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Control of carbonate alkalinity on Mg incorporation in calcite: Insights on the occurrence of high Mg calcites in diagenetic environments

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High Mg calcites (HMC), with up to 25 mol % of Mg, are common features in early diagenetic environments and are frequently associated with bio-induced anaerobic oxidation of methane (AOM). Such archives hold valuable information about the biogeochemical processes occurring in sedimentary environments in the geological past. Despite the frequency AOM-induced HMC observed in marine diagenetic settings and their potential role in dolomitization, only a minor number of experimental studies has been devoted on deciphering their formation conditions.
Thus, in order to improve our understanding on the formation mechanism of HMC induced by elevated carbonate ion concentrations, we precipitated HMC by computer controlled titration of a (Mg,Ca)Cl2 solution at different Mg/Ca ratios into a $NaHCO_3$ solution under precisely defined physicochemical conditions (T = 25.00 $\pm$0.03°C; pH = 8.3 $\pm$0.1). The formation of carbonates was monitored at a high temporal resolution using in situ Raman spectroscopy as well as by continuous sampling and analyzing of precipitates and reactive solutions.
We identified two distinct mechanisms of HMC formation. In solutions with molar Mg/Ca ratios $\leq$ 1/8 calcium carbonate was precipitated as crystalline phases directly from homogeneous solution. In contrast, higher Mg/Ca ratios induced the formation of Mg-rich ACC (up to 10 mol % of Mg), which was subsequently transformed to HMC with up 20 mol % of Mg. Our experimental results highlight that the finally formed HMC has a higher Mg content than the ACC precursor phase. Considering experimental data for Mg containing ACC transformation to crystalline calcium carbonate from literature, the continuous enrichment of Mg in the precipitate throughout transformation of amorphous to crystalline $CaCO_3$ most likely occurs due to the high carbonate alkalinity (DIC about 0.1 M) of our reactive solutions. The Mg incorporation into calcite lattice seems to be favored by intensive supply of carbonate ions as observed in AOM originated HMC in early diagenetic sediments.