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Can a fractured caprock self-heal?

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ABSTRACT

The ability of geologic seals to prevent leakage of fluids injected into the deep subsurface is critical for mitigating risks associated with greenhouse-gas sequestration and natural-gas production. Fractures caused by tectonic or injection-induced stresses create potential leakage pathways that may be further enhanced by mineral dissolution. We present results from reactive-flow experiments in fractured caprock (dolomitic anhydrite), where additional dissolution occurs in the rock matrix adjacent to the fracture surfaces. Preferential dissolution of anhydrite left a compacted layer of dolomite in the fractures. At lower flow rate, rock-fluid reactions proceeded to near equilibrium within the fracture with preferential flow paths persisting over the 6-month duration of the experiment and a negligible change in permeability. At higher flow rate, permeability decreased by a dramatic two orders of magnitude. This laboratory-scale observation of self-healing argues against the likelihood of runaway permeability growth in fractured agrous caprock composed of minerals with different solubilities and reaction kinetics. However, scaling arguments suggest that at larger length scales this self-healing process may be offset by the formation of dissolution channels. Our results have relevance beyond the greenhouse-gas sequestration problem. Chemical disequilibrium at waste injection sites and in hydrothermal reservoirs will lead to reactive flows that may also significantly alter formation permeability.

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1. Introduction

Injection of fluids into deep saline aquifers or depleted oil and gas reservoirs is a promising strategy for CO₂ sequestration (Hitchon et al., 1999; Bachu, 2000; Metz et al., 2005; Kumar et al., 2005) and waste fluids disposal (Tsang et al., 2008). However, fluid injection increases pore pressure in the targeted geologic formation, creating vertical pressure gradients that can drive flow towards the surface. Successful long-term isolation of injected fluids requires geologic seals, or caprocks, with large vertical extent or very low permeability and ideally both. Lab-scale measurements of caprock permeability provide a means for predicting leakage rates through intact caprock. But, preexisting fractures in the caprock layer or fractures induced by the injection process can provide flow paths that are orders-of-magnitude more conductive than the intact rock.

Fracture permeability is influenced by the roughness of the fracture surfaces (e.g., Brown, 1987), the degree of mismatch (or shear displacement) between the surfaces (e.g., Wang et al., 1988; Mallikamas and Rajaram, 2005), the stress applied to the

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fracture (e.g., Cook, 1992; Durham and Bonner, 1994), and the gouge created during fracturing events (e.g., Faoro et al., 2009; Elkhoury et al., 2011). Predicting the potential for fractures to provide leakage pathways is further complicated by the possibility for fracture permeability to change with time due to injectioninduced perturbations in stress and fluid chemistry. Stress perturbations lead to mechanical deformation and fracture propagation that can increase or decrease formation permeability (Bandis et al., 1983; Liu, 2005; Elkhoury et al., 2006, 2011; Candela et al., 2011; Renard et al., 2012; Mason et al., 2013). Chemical disequilibrium between fluids and resident minerals leads to dissolution or precipitation that further alter fracture porosity and permeability (Lowell et al., 1993; Durham et al., 2001; Berkowitz, 2002; Chaudhuri et al., 2008; Wigand et al., 2009; Smith et al., 2013; Mason et al., 2013; Noiriel et al., 2013; Elkhoury et al., 2013).

The relative magnitudes of the timescales associated with advection through a fracture (τ_a) and reactions at mineral surfaces (τ_r) determines the nature of fracture permeability evolution (Fig. S1). When thermodynamics favor mineral dissolution and the residence time is long relative to reaction time scales (i.e., $\tau_a/\tau_r \gg 1$), dissolution-induced aperture growth occurs predominantly along preferential flow paths. The resulting reaction-front instabilities create the potential for runaway permeability increases (Durham et al., 2001; Detwiler et al., 2003; Elkhoury et al., 2013).

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Conversely, at higher flow rates ($\tau_a/\tau_r \ll 1$) dissolution occurs more uniformly throughout the fracture (Elkhoury et al., 2013) and may even be fastest in the smallest aperture regions (Detwiler et al., 2003). Furthermore, computational simulations of reactive alteration show that, because residence times within a fracture depend upon the fluid velocity and fracture length, at higher flow rates reaction instabilities can still occur, but at length scales larger than typical laboratory experiments (Szymczak and Ladd, 2011; Elkhoury et al., 2013).

Under reservoir conditions, both pore pressure and fluid chemistry perturbations are likely. Laboratory-scale experiments aimed at quantifying fracture permeability evolution due to coupled deformation and dissolution demonstrate a confounding range of behaviors: increased permeability (Elkhoury et al., 2013), decreased permeability (Ellis et al., 2013), and nonmonotonic changes in permeability (Polak et al., 2004). Due to either experimental constraints or expediency these studies often involve conditions that differ from those encountered in the reservoir. For example it is difficult to accurately measure small differential pressures in experiments carried out at pore pressures of $O(10^7 \text{ Pa})$, so differential pressures are often $O(10^5 \text{ Pa})$ or larger, resulting in pressure gradients of 10s or even 100s of MPa/m (head gradients from 10^3 to 10⁴). However, it is difficult to imagine the field conditions under which such large pressure gradients would be sustained. This results in experiments where $\tau_a/\tau_r \ll 1$ and dissolution occurs relatively uniformly throughout the fracture (regardless of aperture variability) and may be fastest in small aperture regions. The resulting increasing stresses at eroding contacts leads to pressure solution (Yasuhara et al., 2004) or brittle failure (Detwiler, 2008) and subsequent permeability loss.

The choice of fluid chemistry also influences the evolution of fracture dissolution. When studying the influence of CO₂ injection, a natural choice for injected fluid is a brine in equilibrium with host minerals equilibrated with CO₂ at reservoir pressure, which leads to influent fluids with $pH \lesssim 4$ (Elkhoury et al., 2013; Smith et al., 2013). However, fluids leaking through a fractured caprock are likely to exhibit a wide range of compositions. Monitoring of reservoir fluids over 10 yr of intermittent water and CO_2 injection at the Weyburn site show 5 < pH < 6 (Burrowes and Gilboy, 2001). This is due to a combination of buffering due to dissolution of CaCO₃ in the reservoir, mixing of injected water with CO₂-equilibrated brine, and propagation of increased pressures far beyond CO₂ plumes. This suggests that fluids expected to leak through a caprock will likely exhibit a wide range of compositions with near-equilibrium with CO₂ at reservoir pressure a low-pH end member. Using such low-pH fluids in laboratory experiments increases reaction rates with carbonates and decreases τ_r , thus shortening the required duration of experiments. However, it also has implications on permeability evolution as it will increase the value of τ_a/τ_r , leading to the formation of dissolution channels and increased permeability observed in some experiments (Elkhoury et al., 2013).

Despite the simplifications inherent in laboratory-scale studies, such experiments provide invaluable insights into the competing processes that lead to permeability alteration. However, the relevant simplifications must be placed in the proper context; for example, there is mounting evidence that, though initial fracture aperture variability plays an important role in establishing the initial fracture permeability, it plays a second-order role in controlling the evolution of the fracture aperture relative to the transport and reaction timescales (Szymczak and Ladd, 2011; Elkhoury et al., 2013).

In this study, we performed reactive flow experiments in fractured cores from a dolomitic anhydrite $(CaMg(CaO_3)_2, CaSO_4)$ caprock. We explored the relative importance of advective and reactive timescales during flow of a reactive fluid (water) in a mineral assemblage commonly found in caprocks overlying oil and gas reservoirs. We present results from two experiments in fractured cores with flow rates (and τ_a) differing by two orders of magnitude to determine the potential for caprock selfhealing under this range of parameters. We chose the duration of both experiments so that an equal amount of mass dissolved from each sample, which required the low-flow-rate experiment to run for 6 months. Unlike previous experiments with very reactive fluids in carbonate cores, our experiments exhibited measurable dissolution within the porous matrix adjacent to the fracture suggesting that the diffusion timescale in the porous matrix is faster than the reaction time scale. In both experiments, dissolution weakened the porous matrix adjacent to the fracture, which led to a two-order-of-magnitude permeability decrease at high flow rate and negligible permeability change at low flow rate.

2. Experimental description

To minimize mineralogical heterogeneity between samples, we took both samples from a single 15-cm-long, 9-cm-diameter core from the evaporite that forms the first layer of caprock overlying the Weyburn–Midale reservoir in Saskatchewan, Canada (Wilson and Monea, 2004). We drilled 3.8-cm-diameter sub-cores centered on tensile fractures within the larger core to minimize the impact of damage caused during fracturing. Sample lengths were 6.3 cm (EV1) and 4.9 cm (EV2) and the initial rock masses were 213 g and 163 g for EV1 and EV2, respectively (see Table S1 for complete details of the two experiments).

Although fractures provide preferential flow paths, the permeability of well-mated fractures subjected to a confining stress typical of reservoir conditions have permeabilities that are similar to intact cores (e.g., Durham and Bonner, 1994). For the case of the dolomitic anhydrite cap rock used in our experiments, the intact permeability is on the order of 10^{-19} m² (Smith et al., 2013). Thus, effective leakage pathways will require some shear displacement of the fracture surfaces to create the potential for sustained flow at pressure gradients consistent with those expected in the field. We increased the permeability in our well-mated tensile fractures by shear offsetting the fracture surfaces prior to mating by 0.051 and 0.038 cm for EV1 and EV2, respectively. We placed shims between the endcaps and rock sample (Fig. S2) to maintain these displacements when applying confining stress to the samples. These fractures differ from shear fractures induced under significant normal stress due to the absence of gouge; however, they provide the ability to carefully characterize the surface and fracture geometry prior to flow-through experiments.

Under reservoir conditions, in regions not directly overlying the CO_2 plume, dissolution will be relatively slow due to high ionic concentrations of typical reservoir fluids. To accelerate the dissolution and facilitate lab-scale experiments at reasonable timescales (months), we used de-ionized water as the reactive fluid. Furthermore, speciation calculations using PHREEQC (Parkhurst and Appelo, 1999), show that flowing water instead of CO_2 -equilibrated brine increases the solubility of anhydrite, and only significantly influences the solubility of dolomite relative to anhydrite at very high pCO₂ (low pH) (see Appendix A for details).

To begin the experiments, we jacketed the fractured cores and secured them in a flow-through reactor and applied a 7 MPa confining stress (Figs. S2 and S3). A high-pressure ISCO syringe pump controlled the confining pressure, while two additional ISCO syringe pumps, connected in parallel, continuously pushed de-aired de-ionized water at a temperature of 21 °C through the fractured sample at a constant flow rate ($Q_{EV1} = 0.01$ ml/min and $Q_{EV2} = 0.6$ ml/min) with pressure controlled at the outlet (Figs. S2 and S3

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