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Assessing the impacts of passivation and Ca-silicate formation on lime looping efficiency for direct air capture

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Lime (CaO) based mineral looping is a widely recognised carbon dioxide removal (CDR) technology involving the use of limestone/Ca-based byproducts for sustainable CO_2 capture from industrial point sources. In recent years, the potential of this technology is further being explored for a gigatonne-scale direct air capture (DAC) of atmospheric CO_2. However, scaling up this DAC technology remains subject to recognising limitations to delineate strategies for increasing process efficiency. In this study, we carried out several batch experiments involving calcination-hydration-carbonation of limestone with variable particle sizes and purities to demonstrate two significant inefficiencies in the looping process: i) surface passivation in relation to particle size of the initial limestone powder, and (ii) the presence of silicate impurities.

Passivation limited carbonation extent and rates to <68% and <1.06 wt.% $CaCO_3$/h, respectively, for lime generated from coarser (>25 μm) limestone powder compared to 88% and 1.66 wt.% $CaCO_3$/h for that of finer (<25 μm) particles. Hence, the significance of using very fine limestone powder obtained from proper pulverisation is emphasized for optimising looping efficiency. Additionally, non-carbonate phases that naturally occur in limestones, for example, the 7.5 wt.% redundant fraction in an impure limestone used in this study, remained unreactive to CO_2 and proportionately reduced CDR efficiency. Furthermore, the high temperatures (≥900 °C) required for calcination induced a thermal solid-state reaction between silicates, commonly present in limestones as quartz (SiO_2), and CaO to form relatively inert Ca-silicate phases. These phases can lower CO_2 capture capacity by removing a fraction of freely available CaO and also passivating reactive surfaces, impeding carbonation. This detrimental impact was evidenced by lower reaction extents (86%–75%) in limestones containing 4.4–24.4 wt.% SiO_2, compared to the 93% and 98% carbonation achieved for very high-purity limestone (55.6 wt.% CaO; 0.35 wt.% SiO_2) and synthetic $CaCO_3$ (56.01 wt.% CaO; 0.07 wt.% SiO_2), respectively. Based on these findings, ~10–19% reductions in looping efficiency was estimated when using finely (<25 μm) pulverised, very high-purity to impure limestones. The results also emphasize caution regarding the risks of under-performance of Ca-based byproducts, such as marble wastes and industrial slags, in lime looping.