CO2 mineralization by controlled carbonation of MgO powders via mineral looping

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Anthropogenic climate change is driving researchers to find viable solutions for CO₂ storage. CO₂ mineralization is being adopted in industries such as cement production, turning towards the use of carbonated lime products and alternatives to lime such as MgO. McQueen et al. [1] proposed the use of MgO powder for direct air capture of CO2. Recent experimental studies on direct carbonation of MgO at ambient conditions indicate surface passivation [2]. However, the effect of repeated carbonation cycles on the reactivity of MgO is not well understood. Here we investigate the effect of initial MgO surface area on its reactivity for carbon mineralization at 100% relative humidity in vapor, room temperature, and 1bar of CO₂ in sealed reaction vessels. The MgO sources are three different nanopowders with surface areas of 6.14, 18.97 and 31.01 m²/g after being heated at 450°C for 18 hours prior to carbonation. Three mineral looping cycles are applied to the powders, that are carbonated for two weeks, calcined at 500°C, and carbonated at the same conditions to understand the limitations of the looping process based on reactivity decay. Surface areas of the starting, reacted, and calcined products are measured by BET, particle size and morphology are investigated by TEM imaging, and the extent of reaction by Thermogravimetry and X-ray diffraction. The morphologies of the starting powders are cubic for the one of lower surface area and rounded for the other two. Results show that nesquehonite and brucite are the main mineral phases forming in the carbonation process under high humidity vapor environment, with nesquehonite being the predominant phase. The degree of reaction measured by both thermal decomposition and phase quantification by Rietveld refinement depends on the surface area of the starting powders, being higher for larger surface area.

[1] McQueen, N., Kelemen, P., Dipple, G. *et al.* (2020), Ambient weathering of magnesium oxide for CO₂ removal from air. *Nat Commun* **11**, 3299. https://doi.org/10.1038/s41467-020-16510-3

[2] Rausis, K., Stubbs, A. R., Power, I. M., Paulo, C., (2022), Rates of atmospheric CO2 capture using magnesium oxide powder, International Journal of Greenhouse Gas Control, Volume 119. https://doi.org/10.1016/j.ijggc.2022.103701