

Thermal history of coarse-grained CAIs in the protosolar disk constrained by O-isotope exchange kinetics between CAI melt and water vapor

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Calcium-aluminum-rich refractory inclusions (CAIs) are the sub-millimeter to centimeter-sized oldest objects, consisting of high temperature mineral assemblages, in chondrites (e.g., MacPherson, 2003; Connelly et al., 2012). CAIs would preserve information on high temperature events in the earliest Solar System. Coarse-grained CAIs are considered to experience multiple heating stages after the condensation of their precursors, and exhibit mass-independent O-isotopic variation among their constituent minerals such as spinel, melilite, anorthite and fassaite (e.g., Yurimoto et al., 1998; Kawasaki et al., 2018). An explanation of the O-isotopic variations in coarse-grained CAIs is the partial melting and associated O-isotope exchange between CAI melt and protosolar disk gas (e.g., Yurimoto et al., 1998; Kawasaki et al., 2018). Melilite is the first phase crystallizing from spinel-bearing typical type B CAI melt (Stolper, 1982; Stolper & Paque, 1986) and has typically ¹⁶O-poor isotope composition with a limited isotopic variation (e.g., Yurimoto et al., 2008; Kawasaki et al., 2018). If the precursors of CAIs had ¹⁶O-rich isotopic compositions close to that of spinel, those observations indicate that melilite crystallized from the melt that was isotopically equilibrated with ¹⁶O-poor disk gas above the melilite liquidus. However, because of the lack of quantitative data on O-isotope exchange kinetics between CAI melt and disk gas, the thermal history of CAIs cannot be constrained from O-isotope compositions of constituent minerals. In this study, we conducted O-isotope exchange experiments between type B CAI analogue melt and water vapor.

O-isotope exchange experiments between ~2.5 mm-sized synthetic spherical CAI droplet with the average type B composition (Wark et al., 1979; CAIB hereafter) and ¹⁸O-enriched water vapor (~97% ¹⁸O) were carried out at 1390°C (10°C above the melilite liquidus) and a disk-like low water vapor pressure of 5×10^{-2} Pa using a newly developed high temperature vacuum furnace with a gas flow system. The low $P_{\text{H}_2\text{O}}$ condition was achieved by the supply of water vapor from water ice in the gas flow system (Yamamoto et al., 2018, 2019). The run products were analyzed with FE-SEM (JEOL JSM-7000F) with an EDS. The O-isotope measurements were conducted with a secondary ion mass spectrometry (Cameca-ims 1280HR) at Hokkaido University.

The run products were composed of glass and spinel. The ¹⁸O fraction in the melt increased toward the surface of the sphere, and that at the surface of the sphere gradually increases with time. This suggests that both the gas-melt isotope exchange at the surface and the diffusive transport of O-isotopes into the melt interior occurred simultaneously. The obtained O-isotope profiles were explained by a three-dimensional spherical diffusion equation with a time-dependent surface concentration (Crank, 1975), yielding the O-self diffusion coefficient D of $1.87 \times 10^{-7} \text{ cm}^2 \text{ sec}^{-1}$ and the surface O-isotope exchange efficiency of ~0.25. The surface isotope exchange efficiency of 0.25 requires that one of four colliding water vapor molecules exchanges O-isotopes with the melt surface. The obtained D in the CAIB melt is consistent with that estimated in basaltic melt at 1390°C (Canil and Muehlenbachs, 1990; Leshner et al., 1996). This is likely because the diffusivity of oxygen would be related to the degree of melt

polymerization (NBO/T) (Liang et al., 1996), and the CAIB melt has a similar NBO/T value to that of basaltic melt.

The reaction kinetic suggests that the timescale for O-isotopic equilibration between a 1 cm-sized spinel-bearing CAIB melt droplet and water vapor should be at least a dozen days at plausible range of $P_{\text{H}_2\text{O}}$ in the protosolar disk ($10^{-9} < P_{\text{H}_2\text{O}} \text{ (bar)} < 10^{-6}$). Homogeneous O-isotope compositions of melilite in most type B CAIs would be explained by heating of CAI melt above the melilite liquidus for at least a dozen days during the reheating events in the early Solar System.

Keywords: CAIs, oxygen, isotope exchange, kinetics, protosolar disk