Effect of sub-core scale heterogeneity on relative permeability curves of porous sandstone in a water-supercritical CO₂ system

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Abstract

Relative permeability curves of water and supercritical (sc) CO₂ were obtained for Berea sandstone and Tako sandstone under steady-state flow conditions. The present measurements provide accurate relative permeability values of water and scCO₂, which is important to better understand the behavior of CO₂ injected into the deep saline aquifer for carrying out carbon dioxide capture and storage (CCS). The mixed fluid of CO₂-saturated water and scCO₂ was injected into the rock sample that was previously saturated by CO₂-saturated water. The flow volumes of the water and scCO₂ were measured for different saturation conditions by changing the mixing ratio of the fluids at the sample inlet. The relative permeability curve is then obtained by plotting the relative permeability values with respect to the degree of water saturation estimated by mass balance methods. However, a typical fitting equation (Books and Corey, 1966) cannot adequately approximate relative permeability curves of water-scCO₂ systems. Particularly in Tako sandstone, the deviation of measured relative permeability from the calculated relative permeability curve is most likely to be due to sub-core scale heterogeneity due to inhomogeneous pore size distribution.

Keywords: relative permeability curve; steady-state method; sub-core scale heterogeneity; pore size distribution; X-ray computed tomography (CT) image

1. Introduction

Predicting the fate of injected CO₂ is a major concern for implementing carbon dioxide capture and storage (CCS). The prediction requires relative flow properties between CO₂ and water in a reservoir, which is usually a deep saline aquifer. The volumetric ratio between water and CO₂ in the saline aquifer varies widely during CO₂ migration in the aquifer. Numerical simulations of the multiphase flow in
porous media use the relationship between the fractions of two fluids and their relative permeability, which is called the relative permeability curve. In CCS, CO₂ is in the supercritical (sc) phase for most cases; therefore, for realistic simulations, we need to have an exact relative permeability curve under the scCO₂ conditions.

Relative permeability data have been obtained by steady- or unsteady-state methods. Many of them have been approximated by fitting equations to simulate the flow of CO₂ in a reservoir. For example, Corey [1], Brooks and Corey [2] and van Genuchten [3] showed equations representing relative permeability curves. Especially, the equation by Brooks and Corey [2] has been widely applied to represent the relative permeability for both end members: wetting and non-wetting fluids. However, the experimental data often deviate from fitting curves calculated by the equations. Dana and Skoczylas [4] reported that existing fitting equations tend to overestimate the relative permeability for non-wetting fluid ($k_{rnw}$), and underestimate the relative permeability for wetting fluid ($k_{rw}$), but they had no explanation for the deviation. Therefore, further studies are needed to explain the deviation.

We focused on the effect of sub-core scale heterogeneity on migration of scCO₂ so as to find the reason for the deviation. The term “sub-core scale heterogeneity” usually includes both pore size distribution and micro structures of rock associated with tectonic activities. This paper reports the relative permeability that was experimentally obtained in a water-scCO₂ system under the steady-state flow conditions in Berea sandstone and Tako sandstone. We first evaluated the deviation between experimental relative permeability and calculated relative permeability, calculated from the equation by Brooks and Corey [2] for two sandstones. Then based on another experiment using an X-ray computed tomography (CT) scanner, we revealed the characteristics of scCO₂ flow in Berea sandstone previously saturated by water. Finally we discuss the effect of sub-core scale heterogeneity due to pore size distribution on relative permeability curves.

2. Experiment 1: measurement of relative permeability

2.1. Measurement system

Figure 1 shows a measurement system. A rock sample is settled vertically in the pressure vessel. Flow rate and pressure of fluids are controlled by syringe pumps (SP1–4 in Figure 1). CO₂-saturated water (hereinafter called saturated water) and CO₂ are injected into the rock sample by SP2 and SP3. Saturated water and CO₂ flowing out from the sample are separated in a separator, and the separated CO₂ flows into SP4. Pressure transducers monitor the pressure of each fluid and differential pressure between the samples (PT1–5 in Figure 1). Temporal changes of flow rate, pressure and volume of fluids are recorded to calculate relative permeability. Temperature of fluids is measured by thermocouples. Flexible heaters are used for the separator, pressure vessel and pipes for fluids to keep the temperature constant at 40°C, so that CO₂ is in supercritical phase.
Figure 1. Schematic of the system for measuring relative permeability. “SP” and “PT” stand for syringe pump and pressure transducer, respectively. A rock sample is set at the gray part of the figure. Dotted lines referred to as “A” and “B” show the fluid discharge bypasses through which CO₂-saturated water and scCO₂ were recharged to SP2 and SP3 when the available volume of the cylinder in SP4 was filled with CO₂.

The saturation of CO₂ was estimated by the mass balance method using a pressurized and temperature-controlled separator. The separator has an observation window. Changes of the level of the contact surface between saturated water and CO₂ in the separator are analyzed by photos taken at intervals.

2.2. Rock samples

Rock samples were Berea sandstone and Tako sandstone with diameters of 50 mm and heights of 105 mm (Figure 2). Tako sandstone is an early Miocene sedimentary rock in Japan. Absolute permeability, \( k_{abs} \), was measured prior to the relative permeability measurement: \( 1.0 \times 10^{-14} \) m² (10 mD) for both sandstones. Porosity is 21% for Berea sandstone and 32% for Tako sandstone; therefore, pore volume is 39.9 ml and 60.3 ml, respectively.

Figure 3 shows the microstructures of both rocks. The images were taken by X-ray CT scanning with the samples laid horizontally in the X-ray CT scanner. Image contrasts indicate the CT-values reflecting the densities of pixels: light and dark areas correspond to high and low densities, respectively. CT images of cylindrical sections 1–6 in Figure 3 depict the characteristics of the internal structures: Berea sandstone has a layering structure with high and low densities extending perpendicular to the fluid flow, and Tako sandstone is the mixture of high- and low-density lamellae.

Figure 4 presents pore-size distributions of both sandstones obtained from mercury porosimetry. The small pieces of rocks were taken from the same rock masses from which the cylindrical samples were cored. The rock pieces were used for porosimetry. The shape of the pore size distribution chart for Berea sandstone shows a sharp peak around 10 μm in diameter. In contrast to Berea sandstone, Tako sandstone has pores with a wider diameter range. Therefore, Tako sandstone is more heterogeneous than Berea sandstone in terms of pore size distribution.
Figure 2. Photos of Berea sandstone and Tako sandstone used in this study. A rock sample equipped with end pieces is coated with silicone rubber to prevent the pressure medium (machine oil) intruding into the rock sample.

Figure 3. X-ray CT images of (a) Berea sandstone and (b) Tako sandstone set horizontally in the X-ray CT scanner. The left images show the sectional views, and the images 1–6 show the axial views.

Figure 4. Pore size distribution of (a) Berea sandstone and (b) Tako sandstone. Most of the pores are around 10 μm for Berea sandstone whereas the pores in Tako sandstone have a wide range of diameters.
2.3. Measurement procedure

Relative permeability measurements are made by injecting saturated water and supercritical CO₂ into the rock sample simultaneously. Measurement conditions are 12 MPa for confining pressure, 10 MPa for injection pressure, 9.85 MPa for back pressure and the maximum flow rate of injection fluids is 0.5 ml/min in total. Injection of water and supercritical CO₂ is continued until the flow in the sample become steady-state; that is, the differential pressure between the rock sample and the ratio of water and CO₂ in the inlet and outlet fluids from the sample become equal and constant. After the achievement of steady-state flow, relative permeability is measured. The measurements are continued successively with changing the volume ratios of water and CO₂.

3. Experiment 2: observation of CO₂ flow in Berea sandstone due to X-ray CT scanning

3.1. Measurement system and rock sample

Another experiment using an X-ray CT scanner was conducted to understand the characteristics of CO₂ flow in Berea sandstone. The measurement system is almost the same as shown in Figure 1 with the exception that an X-ray CT scanner is added to the system. Berea sandstone, with a diameter of 35 mm and height of 70 mm (Figure 5), is laid horizontally in the pressure vessel placed inside the X-ray CT scanner. The core has the layering structure as shown in Figure 5, and the layering plane is set vertically. The dotted line in Figure 5 represents the imaging section in Figure 7.

Figure 5. Berea sandstone used in the experiment with an X-ray CT scanner. The dotted line corresponds to the imaging section in Figure 7.

3.2. Procedure

The core is initially saturated with CO₂-saturated water in which a contrast medium is dissolved. Then, scCO₂ is injected twice from opposite directions in different time periods. Saturated water is injected after the first injection of scCO₂ to wash out the scCO₂ from the core. After the injection of saturated water, water saturation in the core is almost the same as in the initial condition. This is confirmed by comparing the CT-values. Measurement conditions are 12 MPa for confining pressure, 10 MPa for injection pressure, 9.85 MPa for back pressure and the maximum flow rate of the injection fluids is 0.5 ml/min.
4. Results and discussions

4.1. Relative permeability

Relative permeability is calculated by Darcy’s law:

\[ Q_i = k_i k_{abs} \frac{A}{\mu_i} \left( \frac{\Delta P}{L} - \rho_i g \right) \quad (i = w \text{ or } \text{CO}_2), \tag{1} \]

where \( Q_i \) is the flow rate of each fluid (m\(^3\)/s), \( k_i \) is the relative permeability, \( k_{abs} \) is the absolute permeability (m\(^2\)), \( A \) is the sectional area of sample (m\(^2\)), \( L \) is the length of sample (m), \( \Delta P \) is the differential pressure across the sample (Pa), \( \mu_i \) is the viscosity of fluid (Pa\(\cdot\)s), \( \rho_i \) is the density of fluid (kg/m\(^3\)) and \( g \) is the gravitational acceleration (m/s\(^2\)). The values of \( \mu_w \) and \( \rho_w \) at 40°C and 10 MPa are \( 6.53 \times 10^{-4} \) Pa\(\cdot\)s and 996.51 kg/m\(^3\), respectively, whereas the values of \( \mu_{\text{CO}_2} \) and \( \rho_{\text{CO}_2} \) are \( 4.78 \times 10^{-5} \) Pa\(\cdot\)s and 628.61 kg/m\(^3\), respectively.

Figure 6 shows relative permeability obtained from the experiments. The values of \( k_w \) and \( k_{\text{CO}_2} \) were calculated from Equation (1) when the flow in the rock sample attained the steady-state condition. The values of \( k_w \) and \( k_{\text{CO}_2} \) were 1.0 and 0 at the start of the experiments, that is, water saturation, \( S_w = 100\% \). Then, \( k_w \) and \( k_{\text{CO}_2} \) gradually decreases and increases, respectively, with the increasing volume of CO\(_2\) injected into the rock sample.

4.2. Fitting relative permeability curves

Brooks and Corey [2] proposed the equations to calculate relative permeability curves for both wetting and non-wetting fluids, \( k_w \) and \( k_{\text{nw}} \), as:

\[ k_w \approx e^{+3\lambda}\lambda \hat{\lambda}, \tag{2} \]

\[ k_{\text{nw}} \approx e^{(-S_e \lambda)}(-S_e \lambda + \lambda) \tag{3} \]

where \( \lambda \) is an index for pore size distribution in a rock, and \( S_e \) presents effective saturation:

\[ S_e = \frac{S_w - S_{\text{wir}}}{1 - S_{\text{wir}}}, \tag{4} \]

where \( S_{\text{wir}} \) is irreducible water saturation.

The relationship between the relative permeability of water and the degree of water saturation can be fairly well approximated by the equations of Brooks and Corey [2] for both Berea and Tako sandstones (solid lines in Figure 6). On the other hand, those for scCO\(_2\) deviate from the curves calculated from the equations (dotted lines in Figure 6). Deviation is larger for Tako sandstone than for Berea sandstone. No values for \( \lambda \) and \( S_{\text{wir}} \) in equations (3) and (4) can well fit the experimental values of \( k_{\text{CO}_2} \) for Tako sandstone.
4.3. Characteristics of CO₂ flow in Berea sandstone

Two X-ray imaging experiments demonstrated that almost the same pattern of the distribution of CO₂ was observed at the final stage of the injection of scCO₂ (A4 and B4 in Figure 7) although the injection directions were opposite. The images are in the vertical plane illustrated by the dotted line in Figure 5. The flow pattern suggests the existence of several narrow pore throats which allow water/scCO₂ migration. Although the section is vertical, there is no upwelling flow pattern. This suggests that the effect of the pore throat distribution is stronger than buoyancy (gravity effect). The narrow pore throat distribution may not be controlled by the tectonic discontinuous planes but by the very small-scale differences in the sand-particle depositions because of the lack of discontinuities associated with tectonic activities in the CT images. This suggests that the sub-core scale heterogeneity due to inhomogeneous pore size distribution affects the flow of scCO₂ in water-saturated porous rocks.
4.4. Effects of sub-core scale heterogeneity on relative permeability curves

Sub-core scale heterogeneity due to inhomogeneous pore size distribution controls the flow of scCO₂ in a water-saturated rock core as mentioned above. The effect of the inhomogeneous pore size distribution is also seen in the results of the relative permeability measurements. The plots of $k_{rco2}$ for scCO₂ in Tako sandstone deviate largely from the calculated curve based on the equations of Brooks and Corey [2] (Figure 6b) while the equation fairly well approximates the $k_{rco2}$ plots in Berea sandstone. This may reflect the difference of the degree of inhomogeneous pore size distribution which is quantitatively verified by the pore size distribution (Figure 4). The inhomogeneous pore size distribution in Tako sandstone may make the paths for fluid flow more complex than Berea sandstone. Therefore, the effect of sub-core scale heterogeneity due to the inhomogeneous pore size distribution on the relative permeability curves in the water-scCO₂ system appears to be larger in Tako sandstone than in Berea sandstone.

5. Conclusions

Through the measurements of relative permeability for the water-scCO₂ system in two sandstones and the X-ray imaging experiments during scCO₂ injection into sandstone, we conclude that: (1) sub-core scale heterogeneity due to inhomogeneous pore size distribution of a rock affects the relative permeability in the water-scCO₂ system, and (2) the equations by Brooks and Corey [2] cannot always approximate relative permeability data obtained in a water-scCO₂ system. For estimating the long-term fate of CO₂ storage, relative permeability curves should be carefully applied with the small-scale heterogeneity.

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References