

## Experimental and Thermodynamic Modeling Study of Multicomponent Ion-Exchange of Alkali and Alkaline-Earth Metals in Clinoptilolite

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Ion-exchange processes in natural systems generally involve more than two cations, but little published data are available for multicomponent ion-exchange reactions. Experimental data on ternary and more complex mixtures are needed to develop and evaluate thermodynamic models that can be used to predict ion-exchange equilibria in natural systems. In this study, ternary and quaternary ion-exchange experiments were conducted involving the alkali and alkaline-earth cations  $\text{Na}^+$ ,  $\text{K}^+$ ,  $\text{Cs}^+$ ,  $\text{Sr}^{2+}$ , and  $\text{Ca}^{2+}$  and clinoptilolite, a zeolite mineral that is locally abundant in the saturated and unsaturated zones of Yucca Mountain, Nevada, which is the potential site for a high-level waste repository. The clinoptilolite used in the experiments was prepared by pulverizing a sample of clinoptilolite-rich tuff from Death Valley Junction, California, and purifying the 200–325 mesh fraction. Homoionic forms of  $\text{Na}^+$ ,  $\text{K}^+$ , and  $\text{Ca}^{2+}$ -clinoptilolite were prepared by reacting at 75 °C [167 °F] the purified zeolite with 3 M chloride solutions of  $\text{Na}^+$ ,  $\text{K}^+$ , or  $\text{Ca}^{2+}$ . The ternary ion-exchange experiments consisted of reacting weighed amounts of homoionic clinoptilolite powder with known volumes of solution mixtures of  $\text{Na}^+\text{K}^+\text{Cs}^+$ ,  $\text{Na}^+\text{K}^+\text{Sr}^{2+}$ , and  $\text{Na}^+\text{K}^+\text{Ca}^{2+}$  at constant solution normality. The quaternary ion-exchange experiment studied a  $\text{Na}^+\text{K}^+\text{Ca}^{2+}\text{Sr}^{2+}$  system.

A thermodynamic model based on the Wilson equation was used to model the ion-exchange data. The parameters for the Wilson equation and the equilibrium constants for the binary ion-exchange reactions were derived from published binary ion-exchange data. A correlation method that has been applied to predictions of formation constants of aqueous hydroxo-metal complexes was used to help constrain the equilibrium constants derived from the binary isotherm data. The Wilson model, with parameters derived only from binary ion-exchange data, was used to predict ternary and quaternary ion-exchange equilibria. A comparison of experimental data for ternary and quaternary systems and thermodynamic model predictions indicates that the Wilson model adequately reproduces multicomponent ion-exchange equilibria. The results suggest that it is possible to use the Wilson model to evaluate multicomponent ion-exchange involving alkali and alkaline-earth metals in natural systems.

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