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Estimation of the equilibrium form of olivine from negative crystals in an equilibrated chondrite

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Olivine is one of the most common minerals both in the solar system and the interstellar and circumstellar environments. Adsorption of hydrogen atoms and water molecules onto the olivine surfaces may play a significant role in the formation of hydrogen molecules in the interstellar medium and the origin of water in the earth [2,3]. The crystal orientations of the olivine surfaces may affect adsorption efficiency and reaction rates, and thus it is important to understand crystal forms of olivine. Although the equilibrium form of olivine has been investigated based on the surface energies of olivine obtained by Ab initio calculations [1], it has not been confirmed by observations of naturally occurred olivine.

Many voids with facets (negative crystals) several um in size have been identified within olivine grains in HAYABUSA samples and equilibrated chondrites [4]. These negative crystals appear to be the equilibrium form because equilibrated chondrites have experienced thermal metamorphism at ~800^{o}C. In this study, we analyze the three-dimensional shapes of negative crystals in an olivine grain to obtain the equilibrium form of olivine using micro-sampling with focused ion beam (FIB) and micro-tomography [5]. The results are compared with the estimated equilibrium form by the Ab initio calculations.

A thin section of Tuxtuac meteorite (LL5) was used in this study. A negative-crystal-rich region of an olivine grain was identified by an optical microscope. The 30x30x60 um^3 sized micro-sample was prepared by an FIB technique (FEI Quanta 200 3DS). X-ray microtomography was performed at the SPring-8 BL47XU beamline at (7 keV, 70.5 nm/voxel). Negative crystals were extracted three-dimensionally by binarization of the CT images. The orientation of the host olivine crystal was determined with an FE-SEM/EBSD (JEOL 7001F/HKL CHANNEL5) and face angles of the negative crystals were measured using the 3D-CT images. Crystal plane indices of negative crystals were determined from the obtained crystal orientation and face angles.

We identified seven negative crystals 2-4 um in size. They aligned in the same plane within the olivine crystal suggesting that they are a healed crack as well as those in the HAYABUSA samples [5]. All negative crystals have developed facets of the same crystal orientations and have similar morphology, and some edges were rounded, which indicate that the morphology of the negative crystals almost represents the equilibrium form of olivine. The (100), (001), and (011) facets were observed and (100) facet was most well-developed. The other facets seem to be (001), (102), and (343) planes although these facets were small with respect to the spatial resolution of the micro-tomography.

Forsterite surface energies determined by Ab initio calculation increase in order of (010), (120), (001), (101), (111), (021), and (110) [1]. The observed (010), (120), and (001) planes have low surface energies while the surface energy of the (100) plane is high [6] and those of (011), (102), and (343) are probably high as well. The possible interpretations of unexpected facet planes with high energies are (1) changing surface energy due to adsorption of molecules onto the crystal surface, (2) temperature dependence of the surface free energy, and (3) kinetic effect. If surface energies change due to adsorption of molecules, anisotropy of the surface structures would play an important role for surface stability. Evaporation rate of forsterite along the a-axis in hydrogen gas at less than 1400^{o}C is smaller than those along the b- and c-axes [7], which is consistent with this study.

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