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Improving periclase and brucite carbonation for scalable MgO looping: Implications for direct air capture

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Mineral looping is a carbon dioxide (CO_2) removal technology that uses reactive oxides, generated by calcining abundant magnesium ($MgCO_3$) or calcium carbonates ($CaCO_3$), to repeatedly capture atmospheric CO_2. The use of Mg-carbonates is often considered advantageous due to their lower calcination temperatures, thereby reducing the overall energy demands of the looping technology. However, slow kinetics and passivation have limited large-scale deployment, and the performance of natural Mg-carbonates remains largely unexplored. Here, we investigated (1) the role of liquid water in reducing MgO passivation and (2) carbonation rates and extents of thin films of calcined and hydrated products of natural Mg-carbonates from Türkiye, as well as chemical grade MgO and $Mg(OH)_2$. Films were either dry, hydrated with deionized water, or hydrated with water equilibrated with 100% CO_2 (i.e. carbonated water; pH ~3.9), then exposed to atmospheric CO_2 concentrations (~420 ppm) at 100% relative humidity and 28 °C for 130 days. Changes in total inorganic carbon content were measured over time using coulometry, and mineralogical and morphological progressions were tracked by X-ray diffraction and scanning electron microscopy, respectively. Among the synthetic MgO, the impact of passivation was highest in dry films (~51% carbonation) and lowest in films hydrated with carbonated water (~92% carbonation), despite similar carbonation rates in the initial time period between day 0 and 18 (0.25 %C/day and 0.24 %C/day, respectively). Films hydrated with deionized water exhibited an initial linear carbonation rate (0.20 %C/day) that plateaued between days 8–14 (13% carbonation), followed by a slow, semi-linear carbonation rate (0.04 %C/day) resulting in ~69% carbonation by day 130. Contrastingly, the carbonation of MgO films produced from calcined natural Mg-carbonates plateaued early beyond day 3, despite complete hydration to $Mg(OH)_2$ within 24 h. $Mg(OH)_2$ films produced from calcined and hydrated natural Mg-carbonates exhibited distinct, highly linear carbonation rates ranging from ~0.05–0.26 %C/day. These results demonstrate that reacting MgO with carbonated water, which can be recycled from captured CO_2, reduces passivation. Additionally, results suggest promising potential for Mg-carbonates derived from natural deposits, as well as high-purity mine wastes, in a MgO looping technology that separates aqueous hydration from carbonation.