ANL/NE-16/43



# Development of ABEC Column for Separation of Tc-99 from NorthStar Dissolved Target Solution

**Nuclear Engineering Division** 

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## Development of ABEC Column for Separation of Tc-99 from NorthStar Dissolved Target Solution

by

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December 2016

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#### DEVELOPMENT OF ABEC COLUMN FOR SEPARATION OF TC-99 FROM NORTHSTAR DISSOLVED TARGET SOLUTION

#### ABSTRACT

Batch and column breakthrough experiments were performed to determine isotherms and mass-transfer parameters for adsorption of Tc on aqueous biphasic extraction chromatographic (ABEC) sorbent in two solutions: 200 g/L Mo, 5.1 M K<sup>+</sup>, 1 M OH<sup>-</sup>, and 0.1 M NO<sub>3</sub><sup>-</sup> (Solution A) and 200 g/L Mo, 9.3 M K<sup>+</sup>, 5 M OH<sup>-</sup>, and 0.1 M NO<sub>3</sub><sup>-</sup> (Solution B). Good agreement was found between the isotherm values obtained by batch and column breakthrough studies for both Solutions A and B. Potassium-pertechnetate intra-particle diffusivity on ABEC resin was estimated by VERSE simulations, and good agreement was found among a series of column-breakthrough experiments at varying flow velocities, column sizes, and technetium concentrations. However, testing of 10 cc cartridges provided by NorthStar with Solutions A and B did not give satisfactory results, as significant Tc breakthrough was observed and ABEC cartridge performance varied widely among experiments. These different experimental results are believed to be due to inconsistent preparation of the ABEC resin prior to packing and/or inconsistent packing.

#### **1 INTRODUCTION**

NorthStar Medical Radioisotopes is pursuing a neutron capture  $[^{98}Mo(n,\gamma)^{99}Mo]$  route for <sup>99</sup>Mo production at the University of Missouri Research Reactor (MURR). Argonne is assisting NorthStar in the development of some aspects of the operation. Once the molybdenum targets are removed from the reactor, they will undergo dissolution. The output of the dissolution results in ~1500 mL of a 200 g/L Mo, 9.3 M  $K^+$ , 5 M OH<sup>-</sup>, 0.1 M NO<sub>3</sub><sup>-</sup> solution containing ~400 Ci of Mo-99, as well as a few other irradiation byproducts, including Tc-99. The solution is pumped from a shielded cask through a chromatography column containing ABEC to remove unwanted Tc present from the target irradiation. This polishing step allows radiopharmacies to use the first aliquot of Tc-99m they elute from their generators. Currently, radiopharmacies discard the first aliquot from the generator, as it contains unacceptable levels of Tc-99. Various ABEC cartridge sizes and flow rates have been tested to optimize the separation of Tc-99 from Mo-99. The results of those experiments along with additional batch and breakthrough experiments were utilized to determine intrinsic parameters, mass transfer, and system parameters for design of a Tc-99 separation process from the irradiated Mo target solution. The ABEC resin consists of polyethylene glycols (PEGs) physisorbed to a 2% crosslinked styrene divinylbenzene copolymer lattice. The amount of resin crosslinkage determines the bead pore size. Therefore, a resin with a lower percentage (2%) of crosslinkage has a more open structure that is permeable to higher molecular-weight substances than a highly crosslinked resin. It also has a lower physical resistance to shrinking and swelling, so it absorbs more water and swells to a larger wet diameter than a highly crosslinked resin of equivalent dry diameter. Therefore, while the kinetics of adsorption on a low crosslinked resin may be superior, consistent packing and channeling may become a challenge in designing a commercial process. The ABEC mode uses high ionic strength salt solutions as mobile phases. In the presence of water-structuring anion salts (e.g.,  $SO_4^{2-}$ , OH<sup>-</sup>), chaotropic ion salts (KTcO<sub>4</sub>) are retained in the sorbent. Stripping is accomplished by washing with water by lowering the ionic strength of the aqueous phase.<sup>1-4</sup>

#### **2 THEORY**

#### 2.1 VERSATILE REACTION-SEPARATION (VERSE) MODEL

The VERSE model and related simulations were developed by Wang and associates in 1991.<sup>5</sup> The general VERSE model takes into account detailed intrinsic (scale independent) parameters, which include the system parameters (particle porosity and bed void fraction), adsorption isotherms, Brownian diffusivities, intra-particle pore diffusivities, axial dispersion coefficient, film mass transfer coefficient, mass-transfer effects, slow adsorption and desorption,<sup>6</sup> and possible chemical reactions in the mobile phase or in the solid phase during the separations.<sup>7-10</sup> Although the general VERSE model can take into account parallel intra-particle pore and surface diffusion, previous studies have shown that at a relatively low concentration region, effects of surface diffusion on frontal or elution chromatography cannot be distinguished from those of pore diffusion. As a result, a pore-diffusion model can be used to predict closely the frontal curves.<sup>11,12</sup> Since the Tc-99 feed concentration in the system of interest is relatively low

 $(\sim 10^{-6} \text{ M})$ , the pore-diffusion model was tested for process simulation and design. The assumptions and equations for the pore-diffusion VERSE model have been reported elsewhere.<sup>5</sup>

#### 2.2 LANGMUIR ADSORPTION ISOTHERM MODEL

When adsorption and desorption rates are much higher than the mass transfer rates, the concentrations in the solid phase and in the solution phase are at equilibrium. Under such conditions, the local solid-phase concentrations are related to the local solution-phase concentrations by an equilibrium isotherm at a given temperature. The Langmuir adsorption isotherm equation used in this study is:

$$q_i = \frac{a_i C_i}{1 + b_i C_i} \tag{1}$$

where  $a_i$  and  $b_i$  are the Langmuir isotherm parameters of species i, and  $q_i$  is the solid phase concentration of species i in equilibrium with the solution phase concentration  $C_i$ . In this study, the unit of  $C_i$  is mmol/L solution, and the unit of  $q_i$  is mmol/L column packing volume.

If bC~1 or bC>>1, the relation between q and C is nonlinear, and the concentration is defined to be in the nonlinear isotherm region. If bC<<1 or  $q \approx aC$ , the concentration in the solution is in the linear isotherm region. The metal ion concentration in the solid phase in equilibrium with the final concentration in the solution can be calculated as follows:

$$q = \frac{(C_0 - C_s)V}{W} \rho_{\text{packing}}$$
(2)

where  $C_0$  is the initial Tc-99 concentration in the solution,  $C_s$  is the final concentration in the solution, and q is the Tc-99 concentration in the solid phase in equilibrium with the final

concentration in the solution. Also, V is the volume of the solution, W is the weight of dry sorbent, and  $\rho_{packing}$  is the packing density of the sorbent, which is defined as the ratio of the sorbent dry weight to the packing volume. Thus, the unit of q is mmol/L packing volume. The ABEC packing density,  $\rho_{packing}$ , is 1000 g/L for Solution A and 1300 g/L for Solution B.

#### 2.3 INTRA-PARTICLE PORE DIFFUSIVITY AND EFFECTIVE CAPACITY

Breakthrough curves for Tc-99 concentration in solution over time result from convection, diffusion, and adsorption effects. The shapes of the curves are affected by the system parameters (column length, particle size, and void fractions), operating conditions (linear velocity and temperature), and the intrinsic parameters (equilibrium adsorption isotherm and mass transfer parameters). Breakthrough curves are useful for verifying the effective column capacity at a given feed concentration (q at C<sub>f</sub>) predicted with the isotherms obtained from the batch tests. The key mass transfer parameter  $D_p$  (intra-particle diffusivity of Tc-99) can be estimated by fitting the experimental Tc frontal curves to the simulation results. The value of  $D_p$ is a function of temperature, sorbent pore structure, and viscosity of the mobile phase. The sorbent capacity at the feed concentration can be calculated from the equation:

$$q_{f} = \frac{C_{f} V_{center}}{V_{column}}$$
(3)

where  $V_{center}$  is the volume of feed solution at the center of the breakthrough curve ( $C_{br}/C_f = 0.5$ ),  $V_{column}$  is the volume of the column,  $q_f$  is the amount of Tc adsorbed per packing volume of sorbent (mmol/LCV), and  $C_f$  is the Tc concentration in the feed solution (mM).

#### **3 EXPERIMENAL**

#### **3.1 MATERIALS**

An ABEC sorbent with particle size of 125-250 µm was obtained from NorthStar and produced by Eichrom Technologies (Lisle, IL). Eichrom pre-packed ABEC 2 cc cartridges (ID, 0.88–0.96 cm; average ID, 0.92 cm; column length, 2.5 cm; and average bed height, 2.125 cm). In addition, NorthStar prepared 10 cc ABEC cartridges (ID, 1.5 cm; length, 6 cm). American Chemical Society (ACS) reagent-grade potassium hydroxide pellets and potassium nitrate crystals were used, as was ACS reagent-grade molybdenum trioxide available from Acros Organics. Ultima Gold XR liquid scintillation cocktail was obtained from Perkin Elmer. Tc-99 was original in the ammonium pertechnetate chemical form and was obtained from Oak Ridge National Laboratory. Omnitfit columns were packed with ABEC resin at Argonne, as described below.

#### **3.2 SOLUTION PREPARATION**

**Solution A** contained 200 g/L Mo, 5.1 M K<sup>+</sup>, 1 M OH<sup>-</sup>, and 0.1 M NO<sub>3</sub><sup>-</sup> in 18 M $\Omega$  deionized distilled (DI) water that had been filtered through a 0.22 µm polyethersulfone (PES) filter available through Fisher Scientific. Subsequently, Tc-99 was added to the Mo solution. An aliquot of this sample was taken for liquid scintillation counting (LSC) as a baseline for the starting solution. It is expected that 170 µg Tc dissolved in 1.5 L of feed solution is the upper limit of Tc in an irradiated natural molybdenum target.

**Solution B** was prepared in the same way and contained 200 g/L Mo, 9.3 M K<sup>+</sup>, 5 M OH<sup>-</sup>, 0.1 M NO<sub>3</sub><sup>-</sup>, and Tc-99.

#### **3.3 EQUIPMENT**

ABEC cartridges were tested on a dispensing unit developed by NorthStar Medical Radioisotopes. The ABEC-packed Omnifit chromatography columns were tested with an AKTA Purifier unit (GE Healthcare, Piscataway, NJ). All column experiments were conducted at room temperature.

#### **3.4 VISCOSITY**

The kinematic viscosity of Solutions A and B was determined in triplicate using a Cross Arm Viscometer from Technical Glass Products, Inc. At room temperature (21.4° C) the viscosity was 0.94 cP for DI water, 1.77 cP for Solution A, and 4.08 cP for Solution B.

#### 3.5 BATCH EQUILIBRIUM TESTS

The uptake of Tc was determined by contacting 1 mL of Solution A or B containing Tc-99 with a known amount  $(10 \pm 1 \text{ mg})$  of sorbent for 30 min at room temperature. After equilibration, the solution was withdrawn and filtered by using a syringe fitted with a PVDF (polyvinylidene fluoride) membrane filter (Millipore, 0.22 µm pore size). Blank experiments demonstrated that the filter does not uptake Tc.

#### **3.6 OMNIFIT COLUMN PACKING**

For a given column size, the amount of sorbent was weighed and placed in a beaker. Water was added to the sorbent to form a 20% (v/v) slurry. The slurry was agitated at 1000 rpm for 3 min to remove air from the sorbent and any impurities which are soluble in water. After the sorbent settled, the excess liquid was decanted. The rinsing and decanting steps were repeated two more times. The slurry was uniformly mixed and poured into the column. Water was pumped downward through the packed bed at a flow rate of 10 mL/min for 30 min. Ten bed volumes of 5 M KOH solution were pumped downward through the packed bed at a flow rate of 10 mL/min. The adjuster was lowered to the surface of sorbent layer and then tightened.

#### **3.7 COLUMN EXPERIMENTS**

The ABEC cartridges were tested with a dispensing unit developed by NorthStar Medical Radioisotopes. The dispensing unit uses pressure to push eluents through the ABEC column. The column was conditioned with ~10 bed volumes of 5 M KOH. After conditioning, an aliquot of 29 mL of Solution A or B, containing Tc-99, was passed through the ABEC cartridge, and a sample was collected for LSC. Either 5 or 50 aliquots were passed through the column, depending upon the experiment. After passing the appropriate number of aliquots through the column, the column was stripped of Tc using 18 M $\Omega$  deionized water. The ABEC packed Omnifit columns were tested with an AKTA Purifier Unit. The columns were pre-equilibrated with ~10 bed volumes of 5 M KOH, feed solution was loaded at 10 mL/min, and aliquots were collected and sampled.

#### **3.8 RADIOMETRIC MEASUREMENTS**

The Tc-99 activity was quantified by measuring its  $\alpha$ -emission on a Perkin-Elmer liquid scintillation counter, and the amount of Tc-99 was calculated by using a calibration curve.

#### **4 RESULTS AND DISCUSSION**

#### 4.1 Tc-99 LANGMUIR ISOTHERM PARAMETERS

Figures 1 and 2 show the absorption isotherms for ABEC uptake of Tc-99 from Solutions A and B at room temperature (RT). The data points were obtained via batch adsorption equilibrium tests calculated using Eq. (2). The Langmuir isotherm model, Eq. (1), was applied to fit the data using the nonlinear analysis tool in Origin 2015. The best fitting of the data indicates that at RT for Solution A, the Tc Langmuir isotherm "a" = 113 L solution volume/L packing volume and "b" = 0 (Figure 1). In Solution B at RT, the Tc Langmuir isotherm constant is nearly an order of magnitude higher, "a" = 970 and "b" = 0 (Figure 2). The driving force for higher Tc extraction from Solution B is the higher ionic strength of as well as increased concentration of  $K^+$ , which coextracts with TcO<sub>4</sub><sup>-</sup> and facilitates uptake on the resin.

#### 4.2 BED VOID FRACTION AND POROSITY

Bed void fraction  $\varepsilon_b=0.3$  was determined empirically. Porosity was calculated by assuming the total void fraction  $\varepsilon_t = 0.6$ . That value was substituted into  $\varepsilon_p = (\varepsilon_t - \varepsilon_b)/(1 - \varepsilon_b)$ , and yielded  $\varepsilon_p=0.4$ . While those numbers are used in VERSE simulations, it needs to be noted that the volume of the stationary phase changes with solution composition and during column loading. Therefore, these numbers may introduce a systematic error and should be treated as arbitrary.



FIGURE 1 ABEC adsorption isotherm of Tc from Solution A (200 g/L Mo, 5.1 M K<sup>+</sup>, 1 M OH<sup>-</sup>, and 0.1 M NO<sub>3</sub><sup>-</sup>) at RT.



FIGURE 2 ABEC adsorption isotherm of Tc from Solution B (200 g/L Mo, 9.3 M K<sup>+</sup>, 5 M OH<sup>-</sup>, and 0.1 M NO<sub>3</sub><sup>-</sup>) at RT.

#### **4.3 INTRA-PARTICLE DIFFUSIVITY**

Brownian diffusivity ( $D_b$ ) of TcO<sub>4</sub><sup>-</sup> in water at 25°C is reported to be  $1.17 \times 10^{-3}$  cm<sup>2</sup>/min.<sup>13</sup> The Stokes-Einstein equation was used to correct for viscosity and temperature effects. The result shows that  $D_b = 5.86 \times 10^{-4}$  cm<sup>2</sup>/min and  $2.54 \times 10^{-4}$  cm<sup>2</sup>/min at RT for Tc in Solutions A and B, respectively. Figures 3-8 present VERSE simulated breakthrough curves along with experimental data obtained from the 2 cc ABEC cartridges in solution A at flow rates of 2, 5, 7, 10, 15, and 20 mL/min, using the NorthStar dispensing unit. The average Tc concentration in Solution A was 0.001 mM.

The VERSE simulations were used to find the intra-particle diffusivity ( $D_p$ ) value that can fit the experimental breakthrough curves for 2 cc ABEC cartridges at 2-20 mL/min. The intrinsic parameters, system parameters, and numerical parameters used in the VERSE simulations are summarized in Table 1. The best fit indicates that  $D_p$  for ABEC Tc in Solution A is  $2.0 \times 10^{-3}$  cm<sup>2</sup>/min (from Figures 3-8). This value is higher than the  $D_b = 5.86 \times 10^{-4}$  cm<sup>2</sup>/min reported in the literature and adjusted for viscosity and temperature effects in Solution A. This increased diffusivity is attributed to an accelerated diffusion effect, where diffusion is increased by coupled diffusion fluxes. Coupled diffusion fluxes occur when the flux of the solute is altered by the concentration gradient of other solutes.<sup>14,15</sup> In addition, the increased diffusivity may be caused by a higher concentration/activity of K<sup>+</sup>, which coextracts with TcO<sub>4</sub><sup>-</sup>. Therefore, it is assumed that in solution A, Tc  $D_p = D_b = 2.0 \times 10^{-3}$  cm<sup>2</sup>/min.



FIGURE 3 Experimental and VERSE simulated breakthrough curves for 2 cc ABEC cartridges at 2 mL/min with 0.00099 mM Tc in Solution A.



FIGURE 4 Experimental and VERSE simulated breakthrough curves for 2 cc ABEC cartridges at 5 mL/min with 0.00096 mM Tc in Solution A.



FIGURE 5 Experimental and VERSE simulated breakthrough curves for 2 cc ABEC cartridges at 7 mL/min with 0.00094 mM Tc in Solution A.



FIGURE 6 Experimental and VERSE simulated breakthrough curves for 2 cc ABEC cartridges at 10 mL/min with 0.0010 mM Tc in Solution A.



FIGURE 7 Experimental and VERSE simulated breakthrough curves for 2 cc ABEC cartridges at 15 mL/min with 0.0011 mM Tc in Solution A.



**FIGURE 8** Experimental and VERSE simulated breakthrough curves for 2 cc ABEC cartridges at 20 mL/min with 0.0011 mM Tc in Solution A.

System Parameters						
Sorbent	R (µm)		ε <sub>b</sub>		ε <sub>p</sub>	
ABEC 85			0.3		0.4	
Mass Transfer Parameters						
Solution	$D_p (cm^2)$	/min)	k <sub>f</sub> (cm/min	n) E <sub>b</sub> (	cm <sup>2</sup> /min)	
А	$2.0 \times$	10 <sup>-3</sup>	From Wilson a	nd From (	Chung and	
В	$3.0 \times$	10 <sup>-3</sup>	Geankoplis (1966) <sup>18</sup>		en (1968) <sup>19</sup>	
Isotherm Parameters (Langmuir)						
Solution		а		b (1/mN	<b>(</b> )	
А		113		0		
В		970		0		
Numerical Parameters						
Axial	Sten Size Collocation F		cation Points	Tolera	nce	
Elements	$(L\epsilon_b/u_s)$	Axia	l Particle	Absolute (mM)	Relative	
100	0.01	4	4	10-7	10-3	

TABLE 1 Parameters used in VERSE simulations forcolumn design

The Tc Langmuir "a" values estimated from breakthrough experiments using Eq. (3) are summarized in Table 2 and are in good agreement with Langmuir isotherm values obtained via batch adsorption equilibrium tests. The results indicate that resin utilization decreases as contact time decreases at higher flow rates. This effect can be explained by the "empty bed contact time" effect,<sup>16,17</sup> where at short contact times KTcO<sub>4</sub> can only diffuse into the polyethylene glycol layer grafted on the resin surface. At the same time, at short contact times, as Tc can only diffuse into the resin surface, the "apparent" D<sub>p</sub> increases. This is exemplified in Figure 9, where at higher flow rates, the actual breakthrough curves become sharper than the simulations. For this reason, as the contact time increases, the resin utilization increases, and the apparent D<sub>p</sub> decreases. The intrinsic parameters, system parameters, and numerical parameters used for the VERSE simulations in Figure 9 are summarized in Table 1.

VERSE simulations were used to find  $D_p$  for ABEC Tc uptake from Solution B by fitting the experimental breakthrough curve for the 2 cc ABEC cartridge at 10 mL/min. The intrinsic parameters, system parameters, and numerical parameters used in VERSE simulations are summarized in Table 1. The best fitting shows that  $D_p$  for ABEC Tc uptake from Solution B is  $0.1 \text{ cm}^2/\text{min}$  (Figure 10), indicating that the Tc diffusivity in Solution B is faster than  $D_b$  for Tc uptake from Solution A. This effect is expected due to accelerated diffusion caused by the concentration gradient of other solutes and the higher concentration/activity of K<sup>+</sup>.<sup>14-15</sup>

TABLE 2 Langmuir "a" values for uptake
of Tc from Solution A calculated from 2 cc
column breakthrough experiments at
different flow rates.

Flow rate (mL/min)	Langmuir a (avg), solution volume/L packing volume
2	112
5	115
7	108
10	58
15	94
20	93



FIGURE 9 Experimental and VERSE simulated breakthrough curves for 10 cc ABEC cartridges at 10, 20, and 30 mL/min for 0.001 mM Tc in Solution A.



FIGURE 10 Experimental and VERSE simulated 0.001 mM Tc breakthrough curve for 2 cc ABEC cartridges at 10 mL/min in Solution B.

Breakthrough curves were obtained from Omnifit columns packed with ABEC sorbent using the AKTA system. Solution B containing either 0.00386 mM Tc-99 or 0.00215 mM Tc-99 was loaded on a column with 0.66 cm ID x 0.4 cm length or 0.66 cm ID x 0.8 cm length, respectively, and tests were run at 10 mL/min and RT. VERSE simulations were used to find the  $D_p$  value that fits the experimental breakthrough curves. The intrinsic parameters, system parameters, and numerical parameters used in VERSE simulations are summarized in Table 1. The best fitting indicates that the  $D_p$  for ABEC Tc uptake in Solution B is  $3.0 \times 10^{-3}$  cm<sup>2</sup>/min (Figures 11 and 12). This value is higher than the  $D_b = 2.0 \times 10^{-3}$  cm<sup>2</sup>/min for ABEC Tc uptake in Solution A. This higher value is caused by an accelerated diffusion effect, where diffusion is increased by coupled diffusion fluxes due to a higher concentration gradient of other solutes and a higher concentration of K<sup>+</sup>.<sup>14,15</sup> The  $D_p$  for Tc in Solution B measured in those experiments is lower than that measured with the 2 cc ABEC cartridge (Figure 10). The Tc Langmuir "a" values were estimated from the breakthrough data in Figures 11 and 12 using Eq. (3) and found to be a = 928 and 939, respectively. These values are in good agreement with the Langmuir isotherm values obtained via batch adsorption equilibrium tests.



FIGURE 11 Experimental and VERSE simulated breakthrough curve for Omnifit ABEC packed column (0.66 cm ID x 0.4 cm length) loaded with Solution B containing 0.00358 mM Tc at 10 mL/min.



FIGURE 12 Experimental and VERSE simulated breakthrough curve for Omnifit ABEC packed column (0.66 cm ID x 0.8 cm length) loaded with Solution B containing 0.00184 mM Tc at 10 mL/min.

#### 4.4 TESTING OF 10 CC ABEC CARTRIDGES AT 10 ML/MIN

Twenty 10 mL ABEC cartridges have been tested at 10 mL/min flow rates using Solution A containing 0.001 mM Tc-99. The results of three experiments were discarded due to low concentrations of Tc-99.

Figure 13 shows the breakthrough curves for 17 ABEC cartridges. The experimental data indicate a wide array of column performances under identical experimental conditions. In addition, VERSE was used to simulate Tc breakthrough on a 10 cc ABEC cartridge (1.5 cm ID x 6 cm long) using the parameters summarized in Table 1. The VERSE simulated breakthrough curves predict the results of PAT 3, PAT 7, PAT 8, PAT 10 and the breakthrough experiments for New Solution Column 3. The VERSE simulation of breakthrough curves generated by reducing the bed height by 25% (to 4.5 cm) or reducing the resin capacity by 25% fits the results of PAT 1, PAT 2, PAT 4, PAT 5, and New Solution Column 2, 4, and 5. Alternatively, VERSE simulation of breakthrough curves generated by increasing the bed height by 17% (to 7 cm) or increasing the resin capacity by 17% fits the results of PAT 6 and PAT 9. The inconsistent experimental results of Tc-99 uptake in Soluiton A are believed to be due to an inconsistent preparation of the ABEC resin prior to packing and/or inconsistent cartridge packing.

Sixteen 10 mL ABEC cartridges have been tested at 10 mL/min flow rates using Solution B containing 0.001 mM Tc-99. Figure 14 shows the breakthrough data for these ABEC cartridges. The data indicate a wide array of column performances under identical experimental conditions. In addition, VERSE was used to simulate Tc breakthrough on a 10 cc ABEC cartridge (1.5 cm ID x 6 cm long) using the parameters summarized in Table 1. The VERSE simulated breakthrough curve predicts that Tc breakthrough is below detection limits. As suggested above for Solution A, the inconsistent experimental results of Tc-99 uptake in Solution B are believed to be due to an inconsistent preparation of the ABEC resin prior to packing and/or inconsistent cartridge packing.



FIGURE 13 Experimental and VERSE simulated breakthrough curves for 10 cc ABEC cartridges with 0.001 mM Tc at 10 mL/min in Solution A.



FIGURE 14 Experimental and VERSE simulated breakthrough curves for 10 cc ABEC cartridges with 0.001 mM Tc at 10 mL/min in Solution B.

#### **5** CONCLUSIONS

Batch and column breakthrough experiments were performed to determine isotherms and mass-transfer parameters for adsorption of Tc on the ABEC sorbent in Solution A (200 g/L Mo, 5.1 M K<sup>+</sup>, 1 M OH<sup>-</sup>, and 0.1 M NO<sub>3</sub><sup>-</sup>) and Solution B (200 g/L Mo, 9.3 M K<sup>+</sup>, 5 M OH<sup>-</sup>, and 0.1 M NO<sub>3</sub><sup>-</sup>). Good agreement was found between the isotherm values obtained by batch and column breakthrough studies for Solutions A and B. The Tc intra-particle diffusivity on ABEC resin was estimated by using VERSE simulations, and good agreement was found among a series of column-breakthrough experiments at varying flow velocities, column sizes, and Tc concentrations. However, testing of 10 cc cartridges provided by NorthStar with Solutions A and B did not give satisfactory results as significant Tc breakthrough was observed, and ABEC cartridge performance varied widely among experiments. The inconsistent experimental results of Tc-99 uptake on the 10 cc ABEC cartridges with Solutions A and B are believed to be due to inconsistent preparation of the ABEC resin prior to packing and/or inconsistent packing.

Based on discussions with NorthStar in November 2015, we determined that the ABEC polishing step should be removed from their front-end processing of the MURR Mo product. The ABEC polishing step was in place to remove Tc that had been produced from Mo-99 decay or been directly produced during irradiation from the Mo product. US Pharmacopeia studies on the Mo product both with and without the ABEC polishing step indicated very little change in purity. The ABEC column was eliminated from the process because the Mo product passed Pharmacopeia testing, and the experimental results on the ABEC column performance were inconsistent.

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