New insights into mantle and crustal processes from high-temperature magnesium isotope fractionation

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Equilibrium isotope fractionation generally decreases with increasing temperature and is expected to be insignificant at high temperatures. By contrast, kinetic isotope fractionation associated with chemical diffusion and Soret effect can be extremely large at high temperatures as shown by recent experiments. However, the extent to which kinetic isotope fractionation may occur in nature remains uncertain.

Here we report large high-temperature kinetic Mg isotope fractionation in zoned olivines and equilibrium Mg isotope fractionation among mantle and crustal minerals. Significant Mg isotope fractionation (>0.4 ‰ variation in $^{26}Mg/^{24}Mg$) was found in over 60 olivine grains from Hawaii, which reflects isotope fractionation associated with inter-diffusion of Mg and Fe during magmatic differentiation. Large Mg isotope fractionation, up to 1.1 ‰ in $^{26}Mg/^{24}Mg$, was found between coexisting garnet and omphacite in 5 orogenic eclogites, and between coexisting spinel, cpx, opx and olivine in 13 mantle peridotites. The large inter-mineral isotope fractionation reflects equilibrium partitioning of Mg isotopes between coexisting minerals as evidenced by the temperature-dependent isotope fractionation, lack of mineral zoning, and absence of intra-mineral isotopic variations, as well as equilibrium oxygen isotope fractionation among these minerals. The direction of inter-mineral equilibrium Mg isotope fractionation among these mantle and crustal minerals agrees with theoretical predictions, suggesting that inter-mineral Mg isotope fractionation is primarily controlled by the Mg-O bond strength, with stronger bonds favoring heavier Mg isotopes.

Our results demonstrate that both equilibrium and kinetic isotope fractionation can occur during high-temperature geological processes, with great potential applications. Kinetic isotope fractionation associated with chemical diffusion can be used to identify diffusive transport in zoned minerals, which is key for using chemical zoning to constrain magmatic timescales. Large equilibrium Mg isotope fractionation among mantle and crustal minerals can potentially be used as a high-precision geothermometry in mantle and crustal geochemistry.

Solvent effect on the precipitation of Mg-carbonate

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Low temperature synthesis of anhydrous magnesite continues to be an outstanding geochemical problem despite our improved understanding in the Mg-CO_3 system (e.g. formation of various hydrated forms and basic hydromagnesites). Presently, the prevalent hypothesis for the lack of magnesite crystallization under ambient conditions seems to focus on the strongly hydrated Mg^{2+} ions (compared to Ca^{2+}). It is hypothesized that the robust hydration shell around Mg^{2+} lowers the cation's activity and provides a barrier for the access of CO_3^{2-}. We intend to explore the possibility of magnesite crystallization in the presence of alcohol which could potentially replace H_2O or dislodge the water cage around Mg^{2+}. We hypothesize that (1) the presence of alcohol in water will distort or weaken the rigid water shell around magnesium ions due to the change in solvent dielectric properties and the water-alcohol interactions to facilitate the interaction of Mg^{2+} and CO_3^{2-}; and (2) dry conditions in pure alcohol may lead to the formation of anhydrous magnesite. We found that $MgCl_2$ and guanidine carbonate (GC) can be used as reactants since both show workable solubility in methanol (me).

Four batches of reactions have been carried out between $MgCl_2$ and GC in water and methanol (Mg-aq + GC-aq; Mg-me +GC-aq; Mg-aq + QC-me; Mg-me + QC-me). The presence of methanol in water greatly accelerates the precipitation, but none of four batches led to the formation of magnesite. FT-IR and XPS revealed the presence of CO_3 in the products, but XRD analysis identified either amorphous phases or could not be matched with the diffraction patterns of any known Mg-CO_3 salts in the database. The sample synthesized in the presence of water were found to be amorphous nanoparticle aggregates, while that formed in pure methanol exhibits a well-crystallized thin platelet assembly morphology. It appears that methanol is able to facilitate Mg-CO_3 crystallization, but the presence of minute amount of H_2O in solvent or in air still found a way to be incorporated in the crystal structure, confirming the high affinity of Mg^{2+} toward water molecules.