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Structure and Hydration of Amorphous Magnesium Carbonate (AMC): Implications for the low-temperature formation of Ca-Mg Carbonates

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Amorphous precursors are considered fundamental intermediates during the formation of calcium and magnesium carbonates in natural settings such as caves, lakes, and marine environments. While the formation of amorphous calcium carbonate (ACC) has been extensively studied and documented, the formation and stability of amorphous magnesium carbonate (AMC) at low temperatures (0-25 °C) remain comparatively less explored, despite its expected critical role in the global carbon cycle. This study investigates the abiotic precipitation of AMC and the impact of temperature and pH on its structure and characteristics, to better understand its potential role in the natural low-temperature formation of anhydrous, Mg-bearing crystalline carbonates like huntite and dolomite.

We synthesized AMC at four temperatures (5, 10, 18, and 22 °C) by slow titration of a $MgCl_2$ solution into a $K_2CO_3/KHCO_3$ buffer, of varying pH (10-11.25). The resulting precipitates were characterized using FTIR, TGA/DSC, TEM, and solid-state MAS NMR. Our results show that temperature and, particularly, pH are key controls on the OH/H_2O content of AMC, which seems to influence its short-range structure and stability in solution. Notably, the stability of AMC decreases with decreasing pH; for pH< 10.5, AMC becomes unstable and transforms immediately after its precipitation into crystalline phases. Additionally, at lower temperatures, AMC nucleation is delayed and recrystallize faster. These findings contrast with those from our research group in Nerja Cave, where stable AMC persists at a circumneutral pH (8.0 ± 0.2) and average temperatures of 18.1 ± 0.1 °C. This implies that the stability of the amorphous phase in such natural environments is likely not determined exclusively by physicochemical conditions of the precipitating fluids (drip water). Instead, the presence of organic compounds associated to microbial activity, and/or the effect of dissolved silica, may play a crucial role in stabilizing AMC as a precursor of mixed Ca-Mg carbonates.