Transport properties of saline CO₂ storage reservoirs with unconnected fractures from brine-CO₂ flow-through tests

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ABSTRACT

CO₂ storage in fractured reservoirs may lead to fast CO₂ flow through interconnected fracture networks; but the role of isolated fractures on brine-CO₂ multiphase flow systems remains unclear. We present the results of a brine-CO₂ flow-through experiment in which we assess the change in transport properties of a synthetic sandstone plug (a surrogate of a saline siliciclastic CO₂ reservoir) containing non-connected fractures aligned 45° from its axis. The test was performed at 40 MPa of constant hydrostatic confining pressure and ~11 MPa of pore pressure, at room temperature (~19.5 °C), using pure liquid-CO₂ and 35 g L⁻¹ NaCl salt solution. The injected CO₂-brine volume fraction was increased from 0 to 1 in 0.2 units-steps (drainage). Upon achievement of the maximum CO₂ saturation (S_CO₂ ~0.6), the plug was flushed-back with the original brine (imbibition). During the test, we monitored simultaneously pore pressure, temperature, axial and radial strains, and bulk electrical resistivity. The fractured sample showed lower values of cross- and end-points in the relative permeability curves to CO₂ compared to non-fractured samples, from comparable experiments performed at similar pressure and brine salinity conditions, but different temperature. Our results suggest that a non-connected fracture network affects the mobility of the individual phases, favouring the trapping of CO₂ in the porous medium and improving the storage efficiency of the reservoir. These evidences show the need of a better understanding of fracture connectivity prior to discard fractured reservoirs as unsuitable geological formations for CO₂ storage.

1. Introduction

The success of large-scale CO₂ geosequestration and long-term storage highly depends on the capacity of the reservoir to avoid potential CO₂ leakage to the surface along permeable underground pathways. Depleted oil and gas reservoirs, saline aquifers, and coal seams have been recognized as suitable formations for CO₂ storage (IPCC, 2005). Among them, saline aquifers are the most attractive sites due to its worldwide distribution, high potential storage capacity and low reactivity to CO₂ (Bachu, 2015; Michael et al., 2010).

Enhanced chemical reactivity induced by the introduction of a non-resident fluid into rocks can also lead to significant modifications of their porosity and permeability (e.g., Canal et al., 2013). Even in reservoirs with low potential reactivity to CO₂ (e.g., sandstone), CO₂-induced salt precipitation can significantly affect the injection efficiency in the CO₂ storage site, as observed in Ketzin (Baumann et al., 2014) or in the Tubåen Formation at the Snøhvit Field (Grude et al., 2014). However, changes in permeability caused by mineral dissolution/precipitation strongly depend on the specific location where these processes occur (i.e., at open pore spaces or in necks or throats; Canal et al. (2013); Stack et al. (2014)) in connection to the main flow paths.

Rock wettability in brine-CO₂ systems affects the efficiency of the CO₂ injection process, the flow and distribution of fluids within the pore space, and the trapping capacity during CO₂ storage (Al-Menhali and Krevor, 2014). The wetting phase (brine) occupies the smallest pores while the non-wetting phase (CO₂) remains in the largest cavities (Dullien, 1992). Then, residual CO₂ saturations are higher in strongly brine-wet sandstones; while in CO₂-brine mixed-wet systems certain parts of the pore surface are preferentially wetted by one of the both fluids, leading to a lower residual trapping (Al-Khdeesawi et al., 2017). A number of factors influence the wettability of rock-fluid systems, such as...
as pressure, temperature and brine salinity (Al-Khdheawi et al., 2017). Water-wettability decreases (reciprocally, CO₂-wettability increases) with pressure and brine salinity, and increases with temperature in the presence of common sandstone minerals such as mica or quartz (Broseta et al., 2012; Chiquet et al., 2007).

Capillary pressure is a key factor impacting fluid transport in the formation and determines the ability of the wetting phase to prevent the non-wetting phase to flow through the sample. It increases with the interfacial tension (IFT), decreases with the pore radius (Chiquet et al., 2007), and also depends on wettability (e.g., Krevor et al., 2015; Wang and Tokunaga, 2015). According to Bachu and Bennion (2009), IFT increases with temperature and water salinity and decreases with pressure. Pore shape and connectivity also influence CO₂ displacement at the pore scale. The relative abundance of necks and throats in the overall rock framework constrains the capillary threshold to be exceeded to initiate CO₂ migration (Zhu et al., 2016). The heterogeneities in the pore space result in heterogeneous trapping of the non-wetting phase (Tsui et al., 2016).

The study of fluid mobility is commonly addressed by flow-through experiments under reservoir conditions in the laboratory, typically complemented with non-intrusive techniques such as computed tomography (CT) scanning (Akkarabadi and Pirsi, 2013; Krevor et al., 2012), magnetic resonance imaging (Almennen et al., 2018), or electrical resistivity (Carrigan et al., 2013; Falcon-Suarez et al., 2016; Nakatsuka et al., 2010). Electrical resistivity is also used as a remote sensing technique for monitoring the CO₂ plume advance in saline aquifers (i.e., controlled-source electromagnetic surveys – CSEM, e.g., Park et al., 2013), due to the strong resistivity contrast between brine (low resistivity) and CO₂ (high resistivity). The degree of brine saturation can be inferred from electrical resistivity using Archie’s law (Archie, 1942). The application of Archie’s law relies on the accurate resistivity estimate of the fully-saturated rock, the resistivity of pore fluid, and the shape properties of the electrically-connected porosity fraction (Cai et al., 2017). The resistivity-saturation conversion has been previously used to map CO₂ distribution in partially saturated porous media in a number of CO₂-brine flow-through tests (e.g., Alemu et al., 2013; Nakatsuka et al., 2010; Wang et al., 2009; Falcon-Suarez et al., 2016), as an alternative to X-ray CT-scanning (e.g., Berg et al., 2013; Krevor et al., 2012).

To some extent, all CO₂ storage formations are fractured reservoirs (Iding and Ringrose, 2010). Therefore, fracture density and distribution, orientation and connectivity need to be precisely assessed to guarantee safe and long-term geological storage of CO₂ (Bond et al., 2013). Different models have been proposed to describe multiphase flow through an individual fracture (e.g., Huo and Benson (2016)), and validated from experimental studies on artificial samples with ideal smooth and parallel surfaces (Dionampo, 2001; Fourar and Bories, 1995). However, experimental studies on natural rock samples with a single fracture highlight the limitations of the proposed models when upsampling results to multiphase flow on real fractured reservoir formations (Norriak et al., 2015).

Assessing the fracture connectivity effect on multiphase flow is challenging both in nature and laboratory scales. Interconnected fracture networks may act as path-flow in low permeability formations (Nooriaepour et al., 2018). In principle, the high hydraulic conductivity of the fractures would lead to preferential path-flows for CO₂, diminishing the storage effectiveness in the rock matrix and hampering the CO₂ migration control (March et al., 2018). However, when a high density of non-connected fractures exists in the reservoir, their effect on the hydrodynamic behaviour of CO₂ storage systems is unclear, and yet to be investigated.

In this contribution, we deepen into this aspect and present the results of a brine-CO₂ flow-through experiment performed on a synthetic sandstone sample containing well-distributed non-connected fractures aligned at 45° from its axis. We focus on assessing the transport properties of non-connected fractured saline siliciclastic CO₂ storage reservoirs, by analysing CO₂-induced changes in electrical resistivity and relative permeability. The experiment is complemented with (i) post-test thin section observations, (ii) reactive transport modelling, and (iii) comparison with previous experiments performed under similar conditions but using non-fractured synthetic sandstone samples.

2. Materials and methods

2.1. Synthetic sandstone

We used a 50 mm diameter 20 mm length silica-cemented synthetic sandstone plug, with spread aligned fractures oriented at 45° from its axis. The sample belongs to the same batch used by Tillotson et al. (2012), and later used by Amalokwu et al. (2015) to study water saturation and stress effects on shear wave splitting at oblique angles, using de-ionized water (DIW) as saturating fluid. Specific details concerning its preparation are given therein, but we explain here the main aspects for completeness. During the manufacturing process, aligned fractures were conformed in a parallelepiped block at the time of sample manufacturing by horizontally spreading 2 mm diameter, 200 μm thick penny-shape aluminium elements (Al-discs), in four layers with a vertical separation of ~4 mm. After the thermal (manufacturing) treatment, the block was cored ensuring an angle of 45° for the Al-disc layers. The resulting sample was flushed with 10% HCl solution to etch the fractures by leaching the aluminium. The morphology of the sample was checked by X-ray CT scan imaging, from which a fracture density $\nu_f = 0.0289 \pm 0.0077$ was determined (Tillotson et al., 2012). Since $\nu_f$ is related to the number of fractures N with radius a per unit volume V as $\nu_f = N a^2 V^{-1}$ (Mavko et al., 2009), we estimate the pore volume fraction of sample 45 A corresponding to fractures is ~10%.

Before the brine-CO₂ flow-through (BCFT) test, porosity ($\phi_0$) was measured by He-pycnometry and intrinsic permeability ($k_{gas,0}$) by N₂ permeameter under low confining pressure conditions (~0.5 MPa).

Table 1 shows the main physical properties of the sample.

2.2. Experimental setup

The experimental rig (Fig. 1) is designed around a modified Hoek-Franklin type triaxial core holder (previously described in Falcon-Suarez et al. (2017)), which allows the simulation of reservoir conditions up to 65 MPa of confining and pore pressure. This system is capable to measure electrical resistivity and axial and radial strains simultaneously, during the co-injection of up to two different pore fluids.

To track the axial and radial displacements occurring during the experiment, a rosette of two 350-Ω strain gages (90° configuration) was epoxy-glued onto the lateral surface of the sample. Inside the triaxial cell, a 6 mm thick, 190 mm height nitrile-hydrogenated butadiene rubber (NHBR) sleeve isolates the sample plug from the mineral oil used to deliver confining pressure. The sleeve is equipped with 16 stainless steel electrodes arranged in two rings around the plug that provide electrical resistivity measurements, once in contact with the sample. The system uses a tetra-polar electrode configuration to minimize electrode polarization artefacts. For any single operational run, the electrical resistivity tomography (ERT) system acquires 208 individual tetra-polar measurements using various permutations of current injection and potential difference sensing electrode pairs. The whole acquisition lasts ~20 s. The gathered data are then inverted using software based upon EIDORS (Adler and Lionheart, 2006) MATLAB toolkit for a

| Table 1 Sample properties before test. |
|----------------------------------------|
| Sample | Length (mm) | Diameter (mm) | $\phi_0$ (m$^{-3}$) | $k_{gas,0}$ (mD) | $\rho_b$ (kg m$^{-3}$) |
| 45 A | 19.85 | 49.54 | 0.273 | 5.48 | 1812 |

$\rho_b$ is the bulk density.
uniform/homogeneous isotropic resistivity and heterogeneous isotropic resistivity distribution. Both inversion schemes employ a finite element forward model of the sample and electrodes (further details in North et al., 2013). For this test, the system was configured to discard the measurements at the electrodes nearby the strain gauges, providing an accurate result for the bulk resistivity but hampering the tomography inversion. Under our experimental P-T- fluid salinity conditions and sample permeability, the associated resistivity error is ~1% (for frequencies between 1 and 500 Hz), for homogenous and isotropic rock samples in the electrical resistivity range 1–100 Ωm. Above this resistivity and for the case of non-isotropic materials, the error may rise up to 5% (North et al., 2013).

Axial stress is applied through two stainless-steel platens, which allow pore fluid circulation through the sample via thru-holes. The platens are specially designed to bypass the leads from the strain gauges glued to the sides of the plug, which are then connected to a 4-channel data acquisition system (Vishay-Model D4). The experiment was conducted under hydrostatic confining stress conditions (σ1 = σ2 = σ3) automatically controlled with a dual ISCO EX-100D system. Upstream pore pressure was also controlled with an additional ISCO EX-100D system, configured to independently deliver brine and CO2. A third ISCO ED100 pump was used as backpressure regulator downstream of the sample. In order to prevent the controller from potential damage due to direct contact with the corrosive brine and CO2 solutions, the pore fluids are indirectly delivered/received using fluid transfer vessels (FTVs). For this experiment, two FTVs supplied pure liquid-CO2 and 35 g L⁻¹ NaCl deairated aqueous solution (brine), representing the resident fluid in a potential reservoir. The dry sample was placed in the saturation chamber and subjected first to brine imbibition under vacuum (~10⁻⁴ Pa), followed by a 5 MPa pore fluid pressurization to enhance the dissolution of any remaining air in the porous media. The test was performed at constant confining pressure (Pc) of 40 MPa at room temperature (~19.5 °C), while pore pressure downstream (Pdown) was set to 9.5 MPa. Because the experiment was performed under constant flow conditions, pore pressure upstream (Pup) was not constant and, on average, it attained a value of 11 ± 1 MPa during the experiment. The pore pressure difference up- and downstream (ΔPp = Pup - Pdown) of the sample, temperature (T), and axial (εa) and radial (εr) strains were measured every second. Electrical resistivity was systematically measured at least every one sample pore volume

2.3. Experimental procedure: Brine-CO2 flow-through test

The test simulates a CO2 injection scenario in a siliciclastic saline reservoir with non-connected aligned fractures. We performed a brine-CO2 flow-through (BCFT) test using the steady-state technique (Dullien, 1992), with drainage (i.e., the progressive displacement of a resident saline fluid with CO2) and imbibition (i.e., the back-displacement of CO2 by saline fluid) episodes, similar to the experimental approach described in Falcon-Suarez et al. (2018, 2017, 2016).

Before starting the experiment, the plug was oven-dried at 60 °C for 24 h and then saturated with a 35 g L⁻¹ NaCl deairated aqueous solution (brine), representing the resident fluid in a potential reservoir. The dry sample was placed in the saturation chamber and subjected first to brine imbibition under vacuum (~10⁻⁴ Pa), followed by a 5 MPa pore fluid pressurization to enhance the dissolution of any remaining air in the porous media. The test was performed at constant confining pressure (Pc) of 40 MPa at room temperature (~19.5 °C), while pore pressure downstream (Pdown) was set to 9.5 MPa. Because the experiment was performed under constant flow conditions, pore pressure upstream (Pup) was not constant and, on average, it attained a value of 11 ± 1 MPa during the experiment. The pore pressure difference up- and downstream (ΔPp = Pup - Pdown) of the sample, temperature (T), and axial (εa) and radial (εr) strains were measured every second. Electrical resistivity was systematically measured at least every one sample pore volume

![Fig. 1. Experimental setup and synthetic sandstone with non-connected fractures aligned at 45° from its axis (sample 45A).](image-url)
3. Permeability, electrical resistivity and degree of saturation

3.1. Absolute and relative permeability

The absolute permeability \( k_{abs} \) to brine was calculated at the initial brine-\( CO_2 \) flow episode \( (X_w = 1) \) before starting \( CO_2 \) injection. During this period the sample was completely saturated with brine. \( k_{abs} \) was derived using the steady-state flow method based on Darcy’s law:

\[
k_{abs} = \frac{\mu Q L}{\Delta P A}
\]

where \( \mu \) is the dynamic viscosity of the fluid \( (Pa \ s) \), \( Q \) is the volumetric flow rate \( (m^3 \ s^{-1}) \), \( \Delta P \) is the pressure drop across the sample \( (Pa) \), \( A \) and \( L \) are the cross sectional area \( (m^2) \) and the length \( (m) \) of the sample, respectively.

We calculated relative permeability to brine and \( CO_2 \) (denoted hereafter as \( k_{r,b} \) and \( k_{r,CO_2} \), respectively) by simultaneously circulating both fluids (with different volume ratios) at controlled pore pressure and flow rates, using a two-phase multiphase flow system \( \) (Dullien, 1992), as follows:

\[
k_{i,r}(S_i) = \frac{k_i \mu_i Q_i L}{\Delta P_i A_{i, mn}}
\]

In Eq. (2), the relative permeability of each phase (denoted with the subscript \( i \)) is a function of their partial contribution to the total saturation, inferred from the electrical resistivity of the sample as explained below. Partial fluid mobility is controlled by the relative permeability of each fluid phase.

3.2. Two-phase relative permeability models

Multiphase flow properties are needed to characterize the mobility of the \( CO_2 \) plume, the injection pressure, the extent of the residual \( CO_2 \) trapping and the far-field pressure distribution \( \) (Krevor et al., 2012; Miller, 2011).

Several empirical relationships for relative permeability in multiphase systems have been developed. The Brooks-Corey’s model is the one more generally used to describe relative permeability curves in water-\( CO_2 \) systems \( \) (e.g., Krevor et al. (2012); Bachu (2013)), although the Corey’s \( \) (Corey, 1954), Purcell’s \( \) (Purcell, 1949) or van Genuchten’s \( \) (van Genuchten, 1980) models are also applied \( \) (e.g. Li and Horne \( \) (2006); Oostrom et al. \( \) (2016)). Multiphase flow simulators such as TOUGHREACT \( \) (Xu et al., 2017), ICSSM \( \) (Shi et al., 2007) or METSIM2 \( \) (Shi et al., 2011), use van Genuchten and Corey models to solve relative permeabilities to water and gas, respectively.

Purcell’s model tends to give better fits for the wetting phase, while Corey’s and Brooks-Corey’s models seem to be appropriate for both phases \( \) (Li and Horne, 2006). Based on this, we compare our experimentally obtained relative permeability curves to those from the modified version of Brooks-Corey \( \) in Krevor et al. \( \) (2012) for the wetting \( \) (brine) and non-wetting \( \) (\( CO_2 \)) phases, with the Purcell and van Genuchten models for the wetting phase, and the Corey model for the non-wetting. These models use the normalized water saturation \( (S_w^*) \) to calculate the relative permeability:

\[
k_{r,i} \left( S_{w^*} \right) = \frac{S_i - S_{i, min}}{S_{i, max} - S_{i, min}}
\]

where \( S_{w^*} \) is the degree of water saturation, \( S_{i, max} \) is the residual water saturation, and \( S_{i, min} \) is the effective saturation, that is, the saturation in the sample within the range which is still available to be saturated. For drainage \( S_{r, CO_2} = 1 - S_{w^*} \), while for imbibition \( S_{r, CO_2} = 1 - S_{r, CO_2} - S_{CO_2} \), where \( S_{CO_2} \) is the residual \( CO_2 \) saturation. During impregnation, as water saturation increases, \( k_{r, CO_2} \) decreases until it reaches its minimum at \( S_{CO_2} \). The equations and parameters of each model are presented in Table 2.

3.3. Electrical resistivity

Archie’s first and second laws \( \) (Archie, 1942) allow to relate electrical resistivity to brine saturation in variably saturated clean (clay-free) reservoir rocks \( \) (Mavko et al., 2009). Archie’s first law relates the formation factor of the rock, \( F_0 \), to the porosity \( (\phi) \) of the porous medium when fully saturated, as follows:

\[
F_0 = \frac{R_w}{R_0} = \phi^{-m},
\]

where \( R_0 \) and \( R_w \) are the resistivities of the (fully saturated) rock and pore fluid, respectively, and \( m \) the cementation exponent.

Archie’s second law extends the previous relationship to partially saturated rocks, through the following expression:

\[
S_w = \left( \frac{R_w}{R_0^{1/m}} \right)^{1/n},
\]

Table 2

| Models | Wetting phase (brine, \( k_{r,b} \)) | Non-wetting phase (\( CO_2 \), \( k_{r,CO_2} \)) |
|--------|-----------------------------------|-----------------------------------------------|
| Purcell (A) | \( k_{r,b} = \left( \frac{(S_w^*)^{2 + \frac{1}{n}}}{1 - S_{w^*}^{2 + \frac{1}{n}}} \right)^{1/n} \) | \( k_{r,CO_2} = \left( \frac{(1 - S_{w^*})^{2 + \frac{1}{n}}}{1 - (S_{w^*})^{2 + \frac{1}{n}}} \right)^{1/n} \) |
| Corey (B) | \( k_{r,b} = \left( \frac{(S_w^*)^{n_k}}{1 - S_{w^*}^{n_k}} \right) \) | \( k_{r,CO_2} = \left( \frac{(1 - S_{w^*})^{n_k}}{1 - (S_{w^*})^{n_k}} \right) \) |
| Brooks-Corey (C) | \( k_{r,b} = \left( \frac{(S_w^*)^{n_k}}{1 - S_{w^*}^{n_k}} \right) \) | \( k_{r,CO_2} = \left( \frac{(1 - S_{w^*})^{n_k}}{1 - (S_{w^*})^{n_k}} \right) \) |
| van Genuchten (D) | \( k_{r,b} = \sqrt{S_{w^*} \left[ 1 - (1 - S_{w^*})^{1/n} \right]^{2}} \) | \( k_{r,CO_2} = \sqrt{S_{w^*} \left[ 1 - (1 - S_{w^*})^{1/n} \right]^{2}} \) |
where $R_a$ is the bulk electrical resistivity of the rock, and $n$ is the saturation exponent. The empirical exponents $m$ and $n$ usually adopt values close to 2 for sandstones (Mavko et al., 2009). This relationship is useful to describe fluid substitution in brine–CO$_2$–rock systems initially saturated with the conducting fluid (brine), where the resistivity increases when brine is replaced by the non-conductive CO$_2$ phase. CO$_2$ saturation ($S_{CO2}$) is computed as the reciprocal of brine saturation (i.e., $S_{CO2} = 1 - S_w$).

For a reservoir with low chemical reactivity to CO$_2$ (e.g., silicic sandstones), $R_n$ can be assumed to be constant. Then, we can normalize the bulk resistivity in terms of partial formation factors ($F_i$), as follows:

$$F = \frac{F_i}{F_n} = \frac{R_a}{R_n} = S_w.$$  

(6)

Our normalized formation factor $F$ is the inverse of the resistivity index (e.g., Dullien (1992)). Using Eq. (6), the $S_w$ in the conventional relative permeability curves can be directly replaced by the measured property ($F$), by adjusting the saturation exponent $n$ (e.g., Li (2008)). In this contribution, we analyse our experimental results from this angle, assuming that the system remains chemically non-reactive to CO$_2$–rock interaction, so that the bulk electrical resistivity of the rock is solely function of the partial saturation of each fluid phase in the porous medium.

Some models account for the excess of conductivity induced by the presence of clay minerals by introducing an additional term into Archie’s law (Mavko et al., 2009). However, here we neglect this effect due to the practical absence of clays, which is limited to the kaolinite used to make the rock and practically consumed in the manufacturing process (Tillotson et al., 2012).

4. Results

Pore pressure difference ($\Delta P_p$) progressively increases (the pressure upstream, $P_{up}$, goes up; the pressure downstream, $P_{down}$, remains constant) as the volume fraction of CO$_2$ ($X_{CO2}$) increases (Fig. 2), during the first two fractional flow episodes (up to $X_{CO2} = 0.4$). Thereafter, the inlet flow rate ($Q_{T,OL}$) is reduced by 50% down to 0.25 mL min$^{-1}$ to satisfy our imposed condition of $\Delta P_p < 2$ MPa and keep the CO$_2$ variations in density and viscosity below 5% across the sample. The reduction in the inlet flow rate decreases the $\Delta P_p$ for the next three drainage episodes. During imbibition, we imposed the same flow rate as that used during the first ($X_{CO2} = 0$) injection episode (i.e., 0.5 mL min$^{-1}$). However, $\Delta P_p$ is higher during imbibition than during the original drainage (100% brine) episode (i.e., more energy required to flow-through), which can be related to the presence of a residual CO$_2$ fraction in the porous medium.

During drainage, the presence of CO$_2$ increases the data dispersion of the resulting flow downstream ($Q_{T,OL}$; the subscript ‘OL’ refers to flow outlet), which decreases towards the end of each episode, eventually satisfying the condition $Q_{T,OL} = Q_w + Q_{CO2}$ (after discounting mutual phases dissolution fractions).

Assuming a good gauge-rock function during the test (i.e., no visual damage evidences), volumetric strain ($\varepsilon_v$) was inferred from axial ($\varepsilon_a$) and radial ($\varepsilon_r$) strains ($\varepsilon_v = \varepsilon_a + 2\varepsilon_r$). We observe no significant changes in the dimensions of the sample during the major part of the experiment, although some axial and minor radial increase were recorded after attaining $X_{CO2} = 0.6$. At the end of the experiment, the maximum volumetric strain computed reached ~0.13% (i.e., axial and diametrical enlargement of ~0.06 and ~0.025%, respectively; Fig. 2). At the sample scale, that corresponds to an apparent swelling of ~12 $\mu$m in both the

![Fig. 2. Brine-CO$_2$ flow-through test on 45 A synthetic sandstone with a controlled distribution of non-connected fractures. Pore pressure difference ($\Delta P_p$), temperature ($T$), partial flows for CO$_2$ ($Q_{CO2}$), brine ($Q_w$) set upstream and the resulting fluid flow outlet ($Q_{T,OL}$) downstream, axial, radial and volumetric strains ($\varepsilon_a$, $\varepsilon_r$ and $\varepsilon_v$, respectively), and electrical resistivity for seven consecutive brine-CO$_2$ injection stages denoted as volume fractions of CO$_2$ ($X_{CO2}$; see text for details), covering six drainage (from $X_{CO2} = 0$ to $X_{CO2} = 1$) and a forced imbibition ($X_{CO2} = R-0$) episodes, plotted versus pore volume (PV). Vertical dashed lines separate consecutive brine-CO$_2$ stages. Notice that we only show effective flow time; interludes between two consecutive episodes are omitted. The duration of the interludes varied from one episode to the next, as follows: $X_{CO2}(0.0-0.2) \sim 0.1 h$; $X_{CO2}(0.2-0.4) \sim 15 h$; $X_{CO2}(0.4-0.6) \sim 0.1 h$; $X_{CO2}(0.6-0.8) \sim 13 h$; $X_{CO2}(0.8-1) \sim 13 h$; and transition from drainage to imbibition, $X_{CO2}(1- R-0), \sim 40 h$.](image-url)
axial and dihedral diameters. Strains remained constant during imbibition. The magnitude of the swelling is small and rather intriguing, considering the significant confining pressure and nearly constant (average) pore pressure conditions, and the absence of swelling considerations. The magnitude of the swelling is small and rather intriguing, considering the significant confining pressure and nearly constant pore pressure conditions. The absence of swelling considerations is important for the estimation of up to one order of magnitude in relative permeability and ~5% in saturation, for our experimental conditions. This aspect limits the extrapolation of our observations regarding the fractures effect on fluid mobility and CO₂ storage capacity to samples with similar geometric characteristics tested at similar experimental conditions.

We compare our results to those obtained from three previous experiments (Falcon-Suarez et al., 2017, 2016; Papageorgiou et al., 2018) performed with non-fractured synthetic sandstones using a similar experimental approach. The datasets of these tests are available at http://www.bgs.ac.uk/discoveryMetadata/. These synthetic sandstones were made using similar manufacturing conditions and materials to those employed in this study, but with variable sand-to-cement ratios. Consequently, the resulting samples S026 (Falcon-Suarez et al., 2016), S038 (Falcon-Suarez et al., 2017) and S045 (Papageorgiou et al., 2018), have different porosities (0.26, 0.38 and 0.45, respectively) and permeabilities (1, 50 and 500 mD, respectively). Considering that all samples have the same grain size and that they were subjected to the same consolidation during manufacturing, the pore size distribution of the three non-fractured samples should be similar but with different cumulative distribution (Falcon-Suarez et al., 2019). Then, the main difference in terms of transport properties should be the presence of fractures. All the experiments were performed at the same brine salinity (35 g L⁻¹ NaCl). The main differences with respect to our test are the temperature, which was set above 31 °C and the variable pore pressure from 7 to 12 MPa (seeking for supercritical-CO₂ conditions). The latter condition caused significant data scattering in the relative permeability values (Falcon-Suarez et al., 2016). Also, although the three experiments present drainage episodes, imbibition is only available for S038 (Falcon-Suarez et al., 2017).

As above, we fit the data using power-functions of the kind \( k_{i,j} = a F^b \). The imbibition data evolution in Fig. 3 reflects that the formation factor increases with very low relative permeability variations. Since the injected brine is unsaturated in CO₂, this trend would be indicating fast CO₂ dissolution (≤3 PV, equivalent to <1 h at the experimental conditions; Fig. 2) until the residual CO₂ saturation is achieved (\( S_{CO₂} = 0.15 \)).

The specific dimensions of the sample, together with the experimental procedure applied, may be affecting our observations. We expect significant capillary end effects (Leverett, 1941) because of the low length-to-diameter ratio (slenderness) of our rock sample, which result in heterogeneous pore pressure and fluid distributions across the plug. Based on the characterization developed by Jackson et al. (2018), neglecting this boundary effect can lead to a crossing point underestimation of up to one order of magnitude in relative permeability and ~5% in saturation, for our experimental conditions. This aspect limits the extrapolation of our observations regarding the fractures effect on fluid mobility and CO₂ storage capacity to samples with similar geometric characteristics tested at similar experimental conditions.

We compare our results to those obtained from three previous experiments (Falcon-Suarez et al., 2017, 2016; Papageorgiou et al., 2018) performed with non-fractured synthetic sandstones using a similar experimental approach. The datasets of these tests are available at http://www.bgs.ac.uk/discoveryMetadata/. These synthetic sandstones were made using similar manufacturing conditions and materials to those employed in this study, but with variable sand-to-cement ratios. Consequently, the resulting samples S026 (Falcon-Suarez et al., 2016), S038 (Falcon-Suarez et al., 2017) and S045 (Papageorgiou et al., 2018), have different porosities (0.26, 0.38 and 0.45, respectively) and permeabilities (1, 50 and 500 mD, respectively). Considering that all samples have the same grain size and that they were subjected to the same consolidation during manufacturing, the pore size distribution of the three non-fractured samples should be similar but with different cumulative distribution (Falcon-Suarez et al., 2019). Then, the main difference in terms of transport properties should be the presence of fractures. All the experiments were performed at the same brine salinity (35 g L⁻¹ NaCl). The main differences with respect to our test are the temperature, which was set above 31 °C and the variable pore pressure from 7 to 12 MPa (seeking for supercritical-CO₂ conditions). The latter condition caused significant data scattering in the relative permeability values (Falcon-Suarez et al., 2016). Also, although the three experiments present drainage episodes, imbibition is only available for S038 (Falcon-Suarez et al., 2017).

As above, we fit the data using power-functions of the kind \( k_{i,j} = a F^b \).
### Table 3
Fitting parameters for Brooks-Corey (BC), Purcell (P) and van Genuchten (vG) models for the fractured sample (45 A), the BC for the three non-fractured samples (S026, S038, S045), and power-function experimental fitting for the four samples.

| Fitting parameters | Fractured sample (45 A) | Non-fractured samples (S026, S038, S045) |
|--------------------|--------------------------|------------------------------------------|
| Brooks-Corey | Nw | 7.5 | 7 | 2.8 | 4.5 |
| | NCO2 | 0.1 | 0.7 | 1.8 | 0.7 |
| Purcell | λP | 0.3 |
| van Genuchten | λvG | 0.55 |
| | S_wi | 0.1 |
| | k_r,CO2 | 0.4 |
| Experimental fit (k_r, | k_r,CO2) | a | 0.95 | 1.10^-3 | 0.97 | 3.10^-3 | 1.05 | 7.10^-5 | 1.05 | 1.19^-3 |
| b | 4.02 | 1.38 | 4.13 | -2.30 | 1.84 | -6.82 | 2.87 | -3.48 |
| R^2 | 0.97 | 0.27 | 0.91 | 0.26 | 0.90 | 0.70 | 0.88 | 0.30 |

a The values were derived from an iterative, error constrained least square fitting.

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**Fig. 4.** Brine-CO2 relative permeability (k_i) versus normalized formation factor (F). Experimental data for the brine (k_r, | k_r,CO2) phases for the three non-fractured synthetic sandstones: (a) S026; (b) S038; (c) S045 (Falcon-Suarez et al., 2017, 2016; Papageorgiou et al., 2018). The plot includes the best fit curves for the wetting (fit-k_r,w) and non-wetting (fit-k_r,CO2) phases based on the experimental data, together with the best fit curves for Brooks-Corey for the wetting (BC-k_r,w) and non-wetting phases (BC-k_r,CO2), with best-fit parameters given in Table 3. To facilitate the visualization, only one error cross bar is displayed: The horizontal bar covers the error associated with the electrical resistivity; the vertical bar, the influence of the (neglected) dissolved CO2 in the relative permeability.
for both the wetting and non-wetting phases (Table 3 and Fig. 4). Based on the results obtained for the fractured sample, we only account for Brooks-Corey to fit the non-fractured samples data. For S038 and S045 the measured relative permeabilities are higher than those from the models for intermediate \( F \) values (within the range 0.35–0.75).

5.2. Hydrodynamic modelling

In Falcon-Suarez et al. (2017), the geochemical analysis of the pore fluid revealed a gradual reduction of the major cation Na with increasing \( \text{CO}_2 \), interpreted as salt precipitation phenomena during \( \text{CO}_2 \)-injection. To assess the hydrodynamics of our test, including potential \( \text{CO}_2 \)-induced salt precipitation/dissolution effects, we performed a 2D axisymmetric radial numerical simulation of our experiment using TOUGHREACT v3.3 code (Xu et al., 2017). TOUGHREACT is a non-isothermal reactive transport code that introduces reactive chemistry into the existing multi-phase fluid and heat flow code TOUGH2 (Xu et al., 2017, 2004). TOUGHREACT uses a fluid property module called ECO2N designed for applications to \( \text{CO}_2 \) storage in saline aquifers (Pruess, 2005). This module reproduces fluid properties of \( \text{H}_2\text{O}-\text{NaCl}-\text{CO}_2 \) mixtures under temperature, pressure and salinity conditions typically encountered in storage reservoirs of interest (10 °C ≤ \( T \) ≤ 100 °C; \( P \) ≤ 60 MPa; salinity up to halite saturation). Here, we consider that the system is chemically non-reactive to \( \text{CO}_2 \)-brine-rock interaction and only solve the multiphase fluid transport processes (find the details of the modelling in Appendix A).

\( \text{CO}_2 \) extends laterally and vertically from the injection point, and after the first \( \text{CO}_2 \) injection episode (\( X_{\text{CO}_2} = 0.2 \)), the upward migration of \( \text{CO}_2 \) occurs quickly and \( S_{\text{CO}_2} \) reaches ~0.15. The maximum \( S_{\text{CO}_2} \) of ~0.35 is achieved after the last drainage episode (\( X_{\text{CO}_2} = 1 \)) (Fig. 5). The model reported no evidences of salt precipitation at the experimental conditions. The maximum \( S_{\text{CO}_2} \) obtained in the model (\( S_{\text{CO}_2} \approx 0.35 \)) significantly differs from that estimated from electrical resistivity, \( S_{\text{CO}_2} \approx 0.55 \). From this 20% difference, the combined effect of the resistivity error and the empirical saturation factor (\( n = 2 \)) used for the conversion (see above) could explain ~10% variability in saturation. The remaining 10% could be related to the pore fabric of the rock. Since the model does not reproduce the non-connected fractures, we interpret this 10% represents \( \text{CO}_2 \) preferentially trapped in the fractures (see discussion below).

6. Discussion

6.1. Hydro-mechanical interpretation

The reactivity of our synthetic sample is very low, but may be able to induce an increase in porosity and permeability (Hangx et al., 2015). Post-test porosity (\( \phi \)) and gas permeability (\( k_{\text{gas,f}} \)) were measured after a week of immersion in DIW; porosity increased from 0.273 to 0.296 (~8% increment) and permeability increased from 5.5 to 12.1 mD (~54% increment). These changes indicate rock weathering during the flow-through experiment, linked to the dissolution of mineral phases or to mechanical damage associated with crack propagation; probably both (e.g., Hangx et al., 2015).

In principle, the low-concentrated (10% HCl) acid solution used originally for dissolving aluminium disks in the sample to create the fractures, should have had a minor effect on the rock matrix, composed by quartzose grains and silica cement. However, this sample was previously used to study stress-induced changes and partial saturations effects on ultrasonic properties of fractured media (Amalokwu et al., 2015; Tillotson et al., 2012). The variable stress history applied in those experiments may have also contributed to develop localized weak points in the microstructure, resulting in grain-to-grain bounds weakening from one test to the next, explaining the porosity and permeability variations.

To characterize the microstructure of the sample, we segmented it into smaller portions to obtain two oriented thin sections: normal and parallel to the dip direction of the cracks (Fig. 6). The thin sections reveal distinct stress-induced features. For instance, section A-A’ (Fig. 6b), which represents the dip direction of the crack, illustrates that,

![Fig. 5. Distribution of CO2 saturations \( S_{\text{CO}_2} \) within the sample after five drainage episodes (from \( X_{\text{CO}_2} = 0.2 \) to \( X_{\text{CO}_2} = 1 \)). For the first drainage episode and imbibition (\( X_{\text{CO}_2} = 0 \) in both cases) the sample was fully saturated in brine. The horizontal (\( x \)) and vertical (\( z \)) axis are the sample radius and thickness, respectively. The symmetry axis of the sample is placed at \( x = 0 \), and the flow inlet (IL) and outlet (OL) points are ~21 mm far from it, at the bottom and the top of the sample, respectively.](image-url)
cracks have grown, they remain non-connected. This observation suggests the permeability changes from before to after the test are not attributed to Al-disk overlapping during the manufacturing process. This effect might be explained by the increased fracture overlap, which could lead to a more complex deformation pattern, preferentially affecting the radial deformation. For hydrostatic stress conditions the orientation of our artificial cracks (45°) should be adequate to compensate for any differences in pore pressure. However, misalignments in the manufacturing process may result in crack orientation at angles different to 45° that could lead to variable stress concentration effects along the sample, and eventually to radial strain increase.

6.2. Effects and implications of non-connected fractures in CO₂ storage reservoirs

A number of methods have been developed to measure the wettability of a rock-fluid system (Castillo et al., 2011). Among them, the permeability curves can be used to qualitatively assess strong changes in wettability, although minor differences may remain unnoticed (Dandekar, 2013). Our fractured sample (45 A) shows a relative permeability cross point at F ~0.23 (Fig. 3), which may be indicative of a weaker water-wet behaviour than the non-fractured samples (S026, S038 and S045) with F values within the range of 0.32–0.38 (Fig. 4). This observation suggests the pore morphology may affect the wettability of the sample, although the differences between the cross points may be insufficient to guarantee this behaviour. The residual trapping is also indicative of wettability. Our data for residual trapping are only available for samples S038 (Falcon-Suarez et al., 2017) and 45 A, with S_{CO2r} ~0.16 and ~0.15, respectively. These two close values would indicate similar wettability in both cases, although the lack of imbibition data for S026 and S045 limit any further interpretations.

Although water-wet rocks are desirable CO₂ reservoirs, the degree of trapping also depends on other aspects besides wettability, such as pore throat size ratio, connectivity and flow rate. The pore scale physics of trapping is governed by the competition between snap-off of wetting layers and piston-like advance (Krevor et al., 2015). During piston-like filling, a displacing phase progressively fills the pores and throats, gaining space over the other phase and forcing it to exit the pore space. Snap-off occurs as the wetting phase on the pore wall and corners swells and separates two pores filled with the non-wetting phase, disconnecting and trapping the non-wetting phase in the centre of the pores. This process only occurs if the movement of the wetting phase is prevented by very large pores (Krevor et al., 2015), as in our 45 A sample. Accordingly, we interpret that the poorly connected fracture network of our 45 A sample facilitates the isolation of the non-wetting phase and benefits the CO₂ trapping in the porous medium, when compared to non-fractured samples.

The data of the four samples compared here were obtained from experiments performed at the same brine salinity (35 g L⁻¹ NaCl), but...
different P-T conditions. The mean pore pressure observed in the 45 A test (i.e., $P_p = 11 \pm 1$ MPa) is within the upper $P_p$ range (7–12 MPa) applied for the tests performed on the three non-fractured samples S026, S038 and S045 (Papageorgiou et al., 2018). Therefore, the most significant difference between experiments is the temperature: room temperature at 19 °C for the 45 A sample, 35 °C for S026 and S045, and 32 °C for S038 (i.e., supercritical CO$_2$ conditions for the non-fractured samples). Hence, a relatively lower water wettability might be expected in the 45 A sample because the temperature difference between experiments would decrease the CO$_2$-brine interfacial tension by ~15% (Bachu and Bennion, 2009). The higher IFT of the non-fractured samples would lead to increase the convexity of the relative permeability curves (Bachu and Bennion, 2008), and to lower the endpoint relative permeability value (Bennion and Bachu, 2006). However, we observe the contrary, which suggests the IFT might have a minor effect on our results.

The end-point relative permeability to CO$_2$ significantly controls injectivity (Yoshida et al., 2016). The extremely low $k_{r,CO_2}$ in the 45 A sample suggests that the presence of fractures is affecting the mobility of the individual phases (brine and CO$_2$), when compared to the non-fractured samples. This interpretation agrees with the observations reported by Reynolds and Krevor (2017), of CO$_2$ preferentially filling high porosity layers in heterogeneous cores. In our experiment, in essence, the lower capillary entry pressure of the large pores (fractures) benefits the displacement of the wetting phase by the non-wetting one, allowing the CO$_2$ to fill the large fractures, preferentially. During imbibition, the CO$_2$ would remain trapped in the fractures due to their lower capillary pressure, provided that they are non-connected.

With certain limitations (arising from sample size and the non-random orientation of fractures), we speculate our fractured sample represents a scaled-down ideal reservoir with non-connected fractures. Our experimental results suggest that this type of reservoirs could represent a scaled-down ideal reservoir with non-connected fractures. This finding might have significant implications in the storage efficiency. Although CO$_2$ mobility would be reduced in non-connected fractured systems, the fractures would be acting as small reservoirs, enhancing the overall CO$_2$ storage efficiency of the system. Based on these observations, an ideal configuration for a potential CO$_2$ storage site could be a reservoir in which (i) the bottom part of the formation (injection point) has high CO$_2$ mobility (i.e., a porosity/permeability formation, which could contain interconnected fractures), to guarantee an adequate injectivity and distribution of the CO$_2$ plume, and (ii) the top part has non-connected fractures that would act as vertical flow barriers, helping to immobilize the CO$_2$ plume and enhancing the whole trapping efficiency of the reservoir (Fig. 7).

7. Conclusions

We have performed a brine-CO$_2$ flow-through experiment using a synthetic sandstone sample with oblique aligned cracks while monitoring electrical resistivity and relative permeability. The experimental results have been compared to previous experiments in which similar synthetic samples without cracks were tested under similar conditions. Based on the newly obtained experimental data, the integration of previous results and rationale described in the text we conclude that:

- The presence of non-connected fractures may hamper the mobility of CO$_2$ in the porous medium and slightly increase the residual CO$_2$ trapping.
- In a weak water-wet sample, large fractures may prevent the movement of the wetting phase and facilitate the partial isolation of the non-wetting phase in the centre of pores (snap-off process). Further experiments to investigate the wettability on fractured samples are required for more robust interpretations.
- Heterogeneities within the porous medium may lead to differences among the various relative permeability models. The Brooks-Corey’s model can be used to describe relative permeability curves for the wetting and non-wetting phase, in both fractured and non-fractured systems.
- The fractures remain unconnected, even though we found evidences of mechanical deformation. Associated microcracks developed at weak structural points would remain saturated in brine during CO$_2$ injection due to the higher surface tension of the wetting phase.

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Fig. 7. Schematic diagram of an ideal CO$_2$ storage reservoir, showing CO$_2$ injection and plume migration. The bottom part of the reservoir is a non-fractured formation with high CO$_2$ mobility, allowing efficient injection and distribution of CO$_2$. Above this formation, a non-connected fractured layer with high CO$_2$ trapping capacity immobilises the CO$_2$ plume.
Appendix A

Reactive transport modelling has been extensively applied to predict the behaviour of aquifers during and after CO\(_2\) sequestration (e.g., Xu and Pruess (2001); Pruess and Müller (2009); Andre et al. (2010); Sung et al. (2014)).

The TOUGHREACT code has been previously used to simulate brine-CO\(_2\) multi-flow processes in continuum idealized porous media (e.g. Pruess and Müller, 2009). We did not consider transport by molecular diffusion and performed an isothermal analysis. In addition, and although this program couples thermal, hydrologic and chemical (THC) processes, reactive transport was not considered since our siliciclastic sample is likely to remain chemically unaltered when exposed to brine and CO\(_2\).

We developed a 2D axisymmetric radial model (20 mm thickness; 25 mm radius) of 500 grid elements. Cells between 0.5 and 2 mm wide were distributed in vertical layers of 1 mm, with a mesh refinement around the injection point and flow outlet to capture more precisely the processes occurring around them. An initial equilibrium simulation was performed to establish the initial pressure and temperature conditions for the BCFT simulation. In this equilibrium simulation, the grid blocks along the right, top, and bottom boundaries are kept constant at P = 100 bar; T = 19.6 °C; salinity = 3.5%; no CO\(_2\) mass fraction. During the BCFT simulation, only the flow outlet element was kept at a constant pressure of 95 bar. The volume of the cell representing the injection point matches that of the diffusor pathway to better capture the injection velocities of the experiments. We used van Genuchten models (van Genuchten, 1980) for both water relative permeability and capillary pressure and Corey’s model (Corey, 1954) for CO\(_2\) relative permeability. Residual liquid and gas saturation values were obtained from experimental data. The simulation covers the six drainage stages and the imbibition episode. Injection rates and partial flows varied according to the experimental conditions. Table A1 shows the parameters adopted in this model.

Table A1

| Modelling parameters                          | Symbol | Value          | References         |
|----------------------------------------------|--------|----------------|--------------------|
| Rock property                                |        |                |                    |
| Sample diameter (m)                          | Ø      | 0.05           |                    |
| Sample thickness (m)                         | T      | 0.02           |                    |
| Rock grain density (kg m\(^{-3}\))           | \(\rho_n\) | 2600          |                    |
| Porosity                                     | \(\psi\) | 0.273         | Experimental       |
| Horizontal permeability (m\(^{2}\))          | \(K_h\) | \(5.410 \times 10^{-16}\) | Experimental       |
| Vertical permeability (m\(^{2}\))            | \(K_v\) | \(5.410 \times 10^{-16}\) | Experimental       |
| Rock thermal conductivity (W m\(^{-1}\) C\(^{-1}\)) | \(K\) | 2              | (Eppelbaum et al., 2014) |
| Rock specific heat (J kg\(^{-1}\) C\(^{-1}\)) | \(C\) | 840            | (Eppelbaum et al., 2014) |
| Pore compressibility (Pa\(^{-1}\))           | \(c_p\) | 8.69E-10       |                    |
| Relative permeability parameters             | \(M\)  |                |                    |
| Residual liquid saturation                   | \(S_{wri}\) | 0.457         | (Pruess and Müller, 2009) |
| Liquid saturation                            | \(S_{wi}\) | 0.1            | Experimental       |
| Residual gas saturation                      | \(S_{CO2i}\) | 0.07          | Experimental       |
| Capillary pressure parameters                | \(m - 1 - 1/n\) | \(n\) | \(1/n\) | \(1/P_0\) | \(P_{max}\) (Pa) | Liquid saturation |
| Residual liquid saturation                   | \(S_{wR}\) | 0.457          | (Pruess and Müller, 2009) |
| \(1/P_0\)                                   | 5.1E-5            | (Pruess and Müller, 2009) |
| \(P_{max}\)                                 | 1E7             |                    |
| Liquid saturation                            | 0.930          |                    |

Appendix B. Supplementary data

Supplementary data to this article can be found online at https://dx.doi.org/10.5285/abc38c58-3a69-42ed-86ac-1502509bd88c.

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