

Heterogeneity of oxygen and carbon isotopic compositions within carbonate grains in the Yamato 791198 CM chondrite and evolution of isotope ratios during aqueous alteration

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Carbonate minerals are ubiquitous in CM chondrites and formed by aqueous alteration in their parent body [1]. Previous studies have revealed a large variation in C isotope ratios of CM carbonates with $\delta^{13}\text{C}$ values ranging from ~ 20 to ~ 90 ‰ [e.g., 2,3]. However, the reason for this variability has remained poorly understood. Here, we conducted in-situ C- and O-isotope measurements by SIMS along crystal growth within single carbonate grains in Yamato 791198 (CM 2.4) to trace the change in $\delta^{13}\text{C}$ values during aqueous alteration.

First, we obtained color CL images of seven calcite (CaCO_3) grains with an FE-EPMA. Then, we measured O and C isotope ratios of the calcite grains with the CAMECA ims-1280HR. Typically, the isotope measurements were carried out on three different points for each calcite grain. Finally, we obtained quantitative chemical compositions and elemental maps of Mg, Si, Ca, Mn, and Fe of the calcite grains using the same FE-EPMA as that for the CL images.

All grain analysed exhibit a clear zoning of Mn, Mg, and Fe. The zoning is well correlated with CL images. The heterogeneous distribution of minor elements and CL intensity likely reflects the crystal growth of these grains.

The O isotope ratios of the calcite grains significantly vary with the $\delta^{18}\text{O}$ values ranging from ~ 22 to ~ 38 ‰. The O isotope ratios plot on a single trend line in an O three-isotope plot with a slope of ~ 0.61 , slightly steeper than the terrestrial fractionation line, reflecting a progressive O-isotope exchange between initially ^{16}O -poor water and ^{16}O -rich rock [4,5]. The $\delta^{18}\text{O}$ values also vary within single calcite grains, demonstrating crystal growth from a relatively dark CL core to a red CL periphery.

The $\delta^{13}\text{C}$ values of the calcite grains range from 17 to 32 ‰. This range is much smaller than that observed for different carbonate grains or bulk meteorite samples by previous studies. Especially, the $\delta^{13}\text{C}$ variations within individual grains are only 4 ‰ at most except for one grain. Previous studies have suggested that the observed $\delta^{13}\text{C}$ variation was produced by the Rayleigh-type isotopic fractionation by the escape of ^{13}C -poor methane [2], or equilibrium isotopic fractionation reflecting variable formation temperatures of carbonates [3]. However, the small intra-grain $\delta^{13}\text{C}$ variations associated with the large $\delta^{18}\text{O}$ change observed in this study cannot be explained by such processes. Instead, here we propose that the $\delta^{13}\text{C}$ values of dissolved carbon species in aqueous solutions were locally heterogeneous, and did not significantly change during carbonate formation.

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Keywords: Carbonate mineral, Carbonaceous chondrite, Oxygen isotope, Carbon isotope