Kinetic Si isotope fractionation factors in chert formation processes

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The variations of $\delta^{30}Si$ values in diagenetic chert and chert- associated BIFs over time can be used to reconstruct the environmental conditions of the early Earth, and become a hot topic in the Si isotope society. However, there are several different views of explaining the variation of $\delta^{30}Si$ values over time. Moreover, there are disputes in explaining the distribution of Si isotope in several main reservoirs in surface systems. Those disagreements are caused by lacking key Si isotope kinetic fractionation factors associated with the formation processes of chert and its altered products.

There are many unexplained observations about Si isotope distributions in Earth's surface systems (i.e., in soil, water or plants) (Opfergelt and Delmelle, 2012). For example, the deduced Si isotope equilibrium fractionation factors by Rayleigh model at ambient temperature between clay and the solution $\Delta^{30}Si_{clay-solution}$ = -1.5 ‰ (Georg et al., 2007) and -2.05‰ (Hughes et al., 2013) obviously disagree with common sense, which dictates that stiffer chemical bonds will enrich heavier isotopes, i.e., the precipitated minerals will preferentially incorporate heavy isotopes relative to aqueous H_4SiO_4 due to their shorter Si-O bonds. Another similar case is the fractionation between quartz and solution. Most field observations suggested that solution will be enriched with heavier Si isotope compared to quartz (e.g., Douthitt, 1982; Basile-Doelsch et al., 2005), conflicting to the fact that quartz is the one with much shorter Si-O bonds than aqueous H_4SiO_4 (ca. 1.610Å vs. 1.639Å). Those strange Si isotope behaviors can be explained by knowing basic Si isotope fractionation parameters related to kinetic processes of Si cycling in surface systems. We provide the most important Si kinetic isotope fractionation (KIE) factors associated with the formation of amorphous quartz and other secondary minerals in polymerization, co-precipitation and adsorption processes. Without the knowledge of KIE of these coupled or competing pathways, we cannot quantitatively evaluate the net Si isotope fractionation during the chert formation processes and cannot figure out the reason of environment change.