

## **Deciphering Amorphous Calcium Carbonate Crystallization Pathway Using Stable Isotope Labels**

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The realization that carbonate biominerals can form by an amorphous to crystalline transformation has prompted the question of when and where chemical signatures are recorded during mineralization. This pathway begins with accumulation of amorphous calcium carbonate (ACC) in a localized environment that subsequently transforms to the crystal/organic composites we know as skeletal structures. It is not yet established if the transformation involves microscopic dissolution and reprecipitation or an altogether different mineralization pathway, such as a solid-state transformation. In this study, we employ stable isotope labeling to directly probe the extent of cation incorporation from the solution during transformation of ACC of varying Mg compositions.

We find that calcites do not retain the isotopic labeling of the initial ACC phase, as predicted for a solid-state transformation pathway. Rather, our analysis of the crystals shows signatures that are explained by a mixing model of the initial ACC phase and the labeled solution. Further, the extent of mixing correlates with rates of transformation, indicating the calcites are recording the changing solution environment during transformation. This study suggests skeletal signatures are not set when ACC forms, but are instead determined at the time of calcite growth.