

Reaction products and evolution of permeability during carbon sequestration in fractures of unaltered and serpentinized basalt

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Basalt rocks contain mafic minerals that will react with CO₂-acidified waters and release metal cations (M²⁺) into solution that may combine with dissolved carbonate ions to form stable carbonate minerals. Such mineral carbonation provides long-term secure CO₂ storage, but as these reactions occur along fractures pathways, they may alter fracture permeability, and ultimately the viability of fractured basalts as CO₂ storage reservoirs.

This study combines detailed petrographic characterization with a series of static and flow-through experiments of CO₂-saturated waters in contact with serpentinized and unserpentinized fractured basalt cores in an effort to understand the role of reaction products in altering fracture permeability. Saw-cut fracture surfaces were either milled or roughened with sand paper, in order to enable flow under confining stress. Experimental temperatures ranged between 45-150°C and a total CO₂ pressure of 10 MPa was used throughout. Flow rates for the core-flooding studies varied between 3-5 ml/hr, with the cores under 20 MPa confining stress. Static experiments were conducted in high-pressure batch reactors to evaluate dissolution-precipitation in zones with transport limited to diffusion. X-ray computed tomography was used to follow fracture microstructure evolution, effluent chemistry was monitored to evaluate mineral reactivity, and fracture surfaces after reaction were examined by Raman spectroscopy, optical microscopy, and total carbon analysis.

Calcium carbonate precipitation was observed in the static experiments, while continued dissolution of reactive mineral phases was observed in the core flooding studies. Carbonate minerals were not uniformly distributed with depth into the static fractures, and the extent and morphology of precipitates were different for the two basalts. These results indicate the importance of fluid transport conditions in generating regions that may favour carbonate precipitation due to the development of pH and (M²⁺) concentration gradients.