Origin of oldhamite in enstatites: evidence from Ca isotope

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Oldhamite (CaS) is a sulfide present in enstatite meteorites: chondrites and achondrites. It has attracted much attention for its enrichment in rare-earth element (REE) and potentially control the REE budget of these meteorites. However, the origin oldhamite, formed by condensation or igneous crystallization, is still unclear but important to understand the evolution history of the early Solar System. In this study, we report Ca isotopic compositions ($\delta^{44/40}\text{Ca}$) of oldhamite (both obtained from water leachate from chondrites and aubrites and mineral separates from the Norton County aubrite) and silicate minerals from different types of enstatite chondrites and aubrite. The enstatite chondrites contain different metamorphism grade, from EH3 to EH6, and three EL6 samples. The aubrite sample, Norton County, is a typical igneous achondrite and two individual oldhamite grain could be separated.

The $\delta^{44/40}\text{Ca}$ of the bulk enstatite chondrites range from 1.06‰ to 1.38‰, with an average of 1.17 ± 0.20‰, higher than that of bulk silicate earth (~0.94‰). Major and trace element analyses show that the leachates of enstatite chondrites are nearly pure oldhamite, and they take over 23-65% Ca of the bulk meteorite Ca budget. The Ca isotope fractionation between oldhamite and silicate ($\Delta^{44/40}\text{Ca}_{\text{oldhamite-silicate}}$) varies and correlate with the metamorphism grade from -0.50‰ in EH6 to +0.20‰ in EH3. The Ca isotope fractionation between the two individual oldhamite grains and silicates in Norton County are -0.55 ± 0.09‰ and -0.58 ± 0.08‰. These $\Delta^{44/40}\text{Ca}_{\text{oldhamite-silicate}}$ correlate well with previous theoretical calculation and suggests that the oldhamites in Norton County are in isotopic equilibrium with co-existing silicates, and therefore were formed during magmatic processes. However, in enstatite chondrites, the large variation on $\Delta^{44/40}\text{Ca}_{\text{oldhamite-silicate}}$ and its opposite trend with metamorphism grade reflects the redistribution and equilibration of Ca isotopes during metamorphism. The variable $\Delta^{44/40}\text{Ca}_{\text{oldhamite-silicate}}$ found in unequilibrated chondrites reflect kinetic Ca isotope fractionation between oldhamite and nebular gas and therefore is evidence for the formation of oldhamite by condensation in the solar nebula.