A theoretical study of diffusional isotope effect in minerals

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Diffusion is a universal phenomenon in minerals. Light and heavy isotopes have different diffusion rates, which will lead to the uneven isotope distribution along a profile in minerals. It is so-called the diffusion isotope effect (DIE) of solid. With the development of high space resolution mass spectrometer (HRMS), the study of isotope fractiontion in minerals becomes a research hotspot recently. Although isotope data in minerals are rapidly accumulating, people still feel difficult to figure out: how long the heating history is? what's the diffusion speed difference for different isotope systems? Therefore, the theoretical basis and basic parameters in these processes are urgently needed now. In this study, on the one hand, we will advance the theory of isotope diffusion in minerals[1,2], including vacancy and interstitial diffusion mechanisms. On the other hand, we can demonstrate that the use of quantum chemistry methods is a good way to explain the DIE in solids.

Classical theory[1,2] suggested that the DIE is affected by two factors, one is the correlation coefficient (*f*) which denotes the degree of deviation from the pure random steps, and another is the coupling constant (K) which denotes the degree of coupling of jump atom and remaining atoms. Although *f* and K have explicit physical significance, it is difficult to quantify them in the quantum chemistry calculation. We will show that 1)The approximation, which is used by previous researchers for dealing with the diffusion passing through a energy barrier (i.e., the TST model), is improper due to the ignoring of kinetic energy differences of different isotopes. Considering the kinetic energy differences at transition state will provide another term in the final DIE formalism besides the *f* and K terms; 2) Quantum chemistry calculation using first-principles molecular dynamics methods can readily provide a lot of DIE coefficients for minerals at high temperature and high pressure.

[1] Schoen. (1958) Physical Review Letters 1, 138-140. [2] Tharmalingam and Lidiard. (1959) Philosophical Magazine 4, 899-906.