Chemical Geology 403 (2015) 1–12

Contents lists available at ScienceDirect

Chemical Geology

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

Numerical simulation of porosity and permeability evolution of Mount Simon sandstone under geological carbon sequestration conditions

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ARTICLE INFO

Article history:
Received 17 August 2014
Received in revised form 11 March 2015
Accepted 12 March 2015
Available online 20 March 2015

Editor: J. Fein

Keywords:
CO_2 sequestration
Sandstone formation
Reactive transport model
Permeability
Porosity

ABSTRACT

A numerical model was developed with the use of reactive transport code CrunchFlow to estimate porosity, permeability and mineral composition changes of Mount Simon sandstone under typical geological carbon sequestration conditions ($P = 23.8$ MPa and $T = 85$ °C). The model predicted a permeability decrease from 1.60 mD to 1.02 mD for the Mount Simon sandstone sample in a static batch reactor after 180 days of exposure to CO_2-saturated brine, which is consistent with measured permeability results. Model-predicted solution chemistry results were also consistent with laboratory-measured solution chemistry data. SiO_2 (am) was the primary mineral that causes permeability decrease, followed by kaolinite. Both SiO_2 (am) formation and kaolinite formation were attributed to the dissolution of quartz and feldspar. This study shows that the formation of SiO_2 (am) and kaolinite in the pore space of host rock is possible under typical CO_2 sequestration conditions. SiO_2 (am) and kaolinite precipitation at the CO_2 plume extent could reduce the permeability of host rock and improve lateral containment of free-phase CO_2, contributing to overall security of CO_2 storage.

1. Introduction

Carbon capture and sequestration (CCS), which involves capture of large quantities of carbon dioxide from flue gas and injecting carbon dioxide into permeable geological formations at depth (Gasda et al., 2008) sufficient to ensure safe long-term storage, is a promising strategy to reduce the emissions of greenhouse gas to the atmosphere. Projections by the Global CCS Institute (2011) suggest that CCS would need to account for 19% of the total CO_2 emission reduction to reach the CO_2 emission target by 2050. Deep saline aquifers have been reported to have the highest estimated CO_2 storage capacity of all candidate geologic storage targets — at least 1738 Gigatonnes of CO_2 in North America alone (The North American Carbon Storage Atlas, 2012). The interaction between injected CO_2 and host rock in deep saline aquifers for CO_2 sequestration is of interest because a) the interaction between injected CO_2 and host rock may result in porosity and permeability changes of host rock, which would directly impact CO_2 storage capacity, CO_2 plume migration, and reservoir pressure response; b) the interaction between injected CO_2 and host rock may result in permanent storage of some of injected CO_2 as carbonate minerals, and could potentially release unwanted ions (e.g., arsenic from arsenopyrite in host rock, Parthasarathy et al., 2011) to the aquifers.

There is a wide range of pressure and temperature for CO_2 sequestration conditions. The pressure can range from 7.3 MPa to 65 MPa and the temperature can range from 31 °C to 132 °C. In this study, the CO_2 sequestration conditions are defined as a pressure of 23.8 MPa and a temperature of 85 °C, which correspond to a formation depth of around 2.4 km given a pressure gradient of 10^4 Pa/m.

There have been a number of experimental studies that investigated the physical and chemical property changes of host CO_2 storage rock when exposed to CO_2 (Luquot and Gouze, 2009; Fu et al., 2009; Morris et al., 2009; Lu et al., 2011; Liu et al., 2012; Luquot et al., 2012; Carroll et al., 2012; Karamalidis et al., 2012; Soong et al., 2014). Important conclusions that can be derived from those previous studies are: a) the nature of physical and chemical changes of host rock is dependent on its mineral composition, and on the progressive dissolution of CO_2 that results in a decrease of pH in brine (Luquot and Gouze, 2009). b) CO_2–sandstone interaction usually does not result in formation of $CaCO_3$ within a short exposure period (Carroll et al., 2012; Soong et al., 2014), but slow dissolution of feldspar minerals would cause $CaCO_3$ to ultimately precipitate (Kharaka et al., 2013). More carbonate minerals would ultimately precipitate from sandstones that have abundant feldspar minerals and where the feldspars are more calcic (Kharaka et al., 2013). c) dissolution of feldspar in feldspar-rich host rock usually results in precipitation of secondary clay minerals (e.g., montmorillinite and kaolinite) (Fu et al., 2009; Liu et al., 2012; Carroll et al., 2012); and precipitation of secondary clay minerals can offset porosity and permeability changes as a result of feldspar dissolution (Soong et al., 2014).

Results from experimental studies help researchers identify key geochemical reactions and enable insights into reaction pathways of host

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http://dx.doi.org/10.1016/j.chemgeo.2015.03.014
0009-2541/

rock–CO_2 interaction under CO_2 sequestration conditions. Results from experimental studies also help develop and constrain geochemical models. However, experimental studies have limitations. First, they are limited to relatively short time scales (typically days to months), and it is difficult to predict long-term physical and chemical property changes of host rock (e.g., 30–1000 years) under CO_2 sequestration conditions. Second, rigorous experimental exploration of parameter space for controlling variables, such as the initial porosity of host rock, feldspar wt.% of host rock and aqueous CO_2 concentration after CO_2 injection, is time-consuming, expensive and sometimes not feasible in the laboratory. Computer-based reactive transport modeling overcomes these limitations, enabling extrapolation of findings from relatively short experiments to longer time scales and parameter sensitivity analyses (Gunter et al., 1997; Gaus et al., 2005; Johnson et al., 2005a, 2005b; Xu et al., 2011; Gherardi et al., 2012). There have been some studies that focused on modeling of interactions between CO_2-saturated brine and reservoir host rock (or caprock). Johnson et al. (2005a, 2005b) used a reactive transport model to assess the evolution of caprock permeability due to CO_2 injection-induced mineral alteration. Zerai et al. (2006) conducted equilibrium, path-of-reaction and kinetic modeling of CO_2–brine–mineral reactions in the Rose Run Sandstone in Ohio to investigate the factors that are likely to influence the capacity of this formation to trap injected CO_2. Xu et al. (2011) applied a generic two-dimensional (2-D) radial TOUGHREACT model to illustrate the temporal evolution and spatial distribution of the injected CO_2 and the subsequent physical and chemical changes of the reservoir host rock under CO_2 sequestration conditions. Liu et al. (2011) performed numerical simulation of large scale CO_2 injection (a million tons per year for 100 years) into Mount Simon sandstone. Gherardi et al. (2012) also applied TOUGHREACT code to predict mineral alteration in both the cement of an idealized abandoned wellbore and the caprock adjacent to the wellbore at the top of a potential CO_2 storage aquifer (Dogger aquifer) in Paris Basin, France.

Most reactive transport models to study the interaction between host rock and CO_2-saturated brine rely on kinetic and equilibrium constants that are uncertain (or maybe even unreliable). So it is prudent to compare modeling results with experimental results to help understand the implications of model predictions, and to explore sensitivity of model results to reasonable perturbation of those key parameters. One study investigated the mineral composition change of Mount Simon sandstone samples after exposure to CO_2-saturated brine with the use of reactive transport modeling, and the mineral composition results from modeling were compared with experimental data (Carroll et al., 2012). However, more studies are needed to compare modeling results with experimental data.

In this study, a numerical model that is able to simulate porosity, permeability and mineral composition changes of host rock under CO_2 storage conditions is presented. The model is compared with experimental results (permeability and solution chemistry data) from Soong et al. (2014). Both the model and the experimental study done by Soong et al. (2014) are representative of the post CO_2 injection period where the CO_2 plume is stabilized and the system pressure returns to pre-injection values. In the experiment of Soong et al., one cylindrical sandstone sample (2.54 cm in diameter × 5.08 cm in length) obtained from a depth of 1769.7 m from a well located in Vermillion County, IN was exposed to CO_2-saturated brine ($P(CO_2) = 23.8$ MPa and temperature $= 85$ °C) in a static (no flow) high pressure vessel with a Teflon liner. The exposure time was 180 days. The initial brine composition in the experiment of Soong et al. (2014) is shown in Table 1. An equilibrium calculation based on initial brine composition and exposure conditions was performed using CrunchFlow to take into account potential mineral super-saturation at elevated temperature under given exposure conditions. The brine composition after equilibrium calculation is reported in Table 1. Despite a significant concentration decrease for Ba^{2+}, H_4SiO_4 (aq) and Al^{3+} shown in Table 1, CrunchFlow does not predict any mineral precipitation when the system reaches equilibrium, which implies that minerals that have potential to precipitate (i.e., $BaSO_4$ and $K_{0.85}Al_{2.85}Si_{3.15}O_{10}(OH)_2$) have not reached super-saturated state. Therefore, the sandstone porosity change after 180 days of exposure is caused by interaction between sandstone and brine, not precipitation of minerals from super-saturated brine. More details about the experiment can be found in Soong et al. (2014).

Table 1
Brine composition reported by Soong et al. (2014) and brine composition used in the model, which takes account of mineral super-saturation at elevated temperature and injection of CO_2.

Brine composition in Soong et al. (2014) (measured at 25 °C; before injection of CO_2) Unit: mol/kg	Brine composition used in the model (calculated at 85 °C; after injection of CO_2) Unit: mol/kg
pH = 5.40	pH = 4.13
$[H^+] = 3.59E-6$	$[H^+] = 7.41E-5$
$[Ca^{2+}] = 0.47$	$[Ca^{2+}] = 0.47$
$[Na^+] = 1.81$	$[Na^+] = 1.80$
$[Mg^{2+}] = 0.099$	$[Mg^{2+}] = 0.099$
$[K^+] = 0.036$	$[K^+] = 0.036$
$[Ba^{2+}] = 5.71E-5$	$[Ba^{2+}] = 9.64E-7$
$[SO_4^{2-}] = 4.84E-3$	$[SO_4^{2-}] = 4.78E-3$
$[H_4SiO_4] = 1.61E-5$	$[H_4SiO_4] = 6.24E-6$
$[Cl^-] = 2.97$	$[Cl^-] = 2.97$
$[Al^{3+}] = 1.00E-5$	$[Al^{3+}] = 1.15E-7$
$[Fe^{2+}] = 1.42E-3$	$[Fe^{2+}] = 1.42E-3$
$[CO_2]_{tot}$ = N/A	$[CO_2]_{tot} = 0.689$

Note: All values reported in Table 1 are in the unit of mol/kg. A brine density of 1.11×10^3 kg/m^3 was used to convert mg/L values in Soong et al. (2014) into mol/kg. The brine density was calculated based on a brine density calculator (Computer Support Group, Inc., 2014).

Most previous studies are focused on the reaction pathways for interaction between host rock and CO_2-saturated brine, while the correlation between chemical reactions and change in porosity and permeability has not been extensively studied. This study demonstrates a methodology to couple reactive transport modeling and permeability evolution modeling, and the results from this study provide a link between chemical reaction and permeability change. A comprehensive sensitivity analysis is conducted to investigate how the changes of important modeling parameters affect permeability evolution, which has not been reported in previous studies. Though the simulations performed in this study are not field scale, the results from this study can be used to demonstrate how chemical reactions might affect the permeability of Mount Simon formation after injection of CO_2.

2. Model description

2.1. Model set-up

A 1-D reactive transport model was developed with the use of the multicomponent reactive transport modeling code CrunchFlow (Steefel, 2009). The model simulates interactions between CO_2-saturated brine and a Mount Simon sandstone sample, and yields mineral composition and porosity changes of the sample. The model is focused on the post injection period, and the simulations performed were able to capture the conditions at locations close to the CO_2 plume after injection of CO_2 (i.e., near-static (no flow) condition, pressure is close to pre-injection reservoir pressure, and brine is saturated with CO_2). Two scenarios with different simulation durations were tested. The first scenario had a modeled reaction time of 180 days, which was the same as the exposure time of the sandstone exposure experiment (Soong et al., 2014); the second scenario had a modeled reaction time of 30 years, which was selected to explore permeability and porosity changes of the sandstone sample after very long exposure time.

A schematic of the modeling region is illustrated in Fig. 1. In the 1-D model, the sandstone sample is divided into 1000 grid blocks and each block has a length of 2.54×10^{-3} cm. The ratio of the brine domain width (296 mm) to sandstone domain width (25.4 mm) equals to the ratio of brine volume to sandstone volume in the high pressure vessel,