

A novel approach to precise evaluation of carbon dioxide flow behaviour in siltstone under tri-axial drained conditions



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ABSTRACT

Since the secure storage of CO₂ in any geological sink is largely dependent on its caprock and reservoir rock flow properties, it is necessary to check the permeability of both in assessing geological reservoirs for CO₂ storage. Siltstone is a common rock type in both. On the other hand, the highly complicated thermodynamic properties of CO₂ cause its flow behaviour through any rock mass to become highly complex. This intention of this study is to understand this complexity and to propose an accurate technique to evaluate the apparent permeability of CO₂ through siltstone under laboratory conditions, considering the possible phase transitions of CO₂ inside the rock mass. A series of tri-axial drained experiments on siltstones at room temperature was therefore performed. According to the results, the proposed method accurately predicts permeability through siltstone in tri-axial drained tests, because it more precisely considers the influence of CO₂ phase transition on its flow performance, and the permeability in the sample is separately evaluated for the liquid and gas CO₂ regions. The new approach shows that the actual CO₂ pressure distribution along the sample has a curvilinear shape. Consideration of the possible phase transition between the sample inlet and the outlet is particularly important for liquid CO₂ injection in tri-axial drained tests, due to the certainty of the occurrence of phase transition under this liquid inlet and gas outlet condition. According to the test results, the apparent permeability calculated for liquid CO₂ injection using the proposed method is around 50% lower than that evaluated using the traditional method. This suggests the importance of the use of more accurate approaches such as that proposed under such situations. In addition, CO₂ permeability in siltstone is found to be significantly increased with increasing injection pressure, and increasing the injection pressure from 3 to 6 MPa (in the gas CO₂ region) caused the CO₂ permeability in the tested siltstone to be increased from 0.00102 to 0.00228 mD at 15 MPa confining pressure, which is related to the related pore structure modification in siltstone.

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1. Introduction

As the greatest contributor, CO₂ accounts for about 64% of the total greenhouse effect and therefore has had a considerable influence on global warming, particularly since the Industrial Revolution (Bryant, 1997; Xie et al., 2015). Geological storage of CO₂ is considered to be an efficient solution to the reduction of anthropogenic CO₂ emissions and has been widely tested in the field (Riddiford et al., 2003; Fischer et al., 2013; Ren et al., 2015a). According to current practices, the main potential geological media for sub-surface storage of CO₂ are depleted oil and gas reservoirs, deep saline aquifers and unmineable coal beds (Bachu, 2000;

Baines and Worden, 2004). The potential to enhance oil and gas recovery from deep unconventional geological formations through CO₂ sequestration is an appealing feature of the process (Olsen, 2011; Perera et al., 2012b; Perera et al., 2013; Perera et al., 2015), and an incremental oil recovery of 10% was achieved after CO₂ injection in the Jilin tight oilfield (Ren et al., 2015b; Ren et al., 2016). The latest trend in using CO₂ in deep geological formations in gas/oil recovery is its use as a fracturing fluid to create hydraulic fractures in the formation to enhance production (Conti et al., 2014; He et al., 2014; Middleton et al., 2015; Wanniarachchi et al., 2015; Yang et al., 2015).

The safe storage of the injected CO₂ in any potential geological sink is greatly dependent on the CO₂'s upward migration through the caprock or low permeable rock layers lying above the formation (Li et al., 2006). In production-enhancement practices such as

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hydro-fracturing, the risk of CO₂ leakage is mainly associated with the permeability characteristics of the reservoir rock (Detournay and Cheng, 1992; Gupta and Bobier, 1998; Ishida et al., 2004; Fjar et al., 2008; Ishida et al., 2012). Reservoir permeability determines the productivity of any unconventional reservoir. In the light of all of these facts, the important role of the permeability of reservoir rock/caprock in deep earth CO₂ injection applications is clear.

Since siltstone is a common type of rock in underground reservoirs as shales and their cap-rocks (Clarkson et al., 2012), a precise understanding of CO₂ permeability behaviour in such a common rock type is certainly important for both geological CO₂ storage and CO₂-enhanced unconventional gas/oil recovery-enhancement. However, the complicated thermodynamic properties of CO₂ have become a major challenge (Perera et al., 2011a), as they cause the physical properties of CO₂ (e.g. density, compressibility) to be highly pressure/temperature-dependent. This is mainly caused by the easy phase transition of CO₂ from gaseous, liquid and supercritical states with changing pressure and temperature conditions. This even causes different phases of CO₂ to exist from the ground surface to the reservoir rock with changing pressure and temperature conditions with geological depth (Pruess, 2011). In addition, the opposite phase transitions can be expected during any CO₂ flow back or leakage into the atmosphere from geological formations. This indicates the importance of identifying the permeability characteristics of CO₂ under its various phase conditions in different reservoir rocks.

The existing experimental studies on CO₂ permeability have been performed using both steady-state and unsteady-state approaches. Of these, the steady-state condition is achieved by opening the downstream to the atmosphere while offering atmospheric downstream pressure conditions. However, in such situations super-critical or liquid CO₂ phase conditions cannot be secured downstream and there is therefore phase variation of CO₂ throughout the sample (Perera et al., 2011a). Such issues can be overcome using the unsteady-state approach, in which the downstream is closed, allowing the opportunity for downstream pressure development (Perera et al., 2012a). The calculation of the rock mass permeability in unsteady-state testing is much more complicated than in steady-state testing, and requires many assumptions to be made, adding uncertainty to the permeability calculation (Carles et al., 2007). Compared to the unsteady-state approach, the steady-state approach offers a more reliable pathway for reservoir rock mass permeability calculations (Boulin et al., 2010). However, steady-state permeability calculations may have considerable errors if the possible CO₂ phase transition through rock mass is not considered (Ranathunga et al., 2015).

This study therefore proposes a more reliable approach to the calculation of reservoir rock's apparent permeability for CO₂, following the conduct of a comprehensive set of permeability experiments on siltstone incorporating the CO₂ phase transition between liquid and gaseous phases. Dry siltstone samples were tested for permeability using a steady-state tri-axial permeability approach at room temperature. Scanning electron microscopy (SEM) analyses were also carried out to clarify the relationship between chemical constitution and pore path channel for CO₂ movement in the rock mass, and the X-ray powder diffraction (XRD) analytical technique was used to quantify the crystalline material content of the siltstone samples.

2. Experimental methodology

2.1. Sample description

Siltstone samples were collected from the Eidsvold formation,

Queensland, Australia. This formation is a mixture of sandstone, siltstone and mudstone and formed in three distinct periods (Triassic, Jurassic and Cretaceous) during the Mesozoic era. The total porosity of the dry siltstone measured using the mercury intrusion method was around 19.45% and bulk and skeletal densities were around 2.24 g/ml and 2.78 g/ml, respectively. This siltstone has around 100 nm average pore diameter, and the pore size distribution is plotted in Figs. 1–2, in terms of pore volume and area. According to these figures, the cumulative pore volume increases significantly below around 1000 nm, and the incremental pore volume peak is at around 625 nm. Therefore, the median pore diameter by volume is 625 nm (see Fig. 3). However, smaller pores contribute more to the surface area of the samples, and the incremental pore area has a clear growth below 100 nm and reaches the culmination at around 10 nm, which indicates that the median pore diameter by surface area is around 17.2 nm, which is much lower than that by volume.

It is well known that the presence of tiny clay particles is responsible for the small pore size of siltstone (Kuila and Prasad, 2011, 2013), and the XRD results showed that the tested dry siltstone is mainly composed of quartz rock mineral (weight ratio: 43%; molecular formula SiO₂) and kaolinite clay mineral (weight ratio: 40%; molecular formula Al₂Si₂O₅(OH)₄) (see Table 1). The kaolinite particles which characterise its stratified structure exist as 1:1 layers combined through tetrahedral and octahedral sheets. These tiny kaolinite particles (<2 μm) separate the quartz-quartz particle contacts and segment the space formed by quartz particle skeletons (Bergaya and Lagaly, 2013). As Fig. 3 shows, small platy kaolinite particles are located between quartz particles, which was validated by performing a chemical analysis (X-ray Spectroscopy (EDS)). The analysis showed that at point 1, the small particles are mainly composed of elements of O, Al and Si and at point 2, the large particles are only composed of elements of Si and O (see Fig. 4). The overall characteristics of the rock mass pore structure determine the low permeability of this sedimentary siltstone.

2.2. Sample preparation

The siltstone samples were cored in the Deep Earth Energy Research Laboratory in the Monash University Civil Engineering Department following the ASTM standards. The samples were first cored to a diameter of 38 mm, and then cut into 76 mm high cylinders to maintain the standard diameter-to-height ratio of 1:2. Both sides of the samples were then ground smooth to ensure the injection pressure was evenly distributed. Before permeability

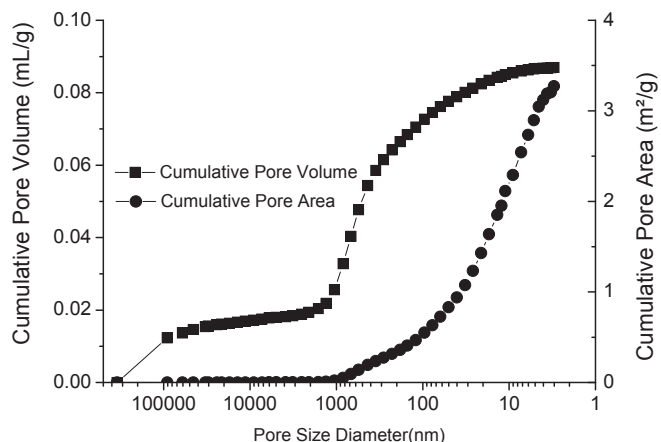


Fig. 1. Pore size distribution defined by cumulative pore volume and pore area.

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