Empirical constraints on the mass dependence of isotope diffusion in minerals by modeling sub-solidus exchange: Calcium isotopes in the two-pyroxene system

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Diffusion is a significant driver of isotope fractionation at high temperatures [1-3], but its precise role is difficult to evaluate in many situations due to the sparsity of data on the isotopic mass dependence of the diffusion coefficient. The ratio of diffusion coefficients of two isotopes of the same element in the same material can be characterized as the inverse mass ratio raised to an empirical exponent β [4], but experiments to determine β are challenging and few measurements have been reported, especially for minerals. Here, a method is developed to empirically determine β based on numerical modelling of the diffusion-controlled elemental and isotopic redistribution between minerals during slow subsolidus cooling, and comparison of the results to data from natural mineral pairs. The method is applied to the Ca redistribution between orthopyroxene (opx) and clinopyroxene (cpx), where the elemental partitioning equilibrium as a function of temperature has been well characterized experimentally and Ca isotope fractionation data are available in natural samples that experienced slow cooling. We show that the Ca isotope fractionation during cooling is insensitive to the initial temperature, cooling rate or grain size over a broad range of conditions, provided that diffusion is sufficient to re-homogenize opx grains at near-peak temperatures during early cooling. Furthermore, we find that the Ca isotope fractionation established during cooling is much more strongly dependent on the value of β in opx than the value of β in cpx, as long as cpx dominates the Ca budget in the system. Based on the largest Ca isotope fractionation that has been documented between opx and cpx in natural samples, it is inferred that for Ca, β_{opx} = 0.04±0.01. Similar behavior during sub-solidus cooling is anticipated for other elements with temperature-dependent inter-mineral partition coefficients, and the method developed here has the potential to be used to place constraints on β for many elements, even in minerals where experimental measurements are not feasible.

References
[1] Richter et al., (2008) GCA
[2] Richter et al., (2014) GCA
[3] Van Orman and Krawczynski (2015) GCA
[4] Richter et al., (1999) GCA