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Direct nano-scale observations of CO_2 sequestration during brucite ($Mg(OH)_2$) dissolution

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Mineralization of CO_2 is regarded as the safest and most permanent option for carbon sequestration (e.g., O'Connor et al. 2005). So far, research has been concentrated on the carbonation of silicates such as olivine ($(Mg,Fe)_2SiO_4$) and serpentine ($Mg_3Si_2O_5(OH)_4$). However, also the less common hydroxide mineral brucite ($Mg(OH)_2$) has recently become a focus of interest, owing to its much higher reactivity relative to silicate minerals at low temperature and $P\mathrm{CO}_2$ conditions(e.g., Zhao et al. 2010; Schaef et al. 2011). Utilization of brucite for carbon sequestration requires an in-depth understanding of the associated reaction mechanisms from the macro- to the nano-scale. Therefore, we have conducted a series of in-situ and ex-situ Atomic Force Microscopy (AFM) experiments enabling direct nano-scale observations of dissolution and precipitation features on brucite (001) cleavage surfaces. In particular, we tested the effects of pH (2-12), aqueous $NaHCO_3$ concentration (1μM – 1M) and $P\mathrm{CO}_2$ (0-1 bar) on brucite dissolution and carbonation.

Brucite dissolution proceeded by the formation and spreading of etch pits with equilateral triangular shapes. Measured etch pit spreading rates increased with decreasing pH (from 0.030 ± 0.008 nm/s at pH 9 to 0.70 ± 0.07 nm/s at pH 2) and increasing $NaHCO_3$ concentration (from 0.038 ± 0.004 nm/s in 1μM to 0.38 ± 0.07 nm/s in 1M solution). In pure $NaHCO_3$ solutions (pH 7.2-9.3) secondary phase precipitation was relatively minor. Enhanced precipitation was observed in slightly acidified $NaHCO_3$ solutions (pH 5) as well as in solutions that were equilibrated with 1 bar CO_2 (pH 4). Nucleation predominantly occurred in areas of high dissolution such as larger step edges. Initially, nucleating particles were only 1-2 nm and weakly attached to the brucite surface as they could be easily pushed away by the scanning tip during in-situ AFM experiments. Growth of the particles was observed after ex-situ AFM experiments lasting for several hours. The size of individual particles increased to about 100 nm within 16 h. Lateral spreading of the particles was rather limited. Instead, we observed the formation of particle clusters reaching total heights of up to 1 μm after 32 h. SEM-EDX investigations confirmed that the precipitate is an Mg-rich carbonate phase (probably hydromagnesite). Our results contribute to an improved understanding of the mechanism of aqueous brucite carbonation at low temperature and $P\mathrm{CO}_2$ conditions and to a further understanding of carbonation reactions in general.

References

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