International Journal of Greenhouse Gas Control 47 (2016) 25–37

Contents lists available at ScienceDirect

International Journal of Greenhouse Gas Control

journal homepage: www.elsevier.com/locate/ijggc

Fracture opening or self-sealing: Critical residence time as a unifying parameter for cement–CO_2–brine interactions

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ARTICLE INFO

Article history:
Received 30 June 2015
Received in revised form
21 November 2015
Accepted 14 January 2016
Available online 6 February 2016

Keywords:
Well integrity
Carbon storage
Reactive transport modeling
Cement fracture alteration
CO_2 leakage

ABSTRACT

Understanding long-term property evolution of cement fractures is essential for assessing well integrity during geological carbon sequestration (GCS). Cement fractures represent preferential leakage pathways in abandoned wells upon exposure to CO_2-rich fluid. Contrasting self-sealing and fracture opening behavior have been observed while a unifying framework is still missing. Here we developed a process-based reactive transport model that explicitly simulates flow and multi-component reactive transport in fractured cement by reproducing experimental observation of sharp flow rate reduction during exposure to carbonated water. The simulation shows similar reaction network as in diffusion-controlled systems without flow. That is, the CO_2-rich water accelerates the portlandite dissolution, releasing calcium that further reacted with carbonate to form calcite. The calibrated model was used for CO_2-flooding numerical experiments in 250 cement fractures with varying initial hydraulic aperture (b) and residence time (τ) defined as the ratio of fracture volume over flow rate. A long τ leads to slow replenishment of carbonated water, calcite precipitation, and self-sealing. The opposite occurs when τ is small with short fracture and fast flow rates. Simulation results indicate a critical residence time τ_c – the minimum τ required for self-sealing – divides the conditions that trigger the opening and self-sealing behavior. The τ_c value depends on the initial aperture size through $\tau_c = 9.8 \times 10^{-4} \times b^2 + 0.254 \times b$. Among the 250 numerical experiments, significant changes in effective permeability – self-healing or opening – typically occur within hours to a day, thus providing supporting argument for the extrapolation of short-term laboratory observation (hours to months) to long-term prediction at relevant GCS time scales (years to hundreds of years).

1. Introduction

Geological carbon sequestration (GCS) has been considered as a promising mitigation strategy to reduce anthropogenic CO_2 emission (Bachu, 2008; Solomon et al., 2007). A major concern in GCS operations is the increase in formation pressure induced by CO_2 injection, leading to potential migration of the injected CO_2 and/or formation brine into the overlying formation or to the surface (Benson and Cole, 2008; Oldenburg et al., 2009; White et al., 2003). Natural analog studies, as well as experimental and modeling work have shown the potential of CO_2 migration into shallow aquifers (Carroll et al., 2014; Keating et al., 2013; Smith et al., 2013), which can result in groundwater acidification (Apps et al., 2010; Carroll et al., 2009), heavy metal mobilization (Frye et al., 2012; Lawter et al., 2015; Zheng et al., 2009), and mineral dissolution (Wang and Jaffe, 2004), all leading to detrimental impacts on water quality. The dissolution of CO_2 acidifies brines in deep saline aquifers, potentially dissolving solid materials and increasing in situ permeability (Li et al., 2006; Qiao et al., 2015). These processes raise the risks of leakage through existing interfaces, fractures, and faults (Gaus, 2010; Keating et al., 2013). Regions with suitable conditions for GCS are commonly characterized by a large number of abandoned wells (Gasda et al., 2004; Nicot, 2009). Portlandite, an important component of cement, is commonly used in well construction and can react with CO_2-saturated brine, thus changing the structural integrity and transport properties of the well cement (Carey et al., 2007; Loizzo et al., 2011). It is essential to thoroughly understand the transport property evolution of portlandite-based cement upon

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http://dx.doi.org/10.1016/j.ijggc.2016.01.024
1750-5836/

exposure to CO_2-rich brine to predict leakage possibilities more accurately.

Extensive modeling and experimental studies in recent years have characterized the evolution of well cement exposed to CO_2-saturated brine (Duguid et al., 2011; Duguid and Scherer, 2010; Fabbri et al., 2009; Huerta et al., 2013; Kutchko et al., 2007; Mason et al., 2013). The high acidity of CO_2-enriched brine leads to the dissolution of calcium bearing minerals, which increases pH and causes carbonate precipitation (typically calcite) (Huerta et al., 2015; Kutchko et al., 2007). Under both diffusion-controlled and dynamic flow conditions, the chemical degradation of cement (decalcification) have led the formation of distinct zones: the amorphous silica zone in direct contact with acidic brine, followed by a calcium carbonate zone, a portlandite depleted zone, and the inner unaltered zone (Kutchko et al., 2007). While the cement degradation mechanisms have now been well known, its impacts on the long-term cement properties and therefore well integrity are not well understood. Although it has been suggested that cement degradation has relatively minor impacts on well integrity due to the slow displacement of the reaction front (Carey et al., 2007; Kutchko et al., 2007), recent experimental observations and long-term simulation has shown that the degraded zone can potentially create preferential pathways, depending on initial cement properties (Brunet et al., 2013; Cao et al., 2013).

Recent studies have documented cement property evolution under flow-through conditions with defective or vulnerable interfaces (cement–cement fractures, cement–rock or cement–steel) continuously exposed to CO_2-rich brine. Those experiments showed either an increase (Cao et al., 2013; Huerta et al., 2013, 2015; Luquot et al., 2013; Scherer et al., 2005; Wigand et al., 2009) or decrease (Carey et al., 2010; Huerta et al., 2013, 2015; Liteanu and Spiers, 2011; Luquot et al., 2013; Mason et al., 2013; Newell and Carey, 2013; Walsh et al., 2012, 2014a) in fracture permeability. Consequently, fractures have been considered to have the potential to self-seal under some conditions while opening under other conditions. Using a composite cement–sandstone core of 76.1 mm in length with a large defect at the cement–sandstone interface, Cao et al. (2013) observed an eight-time increase in sample permeability after 8 days of exposure to carbonated brine. Under similar injection conditions, Cao et al. (2016) observed a permeability increase by three times in a cement-sandstone composite core, and a permeability decrease by half in a fractured cement core (Cao et al., 2015). Similarly, during exposure to carbonated brine, the cement surface degradation was observed resulting in an increase in the channel width (Scherer et al., 2005). In a cement core with an embedded steel rode presenting channels at the cement–steel interface, Carey et al. (2010) observed an increase in channel permeability and calcite precipitates however with no erosion. Luquot et al. (2013) observed three different outcomes in cement fractures exposed to carbonated brine. The fracture with a large aperture (30 μm) subjected to a high initial flow rate of 2 ml/min showed no permeability alteration, although three distinct zones similar to those under diffusion-controlled conditions were observed. In contrast, fractures with smaller apertures (13.6 and 2 μm) at lower flow rates (0.2 ml/min and 0.1 ml/min) were observed to have similar zonation except the absence of high porosity zone at the cement–fracture interface. The fractures subjected to medium flow rates exhibited permeability increase early on followed by a plateau with a final increase in permeability by a factor of 1.5. Similarly, Huerta et al. (2015) conducted a series of experiments with fractured cement exposed to carbonated water and reported permeability increase for short cores with large apertures and flow rates while self-sealing in long fractures with small aperture and flow rates.

It was speculated that the fast flow and the formation of a calcium depleted zone prevent significant change in permeability and maintain aperture. In contrast, low flow rates allow calcite precipitation to an extent that is sufficient for self-sealing to occur. Those mechanisms can offer an explanation for the observations where large flow/fracture apertures minimize the formation of a protective calcite deposit and permeability increase (Cao et al., 2013; Carey et al., 2010; Luquot et al., 2013; Scherer et al., 2005). Several mechanisms, however, have been proposed to explain the self-sealing behavior. Changes in mechanical strength of altered cement and the increased compressibility have been attributed to fracture "collapse" (Mason et al., 2013; Walsh et al., 2014a,b), "swelling" of the cement matrix, a lower density cement matrix (Luquot et al., 2013), or precipitation of leached calcium from the cement as calcium carbonate (calcite) inside the fracture (Huerta et al., 2013, 2015; Liteanu and Spiers, 2011; Luquot et al., 2013; Mason et al., 2013). Parameters such as fracture length, injection rate, residence time, and brine composition have been shown to influence the evolution of the fracture permeability.

In general, however, there is no framework that unifies contrasting observations under different conditions. The objective of this work is to identify and quantify key controlling parameters that can offer general guidelines for predicting conditions of self-sealing and fracture opening.

2. Methodology

Here we develop a reactive transport model constrained by experimental data of fracture permeability alteration during exposure to CO_2-saturated distilled water. Distilled water was used to maximize the dissolved CO_2 concentration and understand the base process independently of variable brine composition. The data reproduction gave a set of kinetic parameters that describe reactive transport processes during cement–CO_2–water interactions. We then predict the evolving fracture transport properties and compositional alteration at time scales well beyond the experimental duration (up to 100 days). To identify conditions leading to fracture opening or self-sealing, the calibrated model was used to carry out a total of 250 numerical experiments with initial apertures ranging from 6 to 90 μm, pressure gradients from 15.74 to 110.15 Pa/mm corresponding to initial flow from 0.0015 to 0.130 ml/min, and core lengths from 110 to 330 mm. The fracture residence time, τ (min), defined as the ratio of the initial fracture volume (ml) by the flow rates (ml/min) in the fracture, was used to integrate the effects of fracture length, aperture size, and pressure gradient.

2.1. Base case experiment

Well leakage pathways can develop at cement–host rock, cement-casing, and cement–cement (due to fractures and voids) interfaces. The focus here is the voids and fractures within the cement. While most CO_2 flooding experiments in fractured cement were conducted using constant flow rate, it is not representative of natural conditions where constant pressure gradients are typical. In this work, flow rate data from a carbonated-water flooding experiment in a fractured cement under constant pressure conditions (Huerta et al., 2015) offer constraints to develop a baseline reactive transport model. In this experiment, four fractured class H cement cores were assembled into a single fractured channel of 234 mm in length. At the confining pressure of 13.7 MPa and the temperature of 21 °C, water with a corresponding dissolved CO_2 concentration of 1.5 mol/L was injected (Duan and Sun, 2003). The flooding experiments were conducted for 11 h using a constant pressure gradient of 13.8 kPa. During the first 11 h, more than 70% reduction in flow rates was observed. In order to reestablish breakthrough, the pressure gradient was then increased to 68.9 kPa for the next 2.5 h and again to 344 kPa for the remaining 44.5 h. Despite this large increase