A physics-based mesoscale phase-field model for predicting the uptake kinetics of radionuclides in hierarchical nuclear wasteform materials

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A B S T R A C T

Multiscale, hierarchical, porous materials are promising nuclear wasteform materials with potential for efficiently adsorbing and sequestering radionuclides. However, fundamental understanding of such complex micro- and meso-structures on radionuclide diffusion and uptake kinetics is lacking, which is essential for designing advanced nuclear wasteform materials. In this work we develop a microstructural-dependent diffusion model that accounts for three-dimensional (3D) microstructural features, including heterogeneous thermodynamic and kinetic properties, to predict radionuclide uptake kinetics in complex porous structures. Na\(^+\) and Sr\(^2+\) ionic exchange in LTA zeolite multiscale materials in batch tests is taken as a model system to demonstrate the model and its application to hierarchical materials with multiscale porosity. It is found that the reduction of ionic mobility associated with chemistry, structure, and/or phase changes has more profound impact on the uptake kinetics in a large 3D porous particle than that in a small one. Uptake kinetics in large particles are limited by two diffusion processes: bulk diffusion and surface layer transport. Decreasing particle size and increasing mesoscale pore volume fraction changes the uptake kinetics from two diffusion processes to a single process and dramatically increases the uptake kinetics. Predicted uptake kinetics and the effect of microstructures on the effective capacity of radionuclides and uptake efficiency are consistent with results observed in Na\(^+\)/Sr\(^2+\) exchange experiments. The results demonstrate that the developed model should be adaptable to transport problems in other hierarchical material systems.

1. Introduction

A number of advanced nuclear wasteform materials have been developed to selectively extract radionuclides from different waste streams (liquid or gaseous) \cite{1,2}. For example, a porous silica or glass-based material (ferrocyanide compounds, zeolitic structures, and silver-nitrate phases), surface functionalized nanoparticles (hexacyanoferrate (HCF) nanoparticles, and multi-metallic nanoparticles) salt inclusion materials (SIMs), and metal-organic frameworks (MOFs), are currently the most widely studied materials to capture and stabilize specific radionuclides. However, nuclear waste management generally overlooks nuclear wasteform materials with high performance (i.e., radionuclide capacity, uptake efficiency, and structure stability) that can be used directly as raw materials for a specific containment stream. Experiments show that the radionuclide capacity and efficiency of the uptake process strongly depend on wasteform multi-scale hierarchical structures \cite{3–7}. A number of factors, including inhomogeneous chemical potentials and mobility of radionuclides in different phases and on interfaces or grain boundaries, the reduction of ionic mobility associated with ionic exchange-induced structure distortion, chemistry change and phase transition, and the effect of multiscale pore structures on fluid field and concentration field, may affect ionic diffusion, hence, wasteform material performance. To optimize wasteform structures for high capacity and efficient uptake, it is crucial to understand the mechanisms of ionic diffusion and to predict the effect of hierarchical structures and multi-physics interactions on ionic diffusion, sorption capacity, and ion uptake kinetics.

Multiscale modeling and simulation, which have emerged as valuable approaches for the development of advanced materials \cite{8–11}, have been applied to both framework and porous materials. Density Functional Theory (DFT) can be used to analyze structures and bonding of cations and water within typical framework molecules, such as

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zeolites \([12,13]\). Molecular Dynamics (MD) simulations are used to study free energies, cation thermodynamics, and kinetic properties \([14–16]\). Monte Carlo (MC) techniques allow calculation of cation selectivity and equilibrium ionic exchange properties \([17–19]\), and thermodynamic calculations can assess the effect of porosity on the free energy and phase stability of porous structures \([20–22]\). With knowledge from atomic-level simulations and thermodynamic calculations of phase stabilities, the mesoscale phase-field (PF) method offers a promising modeling tool to predict the effects of 3D microstructures and processing parameters on microstructure evolution and material performance in radionuclide uptake processes \([10,23]\).

In this work we use the PF method to develop a microstructure dependent diffusion model for ion exchange in hierarchically porous zeolite structures. The model accounts for 3D hierarchically porous microstructures and inhomogeneous thermodynamic and kinetic properties, which allows the study of the radionuclide uptake kinetics in these complex structures. PF models of spinodal decomposition and grain growth are used to computationally generate hierarchically porous structures as our model structures. Na\(^+\) and Sr\(^{2+}\) ion exchange in Na-LTA (Sodium Linde Type A) zeolite monoliths in batch uptake tests are taken as model systems to demonstrate the model and its applications.

2. Description of multi-species diffusion in hierarchical materials

LTA zeolite monoliths typically exhibit a hierarchical porous structure consisting of (1) a SiO\(_2\) skeleton with micro-scale macroscopic pores; (2) aggregates of zeolite crystalline particles with nanosized mesoscopic pores; and (3) zeolite crystals with 4Å microscale channels \([24]\). Fig. 1 shows a typical LTA zeolite microstructure.

When a liquid waste stream containing radioactive ions flows through a porous structure ion exchange can occur. In the case of a Na-LTA zeolite monolith and a Sr-containing waste stream, Na\(^+\) inside the zeolite crystals diffuses out while Sr\(^{2+}\) in the waste stream diffuses into the zeolite crystals. This work mainly focuses on the effects of micro- and meso-structures and heterogeneous thermodynamic and kinetic properties on the Sr\(^{2+}\) (or other ion species) uptake kinetics during an ion exchange process. For simplicity, we consider a batch test situation so that the effects of fluid flow in the multi-scale porous structure on Sr\(^{2+}\) uptake kinetics are ignored for this work, although the PF method can be used to describe fluid flow via the Navier-Stokes equations \([27,28]\). It is also assumed that diffusion of the respective ions (Na\(^+\) and Sr\(^{2+}\)) is charge neutral. The charge neutrality is enforced by chemical potentials ensuring that one Sr\(^{2+}\) replaces two Na\(^+\) in the zeolite crystals. With these assumptions, the microstructure and the inhomogeneous thermodynamic and kinetic properties of the system as well as the ionic diffusion and microstructure evolution can be described by two sets of PF variables (see Fig. 2). One is the order parameter field \(\eta_i\), \((i = 1, 2, 3)\), which describe zeolite, SiO\(_2\), and solution phases, respectively. The order parameter field either represents the atomic structures of different phases or different crystal orientations of a phase \([23,29]\). Variable \(\eta_i\) \((i = 1, 2, 3)\) is, respectively, equal to 1 inside phase \(i\) and 0 outside phase \(i\), and varies smoothly from 1 to 0 across the interfaces between different phases or the grain boundaries between different zeolite crystalline particles. The other variable is the concentration field \(c_i\) \((i = 1, 2)\), which describe the spatial distributions of Na\(^+\) and Sr\(^{2+}\), respectively, within each phase.

The exchange of Sr\(^{2+}\) with Na\(^+\) or diffusion of Na\(^+\) and Sr\(^{2+}\) in the hierarchical structure is described by the following equations:

\[
\frac{\partial c_i(r,t)}{\partial t} = \nabla \cdot (D_i \nabla \left( \frac{\mu_i}{RT} \right)) \quad i = 1, \text{Na}^+; \quad i = 2, \text{Sr}^{2+}
\]  

where \(D_i\) is the diffusivity and \(\mu_i\) is the chemical potential of species \(i\). \(R\) and \(T\) are ideal gas constant and absolute temperature, respectively. \(\nabla = (\frac{\partial}{\partial x}, \frac{\partial}{\partial y}, \frac{\partial}{\partial z})\) and \(\mathbf{r} = (x, y, z)\) is the spatial coordinate and \(t\) is time. Under given boundary conditions and initial conditions, the spatial and temporal evolution of concentration \(c_i\) are obtained by solving Eq. (1).

In the model system, there are three phases: SiO\(_2\), zeolite, and...
liquid. The free energies of the different phases in terms of Na\(^+\) and Sr\(^{2+}\) concentrations and temperature can be assessed by thermodynamic calculations \[18,30\]. The chemical free energy of species \(i\) near equilibrium can be approximated by a parabolic function of Na\(^+\) and Sr\(^{2+}\) concentrations. Thus, the chemical potential of species \(i\) in the multiphase system can be described as:

\[
\mu_i = \sum_{\alpha=1}^{2} A_i(\alpha)(c_i - c_i^{eq}(\alpha))\eta_{i\alpha}, \ i = 1, 2
\]

(2)

where \(c_i^{eq}(\alpha)\) (\(i = 1, 2\)) are the equilibrium concentrations of Na\(^+\) and Sr\(^{2+}\) in phase \(\alpha\), respectively, \(A_i(\alpha)\) is the free energy coefficient of phase \(\alpha\), with \(\alpha = 1, 2, 3\), respectively, corresponding to zeolite, silica, and liquid phases. The equilibrium concentrations \(c_i^{eq}(\alpha)\) and coefficients \(A_i(\alpha)\) can be determined once the free energies of these three phases are known.

The diffusivities of Na\(^+\) and Sr\(^{2+}\) in different phases are treated as separate quantities. The diffusivity along interfaces and grain boundaries is usually much larger than in the bulk. Since Na\(^+\) and Sr\(^{2+}\) diffuse through porosities in the form of channels and only occupy certain porosities, the occupation concentration of Na\(^+\) and Sr\(^{2+}\) is usually much larger than in the bulk. Since Na\(^+\) separate quantities. The diffusivity may strongly depend on the concentrations of Na\(^+\) and Sr\(^{2+}\) inside the zeolite crystals changes the local chemistry, which may result in lattice distortion and possible phase transitions. As a result, the diffusivity can be determined once the concentrations of Na\(^+\) and Sr\(^{2+}\) are known.

Therefore, the diffusivity of Na\(^+\) and Sr\(^{2+}\) is spatial/phase and concentration dependent. In order to reflect the spatial/phase and concentration dependency, we have constructed the following expression for the diffusivity:

\[
D_i = D_{i0}\left(1 + d_i(1)\varphi(c_2\eta_1)\eta_1 + d_i(2)\eta_2 + d_i(3)\eta_3 + d_{gb}(1 - \sum_{i=1}^{3} \eta_i^2)\right)
\]

(3)

where \(d_i(\alpha), \alpha = 1, 2, 3\), and \(d_{gb}\) are the model parameters which can be determined by the bulk diffusivity and interface diffusivity, respectively. For instance, when \(\eta_1 = \eta_2 = 0\) and \(\eta_3 = 1\), \(D_i = D_{i0}[1 + d_i(2)]\) is the diffusivity of species \(i\) in SiO\(_2\). \(\varphi(c_2, \eta_1)\eta_1\) is not equal to zero inside zeolite particles. It describes the Sr\(^{2+}\) concentration dependence of diffusivity. In this work, \(\varphi\) is taken as a kind of Fermi-Dirac function so its value varies from 0 to 1, and has the form of

\[
\varphi(c_2, \eta_1) = \frac{1}{\exp\left(-k\left(\frac{c_2\eta_1 - c_2^{eq}(1)}{c_2^{eq}(2)}\right) - \lambda\right) + 1}
\]

(4)

where \(k\) and \(\lambda\) are coefficients to adjust the profile of \(\varphi\) as illustrated in Fig. 3. For a negative value of \(d_i(1)\) in the zeolite phase it can be seen from Eq. (3) that the diffusivity of species \(i\) decreases from \(D_0\) to \(D_0(1 + d_i(1))\) with the increase of the Sr\(^{2+}\) concentration. How strongly the diffusivity depends on Sr\(^{2+}\) concentration is determined by the two coefficients \(k\) and \(\lambda\) which could be fitted with DFT calculations or experimental data of the diffusivity.

3. Generation of hierarchical microstructures

The simulation requires initial microstructures that preferentially match the experimental materials in terms of particle size, distribution, and porosity. The PF method can directly read data obtained from experimental images such as those shown in Fig. 1(b). However, it is very challenging to obtain 3D images from experiments. In this work we use the PF method to generate model microstructures for use in the ion exchange model. The SiO\(_2\) skeleton can be considered as a type of 3D connected porous structure obtained by spinodal decomposition between PEO and TEO [31]. Therefore, a PF model of spinodal decomposition [32,33] is used to generate an SiO\(_2\) skeleton for the ion exchange model. Fig. 4 shows the microstructure evolution of one of the two phases during a typical PF model of spinodal decomposition. The obtained microstructures are very similar to the SiO\(_2\) structures shown in Fig. 1(a) and are therefore used to mimic an experimental SiO\(_2\) skeleton. Adjusting the two-phase volume fraction and interfacial energy in this PF model of spinodal decomposition creates structures with different macro-pore sizes and skeleton wall thicknesses.

Experimentally, a SiO\(_2\) microstructure is classically transformed into the structure shown in Fig. 1(b) using a pseudomorphic transformation by a dissolution-precipitation process that occurs when silica is mixed with a solution containing NaOH and an aluminum salt (such as NaAlO\(_2\)) [34]. Controlling the synthesis process parameters (aging time, crystallization temperature, etc.) leads to diverse Na-LTA morphologies [3]. With the obtained SiO\(_2\) skeleton microstructure in Fig. 4, a PF model of grain growth [29,35] is then employed to add an aggregation of zeolite particles into the skeleton microstructure. Controlling the nucleation densities, nuclei sizes, and growth rates, a 3D hierarchical structure with different macro- and meso-scale pores, particle sizes, and volume fraction of the zeolite particle phase can be obtained as shown in Fig. 5. Therefore, for given microstructural features, such as the macro- and mesoscale pore sizes, zeolite particle size distributions, and volume fractions of SiO\(_2\) and zeolite phases, this PF modeling capability allow us to generate 3D microstructures that reasonably capture experimental microstructures.

4. Results of ion exchange

The effects of Na-LTA microstructures on ionic exchange kinetics in batch tests where 100-mg Na-LTA monolithic particles are added to 100-ml Sr\(^{2+}\) solution were studied. The particle radius is about 200-\(\mu\)m. We consider the particles having the following microstructures, respectively: (1) large zeolite particles without any pores shown in Fig. 5(d); (2) SiO\(_2\) supported aggregation of dense micrometer-sized zeolite particles with macroscale pores but without mesoscale pores shown in Fig. 5(c); and (3) SiO\(_2\) supported aggregation of loose micrometer-sized zeolite particles with macroscale and mesoscale pores shown in Fig. 5(b). Each simulation only considers one such particle microstructure in the simulation domain, which has the dimension of 256\(\times\)256\(\times\)256\(\mu\)m. Periodic boundary conditions are applied in all directions. The fast Fourier spectral method for spatial discretization and a forward Euler scheme for time derivatives are employed to solve Eq. (1). Table 1 gives the initial Na\(^+\) concentration in the Na-LTA zeolite crystals and the initial Sr\(^{2+}\) concentration in the solution. The Na\(^+\) concentration in the zeolite material corresponds to the formula NaAlSiO\(_4\), which is a representative Na-LTA. The Sr\(^{2+}\) concentration in solution corresponds to the amount of Sr\(^{2+}\) needed for a complete exchange between Na\(^+\) from the solid and Sr\(^{2+}\) from the solution with a ratio of 2 to count for the charge difference. The Na\(^+\)
concentration in Table 1 refers to the zeolite solid phase. The considered zeolite has a cation Na\(^+\) exchange capacity of 5.6 mmol/g and a density of 0.76 g/cm\(^3\) [3]. It equals 4.256 mol/L using the same units as Sr\(^{2+}\) in the solution phase. Thus, \(c_0 = 4.256\) mol/L is used to normalize the concentration variables.

In the simulations, the zeolite particle occupies a spherical volume with a diameter of 200 grids in the simulation cell of 256 x 256 x 256 grids. Thus, the liquid phase volume in the simulation cell is much less than that in a real batch test. Considering the fact that ionic diffusion in the solution is much faster than that in the solid phases (SiO\(_2\) and zeolite) we assume that Sr\(^{2+}\) and Na\(^+\) are always uniformly distributed in solution. The uniform concentration in solution varies with ion exchange time and can be estimated as:

\[
c^{\text{BC}}_j(r^*)|_{\beta_3} = \frac{c^{\text{total}}(r^*)}{V^L_z}, \quad c^{\text{BC}}_j(r^*)|_{\beta_0} = c_0^j - \frac{c^{\text{total}}(r^*)}{V^L_z}
\]  

(5)

The superscript \(*\) indicate the quantity is normalized. The normalization is as \(r^* = \frac{r}{l^*}, t^* = \frac{t D_0}{\zeta_0}, c^{*}_j = \frac{c_j}{c_0}, D_0 = D_0/l^* A^*_0 = A_0/(RT), \) where \(l_0, D_0\) are the characteristic length and diffusivity, respectively. In Eq. (5), \(V^L_z\) is the normalized liquid volume equal to 100 mL in real units for the considered solution. \(\eta_3 > \eta_0\) with \(\eta_0 = 0.5\) represents the liquid solution phase, \(c^{\text{total}}_i\) is the total amount of Na\(^+\) entering into the liquid solution phase, and \(c^{\text{total}}_i\) is the total amount of Sr\(^{2+}\) entering into the zeolite particle phase, respectively. They can be calculated by

\[
c^{\text{total}}_i = c_0^i V^s - \int_{V^s} c^i(r^*, t) dV^s
\]  

(6)

\[
c^{\text{total}}_j = \int_{V^s} c^j(r^*, t) dV^s
\]  

(7)

where \(V^s\) is the normalized solid volume. Table 2 lists the dimensionless parameters used in the simulations with a time step of \(\Delta t^* = 1.0 \times 10^{-5}\).
4.1. Effect of particle size and concentration dependent diffusivity on uptake kinetics

Two particle sizes, 150 and 56 μm diameter, and two concentration dependent diffusivities, $d_i(x) = 0.0$ and $−0.999$, respectively, were studied to determine the effect of particle size and diffusivity on uptake kinetics. Seen from Eq. (3), $d_i(x) = 0.0$ means that diffusivity in zeolite is independent of concentration while $d_i(x) = −0.999$ means that the diffusivity becomes three orders of magnitude smaller when Sr$^{2+}$ concentration in zeolite reaches the equilibrium concentration $c_i^0(1)$. Fig. 6(a) plots the time evolution of Na$^+$ and Sr$^{2+}$ concentrations along a line through the center of a solid zeolite spherical particle as shown in Fig. 5(d). In the simulation, coefficients of $k = 20$ and $λ = 0.5$ were used. It shows that Na$^+$ diffuses out of the zeolite particle and reaches an equilibrium concentration of 0.0 and Sr$^{2+}$ diffuses into the zeolite particle and gradually reaches the dimensionless equilibrium concentration of 0.5. The reduction of Na$^+$ concentration causes a negative charge $−Q(1 − c_i^1)$ while the increase of Sr$^{2+}$ concentration causes a positive charge $2Qc_i^2$, where $Q$ is the charge per Na$^+$. Fig. 6(b) replots the evolution of scaled concentration $(1 − c_i^1)/2$, and $c_i^2$ inside the particle. We found that $(1 − c_i^1)/2$ is almost equal to $c_i^2$, which corresponds to local charge neutrality. Therefore, the results demonstrate that the chemical potentials and mobility used in the simulations ensure local charge neutrality even though electric field equilibrium is not explicitly considered in the current model. However, a quantitative and predictive model needs to take the electric field into account to capture the effect of local charge on ionic diffusion and ion exchange kinetics, especially in hierarchical nuclear wasteform materials of interest. It is straightforward to consider charge effects as did in a previous electrochemistry model of charge and discharge in Li ionic battery materials [37] and we intend to do so in the future.

Fig. 7 shows the evolution of the average concentration of Sr$^{2+}$ inside the particles versus the normalized time $(t^{1/2})$ for different particle sizes and different value of $d_i(x)$. The average concentration is defined as the ratio of the total amount of Sr$^{2+}$ in the particle and the total ideal capacity of Sr$^{2+}$, which is equal to the capacity multiplied by the particle volume. The average concentration is proportional to the total amount of Sr$^{2+}$ because the total ideal capacity is constant for given particle size. The solid lines present the uptake kinetics for the particles with concentration independent diffusivity while the solid symbol lines are for the particles with concentration dependent diffusivity. As expected, for both cases, the larger the particle size, the slower the uptake kinetics (i.e. how fast the system reaches its ideal capacity) because the uptake kinetics are controlled by long-range bulk diffusion. It can be clearly seen that at earlier stages the average concentration and the time $(t^{1/2})$ has the same linear relationship identified by the green lines for a given particle size. Such a linear relationship can only be explained by bulk diffusion control. With increasing Sr$^{2+}$ concentration in the surface layer of the particle, the diffusivity in the surface layer decreases. From Fig. 7 it is seen that a second linear kinetic process appears in the particles with concentration dependent diffusivity. The second linear process is identified by the blue lines. More importantly, the uptake kinetics may be significantly reduced if Sr$^{2+}$ accumulation strongly reduces the diffusivity. The results show that decreased uptake kinetics reduces the effective capacity of Sr$^{2+}$ for a given time. In contrast, there is no second linear kinetic phenomena for the particles with concentration independent diffusion. Many ion exchange experiments [26,38,39] show two diffusion kinetic regimes in agreement with our simulation results. The experimental and simulation results demonstrate that concentration dependent diffusivity plays an important role in Sr$^{2+}$ uptake, and the developed model is able to reasonably capture these effects.

4.2. Effect of microstructure on uptake kinetics

In a microstructure with dense zeolite particles, the particles are in close contact with each other, resulting in no mesoscale pores and no liquid penetration into the aggregation of zeolite particles. On the other hand, in a microstructure consisting of a loose aggregation of zeolite particles, there are mesoscale pores that allow the solution to penetrate into the structure. The interconnecting mesoscale pores filled with liquid are fast channels for Na$^+$ and Sr$^{2+}$ diffusion. Figs. 8 and 9(a)
summarize the effect of microstructures with and without mesoscale pores on Sr\(^{2+}\) uptake kinetics. The average concentration is calculated in the same way as in Fig. 7, i.e., it is the ratio of the total amount of Sr\(^{2+}\) into the aggregated particle and the total ideal particle capacity of Sr\(^{2+}\), which has a maximum of 1.0. In the simulations, coefficients of \(k = 10.0, \lambda = 0.7\) and \(d_{i,1} = -0.999\) were used.

From the results in Figs. 8 and 9(a), several uptake kinetic trends are observed including: (1) during the first linear kinetic process kinetics decrease with increasing zeolite thickness; (2) the thicker the zeolite particle layer, the slower the overall uptake kinetics for both dense and loose zeolite particles; (3) a second linear kinetic process in the microstructure with dense particles has more impact than in the microstructure with loose particles; and (4) the second linear kinetic process becomes less important with decreasing particle size and/or zeolite layer thickness. These results indicate that uptake kinetics strongly depend on the zeolite microstructure.

For comparison, some experimental data from porous materials containing HCF are used. For example, mesoporous silica support loaded with HCF shows very fast kinetics where equilibrium was reached in a few minutes and was attributed to the porosity of the solid support [40]. However, a dense support loaded with the same kind of HCF requires a few days to reach equilibrium [38]. The experimental results show that uptake kinetics strongly depend on microstructure. It can be seen in Fig. 9(a) that in the structure with only one large zeolite particle and the structure with dense small zeolite particles without mesoscale pores the uptake kinetics are controlled by both bulk diffusion and surface layer diffusion. However, the overall uptake kinetics are only controlled by bulk diffusion in the structure with loose small zeolite particles and meso-pores. The predicted microstructure dependence of ion exchange kinetics are consistent with experiments shown in Fig. 9b. We have designed LTA batch tests for more accurate comparisons. The three circles mark the average concentrations of Sr\(^{2+}\) corresponding to about 11\%, 57\% and 94\% of the ideal capacity at the time \((t^*)^{1/2} = \left(\frac{l}{L}\right)^{1/2} = 0.15\) for three different microstructures. The large difference clearly indicates that the effective capacity strongly depends on the microstructure.

5. Summary

A 3D PF microstructure-dependent diffusion model has been developed to study the effects of complex microstructures, inhomogeneous thermodynamics, and diffusion kinetics on radionuclide uptake kinetics and on the capacity of radionuclides and uptake efficiency, as well as highlighting known diffusion mechanisms. PF models of spinodal decomposition and grain growth were employed to generate hierarchical microstructures generally corresponding to experimental materials for the model microstructures in the simulations. The developed PF model demonstrated that uptake kinetics in large particles and aggregation of dense particles are limited by two diffusion processes: bulk diffusion and surface layer transport. The uptake kinetics change from two diffusion processes to a single process as the particle size decreases and the mesoscale pore volume fraction increases. The predicted uptake kinetics and the effect of microstructures on the effective capacity of radionuclides and uptake efficiency agreed reasonably well with published ion exchange experiments in Na-LTA zeolite materials. The results demonstrate that the developed model has the capability to investigate effects of microstructure on uptake kinetics in complex, hierarchically-porous microstructures. However, because thermodynamic and kinetic properties of zeolite structures are lacking, qualitative thermodynamic and kinetic models are used in the simulations, which means that the results are qualitative but can be made quantitative by acquiring the required data, either by experiment or by computation. The chemical potential gradient for the species in the solution phase and the zeolite phase are the driving force for diffusion. A concentration dependent diffusivity strongly affects ion exchange kinetics. It is desired to obtain these thermodynamic and kinetic properties for more quantitative simulations. In addition, some physics, such as electric fields associated with local aggregation of ions and fluid flow in multiscale pores, may play important roles in uptake processes and will eventually be integrated into the model in the future.
CRediT authorship contribution statement

Y.L. Li: Methodology, Investigation, Visualization, Writing - original draft. B.D. Zeidman: Data curation, Investigation. S.Y. Hu: Conceptualization, Supervision, Writing - original draft, Writing - review & editing. C.H. Henager: Conceptualization, Funding acquisition, Project administration, Writing - review & editing. T.M. Besmann: Conceptualization, Writing - review & editing. A. Grandjean: Data curation, Writing - review & editing.

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