

Targeted control of subsurface permeability using mineral carbonation reactions

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The recent boom in oil and gas production from shale formations has led to wide scale concern about risks to overlying freshwater aquifers. Hydraulic fracturing and vertical drilling in these formations could result in seepage and leakage that would impact drinking water resources far from production wells. Here we propose the use of mineral carbonation reactions to selectively seal fractured shale formations and control leakage. Mineral carbonation reactions between CO₂ and magnesium or calcium bearing silicates at high pressures and temperatures in the aqueous phase have been studied extensively as an *ex situ* method for mineralization of CO₂ from power plants. In this work we evaluated the idea of deploying these same reactions *in situ* shale formations under reservoir temperature and pressure conditions. The cation donor could be introduced to the formation as a proppant and the CO₂ could be pumped in using the same infrastructure used for gas production. This approach could be used during production to control leaks or following production to seal the entire formation. The ancillary benefit of this approach is that it would permanently bury significant amounts of CO₂.

Batch and flow experiments were carried out to characterize physicochemical properties of these reactions under the pressure and temperature conditions of the Marcellus Shale in the United States. Conversion efficiency for wollastonite (CaSiO₃) and CO₂ in the presence of ground shale particles was evaluated using semi-quantitative XRD and found to be over 50% after 24 hours. Scanning Electron Microscopy (SEM) reveals that, following reaction, the shale particle surfaces are covered with precipitated calcite crystals ranging in size from 1 to 5 μm. X-ray computed tomography (xCT) characterization of packed column experiments of shale particles in the presence of CaSiO₃ showed clear changes to the pore size and structure. The precipitated CaCO₃ formed on the surface of the shale particles in a manner likely to decrease porosity and fluid conductivity.