

A theoretical and experimental study on kinetic isotope fractionation during evaporation

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Enrichment of heavy isotopes of Mg and Si observed in some igneous CAIs has been attributed to kinetically controlled evaporation during melting. Based on the conventional theory for the evaporation rate (Hertz-Knudsen equation [e.g., 1]), the isotopic fractionation factor is predicted to be a square root of the inverse mass ratio of isotopes, but the experimentally determined values for evaporation of silicate melts and silicates were generally closer to unity than the prediction with unexpected temperature dependence [2, 3].

In the present study, we discuss a theory of evaporation kinetics in nonequilibrium based on the transition state theory [4] and examine the kinetic isotopic fractionation during evaporation. We present two models capable of explaining the existing experimental results [2]: (i) Equilibrium isotopic fractionation between the condensed phase and the transition state which arises from the quantum mechanical effects [5] and (ii) Isotope effects of classical mechanical vibrational motions of the activated complex and their anharmonicity. Because two models predict different temperature dependence of the isotopic fractionation factor at low temperatures, the models can be tested by evaporation experiments at lower temperatures than previous studies [1, 2]. We carried out evaporation experiments of a single crystal of forsterite in ~1 Pa of hydrogen gas at 1600 and 1350 K, and measured Mg-isotope depth profiles of the experimental samples using secondary ion mass spectrometry (SIMS) to obtain the kinetic isotope fractionation factors [2]. The preliminary results suggest a smaller degree of fractionation of Mg isotopes at 1350 K than at 1600 K and higher temperatures, which supports the model (i), although further experiments are necessary for more robust discussions.

[1] Persad and Ward (2016) *Chem. Rev.* 116, 7727. [2] Wang et al. (1999) *GCA* 63, 953. [3] Richter et al. (2007) *GCA* 71, 5544. [4] Mortensen and Eyring (1960) *J. Phys. Chem.* 64, 846. [5] Bigeleisen and Mayer (1947) *J. Chem. Phys.* 15, 261.