

# Experimental calibration of kinetic isotope fractionation of alkaline and alkaline-earth elements during chemical diffusion in aqueous solutions

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Diffusion is a basic process that could cause isotopic variability in nature, diffusion-driven isotope fractionation of cations have been reported in literature but there are still some unclear aspects of this phenomenon. For example, chemical diffusion of  $\text{Mg}^{2+}$  aquo ions in aqueous solution had long been perceived as a process with no isotope fractionation, despite that all other alkaline-earth elements (i.e., Ca, Ba) exhibit significant isotope fractionation during diffusion. We developed a novel “diffusion cell” method that enables efficient and precise determination of kinetic isotope fractionation of ions during chemical diffusion in aqueous solutions, and used this method to investigate the isotopic effect of diffusion of aqueous Mg under various conditions. The experimental results show that in aqueous solutions, light Mg isotopes diffuse faster than heavy Mg isotopes, with  $D_{26\text{Mg}}/D_{24\text{Mg}}$  (ratio of diffusion coefficients between  $^{26}\text{Mg}$  and  $^{24}\text{Mg}$ ) ranging from 0.99990 to 0.99982, depending on experimental conditions. Notably, the magnitude of diffusion-induced Mg isotope fractionation in aqueous solution is higher at higher temperature, and is greater for  $\text{MgCl}_2$  and  $\text{Mg}(\text{NO}_3)_2$  than for  $\text{MgSO}_4$  solutions. We also used the “diffusion cell” method and determined the diffusion-driven kinetic isotope fractionation factors of the cations of Li, K, Rb, Ca, Sr, and Ba in aqueous solutions under room temperatures. Based on a synthesis of these data, we argue that the mass dependence of isotope fractionation associated with chemical diffusion in aqueous solution is fundamentally controlled by the intensity of ion-water interaction, with stronger ion-water interaction leading to more prominent hydrodynamic behavior for diffusing ion at molecular level, and less significant isotope fractionation in bulk solutions.