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Influence of intracrystalline organic macromolecules in shells on the crystal growth of calcite

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Biominerals often possess well-regulated structures with superior properties. Such superiority may be ascribed to the fact that biominerals are not pure inorganic crystals because they contain a certain amount of organic matter. Among the organic matter, ”intracrystalline” organic macromolecules such as proteins and polysaccharides have been studied recently, owing to their important roles during crystal growth. Nanoscopic investigation is essential to understand the interaction between crystals and intracrystalline organic macromolecules because the sizes of the organic macromolecules estimated from their molecular weights are several to dozens nanometers.

We are investigating mollusk shells, especially focusing on the relationships between calcite crystals, which is one of the polymorphs of calcium carbonate, and intracrystalline organic macromolecules. Transmission electron microscopy (TEM) with electron energy-loss spectroscopy (EELS) identifies the intracrystalline organic macromolecules, and X-ray diffraction (XRD) evaluates macroscopic properties of the crystals. Using these nanoscopic and macroscopic techniques, we revealed that the distribution of the intracrystalline organic macromolecules affects the microstructure of the crystals. In the prismatic structure in a pearl oyster (*Pinctada fucata*), which is composed of columnar calcite crystals, intracrystalline organic macromolecules are distributed inhomogeneously, which causes small misorientations and local lattice strain. On the other hand, the organic macromolecules in the prisms in a pen shell (*Atrina pectinata*) are distributed homogeneously in the crystals, and local lattice strain is rarely observed. We thought such difference is originated from the different affinities of the organic macromolecules to the calcite crystals. Therefore we conducted calcium carbonate crystallization *in vitro* with EDTA (ethylenediaminetetraacetic acid) soluble extract from the prisms. It was found that the organic macromolecules in the extract from *P. fucata* and *A. pectinata* are incorporated differently in calcite. Furthermore, we are characterizing the organic molecules in the extract using molecular biological techniques to consider their interaction with their host calcite crystals.

Keywords: organic macromolecules, calcite, biomineral, TEM