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Capillary heterogeneity in sandstone rocks during CO_2/water core-flooding experiments

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Abstract

We have successfully applied a novel experimental technique to measure drainage capillary pressure curves in reservoir rocks with representative reservoir fluids at high temperatures and pressures. The method consists of carrying out 100% CO_2 flooding experiments at increasingly higher flow rates on a core that is initially saturated with water and requires that the wetting-phase pressure is continuous across the outlet face of the sample. Experiments have been carried out on a Berea Sandstone core at 25 and 50°C and at 9 MPa pore pressure, while keeping the confining pressure at 12 MPa. Measurements are in good agreement with data from mercury intrusion porosimetry. The technique possesses a great potential of applicability due to the following reasons: (a) it can be applied in conjunction with steady-state relative permeability measurements, as it shares a very similar experimental configuration; (b) it is faster than traditional (porous-plate) techniques used for measuring capillary pressure on rock cores with reservoir fluids; (c) by comparison with results from mercury porosimetry, it allows for the estimation of the interfacial and wetting properties of the CO_2/water system, the latter being unknown for most rocks; (d) by combination with X-ray CT scanning, the method allows for the observation of capillary pressure–saturation relationships on mm-scale subsets of the rock core. The latter are of high relevance as they directly and non-destructively measure capillary pressure curve heterogeneity in sandstone rocks.

Keywords: capillary heterogeneity; multiphase flow; CO_2 sequestration;

1. Introduction

Capillary pressure and relative permeability functions are characteristic curves that, when coupled to the continuum-scale equations of motion, allow for a description of multiphase displacement processes in porous media. Deep porous geological formations are often referred to as capillary systems due to the important role played by capillary pressure in controlling fluid distribution in their porous structure: in fact, capillary pressure directly affects relative permeability and residual trapping curves; the former controls fluid displacement, while the latter describes the amount of non-wetting phase that can be effectively immobilized in the pore space of the rock. Traditionally, these properties are measured in the laboratory and are implemented into reservoir simulations to predict the behaviour at the field-scale.

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Conventional techniques to measure capillary pressure curves on whole cores and with reservoir fluids are time consuming and as a result, the available experimental data set at representative reservoir conditions is limited. With relevance to CO_2 sequestration in deep saline aquifers, measurements have been performed in the relevant temperature and pressure range on a Berea sample [1] and on unconsolidated sand [2]. In addition, there is an increasing awareness that detailed investigations are required to understand the role of the inherent heterogeneity of the rock samples used in the experiments on the measured multiphase properties. In fact, although simulation studies clearly show the importance of small (sub-core) scale heterogeneity on fluid displacement [3,4], experimental techniques are needed for quantitative observation of this phenomenon under a variety of conditions and on cores from different geological settings. In this work, a recently developed technique [5] is applied to the measurement of drainage capillary pressure curves in a sandstone core with representative reservoir fluids at high pressures and temperatures. Additionally, capillary heterogeneity is quantified through the simultaneous observation of capillary pressure-saturation relationships on mm-scale subset of the rock sample. This is done by observing fluid saturations in 3D throughout the core by using X-ray Computed Tomography (CT). Experiments have been carried out on a Berea Sandstone core at 25 and 50°C and at 9 MPa pore pressure, while keeping the confining pressure on the core at 12 MPa.

2. Materials and methods

2.1. Rock sample

A Berea sandstone sample (diameter 5.1 cm and length 9.6 cm) was used for the core-flooding experiments presented in this study. The sample is homogeneous with no apparent bedding planes and it is dominated by quartz with minor presence of iron oxide. The faces of the sample were machined flat and parallel, so as to ensure good contact with the end-caps. The sample was fired at 700 °C for 2 h to stabilize swelling clays and it was then stored in a vacuum oven at 65 °C until the beginning of the experiment. Relevant petrophysical properties of the sample are summarized in Table 1.

Table 1. Petrophysical properties of the Berea Sandstone sample used in this study.

Property		Technique
Bulk (envelope) density [g/cm^3]	2.129	MICP
Skeletal density [g/cm^3]	2.647	Helium Pycnometry
Porosity [%]	19.5	X-Ray CT
Permeability (water) [mD]	280 mD	Steady-State

Additionally, mercury injection capillary pressure (MICP) measurements were carried out on a small sub-sample (~0.5 cm^3) that was drilled from a section adjacent to the core used in the core-flooding experiments by using a Micromeritics Autopore IV (Norcorss, GA, USA) and by covering the pressure range from vacuum to 228 MPa. To compare them to the capillary pressure data obtained for the CO_2-water system (see later), the MICP measurements are converted to appropriate experimental conditions by the following equation,

$$P_{\mathrm{c,g/w}} = P_{\mathrm{c,m/a}} \frac{\sigma_{\mathrm{g/w}} \cos\theta_{\mathrm{g/w}}}{\sigma_{\mathrm{m/a}} \cos\theta_{\mathrm{m/a}}} \tag{1}$$

where P_c [kPa] is the capillary pressure, σ [mN/m] is the interfacial tension and θ [°] is the contact angle; the subscripts m/a and g/w refer to the mercury/air and gas/water system, respectively. For the mercury/air system, θ and σ take values of 40° and 485 mN/m, respectively, while for the gas/water system, a contact angle $\theta = 0°$ is assumed and the interfacial tension is treated as a fitting parameter (see Results section).

2.2. Core-flooding system

All capillary pressure measurements were performed in an experimental setup designed and built up in-house partially using commercially available components and whose details can be found elsewhere [6,7]. A series of six pumps (Teledyne Isco), a two-phase separator (TEMCO AMS-900), and various heaters allow for the injection of water and CO_2 through the core sample at controlled flow rates. The core is isolated from the confining fluid by a layered jacket (from the core outwards: heat-shrinkable Teflon/nickel foil/heat-shrinkable Teflon/viton rubber sleeve). Two high accuracy pressure transducers (Oil filled Digiquartz Intelligent Transmitter, Model 9000-3K-101) are tapped into the core holder and measure the pressure at the inlet and outlet faces of the core. An electric heater keeps the core at the desired experimental temperature. The core-holder is placed in a medical X-ray CT scanning instrument (General Electric Hi-Speed CT/i X-ray computed tomography) that is used to obtain rock porosity and fluid saturation profiles during the core-flooding experiments (see [5] for relevant equations). To this aim, the following imaging parameters were applied: a voxel dimension of about (0.5 x 0.5 x 1) mm, a tube current of 200 mA, an energy level of the radiation of 120 keV and a display field of view of 25 cm. Under these conditions, a complete scan (about 35 slices) is taken within less than two minutes.

Table 2. Properties of the fluids used in this study.

Property	25°C (9MPa)	50°C (9MPa)
Density [g/cm^3]	0.8 (CO_2) / 1.0 (water)	0.285 (CO_2) / 0.992 (water)
Viscosity [x 10^{-5} Pa.s]	7.07 (CO_2) / 88.8 (water)	0.8 (CO_2) / 54.8 (water)
Interfacial tension [mN/m]	25-30	32-36

2.3. Core-flooding experiments

In this study, experiments were conducted at two temperatures (25 and 50 °C); the core outlet pressure was held constant at 9 MPa, whereas the confining pressure was maintained at about 12 MPa, thus providing a differential radial stress of about 3 MPa on the core. CO_2 (purity of 99.9%, Praxair, Inc., USA) and tap water (salinity less than 50 mg/L) were used as the nonwetting and wetting phases, respectively, and their properties are summarized in Table 2. Prior to an experiment, a series of background scans of the rock core are taken at the experimental temperature and include a dry scan, where the pore space is filled with air at atmospheric pressure as well as two scans that are taken with the core being saturated with CO_2 and water at the experimental pressure, respectively. In the latter case, two sets of scans are taken: one with fresh water and a second one with water that has been allowed to equilibrate with CO_2 for at least 12 h at the experimental conditions by circulation in a close loop that by-passes the core holder. This has the purpose of achieving a condition of immiscible displacement during the experiments. At this point, the actual experiment starts: fluid injection is switched from CO_2-saturated water to 100% CO_2 at a given flow rate and the water pumps are stopped. At least 5 pore volumes (PV) are allowed to flow through the core and the pressure drop is continuously monitored until it reaches a stable reading. Once steady state has been reached, water production stops and water pressure within the core stabilizes at a constant value. This implies that the pressure drop measured across the core, $\Delta P = P_1 - P_2$, corresponds to the capillary pressure at the inlet face of the core [5], i.e.

$$\Delta P = P_1 - P_2 = P_c\big|_{z=0} \tag{2}$$

With a simultaneous measurement of the saturation at the inlet face of the core (by X-ray CT scanning), a direct link between saturation and capillary pressure can be established. After a CT scan is taken the flow rate is increased, and the procedure is repeated, thus allowing for the construction of a capillary pressure curve. In order to cover a sufficiently large portion of the capillary pressure curve, the imposed CO_2 flow rate ranged from 0.5 up to 50 ml/min, thus corresponding to capillary numbers $N_C = u\mu_g/\sigma$ between 1×10^{-9} and 1×10^{-6} for the lowest and highest flow rates, respectively.

3. Results

Figure 1 shows the measured pressure drop across the core as a function of the pore volumes injected for the 100% CO_2 drainage experiments conducted at 25 and 50°C, respectively. As shown in the figure, experiments have been carried out at various flow rates and at each step at least 5 pore volumes (PV) were allowed to flow through the core. Although some noise is present at the lower flow rates (u < 2 ml/min), the pressure measurements are quite stable over the whole range of conditions tested. As expected, the pressure drop increases with imposed injection flow rate: at equally large flow rates, the pressure drop measured at 25°C is consistently larger than at 50°C, as in the latter case fluid viscosities are significantly lower (Table 2).

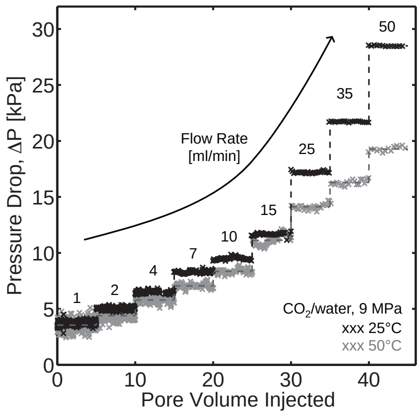

Figure 1. Pressure drop across the core, ΔP, measured at different injection flow rates, *u*, as a function of the pore volumes injected for the experiments carried out at 25 (black) and 50°C (grey). Symbols are experimental points, whereas dashed lines represent average values.

As an example of general validity, Figure 2 shows the two dimensional CO_2 saturation maps observed at the inlet face of the core for the experiment carried out at 50°C as obtained by X-ray CT scanning. In the upper row are reported the slice-averaged images, while the actual saturation (voxel size 2.5x2.5x1 mm^3) and frequency distributions are given in the second and third row, respectively. It can be easily seen that in general CO_2 saturation increases with increasing injection flow rate reaching an average saturation of about 65% at an injection rate of 50 ml/min. This behaviour is expected, as larger flow rates lead to a larger pressure drop (i.e. capillary pressure), thus allowing for the invasion of a larger portion of the pore space.

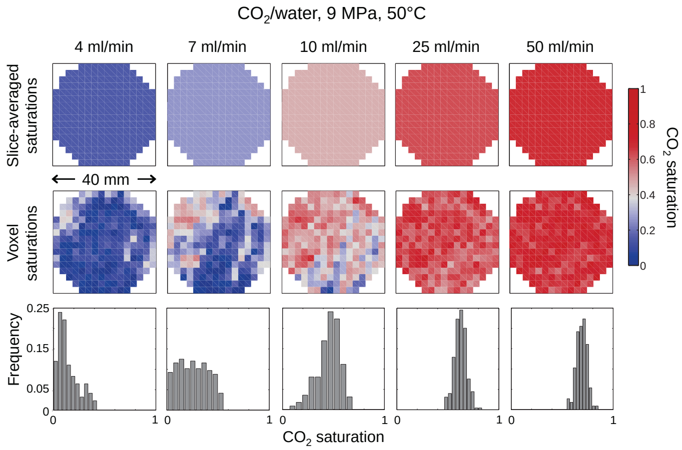

Figure 2. 2D CO_2 saturation maps at various injection flow rates observed at the inlet face of the core, i.e. where $\Delta P = P_c(z = 0)$. The upper row shows slice-averaged maps, while the second and third bottom row shows the actual saturation distribution on a grid with voxel size (2.5x2.5x1) mm^3 and the corresponding frequency distribution, respectively.

Capillary pressure curves representative for the inlet slice of the core can thus be readily obtained by combining the measured pressure drop across the core (Figure 1) with the slice-averaged inlet water saturations (Figure 2) and are shown in Figure 3. In the figure, the filled symbols are the results from the core-flooding experiments (novel technique), while the empty symbols (connected by lines to guide the eye) correspond to the converted mercury intrusion data (Equation 1). The latter have been obtained by assuming strongly water-wet conditions (i.e. $\theta = 0°$) and by using the interfacial tension σ as a fitting parameter; values of 22.3 and 34.9 mN/m have been estimated at 25 and 50°C, respectively. These values are consistent with those obtained at similar P,T conditions that are reported in the literature [8,9] and given in Table 2. It is worth pointing out that a valuable alternative would be to impose a value for the interfacial tension, while fitting the contact angle; for $\sigma = 27$ mN/m (25°C) and 35 mN/m (50°C), the obtained contact angles take values of 30° and 20°, respectively, thus confirming that these systems behave as strongly water wet. Independently of whether the interfacial tension or the contact angle is fitted, there is a very good agreement between the two techniques at both temperatures; additionally, it can be seen that for the same saturation, the capillary pressure data measured at 25°C are consistently lower that those measured at 50°C: in fact, the interfacial tension decreases with decreasing temperature due to a higher CO_2 solubility in the water.

Figure 2 shows that the spatial distribution of CO_2 within each slice varies significantly, this being particularly evident when looking at the frequency distribution plots (third row in the figure). Intuitively, this behaviour suggests that at this scale, the relation between the saturation and the capillary pressure is not unique, a phenomenon that is referred to as capillary heterogeneity. Most importantly, this phenomenon manifests itself in the range of flow rates between 4 and 10 ml/min, i.e. where the average capillary pressure curve goes through a relatively broad plateau (Figure 3); in other words, at these flow rates, small changes in capillary pressure (which might be caused by small-scales heterogeneity) lead to strong variation in the saturation.

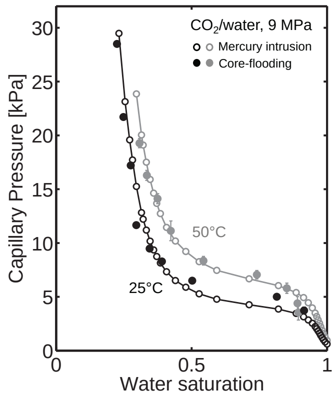

Figure 3. CO_2/water capillary pressure curves for the Berea Sandstone sample investigated in this study.

The novel technique allows obtaining sub-core (mm) scale capillary pressure curves in a straightforward manner, i.e. by linking the measured pressure drop across the core (capillary pressure) to the fluid saturation observed at the voxel scale. Results of this operation are shown in Figure 4, where the capillary pressure curves for four randomly selected voxels are plotted as a function of the water saturation for the experiment carried out at 50°C. It is evident how the mm-scale capillary pressure curves are distinct from each other, thus suggesting that at this scale there is a significant degree of heterogeneity also in an apparently homogeneous Berea Sandstone core. Additionally, it seems that despite this heterogeneity, the shape is preserved at the mm-scale of the average capillary pressure curve, as it would be expected from the Leverett J-Function scaling relationship. Saturation variations such as those shown here have been attributed to spatial variation in capillary pressure characteristic curves based on high-resolution numerical simulations [4] and the present work confirms by independent experiments the existence of these spatially varying capillary pressure curves.

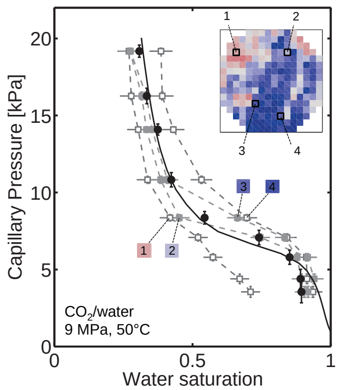

Figure 4. Capillary heterogeneity at the sub-core scale; filled black symbols represent the average capillary pressure curve (Figure 3), while sub-core (mm) scale capillary pressure curves (square grey symbols) are obtained by tracing saturation values at the voxel scale (as indicated in the figure for four randomly selected voxels).

4. Concluding remarks

CO_2/water drainage capillary pressure curves have been measured at representative reservoir pressure and temperature conditions on a Berea sandstone core by applying a recently developed experimental technique [5]. The method consists of carrying out 100% CO_2 flooding experiments at increasingly higher flow rates on a core that is initially saturated with water and requires that the wetting-phase pressure is continuous across the outlet face of the sample. Measurements are in good agreement with results from the widely applied mercury intrusion porosimetry; interestingly, such comparison allows for the estimation of the interfacial and wetting properties of the CO_2/water system. Most importantly, the technique allows for the observation of capillary pressure-saturation relationships on mm-scale subsets of the rock core. The latter are of high relevance as they directly and non-destructively measure capillary pressure curve heterogeneity in sandstone rocks.

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