-6 which depict the experimental percent Tc-99 released with respect to leachate released (in terms of pore volumes) are also presented relative to release curves for more conservative constituents to show that the Tc-99 release reflects a solute (for Tc-99) retardation, which is assumed to be a function of solubility limits. The lack of correlation is likely a function of a kinetically limited dissolution/precipitation process. Because of the lack of correlation between Geochemists Workbench (GWB) solubility limit curves versus DLM test leaching results, a solubility-limit model calibration study was used to evaluate conservative values of solubility limits to be used in the updated SDF PA modeling efforts.

In the GoldSim DLM Optimization Model, the “percent inventory released” values (the percent of the initial mass in place that has been leached from the sample) taken from simulated time histories of the DLM sample cell-pathway elements, are compared to the percent inventory released values measured during the SREL DLM test at the end of each specified sampling interval. The solubility limits used in the GoldSim model optimization runs are calibrated using the mean squared error (MSE) between modeled

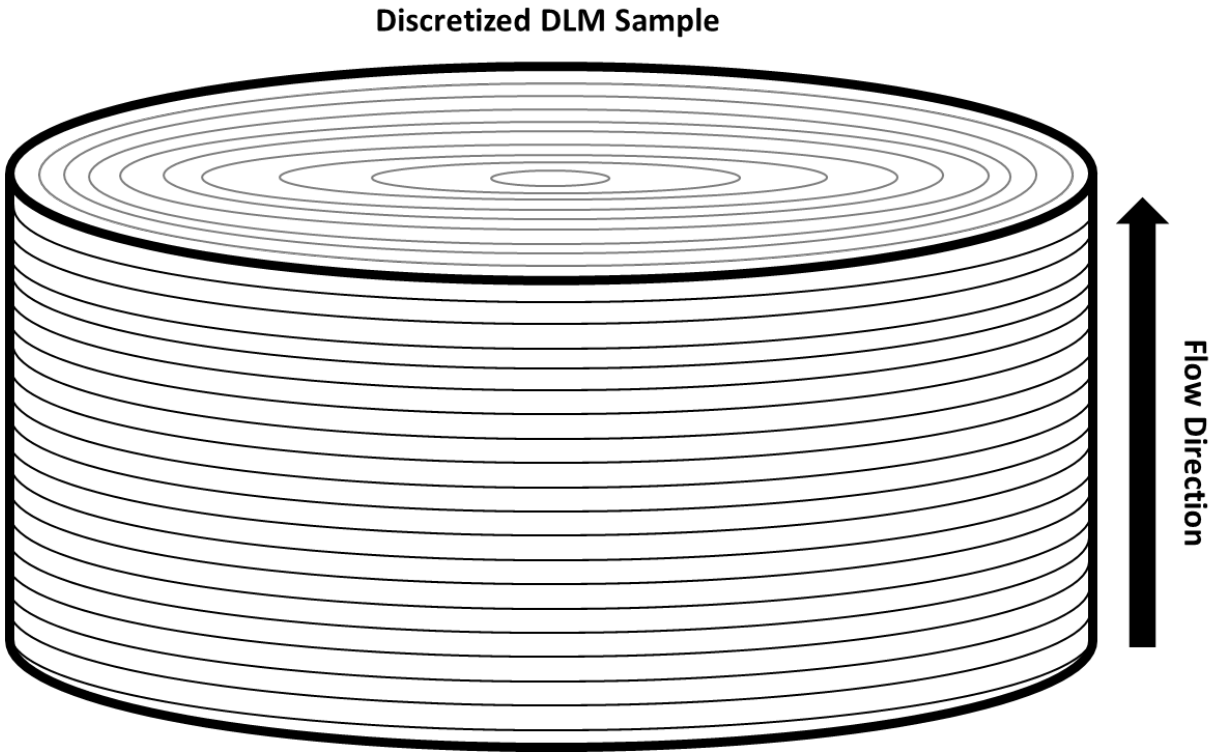
and DLM laboratory test results. The MSE results are used as an objective function that is minimized based on the selection of optimization variables (i.e., solubility limits).

In optimization mode, GoldSim will repeatedly run the model multiple times, systematically selecting combinations of chosen optimization variables. The GoldSim optimizer is based on Box's complex method (Box, 1965). As noted in GoldSim (2017a), Box's complex method begins by generating an initial "complex," which is a set of valid solutions that meet all user-specified requirements. The initial complex contains twice as many valid solutions as the number of optimization variables. Once the initial complex has been developed, the algorithm searches the solution space iteratively, replacing the least optimal members of the complex with more optimal ones until the solution converges. If convergence cannot be achieved a warning message is displayed by the GoldSim software.

In this modeling study, the DLM Optimization Model is used in optimization mode to find the optimal Tc-99 solubility limit(s). Calibration efforts in this study were exercised for the following sets of parameters:

1) searching for an optimal pair of solubility limits in the 45-45-10 Lehigh saltstone simulant,
2) searching for an optimal pair of solubility limits in the SDU 2A saltstone Core A sample,
3) searching for an optimal pair of solubility limits in the SDU 2A saltstone Core B sample, and
4) searching for an optimal single solubility limit in the SDU 2A saltstone Core B sample.

Figure 3: Dynamic Leaching Method Test Apparatus GoldSim Abstraction Model



DLM Optimization Model Structure

As discussed in GTG-2017a, the GoldSim simulation software is designed to be amenable to model development using a "Top-down" approach, starting from the top (i.e., the ultimate objective of the modeling exercise) and concentrating on the integration and coupling of all system components. This section describes the basic structure used in the model. In a GoldSim model, the model's "top" level is the referred to as the Model Container. The inside of the Model Container, is referred to as the Model Root and is analogous to the top directory on a computer. The Model Root of the DLM Optimization model (Figure 4), is comprised of nine active GoldSim containers used to organize the model into six input containers (*ControlInput*, *TransportProcessParameters, Source, Geometry, Materials, and Archive*) where model parameter values are assigned, a system model container (*DLM_Model*) in which the release of Tc-99 from a saltstone simulant core during a DLM test is calculated, a *Clones* container where a set of all cloned GoldSim elements can be found and updated, changing all incidences of like clones throughout the rest of the model, and a calibration container (*Calibration*) where the MSE objective function values are generated. As discussed above, the calculated MSE objective functions, are in turn used to calibrate parameters (solubility limits for this model) needed to replicate the results of the SREL saltstone simulant DLM test. The calibrated set of solubility limits obtained by comparing SREL's DLM laboratory analysis results with model results closely reflect the influence of changes in chemical environment on the migration of Tc-99 within a modeled saltstone simulant under DLM test conditions. The calibrated parameters can then be used to simulate the migration of Tc-99 in a modeled SDU during the Reduced-Young and Reduced-Moderately Aged stages.

Calculations within the *DLM_Model* container are performed in *SaltstoneMonolithContainer*, shown in Figure 5, which is comprised of 20 containers (see Figure 6) each made up of the GoldSim elements representing one of 20 layers of equal thickness in the discretized abstraction model of the DLM test samples. Each arrow between containers represents 10 vertically-oriented advective and diffusive mass-flux links between each of 10 mixing cells and the one in the layer immediately above it. Each layer of mixing cells is contained in a *PuckCells#* container (Figure 7) is comprised of 10 containers (one for each layer-specific concentric cylindrical mixing cell), making up the DLM simulant abstraction model (see Figure 8). Each arrow between containers represents a single radially oriented diffusive mass-flux link between the mixing cells. The content of each container depicted in Figure 8 is presented in Figure 9. The use of individual closed-containers for each mixing cell allows the model to simulate different chemical environments for each cell. The mixing cells in Layer 20 (see Figure 10) use a set of 10 GoldSim advective mass-flux links to release the leachate to the sink cell *Sink_PO* to approximate the leachate release from the DLM simulant sample. The leachate flow rates from the DLM simulant to the sink cell are based on the inputted time-variant volumetric flow rate derived from the DLM simulant lab testing data. This model can follow the oxidation fronts both vertically (based on advection and diffusion) and horizontally (based on diffusion only) allowing for a change from a reduced to oxidized environment on a cell-by-cell basis, but this option is not used reflecting the negligible permeant oxygen concentration used in the DLM tests.

Figure 4: "Top" level of the GoldSim Tc-99 DLM Optimization Model

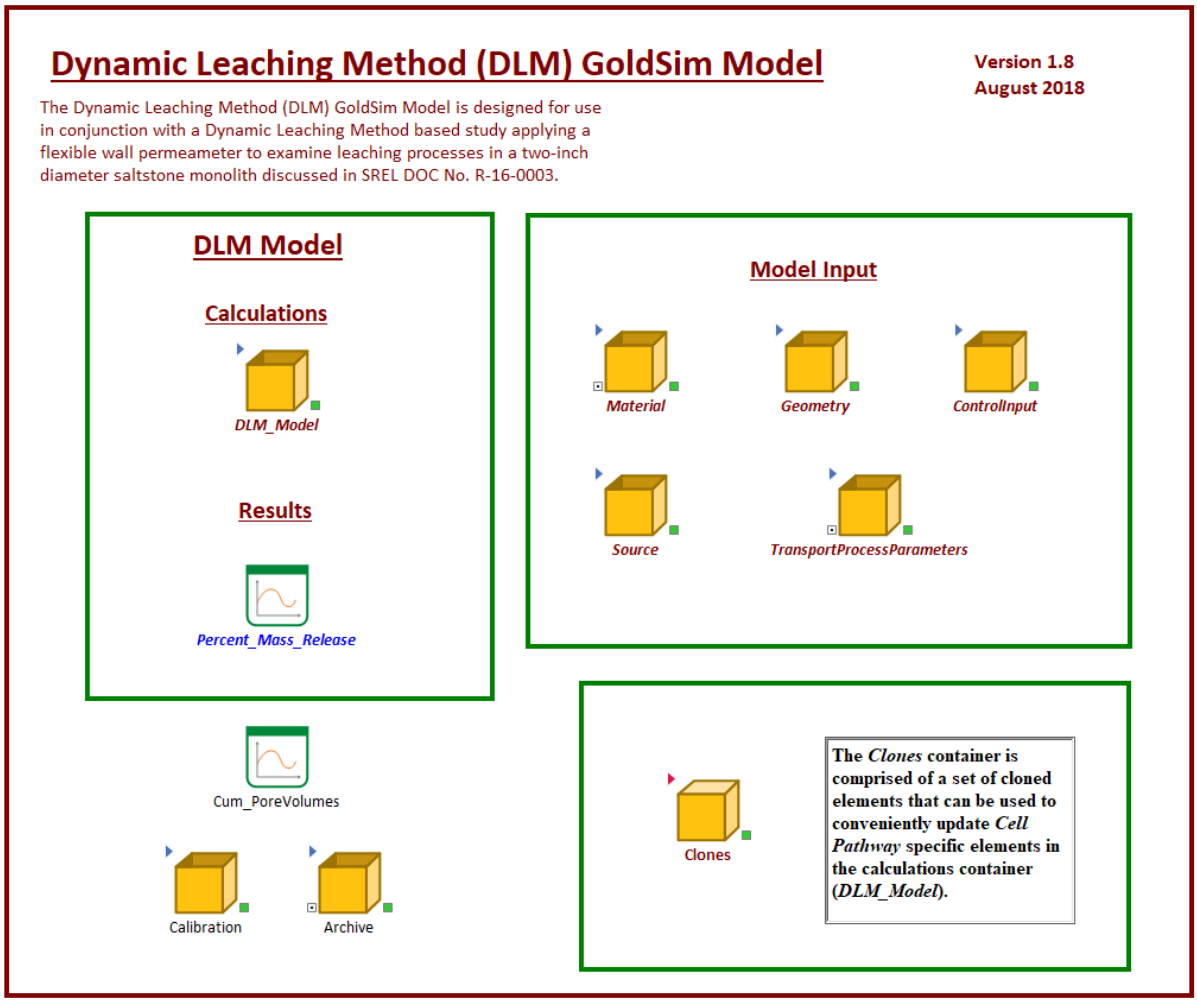


Figure 5: DLM Optimization Model Calculation Container

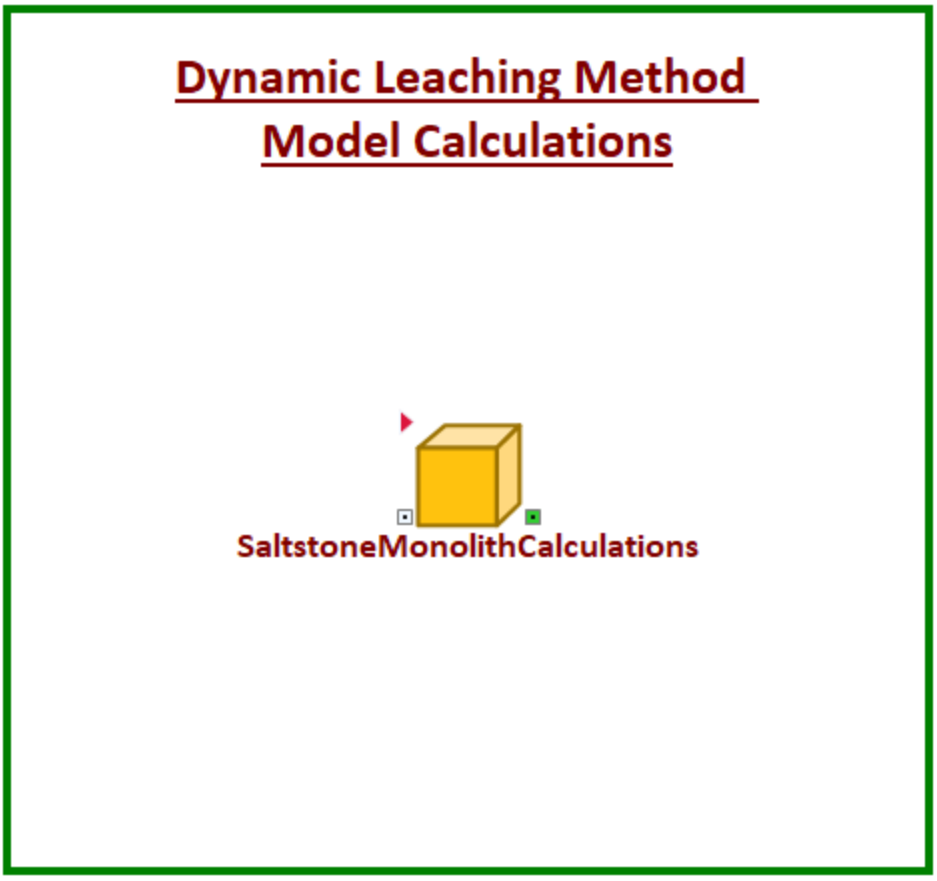


Figure 6: DLM Optimization Model Layer Specific Calculation Containers

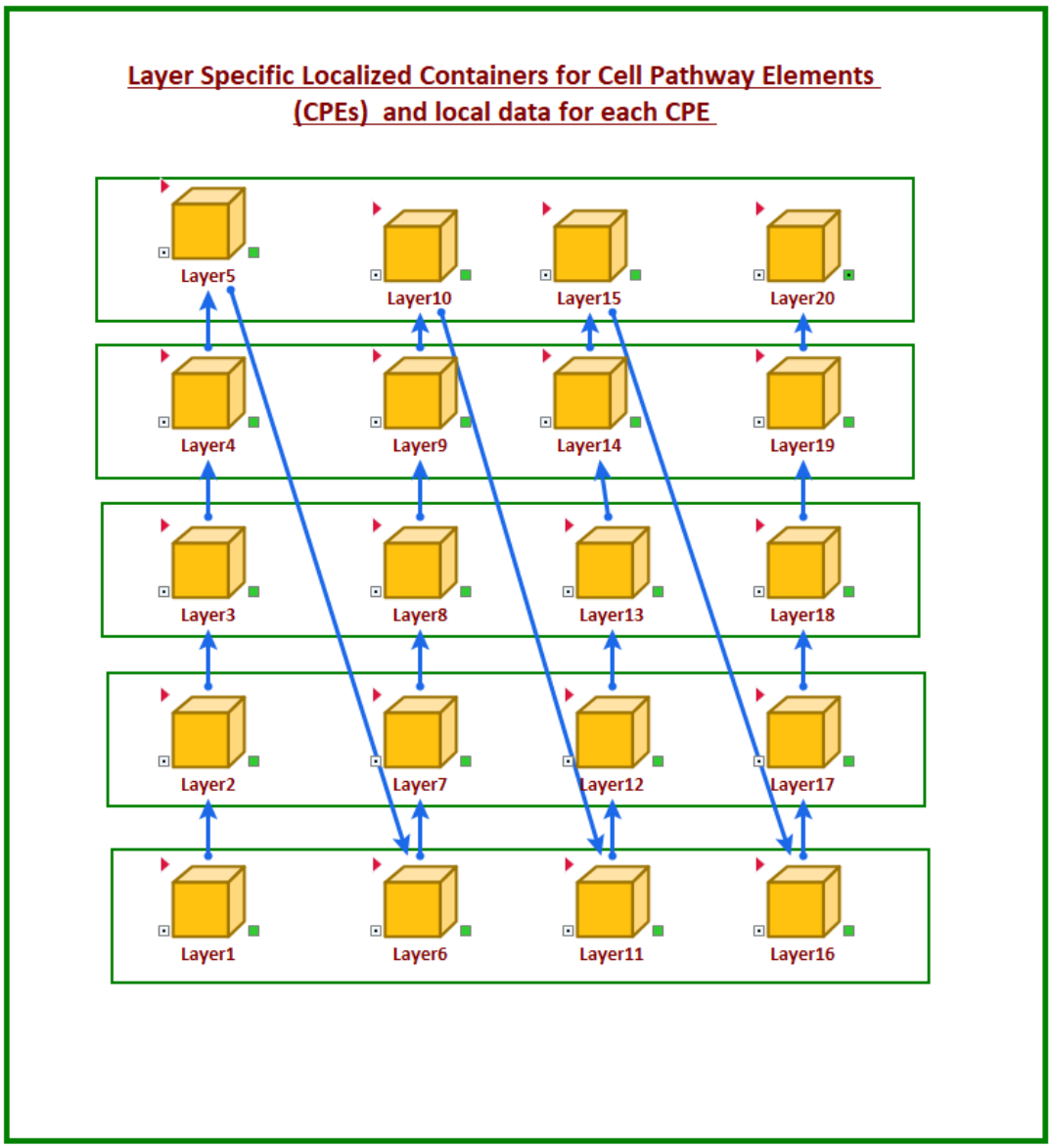


Figure 7: DLM Optimization Model Container Layer 1 Calculations

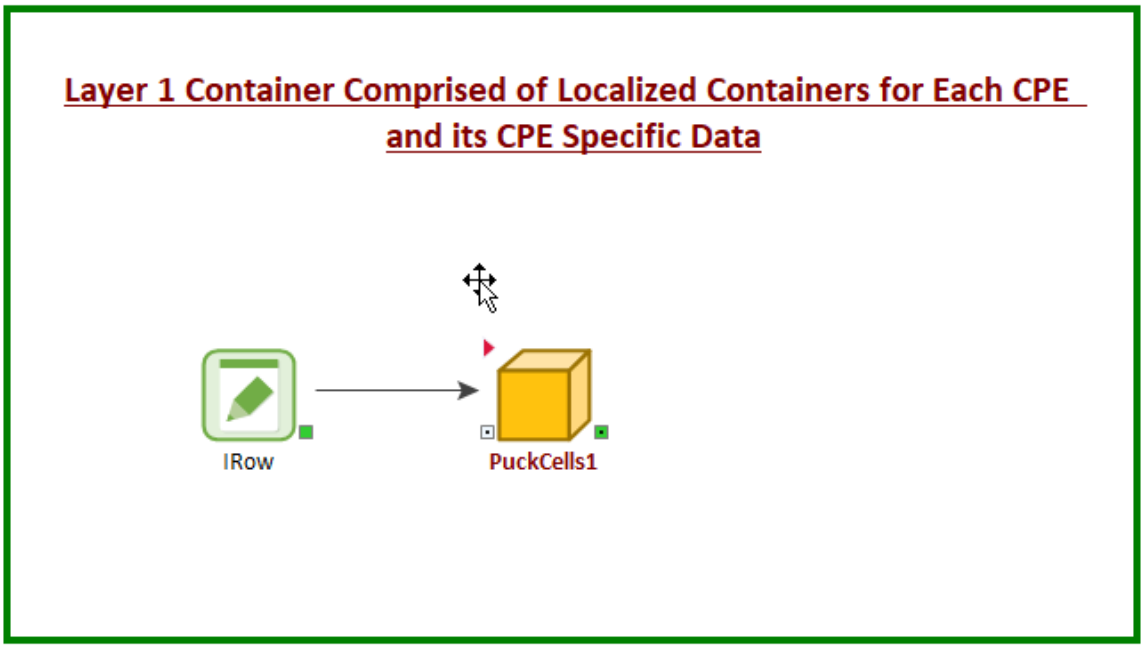


Figure 8: DLM Optimization Model Containers for 10-Concentric Cylindrical Cells Comprising Layer 1

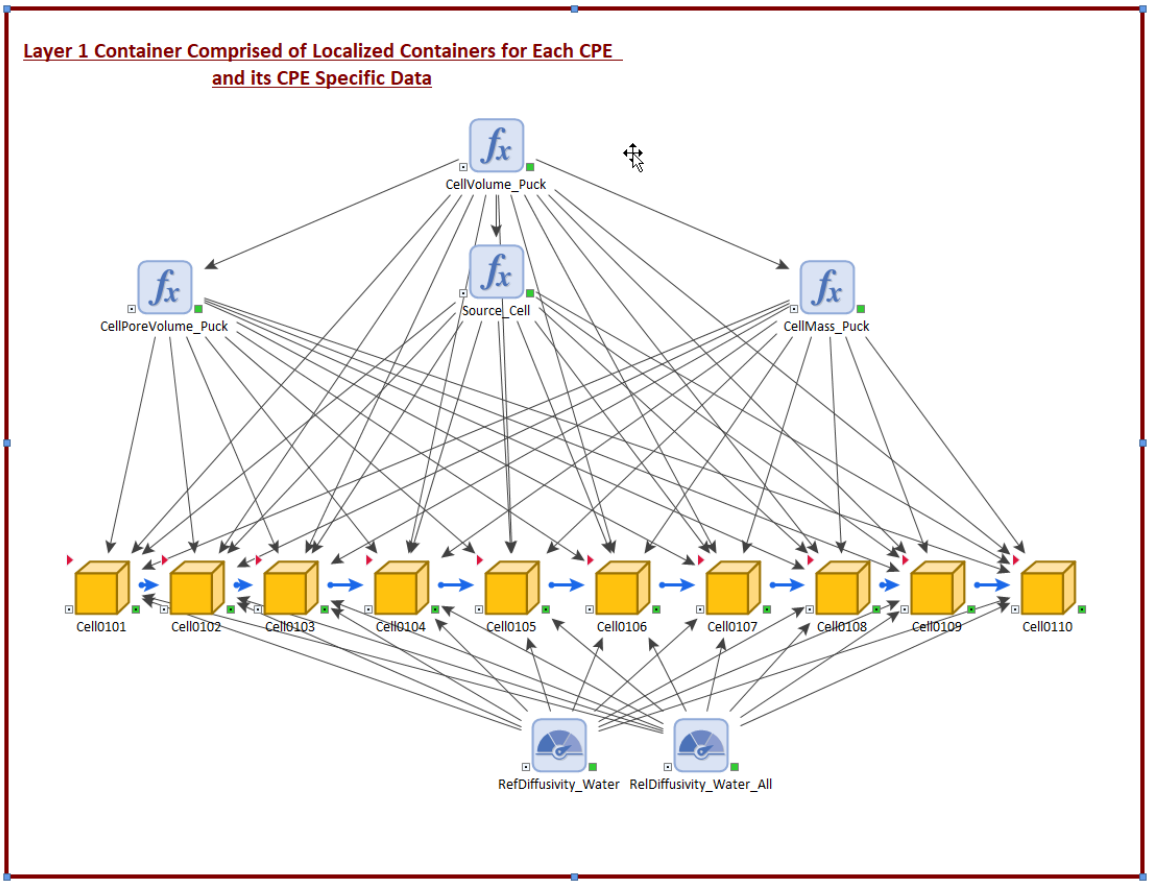


Figure 9: DLM Optimization Model Calculation Container for Layer 1: Column 1

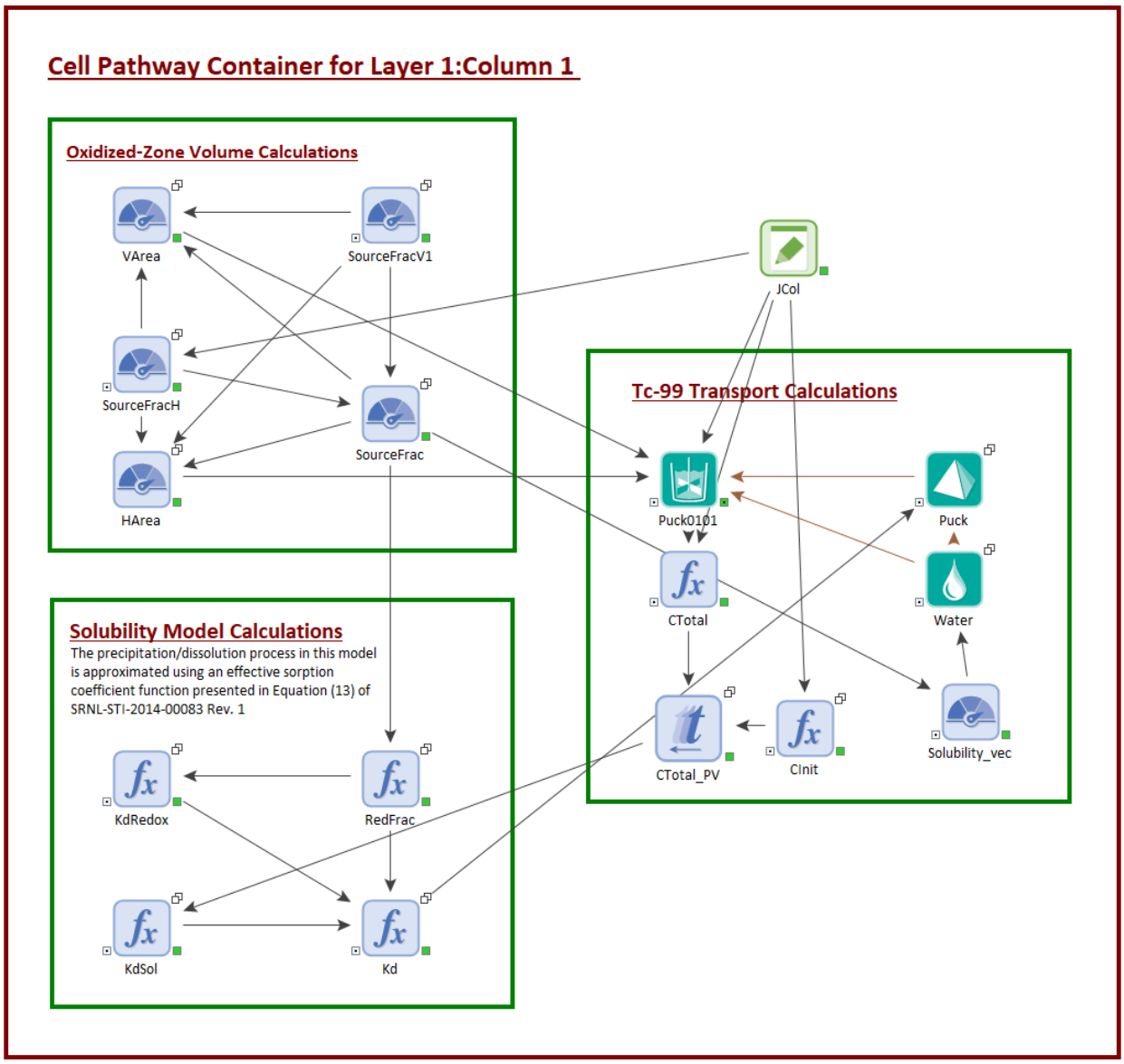
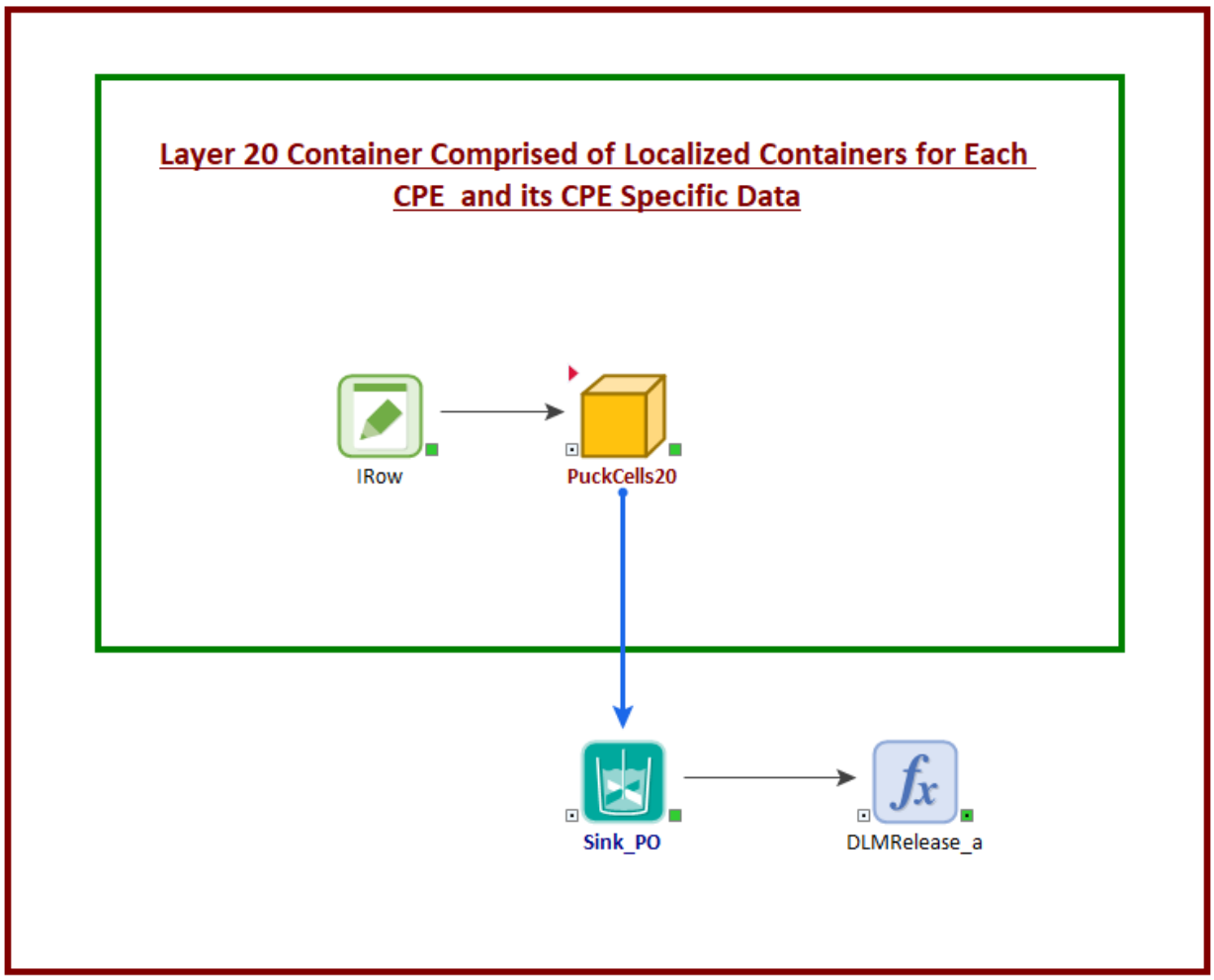


Figure 10: Sink Cell Pathway element for Capturing Releases from DLM Optimization Model Cell Network Layer 20



Model Input

Except for the solubility limits which are calibrated with this model, most of the input values used to simulate Tc-99 release using the DLM Optimization Model are all obtained from the SREL DLM studies for the 45-45-10_LeHigh saltstone sample, SDU 2A (Core A), and SDU 2A (Core B) samples as noted below in Table 1, Table 2, and Table 3. The reducing capacity of the slag and the effective Tc-99 sorption coefficient function exponent are that same that have been used in the FY2016 SDF SA (SRR-CWDA-2016-00072). The permeant oxygen concentration is effectively set to zero to reflect the negligible oxygen content of the permeant used in the DLM tests. Single-valued parameters used in the model are listed in Table 1, Table 2, and Table 3, along with the names of the GoldSim data elements in which they are found and their locations within the model as well as the GoldSim Container Elements in which they are located (see Figure 3). Note for this modeling study because of the relatively short duration of the ongoing testing and the slow leaching of Tc-99 from the simulant samples, chemically the samples are assumed to be in a Reduced Young stage or Reduced Moderately-Aged stage therefore, solubility constraints not sorption (and the associated K_d value listed in Table 1, Table 2, and Table 3), are assumed to control the Tc-99 release during the optimization runs.

The experimental data used for defining the system-model flow field and for comparing the leaching study results with the modeling results are presented in Table 4 for the 45-45-10_LeHigh_B saltstone, Table 5 for the SDU 2A (Core A), and Table 6 for the SDU 2A (Core B). The data presented in Table 4, Table 5, and Table 6, includes the test duration at the end of each sampling interval, the cumulative pore volumes of water that have passed through the saltstone simulant by the end of each interval, the average volumetric flow rate over each sampling interval, the cumulative percentage of the Tc-99 inventory released from the simulant by the end of each sampling interval, and the leachate's Tc-99 concentration in each sample. The volumetric flow rates used in the model simulations (see Table 4, Table 5, and Table 6) are also plotted with respect to test duration time and pore volumes of water that have passed through the DLM sample with the plots depicted in Figure 11 and Figure 12 for the 45-45-10_LeHigh saltstone sample, Figure 13 and Figure 14 for the SDU 2A (Core A) sample, and Figure 15 and Figure 16 for the SDU 2A (Core B) sample, respectively.

Table 1: Parameter Values from SREL DLM Test of 45-45-10 LeHigh_B Saltstone Simulant Core

Parameter Description or Variable Name	Parameter Value	GoldSim Container	GoldSim Element
Bulk Density (g/cm^3)[1]	1.73	*Material\Solid_Puck*	*BulkDensity_Puck*
Porosity[1]	0.52	*Material\Solid_Puck*	*Porosity*
Kd (mL/g) - Reducing Environment[2]	0.01	*Material\Solid_Puck*	*Kd_red*
Kd (mL/g) - Oxidizing Environment[2]	0.5	*Material\Solid_Puck*	*Kd_ox*
Multiple_Solubilities	True	*ControlInput*	*Multiple_Solubilities*
Solubility Limit #1 (mol/L)[3]	9.7E-07	*Material\Solid_Puck*	*PuckSolubility1*
Solubility Limit #2 (mol/L)[3]	4.5E-07	*Material\Solid_Puck*	*PuckSolubility2*
Solubility Limit Transition Pore Volumes	1.5	*Material\Solid_Puck*	*Sol1_PV*
Slag Reduction Capacity (meq e-/g)[4]	0.607	*Material\Solid_Puck*	*CSlag*
Water Oxygen Concentration (meq e-/g)	1.0E-15[5]	*Material\Solid_Puck*	*Coxy*
Sorption Coefficient Function Exponent	200	*Material\Solid_Puck*	*SolPower*
Effective Diffusion Coefficient (cm^2/s)	3.2E-07	*Material*	*RefDiffusivity_Water_Puck*
Inventory (pCi)[1]	655610	*Source*	*Source_pCi*
Puck Dameter (cm)[1]	5.1	*Geometry\SaltstoneMonolithData*	*PuckDiameter*
Puck Height (cm)[1]	2.5	*Geometry\SaltstoneMonolithData*	*PuckThickness*

[1] This data was taken from DLM_Data_Consolidated 8-5-18.xlsx.
[2] This data was taken from SRNL-STI-2013-00280.
[3] This data was calibrated using the GoldSim optimization option.
[4] This data was taken from SRNL-STI-2014-00083.
[5] Effectively 0 meq e-/g, but circumvents a zero-divide.

Table 2: Parameter Values from SREL DLM Test of SDU 2A (Core A) Saltstone Simulant

Parameter Description or Variable Name	Parameter Value	GoldSim Container	GoldSim Element
Bulk Density (g/cm^3)[1]	1.7	*Material\Solid_Puck*	*BulkDensity_Puck*
Porosity[1]	0.60	*Material\Solid_Puck*	*Porosity*
Kd (mL/g) - Reducing Environment[2]	0.01	*Material\Solid_Puck*	*Kd_red*
Kd (mL/g) - Oxidizing Environment[2]	0.5	*Material\Solid_Puck*	*Kd_ox*
Multiple_Solubilities	True	*ControlInput*	*Multiple_Solubilities*
Solubility Limit #1 (mol/L)[3]	2.0E-07	*Material\Solid_Puck*	*PuckSolubility1*
Solubility Limit #2 (mol/L)[3]	7.4E-08	*Material\Solid_Puck*	*PuckSolubility2*
Solubility Limit Transition Pore Volumes	6.3	*Material\Solid_Puck*	*Sol1_PV*
Slag Reduction Capacity (meq e-/g)[4]	0.607	*Material\Solid_Puck*	*CSlag*
Water Oxygen Concentration (meq e-/g)	1.0E-15[5]	*Material\Solid_Puck*	*Coxy*
Sorption Coefficient Function Exponent	200	*Material\Solid_Puck*	*SolPower*
Effective Diffusion Coefficient (cm^2/s)	3.2E-07	*Material*	*RefDiffusivity_Water_Puck*
Inventory (pCi)[1]	6.22E+05	*Source*	*Source_pCi*
Puck Dameter (cm)[1]	5.0	*Geometry\SaltstoneMonolithData*	*PuckDiameter*
Puck Height (cm)[1]	2.5	*Geometry\SaltstoneMonolithData*	*PuckThickness*

[1] This data was taken from DLM_Data_Consolidated 8-5-18.xlsx.
[2] This data was taken from SRNL-STI-2013-00280.

[3] This data was calibrated using the GoldSim optimization option.
[4] This data was taken from SRNL-STI-2014-00083.
[5] Effectively 0 meq e-/g, but circumvents a zero-divide.

Table 3: Parameter Values from SREL DLM Test of SDU 2A (Core B) Saltstone Simulant

Parameter Description or Variable Name	Parameter Value	GoldSim Container	GoldSim Element
Bulk Density (g/cm^3)[1]	1.95	*Material\Solid_Puck*	*BulkDensity_Puck*
Porosity[1]	0.60	*Material\Solid_Puck*	*Porosity*
Kd (mL/g) - Reducing Environment[2]	0.01	*Material\Solid_Puck*	*Kd_red*
Kd (mL/g) - Oxidizing Environment[2]	0.5	*Material\Solid_Puck*	*Kd_ox*
Multiple_Solubilities	True	*ControlInput*	*Multiple_Solubilities*
Solubility Limit #1 (mol/L)[3]	3.3E-07/4.7E-07	*Material\Solid_Puck*	*PuckSolubility1*
Solubility Limit #2 (mol/L)[3]	6.5E-07/NA	*Material\Solid_Puck*	*PuckSolubility2*
Solubility Limit Transition Pore Volumes	0.553/NA	*Material\Solid_Puck*	*Sol1_PV*
Slag Reduction Capacity (meq e-/g)[4]	0.607	*Material\Solid_Puck*	*CSlag*
Water Oxygen Concentration (meq e-/g)	1.0E-15[5]	*Material\Solid_Puck*	*Coxy*
Sorption Coefficient Function Exponent	200	*Material\Solid_Puck*	*SolPower*
Effective Diffusion Coefficient (cm^2/s)	3.2 E-07	*Material*	*RefDiffusivity_Water_Puck*
Inventory (pCi)[1]	6.22E+05	*Source*	*Source_pCi*
Puck Dameter (cm)[1]	5.0	*Geometry\SaltstoneMonolithData*	*PuckDiameter*
Puck Height (cm)[1]	2.5	*Geometry\SaltstoneMonolithData*	*PuckThickness*

[1] This data was taken from DLM_Data_Consolidated 8-5-18.xlsx.
[2] This data was taken from SRNL-STI-2013-00280.
[3] This data was calibrated using the GoldSim optimization option.
[4] This data was taken from SRNL-STI-2014-00083.
[5] Effectively 0 meq e-/g, but circumvents a zero-divide.

Table 4: Time Series of Leachate Release Data Parameter Values from SREL DLM Test of 45-45-10 LeHigh Saltstone Simulant Core

Test[2, 4] Duration (hours)	Cumulative[1] Pore Volumes	Flow Rate [2] (mL/hour)	Tc-99[3, 4] Release	Leachate Concentration (mol/L)
0	0	1.797E-02	0.0%	0
105.2	0.071	2.258E-02	0.1%	2.42E-07
244.7	0.190	1.631E-02	0.7%	7.61E-07
412.7	0.293	8.656E-03	1.2%	6.09E-07
598.7	0.354	1.061E-02	1.6%	9.51E-07
746.7	0.413	8.036E-03	1.9%	8.48E-07
914.7	0.464	5.476E-03	2.2%	9.66E-07
1082.7	0.498	7.179E-03	2.4%	8.54E-07
1199.7	0.530	5.863E-03	2.7%	9.73E-07
1535.7	0.604	5.030E-03	3.2%	1.09E-06
1700.7	0.635	1.393E-02	3.4%	9.89E-07
1868.7	0.723	1.030E-02	4.1%	1.16E-06
2372.7	0.919	1.554E-02	5.1%	7.50E-07
2540.7	1.017	2.399E-02	5.7%	8.94E-07
2708.7	1.169	1.815E-02	6.4%	6.79E-07
2876.7	1.284	2.554E-02	6.9%	5.30E-07
3044.7	1.445	2.554E-02	7.4%	4.71E-07
3212.7	1.607	2.708E-02	7.9%	4.44E-07
3380.7	1.778	2.679E-02	8.3%	3.69E-07
3548.7	1.948	2.679E-02	8.7%	3.09E-07
3716.7	2.117	2.131E-02	9.0%	2.71E-07
3884.7	2.252	2.113E-02	9.2%	2.55E-07
4052.7	2.385	1.976E-02	9.4%	2.47E-07
4220.7	2.510	2.060E-02	9.7%	3.14E-07
4388.7	2.641	2.106E-02	9.9%	2.37E-07
4604.7	2.812	2.177E-02	10.3%	2.90E-07
4892.7	3.048	2.159E-02	10.7%	2.63E-07
5564.7	3.595	2.119E-02	11.8%	3.07E-07
5900.7	3.863	2.138E-02	12.4%	3.1E-07
6284.7	4.172	1.786E-02	13.0%	3.1E-07
6452.7	4.285	1.786E-02	13.3%	3.1E-07

[1] Found in the data properties element *Exp_CumPV* located in *CalibrationParameters*.
[2] Found in the lookup *Infiltration_transient* located in *TransportProcessParameters*.
[3] Found in the data properties element *Exp_Tc_Released* located in *CalibrationParameters*.
[4] Also found in the lookup table *CumPctReleased* located in *Archive*.

Table 5: Time Series of Leachate Release Data Parameter Values from SREL DLM Test of SDU 2A (Core A) Saltstone Simulant

Test[2, 4] Duration (hours)	Cumulative[1] Pore Volumes	Flow Rate [2] (mL/hour)	Tc-99[3, 4] Release	Leachate Concentration (mol/L)
0	0	3.750E-01	0%	0
24.0	0.306	3.123E-01	0.6%	2.51E-07
47.5	0.555	1.931E-01	0.9%	1.66E-07
78.0	0.755	2.000E-01	1.2%	1.51E-07
102.0	0.918	1.505E-01	1.4%	1.28E-07
124.0	1.030	1.321E-01	1.5%	1.09E-07
150.5	1.149	9.946E-02	1.6%	1.09E-07
169.0	1.211	8.803E-02	1.6%	1.09E-07
191.8	1.280	6.476E-02	1.7%	1.09E-07
223.3	1.349	5.079E-02	1.7%	1.13E-07
290.1	1.464	1.505E-02	1.9%	1.29E-07
506.1	1.574	1.800E-02	2.0%	1.51E-07
618.8	1.643	1.175E-02	2.0%	1.13E-07
789.8	1.712	9.913E-03	2.1%	1.35E-07
1077.3	1.808	5.062E-03	2.2%	1.57E-07
1219.6	1.833	1.805E-02	2.3%	1.82E-07
1388.6	1.936	1.395E-02	2.4%	1.59E-07
1721.1	2.094	9.349E-03	2.7%	2.38E-07
1936.1	2.162	1.052E-02	2.8%	2.35E-07
2349.6	2.310	1.109E-02	3.0%	1.75E-07
2541.6	2.382	1.043E-02	3.2%	1.92E-07
2799.6	2.473	1.246E-02	3.3%	1.91E-07
3165.6	2.628	1.269E-02	3.6%	2.36E-07
3332.6	2.700	1.275E-02	3.7%	2.41E-07
3666.6	2.845	1.427E-02	4.0%	2.09E-07
3954.6	2.984	1.295E-02	4.2%	2.05E-07
5236.6	3.548	1.398E-02	5.1%	1.95E-07
6148.6	3.981	1.929E-03	5.7%	1.85E-07
6796.6	4.023	1.786E-02	5.8%	1.91E-07
6964.6	4.125	4.464E-02	6.0%	2.02E-07
7300.6	4.635	7.523E-03	6.8%	2.07E-07
9028.6	5.076	7.292E-03	7.5%	1.85E-07
9988.6	5.314	8.482E-03	7.8%	2.01E-07
10324.6	5.410	8.631E-03	8.0%	1.78E-07
10660.6	5.509	8.631E-03	8.2%	2.22E-07
10996.6	5.607	5.327E-03	8.3%	1.69E-07
11332.6	5.668	8.929E-03	8.4%	1.65E-07
11668.6	5.770	9.524E-03	8.4%	9.41E-08
11836.6	5.824	1.065E-02	8.5%	1.39E-07
12052.6	5.902	9.167E-03	8.6%	1.56E-07
12172.6	5.940	8.601E-03	8.6%	1.47E-07
12508.6	6.038	4.464E-03	8.7%	1.32E-07
12844.6	6.089	7.981E-03	8.8%	1.19E-07
13057.6	6.147	8.130E-03	8.9%	1.22E-07

13180.6	6.180	9.524E-03	8.9%	1.15E-07
13348.6	6.235	8.929E-03	8.9%	1.11E-07
13516.6	6.286	3.571E-02	9.0%	1.22E-07
13684.6	6.489	3.125E-02	9.2%	1.39E-07
13876.6	6.693	3.472E-02	9.5%	1.52E-07
14020.6	6.863	3.869E-02	9.5%	6.61E-08
14188.6	7.084	3.869E-02	9.6%	5.40E-08
14356.6	7.304	2.679E-02	9.7%	5.05E-08
14524.6	7.457	2.976E-02	9.8%	5.10E-08
14692.6	7.627	2.679E-02	9.9%	4.90E-08
14860.6	7.780	2.083E-02	9.9%	5.17E-08
15196.6	8.017	2.127E-02	10.0%	4.02E-08
15361.6	8.137	2.024E-02	10.0%	4.13E-08
15697.6	8.367	2.030E-02	10.1%	4.93E-08
16201.6	8.715	1.679E-02	10.3%	4.82E-08
16369.6	8.810	2.512E-02	10.3%	4.73E-08
16537.6	8.954	2.536E-02	10.4%	4.90E-08
16705.6	9.098	2.583E-02	10.4%	4.31E-08
16873.6	9.246	2.619E-02	10.5%	4.28E-08
17041.6	9.395	2.440E-02	10.5%	6.18E-08
17209.6	9.534	2.560E-02	10.6%	5.81E-08
17377.6	9.680	2.929E-02	10.7%	4.49E-08
17545.6	9.847	2.661E-02	10.7%	4.33E-08
17713.6	9.999	3.046E-02	10.8%	4.10E-08
17929.6	10.223	3.160E-02	10.8%	4.40E-08
18217.6	10.532	1.756E-02	10.9%	4.09E-08
18889.6	10.932	1.949E-02	11.1%	5.82E-08
19225.6	11.155	1.857E-02	11.2%	5.35E-08
19609.6	11.397	1.815E-02	11.3%	6.26E-08
19777.6	11.500	1.815E-02	11.4%	6.85E-08

[1] Found in the data properties element *Exp_CumPV* located in *CalibrationParameters*.
[2] Found in the lookup *Infiltration_transient* located in *TransportProcessParameters*.
[3] Found in the data properties element *Exp_Tc_Released* located in *CalibrationParameters*.
[4] Also found in the lookup table *CumPctReleased* located in *Archive*.

Table 6: Time Series of Leachate Release Data Parameter Values from SREL DLM Test of SDU 2A (Core B) Saltstone Simulant

Test[2, 4] Duration (hours)	Cumulative[1] Pore Volumes	Flow Rate [2] (mL/hour)	Tc-99 [3, 4] Release %	Leachate Concentration (mol/L)
0	0	6.349E-02	0.0%	0
43.0	0.093	1.690E-02	0.2%	2.05E-07
97.5	0.124	7.528E-03	0.2%	2.60E-07
165.2	0.141	6.071E-03	0.3%	2.68E-07
193.2	0.147	1.323E-02	0.3%	3.34E-07
287.0	0.189	1.292E-02	0.4%	2.70E-07
503.0	0.284	4.464E-03	0.6%	3.25E-07
671.0	0.309	2.431E-03	0.7%	2.69E-07
815.0	0.321	2.439E-03	0.7%	2.52E-07
856.0	0.325	1.517E-03	0.7%	2.35E-07
1093.2	0.337	2.250E-03	0.7%	4.46E-07
1235.5	0.348	4.497E-03	0.8%	2.62E-07
1404.5	0.373	4.361E-03	0.8%	3.50E-07
1737.0	0.423	4.140E-03	1.0%	3.70E-07
1952.0	0.453	3.628E-03	1.1%	3.60E-07
2365.5	0.504	3.385E-03	1.2%	3.76E-07
2557.5	0.526	3.140E-03	1.3%	4.09E-07
2815.5	0.553	2.596E-03	1.4%	4.32E-07
3181.5	0.586	2.934E-03	1.5%	4.85E-07
3348.5	0.602	2.784E-03	1.6%	4.90E-07
3682.5	0.634	2.569E-03	1.7%	4.64E-07
3970.5	0.659	6.318E-03	1.8%	4.72E-07
5252.5	0.934	8.224E-04	2.8%	4.64E-07
6164.5	0.960	3.086E-04	2.9%	4.84E-07
6812.5	0.966	4.464E-03	2.9%	4.26E-07
6980.5	0.992	2.976E-03	3.0%	3.69E-07
7316.5	1.026	2.025E-03	3.1%	4.36E-07
9044.5	1.145	1.927E-03	3.5%	4.30E-07
10004.5	1.207	2.381E-03	3.8%	5.01E-07
10340.5	1.235	2.440E-03	3.9%	4.33E-07
10676.5	1.262	3.571E-03	4.0%	4.92E-07
11012.5	1.303	2.440E-03	4.1%	4.29E-07
11348.5	1.331	2.976E-03	4.2%	3.11E-07
11684.5	1.365	3.571E-03	4.3%	2.97E-07
11852.5	1.385	3.241E-03	4.3%	2.46E-07
12068.5	1.409	3.083E-03	4.4%	2.36E-07
12188.5	1.422	2.887E-03	4.4%	2.41E-07
12524.5	1.455	1.518E-03	4.5%	2.32E-07
12860.5	1.472	2.560E-03	4.5%	2.10E-07
13196.5	1.501	1.161E-03	4.5%	2.06E-07
13532.5	1.514	2.560E-03	4.6%	2.17E-07
13700.5	1.529	2.500E-03	4.6%	2.29E-07
13868.5	1.543	2.500E-03	4.6%	2.06E-07

[1] Found in the data properties element *Exp_CumPV* located in *CalibrationParameters*.

[2] Found in the lookup *Infiltration_transient* located in *TransportProcessParameters*.

[3] Found in the data properties element *Exp_Ic_Released* located in *CalibrationParameters*.

[4] Also found in the lookup table *CumPctReleased* located in *Archive*.

Figure 11: Time-Averaged Volumetric Flow Rates Versus Test Duration Time for 45-45-10 LeHigh_B

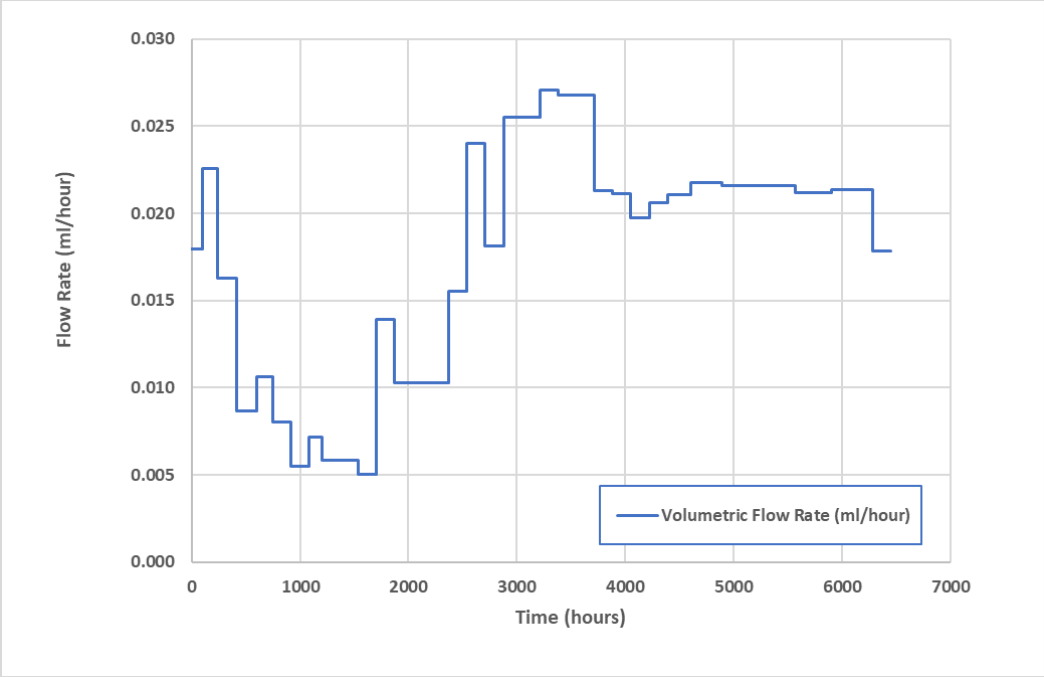


Figure 12: Time-Averaged Volumetric Flow Rates Versus Cumulative Pore Volumes for 45-45-10 LeHigh_B

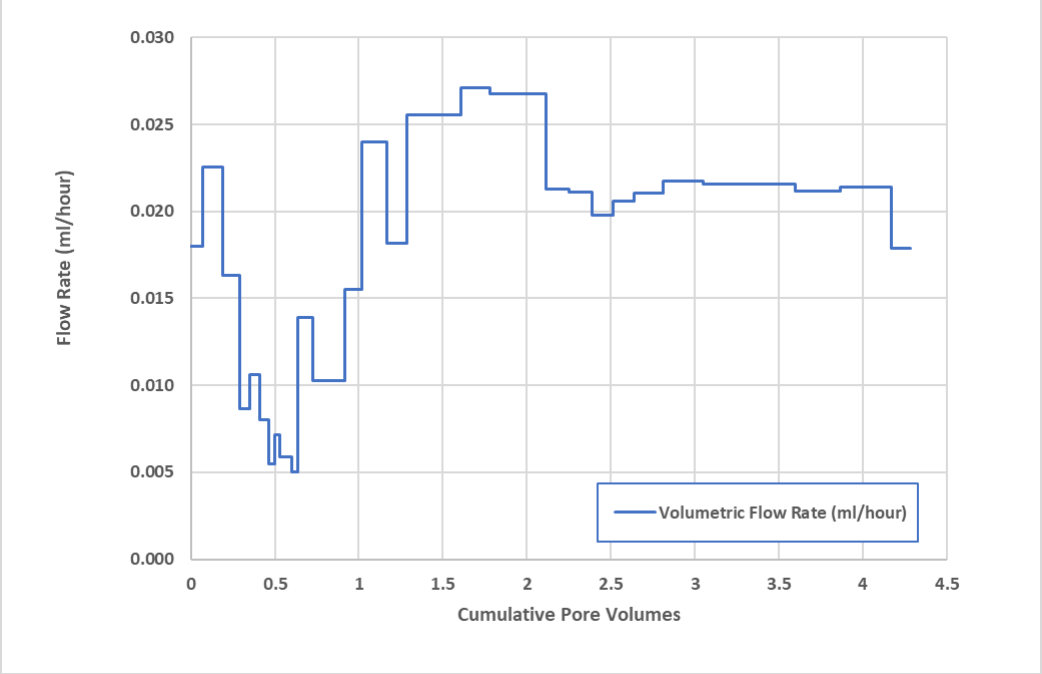


Figure 13: Time-Averaged Volumetric Flow Rates Versus Test Duration Time for SDU 2A (Core A)

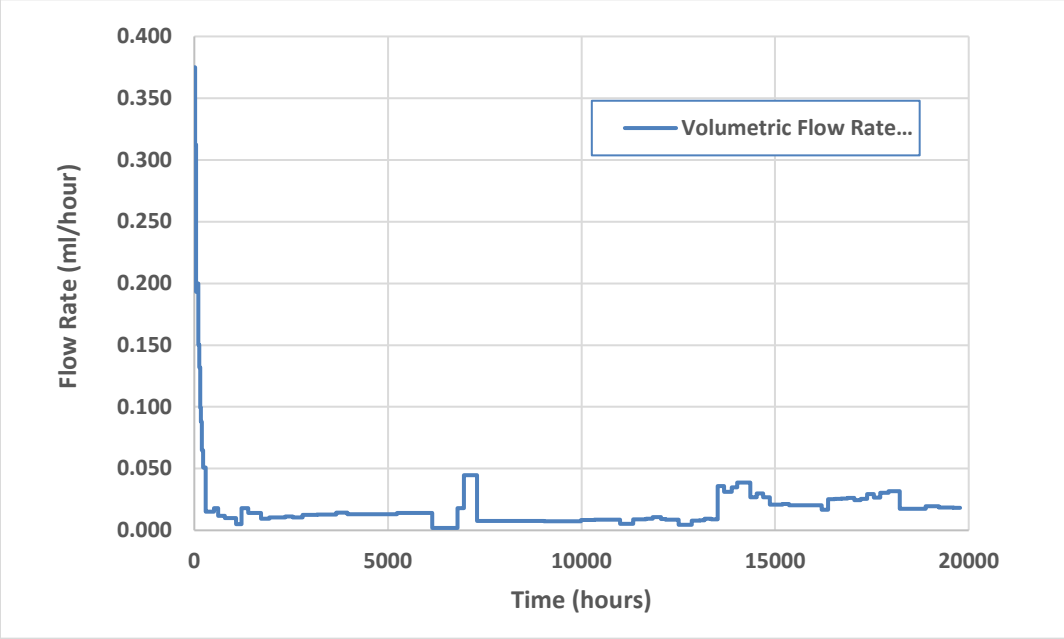


Figure 14: Time-Averaged Volumetric Flow Rates Versus Cumulative Pore Volumes for SDU 2A (Core A)

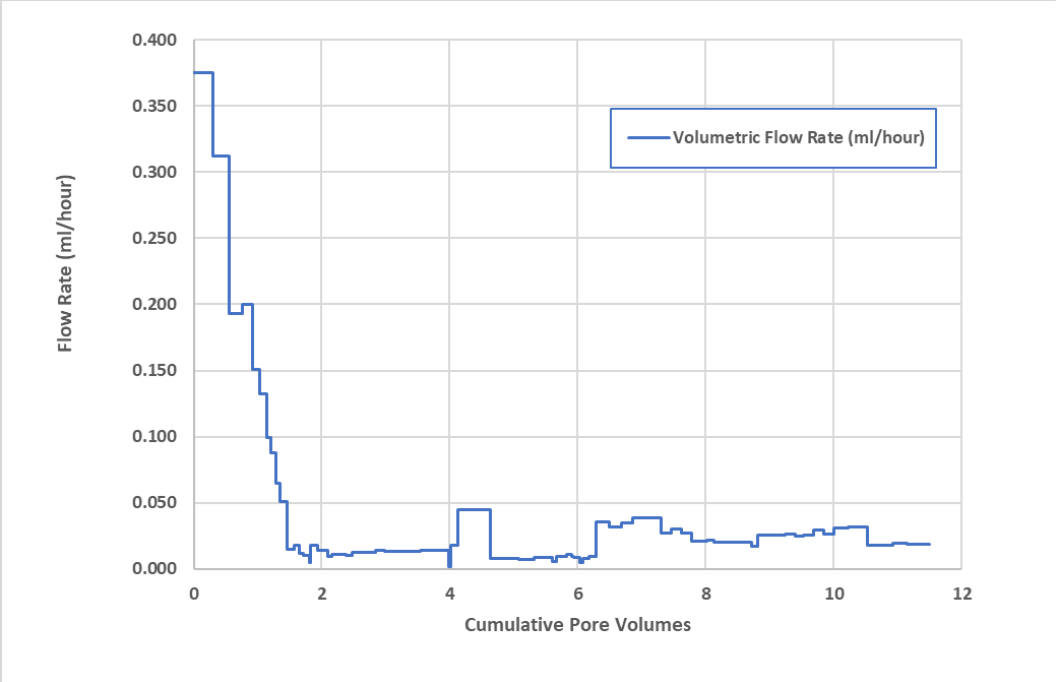


Figure 15: Time-Averaged Volumetric Flow Rates Versus Test Duration Time Volumes for SDU 2A (Core B)

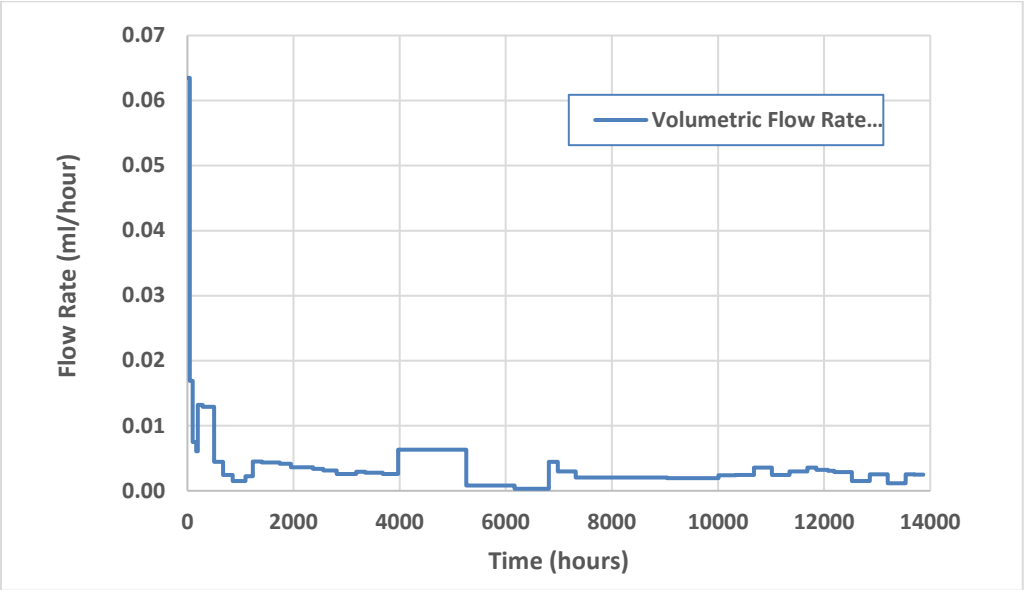
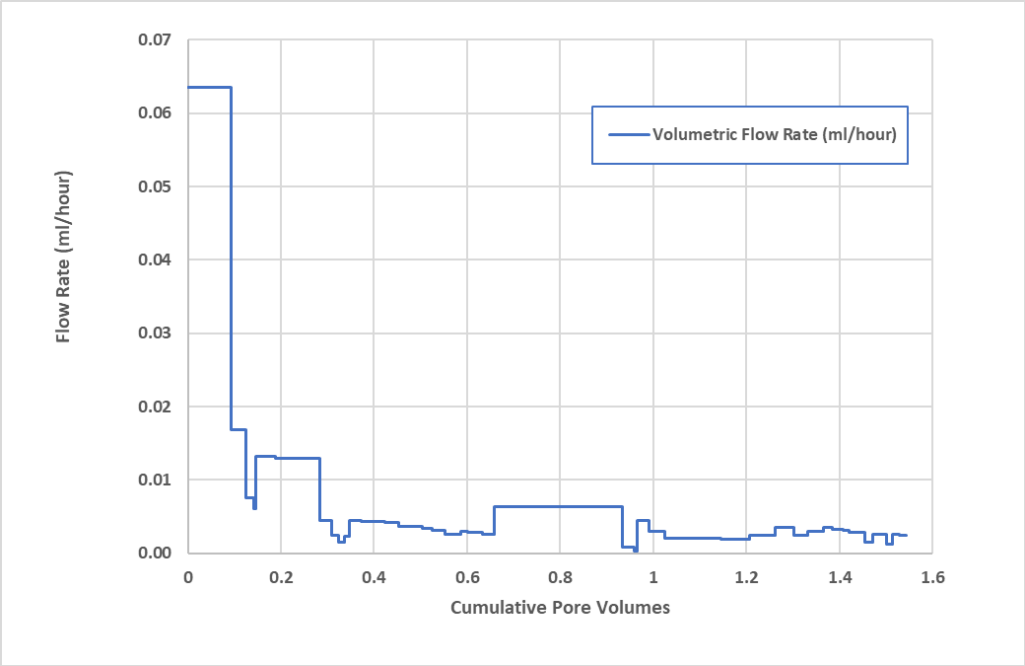


Figure 16: Time-Averaged Volumetric Flow Rates Versus Cumulative Pore Volumes for SDU 2A (Core B)



Optimization Runs

This section describes the results of four optimization simulations that were used to calibrate and compare calibrated values of solubility limits in conjunction with the SREL DLM study test results pertaining to Tc-99 releases from the 45-45-10_LeHigh (Sample B) saltstone simulant and SDU 2A core samples (Core A and Core B). The first three optimization runs assume the solubility limit transitions as the pH falls below 11. The fourth run is used to compare how well the test data can be fitted omitting the solubility limit transition when calibrating to the SDU 2A (Core B) test data.

The first optimization run (Run #1) was used to calibrate two solubility limits for the GoldSim model of the 45-45-10_LeHigh (Sample B) saltstone simulant sample, one reflecting leachate samples with pH values at or above 11 and the second for samples with pH values below 11. The transition was implemented at a cumulative leachate release of 1.5 pore-volumes which is consistent with the sampling event in which the pH drops below 11. The optimal initial solubility limit calculated by the GoldSim optimizer was 9.7E-07 mol/L, and the optimal transition solubility limit calculated by the GoldSim optimizer, was 4.5E-07 mol/L (Table 5) and provided a good match with the SREL results (see Figure 17).

The second optimization run (Run #2) was used to calibrate two values of solubility limits for the GoldSim model of the SDU 2A (Core A) saltstone sample, one reflecting leachate samples with pH values at or above 11 and the second for samples with pH values below 11. The transition was implemented at a cumulative leachate release of 6.3 pore-volumes which is consistent with the sampling event in which the pH drops below 11. The optimal initial solubility limit calculated by the GoldSim optimizer was 2.0E-07 mol/L, and the optimal transition solubility limit calculated by the GoldSim optimizer was 7.4E-08 mol/L (Table 5) and provided a good match with the SREL results (see Figure 17).

The third optimization run (Run #3) was used to calibrate two values of solubility limits for the GoldSim model of the SDU 2A (Core B) saltstone sample, one reflecting leachate samples with pH values at or above 11 and the second for samples with pH values below 11. The transition was implemented at a cumulative leachate release of 0.553 pore-volumes which is consistent with the sampling event in which the pH drops below 11. The optimal initial solubility limit calculated by the GoldSim optimizer was 3.3E-07 mol/L, and the optimal transition solubility limit calculated by the GoldSim optimizer was 6.5E-07 mol/L (Table 5) and provided a good match with the SREL results (see Figure 18). Noting that the transition solubility limit increased instead of decreasing in a manner similar (and more expected) to the first two runs, it is possible that the pH values for this run may not be reliable. It is also possible that there was an unexpected change in Eh. Since the Eh sampling may not be reliable due to oxygen exposure of the leachate samples this possibility is speculative. In case the sampled pH values are not reliable and a true transition has not been reached, rerunning the model without transitioning the solubility limit was tried. The fourth optimization run (Run #4) was performed calibrating only a single solubility limit for the GoldSim model of the SDU 2A (Core B) saltstone sample. The optimal solubility limit calculated by the GoldSim optimizer for the test duration was 4.7E-07 mol/L as shown in Table 5 and provided a good match with the SREL results (see Figure 19), but not as good as for the 2-parameter case.

The final MSE values for the 4 simulations are also presented in Table 5 to show that the calibrated parameters provide a reasonable approximation of the experimental results.

Table 5: Optimization Run Results for Calibrating Solubility Limit Values Based on Results of SREL Tc-99 DLM Simulant Sample Releases

Optimization Run Number	Sample	Initial Solubility Limit (mol/L)	Transition Solubility Limit (mol/L)	Solubility Transition Pore-Volume Count	Optimal Mean-Square Error
1	45-45-10 LeHigh (Sample B)	9.7E-07	4.5E-07	1.5	6.4E-06
2	SDU 2A (Core A)	2.0E-07	7.4E-08	6.3	6.0E-06
3	SDU 2A (Core B)[1]	3.3E-07	6.5E-07	0.553	5.1E-07
4	SDU 2A (Core B)[2]	4.7E-07	NA	NA	8.1E-06

[1] Two solubility limit model.
[2] One solubility limit model.

Figure 17: Comparisons of Tc-99 Percent Releases from 45-45-10_LeHigh Saltstone Simulant Sample B and SDU 2A Core A Sample

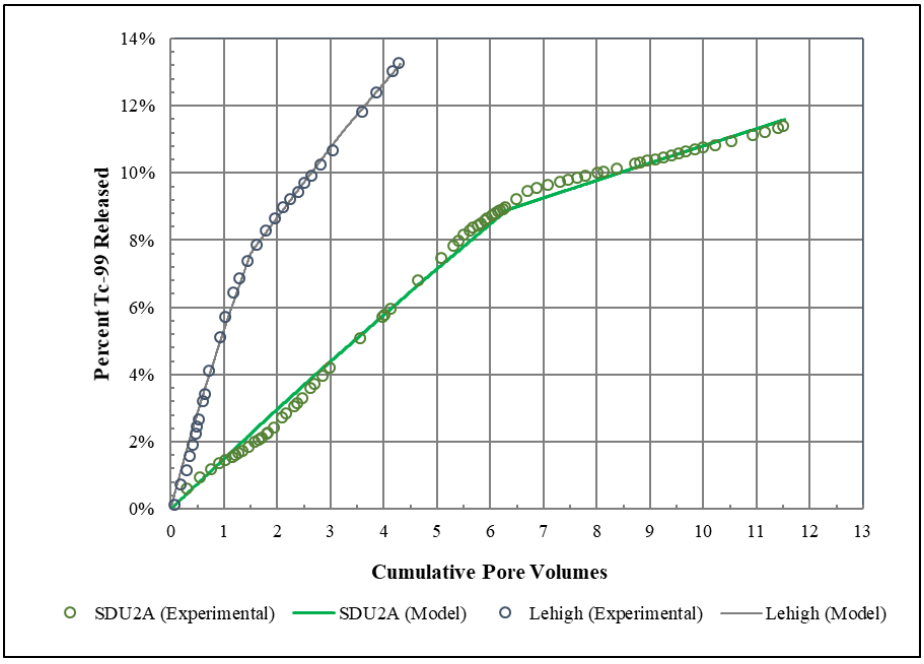


Figure 18: Comparisons of Tc-99 Percent Releases from SDU 2A Core B Sample Using Assuming a Transition in Solubility Limits When the Leachate pH Falls Below 11.0

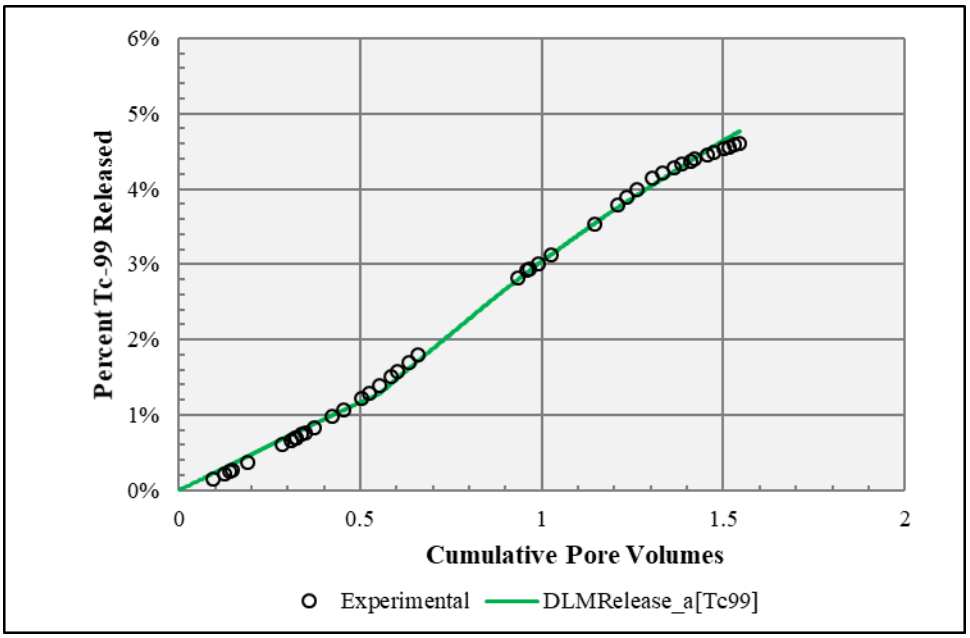
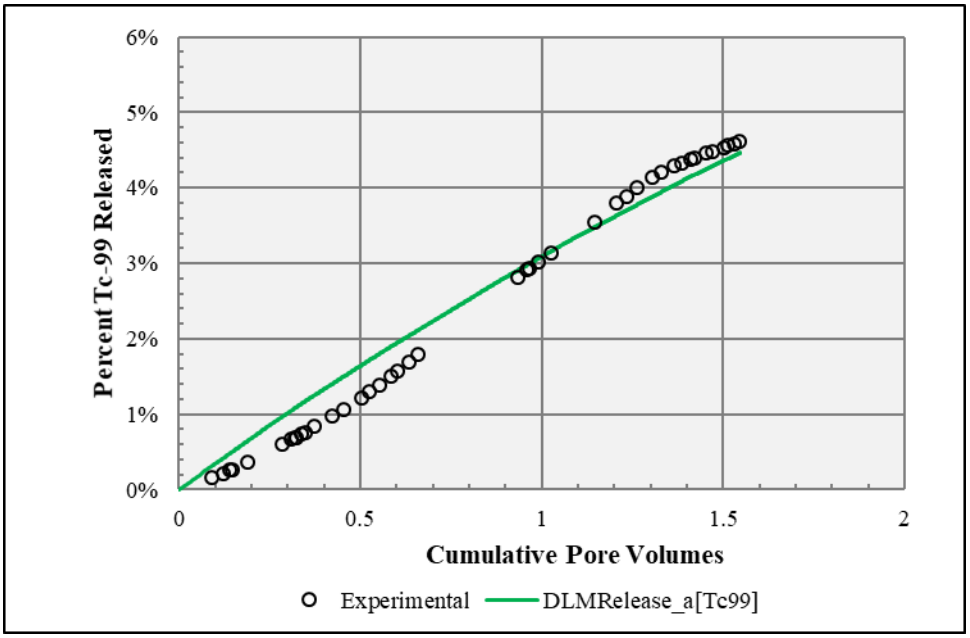


Figure 19: Comparisons of Tc-99 Percent Releases from SDU 2A Core B Sample Using Assuming No Transition in Solubility Limits When the Leachate pH Falls Below 11.0



Recommendations

The recommended path forward for choosing the Tc-99 solubility limits for the SDF PA model is to use the more conservative calibrated parameter results from Run #1 (based on 45-45-10_LeHigh Sample B saltstone simulant test) for the Compliance Model (MPAD Values) and Defense-in-Depth (Conservative) modeling cases. For the Best Estimate (Realistic) modeling case, the SDU 2A (Core A) calibrated solubility limits are recommended (see Table 6).

Table 6: Optimization Run Results for Calibrating Solubility Limit Values Based on Results of SREL Tc-99 DLM Simulant Sample Releases

Modeling Case	Saltstone pH >=11	Saltstone pH <11
Best Estimate (Realistic)	2.0E-07 mol/L	7.4E-08 mol/L
Compliance (MPAD)	9.7E-07 mol/L	4.5E-07 mol/L
Defense-in-Depth (Conservative)	9.7E-07 mol/L	4.5E-07 mol/L

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APPENDIX A

Geochemist Workbench (GWB) Results

Following are a set of figures reflecting the DLM testing results for Tc-99 releases with solubility limit curves generated using the Geochemist Workbench (SREL-R-18-004). Note solubility plots are generated for a pore solution in equilibrium with saltstone after 5 pore-volumes of permeant have been pushed through the saltstone. The pore solution chemistry is similar to what is seen after one pore-flush, and an ideal (i.e., ionic strength ≈ 0) condition solution (SREL-R-18-004).

Figure A-1: Comparison of Tc-99 Leachate Release Concentrations for LeHigh (Sample B) with GWB Generated Solubility of $TcO_2 \cdot 1.6H_2O$(s) as a Function of pH and Eh (5th PV DLM) (all other Tc minerals suppressed)

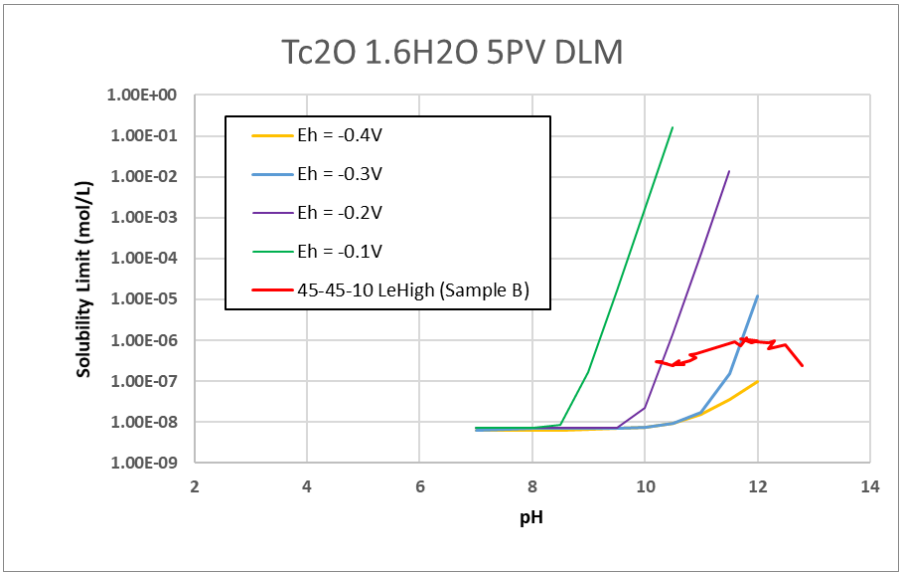

Figure A-2: Comparison of Tc-99 Leachate Release Concentrations for SDU 2A (Core A) with GWB Generated Solubility of $TcO_2 \cdot 1.6H_2O$(s) as a Function of pH and Eh (5th PV DLM) (all other Tc minerals suppressed)

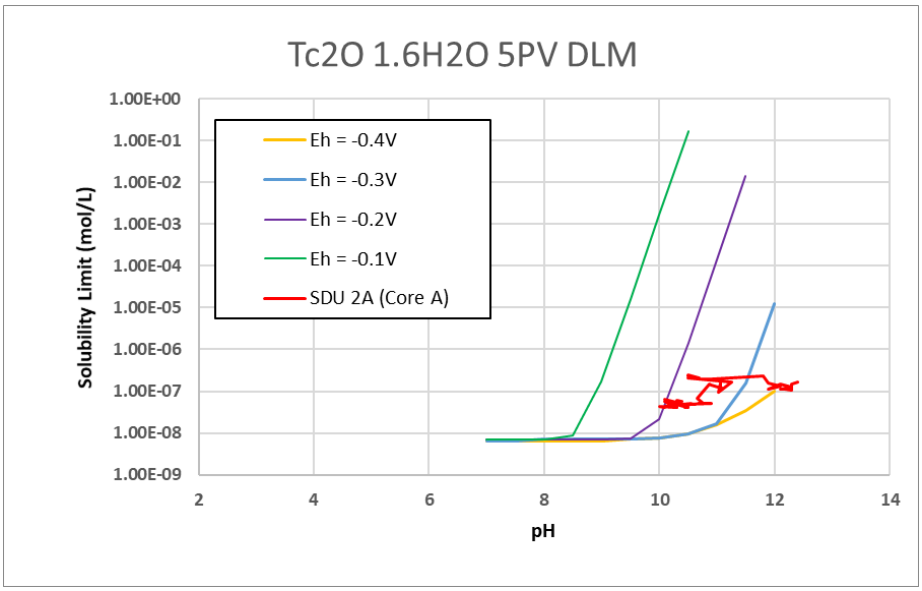

Figure A-3: Comparison of Tc-99 Leachate Release Concentrations for LeHigh (Sample B) with GWB Generated Solubility of $TcO_2 \cdot 2.0H_2O$(s) as a Function of pH and Eh (5th PV DLM) (all other Tc minerals suppressed)

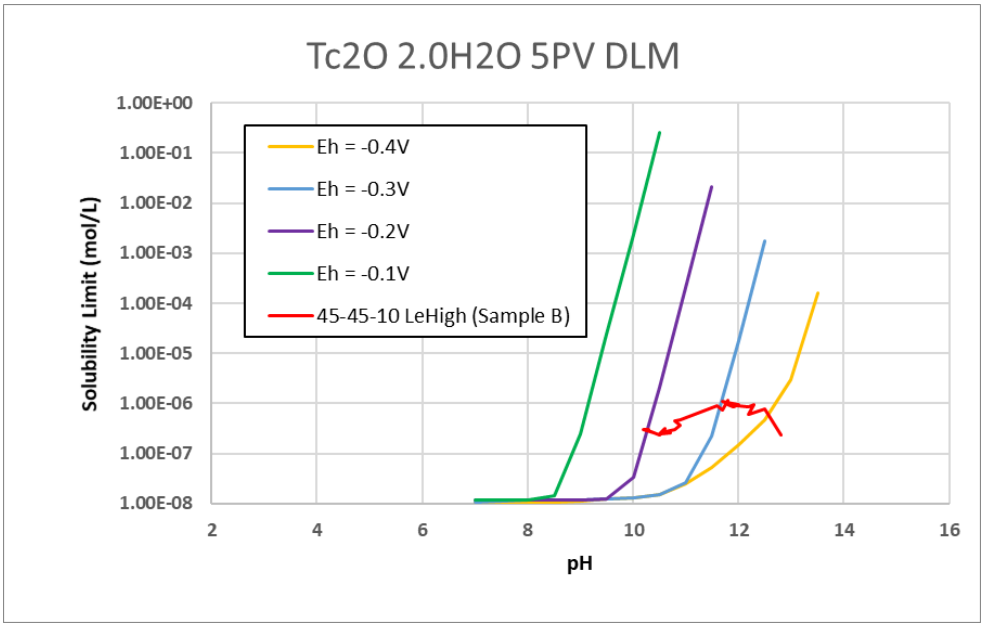

Figure A-4: Comparison of Tc-99 Leachate Release Concentrations for SDU 2A (Core A) with GWB Generated Solubility of $TcO_2 \cdot 2.0H_2O$(s) as a Function of pH and Eh (5th PV DLM) (all other Tc minerals suppressed)

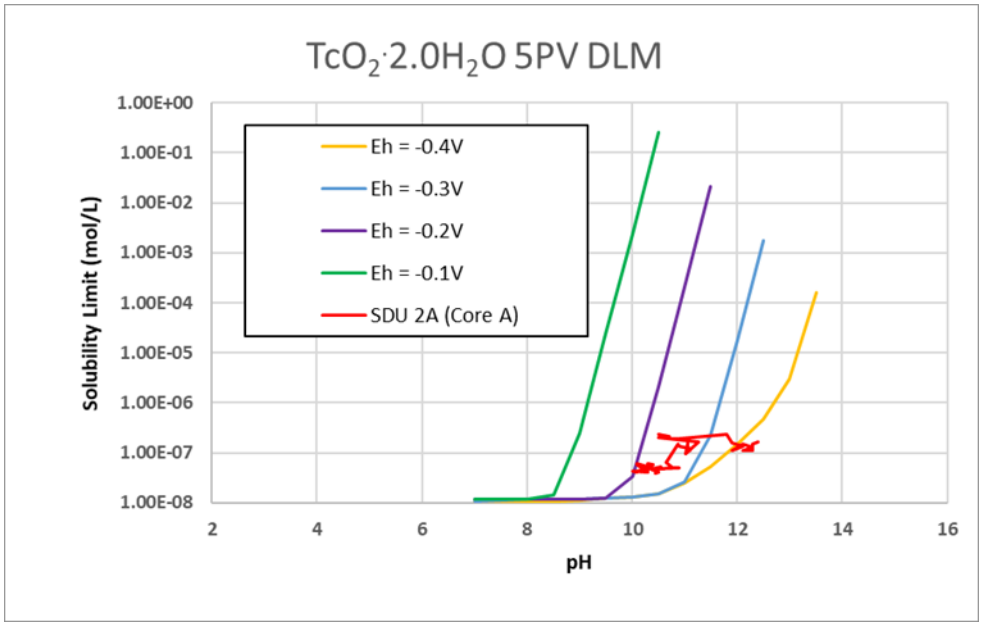

Figure A-5: Comparison of Tc-99 Percent Release for LeHigh (Sample B) with NO3 and Na Releases

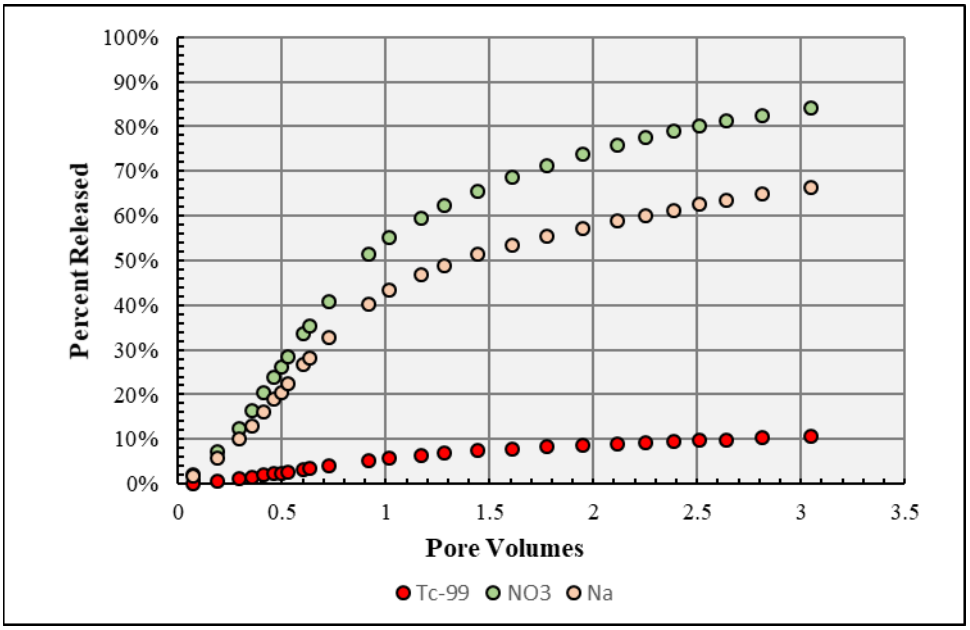

Figure A-6: Comparison of Tc-99 Percent Release for SDU 2A (Core A) with Cs-137 and NO3 Releases