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3D microfluidic investigation of crystallization behavior in porous media for carbon storage application

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We investigate the impact of multiphase fluid transport on nucleation and crystallization (precipitation) reactions under pore confinement and the influence of that behavior on permeability and accessible pore space in porous media. Experiments at the intersection of geochemistry and microfluidics have the potential to enable a step change in understanding CO2 storage and mineralization mechanisms by investigating the influence of transport on mineralization rates reported from batch reactor experiments. To study precipitation of silicate and carbonate minerals in mafic/ultramafic formations for carbon storage applications, we create models of 3D unconsolidated or sintered glass bead "grains"in glass tubes and planar cells with varied reactive inclusion "grains"of (1) mafic/ultramafic minerals including MgO, brucite, and olivine and/or (2) water-soluble seed crystals (e.g., sodium chloride, sodium acetate, copper sulfate) that are mounted in a microfluidic setup. The latter form from supersaturated brines at faster rates than geologic crystals and are used as mineralization proxies to explore crystallization dynamics in a lab microfluidic setting on a reasonable timescale. Mineralization reactions of mafic/ultramafic inclusions are slower, in part because gaseous CO2 must dissolve into the brine and the minerals must dissolve to release ions before the crystal formation can proceed. The microfluidic devices are pre-saturated with a brine of the necessary ionic concentration to promote crystallization. Then, either air or CO2 gas is injected depending on the desired reaction. This multiphase flow produces liquid-gas interfaces that influence crystallization. Flow dynamics and the habits of crystal nucleation and growth under pore-confinement in these engineered porous media are captured with time-lapse microscopy (e.g., stereomicroscope, confocal microscope, microCT) while keeping the samples intact. On select samples, cross-sections (e.g., billets and thin sections) of the samples are examined with microscopy (e.g., thin section, SEM, EDS) and bulk sample measurements (e.g., helium pycnometer, imbibition capacity) are acquired after reactive transport to quantify porosity changes with crystallization.

This presentation first details development of fabricated "lab-on-a-chip"miniature 3D synthetic rocks with varied reactive properties. The workflow enables control of porosity, permeability, microstructure, mineral composition, and accessible reactive surface area and allows for easier in-situ observations and measurements as opposed to real core experiments. Next, the presentation provides early insights on how formation of carbonates and carbonate mineralization proxies alters the microstructure of a rock and how to optimize injection strategies to maximize crystallization reaction and minimize permeability reduction. Results suggest that precipitation favors diffusion-dominated zones and that nucleation behavior and location will be influenced by liquid/gas interfaces and the presence of thin films. Results are cast in terms of dimensionless transport numbers (e.g., Capillary, Damkohler, Peclet numbers) and provide ideal benchmarks for numerical modeling and functional relationships for accelerating or inhibiting carbon mineralization within a porous media. Going forward, the 3D microfluidics setups will enable investigation of a host of coupled pore-scale alteration phenomena, including dissolution, carbonation/precipitation, and swelling/shrinking as a function of fluid flow/injection rates, pH, total dissolved solids, temperature, and pressure.

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References

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