EXPERIMENTAL INVESTIGATION OF THE IMPACT OF SALT PRECIPITATION ON CO_2 INJECTIVITY

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ABSTRACT

Adequate CO_2 injectivity is required to inject large volumes of CO_2 into the reservoir at acceptable injection flow rates through a minimum number of wells. Salt precipitation induced by brine vaporization in the wellbore vicinity could impair CO_2 injectivity especially in deep saline reservoirs. We conducted core-flood experiments to investigate the development of salt cake at the injection inlet during CO_2 injection into sandstone core samples. We also attempted to quantify the impact of drying and salt precipitation on CO_2 injectivity. We observed severe salt cake deposition at the injection inlet even when the core was still wet with immobile brine. The amount of salt cake deposited at the injection inlet depended on the resident brine salinity and the supercritical CO_2 injection rate. About 29 per cent CO_2 injectivity impairment was induced by drying and salt precipitation. Injectivity impairment was also found to be dependent on CO_2 injection flow rate. The present work provides insight into the underlying mechanisms of salt cake development at the injection inlet during CO_2 injection into deep saline formations.

INTRODUCTION

A successful Carbon Capture, Utilization and Storage (CCUS) candidate must have good containment efficiency, adequate storage capacity and threshold well injectivity, to inject the desired quantity of CO_2 at acceptable rates through a minimum number of wells [1]. Deep saline reservoirs standout in terms of storage capacity and containment efficiency. However, salt precipitation induced by brine vaporization near the wellbore, could threaten CO_2 injectivity in deep saline formations and render them unsuitable for CCUS [2–5]. Miri and Hellevang [6] identified initial rock permeability, CO_2 injection rate, saturating brine salinity, temperature and pressure as important parameters underlying CO_2 injectivity impairment caused by salt precipitation.

The underlying mechanisms of formation brine dry-out and salt precipitation include: (1) immiscible two-phase displacement of brine by injected CO_2, (2) vaporization of brine into the flowing CO_2 stream, (3) capillary-driven back-flow of brine toward the injection inlet, (4) diffusion of dissolved salt in the aqueous phase, and (5) gravity override of injected CO_2 [6]. Low brine evaporation rate in the drying front, may result in homogeneous distribution of precipitated salt throughout the porous medium [3,4,6,7]. For high evaporation rates, there are no sufficient time for the salt concentration gradient to diffuse away from the drying front, resulting in nonhomogeneous accumulation of salt [4,8]. While numerical experiments by Roels et al.[9] suggested that local salt accumulation occurs far from the wellbore, several research works [3,10–12] suggest that precipitated salt accumulates near the wellbore.

As brine is vaporized, the concentration of salt in the brine increases. Zuluaga et al.[13] explains that salt precipitates out when the brine concentration exceed supersaturation. Several researchers have observed the formation of salt cake at the core inlet in CO_2 core-flood experiments [10,14]. However, the governing mechanism of salt cake development at the injection inlet have not been studied thoroughly but it is believed that capillary backflow of brine towards the injection inlet may result in formation of salt cake if the brine salinity is high enough to reach supersaturation before the brine is swept into the formation. Insight into the development of salt cake during the drying process will improve understanding of its impact on CO_2 injectivity.

In the present work, we have conducted various core-flood experiments using sandstone core plugs where the development of salt cake at the core inlet was monitored. We also present quantitative effect of drying and salt precipitation on CO_2 injectivity.

MATERIALS AND METHODS

Materials

The experiments were conducted on two types of homogeneous cylindrical sandstone core plugs. Berea sandstone core samples were used to study the quantitative effect of drying and salt precipitation because of their suitable range of permeability and porosity. Bentheimer core plugs were used to investigate salt cake development at the injection inlet because of their high permeability. Each cylindrical core samples were 20cm long and 3.81cm in diameter.

The non-wetting fluid used was liquefied CO_2 with purity percentage of about 99.7 %. The liquid CO_2 was injected at 80 bar and 25 ^{o}C. During supercritical CO_2 injection, the operational conditions were set to 80 bar and 45 ^{o}C. To investigate the effect of brine vaporization and salt precipitation on CO_2 injectivity, we used synthetic Formation Water (FW) with salinity of 105.5 g/l (NaCl 77.4 g/L; $CaCl_2.2H_2O$ 21.75 g/L; $MgCl_2.6H_2O$ 3.56g/L; $SrCl_2.6H_2O$ 2.25 g/L; Na_2SO_4 0.13 g/L; KCl 0.42 g/L)[15]. The Berea sandstone cores were vacuum-saturated with FW prior to CO_2 injection. NaCl brine with equivalent ionic strength as FW (120 g/l) was used to investigate the development of salt cake at the injection inlet. Jeddizahed and Rostami [16] have shown that the total dissolved salt is the main determinant of the precipitated solid salt saturation. Therefore, we expect NaCl brine with the same ionic strength as FW to precipitate solid salt comparable to FW under the same conditions.

Setup and Procedure

The schematics of core flooding apparatus used in the tests are shown in Figure 1. Prior to the test, the core was loaded in a horizontal core-holder. The Quizix pump delivers brine through the connected piston cell into the core inlet. The ISCO CO_2 pump receives liquid CO_2 from the gas container through a pressure regulator. Depending on the injection conditions, either liquid or supercritical CO_2 are injected. The injected fluid passes a piston cell, positioned in the oven, to hold the fluid and secure a pre-set temperature in the oven. A differential pressure gauge and a pressure transducer are connected across the core to monitor the pressure drop and record the absolute pore pressure. A backpressure of 80 bar was set at the outlet during CO_2 injection and effluent fluid was safely collected in a piston cell for analysis and safe disposal.

The core was cleaned and dried at 65 ^oC for about 24 hours. It was then wrapped in shrinking Teflon and rubber sleeves to prevent CO_2 leakage and mounted horizontally in the core holder. A confinement pressure of about 150 bar was applied in the annular space between the core and the core-holder. The experimental procedure consists of the following steps:

1. Initial liquid CO_2 pressure drop (ΔP_i) across the clean dry core sample were measured and the permeability (K_i) calculated.
2. The core was saturated with either FW or NaCl brine.
3. The saturated core sample was flooded with supercritical CO_2 to vaporize brine.
4. The core was then inspected for salt cake development at the inlet.
5. Final liquid CO_2 pressure drop (ΔP_f) was measured and permeability (K_f) calculated.

In Step 1 and Step 5, liquid CO_2 was injected at 2 ml/min to measure permeability before and after brine vaporization, and salt precipitation. In step 3, about 100 – 300 pore volumes (PV) of supercritical CO_2 was injected at various injection flow rates to vaporize brine and dry the core.

Theory

Fluid injectivity, I is defined as the ratio of volumetric injection flow rate, q to the pressure drop, Δp. Assuming the core has constant absolute permeability k_i and k_f before and after it has been exposed to precipitated minerals respectively and that the viscosity of the fluid used in the measurement (liquid CO_2) is constant, the injectivity before and after salt deposition can be expressed from Darcy's law as:

$$I_i = \frac{q_i}{\Delta p_i} = k_i.C \tag{1}$$

$$I_f = \frac{q_f}{\Delta p_f} = k_f.C \tag{2}$$

In Eq. (1) and (2), C is a constant defined as $C = \frac{A}{\mu L}$, for constant A and L. If liquid CO_2 is injected at a constant rate during injectivity measurements ($q_i = q_f$), we can define a Relative Injectivity Change (RIC) index as:

$$RIC = \left(\frac{I_i - I_f}{I_i}\right) = 1 - \left(\frac{I_f}{I_i}\right) \tag{3}$$

Substituting Eq. (1) and (2) into (3) yields:

$$RIC = 1 - \left(\frac{\Delta p_i}{\Delta p_f}\right) = 1 - \left(\frac{K_f}{K_i}\right) \tag{4}$$

Salt precipitation would reduce the flow area and increase Δp. Thus, $\Delta p_f > \Delta p_i$ and $K_i > K_f$ after salt deposition. Consequently, a positive RIC value indicates injectivity impairment. In the present work, RIC is expressed as a percentage.

RESULTS AND DISCUSSION

Salt Cake Development

To investigate the development of salt cake at the injection inlet during CO_2 injection, a clean Bentheimer core was vacuum-saturated with 120 g/l NaCl brine and flooded with about 100 PV of supercritical CO_2 at a rate of 1 ml/min. The core was then taken out and inspected for salt cake. Figure 2 shows pictures of the core taken after the core-flood process.

Figure 2 (A) shows that no salt was observed at the injection outlet. In Figure 2 (B), we observe massive filter salt cake development at the core inlet. Figure 2 (C) shows that the entire core was still wet. When CO_2 is injected into the core, capillary backflow draws the aqueous phase towards the core inlet. Although the same fittings were used at the inlet and the outlet, no salt was precipitated at the injection outlet. This affirms the role played by capillary backflow of brine in salt cake development. The dry supercritical CO_2 removes water from the brine through evaporation. As more water is removed, the salinity of brine increases. When the concentration of salt in the brine exceed supersaturation, salt precipitates out. A filter salt cake could be deposited at the injection inlet if the inlet brine salinity exceed supersaturation before it is swept into the core by the injected gas. For salt cake development at the injection inlet, two conditions must be fulfilled: (1) The brine salinity must be sufficiently high and (2) capillary backflow dominate flow around the injection inlet.

To investigate the impact of brine vaporization rate on salt cake development, we increased the supercritical CO_2 injection flow rate from 1 ml/min to 5 ml/min. We observed that, the amount of deposited salts at the injection inlet decreased when the brine vaporization rate was increased from 1 ml/min to 5 ml/min (Figure 3). At higher supercritical CO_2 injection flow rate, viscous forces overcome capillary forces. The net effect is reduced capillary backflow of brine at the injection inlet. Thus, brine available at the inlet for salt dropout is reduced and lower amount of salt cake is deposited at the inlet.

We then reduced the brine salinity from 120 g/l to 75 g/l, keeping the CO_2 injection flow rate at 5 ml/min. We observed the amount of salt cake precipitated at the injection inlet further decreased significantly when the brine salinity was decreased (Figure 4). At constant injection flow rate, it will take a longer time for the low salinity brine to reach supersaturation. Therefore, a significant portion of the producible brine at the injection inlet could be swept into the core, reducing the chances of salt cake development. Therefore, salt cake could be deposited at the injection inlet during CO_2 injection into saline porous media if the saturating brine salinity is above a certain threshold value and capillary backflow of brine at the injection inlet is high enough.

Effect of Formation Brine Dry-out

It is important to note that in this section, all salt cake at the core inlet was removed before injectivity effects were quantified. Only the effect of complete dry-out and salt precipitation within the core was studied. The clean Berea core with known permeability was initially saturated with FW and flooded with about 300 PV of supercritical CO_2 at a rate of 1 ml/min until the core was completely dried. The permeability of the core after drying was measured and RIC was calculated. The experiment was then repeated for CO_2 injection flow rate of 5 ml/min and 10 ml/min, keeping all other parameters constant, to study the effect of injection flow rate. Figure 5 shows the results of injectivity impairment induced by brine vaporization at varying CO_2 injection flow rates.

CO_2 injectivity was impaired by about 36 % for drying rate of 1 ml/min (Figure 5). Injectivity impairment then decreased from 36% to about 25% when drying rate was increased to 5 ml/min and remained practically unchanged when the drying rate was further increased to 10 ml/min. Several researchers [2,5,17–19] have reported CO_2 injectivity impairment within a range (13% - 83%) that agree favorably with our findings. During CO_2 injection, water is removed from the core through immiscible CO_2-brine displacement and brine vaporization. As brine is vaporized by supercritical CO_2, the concentration of salt in the brine increases within the pore spaces. When the concentration of brine exceed supersaturation, salt precipitates into the pore channels as observed by Zuluaga et al., (2001). The precipitated salts drop out into the pore channels, reducing the CO_2 flow area and consequently the permeability and injectivity [4,16,20,21].

At very low CO_2 injection rate, immiscible CO_2-brine displacement is delayed and the drying rate is lowered, thus increasing the period available for salt precipitation. At high CO_2 injection rates, the resident brine is quickly swept out of the core, leaving out only immobile brine for salt precipitation. With pore volume of about 45 ml, the time taken to sweep out all producible brine will be close for injection rates of 5 ml/min and 10 ml/min and probably that is why injectivity impairment remained practically the same within this interval of CO_2 injection flow rate.

CONCLUSION

Storage capacity and well injectivity are the two most important parameters required to define the storage potential of a geological CCUS candidate. Deep saline reservoirs have the largest storage capacity. However, brine vaporization and salt precipitation in the wellbore region during CO_2 injection, could impair injectivity, thus rendering deep saline formations unsuitable for CO_2 storage. Most of the experimental and theoretical works have investigated salt precipitation in the dry-out region. It is therefore generally assumed that salt precipitation commences when the near well region start to dry-out.

We have conducted laboratory core-flood experiments using sandstone cores to study the mechanisms of salt precipitation from the onset of brine vaporization to the end of the drying process. Some of the highlights of our work include the following:

- Salt could be precipitated in the form of salt cake at the core inlet when the core is still wet with immobile brine. Capillary backflow of brine towards the core inlet could be the main driving force of salt cake development.
- The magnitude of precipitated salt cake depends on the injection flow rate of supercritical CO_2 and the salinity of the resident brine. High brine salinity and low CO_2 injection rate favor salt cake development.
- At complete dry-out, the effect of salt precipitation on CO_2 injectivity is dependent on the drying rate. An average of about 29% injectivity impairment was induced by drying effects.

The present work provides experimental evidence of salt precipitation prior to drying out. Some of the influential parameters underlying the development of salt cake in the inlet region have been identified and studied.

NOMENCLATURE

CCUS	Carbon capture, Utilization and Storage
FW	Formation water
I	CO_2 well Injectivity
K_i	Initial Permeability
K_f	Final Permeability
PV	Pore Volume

Δp_i	Initial pressure drop across the core
Δp_f	Final pressure drop across the core
q_i	Initial injection volumetric flow rate
q_f	final injection volumetric flow rate

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APPENDIX

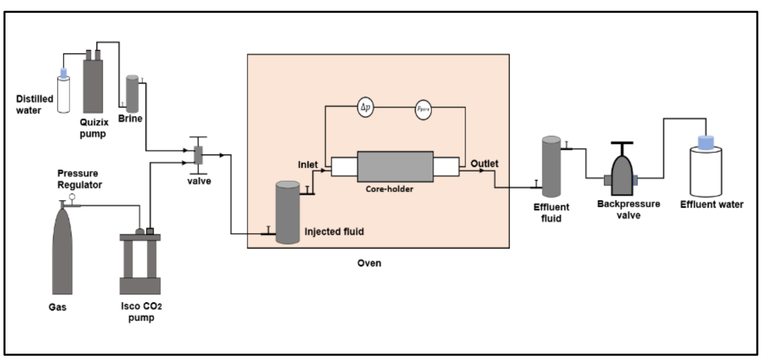

Figure 1. Experimental setup - the CO2 flow rig.

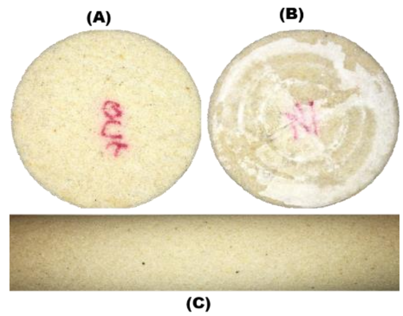

Figure 2. Pictures showing Salt cake development at 1 ml/min CO_2 injection rate into Bentheimer core saturated with 120 g/l NaCl brine. (A) No salt cake observed at the outlet. (B) Massive salt cake developed at the inlet. (C) The entire core is still wet.

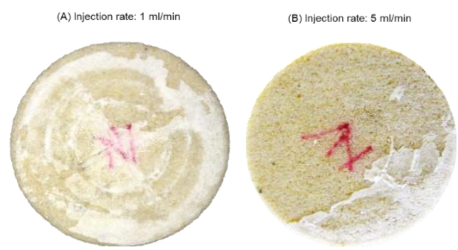

Figure 3. Pictures showing salt cake development as supercritical CO_2 injection rate was increased from 1 ml/min (A) to 5 ml/min (B).

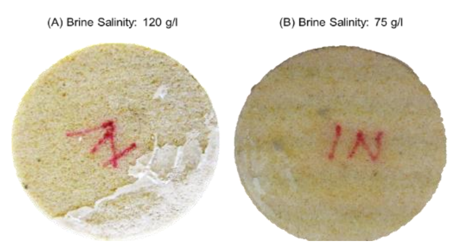

Figure 4. Pictures showing salt cake development as brine salinity was decreased from 120 g/l (A) to 75 g/l (B).

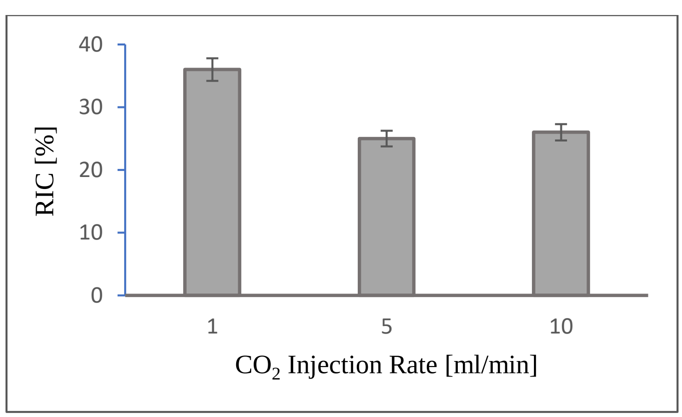

Figure 5. Effect of brine vaporization and salt precipitation on CO_2 injectivity. Injectivity impairment increased with decreasing drying rate.