



## Assessment of two-phase flow on the chemical alteration and sealing of leakage pathways in cemented wellbores

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### ABSTRACT

Wellbore leakage tops the list of perceived risks to the long-term geologic storage of CO<sub>2</sub>, because wells provide a direct path between the CO<sub>2</sub> storage reservoir and the atmosphere. In this paper, we have coupled a two-phase flow model with our original framework that combined models for reactive transport of carbonated brine, geochemistry of reacting cement, and geomechanics to predict the permeability evolution of cement fractures. This addition makes the framework suitable for field conditions in geological storage sites, permitting simulation of contact between cement and mixtures of brine and supercritical CO<sub>2</sub>.

Due to lack of conclusive experimental data, we tried both linear and Corey relative permeability models to simulate flow of the two phases in cement fractures. The model also includes two options to account for the inconsistent experimental observations regarding cement reactivity with two-phase CO<sub>2</sub>-brine mixtures. One option assumes that the reactive surface area is independent of the brine saturation and the second option assumes that the reactive surface area is proportional to the brine saturation.

We have applied the model to predict the extent of cement alteration, the conditions under which fractures seal, the time it takes to seal a fracture, and the leakage rates of CO<sub>2</sub> and brine when damage zones in the wellbore are exposed to two-phase CO<sub>2</sub>-brine mixtures. Initial brine residence time and the initial fracture aperture are critical parameters that affect the fracture sealing behavior. We also evaluated the importance of the model assumptions regarding relative permeability and cement reactivity. Our results illustrate the need to understand how mixtures of carbon dioxide and brine flow through fractures and react with cement to make reasonable predictions regarding well integrity. For example, a reduction in the cement reactivity with two-phase CO<sub>2</sub>-brine mixture can not only significantly increase the sealing time for fractures but may also prevent fracture sealing.

### 1. Introduction

Depleted oil and gas reservoirs are prime candidates for injection of supercritical CO<sub>2</sub> for carbon dioxide storage (IPCC, 2005). However, sites with a history of oil and gas exploration and production activities typically contain abandoned wells, which may compromise the integrity of the storage site. While such abandoned wells are plugged with cement to prevent leakage, poor cement bonding or failure due to stresses (Zhang and Bachu, 2011; Nygaard et al., 2014) can create fractures in the cement, or at the cement-formation or cement-casing interfaces. These damage zones may allow carbon dioxide to leak from the reservoir, bringing single-phase and two-phase mixtures of supercritical/gaseous CO<sub>2</sub> and brine in contact with the wellbore cement. The purpose of this study is to understand the behavior of these multiphase mixtures and their interaction with wellbore cement.

Reactions between cement and carbonated brine alter cement's chemical, mechanical, and hydraulic properties. Under different circumstances these coupled processes can either hamper or even improve the cement's ability to seal the well. While several experiments have shown that reactions between CO<sub>2</sub>-saturated brine and cement result in altered cement layers (Zhang and Bachu, 2011; Carey, 2013; Carroll et al., 2016), it is unclear how cement reacts with a two-phase mixture of brine and supercritical CO<sub>2</sub>, or with brine-saturated supercritical CO<sub>2</sub> (*i.e.* "wet" supercritical CO<sub>2</sub>). There have been insufficient experimental studies to determine how cement fractures respond to flows involving two-phase mixtures of brine and CO<sub>2</sub>. Several batch experimental studies have attempted to characterize the interactions between cement and supercritical CO<sub>2</sub> but their results have been inconclusive. While some studies have shown that cement exposed to wet supercritical CO<sub>2</sub> results in the formation of reaction fronts within the cement (Barlet-

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Gouédard et al., 2007; Rimmelé et al., 2008; Fabbri et al., 2009; Mito et al., 2015), others have observed uniform carbonation instead (Kutchko et al., 2008; Jung and Um, 2013). In the latter case, Kutchko et al. (2008) attributed the absence of reaction fronts to the lack of water. With regards to the rates of reactions, some studies showed no significant difference in the rate of alteration of cement upon exposure to carbonated brine or wet supercritical CO<sub>2</sub> (Barlet-Gouédard et al., 2007; Rimmelé et al., 2008; Lesti et al., 2013). Others have observed faster (Mito et al., 2015) or slower (Kutchko et al., 2008; Jung and Um, 2013) rates of alteration with wet supercritical CO<sub>2</sub> compared to carbonated brine. Kutchko et al. (2008) attributed the slow alteration rate with wet supercritical CO<sub>2</sub> to the lack of an aqueous phase to facilitate the diffusion of ions. Conversely, Mito et al. (2015) attributed the fast alteration rate with wet supercritical CO<sub>2</sub> to the lack of a diffusion barrier in the absence of water. In a different set of experiments, where wet and oven-dried cement samples were exposed to wet supercritical CO<sub>2</sub>, Fabbri et al. (2009) showed that reaction fronts are present when wet cement is exposed to wet supercritical CO<sub>2</sub> and are absent when the cement was dried. They also found that the rate of degradation was lower in wet cement compared to dry cement, which they attributed to the diffusion barrier created by water in the wet cement. Although water plays a key role in the alteration of cement by carbon dioxide, the conflicting experimental observations make it difficult to identify the conditions under which water facilitates rather than inhibits these reactions. Differences in both the preparation and composition of cement samples, the details of which are often difficult to identify in the published literature, further muddles this issue.

It is essential to evaluate the behavior of mixtures of supercritical CO<sub>2</sub> and brine to accurately quantify the risk of carbon dioxide and brine leaking from a storage reservoir through damage in well cement. Differences in density, viscosity and relative permeability of the two phases have significant impacts on the flow rates of the individual phases in multiphase flow.

Over the last few years, we have developed an experimentally calibrated framework that couples reactive transport of carbonated brine, geochemistry of reacting cement, and geomechanics to predict the permeability evolution of leakage paths in cement exposed to carbonated brine (Walsh et al., 2013, 2014a,b; Iyer et al., 2017). In this paper, we extend our model to include two-phase flow of supercritical CO<sub>2</sub> and brine through leakage paths to make the model applicable to field conditions. We use the model to evaluate how two-phase flow impacts the chemical alteration of cement compared to carbonated brine. We also use the model to predict carbon dioxide and brine leakage rates through damage zones. The assumptions adopted for the relative permeability of CO<sub>2</sub> and brine flowing in fractures, and their reactivity with cement have a significant impact on the model's predictions. In light of the current lack of consensus regarding both of these factors, we investigate extremes in both cases in order to evaluate their impact on the chemical alteration of cement and the leakage rate of carbon dioxide and brine.

## 2. Model description

The present model builds on earlier work done to simulate the interaction between well cement and CO<sub>2</sub>-saturated brine flowing through a fracture (Walsh et al., 2013, 2014a,b; Iyer et al., 2017). The original framework coupled a single-phase flow model for brine flow through a fracture, a reactive-transport model to predict the brine concentrations, a geochemical model for the CO<sub>2</sub>-cement reactions, and a mechanical model to predict the change in fracture aperture due to the chemically altered cement. The models are implemented in the GEOS multi-physics framework developed at Lawrence Livermore National Laboratory (Settgast et al., 2017). Here, we describe how this framework can be extended to include a multiphase flow model to capture the flow of two-phase mixtures of brine and supercritical CO<sub>2</sub>. We also describe the assumptions made with regards to the uncertainty

surrounding (i) the relative permeability model for two-phase flow through fractures, and (ii) the impact of saturation on cement alteration rate.

### 2.1. Two-phase flow model

To model the two-phase flow of brine and carbon dioxide through a fracture, we first perform a mass balance for both brine and carbon dioxide. A depth-averaged mass balance for brine yields:

$$\frac{\partial}{\partial t}(b\rho_w\phi S_w) + \nabla \cdot (b\rho_w \mathbf{v}_w) = 0, \quad (1)$$

where the subscript *w* denotes the brine phase, *b* is the fracture aperture,  $\rho_i$  is the density of phase *i*,  $\phi$  is the porosity of the medium, which is 1 in the case of a fracture,  $S_i$  is the saturation of phase *i*, and  $\mathbf{v}_i$  is the superficial velocity of phase *i*. In this manuscript, our analysis is confined to one-dimensional flows for which the depth-averaged velocity vector can be represented as a scalar. Nevertheless, the approach and the code itself is applicable to flows through two-dimensional fractures, and consequently we leave the depth-averaged velocity as a vector. In addition, we have neglected the mass of brine diffusing in or out of the cement or solubilizing in the CO<sub>2</sub> phase, as these terms are expected to be significant only when the brine saturation is very low.

As carbon dioxide flows through the fracture, some of it is dissolved in the brine which subsequently diffuses into the cement and reacts with it. A mass balance of the carbon dioxide present in the CO<sub>2</sub> and the brine phases yields

$$\frac{\partial}{\partial t}(b\phi(\rho_{g,w}S_w + \rho_g(1 - S_w))) + \nabla \cdot (b(\rho_{g,w}\mathbf{v}_w + \rho_g\mathbf{v}_g)) + R_C = 0, \quad (2)$$

where the subscript *g* denotes the CO<sub>2</sub> phase,  $\rho_{g,w}$  is the solubility of carbon dioxide in the brine phase, and  $R_C$  is the rate at which carbon dioxide is consumed in the reactions with cement.

The velocities of the two phases in Eqs. (1) and (2) are calculated using the extension of Darcy's law for multiphase flow:

$$\mathbf{v}_i = -\frac{kk_{r,i}}{\mu_i}(\nabla p_i - \rho_i \mathbf{g}), \quad (3)$$

where the subscript *i* denotes the brine and CO<sub>2</sub> phases, *k* is the permeability of the fracture,  $k_{r,i}$  is the relative permeability of phase *i*,  $\mu_i$  is the viscosity of phase *i*,  $p_i$  is the pressure of phase *i*, and  $\mathbf{g}$  is the gravity vector. As carbon dioxide and brine are immiscible, the two phases will have different pressures due to capillary forces:

$$p_w - p_g = \frac{2\gamma \cos \theta}{r_c}, \quad (4)$$

where the capillary pressure,  $p_w - p_g$ , is a function of the wetting angle  $\theta$ , the surface tension between the phases  $\gamma$ , and the radius of curvature  $r_c$ . In a fracture the radius of curvature is well approximated as half the aperture ( $r_c = b/2$ ), hence the smaller the aperture the larger the capillary pressure. Mixtures of supercritical CO<sub>2</sub> and brine have an interfacial tension of around 30 mN/m (Chalbaud et al., 2009). An aperture of 50  $\mu\text{m}$  and a wetting angle of 30° (Saraji et al., 2013) gives a capillary pressure of 2 kPa. As we are applying this formulation to fractures with an aperture of 50–500  $\mu\text{m}$  at average pressures around 10 MPa, we have assumed that the capillary pressure is small and have neglected it. Therefore, the pressure in both phases is the same and is denoted by *p*.

The permeability of the fracture in Eq. (3) is calculated using the following approximation (Witherspoon et al., 1980; Zimmerman and Bodvarsson, 1996):

$$k = \frac{b^2}{12}. \quad (5)$$

Modeling two phase flow in fractures is still an active area of research. In the microfluidics literature two-phase flow is modeled by

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