

The influence of humidity and CO₂ supply on calcium oxide looping: Implications for passive direct air capture

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Calcium oxide (CaO) looping is a proposed negative emissions technology that passively captures carbon dioxide (CO₂) from the atmosphere to form calcium carbonate (CaCO₃), which is calcined to regenerate the CaO powder for further reaction while a high-purity CO₂ is utilized or stored. Currently, the influence of relative humidity (RH), CO₂ supply, and deposit thickness on CaO hydration and carbonation under ambient conditions is not well understood, limiting potential implementation. This study aims to determine 1) the effects of relative humidity (RH) and CO₂ concentrations on the rates and extents of CaO hydration and carbonation, 2) the CO₂ removal rates of varying CaO deposit thicknesses and, 3) the impacts of CO₂ supply rates on passive direct air capture systems. Controlled relative humidity (20, 40, 60, 80, 95% RH) and humidity swing (60–80/95% and 40–60/95% RH) experiments were conducted to quantify reaction rates and extent over 14 days. Although complete hydration to Ca(OH)₂ was always attained, full carbonation (>90%) was not achieved under most experimental conditions, likely owing to passivation as indicated by electron microscopy. Flow-through experiments tested different CO₂ concentrations (100, 200, 300, 400–500 ppm). Slower reaction rates, yet greater carbonation extents, were achieved at lower CO₂ concentrations, allowing greater CaO hydration to Ca(OH)₂ prior to carbonation. Our findings indicate that complete CaO hydration must occur prior to carbonation in addition to greater water availability to minimize passivation. Based on CO₂ fluxes, CaO deposits with 0.5, 1.0, and 1.5 cm thicknesses and exposed to 85–95% RH had similar CO₂ removal rates of -32, -35, -33 kg CO₂/m²/yr, respectively, after 24 days. Lastly, a flow-through experiment assessed the air exchange rates required to maintain atmospheric CO₂ concentrations for a given mass of CaO. This experiment details how spreading CaO powder in units of vertical stacks/plates would yield a significantly greater CO₂ supply rate for an equal amount of mass compared to one horizontal layer of powder, leading to faster carbonation rates based on our previous results. Overall, a new understanding of the influences on humidity-driven reactions demonstrates high carbonation efficiency and the knowledge necessary for pilot-scale implementation.