

Negative crystals of calcite and empty crystals in the shape of hexagonal plate in carbonaceous chondrites.

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CI, CM and CR chondrites have calcite formed by aqueous alteration. Aqueous fluid inclusions have been searched from these mineral grains using an X-ray micro-tomography technique combined with FIB micro-sampling [1]. During the course of this research, we found pores with facets (negative crystals) in the shape of hexagonal plate in calcite and hexagonal platelet cavities (empty crystals) in matrix. Calcite has a variety of crystal morphology, which reflects their formation conditions such as temperature and partial pressure of CO₂ [2]. In this study, the shapes of these negative and empty crystals, which might have their formation conditions during aqueous alteration, were investigated based on crystallographic consideration. The shapes of negative crystals in terrestrial calcite were also examined for comparison.

Samples used in this study are a calcite grain (about 30 mm) with negative crystals in the Sutter's Mill meteorite (CM) and a grain of unknown phase (about 40 mm) with hexagonal platelet cavities in a matrix of the Ivuna meteorite (CI). Two single crystals with different morphologies, hexagonal plate and stud-like shape, which contain fluid inclusions, from Kamioka Mine were used as the terrestrial samples. Cubes or a cylinder 20 to 30 μm in size were sampled from thin sections using SEM/FIB and imaged by x-ray absorption imaging tomography with the effective spatial resolution of approximately 150 nm at BL47XU of SPring-8, Japan (e.g., [1]). The 3D shape of negative and empty crystals were extracted from CT images by binarization. The crystallographic orientations of the terrestrial calcite were determined with an FE-SEM/EBSD and the Miller indices of negative crystal facets were determined.

Two negative crystals about 2 mm are found in the Sutter's Mill sample. One clearly has a hexagonal plate shape, while the other not. As the calcite grain is a single crystal based on X-ray diffraction, the Miller indices of negative crystal facets were estimated by assuming that the hexagonal face is parallel to (001) and secondary face corresponds to (101). Both of the negative crystals have almost common combination of crystal faces but are different from a popular hexagonal shape composed of {001} and {104}. A large number of small inclusions (<1mm) are distributed in the calcite grain as a band, which is almost parallel to (001) of the negative crystals. This suggests that calcite crystal grew with (001) surface.

Several cavities (about 3 mm) with hexagonal plate shape were observed in the Ivuna sample. Miller indexing by assuming the hexagonal face as (001) suggest that the cavity is an empty crystal of carbonate such as calcite with (001) and (100) facets, although we could not eliminate a possibility of different minerals such as pyrrhotite.

Negative crystals in both of the terrestrial calcite crystals are distributed along planes, suggesting that they are healed cracks. The hexagonal plate crystal with {001} and {104} has a negative crystal (5 mm) with common {104} facets but {001} facets are absent. The stud-like calcite with {018} has a negative crystal (5 mm) with different facets of {1-12}. These results can be explained by that negative crystals in healed cracks have, faces with low surface free energies by dissolution and recrystallization during the course of healed crack formation, and (001) facet with high free energy disappeared or was originally absent. In contrast, (001) in the Sutter's Mill

negative crystals corresponds to growing crystal surface. Thus, it is important to examine conditions for growth with (001) surface.

[1] Tsuchiyama et al. 2014, *MAPS*, 49: A404. [2] Krasnova & Petrov 1997, Genesis of mineral individuals and aggregates. Nevsky kuryer, St.-Petersburg.

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