

# 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. <sup>16,17,18</sup>O<sup>-</sup> ions (in O-isotope measurement), or <sup>12,13</sup>C<sup>-</sup>, <sup>18</sup>O<sup>-</sup>, <sup>12</sup>C<sup>14</sup>N<sup>-</sup>, and <sup>28</sup>Si<sup>-</sup> ions (in C-isotope measurement) produced by a 20-30 pA Cs<sup>+</sup> ion beam were detected with a FC and two EMs, or with five EMs, respectively. Typical errors on  $\delta^{17,18}\text{O}$  and  $\delta^{13}\text{C}$  values were 5.3 ‰ and 6.4 ‰ (2 $\sigma$ ), 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 ( $\delta^{18}\text{O}_{\text{SMOW}} = 35.1$  ‰ (type I) and 18.0 ‰ (type II) on average). The average  $\Delta^{17}\text{O}$  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  $\delta^{18}\text{O}$  and  $\Delta^{17}\text{O}$  values than water. In contrast, C isotopic compositions of two types of grains are similar ( $\delta^{13}\text{C}_{\text{PDB}} = 32.5$  ‰ on average), except for one type II grain with  $\delta^{13}\text{C}$  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  $\delta^{18}\text{O}$  variation must reflect both change in temperatures and O-isotope exchange between water and anhydrous silicate. Temperature-dependent C isotopic fractionation between  $\text{CO}_3^{2-}$  and gaseous species such as  $\text{CH}_4$  may have been compensated by the Rayleigh-type fractionation resulted from escape of the gaseous phases.

**References:** [1] Grady M. M. et al., 1988. *Geochim. Cosmochim. Acta* 52:2855-2866. [2] Guo W. and Eiler J. M., 2007. *Geochim. Cosmochim. Acta* 71:5565-5575. [3] Alexander C. M. O'D. et al., 2014. *Meteorit. Planet. Sci.* 50:810-833. [4] Lee M. R. et al., 2014. *Geochim. Cosmochim. Acta* 144:126-156.