

Kinetic isotope effects in high-temperature evaporation in protoplanetary disks

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Interstellar dust grains are building blocks of planetary materials but are likely to be thermally processed in protoplanetary disks before they accrete to form planets. The most intensive processing in inner hot regions of disks results in vaporization of dust grains, and their chemical compositions are significantly altered when they are partially vaporized. The metal-bearing molecules (e.g., SiO, AlO and NaCl) observed in a high mass young protostar candidate (Orion Source I) are likely to be the molecules evaporated from dust [1-3].

Evaporation is a kinetic process that governs the timescales of such solid-vapor partitioning and thus is important in understanding chemical evolution of planetary materials. Kinetic isotope effects (KIEs) associated with evaporation lead to isotopic fractionation of the residual phase, providing information about evaporation experienced by extraterrestrial samples. For example, calcium-, aluminum-rich inclusions (CAIs) with igneous textures found in chondrites show enrichment of heavy isotopes of Mg and Si, which is attributed to evaporative isotopic fractionation in the protosolar disk [4]. The KIE in evaporation can be theoretically predicted by the ratio of the thermal velocity of the evaporating molecules, i.e., the square root of their inverse mass ratio. However, experimentally determined KIEs are usually smaller than the theoretical prediction [5,6].

In this study, we newly formulated the KIE in evaporation by using the expression of the evaporation rate based on conventional transition state theory (CTST) [7,8]. We found that the KIE in evaporation at the high-temperature limit is expressed by the square root of the inverse ratio of the reduced mass for the motion along the reaction coordinate at the transition state. This result is consistent with the theory previously proposed for KIEs in general chemical reactions [9] and explains the experimental KIEs in evaporation smaller than the conventional prediction. By considering the motion along the reaction coordinate as vibration with an imaginary frequency, we further evaluated the KIE in evaporation using normal-mode vibration analyses for the transition state. The result suggested that the KIE is smaller than the conventional prediction when the activation barrier exists for the bond dissociation. This suggests that the different degrees of KIEs of metallic iron and silicates obtained by experiments might indicate the presence of an activation barrier for the dissociation of an ionic bond that originates from the avoided crossing of potential energy curves.

References

- [1] T. Hirota et al., 2017, *Nature Astronomy* 1, 0146
- [2] S. Tachibana et al., 2019, *ApJ* 875, L29
- [3] A. Ginsburg et al., 2019 *ApJ* 872 54.
- [4] L. Grossman et al., 2008, *GCA* 72, 3001.
- [5] M. Yamada, S. Tachibana, H. Nagahara, & K. Ozawa, 2006, *Planet. Space Sci.* 54, 1096.
- [6] F. M. Richter, P. E. Janney, R. A. Mendybaev, A. M. Davis, & M. Wadhwa, 2007, *GCA* 71, 5544
- [7] E. Mortensen & H. Eyring, 1960, *J. Phys. Chem.* 64, 846.
- [8] S. Inada, T. Hama, & S. Tachibana, *J. Chem. Phys.*, in revision.
- [9] J. Bigeleisen, 1949, *J. Chem. Phys.* 17, 675.