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First-principle calculations of equilibrium Barium isotope fractionation between silicate minerals and its implications

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Barium (Ba) isotopes could be useful to trace geochemical processes, such as crustal recycling into the mantle, evolution of the continental crust, and the paleo-oceanic primary productivity. A recent study observed large Ba isotope fractionation during the evolution of granitic magma [1], but the fractionation mechanism is not well known due to the lack of the equilibrium Ba isotopes fractionation factors ($\Delta^{138/134}$Ba). Here, we conducted first-principles calculations based on density functional theory to estimate the $\Delta^{138/134}$Ba between Ba-bearing minerals, including phlogopite, muscovite, microcline, albite, tremolite, richterite, epidote, witherite, and barite. Our results show that the enrichment of heavy Ba isotopes follows the order of phlogopite (Mg substituted) > tremolite > epidote > albite > witherite ~ richterite ~ microcline ~ barite ~ muscovite > phlogopite (K substituted). Within the explored compositional space that is comparable to or higher than Ba concentrations in naturally occurring silicate minerals, there is no significant Ba concentration effect on the reduced partition function ratios of Ba isotopes ($10^3\ln\beta$). The $10^3\ln\beta$ are proportional to the force constants of Ba, which are mainly affected by both bond lengths and coordination numbers. We also find that the Ba-doped structures in which Ba occupies K sites are enriched in light Ba isotopes than other configurations, suggesting that the substitution mechanism for Ba will significantly affect the equilibrium Ba isotope fractionation.

This work suggests that the equilibrium Ba isotope fractionation between minerals is limited at high temperature, e.g., < 0.1‰ at 1000 K. Such a magnitude of fractionation is much smaller than the variation of Ba isotope composition observed in Huili granitic pluton [eg., 1], suggesting that the Ba isotope fractionation associated with the evolution of granitic magma is likely caused by disequilibrium processes. Combining previously published studies on fluid systems [2], we also estimate the $\Delta^{138/134}Ba_{\text{fluid-silicate minerals}}$, which provides a guideline for our understanding of Ba isotope behaviors during fluid-involved processes.

Reference:

[1] Deng, G., et al., (2021), Geochimica et Cosmochimica Acta, **292**: p. 115-129.

[2] Wang, W., Z. Wu, and F. Huang (2021), Geochimica et Cosmochimica Acta, **292**: p. 64-77.