OXYGEN- AND CARBON-ISOTOPE EVOLUTION IN FLUIDS DURING AQUEOUS ALTERATION.
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Introduction: CM chondrites exhibit evidence for aqueous alteration. Carbonates formed during aqueous alteration and recorded O and C isotopic compositions of water and dissolved inorganic C species. Although O and C isotopic compositions of carbonates are highly variable [1-3], much is unknown about the controlling factor(s) of the isotopic variation. In this study, we conducted in-situ O- and C-isotope measurements on calcite grains in the Nogoya CM chondrite in order to investigate O and C isotopic evolution during aqueous alteration.

Experimental: Oxygen- and C-isotope measurements were performed with the NanoSIMS 50 at AORI, UTokyo. 16,17,18O ions (in O-isotope measurement), or 12,13C, 16O, 12C18N, and 28Si ions (in C-isotope measurement) produced by a 20-30 pA Cs+ ion beam were detected with a FC and two EMs, or with five EMs, respectively. Typical errors on δ18O and δ13C values were 5.3 ‰ and 6.4 ‰ (2σ), respectively.

Results and discussion: We found calcite grains with distinct mineralogical characteristics (type I and II grains). Type I grains are likely to have precipitated in pore fluids while type II grains appear to have replaced other minerals. These observations have also been reported by previous studies (e.g., [4]).

Oxygen isotopic compositions are highly different between type I and II grains, but similar within each type (δ18O (SMOW) = 35.1 ‰ (type I) and 18.0 ‰ (type II) on average). The average Δ18O values are -3.0 ‰ (type I) and -5.7 ‰ (type II). These observations suggest that type II calcite formed later than type I calcite, because progressive alteration led to O-isotope exchange between water and anhydrous silicate with lower δ18O and Δ18O values than water. In contrast, C isotopic compositions of two types of grains are similar (δ13C (PDB) = 32.5 ‰ on average), except for one type II grain with δ13C of 53.8 ‰.

The variation in O and C isotopic compositions cannot be explained by change in formation temperatures [3], or the Rayleigh-type fractionation [2] alone. The δ18O variation must reflect both change in temperatures and O-isotope exchange between water and anhydrous silicate. Temperature-dependent C isotopic fractionation between CO32- and gaseous species such as CH4 may have been compensated by the Rayleigh-type fractionation resulted from escape of the gaseous phases.