



# Fines migration during supercritical CO<sub>2</sub> injection in sandstone

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## ABSTRACT

Sandstone formations are widely considered for underground sequestration of CO<sub>2</sub>. During CO<sub>2</sub> injection, fines migration can cause pore blockage, which reduces sandstone permeability and CO<sub>2</sub> injectivity. To better the understanding of the mechanism of fines migration during CO<sub>2</sub> injection, this study characterizes the fines mobilized.

This paper presents a CO<sub>2</sub> injection experiment with detailed characterization of the rock and fluids produced. A Berea sandstone core sample is used. To characterize the core sample, X-Ray powder Diffraction (XRD), X-Ray Fluorescence (XRF), and Scanning Electron Microscopy (SEM) analyses are performed. The core receives injection first of brine (10 g/l NaCl), then of CO<sub>2</sub>-saturated brine, and finally of brine-saturated supercritical CO<sub>2</sub> (scCO<sub>2</sub>). During the injection, pressure difference between the core's injection face and production face is recorded. Samples of produced water are used for calculating produced fines concentration and for ionic chromatography. Then fines are separated from the produced water samples for further characterization. After the experiment, SEM images of the core are taken and compared with the pre-injection images to assess fines migration. Energy Dispersive X-ray Spectroscopy (EDS) is run on the post-injection SEM images to identify the fines blocking the pores.

SEM-EDS analysis of the produced fines and blocked pores in the core show that blockage is caused by clay, quartz, and cement. Ionic chromatography for produced water during CO<sub>2</sub>-saturated brine and brine-saturated scCO<sub>2</sub> injection show an increase in the ions present in the intergranular cement of Berea core. This indicates that CO<sub>2</sub>-saturated brine dissolved the cement, resulting in dislodgement of clay and quartz particles, some of which blocked the pore space near the core production end and thereby reduced permeability.

## 1. Introduction

Geological storage of CO<sub>2</sub>, known as CO<sub>2</sub> sequestration, can reduce greenhouse gas emissions (Zhang et al., 2018; Xie et al., 2017a; Bradshaw et al., 2007; Bachu, 2000). For CO<sub>2</sub> sequestration to be feasible, sustained CO<sub>2</sub> injectivity is essential. Injectivity loss during CO<sub>2</sub> injection can result from wettability (Kamali et al., 2017; Li et al., 2015; Berg et al., 2013; Stern, 1991), fluid bypassing (Krevor et al., 2012; Perrin and Benson, 2010; Yellig, 1982), mineral dissolution and precipitation (Miri et al., 2015; Ott et al., 2011; Patel et al., 1985), or fines migration (Sokama-Neuyam et al., 2017; Sbai and Azaroual, 2011; Sokama-Neuyam and Ursin, 2015; Olsen, 2011). The last of these factors, fines migration, is the focus of this paper. Several studies have investigated fines migration during water flow in coal (Guo et al., 2015; Guo et al., 2016; Keshavarz et al., 2016) and sandstone rocks (Russell et al., 2017; Hussain et al., 2013; Bedrikovetsky et al., 2011). However, understanding of fines migration during CO<sub>2</sub> sequestration is still limited.

To understand fines-migration during CO<sub>2</sub> sequestration, Sokama-Neuyam et al. (2017) studied the displacement of colloidal suspension by supercritical carbon dioxide (scCO<sub>2</sub>). Sandstone core samples were saturated with colloidal suspension. Then scCO<sub>2</sub> was injected to displace colloidal suspension in the core samples. The results demonstrated severe injectivity impairment due to pore blockage caused by colloidal particles. However, both studies used alumina latex particles, which are not naturally present in sandstones (Hussain et al., 2013). Therefore, artificially injected colloidal particles might not fully represent the in-situ fines migration mechanism.

Physical migration of naturally present fines has been investigated during CO<sub>2</sub>-saturated brine interaction with sandstone samples (Sayegh et al., 1990; Dawson et al., 2015; Pudlo et al., 2015). Sayegh et al. (1990) observed the chemical reactions during the injection of CO<sub>2</sub>-saturated brine into sandstone core samples. They found that fines migration caused permeability to drop by 15–30%. Dawson et al. (2015) conducted static dissolution experiments on Berea sandstone blocks kept in CO<sub>2</sub>-saturated brine and confirmed the chemical

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**Nomenclature**

EDS	Energy-Dispersive X-ray Spectroscopy	$k_0$	Initial permeability
$F_d$	Drag force	$r_p$	Radius of the particle
$F_e$	Electrostatic force	scCO <sub>2</sub>	Supercritical carbon dioxide
$F_g$	Gravity force	SEM	Scanning Electron Microscope
$F_l$	Lift force (N)	XFD	X-Ray powder Diffraction
$h$	Vertical distance from the interface	XRF	X-Ray Fluorescence
ICP-OES	Inductively Coupled Plasma Optical Emission Spectroscopy	$\alpha$	Position of the particle with the interface
$I_d$	Levers of drag force	$\theta$	Three-phase contact angle
$I_n$	Levers of normal force	$\sigma$	Interfacial tension
$k$	Final permeability	$p$	Perimeter of the contact between the particle and the interface
		PVI	Pore Volume Injected

dissolution of cement minerals. Comparison of SEM images taken before and after the experiments showed dissolution of carbonate minerals. Pudlo et al. (2015) conducted static experiments on a reservoir sandstone and concluded that the chemical dissolution lead to an increased exposure of clay mineral surfaces. This exposure of clay can result in fines migration. Jean et al. (2015) investigated changes in water chemistry during static experiments conducted on a storage formation and its overlying cap rock. The results confirmed that most minerals in the sandstone and caprock can be mobilized and dissolved in the presence of CO<sub>2</sub>-saturated brine under supercritical conditions. Tang et al. (2016) conducted both static and dynamic experiments on low and high permeable sandstones. Static experiments confirmed the mineral dissolution and dynamic injection of CO<sub>2</sub>-saturated brine demonstrated that minerals can migrate in sandstone with any permeability. However, the above experiments are single-phase CO<sub>2</sub>-saturated brine-rock interaction experiments, which might not manifest the injectivity issues found in two-phase CO<sub>2</sub> injection (Xie et al., 2017b).

Berrezueta et al. (2013) performed static experiments to investigate dry CO<sub>2</sub> interaction with a sandstone sample. Because CO<sub>2</sub>-saturated brine was not injected, the chemical dissolution of cement minerals was not observed. Berrezueta et al. (2013) argued that in a field-scale CO<sub>2</sub> injection scenario, near CO<sub>2</sub> injection wells, CO<sub>2</sub> displaces brine in near dry conditions. Therefore, chemical reactions or dissolution phenomena do not exist and fines migrate only due to drag applied by the injected CO<sub>2</sub>. Other experimental and theoretical studies have concurred that chemical reactions in the presence of dry CO<sub>2</sub> can be ignored (Gaus et al., 2008; Sterpenich et al., 2009). However, low pH CO<sub>2</sub>-saturated brine can affect CO<sub>2</sub> injectivity (Gaus et al., 2008). Sterpenich et al. (2009) performed static experiments with CO<sub>2</sub>-saturated brine and concluded that calcite dissolution should not occur during CO<sub>2</sub> injection. Jin et al. (2016) modelled geochemical reactions during CO<sub>2</sub> injection and concluded that the reactions are very slow and do not affect CO<sub>2</sub> injectivity. Furthermore, in a realistic CO<sub>2</sub> sequestration scenario, chemical reactions are usually considered for long-term (hundreds of years) simulations to model CO<sub>2</sub> trapping (Bachu, 2008). Although a number of studies have supported absence of chemical reactions during

CO<sub>2</sub> injection, Berrezueta et al. (2013) is in conflict with previously cited work that suggests a strong influence of chemical reactions on fines migration (Sayegh et al., 1990; Dawson et al., 2015; Pudlo et al., 2015; Jean et al., 2015; Tang et al., 2016). Recent field (Bickle et al., 2017) and laboratory (Mohamed et al., 2012) studies show direct evidence of rapid chemical reactions during scCO<sub>2</sub> injection.

In Mohamed et al. (2012), a Berea sandstone core fully saturated with brine was injected with 15 pore volumes of dry scCO<sub>2</sub>. Ionic concentration of produced brine showed the chemical dissolution of the sandstone minerals. A reduction of 39% was observed in the initial permeability of the Berea core. A limitation in Mohamed et al. (2012) is that the brine was not saturated with CO<sub>2</sub>. CO<sub>2</sub>-saturated brine can have a pH of 3–4, which can strongly affect the chemical reaction rates (Zhang et al., 2015). Although it might seem that the injected CO<sub>2</sub> should mix with brine and saturate it, the mixing of CO<sub>2</sub> and brine has been shown to be too slow to occur in a short dynamic experiment (Zhang et al., 2015). Hence the chemical reactions noted by Mohamed et al. (2012) might not represent a realistic scenario.

In more recent studies, sandstone core samples received injection first of brine, then of CO<sub>2</sub>-saturated brine, and finally of CO<sub>2</sub> (Xie et al., 2017b; Al-Yaseri et al., 2017). This series of injections is closer to a realistic scenario and usually adopted in experimental modelling of CO<sub>2</sub> relative permeability tests (Berg et al., 2013). However, in contrast to Mohamed et al. (2012), Xie et al. and Al-Yaseri et al. did not measure the dissolved ionic concentration in the produced water. Hence, there was no direct evidence of chemical reactions. Consequently, the authors' discussion of chemical reactions was qualitative only. Xie et al. (2017b) derived analytical models based on their experiments. But their models did not consider chemical reactions.

The use of incompatible experimental methods in the above studies has led to contradictory inferences about fines migration mechanisms, particularly regarding chemical reactions. In addition, none of the studies tried to characterize the types of migrating fine. Characterization of the produced fines can help in identifying mechanisms associated with fines migration. This paper presents a well-characterized experimental study to model fines migration during CO<sub>2</sub>-

**Table 1**  
XRD results for the sample of Berea sandstone.

Compound name	Mineral name	Chemical formula	Quantity [%wt]
Silicon Oxide	Quartz	SiO <sub>2</sub>	84.1 (± 0.7)
Potassium Aluminum Silicate Hydroxide	Muscovite -2M1	Al <sub>3</sub> H <sub>2</sub> KO <sub>12</sub> Si <sub>3</sub> [(K <sub>0.959</sub> Ba <sub>0.012</sub> Na <sub>0.029</sub> ) (Al <sub>1.826</sub> Ti <sub>0.033</sub> Fe <sub>0.102</sub> Cr <sub>0.108</sub> Mg <sub>0.11</sub> Mn <sub>0.0009</sub> ) (Si <sub>3.135</sub> Al <sub>0.865</sub> ) O <sub>10</sub> ) (O <sub>0.502</sub> F <sub>0.041</sub> Cl <sub>0.001</sub> (OH) <sub>1.456</sub> )]	6.3 (± 0.6)
Aluminum Silicate Hydroxide	Kaolinite-1A	Al <sub>2</sub> (Si <sub>2</sub> O <sub>5</sub> ) (O H) <sub>4</sub>	4.8 (± 1.1)
Potassium Aluminum Silicate (K-Feldspar)	Microcline	AlK O <sub>8</sub> Si <sub>3</sub>	2.9 (± 0.4)
Calcium Magnesium Carbonate	Dolomite	CaMg(CO <sub>3</sub> ) <sub>2</sub>	0.9 (± 0.0)
Titanium Oxide	Rutile	TiO <sub>2</sub>	0.5 (± 0.1)
Iron Carbonate	Siderite	FeCO <sub>3</sub>	0.5 (± 0.0)

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