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From batch to flow experiments: advancing basalt carbonatation for in-situ CO_2 sequestration at temperature below 120°C

MR. VALENTIN FORTIER[1], **DR. PIERRE-ALEXANDRE TEBOUL**[2], IMANE GUETNI[2], ALEXANDRA KLIMENKO[2], MARGUERITE GODARD[1], RICHARD LEPROVOST[1], PHILIPPE GOUZE[1] AND SYLVAIN CALASSOU[2]

[1]Géosciences Montpellier, CNRS, Univ. Montpellier
[2]TotalEnergies

Storing CO_2 in mafic rocks by carbonate mineralization is a promising solution for long-term sequestration. While basalt carbonatation is chemically feasible, there are few experimental studies integrating CO_2-rich seawater transport, mineral dissolution, and carbonate precipitation below 120 °C without using alkaline bases or depressurization. Such studies are essential to fully understand the chemical pathway to be modelled for low temperatures pilot-scale applications. To investigate reaction kinetics, both static batch and flow-through experiments were conducted.

Batch tests at 70 °C and 120 °C used crystalline basalt crushed at 80-150 µm and synthetic seawater under a CO_2 partial pressure of 20 bar. After four months, Fe-Mg carbonates formed. Temperature and reactive surfaces mainly controlled dissolution rates. Experiments with seawater and those with desulfated seawater produced similar results.

A slim-tube apparatus was designed to evaluate transport limitations during carbonation. The apparatus consisted of six titanium tubes with a combined length of 60 cm and an internal diameter of 3.85 mm. The tubes were packed with crystalline basalt powder, achieving ~28% porosity and initial permeability of ~50 mD. Synthetic desulfated seawater was injected at 1.5 µl/min (interstitial velocity of 0.7 m/day) under conditions of 120 °C, 100 bar total pressure, and 50 bar PCO_2. During a 41-day experiment (45 pore volumes), spatial variations in mineralogical changes were observed. SEM-EDS analyses reveal olivine dissolution with characteristic skeletonized structures. Pyroxenes and plagioclase remained stable. Secondary Fe-Mg carbonate crystals formed within primary pore spaces; these crystals were similar in morphology and size to those observed in batch experiments. The absence of precipitation in secondary porosity suggests that pore-scale transport processes and local saturation conditions control nucleation site selection.

Dynamic experiments show that basalt carbonation is feasible at 120 °C under flow, but the detection of calcium in carbonates suggests more complex ion exchange in pores than the batch experiments indicate. Numerical simulations confirm the kinetic rate values found in common databases, yet they also emphasize the importance of more accurately identifying effective reactive surfaces. Ongoing static and dynamic experiments at 45 °C aim to clarify how temperature influences these transport-limited reactions under conditions relevant for pilot-scale applications.