SAND2018-11963C

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Chemical effects on fracture in calcite single crystals and in carbonate-rich shale

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Carbonates, such as calcite ($CaCO_3$) and dolomite $CaMg(CO_3)_2$, are abundant minerals in the Earth's crust, common in sedimentary formations. These minerals are susceptible to subcritical chemically-driven fracturing. Understanding chemically-driven fracture is necessary for carbon dioxide (CO_2) and nuclear waste storage, resource extraction, and predicting earthquakes. We examined chemical processes controlling subcritical fracture in calcite single crystals, and in carbonate-rich shale caprock, following its reaction with CO_2.

We developed a novel approach for quantifying the growth rate of subcritical fracture in calcite. We used a Vickers-geometry indenter tip to create identical micron-scale fractures on the calcite (100) surface. These pre-fractured samples were submerged in a series of aqueous fluids and an optical microscope was used to track the fracture growth *in-situ*. The fracture propagation rate depended on the type of aqueous fluid, and varied from 1.6×10^{-8} m s^{-1} to 2.4×10^{-10} m s^{-1}. The fracture growth rate correlated with the strength of the calcium-ligand complex, and did not depend on the measured dissolution rate of calcite or trends in zeta-potential. We conclude that the growth of subcritical fracture in calcite is limited by chemical complexation reactions at the fracture tip.

To examine the coupled chemical-mechanical effects in calcite- and dolomite-rich sedimentary rock we used Mancos Shale. We quantified the changes in mineralogy and micromechanical properties before and after reacting this shale with CO_2. The mineralogical changes depended on the pressure of CO_2 during shale exposure to the CO_2-brine mixtures. Dedolomitization (replacement of dolomite by calcite and gypsum) was observed in the reactors pressurized with 100 psi of CO_2, while at the higher pressure (2,500 psi of CO_2), the complete dissolution of calcite, partial dissolution of dolomite, and precipitation of magnesite and anhydrite were observed. The mechanical weakening of shale was localized: for dolomite-rich laminae a decrease of up to 50 % in scratch toughness was observed, while the quartz-calcite-rich laminae did not undergo mechanical degradation.

Sandia National Laboratories is a multimission laboratory managed and operated by National Technology and Engineering Solutions of Sandia, LLC., a wholly owned subsidiary of Honeywell International, Inc., for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-NA-0003525.