Face-specific replacement of calcite by amorphous silica nanoparticles

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Natural nanoparticles are fundamental building blocks of Earth´s bio- and geosphere. Amorphous silica nanoparticles are ubiquitous in nature, but fundamental knowledge of their interaction mechanisms and role in mineral replacement reactions is limited. Here we show how silica nanoparticles replace Cretaceous calcite bivalve shells in a volume- and texture-preserving process. Electron microscopy and tomography show that during mineral replacement calcite crystallographic orientations are transferred to twinned photonic crystals composed of face-centered cubic silica sphere stacks. These sphere arrays perfectly replicate calcite twin lamellae in periodically changing orientations and the intersection angle of {018} twin and {104} cleavage planes. During the face-specific replacement process, silica nanoparticles continuously nucleate, aggregate, and form a lattice of uniform spheres parallel to calcite low-energy facets. We explain the replacement process with a novel model that unifies recently proposed, probably universal mechanisms of interface-coupled dissolution-precipitation and crystallization by particle attachment; both are key mechanisms in geological processes and nanomaterials design and synthesis.