Crystal of Li-Fe: The Impact of Crystallochemistry on Lithium Isotope Signatures during Interaction with Fe Oxides

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Lithium isotopes ($\delta^7 \text{Li}$) have proven to be robust tracers of clay formations during water-rock interactions. Although coupling between Li and Fe oxides and the associated Li isotope fractionation have long been speculated upon, only a handful of experimental studies have investigated this topic. As a result, the effect of Fe oxides on the Li isotope system remains undetermined, obscuring the understanding of geological profiles enriched in Fe oxides, such as laterite.

We conducted experiments to explore (1) the sorption of Li by Fe oxides, and (2) the co-precipitation of Li with Fe oxides. In addition to isotope analysis, we applied advanced imaging techniques such as SEM and TEM to better understand the processes affecting the geochemical behaviour of Li.

Contrary to previous suggestions, Fe oxides appear to have a smaller-than-assumed impact on the low-temperature Li isotope system. Imaging results show that crystallochemistry plays an important yet often overlooked role in water-rock interactions. Poorly crystallized goethite undergoes possible dissolutionreprecipitation reactions at defect sites, which take up dissolved Li and result in Li isotope fractionation of ~20%, particularly under alkaline conditions (pH ~12). Compared to clay minerals, only a small amount of Li is taken up during goethite precipitation at room temperature, with an associated Li isotope fractionation of ~20%. At elevated temperatures, Li uptake becomes significant during the formation of well-crystallized goethite (70°C), but Li isotope fractionation is weakened (\sim 7%). The formation of hematite at high temperatures (98°C) consumes a small amount of Li, with an associated Li isotope fractionation of ~7%. Coupled with imaging results, the uptake of Li and its isotope fractionation appear to be associated with nucleationcrystallization processes. Little Li isotope fractionation is observed during the early stage of precipitation just after nucleation, whereas significant fractionation occurs during recrystallization, as indicated by the presence of lattice structures observed under TEM.