

The Potential of Ca-Bearing Zeolites in CO₂ Mineralization

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Abstract

Although there are many parts of the world where there are saline aquifers or depleted hydrocarbon reservoirs available for large-scale subsurface carbon storage, these may not be present in other areas near major point sources. In such cases, the mineralization of carbon injected into mafic or ultramafic rocks may provide an alternative. Carbon mineralization has been successful in fresh basalts. More common are basalts that have been hydrothermally altered over time by their interaction with warm waters. The most common Ca-bearing silicate alteration phase of basalts altered at temperatures less than 200 oC are zeolite phases. Zeolites are noted for rapidly exchanging their interstitial cations to the aqueous solution. This process suggests zeolite-bearing altered basalts may be a favorable target for subsurface mineral carbonation efforts. This study was designed to assess this possibility.

Closed system batch reactor experiments were conducted at 60°C and 1 bar using stilbite, a Ca-bearing zeolite member. Each reactor contained 0.3 g of ground stilbite and 30 g of a 0.1 mol/kg sodium carbonate solution with a pH of 11.48. The fluids and solids of each reactor were collected and analyzed at the end of each reactor's preselected duration.

Stilbite dissolved immediately at the start of the experiment provoking the precipitation of calcium carbonate. SEM images show the presence of extensive calcite crystals suggesting rapid and efficient carbonation of stilbite. The total CO₂ concentration of the recovered solids was measured by TGA, and the amount of carbonation was quantified at an average of 5% of the original stilbite mass. This indicates that almost all the calcium in the stilbite was released during the carbonation process. Corresponding mixed-flow reactor experiments demonstrate the preferential release rate of calcium of this mineral at 60 oC and a function of pH.

Measured reaction rates have been used in reactive transport calculations to assess the efficiency of zeolite carbonation in the subsurface. The results of these experiments and calculations indicate that Ca-bearing zeolites strongly favor the fixation of injected CO₂ through the formation of calcite and aragonite. This result suggests the targeting of subsurface carbon mineral storage efforts into zeolites.