Towards Novel, Scalable and Realizable Technologies for Carbon Capture, Utilization, and Storage: Hybrid Absorption-Crystallization Pathways for Inorganic and Organic Carbonate Synthesis and H_2 Generation

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The urgency to meet our societal targets for net zero emissions calls for developing a portfolio of technologies for carbon capture, utilization, and storage (CCUS). Novel solutions and systems are needed to permanently remove CO_2 from our point source emissions and from the atmosphere. In this context, capturing and removing CO_2 as inorganic carbonates, also known as carbon mineralization, is an emerging and versatile strategy that can be harnessed for carbon removal or for producing high purity H_2 through the reactive separation of CO_2 and H_2. In these multiphase reactive environments, the low solubility of CO_2 in water is a challenge. To overcome this challenge and advance process-intensified strategies, we are developing integrated approaches that combine the use of dilute CO_2 concentrations with aqueous amine and amino acid solvents to enhance the absorption of CO_2 into the aqueous phase and increase the supply of carbonate ions for producing solid carbonates in fewer unit operations. Calcium and magnesium content that is abundant in naturally occurring silicate minerals and alkaline industrial residues is harnessed for producing crystalline carbonates. Alternatively, the higher solubility of pressurized CO_2 and the favorable kinetics of calcium and magnesium carbonates formation at elevated temperatures can be harnessed to use carbon mineralization as a separation pathway to produce high purity H_2 from various carbon-bearing sources including syngas and biomass.

The concept of hybrid absorption-crystallization can also be harnessed to accelerate CO_2 capture from dilute sources by developing novel phase changing guanidine bases. These bases react with CO_2 in the aqueous phase to produce solid carbonate-bearing crystalline solids which readily precipitate from the aqueous phase. To advance mechanistic insights into these hybrid absorption-crystallization pathways as they occur, we developed micro-reactor environments to probe the structural and morphological evolution of the crystalline phases using *in-operando* X-ray scattering and spectroscopy methods. These studies demonstrate the need to advance measurement science in multiphase reactive environments to accelerate the development of novel, scalable, and realizable technologies for carbon capture, utilization, and storage.