New recipes for CO₂ mineralization: accounting for crystal chemistry and crystallization pathway

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Previous estimates of CO_2 mineralization potential have typically assumed ideal stoichiometry for feedstock minerals and carbonate mineral products. However, the silicate and hydroxide minerals used for CO_2 mineralization are generally Ca-Fe-Mg solid solutions. As a result, CO_2 mineralization reactions are dependent on redox conditions and differences in solubility that occur for non-endmember mineral compositions. Both the stability and efficiency of CO_2 storage in minerals may also depend on crystallization pathway: whether a carbonate mineral forms classically, ion-by-ion from solution, or non-classically by particle-based assembly.

Here, we show that Ca-bearing magnesite $[(Mg,Ca)CO_3]$ with ~10 mol% Ca is favoured to form from amorphous Ca-Mgcarbonate (ACMC) at temperatures of 60 and 80 °C. Lowtemperature magnesite with similar composition and microstructure is consistently observed in sediments from alkaline lakes in the Basque, Atlin and Cariboo Plateau regions in Canada and the Coorong, Australia. Work from the Coorong lakes suggests that very high magnesium calcite or VHMC (Ca_{0.5}Mg_{0.5}CO₃, also called disordered dolomite) and Ca-bearing magnesite form non-classically, as has been observed in the laboratory, when lake waters reach the extreme supersaturations required to form ACMC [1].

We also report new results from low-temperature carbonation experiments using synthetic, Fe(II)-bearing brucite [(Mg,Fe) (OH)₂]. Synthetic brucite containing 5, 20 or 40 mol% Fe(II) was carbonated using 10% CO₂ gas under oxic and anoxic conditions in the presence of different background anions (Cl⁻, SO₄²⁻, H₄SiO₄⁰). Rather than forming a single carbonate mineral phase, complex assemblages of amorphous and crystalline hydrated and hydroxylated Mg-Fe-carbonate, layered double hydroxide, Fe-(hydr)oxide and Mg-Fe-silicate minerals are produced. The efficiency of carbonation depends strongly on both the Fe(II) content of brucite and the identity of the background anions in solution, with the amount of CO₂ mineralized decreasing with increasing Fe substitution and addition of silica to solution.

A better understanding of how crystal chemistry and crystallization pathway affects CO₂ storage will provide us with

greater predictive power and new recipes to capture and/or store CO₂ more stably and efficiently in minerals.

[1] Raudsepp et al. (2022) Sedimentology, 69, 2246.