

Quantifying carbonation rates of periclase and brucite powders at elevated $p\text{CO}_2$: Implications for MgO looping

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Mineral oxide looping is a negative emissions technology, initially proposed by McQueen et al. (2020),¹ which involves mining and calcining either magnesite (MgCO_3) or calcite (CaCO_3) to reactive oxides that can be repeatedly used to capture CO_2 . Despite slower kinetics,² periclase (MgO) carbonation is favourable due to the lower calcination temperatures of the magnesium carbonate products, thereby lowering energy requirements. This study aimed to determine (1) MgO hydration rates, (2) periclase and brucite [$\text{Mg}(\text{OH})_2$] carbonation rates, and (3) calcination temperatures of the Mg-carbonate products. Experiments evaluated hydration rates of MgO powders in liquid water exposed to different treatments (still, mixing, and sonication) and at 100% relative humidity (RH). Carbonation experiments were conducted at variable CO_2 concentrations (0.04%–9.0%), and changes in total inorganic carbon content were analyzed over time to derive carbonation rates. Mineralogical changes were monitored using X-ray diffraction (XRD) with MgO hydration rates determined by Rietveld refinement using XRD data. The thermal degradation behaviour of the carbonation products was assessed by thermogravimetric analysis. MgO in aqueous still, mixing, and sonication experiments reached 90 wt.% $\text{Mg}(\text{OH})_2$ within 16, 20, and 12 days, respectively, with hydration rates ranging from 4.5–7.4 wt.% $\text{Mg}(\text{OH})_2$ /day. Hydration of MgO exposed to 100% RH was incomplete, reaching a maximum extent of 62 wt.% $\text{Mg}(\text{OH})_2$ after 9 days. Carbonation of $\text{Mg}(\text{OH})_2$ consistently reached complete conversion, while MgO never reacted completely, likely due to passivation.² Carbonation rates of MgO and $\text{Mg}(\text{OH})_2$ increased logarithmically with increasing CO_2 concentration, ranging from 0.25–3.0 %C/day and 0.15–3.4 %C/day, respectively. Dypingite [$\text{Mg}_5(\text{CO}_3)_4(\text{OH})_2 \cdot 5\text{--}8\text{H}_2\text{O}$] was the magnesium carbonate that formed in all experiments and had a thermal degradation behaviour similar to natural hydromagnesite, calcining to MgO at $\sim 500^\circ\text{C}$, which is markedly lower than magnesite (600°C) and calcite (900°C). These results suggest the promising implementation of Mg-based materials in a mineral looping technology that separates aqueous hydration from carbonation and harnesses CO_2 -rich gas streams.³

[1] McQueen et al. (2020). *Nat. Commun* 11, 3299. [2] Rausis et al. (2022) *Int. J. Greenhouse Gas Control* 119, 103701. [3] IPCC (2005). Special Report on Carbon Dioxide Capture and Storage.