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VERSE-LC TCCR Model Calibration using Batch 1A, Batch 2, and Batch 3 Column Data

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EXECUTIVE SUMMARY

The objective of this work is to utilize on-line gamma monitoring data collected during Tank Closure Cesium Removal (TCCR) Column Batch 3 operations to calibrate the parameters (essentially, the correction/dilution factors (CF or DF)) used in the VERSE-LC models for the prediction of cesium breakthrough while processing Savannah River Site (SRS) Tank 10H dissolved saltcake High-Level Waste through a packed cylindrical bed of Crystalline Silicotitanate (CST). Calibrated VERSE-LC models could be applied to aid in evaluating future TCCR operations. The following options are applied for calibrations:

a) VERSE-LC Batch 3 / Column D: Compare column D computed cesium effluent concentration with measured Batch 3/Column D effluent concentration to determine CF/DF for Batch 3. This simple option should provide the best comparison between prediction and measurements because Column D was a fresh (unused) column.
b) In addition to Option a), another approach can be pursued as follows:
    i. VERSE-LC Batch 1A / Columns A and B predictions could be utilized to provide effluent concentrations and determine CST loadings for columns A and B.
    ii. VERSE-LC Batch 2 / Columns B and C predictions could be utilized to provide effluent concentrations and determine CST loadings for columns B and C.
    iii. VERSE-LC Batch 3 / Column A: Compare column A predicted effluent concentration from Batch 1A with measured Batch 3/Column A effluent concentration to determine CF/DF for Batch 1A run and Batch 3.
    iv. VERSE-LC Batch 3 / Columns A and B: Compare column B effluent concentration from Batch 3/Columns A&B model run with measured Batch 3/Columns A&B effluent concentration. Since the CF factor for Batch 3 is selected based on the tea bag test, this comparison/adjustment Step allows the CF determination for Batch 2 run.

Option b) involves Columns A and B for Batch 3. Columns A and B were also used to process TCCR Batch 1A and Batch 2 and were partially loaded with cesium at the end of these operations. Therefore, VERSE-LC models must be set up to run Batch 1A and Batch 2 (steps i. and ii.) to establish the cesium concentration profiles in both columns A and B prior to performing steps iii. and iv.

<u>Calibration Results Summary</u>

1. Overall, the correction/dilution factors determined from teabag batch contact tests provide a good match to gamma monitoring data. Based on the results, a correction/dilution factor near 0.3 appears to be required to model the cesium removal performance.
2. Calibration of Batch 3/Column D indicates that based on the measured data it is not possible to determine which pore tortuosity is better.
3. Measured data for Batch 3/Columns A&B are low and inconsistent with gamma monitoring data for Batch 3/Column A.
4. Improvement in the gamma monitoring device is essential to future model calibrations.
5. Another approach for evaluating the data would be to use cumulative TCCR product sample data from the receipt tank (i.e., Tank 11H) to track the volume and Curie content over time rather than using on-line gamma data. VERSE-LC model can be calibrated to generate the same volume and Curie content.

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LIST OF ABBREVIATIONS

CF	Correction Factor
CST	Crystalline Silicotitanate
DF	Dilution Factor
HLW	High Level Waste
IX	Ion Exchange
SRNL	Savannah River National Laboratory
SRR	Savannah River Remediation
SRS	Savannah River Site
TCCR	Tank Closure Cesium Removal

1.0 Introduction

1.1 Background

The Tank Closure Cesium Removal (TCCR) at the Savannah River Site (SRS) is an “at-tank” process designed to remove cesium from aqueous tank waste. The High-Level Waste (HLW) requiring treatment was prepared from saltcake solids (primarily sodium hydroxide, nitrate, nitrite, carbonate, and sulfate salts) dissolved by the addition of water. Cesium is removed from the waste by ion exchange (IX) using the engineered IONSIV® R9120-B[1] form of the Crystalline Silicotitanate (CST) media.

The TCCR modular enclosure is deployed at SRS Tanks 10H and 11H in the H Tank Farm to remove cesium from Tank 10H. To date, three dissolved saltcake waste batches (i.e., Batch 1A, Batch 2, and Batch 3) have been characterized and processed through the TCCR unit.

In the current operation, Tank 10H serves dual functions as both the dissolution tank and the feed tank for the TCCR system. The dissolved salt solution waste is pumped out of Tank 10H, through pre-filters and ion exchange columns. The decontaminated salt solution is then transferred to nearby Tank 11H (see Figure 1-1) and on to Tank 50H for final disposal in the Saltstone Production Facility.

The number of IX columns utilized can vary from single column to four columns in sequence. For the TCCR demonstration, 2 columns were mainly operated, however the last batch of feed processed had several IX column (IXC) evolutions including 3 and 4 IXC operations. In a typical TCCR operation, for example if two columns are on-line in a so-called lead-lag configuration, once the lead column is saturated with cesium indicated by the lag column effluent concentration reaching the target decontamination threshold (usually at a decontamination factor of 1000), the lead column may be removed from service, the lag column can be rotated into the lead position, and a new column can be placed into the lag position. The TCCR process for cesium removal from Tank 10H is detailed in X-SOW-H-00002 (Caldwell, 2017).

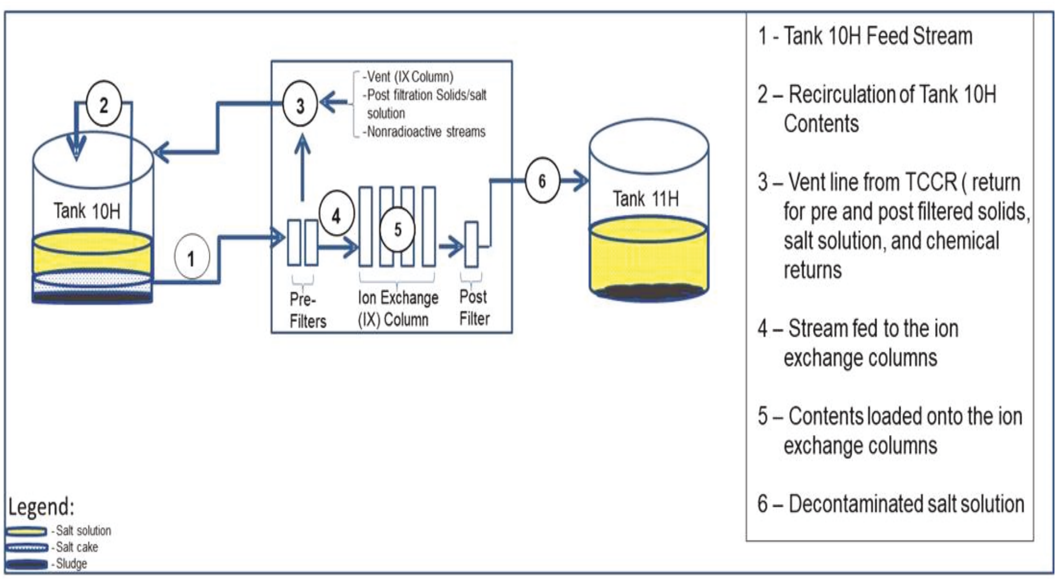

Figure 1-1. Cesium Removal from Tank 10H

To model IX column performance, the dynamic simulation VERSE-LC (VErsatile Reaction SEparation Simulation for Liquid Phase Adsorption and Chromatography Processes) code was chosen based on its

[1] IONSIV is a registered trademark of Honeywell UOP, Des Plaines, IL, U.S.A.

availability and widespread (and accepted) use in this field. VERSE-LC, developed by Professor Linda Wang of Purdue University in the 1990s, which models ion exchange columns including ion transport into porous media particles, was written in FORTRAN 90 and available on various platforms (Berninger et al., 1991). Savannah River National Laboratory (SRNL) procured an executable file running on the PC/Windows platform. SRNL has used VERSE-LC previously to predict IX column performance of cesium loading on CST for both SRS and Hanford tank waste applications (Hamm et al., 2001; Smith 2011).

1.2 Task Objective

Per request of Savannah River Remediation (SRR) (Fellinger, 2020), the task objective is to utilize on-line gamma monitoring data collected during TCCR Column Batch 3 operations to calibrate the parameters used in the VERSE-LC models. Calibrated VERSE-LC models could be applied to aid in evaluation of future TCCR operations. Column model calculations provide SRR with information on column breakthrough, CST bed utilization, and crucial parameters (e.g., flow rate, temperature, media particle size etc.) that affect the TCCR process to assist in the development of waste processing strategies.

1.3 Technical Reviews and Quality Assurance

Requirements for performing reviews of technical reports and the extent of review are established in manual E7 2.60.

The reviewers of this report include W. D. King and F. G. Smith, III. King provides an overall review. Smith design-checks the VERSE-LC predictions.

2.0 Model Formulations

2.1 Modeling Approach

- The OLI Studio™ software (Version 10) from OLI Systems, Inc. (OLI Systems, 2019) was used to calculate charge balanced feed compositions and to estimate feed solution density, viscosity and diffusivity required as input data to VERSE-LC.
- CST column performance in the TCCR process was modeled by VERSE-LC (Version 7.8). The VERSE-LC Fortran 90 code was developed by a research group of chemical engineers at Purdue University (Berninger et al., 1991). The code has been utilized in many IX projects at SRS and Hanford (Hamm et al., 2001; Smith 2011).

2.2 IX Column Model by VERSE-LC

The mathematical model utilized in the IX column simulations is a porous particle model that accounts for competitive adsorption (i.e., $Cs^+ >> K^+ > Na^+$), bulk advection, axial dispersion, film mass transfer, and pore diffusion. The numerical solutions of the governing equations and boundary conditions are performed by the VERSE-LC simulation package. The pore diffusion assumes uniform spherical adsorbent particles, plug flow with constant linear velocity, local equilibrium with the adsorbent, and constant diffusivities.

Early column performance (the first 5 to 10 bed volumes) may require the use of a multi-component model. Long-term performance, however, should be adequately handled using the single-component formulation as discussed below (Hamm et al., 2001).

In this model the kinetics associated with local ion exchange at an active site are assumed to be very fast (faster than the various liquid mass transfer mechanisms that transport ions to that site). Assuming radial effects to be negligible within the active region of the packed bed (i.e., a large column-to-particle diameter ratio), a one-dimensional solute transport equation for the mobile phase becomes

$$\frac{\partial C}{\partial t} = E_b \frac{\partial^2 C}{\partial z^2} - u_o \frac{\partial C}{\partial z} - \frac{3(1-\varepsilon_b)k_f}{R_p \varepsilon_b}(C - C_{p,r=R_p})$$

storage | **axial dispersion** | **advection** | **liquid film diffusion (mass transfer)**

With boundary and initial conditions

z = 0: $E_b \frac{\partial C}{\partial z} = u_o (C(t,0) - C_o)$

z = L: $\frac{\partial C}{\partial z} = 0$

t = 0: $C = C(0,z)$

C: Concentration in bed fluid, mol/L
E_b: Axial dispersivity, cm^2/min
u_o: Linear interstitial velocity, cm/min
R_p: Average particle radius, μm
ε_b: Bed porosity
k_f: Liquid film mass transfer coefficient, cm/min
L: Axial length of active bed of column, cm

Assuming uniformly sized spherical particles with a homogeneous distribution of pores, a one-dimensional species transport equation for the pore phase (within an average sized particle of media) becomes

$$\varepsilon_p \frac{\partial C_p}{\partial t} \quad + \quad (1-\varepsilon_p)\left(\frac{\partial Q}{\partial C_p}\right)\frac{\partial C_p}{\partial t} \quad = \quad \varepsilon_p \frac{D_p}{r^2}\frac{\partial}{\partial r}(r^2 \frac{\partial C_p}{\partial r})$$

storage **surface adsorption** **Fickian pore diffusion**

Subjected to boundary and initial conditions

r = 0: $\frac{\partial C_p}{\partial r} = 0$

r = R_p: $\varepsilon_p D_p \frac{\partial C_p}{\partial r} = k_f (C - C_{p,r=R_p})$

t = 0: $C_p = C_p(0,r)$

C_p: Concentration in pore fluid, mol/L
Q: Solid-phase solute concentration, mol/L_{Bed}
ε_p: Particle porosity
D_p: Pore diffusion coefficient, cm^2/min

Note that the second term on the left-hand side of the pore-phase transport equation includes the relationship of species solid-phase concentration and liquid-phase concentration. As a result, an isotherm equation is required.

The pore diffusion coefficient, D_p, relates to the molecular liquid-phase diffusion coefficient, D_∞, by a particle tortuosity factor, τ, accounting for bends along the pore paths as follows (Hamm et al., 2001)

$$D_p \cong \frac{1}{\tau} D_\infty$$

2.3 Software and Quality Assurance

The software and program code used in this report comply with requirements by Manual 1Q, Procedure 20-1 (Software Quality Assurance).

The OLI Studio™ is an acquired software that meets the commercial grade definition criteria in accordance with Manual E7 Procedure 3.46 and is accepted from the vendor by verifying the parts identifiers are correct. Dedication of the commercial grade software in accordance with Manual E7, Procedure 5.07 is not required for the OLI software, which was classified as Level D (Choi, 2019). All the activities related to the verification and validation of the OLI software database and the resulting models were documented in accordance with Manual E7 Procedure 5.40, Software Testing, Acceptance and Turnover.

Prior to applying VERSE-LC to the ion exchange modeling a verification process was completed and the results of that effort were reported in Hamm et al. (2000). The verification process ensures that the installed Windows version of VERSE-LC (i.e., version 7.80) was capable of adequately solving the above-mentioned governing equations and provided guidelines on how to accurately use the VERSE-LC code (e.g., mesh refinement requirements and input/output options). For all column simulations, numerical errors associated with the results of VERSE-LC should be very small when compared to the uncertainties associated with various model input parameters (bed density, particle size, pore diffusion, etc.). VERSE-LC was classified as Level D (Hang 2017).

Note that all software (OLI, VERSE-LC) are classified as Level D. Therefore, they cannot be used for safety-related calculations. The customer specified that the applicable Quality Assurance classification for the modeling efforts is Production Support; Level D software is compliant with these requirements.

3.0 Waste Compositions and Properties

3.1 Tank 10H Batch 1A, Batch 2 and Batch 3 Operations

As discussed under Section 1.1, Tank 10H serves dual functions as both the salt dissolution tank as well as the feed tank for the TCCR system. In each batch of the TCCR operation, Tank 10H undergoes dissolution campaigns, dissolving the salt cake to form an aqueous salt solution (i.e., supernate). Once the salt cake is dissolved, the contents were mixed by recirculation to achieve the target Na^+ concentration. Additionally, in each batch, in-tank CST batch contact equilibrium (or "teabag") samples were collected and analyzed. The teabag samples refer to a test where a small amount of CST was placed in a special container that remained submerged in the unagitated tank liquid over a number of days (usually a 10-day period) with the goal of loading cesium on the CST to saturation. The cesium-loaded CST was retrieved from the tank and analyzed by SRNL. In addition, traditional CST batch contact tests were conducted in SRNL under controlled conditions using Tank 10H supernate samples. Note that engineered CST from the same IONSIV™ R9120-B batch used in the TCCR columns was employed in both teabag and SRNL batch contact tests after preconditioning. The compositions of the three supernate batches are listed in Table 3-1. OLI calculations were performed at 34 °C for Batch 1A and Batch 2 and at 35 °C for Batch 3. For Batches 1A and Batch 2, chloride was adjusted for charge balance. For Batch 3, all anions were adjusted to balance the charge. A detailed description of anion adjustment of Batch 3 compositions is provided elsewhere (Taylor-Pashow et al, 2020).

Table 3-1. Supernate Compositions

Component	Batch 1A (1)	Batch 2 (2)	Batch 3 (3)
Na^{+1}	3.79	3.61	3.54
K^{+1}	2.21x10^{-3}	9.65x10^{-4}	1.74x10^{-3}
Cs^{+1}	1.131x10^{-5}	1.18x10^{-5}	1.16x10^{-5}
$SrOH^{+1}$	1.6263x10^{-7}	9.4665x10^{-8}	n/a
Sr^{+2}	4.6971x10^{-8}	1.1534x10^{-7}	1.12x10^{-7}
Ca^{+2}	7.14x10^{-5}	2.84x10^{-5}	n/a
Fe^{+3}	4.99x10^{-5}	0	n/a
OH^{-1}	1.82	0.666	0.10001
NO_3^{-1}	0.727	1.19	1.82451
NO_2^{-1}	0.0755	0.0694	0.4737
$Al(OH)_4^{-1}$	0.0422	0.039	0.0303
CO_3^{-2}	0.322	0.43	0.3333
SO_4^{-2}	0.174	0.282	0.2149
Cl^{-1}	0.12727 (4)	0.20068 (4)	0
F^{-1}	0	0	n/a
PO_4^{-3}	0	0	6.6312x10^{-4}
$C_2O_4^{-2}$	0.00427	0.0108	7.3154x10^{-3}
CrO_4^{2-}	n/a	n/a	1.0877x10^{-4}
OLI Diffusivity (cm^2/min)	7.9343x10^{-4}	9.387x10^{-4}	1.163x10^{-3}
OLI Viscosity (cP)	1.6294	1.5032	1.238
OLI Density (g/cm^3)	1.1616	1.1674	1.17

(1): Taylor-Pashow et al, 2019a
(2): Taylor-Pashow et al, 2019b
(3): Taylor-Pashow et al, 2020
(4): Adjusted for charge balance

3.2 Isotherms

The isotherm for each batch was obtained by use of the Langmuir model to fit the data generated by the ZAM computer model (Zheng et al, 1997). The isotherm parameters are listed in Table 3-2. The type of isotherm and its corresponding parameters are input to the VERSE-LC model (See 0).

Langmuir isotherm:

$$Q = \frac{\eta C_T \rho_{Bed} C_p}{\beta + C_p}$$

Q: Cesium loading on CST (mol_{Cs}/L_{Bed})
C_T: Bed cesium capacity (mol_{Cs}/L_{Bed})
C_p: Liquid-phase cesium concentration (mol_{Cs}/L)
η: Correction/dilution factor
ρ_{Bed}: Bed density (g_{CST}/mL_{Bed})
β: Isotherm parameter (mol_{Cs}/L)

Table 3-2. Isotherm Parameters

Feed	T (°C)	η	C_T ($mmol_{Cs}/g_{CST}$)	ρ_{Bed} (g_{CST}/mL_{Bed})	β
Batch 1A [(1)]	34	0.251	0.58	0.987	1.2358E-4
Batch 2 [(2)]	34	0.2457	0.58	0.987	1.343E-4
Batch 3 [(3)]	35	0.305	0.58	0.987	1.7485E-4

[(1)]: Taylor-Pashow et al, 2019a
[(2)]: Taylor-Pashow et al, 2019b
[(3)]: Taylor-Pashow et al, 2020

A correction factor (CF), referred to in the past as "dilution factor" (DF), is used in the IX modeling to offset the difference in performance between the powdered form of CST (which is what the ZAM model is based on) and the engineered media that dilutes the powdered CST with a binder.

The correction/dilution factors were determined by adjusting the ZAM isotherms to match the teabag test data as shown in Figure 3-1, **Error! Reference source not found.**, and **Error! Reference source not found.**. In all three TCCR batches, the correction factor values appear to be much lower than traditionally used binder dilution factors (i.e., 0.68) probably due to competing ions or precipitates rather than an increased amount of binder.

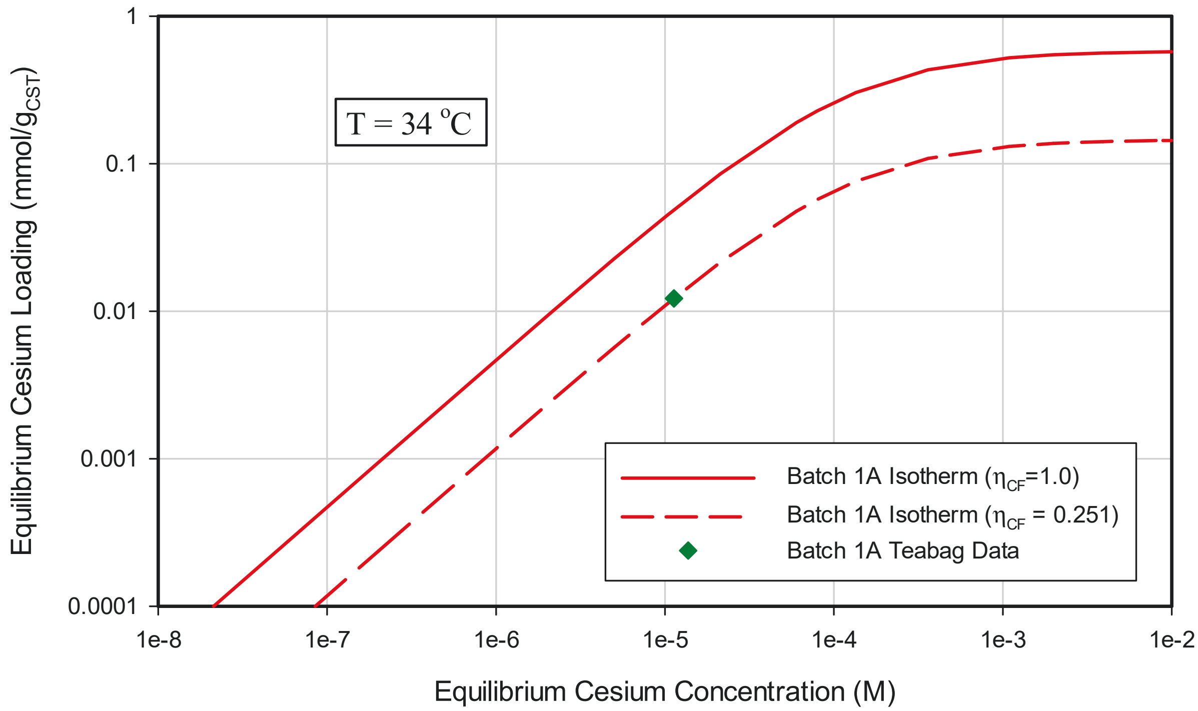

Figure 3-1. Batch 1A Isotherm versus Teabag Result at 34 °C

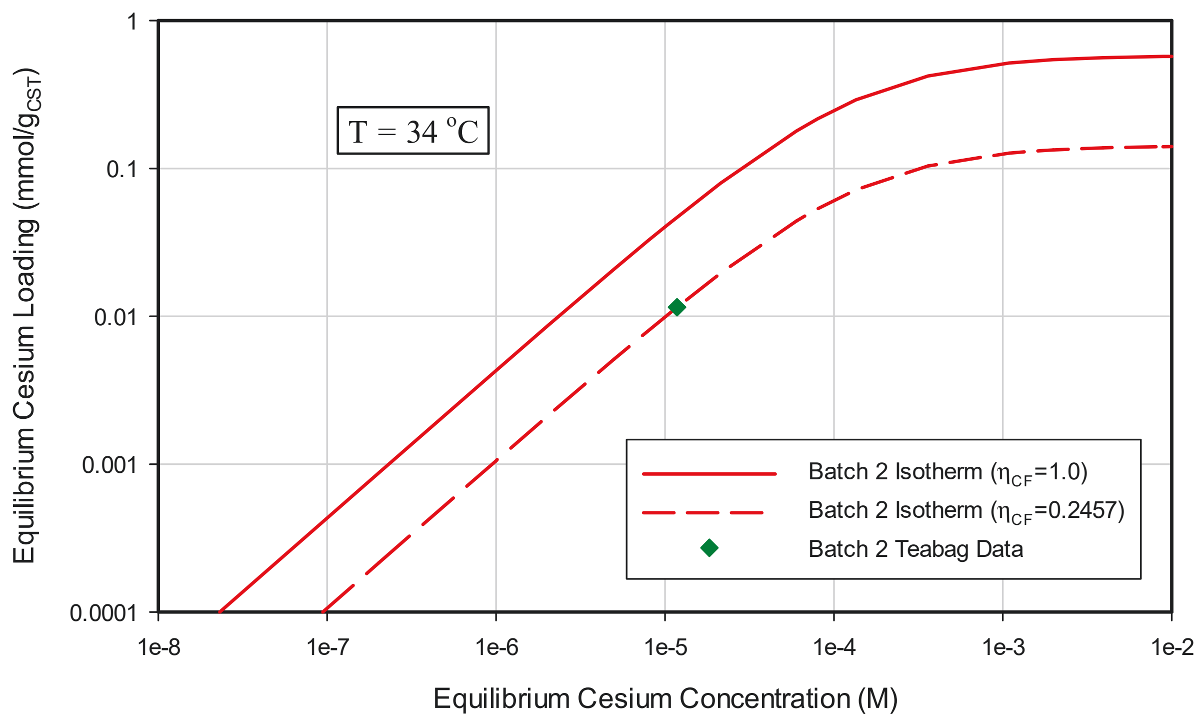

Figure 3-2. Batch 2 Isotherm versus Teabag Result at 34 °C

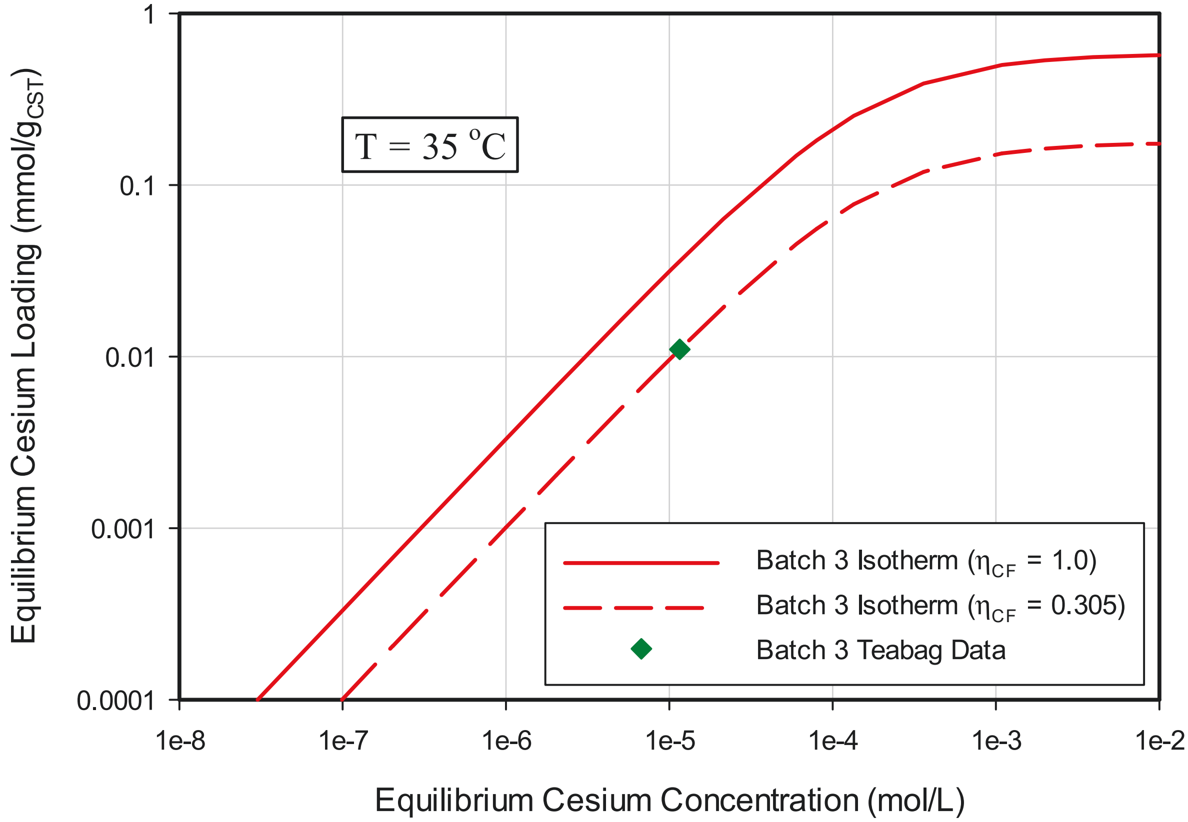

Figure 3-3. Batch 3 Isotherm versus Teabag Result at 35 °C

4.0 CST Columns

Table 4-1 summarizes the CST nominal bed properties and TCCR column design parameters used in the VERSE-LC column simulations.

Table 4-1. CST Bed Properties and TCCR Column Design

Properties	Values
CST Form (R9120-B)	Na^+
Bed Density (g_{CST}/mL_{Bed})	1.2097 (1)
F-Factor	0.8155 (2)
Dry Bed Density (g_{CST}/mL_{Bed})	0.987 (3)
Bed Porosity	0.548 (4)
Particle Porosity	0.24 (4)
Particle Diameter (μm)	572 (4)
Particle Tortuosity	4 (4)
Bed Diameter (cm)	48.7
Bed Length per column (cm)	263.6
Bed Volume (L)	491

(1): Based on pretreated R9120-B (King et al, 2019)
(2): Average dry mass correction factor for CST samples at 410 °C (King et al, 2020)
(3): (Bed density) * (F-Factor)
(4): SRNL-STI-2019-00147, Revision 0

5.0 TCCR Column Operations

The TCCR modular enclosure is deployed in the H Tank Farm to remove cesium from Tank 10H. Thus far, the TCCR system has processed three dissolved saltcake batches, i.e., Batch 1A, Batch 2, and Batch 3. A description of the conditions utilized in these treatment operations are provided chronologically below:

1. Batch 1A: Fresh (i.e., unused) columns A and B only
2. Batch 2: Partially cesium-loaded column B and fresh column C
3. Batch 3: Partially cesium-loaded column only
4. Batch 3: Partially cesium-loaded columns A and B
5. Batch 3: Fresh column D only

Processing of Batch 1A and Batch 2 was discussed in detail elsewhere (Hang, 2020). The processing strategy for Batch 3 (Silker, 2021) relevant to this calibration study is summarized below in Table 5-1.

Table 5-1. Batch 3 Processing Strategy

Column Evolution	Flow Rate (gpm)	Volume Processed (gal)	Start Date	Stop Date
A	5	2,981	7/10/20 11:33	7/30/2020 19:25
AB	5	22,222	8/1/2020 15:07	8/4/2020 10:04
	4	7,317	8/4/2020 10:04	8/5/2020 07:55
	8	5,691	8/5/2020 07:55	8/5/2020 21:18
D	5	4,065	8/17/2020 08:45	8/17/2020 19:27
	4	10,027	8/17/2020 19:27	8/20/2020 04:30

Note: For Batch 3, more IXC evolutions were performed, however these specific evolutions were used in modeling

During Batch 3 processing, Cs-137 activity at the outlet of the last column was detected by a gamma monitor device (Operations 3-5 above). Total cesium concentration can be estimated from the ratio of Cs-137/total Cs (i.e., 0.159) measured in the Batch 3 sample (Taylor-Pashow and Nash, 2020). No gamma monitoring data are available for Batch 1A and Batch 2. Note that the measured Cs-137 concentrations were corrected for full flow by applying a volume correction factor based on the recorded flow rate (Silker, 2021).

6.0 Calibration Methodology

The objective of this work is to utilize on-line gamma monitoring data collected during the TCCR column operations to calibrate the parameters (essentially, the correction/dilution factor, and possibly the pore tortuosity factor) used in the VERSE-LC models. Calibrated VERSE-LC models could be applied to aid in evaluating and planning future TCCR operations. Based on the TCCR columns utilization in the three batches for Tank 10H as outlined in Section 5.0 the following options are applied for calibration:

a) VERSE-LC Batch 3 / Column D: Compare column D computed cesium effluent concentration with measured Batch 3/Column D effluent concentration to determine CF/DF for Batch 3. This simple option should provide the best comparison between prediction and measurements because Column D is a fresh column.
b) In addition to Option a), another approach can be pursued:
    i. VERSE-LC Batch 1A / Columns A and B predictions could be utilized to provide effluent concentrations and determine CST loadings for columns A and B.
    ii. VERSE-LC Batch 2 / Columns B and C predictions could be utilized to provide effluent concentrations and determine CST loadings for columns B and C.
    iii. VERSE-LC Batch 3 / Column A: Compare column predicted A effluent concentration from Batch 1A with measured Batch 3/Column A effluent concentration to determine CF/DF for Batch 1A run and Batch 3.
    iv. VERSE-LC Batch 3 / Columns A and B: Compare column B effluent concentration from Batch 3/Columns A&B model run with measured Batch 3/Columns A&B effluent concentration. Since the CF factor for Batch 3 is selected based on the tea bag test, this comparison/adjustment Step allows the CF determination for Batch 2 run.

Note that Option b) involves Columns A and B for Batch 3. Columns A and B were also used to process Batch 1A and Batch 2 and were partially loaded with cesium at the end of these operations. Therefore, VERSE-LC models must be set up to run Batch 1A and Batch 2 (Steps i. and ii.) to establish the concentration profiles in both columns A and B prior to performing Steps iii. and iv.

7.0 Results and Discussion

The following sections provide the calibration results and some discussion of the options proposed in Section 6.0 above.

7.1 Batch 3/Column D Comparison (Calibration Option a)

In Batch 3, the fresh Column D was operated at two different flow rates (4 gpm and 5 gpm) as shown in Figure 7-1.

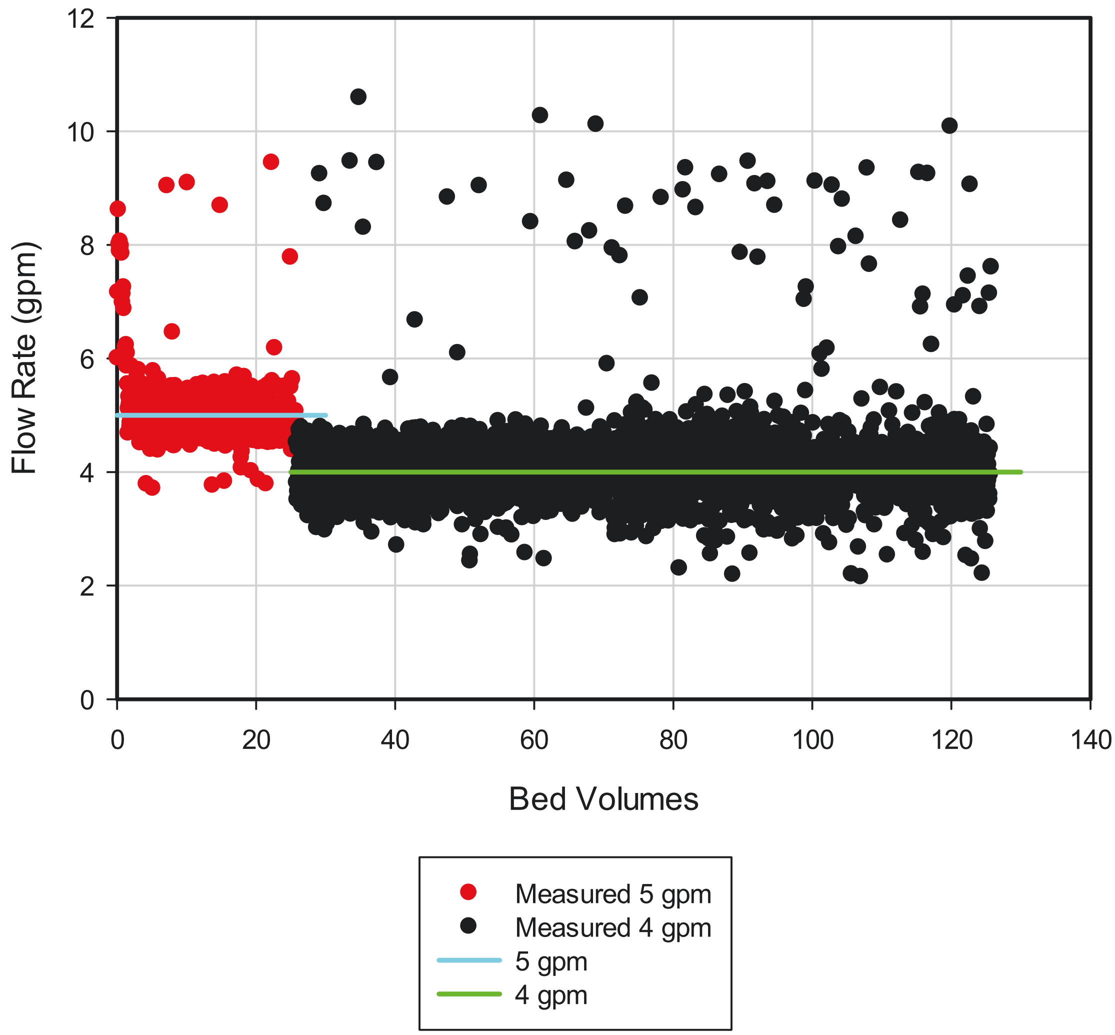

Figure 7-1. Batch 3/Column D - Measured Flow Rates

The cesium concentrations (total and Cs-137) in the effluent and the total Cs in the feed are displayed in Figure 7-2 for comparison. Note that Cs-137 effluent concentration was obtained from the on-line gamma monitoring data. The total Cs effluent concentration was computed from Cs-137 using the ratio of Cs-137/total Cs.

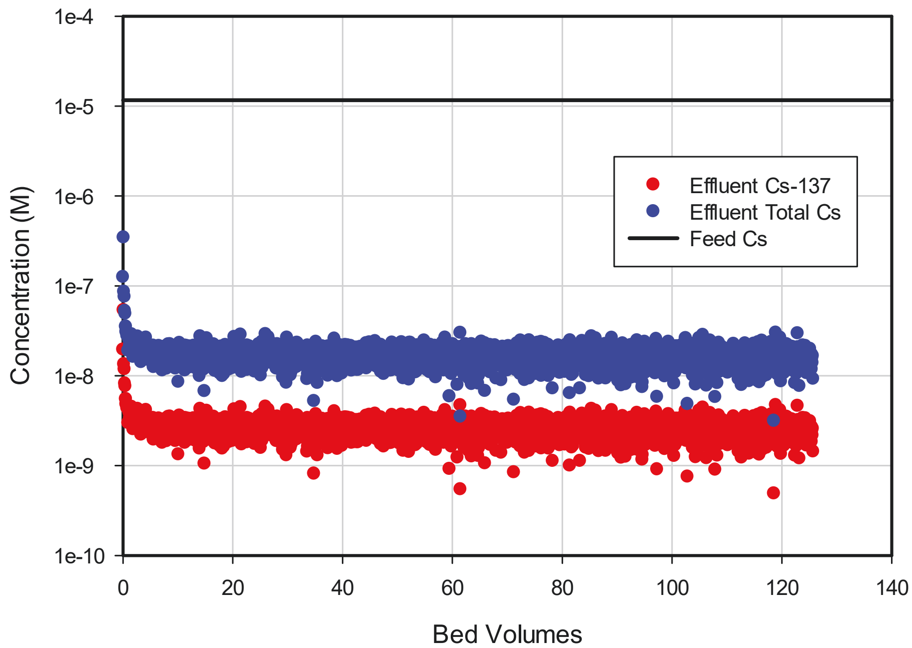

Figure 7-2. Batch 3/Column D - Measured Cesium Concentrations

In Figure 7-3, the cesium column effluent concentrations are shown for VERSE-LC results at CF factor of 0.305 and at different tortuosity τ values (i.e., 4, and 10) in comparison with the measured concentrations. The measurements show that the cesium concentrations remain flat for the entire operation. A gradual increase in effluent concentrations over time would be expected for early cesium breakthrough. The results indicate that the cesium breakthrough may be below the detectable limit of the gamma monitoring method. Based on the data, it is not possible to determine which tortuosity factor is correct, since no indication of cesium breakthrough is observed in the data.

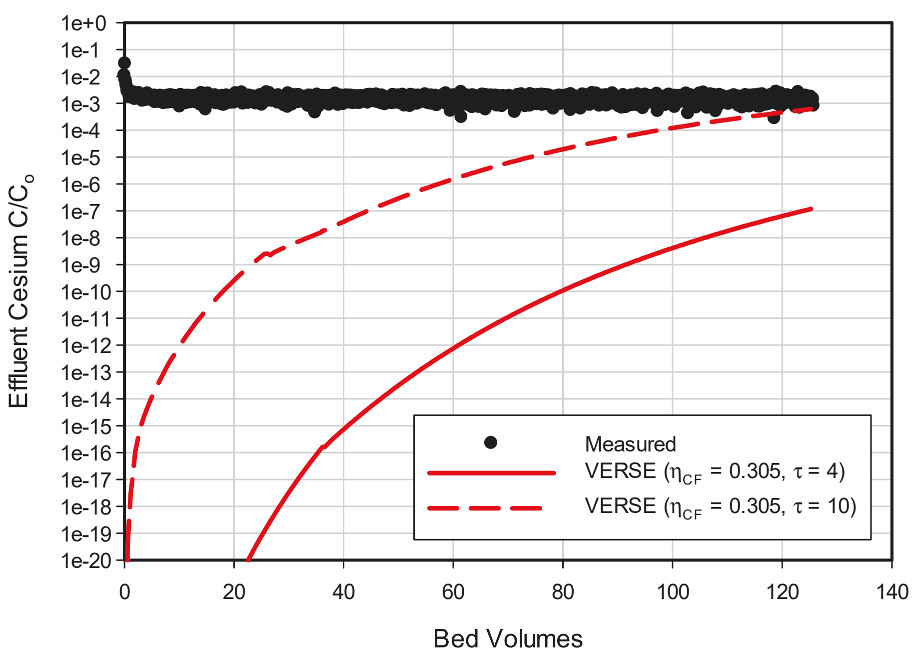

Figure 7-3. Batch 3/Column D - VERSE-LC Model Calibration

7.2 Comparison of Data from Batch 1A, Batch 2, Batch 3 / Columns AB&C (Calibration Option b)

Following Calibration Option b), step by step, CF results based on the on-line gamma data were obtained as shown below:

- Batch 1A: Selected CF based on effluent data analysis = 0.35 (Tea bag test: CF = 0.251)
- Batch 2: Selected CF based on effluent data analysis = 0.2457 (Tea bag test: CF = 0.2457)
- Batch 3: Selected CF based on effluent data analysis = 0.305 (Tea bag test: CF = 0.305)

Based on these derived correction factors from the processing of Batches 1A and 2 and using the measured correction factor for Batch 3, the following were generated for processing Batch 3. In Figure 7-4, the calculated and measured cesium breakthrough profiles for processing through the partially loaded Column A only (Step iii. in Section 6.0 above) are provided. A gradual increase in the cesium effluent concentration was observed for this processing step. The pore tortuosity of 10 seems to produce VERSE-LC data in better agreement with the monitoring data. Overall, the model predictions and the rad data are in reasonable alignment for Batch3/Column A.

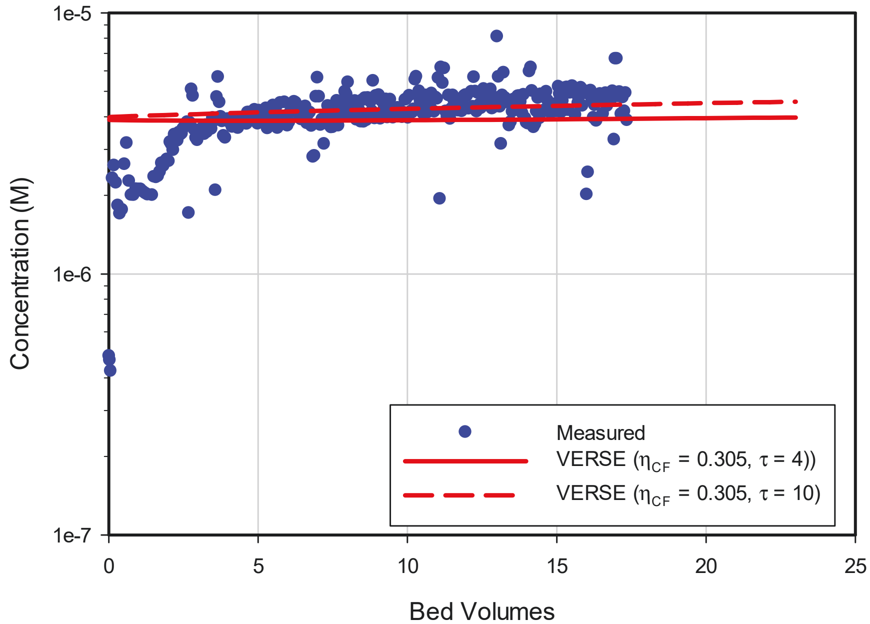

Figure 7-4. Batch 3/Column A - Effluent Concentrations

In Figure 7-5, the calculated and measured cesium breakthrough profiles for processing through partially loaded Columns A and B (Step iv in Section 6.0) are provided using the correction factors identified for each batch above. As shown in Figure 7-5, the data and predictions do not match well (greater than order-of-magnitude difference) and the measured values are well below the VERSE-LC predictions. The pore tortuosity of 10 produces worse VERSE-LC prediction in comparison with the monitoring data. A steady increase in the cesium effluent concentration was observed during processing through the two columns.

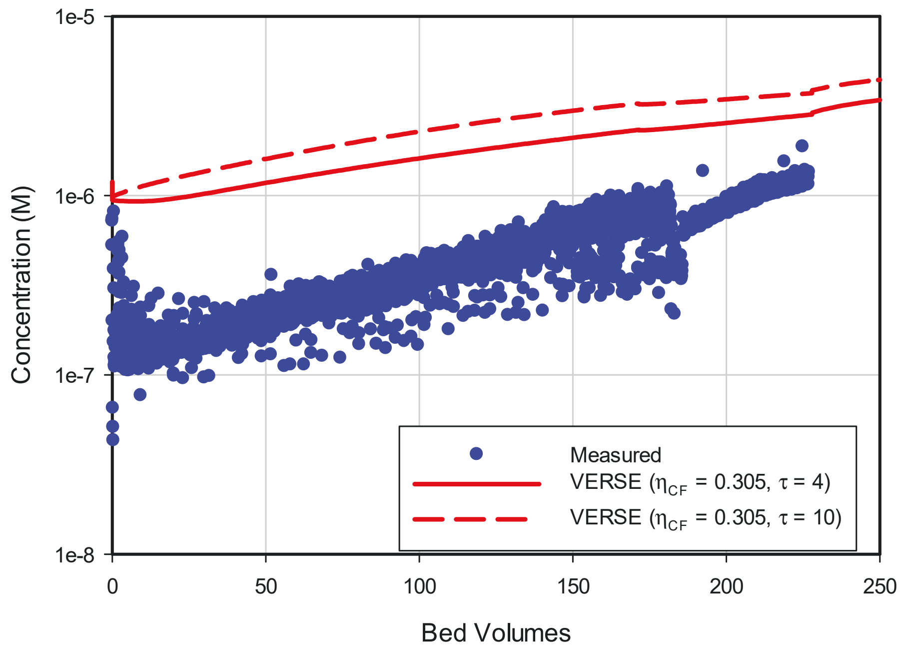

Figure 7-5. Batch 3/Columns A&B (in Sequence) - Effluent Concentrations

There appears to be an issue with gamma monitoring data for Batch 3/Columns A&B. For most of the operation, the measured effluent concentrations remain below 1E-6 M (Figure 7-5). But the measured effluent concentrations for Batch 3/Column A which involved the same feed display the values of ~4.5E-6 M (Figure 7-4) at the end of the operation, meaning the concentration profile in Column A would be above 1E-6 M as shown in Figure 7-6 and Figure 7-7. The same applies to Column B. Hence, the measured data for Batch 3/Columns A&B seem low and inconsistent.

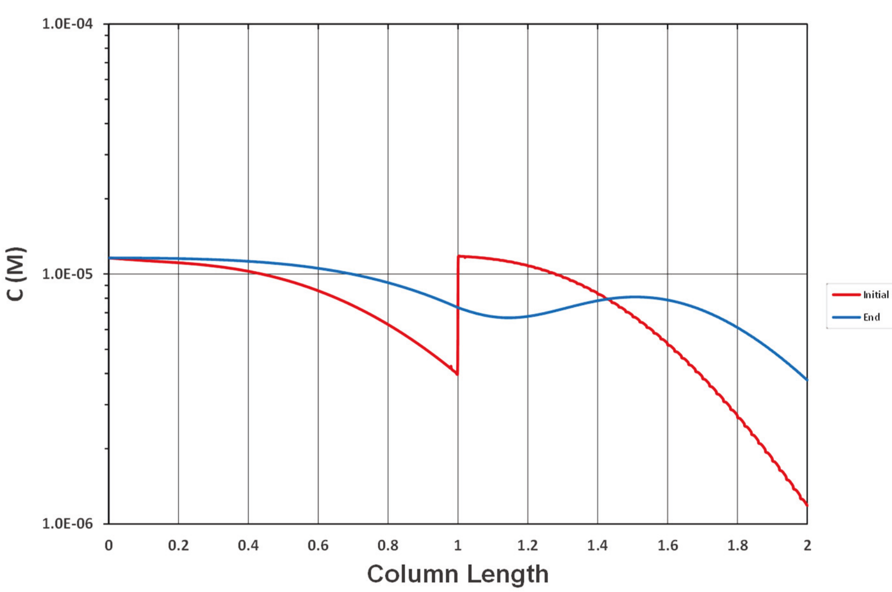

Figure 7-6. Batch 3/Columns A&B - Concentration Profiles calculated by VERSE-LC (τ = 4)

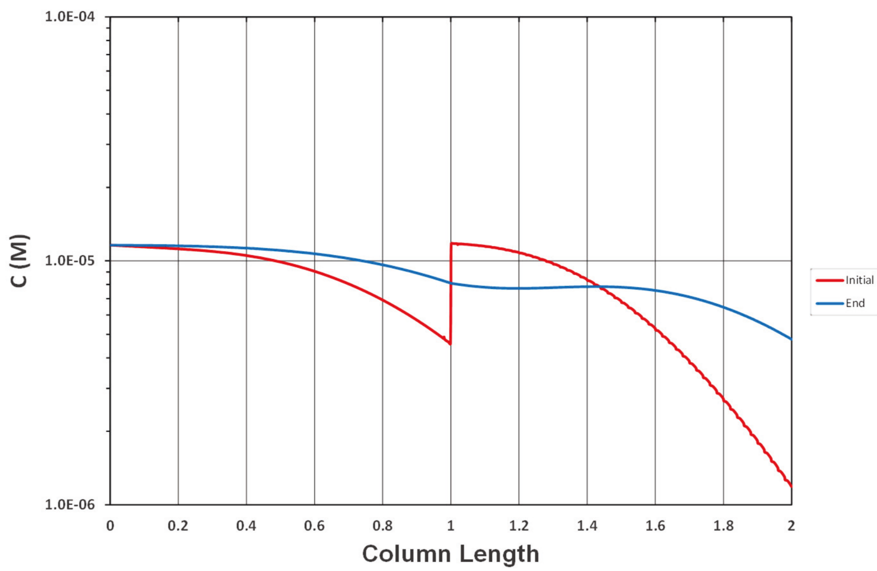

Figure 7-7. Batch 3/Columns A&B - Concentration Profiles calculated by VERSE-LC (τ = 10)

7.3 Bed Utilization

The bed utilization of columns used in Batch 1A, Batch 2 and Batch 3 is estimated and summarized in Table 7-1. Table 7-1 also displays the dilution/correction factor CF and the pore tortuosity for each batch operation. The bed utilization is provided for individual columns together with an overall utilization for the combining columns if multiple columns are utilized.

Note that the CST bed utilization varies between processing batches, depending on the waste feed compositions that result in varying isotherms. In general, it can be seen in Table 7-1 that the capacity of each column utilized increases with the increasing total volume processed when processing batches 1A, 2, and 3.

Table 7-1. Estimated Bed Utilization

Batches	Configuration	CF	Tortuosity	Column Utilization (%)				
				A	B	C	D	Overall
Batch 1A	A & B	0.35	4	77.8	9.5			43.7
Batch 2	B & C	0.2457	4		58.2	2.2		30.2
Batch 3	A	0.305	4	76.1				76.1
			10	79.2				79.2
	A & B	0.305	4	88.5	57.9			73.2
			10	90	61.1			75.6
	D	0.305	4				19	19
			10				26	26

8.0 Conclusions

On-line gamma monitoring data for Batch 3 during the TCCR operations were used to attempt to calibrate the correction/dilution factor to be used in the VERSE-LC models. The calibration findings are summarized as follows:

1. Overall, the correction/dilution factors determined from the teabag tests provide a good match to gamma monitoring data. Based on the results, a correction/dilution factor near 0.3 appears to be required to model the cesium removal performance.
2. Calibration of Batch 3/Column D indicates that based on the measured data it is not possible to determine which pore tortuosity is better.
3. Measured data for Batch 3/Columns A&B apparently seem low and inconsistent with gamma monitoring data for Batch 3/Column A.
4. Improvement in gamma monitoring device is essential to future VERSE-LC model calibrations.
5. Another option to gamma monitoring data would be to use actual sample data for the receipt tank (i.e., Tank 11H) to track the volume and Curie content over time. The VERSE-LC model can be calibrated to generate the same volume and Curie content.

9.0 References

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Appendix A. VERSE-LC Model Calibration Work Scope

Original Scope

From: Terri Fellinger
Sent: Friday, September 11, 2020 12:54 PM
To: Thong Hang
Cc: Boyd Wiedenman; Frank Pennebaker; Patricia Lee; Aubrey Silker
Subject: RE: Batch 3 VERSE Modeling

Thong,

Sorry for the late response. Here is the run information for Batch 3. I highlighted Column D. Here is what I would like you to do.

- You have the ZAM model run for Batch 3 that you completed for Kathryn. Please use the dilution factor that was observed from the Batch Equilibrium contact test sample taken from Tank 10.
- I would like to use Column D for this comparison because the IX column was not used during Batch 1A or 2. o Use the TCCR demonstration IX column design (20" column) o The column is filled with 515kgs of CST o The IX column D has been held full with 3M NaOH solution.
    - The flows and times are provided below (highlighted in yellow). Please model both.
    - The temperature of salt solution coming from Tank 10 was 35C during this time.
    - Would like to know what the column profile looks like and how much of the CST has been utilized based on your modeling.

Aubrey is working on processing the rad monitor data for the entire run. Once she has both inlet and outlet rad monitor data, we can compare what we observed to the modeling efforts.

Thanks,
Terri

Evolution	Feed Volume (gallons)	Gallons Available	Gallons Processed	Start Date	Stop Date
Initial Batch 3 Volume		91,327			
Run Column A (5 gpm)					
Run 5 gpm	2,981	88,346	2,981	7/30/20 11:33	7/30/20 19:25
Shutdown when cesium breakthrough is reached on the column					
Run Column A, B (5, 4, and 8 gpm)					
Run 5 gpm	22,222	66,124	25,203	8/1/20 15:07	8/4/20 10:04

Run 4 gpm	7,317	58,807	32,520	8/4/20 10:04	8/5/20 7:55
Run 8 gpm	5,691	53,116	38,211	8/5/20 7:55	8/5/20 21:18
Shutdown					
Sample Tank 11				8/6	
Run Column A, B, C (8 and 5 gpm)					
Run 8 gpm	8,130	44,986	46,341	8/6/20 11:20	8/7/20 9:13
Run 5 gpm	9,485	35,501	55,826	8/7/20 9:13	8/9/20 20:35
Shutdown					
Sample Tank 11				8/13	

1

Outage: Tank 9 Work					
Run Column D (5 and 4 gpm)					
Run 5 gpm	4,065	31,436	59,891	8/17/20 8:45	8/17/20 19:27
Run 4 gpm	10,027	21,409	69,918	8/17/20 19:27	8/20/20 4:30
Shutdown					
Sample Tank 11				8/21	
Run Column A, B, C, D (5 and 4 gpm)					
Run 5 gpm	2,168	19,241	72,086	8/24/20 14:46	8/24/20 16:23
Run 4 gpm	17,344	1,897	89,430	8/24/20 16:23	8/28/20 16:03
Batch 3 complete			89,430		
Shutdown					
Sample Tank 11				9/1	

From: Thong Hang <thong.hang@srnl.doe.gov>
Sent: Thursday, September 3, 2020 2:30 PM
To: Terri Fellinger <terri.fellinger@srs.gov>
Cc: Boyd Wiedenman <Boyd.Wiedenman@srnl.doe.gov>; Frank Pennebaker <frank.pennebaker@srnl.doe.gov>; Patricia Lee <patricia.lee@srnl.doe.gov>
Subject: Batch 3 VERSE Modeling

Terri,

In the TCCR Monthly meeting last week, you said that there may be some scenarios for Batch 3 operation to be evaluated in VERSE. Would you please outline those scenarios? Also, when do we expect to have Batch 3 operation data available for model benchmark?

I should have time in September and probably early October to work on TCCR.

Thanks,
Thong

Thong Hang, PhD
Advanced Modeling, Simulation and Analytics
Savannah River National Laboratory
Email: thong.hang@srnl.doe.gov
Work: 803-725-8204

Additional Scope

From: Aubrey Silker
Sent: Tuesday, February 02, 2021 3:04 PM
To: Thong Hang
Cc: Terri Fellinger
Subject: Re: TCCR Batch 3 Column D data
Attachments: Data_for Thong.xlsx

Hi Thong!

Attached is the requested data from the A and AB Run. The setup for the sheet is the same as before. Below is the Batch 3 processing strategy. For the A run, we ran at 5 gpm the entire time. The total volume processed was 2,981 gallons. For the AB Run, we ran at 5, 4, and 8 gpm (in that order) and the total volume processed was 35,230 gallons.

Table 3. Batch 3 Processing Strategy.

Column Evolution	Flow rate (gpm)	Volume Processed (gal)	Start Date
A	5	2,981	7/30 11:33
AB	5	22,222	8/1 15:07
	4	7,317	8/4 10:04
	8	5,691	8/5 07:55
ABC	8	8,130	8/6 11:20
	5	9,485	8/7 09:13
D	5	4,065	8/17 08:45
	4	10,027	8/17 19:27

Best,

Aubrey Silker
766-H 2040
O: (803) 208-0235
C: (407) 221-7389

From: Aubrey Silker
Sent: Monday, December 14, 2020 11:12 AM
To: Thong Hang <thong.hang@srnl.doe.gov>
Cc: Terri Fellinger <terri.fellinger@srs.gov>
Subject: TCCR Batch 3 Column D data

Good morning Thong,

Attached is the Batch 3 column D run DSS data. I have transferred this data from the original spreadsheet into a new one, since it was a large file. Some things to note--

Column A includes the date and time during processing. The column D run occurred from 8/17 08:45 - 8/20 04:30. The Cs-137 activity was measured about every 60 seconds. From 8/17 10:18 - 10:35 no data was recorded from the new radiation monitors.

Column B is the flow rate (in gallons per minute, gpm) during processing. At startup (for approximately 25 minutes), the process was run at 8 gpm to remove air from the system. Then the flow rate was reduced to 5 gpm. On 8/17 19:25, the flow rate was reduced again to 4 gpm for the rest of the run. The flow rate can vary significantly from the setpoint but is usually within +/- 0.25 gpm. Any flow rate below 2 gpm was removed from the data set and replaced with "outlier." This flow rate drop occurred during pre-filter flushes.

Column C is the gallons processed based on the flow rate data and the change in time. There is good agreement between the flow rate data and the measured Tank 10 level. Per the tank farm morning report, 14,092 gallons was processed during the column D run. Based on the flow rate data, 14,149 gallons was processed during the column D run.

Column D is the Cs-137 Ci added to Tank 11 during processing. I calculated this by applying the trapezoidal rule for finding the area under the curve and using the data from Column C and Column E. Per the new radiation monitors, we added ~1.42 Ci Cs-137 to Tank 11 during the column D run.

Column E is the measured Cs-137 activity (in Ci/gal) for the DSS line. This data has been scrubbed and corrected. First I removed any "zero" readings from the data (any values below 3E-06 Ci/gal). Then I removed any readings that were taken when the flow rate was less than 2 gpm/during pre-filter flushes. Last, I applied a volume correction factor based on the recorded flow rate in Column B. For the column D run, the average Cs-137 activity in the DSS line was 1.32E-04 Ci/gal at 5 gpm and 1.13E-04 Ci/gal at 4 gpm.

Column F is the DF based on the DSS line readings in Column E. The Batch 3 qualification sample data was used for the inlet Cs-137 concentration (0.081 Ci/gal). The average DF was 686 at 5 gpm and 750 at 4 gpm.

Please let me know if you have any questions! Thank you for your patience!

Best,

Aubrey Silker
766-H 2040
O: (803) 208-0235
C: (407) 221-7389

Appendix B. VERSE-LC Input and Output Files

1) Batch 3/Column D (Option a)

- CF = 0.305 & τ = 4

VERSE-LC input:

```
TCCR Simulation of Cs removal on CST material single column
Case 1 - TCCR Batch 3, 5 gpm & 4 gpm, 35 C, D Column
1, 50, 4, 6                          ncomp, nelem, ncol-bed, ncol-part
FCWNA                                isotherm,axial-disp,film-coef,surf-diff,BC-col
NNNNN                             input-only,perfusable,feed-equil,use datafile.yio,generate/update datafile.yio
M                                    comp-conc units
228.74, 48.68, 18927, 6d+4           Total bed length(cm),Diam(cm),Q-flow(ml/min),CSTR-vol(ml)
286.0, 0.50, 0.24,  0.0              part-rad(um), bed-void, part-void, sorb-cap()
0.0                                  initial concentrations (M)
S                                    COMMAND - inlet conc step change
1, 0.0, 1.16d-5, 1, 0.0              spec id, time(min), conc(M), freq, dt(min)
V                                    COMMAND - viscosity/density change
0.01238, 1.17                        fluid viscosity(poise), density(g/cm^3)
s                                    COMMAND - step change flow
813, 15142, 1, 0.0                   te, Fnew, Ne, tr
h                                    COMMAND - effluent history dump
2, 1.0, 1.0, 0.50, 0.5               unit op#, ptscale(1-4) filtering
D                                    COMMAND - dump column profile at 813 minutes
-1, 813, 1, 0                        particle point (-1 for all), te, Ne, tr
D                                    COMMAND - dump column profile at 3320 minutes
-1, 3320, 1, 0                       particle point (-1 for all), te, Ne, tr
-                                    end of commands
 3320, 1.0                           end time(min), max step size (B.V.)
1.0d-7, 1.0d-4                       abs-tol, rel-tol
-                                    non-negative conc constraint
1.0d0                                size exclusion factor
2.908d-4                             part-pore diffusivities(cm^2/min) 25% of free value
1.163d-3                             Brownian diffusivities(cm^2/min)
0.1746                               Freundlich/Langmuir Hybrid a    (moles/L B.V.) rhob=0.987 g/ml
1.0                                  Freundlich/Langmuir Hybrid b    (1/M)          Batch specific isotherm
1.0                                  Freundlich/Langmuir Hybrid Ma   (-)            a = 0.305 x 0.58 x rhob
1.0                                  Freundlich/Langmuir Hybrid Mb   (-)
1.7485d-4                            Freundlich/Langmuir Hybrid beta (-)
```

VERSE-LC output:

```
===============================================================================
 VERSE v7.80  by R. D. Whitley and N.-H. L. Wang, c1999 PRF
 ===============================================================================
 Input file: Case1
 TCCR Simulation of Cs removal on CST material single column
 Case 1 - TCCR Batch 3, 5 gpm & 4 gpm, 35 C, D Column
 Begin Run:  16:28:41 on 08-30-2021   running under Windows 95/8
 Finite elements    - axial: 50  particle: 1
 Collocation points - axial:  4  particle: 6 => Number of eqns:  2010
 Inlet species at equilib.? N   Perfusable sorbent? N   Feed profile only? N
 Use Profile File? N   Generate Profile File? N
 Axial dispersion correlation:   Chung & Wen (1968)
 Film mass transfer correlation: Wilson & Geankoplis (1966)
 ===============================================================================
 SYSTEM PARAMETERS (at initial conditions):

 t(stop)        =   3320.00000 min         dtheta max     =      1.00000 BV
 abs. tol.      =       .10000E-06         rel. tol.      =       .10000E-03
 Total Length   =    228.74000 cm          D              =     48.68000 cm
 Tot. Capacity  =       .00000 eq/L solid  Col. Vol.      = 425728.90452 mL
 F              =  18927.00000 mL/min      Uo (linear)    =     20.33859 cm/min
 R              =    286.00000 microns     L/R            =   7997.90210
 Bed Void frac. =       .50000             Pcl. Porosity  =       .24000
 Spec. Area     =     52.44755 1/cm        Time/BV        =     11.24660 min
 Vol CSTRs      =  60000.00000 mL

 Component no.  =       1
 Ke  [-]        =  .10000E+01
 Eb  [cm2/min]  =  .27627E+01
```

```
Dp  [cm2/min]  =  .29080E-03
Doo [cm2/min]  =  .11630E-02
kf  [cm/min]   =  .35184E+00
Ds  [cm2/min]  =  .00000E+00

Dimensionless Groups:
Re             =  .91622E+00
Sc(i)          =  .54589E+03
Peb(i)         =  .16839E+04
Bi(i)          =  .14418E+03
Nf(i)          =  .41507E+03
Np(i)          =  .95961E+00
Pep(i)         =  .83345E+04

Isotherm       =  Freundlich/Langmuir Hybrid
Iso. Const. 1  =  .17460E+00
Iso. Const. 2  =  .10000E+01
Iso. Const. 3  =  .10000E+01
Iso. Const. 4  =  .10000E+01
Iso. Const. 5  =  .17485E-03
Init. Conc.    =  .00000E+00
Conc. at eqb.  =  .00000E+00
Conc. units          M
===============================================================================
COMMAND LIST:
  1: Step conc. of component 1 at .0000     min to .1160E-04 M
     Execute   1 times, every .0000     mins.
  2: User set viscosity to .1238E-01 poise and density to 1.170     g/cm3
  3: Step change flow at 813.0     min to  .151E+05 mL/min
     Execute   1 times, every .0000     mins.
  4: Monitor conc. history at stream  2.  Filename = Case1.h01
     Output density adjustments:
     1.0    *default abs conc delta,      1.0    *default rel conc delta,
     .50    *default force w/ conc delta, .50    *default force w/o conc delta
  5: Dump full profile file at  813.0     min
     Execute   1 times, every .0000     mins.
  6: Dump full profile file at  3320.     min
     Execute   1 times, every .0000     mins.
===============================================================================
VERSE-LC finished in   411 steps.  Average step size 8.078     minutes
End run:  16:28:53 on 08-30-2021
Integrated Areas in History Files:
Case1.h01                  .305586E-09
```

- CF = 0.305 & τ = 10

VERSE-LC input:

```
TCCR Simulation of Cs removal on CST material single column
Case 1 - TCCR Batch 3, 5 gpm & 4 gpm, 35 C, D Column
1, 50, 4, 6                          ncomp, nelem, ncol-bed, ncol-part
FCWNA                                isotherm,axial-disp,film-coef,surf-diff,BC-col
NNNNN                             input-only,perfusable,feed-equil,use datafile.yio,generate/update datafile.yio
M                                    comp-conc units
228.74, 48.68, 18927, 6d+4           Total bed length(cm),Diam(cm),Q-flow(ml/min),CSTR-vol(ml)
286.0, 0.548, 0.24,  0.0              part-rad(um), bed-void, part-void, sorb-cap()
0.0                                  initial concentrations (M)
S                                    COMMAND - inlet conc step change
1, 0.0, 1.16d-5, 1, 0.0              spec id, time(min), conc(M), freq, dt(min)
V                                    COMMAND - viscosity/density change
0.01238, 1.17                        fluid viscosity(poise), density(g/cm^3)
s                                     COMMAND - step change flow
813, 15142, 1, 0.0                    te, Fnew, Ne, tr
h                                    COMMAND - effluent history dump
2, 1.0, 1.0, 0.50, 0.5               unit op#, ptscale(1-4) filtering
D                                   COMMAND - dump column profile at 813 minutes
-1,  813, 1, 0                      particle point (-1 for all), te, Ne, tr
D                                   COMMAND - dump column profile at 3320 minutes
-1, 3320, 1, 0                      particle point (-1 for all), te, Ne, tr
-                                    end of commands
 3320, 1.0                           end time(min), max step size (B.V.)
1.0d-7, 1.0d-4                       abs-tol, rel-tol
-                                    non-negative conc constraint
1.0d0                                size exclusion factor
1.163d-4                             part-pore diffusivities(cm^2/min) 10% of free value
1.163d-3                             Brownian diffusivities(cm^2/min)
0.1746                               Freundlich/Langmuir Hybrid a    (moles/L B.V.) rhob=0.987 g/ml
1.0                                  Freundlich/Langmuir Hybrid b    (1/M)          Batch specific isotherm
1.0                                  Freundlich/Langmuir Hybrid Ma   (-)            a = 0.305 x 0.58 x rhob
1.0                                  Freundlich/Langmuir Hybrid Mb   (-)
1.7485d-4                            Freundlich/Langmuir Hybrid beta (-)
```

VERSE-LC output:

```
==============================================================================
 VERSE v7.80  by R. D. Whitley and N.-H. L. Wang, c1999 PRF
 ==============================================================================
 Input file: Case1
 TCCR Simulation of Cs removal on CST material single column
 Case 1 - TCCR Batch 3, 5 gpm & 4 gpm, 35 C, D Column
 Begin Run:  15:54:18 on 08-30-2021   running under Windows 95/8
 Finite elements    - axial: 50  particle: 1
 Collocation points - axial:  4  particle: 6 => Number of eqns:  2010
 Inlet species at equilib.? N   Perfusable sorbent? N   Feed profile only? N
 Use Profile File? N   Generate Profile File? N
 Axial dispersion correlation:   Chung & Wen (1968)
 Film mass transfer correlation: Wilson & Geankoplis (1966)
 ==============================================================================
 SYSTEM PARAMETERS (at initial conditions):

 t(stop)        =   3320.00000 min         dtheta max     =      1.00000 BV
 abs. tol.      =       .10000E-06         rel. tol.      =       .10000E-03
 Total Length   =    228.74000 cm          D              =     48.68000 cm
 Tot. Capacity  =       .00000 eq/L solid  Col. Vol.      = 425728.90452 mL
 F              =  18927.00000 mL/min      Uo (linear)    =     18.55710 cm/min
 R              =    286.00000 microns     L/R            =   7997.90210
 Bed Void frac. =       .54800             Pcl. Porosity  =       .24000
 Spec. Area     =     47.41259 1/cm        Time/BV        =     12.32628 min
 Vol CSTRs      =  60000.00000 mL

 Component no.  =      1
 Ke  [-]        =  .10000E+01
 Eb  [cm2/min]  =  .27627E+01
 Dp  [cm2/min]  =  .11630E-03
 Doo [cm2/min]  =  .11630E-02
 kf  [cm/min]   =  .32102E+00
 Ds  [cm2/min]  =  .00000E+00

 Dimensionless Groups:
 Re             =  .91622E+00
 Sc(i)          =  .54589E+03
```

```
Peb(i)          =  .15364E+04
Bi(i)           =  .32893E+03
Nf(i)           =  .34236E+03
Np(i)           =  .42062E+00
Pep(i)          =  .19015E+05

Isotherm        =  Freundlich/Langmuir Hybrid
Iso. Const. 1   =  .17460E+00
Iso. Const. 2   =  .10000E+01
Iso. Const. 3   =  .10000E+01
Iso. Const. 4   =  .10000E+01
Iso. Const. 5   =  .17485E-03
Init. Conc.     =  .00000E+00
Conc. at eqb.   =  .00000E+00
Conc. units           M
===============================================================================
COMMAND LIST:
  1: Step conc. of component 1 at .0000     min to .1160E-04 M
     Execute   1 times, every .0000     mins.
  2: User set viscosity to .1238E-01 poise and density to 1.170     g/cm3
  3: Step change flow at 813.0     min to  .151E+05 mL/min
     Execute   1 times, every .0000     mins.
  4: Monitor conc. history at stream  2.  Filename = Case1.h01
     Output density adjustments:
     1.0    *default abs conc delta,      1.0    *default rel conc delta,
     .50    *default force w/ conc delta, .50    *default force w/o conc delta
  5: Dump full profile file at  813.0     min
     Execute   1 times, every .0000     mins.
  6: Dump full profile file at  3320.     min
     Execute   1 times, every .0000     mins.
===============================================================================
VERSE-LC finished in   392 steps.  Average step size 8.469     minutes
End run:  15:54:30 on 08-30-2021
Integrated Areas in History Files:
Case1.h01                  .390313E-05
```

2) Batch 1A/Columns A&B (Option b, Step i.)

- CF = 0.35 & τ = 4

VERSE-LC input:

```
TCCR Simulation of Cs removal on CST material lead-lag columns A/B
Case 1 - Tank 10H Batch 1A, 5gpm, 34C
1, 100, 4, 6                         ncomp, nelem, ncol-bed, ncol-part
FCWNF                                isotherm,axial-disp,film-coef,surf-diff,BC-col
NNNNY                             input-only,perfusable,feed-equil,use datafile.yio,generate/update datafile.yio
M                                    comp-conc units
527.18, 48.68, 18927, 0.0d+0         Total bed length(cm),Diam(cm),Q-flow(ml/min),CSTR-vol(ml)
286, 0.548, 0.24,  0.0               part-rad(um), bed-void, part-void, sorb-cap()
0.0                                  initial concentrations (M)
S                                    COMMAND - inlet conc step change
1, 0.0, 1.13051d-5, 1, 0.0           spec id, time(min), conc(M), freq, dt(min)
V                                    COMMAND - viscosity/density change
0.0163, 1.1616                       fluid viscosity(poise), density(g/cm^3)
m                                    COMMAND - subcolumns (carousel-concentration driven)
50, 100, 0, 1, 1.13051d-5, 0.0, 1d+6   Nelem shift, Nelem watch, Npp watch, Nc watch, Cthresh, te, tee
h                                    COMMAND - effluent history dump
2, 1.0, 1.0, 0.50, 0.5               unit op#, ptscale(1-4) filtering
h                                    COMMAND - effluent history dump
3, 1.0, 1.0, 0.50, 0.5               unit op#, ptscale(1-4) filtering
D                                    COMMAND - dump column profile (First Lag Col Breakthrough)
-1, 30406, 1, 0                      particle point (-1 for all), time(min), freq, dt(min)
-                                    end of commands
30406.2, 1                           end time(min), max step size (B.V.)
1.0d-7, 1.0d-4                       abs-tol, rel-tol
-                                    non-negative conc constraint
1.0d0                                size exclusion factor
1.9836d-4                            part-pore diffusivities(cm^2/min) 25% of free value
7.9343d-4                            Brownian diffusivities(cm^2/min) (calc. by OLI)
0.2                                  Freundlich/Langmuir Hybrid a    (moles/L B.V.) rhob=0.987 g/ml
1.0                                  Freundlich/Langmuir Hybrid b    (1/M)          Batch specific isotherm
1.0                                  Freundlich/Langmuir Hybrid Ma   (-)            a = 035 x 0.58 x rhob
1.0                                  Freundlich/Langmuir Hybrid Mb   (-)
1.2358d-4                            Freundlich/Langmuir Hybrid beta (-)
```

VERS-LC output:

```
==============================================================================
 VERSE v7.80  by R. D. Whitley and N.-H. L. Wang, c1999 PRF
 ==============================================================================
 Input file: Case1
 TCCR Simulation of Cs removal on CST material lead-lag columns A/B
 Case 1 - Tank 10H Batch 1A, 5gpm, 34C
 Begin Run:  14:55:38 on 09-01-2021   running under Windows 95/8
 Finite elements    - axial:100  particle: 1
 Collocation points - axial:  4  particle: 6 => Number of eqns:  4010
 Inlet species at equilib.? N   Perfusable sorbent? N   Feed profile only? N
 Use Profile File? N   Generate Profile File? Y
 Axial dispersion correlation:   Chung & Wen (1968)
 Film mass transfer correlation: Wilson & Geankoplis (1966)
 Sub-Column Boundary Conditions: Flux Continuity
 ==============================================================================
 SYSTEM PARAMETERS (at initial conditions):

 t(stop)        =  30406.20000 min         dtheta max     =      1.00000 BV
 abs. tol.      =       .10000E-06         rel. tol.      =       .10000E-03
 Total Length   =    527.18000 cm          D              =     48.68000 cm
 Tot. Capacity  =       .00000 eq/L solid  Col. Vol.      = 981182.84466 mL
 F              =  18927.00000 mL/min      Uo (linear)    =     18.55710 cm/min
 R              =    286.00000 microns     L/R            =  18432.86713
 Bed Void frac. =       .54800             Pcl. Porosity  =       .24000
 Spec. Area     =     47.41259 1/cm        Time/BV        =     14.20426 min
 Vol CSTRs      =       .00000 mL

 Component no.  =      1
 Ke  [-]        =  .10000E+01
 Eb  [cm2/min]  =  .27804E+01
 Dp  [cm2/min]  =  .19836E-03
 Doo [cm2/min]  =  .79343E-03
 kf  [cm/min]   =  .24878E+00
```

```
Ds  [cm2/min]  =  .00000E+00

Dimensionless Groups:
Re             =  .69088E+00
Sc(i)          =  .10611E+04
Peb(i)         =  .17593E+04
Bi(i)          =  .14946E+03
Nf(i)          =  .30574E+03
Np(i)          =  .82671E+00
Pep(i)         =  .11148E+05

Isotherm       =  Freundlich/Langmuir Hybrid
Iso. Const. 1  =  .20000E+00
Iso. Const. 2  =  .10000E+01
Iso. Const. 3  =  .10000E+01
Iso. Const. 4  =  .10000E+01
Iso. Const. 5  =  .12358E-03
Init. Conc.    =  .00000E+00
Conc. at eqb.  =  .00000E+00
Conc. units          M
==============================================================================
COMMAND LIST:
  1: Step conc. of component 1 at .0000     min to .1131E-04 M
     Execute   1 times, every .0000     mins.
  2: User set viscosity to .1630E-01 poise and density to 1.162     g/cm3
  3: Carousel (conc.). Active between t = .0000     and .1000E+07 min.
     When comp.  1 reaches .1131E-04 M     at end of node 100,
     shift  50 axial elements out the feed end
  4: Monitor conc. history at stream  2.  Filename = Case1.h01
     Output density adjustments:
     1.0    *default abs conc delta,      1.0    *default rel conc delta,
     .50    *default force w/ conc delta, .50    *default force w/o conc delta
  5: Monitor conc. history at stream  3.  Filename = Case1.h02
     Output density adjustments:
     1.0    *default abs conc delta,      1.0    *default rel conc delta,
     .50    *default force w/ conc delta, .50    *default force w/o conc delta
  6: Dump full profile file at  .3041E+05 min
     Execute   1 times, every .0000     mins.
==============================================================================
VERSE-LC finished in  2675 steps.  Average step size 11.37     minutes
End run:  14:57:45 on 09-01-2021
Integrated Areas in History Files:
Case1.h01                  .304786E-01
Case1.h02                  .122197E-03
```

3) Batch 2/Columns B&C (Option b, Step ii.)

- CF = 0.2457 & τ = 4

VERSE-LC input:

```
TCCR Simulation of Cs removal on CST material lead-lag columns B/C
Case 1 - Tank 10H Batch 2, 34C, large particle size
1, 100, 4, 6                          ncomp, nelem, ncol-bed, ncol-part
FCWNF                                 isotherm,axial-disp,film-coef,surf-diff,BC-col
NNNYY                              input-only,perfusable,feed-equil,use datafile.yio,generate/update datafile.yio
M                                     comp-conc units
527.18, 48.68, 18927, 0.0d+0          Total bed length(cm),Diam(cm),Q-flow(ml/min),CSTR-vol(ml)
286.0, 0.548, 0.24,  0.0              part-rad(um), bed-void, part-void, sorb-cap()
0.0                                   initial concentrations (M)
S                                     COMMAND - inlet conc step change
1, 0.0, 1.18d-5, 1, 0.0               spec id, time(min), conc(M), freq, dt(min)
V                                     COMMAND - viscosity/density change
0.015032, 1.16741                     fluid viscosity(poise), density(g/cm^3)
s                                     COMMAND - inlet flow rate change
5400, 30283, 1, 0.0                   time(min), Q-flow(ml/min), freq, dt(min)
s                                     COMMAND - inlet flow rate change
5580, 37854, 1, 0.0                   time(min), Q-flow(ml/min), freq, dt(min)
s                                     COMMAND - inlet flow rate change
5700, 18927, 1, 0.0                   time(min), Q-flow(ml/min), freq, dt(min)
s                                     COMMAND - inlet flow rate change
8130, 30283, 1, 0.0                   time(min), Q-flow(ml/min), freq, dt(min)
s                                     COMMAND - inlet flow rate change
8550, 18927, 1, 0.0                   time(min), Q-flow(ml/min), freq, dt(min)
m                                     COMMAND - subcolumns (carousel-concentration driven)
50, 100, 0, 1, 1.18d-4, 0.0, 1d+6     Nelem shift, Nelem watch, Npp watch, Nc watch, Cthresh, te, tee
h                                     COMMAND - effluent history dump
2, 1.0, 1.0, 0.50, 0.5                unit op#, ptscale(1-4) filtering
h                                     COMMAND - effluent history dump
3, 1.0, 1.0, 0.50, 0.5                unit op#, ptscale(1-4) filtering
D                                       COMMAND - dump column profile
-1, 1d-4, 1, 0                          particle point (-1 for all), time(min), freq, dt(min)
D                                       COMMAND - dump column profile
-1, 11119, 1, 0                         particle point (-1 for all), time(min), freq, dt(min)
-                                     end of commands
11119, 1                              end time(min), max step size (B.V.)
1.0d-7, 1.0d-4                        abs-tol, rel-tol
-                                     non-negative conc constraint
1.0d0                                 size exclusion factor
2.3468d-4                             part-pore diffusivities(cm^2/min) 25% of free value
9.38712d-4                            Brownian diffusivities(cm^2/min) (calc. by OLI)
0.14065                               Freundlich/Langmuir Hybrid a    (moles/L B.V.) rhob=0.987 g/ml
1.0                                   Freundlich/Langmuir Hybrid b    (1/M)          Batch specific isotherm
1.0                                   Freundlich/Langmuir Hybrid Ma   (-)            a = 0.2457 x 0.58 x rhob
1.0                                   Freundlich/Langmuir Hybrid Mb   (-)
1.343d-04                             Freundlich/Langmuir Hybrid beta (-)
```

VERSE-LC output:

```
=============================================================================
 VERSE v7.80  by R. D. Whitley and N.-H. L. Wang, c1999 PRF
 =============================================================================
 Input file: Case1
 TCCR Simulation of Cs removal on CST material lead-lag columns B/C
 Case 1 - Tank 10H Batch 2, 34C, large particle size
 Begin Run:  15:53:20 on 09-01-2021   running under Windows 95/8
 Finite elements    - axial:100  particle: 1
 Collocation points - axial:  4  particle: 6 => Number of eqns:  4010
 Inlet species at equilib.? N   Perfusable sorbent? N   Feed profile only? N
 Use Profile File? Y   Generate Profile File? Y
 Axial dispersion correlation:   Chung & Wen (1968)
 Film mass transfer correlation: Wilson & Geankoplis (1966)
 Sub-Column Boundary Conditions: Flux Continuity
 =============================================================================
 SYSTEM PARAMETERS (at initial conditions):

 t(stop)        =  11119.00000 min         dtheta max     =      1.00000 BV
 abs. tol.      =       .10000E-06         rel. tol.      =       .10000E-03
 Total Length   =    527.18000 cm          D              =     48.68000 cm
 Tot. Capacity  =       .00000 eq/L solid  Col. Vol.      = 981182.84466 mL
```

```
F              =  18927.00000 mL/min      Uo (linear)    =     18.55710 cm/min
R              =    286.00000 microns     L/R            =  18432.86713
Bed Void frac. =       .54800             Pcl. Porosity  =       .24000
Spec. Area     =     47.41259 1/cm        Time/BV        =     14.20426 min
Vol CSTRs      =       .00000 mL

Component no.  =      1
Ke  [-]        =  .10000E+01
Eb  [cm2/min]  =  .27752E+01
Dp  [cm2/min]  =  .23468E-03
Doo [cm2/min]  =  .93871E-03
kf  [cm/min]   =  .27829E+00
Ds  [cm2/min]  =  .00000E+00

Dimensionless Groups:
Re             =  .75291E+00
Sc(i)          =  .82302E+03
Peb(i)         =  .17626E+04
Bi(i)          =  .14131E+03
Nf(i)          =  .34200E+03
Np(i)          =  .97808E+00
Pep(i)         =  .94230E+04

Isotherm       =  Freundlich/Langmuir Hybrid
Iso. Const. 1  =  .14065E+00
Iso. Const. 2  =  .10000E+01
Iso. Const. 3  =  .10000E+01
Iso. Const. 4  =  .10000E+01
Iso. Const. 5  =  .13430E-03
Init. Conc.    =  .00000E+00
Conc. at eqb.  =  .00000E+00
Conc. units          M
===============================================================================
COMMAND LIST:
  1: Step conc. of component 1 at .0000     min to .1180E-04 M
     Execute   1 times, every .0000     mins.
  2: User set viscosity to .1503E-01 poise and density to 1.167     g/cm3
  3: Step change flow at 5400.     min to  .303E+05 mL/min
     Execute   1 times, every .0000     mins.
  4: Step change flow at 5580.     min to  .379E+05 mL/min
     Execute   1 times, every .0000     mins.
  5: Step change flow at 5700.     min to  .189E+05 mL/min
     Execute   1 times, every .0000     mins.
  6: Step change flow at 8130.     min to  .303E+05 mL/min
     Execute   1 times, every .0000     mins.
  7: Step change flow at 8550.     min to  .189E+05 mL/min
     Execute   1 times, every .0000     mins.
  8: Carousel (conc.). Active between t = .0000     and .1000E+07 min.
     When comp.  1 reaches .1180E-03 M     at end of node 100,
     shift  50 axial elements out the feed end
  9: Monitor conc. history at stream  2.  Filename = Case1.h01
     Output density adjustments:
     1.0    *default abs conc delta,      1.0    *default rel conc delta,
     .50    *default force w/ conc delta, .50    *default force w/o conc delta
 10: Monitor conc. history at stream  3.  Filename = Case1.h02
     Output density adjustments:
     1.0    *default abs conc delta,      1.0    *default rel conc delta,
     .50    *default force w/ conc delta, .50    *default force w/o conc delta
 11: Dump full profile file at  .1000E-03 min
     Execute   1 times, every .0000     mins.
 12: Dump full profile file at  .1112E+05 min
     Execute   1 times, every .0000     mins.
===============================================================================
VERSE-LC finished in  1160 steps.  Average step size 9.585     minutes
End run:  15:53:46 on 09-01-2021
Integrated Areas in History Files:
Case1.h01                  .426492E-02
Case1.h02                  .767992E-05
```

4) Batch 3/Column A (Option b, Step iii.)

- CF = 0.305 & τ = 4

VERSE-LC input:

```
TCCR Simulation of Cs removal on CST material column A
Case 1 - TCCR Batch 3, 5gpm, 35C
1, 50, 4, 6                          ncomp, nelem, ncol-bed, ncol-part
FCWNF                                isotherm,axial-disp,film-coef,surf-diff,BC-col
NNNYY                             input-only,perfusable,feed-equil,use datafile.yio,generate/update datafile.yio
M                                    comp-conc units
263.59, 48.68, 18927, 0.0d+0         Total bed length(cm),Diam(cm),Q-flow(ml/min),CSTR-vol(ml)
286, 0.548, 0.24,  0.0               part-rad(um), bed-void, part-void, sorb-cap()
0.0                                  initial concentrations (M)
S                                    COMMAND - inlet conc step change
1, 0.0, 1.16d-5, 1, 0.0              spec id, time(min), conc(M), freq, dt(min)
V                                    COMMAND - viscosity/density change
0.01238, 1.17                        fluid viscosity(poise), density(g/cm^3)
h                                    COMMAND - effluent history dump
2, 1.0, 1.0, 0.50, 0.5               unit op#, ptscale(1-4) filtering
D                                      COMMAND - dump column profile (First Lag Col Breakthrough)
-1, 596.2, 1, 0                        particle point (-1 for all), time(min), freq, dt(min)
-                                    end of commands
596.2, 1                               end time(min), max step size (B.V.)
1.0d-7, 1.0d-4                       abs-tol, rel-tol
-                                    non-negative conc constraint
1.0d0                                size exclusion factor
2.908d-4                             part-pore diffusivities(cm^2/min) 25% of free value
1.163d-3                             Brownian diffusivities(cm^2/min) (calc. by OLI)
0.1746                               Freundlich/Langmuir Hybrid a    (moles/L B.V.) rhob=0.987 g/ml
1.0                                  Freundlich/Langmuir Hybrid b    (1/M)          Batch specific isotherm
1.0                                  Freundlich/Langmuir Hybrid Ma   (-)            a = 0.305 x 0.58 x rhob
1.0                                  Freundlich/Langmuir Hybrid Mb   (-)
1.7485d-4                            Freundlich/Langmuir Hybrid beta (-)
```

VERSE-LC output:

```
==============================================================================
 VERSE v7.80  by R. D. Whitley and N.-H. L. Wang, c1999 PRF
 ==============================================================================
 Input file: Case1
 TCCR Simulation of Cs removal on CST material column A
 Case 1 - TCCR Batch 3, 5gpm, 35C
 Begin Run:  08:47:58 on 09-02-2021   running under Windows 95/8
 Finite elements    - axial: 50  particle: 1
 Collocation points - axial:  4  particle: 6 => Number of eqns:  2010
 Inlet species at equilib.? N   Perfusable sorbent? N   Feed profile only? N
 Use Profile File? Y   Generate Profile File? Y
 Axial dispersion correlation:   Chung & Wen (1968)
 Film mass transfer correlation: Wilson & Geankoplis (1966)
 ==============================================================================
 SYSTEM PARAMETERS (at initial conditions):

 t(stop)        =    596.20000 min         dtheta max     =      1.00000 BV
 abs. tol.      =       .10000E-06         rel. tol.      =       .10000E-03
 Total Length   =    263.59000 cm          D              =     48.68000 cm
 Tot. Capacity  =       .00000 eq/L solid  Col. Vol.      = 490591.42233 mL
 F              =  18927.00000 mL/min      Uo (linear)    =     18.55710 cm/min
 R              =    286.00000 microns     L/R            =   9216.43357
 Bed Void frac. =       .54800             Pcl. Porosity  =       .24000
 Spec. Area     =     47.41259 1/cm        Time/BV        =     14.20426 min
 Vol CSTRs      =       .00000 mL

 Component no.  =      1
 Ke  [-]        =  .10000E+01
 Eb  [cm2/min]  =  .27627E+01
 Dp  [cm2/min]  =  .29080E-03
 Doo [cm2/min]  =  .11630E-02
 kf  [cm/min]   =  .32102E+00
 Ds  [cm2/min]  =  .00000E+00

 Dimensionless Groups:
 Re             =  .91622E+00
 Sc(i)          =  .54589E+03
```

```
Peb(i)         =  .17705E+04
Bi(i)          =  .13155E+03
Nf(i)          =  .39452E+03
Np(i)          =  .12120E+01
Pep(i)         =  .76045E+04

Isotherm       =  Freundlich/Langmuir Hybrid
Iso. Const. 1  =  .17460E+00
Iso. Const. 2  =  .10000E+01
Iso. Const. 3  =  .10000E+01
Iso. Const. 4  =  .10000E+01
Iso. Const. 5  =  .17485E-03
Init. Conc.    =  .00000E+00
Conc. at eqb.  =  .00000E+00
Conc. units           M
===============================================================================
COMMAND LIST:
  1: Step conc. of component 1 at .0000     min to .1160E-04 M
     Execute   1 times, every .0000     mins.
  2: User set viscosity to .1238E-01 poise and density to 1.170     g/cm3
  3: Monitor conc. history at stream  2.  Filename = Case1.h01
     Output density adjustments:
     1.0    *default abs conc delta,      1.0    *default rel conc delta,
     .50    *default force w/ conc delta, .50    *default force w/o conc delta
  4: Dump full profile file at  596.2     min
     Execute   1 times, every .0000     mins.
===============================================================================
VERSE-LC finished in    47 steps.  Average step size 12.69     minutes
End run:  08:47:59 on 09-02-2021
Integrated Areas in History Files:
Case1.h01                  .232480E-02
```

- CF = 0.305 & τ = 10

VERSE-LC input:

```
TCCR Simulation of Cs removal on CST material column A
Case 1 - TCCR Batch 3, 5gpm, 35C
1, 50, 4, 6                          ncomp, nelem, ncol-bed, ncol-part
FCWNF                                isotherm,axial-disp,film-coef,surf-diff,BC-col
NNNYY                             input-only,perfusable,feed-equil,use datafile.yio,generate/update datafile.yio
M                                    comp-conc units
263.59, 48.68, 18927, 0.0d+0         Total bed length(cm),Diam(cm),Q-flow(ml/min),CSTR-vol(ml)
286, 0.548, 0.24,  0.0               part-rad(um), bed-void, part-void, sorb-cap()
0.0                                  initial concentrations (M)
S                                    COMMAND - inlet conc step change
1, 0.0, 1.16d-5, 1, 0.0              spec id, time(min), conc(M), freq, dt(min)
V                                    COMMAND - viscosity/density change
0.01238, 1.17                        fluid viscosity(poise), density(g/cm^3)
h                                    COMMAND - effluent history dump
2, 1.0, 1.0, 0.50, 0.5               unit op#, ptscale(1-4) filtering
D                                    COMMAND - dump column profile (First Lag Col Breakthrough)
-1, 596.2, 1, 0                      particle point (-1 for all), time(min), freq, dt(min)
-                                    end of commands
596.2, 1                             end time(min), max step size (B.V.)
1.0d-7, 1.0d-4                       abs-tol, rel-tol
-                                    non-negative conc constraint
1.0d0                                size exclusion factor
1.163d-4                             part-pore diffusivities(cm^2/min) 10% of free value
1.163d-3                             Brownian diffusivities(cm^2/min) (calc. by OLI)
0.1746                               Freundlich/Langmuir Hybrid a    (moles/L B.V.) rhob=0.987 g/ml
1.0                                  Freundlich/Langmuir Hybrid b    (1/M)          Batch specific isotherm
1.0                                  Freundlich/Langmuir Hybrid Ma   (-)            a = 0.305 x 0.58 x rhob
1.0                                  Freundlich/Langmuir Hybrid Mb   (-)
1.7485d-4                            Freundlich/Langmuir Hybrid beta (-)
```

VERSE-LC output:

```
===============================================================================
 VERSE v7.80  by R. D. Whitley and N.-H. L. Wang, c1999 PRF
 ===============================================================================
 Input file: Case1
 TCCR Simulation of Cs removal on CST material column A
 Case 1 - TCCR Batch 3, 5gpm, 35C
 Begin Run:  15:25:48 on 09-01-2021   running under Windows 95/8
 Finite elements    - axial: 50  particle: 1
 Collocation points - axial:  4  particle: 6 => Number of eqns:  2010
 Inlet species at equilib.? N   Perfusable sorbent? N   Feed profile only? N
 Use Profile File? Y   Generate Profile File? Y
 Axial dispersion correlation:   Chung & Wen (1968)
 Film mass transfer correlation: Wilson & Geankoplis (1966)
 ===============================================================================
 SYSTEM PARAMETERS (at initial conditions):

 t(stop)        =    596.20000 min         dtheta max     =      1.00000 BV
 abs. tol.      =       .10000E-06         rel. tol.      =       .10000E-03
 Total Length   =    263.59000 cm          D              =     48.68000 cm
 Tot. Capacity  =       .00000 eq/L solid  Col. Vol.      = 490591.42233 mL
 F              =  18927.00000 mL/min      Uo (linear)    =     18.55710 cm/min
 R              =    286.00000 microns     L/R            =   9216.43357
 Bed Void frac. =       .54800             Pcl. Porosity  =       .24000
 Spec. Area     =     47.41259 1/cm        Time/BV        =     14.20426 min
 Vol CSTRs      =       .00000 mL

 Component no.  =      1
 Ke  [-]        =  .10000E+01
 Eb  [cm2/min]  =  .27627E+01
 Dp  [cm2/min]  =  .11630E-03
 Doo [cm2/min]  =  .11630E-02
 kf  [cm/min]   =  .32102E+00
 Ds  [cm2/min]  =  .00000E+00

 Dimensionless Groups:
 Re             =  .91622E+00
 Sc(i)          =  .54589E+03
 Peb(i)         =  .17705E+04
 Bi(i)          =  .32893E+03
 Nf(i)          =  .39452E+03
 Np(i)          =  .48471E+00
```

```
Pep(i)          =  .19015E+05

Isotherm        =  Freundlich/Langmuir Hybrid
Iso. Const. 1  =  .17460E+00
Iso. Const. 2  =  .10000E+01
Iso. Const. 3  =  .10000E+01
Iso. Const. 4  =  .10000E+01
Iso. Const. 5  =  .17485E-03
Init. Conc.     =  .00000E+00
Conc. at eqb.  =  .00000E+00
Conc. units           M
===============================================================================
COMMAND LIST:
  1: Step conc. of component 1 at .0000     min to .1160E-04 M
     Execute   1 times, every .0000     mins.
  2: User set viscosity to .1238E-01 poise and density to 1.170     g/cm3
  3: Monitor conc. history at stream  2.  Filename = Case1.h01
     Output density adjustments:
     1.0    *default abs conc delta,      1.0    *default rel conc delta,
     .50    *default force w/ conc delta, .50    *default force w/o conc delta
  4: Dump full profile file at  596.2     min
     Execute   1 times, every .0000     mins.
===============================================================================
VERSE-LC finished in    47 steps.  Average step size 12.69     minutes
End run:  15:25:49 on 09-01-2021
Integrated Areas in History Files:
Case1.h01                  .256943E-02
```

5) Batch 3/Columns A&B (Option b, Step iv.)

- CF = 0.305 & $\tau = 4$

VERSE-LC input:

```
TCCR Simulation of Cs removal on CST material lead-lag columns A/B
Case 1 - Tank 10H Batch 3, 34C, large particle size
1, 100, 4, 6                          ncomp, nelem, ncol-bed, ncol-part
FCWNF                                 isotherm,axial-disp,film-coef,surf-diff,BC-col
NNNYY                              input-only,perfusable,feed-equil,use datafile.yio,generate/update datafile.yio
M                                     comp-conc units
527.18, 48.68, 18927, 0.0d+0          Total bed length(cm),Diam(cm),Q-flow(ml/min),CSTR-vol(ml)
286.0, 0.548, 0.24,  0.0              part-rad(um), bed-void, part-void, sorb-cap()
0.0                                   initial concentrations (M)
S                                     COMMAND - inlet conc step change
1, 0.0, 1.16d-5, 1, 0.0               spec id, time(min), conc(M), freq, dt(min)
V                                     COMMAND - viscosity/density change
0.01238, 1.17                         fluid viscosity(poise), density(g/cm^3)
s                                     COMMAND - inlet flow rate change
4444.4, 15142, 1, 0.0                 time(min), Q-flow(ml/min), freq, dt(min)
s                                     COMMAND - inlet flow rate change
6274, 30283, 1, 0.0                   time(min), Q-flow(ml/min), freq, dt(min)
m                                     COMMAND - subcolumns (carousel-concentration driven)
50, 100, 0, 1, 1.16d-4, 0.0, 1d+6     Nelem shift, Nelem watch, Npp watch, Nc watch, Cthresh, te, tee
h                                     COMMAND - effluent history dump
2, 1.0, 1.0, 0.50, 0.5                unit op#, ptscale(1-4) filtering
h                                     COMMAND - effluent history dump
3, 1.0, 1.0, 0.50, 0.5                unit op#, ptscale(1-4) filtering
D                                     COMMAND - dump column profile
-1, 1.0d-4, 1, 0                      particle point (-1 for all), time(min), freq, dt(min)
D                                     COMMAND - dump column profile
-1, 6985, 1, 0                        particle point (-1 for all), time(min), freq, dt(min)
-                                     end of commands
6985, 1                               end time(min), max step size (B.V.)
1.0d-7, 1.0d-4                        abs-tol, rel-tol
-                                     non-negative conc constraint
1.0d0                                 size exclusion factor
2.908d-4                              part-pore diffusivities(cm^2/min) 25% of free value
1.163d-3                              Brownian diffusivities(cm^2/min)
0.1746                                Freundlich/Langmuir Hybrid a    (moles/L B.V.) rhob=0.987 g/ml
1.0                                   Freundlich/Langmuir Hybrid b    (1/M)          Batch specific isotherm
1.0                                   Freundlich/Langmuir Hybrid Ma   (-)            a = 0.305 x 0.58 x rhob
1.0                                   Freundlich/Langmuir Hybrid Mb   (-)
1.7485d-4                             Freundlich/Langmuir Hybrid beta (-)
```

VERSE-LC output:

```
==============================================================================
 VERSE v7.80  by R. D. Whitley and N.-H. L. Wang, c1999 PRF
 ==============================================================================
 Input file: Case1
 TCCR Simulation of Cs removal on CST material lead-lag columns A/B
 Case 1 - Tank 10H Batch 3, 34C, large particle size
 Begin Run:  09:38:01 on 09-02-2021   running under Windows 95/8
 Finite elements    - axial:100  particle: 1
 Collocation points - axial:  4  particle: 6 => Number of eqns:  4010
 Inlet species at equilib.? N   Perfusable sorbent? N   Feed profile only? N
 Use Profile File? Y   Generate Profile File? Y
 Axial dispersion correlation:   Chung & Wen (1968)
 Film mass transfer correlation: Wilson & Geankoplis (1966)
 Sub-Column Boundary Conditions: Flux Continuity
 ==============================================================================
 SYSTEM PARAMETERS (at initial conditions):

 t(stop)        =   6985.00000 min        dtheta max     =      1.00000 BV
 abs. tol.      =       .10000E-06        rel. tol.      =       .10000E-03
 Total Length   =    527.18000 cm         D              =     48.68000 cm
 Tot. Capacity  =       .00000 eq/L solid  Col. Vol.      = 981182.84466 mL
 F              =  18927.00000 mL/min     Uo (linear)    =     18.55710 cm/min
 R              =    286.00000 microns    L/R            =  18432.86713
 Bed Void frac. =       .54800            Pcl. Porosity  =       .24000
 Spec. Area     =     47.41259 1/cm       Time/BV        =     14.20426 min
 Vol CSTRs      =       .00000 mL
```

```
Component no.  =      1
Ke  [-]        =  .10000E+01
Eb  [cm2/min]  =  .27627E+01
Dp  [cm2/min]  =  .29080E-03
Doo [cm2/min]  =  .11630E-02
kf  [cm/min]   =  .32102E+00
Ds  [cm2/min]  =  .00000E+00

Dimensionless Groups:
Re             =  .91622E+00
Sc(i)          =  .54589E+03
Peb(i)         =  .17705E+04
Bi(i)          =  .13155E+03
Nf(i)          =  .39452E+03
Np(i)          =  .12120E+01
Pep(i)         =  .76045E+04

Isotherm       =  Freundlich/Langmuir Hybrid
Iso. Const. 1  =  .17460E+00
Iso. Const. 2  =  .10000E+01
Iso. Const. 3  =  .10000E+01
Iso. Const. 4  =  .10000E+01
Iso. Const. 5  =  .17485E-03
Init. Conc.    =  .00000E+00
Conc. at eqb.  =  .00000E+00
Conc. units          M
===============================================================================
COMMAND LIST:
  1: Step conc. of component 1 at .0000     min to .1160E-04 M
     Execute   1 times, every .0000     mins.
  2: User set viscosity to .1238E-01 poise and density to 1.170     g/cm3
  3: Step change flow at 4444.     min to  .151E+05 mL/min
     Execute   1 times, every .0000     mins.
  4: Step change flow at 6274.     min to  .303E+05 mL/min
     Execute   1 times, every .0000     mins.
  5: Carousel (conc.). Active between t = .0000     and .1000E+07 min.
     When comp.  1 reaches .1160E-03 M     at end of node 100,
     shift  50 axial elements out the feed end
  6: Monitor conc. history at stream  2.  Filename = Case1.h01
     Output density adjustments:
     1.0    *default abs conc delta,      1.0    *default rel conc delta,
     .50    *default force w/ conc delta, .50    *default force w/o conc delta
  7: Monitor conc. history at stream  3.  Filename = Case1.h02
     Output density adjustments:
     1.0    *default abs conc delta,      1.0    *default rel conc delta,
     .50    *default force w/ conc delta, .50    *default force w/o conc delta
  8: Dump full profile file at  .1000E-03 min
     Execute   1 times, every .0000     mins.
  9: Dump full profile file at  6985.     min
     Execute   1 times, every .0000     mins.
===============================================================================
VERSE-LC finished in   516 steps.  Average step size 13.54     minutes
End run:  09:38:12 on 09-02-2021
Integrated Areas in History Files:
Case1.h01                  .381331E-01
Case1.h02                  .138107E-01
```

- CF = 0.305 & τ = 10

VERSE-LC input:

```
TCCR Simulation of Cs removal on CST material lead-lag columns A/B
Case 1 - Tank 10H Batch 3, 34C, large particle size
1, 100, 4, 6                         ncomp, nelem, ncol-bed, ncol-part
FCWNF                                isotherm,axial-disp,film-coef,surf-diff,BC-col
NNNYY                             input-only,perfusable,feed-equil,use datafile.yio,generate/update datafile.yio
M                                    comp-conc units
527.18, 48.68, 18927, 0.0d+0         Total bed length(cm),Diam(cm),Q-flow(ml/min),CSTR-vol(ml)
286.0, 0.548, 0.24,  0.0             part-rad(um), bed-void, part-void, sorb-cap()
0.0                                  initial concentrations (M)
S                                    COMMAND - inlet conc step change
1, 0.0, 1.16d-5, 1, 0.0              spec id, time(min), conc(M), freq, dt(min)
V                                    COMMAND - viscosity/density change
0.01238, 1.17                        fluid viscosity(poise), density(g/cm^3)
s                                    COMMAND - inlet flow rate change
4444.4, 15142, 1, 0.0                time(min), Q-flow(ml/min), freq, dt(min)
s                                    COMMAND - inlet flow rate change
6274, 30283, 1, 0.0                  time(min), Q-flow(ml/min), freq, dt(min)
m                                    COMMAND - subcolumns (carousel-concentration driven)
50, 100, 0, 1, 1.16d-4, 0.0, 1d+6    Nelem shift, Nelem watch, Npp watch, Nc watch, Cthresh, te, tee
h                                    COMMAND - effluent history dump
2, 1.0, 1.0, 0.50, 0.5               unit op#, ptscale(1-4) filtering
h                                    COMMAND - effluent history dump
3, 1.0, 1.0, 0.50, 0.5               unit op#, ptscale(1-4) filtering
D                                    COMMAND - dump column profile
-1, 1.0d-4, 1, 0                     particle point (-1 for all), time(min), freq, dt(min)
D                                    COMMAND - dump column profile
-1, 6985, 1, 0                       particle point (-1 for all), time(min), freq, dt(min)
-                                    end of commands
6985, 1                              end time(min), max step size (B.V.)
1.0d-7, 1.0d-4                       abs-tol, rel-tol
-                                    non-negative conc constraint
1.0d0                                size exclusion factor
1.163d-4                             part-pore diffusivities(cm^2/min) 10% of free value
1.163d-3                             Brownian diffusivities(cm^2/min)
0.1746                               Freundlich/Langmuir Hybrid a    (moles/L B.V.) rhob=0.987 g/ml
1.0                                  Freundlich/Langmuir Hybrid b    (1/M)          Batch specific isotherm
1.0                                  Freundlich/Langmuir Hybrid Ma   (-)            a = 0.305 x 0.58 x rhob
1.0                                  Freundlich/Langmuir Hybrid Mb   (-)
1.7485d-4                            Freundlich/Langmuir Hybrid beta (-)
```

VERSE-LC output:

```
==============================================================================
 VERSE v7.80  by R. D. Whitley and N.-H. L. Wang, c1999 PRF
 ==============================================================================
 Input file: Case1
 TCCR Simulation of Cs removal on CST material lead-lag columns A/B
 Case 1 - Tank 10H Batch 3, 34C, large particle size
 Begin Run:  17:00:25 on 09-01-2021   running under Windows 95/8
 Finite elements    - axial:100  particle: 1
 Collocation points - axial:  4  particle: 6 => Number of eqns:  4010
 Inlet species at equilib.? N   Perfusable sorbent? N   Feed profile only? N
 Use Profile File? Y   Generate Profile File? Y
 Axial dispersion correlation:   Chung & Wen (1968)
 Film mass transfer correlation: Wilson & Geankoplis (1966)
 Sub-Column Boundary Conditions: Flux Continuity
 ==============================================================================
 SYSTEM PARAMETERS (at initial conditions):

 t(stop)        =   6985.00000 min        dtheta max     =      1.00000 BV
 abs. tol.      =       .10000E-06        rel. tol.      =       .10000E-03
 Total Length   =    527.18000 cm         D              =     48.68000 cm
 Tot. Capacity  =       .00000 eq/L solid  Col. Vol.     = 981182.84466 mL
 F              =  18927.00000 mL/min     Uo (linear)    =     18.55710 cm/min
 R              =    286.00000 microns    L/R            =  18432.86713
 Bed Void frac. =       .54800            Pcl. Porosity  =       .24000
 Spec. Area     =     47.41259 1/cm       Time/BV        =     14.20426 min
 Vol CSTRs      =       .00000 mL

 Component no.  =      1
 Ke  [-]        =  .10000E+01
```

```
Eb  [cm2/min]  =  .27627E+01
Dp  [cm2/min]  =  .11630E-03
Doo [cm2/min]  =  .11630E-02
kf  [cm/min]   =  .32102E+00
Ds  [cm2/min]  =  .00000E+00

Dimensionless Groups:
Re             =  .91622E+00
Sc(i)          =  .54589E+03
Peb(i)         =  .17705E+04
Bi(i)          =  .32893E+03
Nf(i)          =  .39452E+03
Np(i)          =  .48471E+00
Pep(i)         =  .19015E+05

Isotherm       =  Freundlich/Langmuir Hybrid
Iso. Const. 1  =  .17460E+00
Iso. Const. 2  =  .10000E+01
Iso. Const. 3  =  .10000E+01
Iso. Const. 4  =  .10000E+01
Iso. Const. 5  =  .17485E-03
Init. Conc.    =  .00000E+00
Conc. at eqb.  =  .00000E+00
Conc. units          M
===============================================================================
COMMAND LIST:
  1: Step conc. of component 1 at .0000     min to .1160E-04 M
     Execute   1 times, every .0000     mins.
  2: User set viscosity to .1238E-01 poise and density to 1.170     g/cm3
  3: Step change flow at 4444.     min to  .151E+05 mL/min
     Execute   1 times, every .0000     mins.
  4: Step change flow at 6274.     min to  .303E+05 mL/min
     Execute   1 times, every .0000     mins.
  5: Carousel (conc.). Active between t = .0000     and .1000E+07 min.
     When comp.  1 reaches .1160E-03 M     at end of node 100,
     shift  50 axial elements out the feed end
  6: Monitor conc. history at stream  2.  Filename = Case1.h01
     Output density adjustments:
     1.0    *default abs conc delta,      1.0    *default rel conc delta,
     .50    *default force w/ conc delta, .50    *default force w/o conc delta
  7: Monitor conc. history at stream  3.  Filename = Case1.h02
     Output density adjustments:
     1.0    *default abs conc delta,      1.0    *default rel conc delta,
     .50    *default force w/ conc delta, .50    *default force w/o conc delta
  8: Dump full profile file at  .1000E-03 min
     Execute   1 times, every .0000     mins.
  9: Dump full profile file at  6985.     min
     Execute   1 times, every .0000     mins.
===============================================================================
VERSE-LC finished in   535 steps.  Average step size 13.06     minutes
End run:  17:00:37 on 09-01-2021
Integrated Areas in History Files:
Case1.h01                  .440269E-01
Case1.h02                  .187570E-01
```

Distribution:

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