Lithium incorporation in olivine

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The relative concentrations of the two stable isotopes of lithium are increasingly being used to investigate chemical homogeneity and evolution of the upper mantle. The principal host for lithium in the mantle is olivine, but how univalent species, such as Li and Na, are incorporated into the crystal structure remains poorly understood. Here, we describe the results of isobaric crystal-melt partitioning experiments which, together with thermodynamic modelling, provide insight into the mechanisms of incorporation of Li in forsterite and natural olivine of mantle composition.

The results of partitioning experiments conducted on end-member forsterite doped with varying concentrations of lithium and trivalent cations show that the dominant substitution mechanism depends strongly on the concentrations of dopant elements. At relatively high concentrations (>20 ppm Li in the crystal), lithium is principally incorporated into near end member forsterite via coupled substitution with trivalent cations such as Al and Fe\(^{3+}\). At <20 ppm Li in the crystal, however, there is no correlation between Li partitioning and trivalent element concentration and results indicate charge-balance via vacancy or interstitial defect formation.

The lithium contents of natural olivine from the spinel stability field were measured using LA-ICPMS. The amount of Li in olivine is low (<5 ppm) and, unlike Na, does not correlate with the concentration of any trivalent cation species. We therefore conducted a series of additional experiments in order to investigate Li incorporation in natural, Fe-bearing olivine. In these experiments, crystals were grown at 1 atm from a natural basanite melt, at a range of oxygen fugacities. The results demonstrate that the partitioning of lithium between iron-bearing olivine (approximately Fo89) and melt is strongly dependent on oxygen fugacity (\(f_\text{O}_2\)). The concentrations of lithium in mineral-melt pairs from our experiments show that \(D_{\text{Li}}\)\(_{\text{olivine-melt}}\) decreases from 0.3 at IW to 0.15 at NNO. This negative dependence on \(f_\text{O}_2\) suggests that lithium is incorporated into Fe-bearing olivine either through formation of oxygen vacancies or coupled to tetrahedral Fe\(^{3+}\). The consequences are that partitioning and diffusion of Li in natural materials is strongly dependent on the activities of major components in the medium.