Insights into Silicate Carbonation Processes in Water-Bearing Supercritical CO2 Fluids

Abstract

Long-term geologic storage of carbon dioxide (CO2) is considered an integral part to moderating CO2 concentrations in the atmosphere and subsequently minimizing effects of global climate change. Although subsurface injection of CO2 is common place in certain industries, deployment at the scale required for emission reduction is unprecedented and therefore requires a high degree of predictability. Accurately modeling geochemical processes in the subsurface requires experimental derived data for mineral reactions occurring between the CO2, water, and rocks. Most work in this area has focused on aqueous-dominated systems in which dissolved CO2 reacts to form crystalline carbonate minerals. Comparatively little laboratory research has been conducted on reactions occurring between minerals in the host rock and the wet supercritical fluid.
phase. In this work, we studied the carbonation of wollastonite 
[CaSiO₃] exposed to variably hydrated supercritical CO₂ (scCO₂) at a 
range of temperatures (50, 55 and 70 °C) and pressures (90,120 and 
160 bar) that simulate conditions in geologic repositories. Mineral 
transformation reactions were followed by three novel in situ high 
pressure techniques, including x-ray diffraction that tracked the rate 
and extents of wollastonite conversion to calcite. Increased 
dissolved water concentrations in the supercritical CO₂ resulted in 
increased silicate carbonation approaching ~50