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The amorphous-to-crystalline transition in the Ca–Mg–carbonate system as a function of composition, time and temperature

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Calcium and magnesium carbonate minerals play an important role in regulating Earth's climate, they are amongst the most common biominerals, and they are known to precipitate from amorphous precursors. Previous work has focused on characterization of amorphous precursors such as amorphous Ca-Mg-carbonate (ACMC), amorphous Ca-carbonate (ACC) and amorphous Mg-carbonate (AMC) [1,2]. Our experiments shed new light on how the composition of amorphous carbonate phases influences the mineralogy of their crystalization products.

We synthesized amorphous carbonates with different molar ratios of Ca and Mg (n=6, compositons from 0–100 mol% Mg, spanning the range from ACC to AMC), and then examined the crystallization process as a function of time at two temperatures (i.e., 20–23˚C and 75˚C). The Ca/Mg ratio is an important parameter controlling minerology: endmember phases (i.e., $CaCO_3$ and $MgCO_3$) tend to crystallize first, and Mg-carbonates crystallize more quickly in Ca-rich ACMC. Room temperature crystallization is faster for Ca-bearing amorphous phases: one day for ACC (100 mol% Ca) compared to 6 weeks for ACMC with 61 mol% Ca and ~10 weeks for AMC (0 mol% Ca). The Ca/Mg ratio also determines Ca-carbonate polymorphism: calcite and vaterite were only found together in ACC and aragonite formed only in ACMC with 15–53 mol% Mg. At 75˚C, the tendency for Mg to substitute for Ca in the calcite group structure increased. Higher temperature incubations allowed for the formation of magnesite or VHMC alongside hydromagnesite. Increased crystallization rates were also observed at 75˚C. The crystallization products of amorphous carbonates depend on the initial Ca/Mg ratio such that final mineral assemblages can be used to identify the precursor phase. Our results provide new insights into the complex mineralogy of biomineralization, carbonate playa lakes and carbonate sediments at the seafloor.

[1] Purgstaller et al. (2019) *CrystEngComm* 21, 155–164.
[2] Radha et al. (2012) *GCA* 90, 83–95.