Evolution of disorder in crystalline structures between single crystal and polycrystal via chemical and physicochemical approaches

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In accordance with the modern definition [1,2], a mesocrystal is delineated as a nanostructured material (a mosaic structure of a crystalline solid) that consists of individual nanoparticle building units with a defined order on the atomic scale in at least one direction. The crystallographic regularity of mesocrystals consisting of iso-oriented crystalline units is regarded as an intermediate structure between a homogeneous single crystal and a polycrystal, which is a random aggregate of small crystals. Most of carbonate- and phosphate-based biominerals are composed of mesocrystals, in which nanoscale crystalline units are ordered in approximately the same crystallographic direction [3]. Various elaborate architectures consisting of mesocrystals are constructed by the combination of soluble and insoluble biological organic agents in biotic aqueous systems [4]. However, the variation of the crystallographic regularity of mesocrystals has not been clarified using experimental evidence.

This work demonstrated the gradual evolution of disorder in crystalline structures between single crystal and polycrystal by monitoring the crystallographic regularity using electron diffraction patterns [5]. Single-crystal rods of fluorapatite elongated in the *c* direction are switched to mesocrystals consisting of iso-oriented nanorods by adding a specific molecule having a carboxy group (chemical approach) and to roughly arranged nanograins by increasing the degree of supersaturation at the growth front (physicochemical approach). Lateral and vertical miniaturization of the growth units is achieved through stepwise nucleation and near-epitaxial growth. The crystallographic regularity gradually decreases with deviation of the *c* axis and rotation of the *a* axes through reduction of the growth unit size. Here we can tune the disorder in crystalline structures via the chemical and physicochemical approaches.

References

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