Effect of CO₂ Flooding on the Wettability Evolution of Sand-Stone

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Abstract: Wettability is one of the main parameters controlling CO₂ injectivity and the movement of CO₂ plume during geological CO₂ sequestration. Despite significant research efforts, there is still a high uncertainty associated with the wettability of CO₂/brine/rock systems and how they evolve with CO₂ exposure. This study, therefore, aims to measure the contact angle of sandstone samples with varying clay content before and after laboratory core flooding at different reservoir pressures, of 10 MPa and 15 MPa, and a temperature of 323 K. The samples’ microstructural changes are also assessed to investigate any potential alteration in the samples’ structure due to carbonated water exposure. The results show that the advancing and receding contact angles increased with the increasing pressure for both the Berea and Bandera Gray samples. Moreover, the results indicate that Bandera Gray sandstone has a higher contact angle. The sandstones also turn slightly more hydrophobic after core flooding, indicating that the sandstones become more CO₂-wet after CO₂ injection. These results suggest that CO₂ flooding leads to an increase in the CO₂-wettability of sandstone, and thus an increase in vertical CO₂ plume migration and solubility trapping, and a reduction in the residual trapping capacity, especially when extrapolated to more prolonged field-scale injection and exposure times.

Keywords: CO₂ injectivity; wettability; contact angle; sandstone; CO₂ sequestration

1. Introduction

Carbon geological sequestration (CGS) has been proposed as an efficient method to reduce anthropogenic CO₂ emissions into the atmosphere and thus mitigate global climate change [1]. In essence, the technique involves capturing CO₂ from large stationary emission sources and locking it into some natural geological formations [1–3]. There are three geological formations that have attained a wide consideration. They include (1) depleted oil and gas reservoir, (2) deep saline aquifers, and (3) coal seams [1]. In saline aquifers and oil and gas reservoirs, CO₂ storage is typically placed at depths below 800 m, where CO₂ becomes liquid or supercritical because of the ambient pressure and temperature conditions [1]. Therefore, the vertical migration of CO₂ is the main problem involved in CO₂ injection due to the density differences between the brine and CO₂ [4,5]. It is also essential to assess the different functional trapping mechanisms, which prevent the buoyant CO₂ from flowing upwards [1]. The CO₂ can be trapped in geological formations utilising...
four mechanisms, including structural trapping [6,7], capillary trapping [8–11], solubility trapping [12–14], and mineral trapping [15–17]. Furthermore, coal seams are considered as another option for the underground storage, in which the CO$_2$ injection into coal seams will have advantages for both CO$_2$ storage and enhance methane recovery [1,2]. A number of large-scale CO$_2$ storage projects are currently in operation worldwide. These have captured and stored millions of tonnes of CO$_2$ annually. Many more projects have been planned. Specifically, oil and gas companies have been operating geological CO$_2$ storage projects for a number of years. They have successfully demonstrated that securely storing a large quantity of CO$_2$ in a deep underground area is possible [1]. For instance, the active CGS projects are (1) Sleipner (Norway), (2) Weyburn Midale (Canada), and (3) Cranfield (US), established in 1996, 2000, and 2008, with CO$_2$ capture capacities of 1, 3, and 1.5 Mt/year, respectively. The planned CGS projects include (1) Gorgon (Australia), (2) Quest (Canada), and (3) GreenGen (China), established in 2016, 2015, and 2011, respectively [1,5].

In carbon geo-sequestration, wettability is a crucial factor that intensely and directly influences containment security, injectivity, structural, dissolution, and residual trapping capacities [18,19]. Five different wettability states can be conceptualised in a real reservoir, i.e., strongly water-wet, weakly water-wet, intermediate-wet, weakly CO$_2$-wet, and strongly CO$_2$-wet (where complete wetting occurs), with approximate contact angles of 0°–50°, 50°–70°, 70°–110°, 110°–130°, and 130°–180°, respectively [20]. These differences in wettability are caused by geological and chemical factors, such as surface chemistry (e.g., organic content) [21–23], reservoir pressure (the increase in pressure leads to a decrease in water wettability) [18,24,25], reservoir temperature [26–28], salinity, and ion type (salinity increases as CO$_2$ wettability increases) [18,29–32]. Therefore, it is essential to understand the fluid-rock interaction, as these interactions clearly can affect the capillary pressure and aquifer permeability, and hence the injectivity and storage capacities [33,34].

Injected CO$_2$ forms carbonic acid in the brine phase [35,36]. It interacts with rock minerals, which leads to mineral alterations and ion dissolution–precipitation [37]. Deep saline sandstone reservoirs are potential candidates for CO$_2$ sequestration [1]. Sandstones generally consist of siliceous minerals, clays, and various carbonates, along with quartz [38]. These minerals react differently to the changing environment when CO$_2$ is injected, e.g., calcite cement is highly reactive in an acidic environment [35,39,40]. In fact, pH decreases to 3–4 when CO$_2$ mixes with brine at reservoir conditions [41,42], and such an acidic condition can considerably affect the permeability and pore morphology [43]. Alternatively, CO$_2$ can be stuck in the target reservoir’s pore space for hundreds or thousands of years because of the slow dissolution kinetics caused by the partial mixing of CO$_2$ and brine [44].

Some studies have reported that such water–CO$_2$–rock interactions could change the sandstone pore structures due to fines migration and precipitation or reaction with sensitive materials [45,46]. Such a change can strongly affect the rock porosity and permeability performance [36,47,48]. Furthermore, the influence of the temperature and injection rate on the permeability reduction after CO$_2$ injection have been examined on Berea sandstone [36] and sandstones from the Pembina Cardium field, Canada [49], whereas other studies investigated the factors controlling the permeability changes in sandstone during core flooding [50]. However, the effect of CO$_2$ injection on the wettability changes has received less attention. Thus, this study analyses how CO$_2$ injection changes sandstone’s wettability. This change was correlated with a microstructural alteration in the sandstone caused by CO$_2$ flooding. Subsequently, we determined how CO$_2$ flooding affects the CO$_2$ trapping capacities (i.e., residual and dissolution) and the amount of free CO$_2$ in saline aquifers (i.e., mobile CO$_2$).

2. Materials and Methods

2.1. Materials

Two homogeneous Berea sandstone samples (low clay content) and two Bandera Gray (high clay content) were used in this study. The sandstones were thoroughly characterised by scanning electron microscopy (SEM) to measure the surface morphology and
quantitative X-ray diffraction (XRD—Bruker-AXS D8) to measure the mineral composition before/after the flooding experiment. The samples’ petrophysical properties, including porosity and permeability, were measured before and after flooding and are reported in Table 1.

Table 1. Petrophysical and mineralogical sandstone properties.

| Sample        | Porosity a (%) | Brine Permeability (mD) | Length (mm) | Diameter (mm) | Mineral Constituents b |
|---------------|----------------|--------------------------|-------------|---------------|-----------------------|
|               |                |                          |             |               |                      |
| Before Flooding |                |                          |             |               |                      |
| Berea         | 20             | 69                       | 50.88       | 30.78         | Quartz: 84.3          |
|               |                |                          |             |               | Kaolinite: 4.1        |
|               |                |                          |             |               | Illite: 1.9           |
|               |                |                          |             |               | Albite: 4.2           |
|               |                |                          |             |               | Microcline: 4.1       |
| Bandera Gray  | 19             | 9                        | 60.32       | 30.80         | Quartz: 58.2          |
|               |                |                          |             |               | Kaolinite: 3.2        |
|               |                |                          |             |               | Illite: 3.6           |
|               |                |                          |             |               | Albite: 12.4          |
|               |                |                          |             |               | Muscovite: 1.6        |
|               |                |                          |             |               | Chlorite: 5.7         |
|               |                |                          |             |               | Ankerite: 15.3        |
| After Flooding |                |                          |             |               |                      |
| Berea         | 22             | 80                       | 50.88       | 30.78         | Quartz: 84.9          |
|               |                |                          |             |               | Kaolinite: 3.9        |
|               |                |                          |             |               | Illite: 1.8           |
|               |                |                          |             |               | Albite: 4.2           |
|               |                |                          |             |               | Microcline: 4.1       |
| Bandera Gray  | 20             | 7.3                      | 60.32       | 30.80         | Quartz: 58.4          |
|               |                |                          |             |               | Kaolinite: 3.1        |
|               |                |                          |             |               | Illite: 3.2           |
|               |                |                          |             |               | Albite: 12.2          |
|               |                |                          |             |               | Muscovite: 3.1        |
|               |                |                          |             |               | Chlorite: 5.2         |
|               |                |                          |             |               | Ankerite: 14.8        |

a Porosity was measured with AP-608 Coretest Instrument. b The mineral composition of the samples was measured by X-ray Diffraction (XRD) Bruker—AXS D8.

2.2. CO₂ Core Flooding Experiment

The Berea and Bandera Gray core plugs were wrapped in polytetrafluoroethylene (PTFE) tape, aluminium foil, and PTFE tape again to be prepared for plugging. Subsequently, the samples were sealed with a PTFE heat-shrink sleeve and were placed in a rubber sleeve in a high-pressure and a high-temperature core holder (Figure 1). The samples were next vacuumed for more than 20 h [50]. In the next step, the sandstone samples were saturated with dead brine (5 wt% NaCl and 1 wt% KCl) using a high-precision syringe pump (ISCO 500D). The dead brine was then displaced by 5 pore volumes of live brine (5 wt% NaCl and 1 wt% KCl equilibrated with CO₂) at 1 mL/min [51]. The injection rate was reduced to 0.05 mL/min, and the injection continued for 7 days at reservoir conditions (pore pressure of 10 MPa, confining pressure of 15 MPa, and temperature of 323 K). This simulates the sinking of CO₂-saturated brine deep into the reservoir, i.e., the dissolution trapping [52–54]. Finally, five-pore volumes of supercritical CO₂ (scCO₂) were injected to displace the live brine and to simulate the CO₂ injection into the reservoir. Figure 1 presents a schematic of the core flooding apparatus used in this study.
2.3. Contact Angle Measurements

For the CO₂-wettability tests, the samples were cut with a high-speed diamond blade (5 mm thick cuboids, with a 38 mm diameter), and each sample was exposed to air plasma (model Diener plasma, Ebhausen, Germany, Yocto) for 5 min to remove any potential organic surface contaminations [55,56]. Subsequently, the contact angle was measured using the tilted plate method (as it can quantify simultaneously the advancing and receding contact angles) [37] at storage conditions. For contact angle measurements, the sample was placed inside the pressure cell at a set temperature (323 °K). CO₂ pressure was raised to the desired pressure (10 MPa and 15 MPa), using a high precision syringe pump (ISCO 500D; pressure accuracy of 0.1% FS). A droplet of the brine (5 wt% NaCl and 1 wt% KCl in deionised water) with an average volume of ~6 µL ± 1 µL was released onto the tilted (tilted angle of 12°) sample (Berea and Bandera Gray) surface through a needle. The advancing and receding contact angles were then calculated at the leading and trailing edge. A high-resolution video camera (with specification of Basler scA (640–70) fm, pixel size = 7.4 µm; frame rate = 71 frames per second; Fujinon CCTV lens: HF35HA-1B; 1.6/35 mm) recorded the whole process and the images extracted from the video files to measure the contact angles. Figure 2 illustrates the experimental setup of contact angle measurement. The standard deviation in the contact angle result was determined as ±3° based on replicated measurements.
3. Results and Discussion

3.1. Controlling Factors on Sandstone Wettability

The wettability of sandstone samples (Berea with low clay content and Bandera Gray with high clay content) was measured before and after CO$_2$ flooding at 10 and 15 MPa at a constant temperature of 323 K. The results clearly indicate that the contact angles after flooding were higher than before the flooding for both samples. This shows that Berea and Bandera Gray sandstones became more CO$_2$-wet after CO$_2$ injection. Besides, the advancing and receding contact angles increased with the increasing pressure for both the Berea and Bandera Gray samples (Figure 3), which is consistent with the literature data [18,20,59,60]. As an example, the advancing contact angle of Bandera Gray before flooding increased from 86° to 105° at 323 K, for a pressure of 10 MPa and 15 MPa, respectively. The results also indicate that Bandera Gray has a higher contact angle, compared with Berea, for all test pressures and both before and after CO$_2$ flooding conditions.

Clay minerals can be distributed in different ways within the reservoirs—in the form of laminations in between the grains (laminar clays), dispersed in the reservoir, or structurally coating the grains (structural clays) [50,61,62]. Moreover, the specific clay type is also crucial in controlling the petro-physical properties of sandstone [62–64]. The scanning electron microscopy (SEM) and XRD analysis revealed that the Berea and Bandera Gray sandstones comprise different clay types and distributions. The clay types present in both sandstone samples are shown in Table 1. Studies by other researchers have also demonstrated that, besides smectite, all clay minerals adsorb significant amounts of CO$_2$ [65]. As seen from the SEM analysis, both sandstone samples contain the CO$_2$-adsorbing clays. Therefore, the high contact angle in Bandera Gray (CO$_2$-wet) is attributed to its high clay content.

Moreover, changes in albite and ankerite surfaces have been reported earlier [66]. Dissolution textures have been shown on the surfaces of detrital albite grains. Smooth surface and step-like structures of ankerite grains showed corrosion pits at grain boundaries post-experiment [66]. The dissolution of illite and chlorite can also occur [67], and chlorite dissolution following a subsequent reaction with pre-existing calcite can lead to kaolinite and CO$_2$-rich ankerite production [68]. Since the presence of calcite and its dissolution is a rate-limiting step of this reaction, kaolinite and ankerite production and their precipitation depend on the calcite content of the rock samples [68]. Kaolinite precipitation was reported earlier [66] and was evident in this work for both the Berea and Bandera Grey samples (Figures 4 and 5). Kaolinite precipitation can also be due to the interaction between CO$_2$-saturated brine and feldspar (k-feldspar, albite, microcline) present in the rock [66]. Thus, it can be inferred that a larger content of albite and ankerite in Bandera Gray samples is responsible for more significant interactions with CO$_2$-saturated brine. This phenomenon explains the higher brine contact angle, and thus the higher CO$_2$ wetting, for Bandera Gray as compared to Berea sandstone at the same conditions.

The XRD results (Figure 6) and XRD images (Figure 7) were also employed, showing no significant change in mineral composition before and after CO$_2$ flooding. This can be due to the dissolution of minerals corresponding to their stoichiometry, keeping the overall mineralogy unchanged (or with an insignificant change beyond the detection limit). The additional peak in the XRD results after the experiments corresponds to NaCl, and thus salt precipitation post-drying cannot be ruled out.
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Figure 3. Berea and Bandera Gray sandstone/CO₂/brine advancing (a) and receding (b) contact angles as a function of pressure (measured at 10 and 15 MPa, and 323 K).
Figure 4. SEM images of (a) Berea before flooding (b) Berea after flooding: pore filling dispersed clay (illite/kaolinite based on the XRD) (c) Bandera Gray before flooding (d) Bandera Gray after flooding, quartz-grain-coated structural clay (mostly chlorite based on the XRD).

Figure 5. SEM images of (a) Berea before flooding, smooth surface (b) Clutter Berea after flooding: pore filling dispersed clay (c) Bandera Gray before flooding (d) Clutter Bandera Gray after flooding, quartz-grain-coated structural clay.
Figure 6. Differences in mineral compositions due to CO$_2$ flooding in: (a) Berea, (b) Bandera Gray.
3.2. Effect of Clay Content on Sandstone Trapping Capacity

Rock wettability highly affects CO$_2$ vertical migration and CO$_2$ trapping capacities [6,9]. CO$_2$-wet rock has a significantly higher CO$_2$ upward mobility [69] and a much lower residual trapping capacity [9,24,70] for formations with adequate flow properties (such as permeability). Our results, presented in Section 3.1, show that CO$_2$ flooding in sandstones with a high clay content leads to the reservoir being CO$_2$-wet, which may lead to the above-listed detrimental mobility and storage effects. For example, based on the previous simulation study by [71], the CO$_2$ mobility is found to contribute by 0.5%, dissolution trapping capacity by 18.3%, and residual trapping capacity by 81.2% in the storage capacity of strongly water-wet rocks. By contrast, the strongly CO$_2$-wet rocks have a CO$_2$ mobility of 20.7%, dissolution trapping capacity of 28.6%, and residual trapping capacity of 50.7% after 10 years of storage (Figure 8).
On the contrary, in formations with low permeability where the biggest challenge is the injection of CO$_2$, the enhanced CO$_2$ wetting of the rock’s surface could be advantageous for pressure management provided there is a cap-rock that can make a good seal for containment security. The presence of high clay fractions in such low permeability formations will be beneficial for the enhanced CO$_2$ storage capacity by an increase in mobility, dissolution, and residual trapping.

4. Conclusions

Rock wettability has a significant role in carbon geo-sequestration (CGS). This is because the fluid flow through porous media is strongly controlled by rock wettability. Despite previous research on the area, the parameters influencing the CO$_2$/brine/rock wettability variation are still not fully understood. We thus systematically measured the contact angle (i.e., wettability) of two sandstones (i.e., low clay content (Berea) and high clay content (Bandera Gray)) before and after CO$_2$ flooding with brine (5 wt% NaCl + 1 wt% KCl in deionised water), CO$_2$-saturated (live) brine, and supercritical CO$_2$ (scCO$_2$), at 10 MPa and 15 MPa for a constant temperature (323 K). The results show that CO$_2$ flooding leads to an increase in the advancing and receding contact angles of both Berea and Bandera Gray sandstones (i.e., CO$_2$ flooding leads to increased CO$_2$-sandstone wettability). Our results also show that the CO$_2$/brine/rock contact angle increases with the pressure increase, which is in line with most of the literature data [18,20,59,60].

Moreover, our measurements demonstrate, for all tested conditions (both before and after the CO$_2$ flooding scenarios), that Berea sandstone has lower contact angles (i.e., more water wettability) than Bandera Gray (i.e., Bandera Gray tends to be more intermediate-wet to CO$_2$-wet), due to the higher clay content of Bandera Gray. Our SEM results show that the Bandera Gray sandstones, which became more CO$_2$-wet, contained a high clay content. The published literature indicates that, except for smectites, all clays are CO$_2$-adsorbing. Hence, sandstones with a high clay content become CO$_2$-wet when flooded with CO$_2$, which results in the upward mobility of CO$_2$ in the reservoir, and consequently a reduced capillary trapping capacity. However, low permeability formations with significant CO$_2$ injection issues, CO$_2$-wetting of the reservoir rock surface, and an adequate seal for containment security could help improve the pressure management. Therefore, high clay fractions in such formations will be an advantage for the enhanced CO$_2$ storage capacity.
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