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Coupled effects of relative humidity and temperature on portlandite carbonation

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Mineral carbonation, a naturally occurring process wherein CO_2 reacts with alkaline minerals, is a promising method for CO_2 sequestration to help mitigate climate change. Portlandite [$Ca(OH)_2$] is one of the most reactive alkaline minerals for mineral carbonation which efficiently stores CO_2 as anhydrous $CaCO_3$. Portlandite adsorbs water vapour from the atmosphere onto its surfaces, where the water acts as a medium for CO_2 - mineral interactions, allowing portlandite dissolution and carbonate mineral precipitation even in the absence of liquid water. In this study, *in situ* X-ray diffraction (XRD) was employed to determine portlandite carbonation kinetics under controlled relative humidity (RH, at 90%, 75%, 60% and 43%) and temperature (60°C, 40°C and 20°C). The water sorption behaviour of portlandite in an RH range of 0 - 98% was assessed by dynamic vapour sorption (DVS) analyses at the same temperatures as in XRD experiments. The results suggest that both carbonation rate and maximum carbonation extent of portlandite, as well as mineralogy and morphology of $CaCO_3$, are influenced by RH and temperature. Carbonation kinetics were determined using the JMAK model, and apparent activation energies of calcite precipitation during portlandite carbonation at 75% and 60% RH were determined to be 93 kJ mol^{-1} and 206 kJ mol^{-1}, roughly two and four times higher compared to those estimated during carbonation in bulk solutions, respectively. These findings enhance our understanding of CO_2 - mineral interaction and $CaCO_3$ growth in water-limited environments and provide insights into CO_2 removal and storage in industrial applications under near-Earth's surface conditions.