Multi-component Ion Exchange Kinetics with PAN-KCoFC Composite Ion Exchanger

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Abstract−An ion exchange kinetic study was performed using PAN-KCoFC for removal of cesium ion from mixed solution of Cs, Sr, Ni and Ba ions. Uptake curves were obtained for a multi-component ion exchange system as well as binary system. A PAN-KCoFC composite ion exchanger showed higher selectivity for Cs ion over the Sr, Ba, Ni ions. A homogeneous model could predict accurately the uptake curve for both the binary and multi-component systems. Solid phase effective pore diffusivities obtained by modeling ranged between 10^{-5} cm^2/min and 10^{-6} cm^2/min.

Key words: Composite Ion Exchanger, Ion Exchange Kinetics, Cesium Ion, Radionuclides, PAN, KCoFC

INTRODUCTION

Selective removal of cesium from radioactive waste solution has long been an important issue, due to its hazardous effect on the environment [Enarsson et al., 1998]. Various inorganic ion exchangers have been applied to separate cesium [Anthony et al., 1993; Sinha et al., 1994, 1995; Ernest et al., 1997; Mimura et al., 1997; Tarasevich et al., 1997; Mardan et al., 1999; Marageh et al., 1999; Moon et al., 2004].

Among them, transition metal hexacyanoferrates such as NiFC, KNiFC and KCoFC are reportedly one of the selective ion exchangers for cesium [Rao et al., 1999; Mimura et al., 1997; Moon et al., 2004]. However, as in most inorganic ion exchangers, they are synthesized in powder form and can cause operational problems such as pressure drop and filtration in a column if used as they are. To solve this problem, they should be pelletized or immobilized to the other supporting material. We introduced a polyacrylonitrile (PAN) for binding KCoFC powders to form a PAN-KCoFC composite bead. In this composite ion exchanger, PAN is merely an inert binder and all the cesium ions are adsorbed on the KCoFC. No radionuclides are released even when the PAN is decomposed so that it can be disposed underground safely after cementation or vitrification [John et al., 1997]. In our previous studies [Moon et al., 2000, 2002; Kim et al., 2003], we prepared several PAN based composite ion exchangers such as PAN-zeolite 4A, PAN-K$_2$Ti$_4$O$_9$ and PAN-NiFC successfully. And Sebesta et al. [1995] reported possible application of the various PAN based composite ion exchangers for radionuclides removal. Although PAN-based composite ion exchangers are recently reviewed as the promising ones for removal of radionuclides, more evaluation is required on its performance, especially equilibrium and kinetic behavior. For an optimal design of ion exchange process, it is important to evaluate the dynamic behavior of the ion exchange system. In this study we evaluated ion exchange kinetics of a PAN-KCoFC for multicomponent solution as well as binary solution. A homogeneous model was used to predict ion exchange kinetic parameters.

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MATHEMATICAL MODELING

A mathematical model was developed to simulate the diffusion kinetics for a batch ion exchange system, which is considered to be a completely mixed batch reactor where the solid particles are suspended in the liquid phase containing ionic species diffusing into the particles. We used the homogeneous diffusion model, which assumed that the particle is a homogeneous solid, as shown in Fig. 1. The homogeneous model incorporates mathematical descriptions of the following processes: 1) mass transfer resistance in the liquid film surrounding the ion exchanger particle, 2) local equilibrium adjacent to the exterior surface of the ion exchanger, and 3) diffusion resistance within the particle. In Fig. 1, c_b is the cation concentration in the bulk fluid, c_s is the cation concentration in liquid phase at the particle surface, and q is the cation concentration in the solid phase. At the particle surface, c_s is in equilibrium with q. It is assumed that the system is isothermal and the liquid phase concentration is uniform throughout the reactor except for the liquid film. The ion exchange rate is assumed to be much faster than the diffusion rate. Therefore, the ion exchange rate does not affect the kinetics of the system. The diffusion in the solid particle is assumed to follow Fick's law.

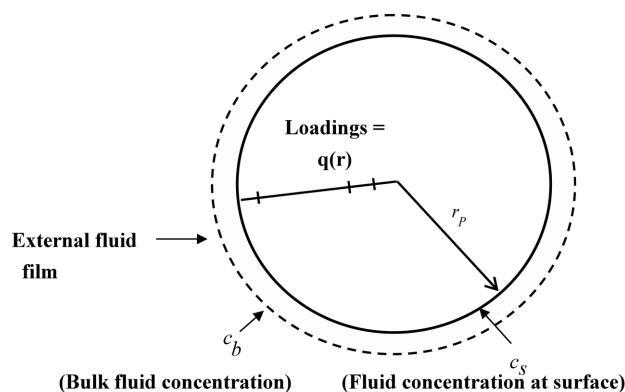The unsteady-state diffusion in a solid sphere can be described by the following partial differential equation:

Fig. 1. Homogeneous diffusion model.

$$\frac{\partial q}{\partial t}=\frac{1}{r^2}\frac{\partial}{\partial r}\left(r^2 D_c\frac{\partial q}{\partial r}\right) \tag{1}$$

where D_c is the solid phase diffusivity for a cation. If D_c is concentration-dependent, it can be represented by Darken's law:

$$D_c=D_c^0\frac{d\ln c}{d\ln q}=D_c^0 H(q) \tag{2}$$

where D_c^0 is the corrected diffusivity, independent of concentration. Substituting Eq. (2) into Eq. (1) gives

$$\frac{\partial q}{\partial t}=D_c^0\left[H(q)\frac{1}{r^2}\frac{\partial}{\partial r}\left(r^2\frac{\partial q}{\partial r}\right)+\frac{\partial H(q)}{\partial r}\frac{\partial q}{\partial r}\right] \tag{3}$$

At the particle surface, $r=r_c$, equilibrium exists between the liquid phase concentration (c_s) and the solid phase concentration (q), expressed by the following equilibrium isotherm equation:

$$q=f(c_s) \qquad\qquad \text{at } r=r_c \tag{4}$$

For the Langmuir isotherm, Eq. (4) becomes

$$q=\frac{ac_s}{1+bc_s} \qquad\qquad \text{at } r=r_c \tag{5}$$

Thus, Darken's law in Eq. (2) is given by the following equation:

$$H(q)=\frac{a}{a-bq} \tag{6}$$

A material balance at the particle surface leads to the boundary condition:

$$\frac{\partial\bar{q}}{\partial t}=\frac{3}{r_c^3}\int_0^{r_c}\frac{\partial q}{\partial t}r^2 dr=\frac{3k_f}{\rho r_c}(c_b-c_s) \qquad \text{at } r=r_c \tag{7}$$

where $\bar{q}$ is the average concentration in the solid phase and k_f is the mass transfer coefficient in the liquid film. Eq. (7) can be modified by substituting Eq. (1) into the integral in Eq. (7).

$$\left[H(q)\frac{\partial q}{\partial r}\right]_{r=r_c}=\frac{k_f}{\rho D_c^0}(c_b-c_s) \qquad \text{at } r=r_c \tag{8}$$

Thus,

$$c_s=c_b-\frac{\rho r_c}{Bi}\left[H(q)\frac{\partial q}{\partial r}\right]_{r=r_c} \qquad \text{at } r=r_c \tag{9}$$

where Bi is the non-dimensional Biot number, defined as:

$$Bi=\frac{k_f/r_c}{D_c^0/r_c^2} \tag{10}$$

The Biot number measures the relative resistance contributed by the liquid film surrounding the particle to the internal diffusion resistance. For most systems, the internal resistance is more important than the film resistance. If the film mass transfer resistance is insignificant, Eq. (9) is replaced by the equation:

$$c_b=c_s \qquad\qquad \text{at } r=r_c \tag{11}$$

Under this condition, q at r_c is in equilibrium with c_b.

The overall mass balance leads to:

$$\varepsilon\frac{\partial c_b}{\partial t}+(1-\varepsilon)\rho\frac{\partial\bar{q}}{\partial t}=0 \tag{12}$$

where ε is the void fraction in the batch reactor and ρ is the particle density. Substituting Eq. (7) into Eq. (12) and then combining it with Eq. (8) gives

$$\frac{\partial c_b}{\partial t}=-\frac{3(1-\varepsilon)k_f}{\varepsilon r_c}(c_b-c_s)=-\frac{3(1-\varepsilon)\rho D_c^0}{\varepsilon r_c}\left[H(q)\frac{\partial q}{\partial r}\right]_{r=r_c} \tag{13}$$

Finally, the initial conditions are:

$$c_b=c_0,\ c_s=q=0 \qquad\qquad \text{at } t=0. \tag{14}$$

The above equations can be rewritten in dimensionless form by introducing the following dimensionless variables:

$$C_b=\frac{c_b}{c_0},\ \ C_s=\frac{c_s}{c_0},\ \ Q=\frac{q}{q_0},\ \ \lambda=\frac{r}{r_c} \tag{15}$$

where q_0 is in equilibrium with c_0. The symmetry of the problem at $\lambda=0$ suggests the transformation of the independent variable [Rice and Do, 1995], $\eta=\lambda^2$. With these new variables, Eqs. (3), (5), (6), (9), (13), and (14) become

$$\frac{\partial Q}{\partial t}=\frac{D_c^2}{r_c^2}\left[H(Q)\left(4\eta\frac{\partial^2 Q}{\partial\eta^2}+6\frac{\partial Q}{\partial\eta}\right)+4\eta\frac{\partial H(Q)}{\partial\eta}\frac{\partial Q}{\partial\eta}\right] \tag{16}$$

$$Q=\frac{ac_0 C_s}{q_0(1+bc_0 C_s)} \qquad\qquad \text{at } \eta=1 \tag{17}$$

$$H(Q)=\frac{a}{a-bq_0 Q}=1+bc_0 C_s \tag{18}$$

$$C_s=C_b-\frac{2\rho q_0}{c_0 Bi}\left[H(Q)\frac{\partial Q}{\partial\eta}\right]_{\eta=1} \qquad \text{at } \eta=1 \tag{19}$$

In the case of the insignificant film mass transfer resistance, this must be replaced by the equation:

$$C_s=C_b \qquad\qquad \text{at } \eta=1 \tag{20}$$

$$\frac{\partial C_b}{\partial t}=-\frac{3(1-\varepsilon)k_f}{\varepsilon r_c}(C_b-C_s)=-\frac{6(1-\varepsilon)\rho q_0 D_c^0}{\varepsilon c_0 r_c^2}\left[H(Q)\frac{\partial Q}{\partial\eta}\right]_{\eta=1} \tag{21}$$

$$C_b=1,\ C_s=Q=0 \qquad\qquad \text{at } t=0 \tag{22}$$

Eqs. (16) to (22) constitute a set of the partial differential equations (PDEs) which should be solved by a numerical method. In this work, we converted the PDEs into ordinary differential equations (ODEs) by using the orthogonal collocation method [Villadsen and Michelsen, 1978]. The domain $\eta\in(0,1)$ is now represented discretely by N interior collocation points. Taking the boundary point ($\eta=1$) as the (N+1)-th point, we have a total N+1 interpolation points. According to the orthogonal collocation method, the first and second derivatives at these interpolation points are related to the functional values at all points as given below:

$$\left.\frac{\partial Q}{\partial\eta}\right|_k=\sum_{j=1}^{N+1}A_{kj}^\eta Q_j,\ \ \left.\frac{\partial^2 Q}{\partial\eta^2}\right|_k=\sum_{j=1}^{N+1}B_{kj}^\eta Q_j \tag{23}$$

for $k=1, 2, \ldots, N+1$. The matrices A_{kj} and B_{kj} are constant matrices once N+1 interpolation points have been chosen.

Eq. (16) is valid at any point within the η-domain. Thus, evaluating that equation at the k-th interior collocation point we get:

$$\frac{dQ_k}{dt}=\frac{D_c^0}{r_c^2}\left\{H(Q_k)\sum_{j=1}^{N+1}C_{kj}^\eta Q_j+4\eta_k\left[\sum_{j=1}^{N+1}A_{kj}^\eta H(Q_j)\right]\left[\sum_{j=1}^{N+1}A_{kj}^\eta Q_j\right]\right\} \tag{24}$$

for k=1, 2, …, N, where $C_{kj}^{\eta}=4\eta_k B_{kj}^{\eta}+6A_{kj}^{\eta}$.

The isotherm equation, Darken's law, and the boundary conditions, Eqs. (17) to (20), become

$$Q_{N+1}=\frac{ac_0C_s}{q_0(1+bc_0C_s)} \qquad \text{at k=N+1} \qquad (25)$$

$$H(Q_k)=\frac{a}{a-bq_0Q_k} \qquad \text{for k=1, 2, …, N} \quad (26)$$

$$H(Q_{N+1})=1+bc_0C_s \qquad \text{at k=N+1} \qquad (27)$$

$$C_s=C_b-\frac{2\rho q_0}{c_0 Bi}\left[H(Q_{N+1})\sum_{j=1}^{N+1}A_{N+1,j}^{\eta}Q_j\right] \qquad \text{at k=N+1} \qquad (28)$$

$$C_s=C_b \qquad \text{at k=N+1 (insignificant film mass transfer resistance)} \qquad (29)$$

In the case of using Eq. (28), we should solve it for the concentration at the boundary (C_s) in terms of other dependent variables, C_b, Q_1, Q_2, …, Q_{N+1}. However, as Q_{N+1} is a function of C_s, Eq. (28) becomes a nonlinear algebraic equation for C_s. To solve Eq. (28), we used the Newton-Raphson method.

Eq. (21) becomes

$$\frac{dC_b}{dt}=-\frac{3(1-\varepsilon)k_f}{\varepsilon r_c}(C_b-C_s) \qquad (30)$$

or

$$\frac{dC_b}{dt}=-\frac{6(1-\varepsilon)\rho q_0 D_c^0}{\varepsilon c_0 r_c^2}\left[H(Q_{N+1})\sum_{j=1}^{N+1}A_{N+1,j}^{\eta}Q_j\right] \qquad (31)$$

Finally, Eq. (22) becomes

$$C_b=1, C_s=0, Q_k=0 \text{ (k=1, 2, …, N+1)} \qquad \text{at t=0} \qquad (32)$$

Eqs. (24) to (32) represent a set of (N+1) ODEs: the N equations for the diffusion within the particle, and one equation for the overall mass balance. The system of equations has (N+1) unknowns: Q_1, Q_2, …, Q_N along the particle radius, and C_b. The remaining equations are the equilibrium isotherm equations and the boundary condition at the particle surface. This set of the ODEs was solved by utilizing the IMSL/Math subroutine IVPAG, which is based on the Gear's stiff integration algorithm.

EXPERIMENTAL

Ion exchange kinetic tests were performed in a batch reactor using binary and multicomponent solutions containing Cs, Sr, Ni and Ba in isothermal condition of 25 °C. In each experiment, 1 g of preconditioned PAN-KCoFC was added to the reactor which contained 100 mL of the testing solutions. The initial concentrations and pHs of each testing solutions were held constant at 0.01 N and 2, respectively. Impeller speeds were varied between 500 and 1,000 rpm. The equilibrated solutions were decanted by using 0.2 μm syringe filter and analyzed by AAS (Perkin-Elmer, Model 1100B).

RESULTS AND DISCUSSION

1. Uptake Behaviors

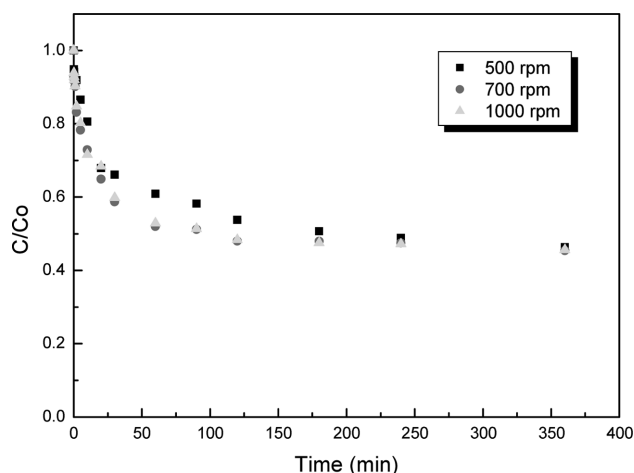The effect of stirring rate on the uptake rate of PAN-KCoFC com-

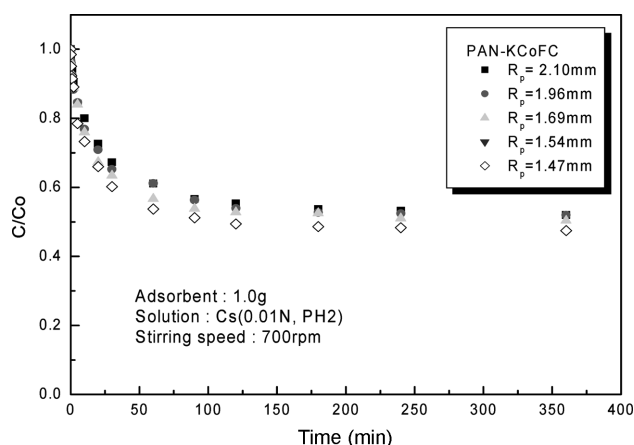Fig. 2. Effect of stirring speed on the uptake rate of cesium ion with PAN-KCoFC.

Fig. 3. Effect of particle size on the uptake rate of Cs ion with PAN-KCoFC.

posite ion exchanger for Cs ion was investigated and the result is represented in Fig. 2. It shows that the stirring speed did not affect the uptake rate in the case of more than 700 rpm. Therefore, most experiments were carried out at 700 rpm and film mass transfer resistance was not considered in modeling the uptake curve.

Fig. 3 shows the uptake rate of cesium ion for the different sizes of PAN-KCoFC particles. The particle radii were varied in the range between 1.47 mm and 2.10 mm. It shows that the particle size variation did not affect the uptake rate seriously. Considering that the PAN-KCoFC beads are mainly composed of macropores of average size of 0.078 μm and nonporous structure of KCoFC, it possibly means that the mass transfer resistance in macropore in the given particle size range is not significant.

Fig. 4 shows uptake curves for binary systems with the 1.47 mm radius PAN-KCoFC particles. The loading of cesium is much higher than the other cations as expected in the equilibrium isotherms obtained in our previous study [Moon et al., 2004]. The uptake behaviors for the ternary system are shown in Fig. 5, which shows much more favorable uptake for cesium over strontium ion. Separation of Cs and Sr is essential since they are importantly reused for irradiation sources rather than disposal. In the case of four compo-
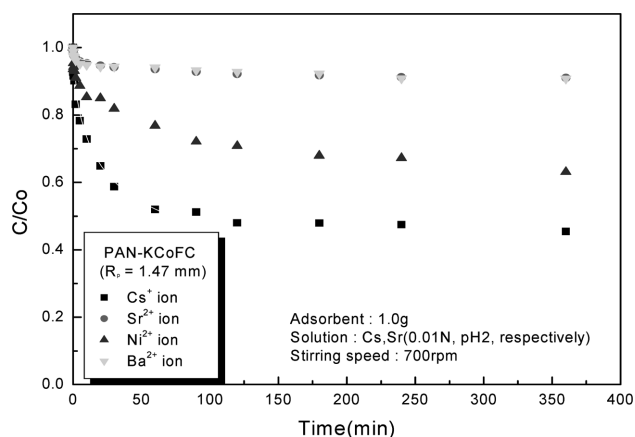
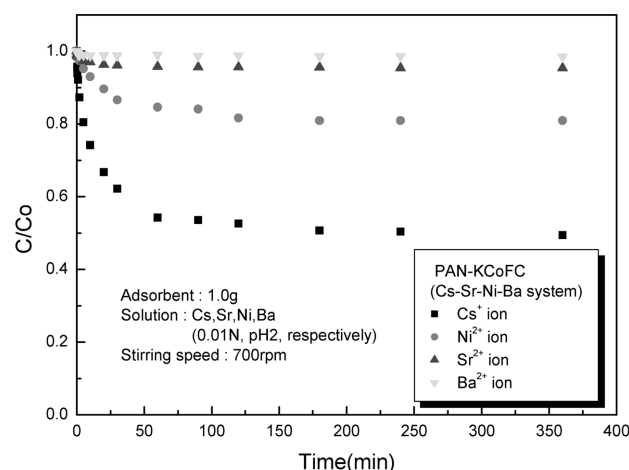**Fig. 4. Uptake rates for binary ion exchange system with PAN-KCoFC.**

Fig. 5. Uptake rates for ternary ion exchange system with PAN-KCoFC.

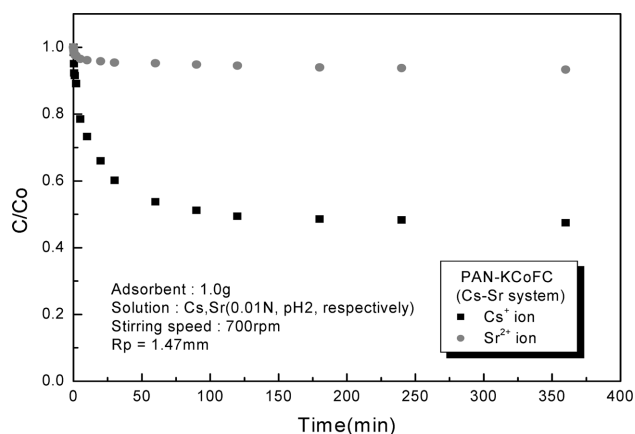**Fig. 6. Uptake rates for multicomponent ion exchange system with PAN-KCoFC.**

nent solution, uptake behaviors as in Fig. 6 are similar to those for a binary system, except that the loadings for Cs and Ni are lower and the favorability for Cs is higher than in the binary system. The lower loadings for the specific ions in multi-component system are obviously due to the competition among the coexisting cations.

2. Modeling of Experimental Data

A homogeneous model was used to model the kinetic data and to evaluate the solid phase effective diffusivities. Here, the film mass transfer coefficient is assumed to be negligible since the uptake rates are not a function of the stirring speed. The effective diffusivity is considered to be dependent on the solute concentration in solid phase and the model is incorporated into to Darkens Law as in Eq. (2). A

Langmuir isotherm was used and the equilibrium parameters together with the physical properties used for modeling are represented in Table 1. Here the particle void fraction and density were measured by using a mercury porosimeter (Micromeritics, AutoPoreIII). The examples of modeling results for the binary and multi-component systems are shown in Figs. 7 and 8.

In the case of Cs and Ni ion exchange system, a homogeneous model predicts the experimental curves quite accurately both in the binary and multi-component system. It might indicate that the PAN-KCoFC beads have homogeneous pore structures, even though the PAN-KCoFC beads are composed of the two phases of KCoFC powder and PAN binder. This result is consistent with the reference [Gu et al., 1997] which used two phase homogeneous model for a silicotitanate ion exchanger, and comparable with other results for zeolite particles [Robinson et al., 1994] which have obviously heterogeneous macro and micro pore structures. Whereas, in the case of Ba and Sr ions the model does not predict the uptake curve so accurately, possibly due to the very low uptake behaviors and inaccurate evaluation of the equilibrium parameters. Considering that the PAN-KCoFC is mainly designed for selective separation of Cs ion, a little deviation is not considered to be so meaningful. The solid phase effective diffusivities obtained for binary systems are shown to range between 10^{-5} cm^2/min and 10^{-6} cm^2/min and on the order of 10^{-5} cm^2/min for multi-component systems. The effective diffusivities for PAN-KCoFC system are rather new so that they cannot be compared with the other results directly. However, if we look into the fact that the macropore diffusivities for clay-binder zeolite particles are reportedly on the order of 10^{-4} cm^2/min and in-

Table 1. Equilibrium parameters and physical data used for modeling

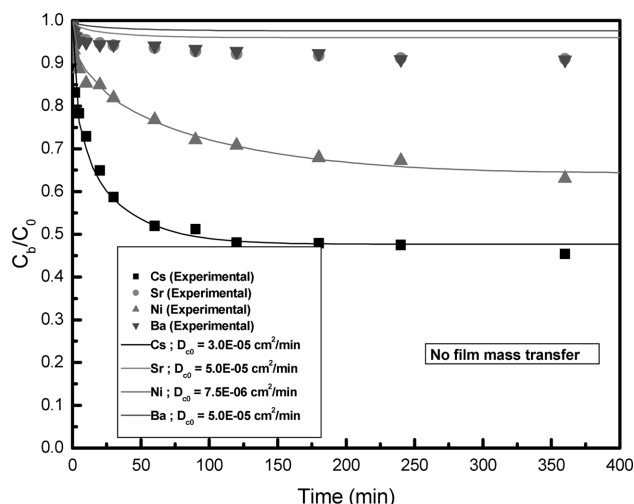| Parameters	System	Binary system				Multi-component system			
		Cs	Sr	Ni	Ba	Cs	Sr	Ni	Ba
Langmuir	a	189.66	5.85	255.86	2.96	531.06	3.83	94.00	2.25
parameter	b	176.92	45.11	563.58	23.28	761.60	38.24	396.41	23.74
Particle void fraction		0.73				0.73			
Particle density		0.18				0.18			

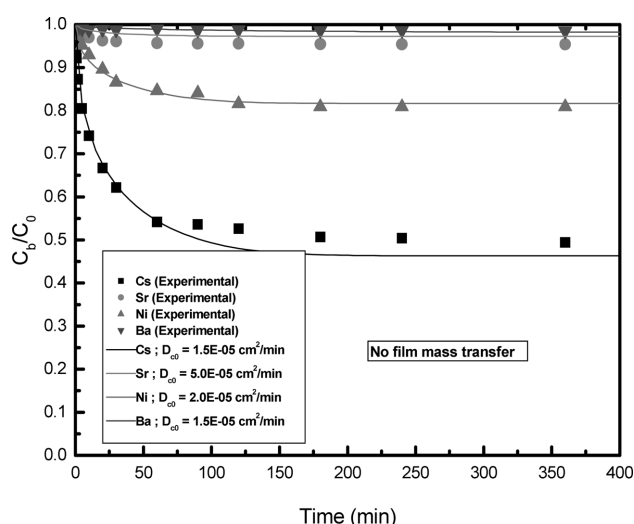**Fig. 7. Modeling examples for binary ion exchange system with PAN-KCoFC.**

Fig. 8. Modeling examples for multi-component ion exchange system with PAN-KCoFC.

tracrystalline diffusivities are normally less than 10^{-8} cm^2/min, those for PAN-KCoFC particles are considered to range between macro and intracrystalline diffusivities. And also considering the macropore resistances for PAN-KCoFC system are not significant and KCoFC powder are almost nonporous, the main mass-transfer resistances are possibly due to the diffusion through the very thin layer which is formed on the surface of the beads by the PAN binder, and this fact would require further investigation.

CONCLUSION

An ion exchange kinetic study was performed using PAN-KCoFC which was newly prepared for selective removal of cesium in high level waste solution. The uptake curves of a PAN-KCoFC composite ion exchanger were obtained for multi-component ion solution as well as binary solution. They showed higher selectivity for Cs ion over the Sr, Ba and Ni ions. The particle radii variation in

the range between 1.47 mm and 2.10 mm did not affect the uptake rate seriously, which means little macropore resistance. A homogeneous model, assuming no film mass transfer resistances, could predict accurately the uptake curve for both the binary and multi-component systems. Solid phase effective pore diffusivities for Cs, Ni, Sr and Ba ions were found to range between 10^{-5} cm^2/min and 10^{-6} cm^2/min.

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