Goldschmidt 2023 Abstract
https://doi.org/10.7185/gold2023.17064

The roles of precursor (Mg)-ACC in the formation of aragonite

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Amorphous calcium carbonate (ACC) is often referred to as a phase that can transform into either of the three anhydrous crystalline $CaCO_3$ forms (calcite, aragonite and vaterite). Here, we examined the conditions that make ACC transform into aragonite. Even though aragonite is the stable anhydrous crystalline $CaCO_3$ polymorph at high pressure (4–40 GPa), it is also widespread on Earth's surface. Nevertheless, the mechanism of its formation and the roles of solid precursors (such as ACC) in the process have not been studied as extensively as in the case of calcite.

We studied aragonite formation by synthesizing it under laboratory conditions, focusing on the two, environmentally most relevant factors that are known to enhance its formation: (*i)* the presence of dissolved Mg^{2+} ions and (*ii)* the temperature of the mother solution (up to 80 °C). We used different experimental approaches in the two cases, and studied the precipitated materials with various scanning transmission electron microscopy (STEM) techniques.

In Mg-rich solutions the first solid product was Mg-bearing ACC that transformed into aragonite aggregates with a typical, spindle-like appearance. Each spindle was an aggregate of crystallographically oriented nanocrystals, and two or more spindles could form star-like assemblies, with their members in twin orientation. Whereas the remarkable hierarchical structure suggested a growth mechanism by nanoparticle attachment, chemical differences observed between Mg-bearing ACC and aragonite indicated that the process must have involved dissolution and re-precipitation (DRP) as well.

At elevated temperature ACC particles formed in the highly supersaturated (Mg-free) solutions, and their electron diffraction analysis suggested a trend of structural change with increasing temperature. The increasing temperature accelerated the transformation of ACC into crystalline forms (both calcite and aragonite), with aragonite formation favored at 80 °C. Morphological features suggested that the ACC could transform into either calcite or aragonite in solid state (preserving the globular shape of ACC), while simultaneously DRP produced aragonite crystals with aggregate or needle-like appearance. Thus, aragonite formation from ACC involved particle aggregation coupled with DRP in Mg-rich solutions, and solid-state transformation coupled with DRP at elevated temperatures. [1]

[1] Support from NKFIH under grants RRF-2.3.1-21-2022-00014 and SNN-139585 is acknowledged.