Nanoscale basis of CO_2-brine multiphase flow and geochemistry in carbon dioxide storage repositories

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Abstract

Geological carbon sequestration (GCS) is an emerging technology that could potentially offset up to 12 % of global CO_2 emissions by 2020 and up to 45 % by 2050. Pilot studies over durations of up to 15 years have established the feasibility of injecting CO_2 in geological formations in quantities of up to several Mt per year. Commercial-scale implementation of GCS will require demonstration that much larger plumes of buoyant CO_2 (hundreds of Mt) can be stored with negligible leakage for durations of thousands of years, as required to achieve permanent CO_2 sequestration through naturally occurring rock weathering reactions. Such demonstrations are routinely carried out with field-scale geophysical models, but model predictions are sensitive to poorly constrained input parameters such as capillary pressure and relative permeability curves and the rate constants of geochemical reactions. Fundamental studies of CO_2-brine multiphase flow and geochemical properties in reservoir and caprocks can play an important role in informing the parameterization of these field-scale models. Here, we describe how molecular- and pore-scale studies carried out within the Center for Nanoscale Control of Geologic CO_2 (an Energy Frontiers Research Center supported by the U.S. Department of Energy) provide fundamental insights into the properties of confined fluids (CO_2 and brine in caprock nanopores, residual water films coating mineral surfaces in pores invaded by CO_2) and the manner in which wetting properties (wetting angles, capillary entry pressures) arise from molecular scale phenomena. We focus on studies that combine macroscopic (gravimetric adsorption, vibrating tube densimetry, goniometric contact angle) and molecular scale experiments (neutron scattering and x-ray fluorescence) with molecular dynamics simulation approaches to elucidate the geochemical properties of CO_2-brine-mineral systems at conditions relevant to CO_2 sequestration in terrestrial brine aquifers (high temperatures, pressures, and salinities).