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Scalable atmospheric CO_2 capture using calcium oxide powder

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Calcium oxide (CaO) looping is a negative emissions technology that uses CaO powder to passively capture carbon dioxide (CO_2) from the atmosphere to form calcium carbonate ($CaCO_3$), which is then calcined to regenerate the CaO powder for further CO_2 capture and the high-purity CO_2 is utilized or geologically stored. Presently, the control of relative humidity (RH), CO_2 supply, and deposit thickness on CaO hydration and carbonation under ambient conditions are not well understood, limiting potential scaling. This study aimed to determine 1) the control of RH on the rates and extents of CaO hydration and carbonation, 2) the impacts of CO_2 supply rates on CO_2 removal rates and passive direct air capture system designs, and 3) the CO_2 removal rates of varying CaO deposit thicknesses. Controlled RH experiments (20, 40, 60, 80, 95%) demonstrated that hydration and carbonation rates were fastest at high humidity, yet passivation limited carbonation extent, as seen using electron microscopy. In contrast, humidity swing experiments (e.g., 40% to 95%) showed that nearly full carbonation (>85%) can be achieved by separating hydration and carbonation reactions using a swing from low to high RH. Subsequently, a flow-through experiment exposed CaO (0.5 cm; 100 g) to 0.1–20 L/h of humid air for 21 days; the effluent CO_2 concentration never reached laboratory CO_2 concentrations, suggesting larger scale deployments of CaO may be CO_2 supply limited. A second series of flow-through experiments exposed CaO and $Ca(OH)_2$ to different CO_2 concentrations (100, 200, 300, 400–500 ppm). While CaO carbonation was again limited by passivation, all $Ca(OH)_2$ experiments reached >90% $CaCO_3$. Lastly, CO_2 flux experiments used CaO deposits with 0.5, 1.0, and 1.5 cm thicknesses and were exposed to 85–95% RH for 24 days, resulting in CO_2 removal rates ranging between 12 to 117 kg $CO_2/m^2/yr$. Our study provides a foundational understanding, previously absent in the literature, that elucidates the relationships between two critically important variables, RH and CO_2 supply, for passive direct air capture using calcium oxide powder, thereby helping enable future laboratory studies and pilot-scale deployments.