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An experimental evaluation of unique CO_2 flow behaviour in loosely held fine particles rich sandstone under deep reservoir conditions and influencing factors

G.P.D. De Silva [a], P.G. Ranjith [a,*], M.S.A. Perera [a], Z.X. Dai [b], S.Q. Yang [c]

[a] Deep Earth Energy Research Laboratory, Building 60, Monash University, Victoria 3800, Australia
[b] Computational Earth Sciences Group (EES-16), Los Alamos National Laboratory, Los Alamos, NM 87545, USA
[c] State Key Laboratory for Geomechanics and Deep Underground Engineering, School of Mechanics and Civil Engineering, China University of Mining and Technology, Xuzhou 221116, PR China

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Lack of understanding of CO_2 flow behaviour in loosely bonded fine particles (clay and mineral fragments) rich sandstone formations has limited the optimum usage and the operational efficiency of various CO_2 injection-related field applications in these formations. A comprehensive experimental study including core flooding tests, XRD and SEM image analysis was therefore conducted precisely to understand the CO_2 flow behaviour in sandstone formations rich with loosely bonded clay and detrital particles. 210 mm long sandstone cores obtained from the Marburg Formation, eastern Australia were flooded with CO_2 at a range of temperatures (24–54 °C) and confining pressures (10–60 MPa). Pressure developments along the cores were monitored to identify the fluid migration patterns through the samples. According to the results, CO_2 permeability in tested sandstone has a high tendency to decrease with increasing injection pressure, depth (confining pressure) and temperature. Increased confining pressure and temperature caused 40–50% and 10–30% reductions in the CO_2 permeability. This is because the permeability of fine-rich sandstone is highly affected by fine particle migration associated increased flowing fluid viscosity, pore shrinkage with fine clay particle accumulations and easy compaction of soft clay minerals. Moreover, the closure of micro-cracks under high confining stresses, CO_2 adsorption created by clay swelling, the occurrence of electric double layers around clay minerals and a reduced CO_2 slip effect are also affect the permeability reduction. Many of these effects were identified in the micro-scale study conducted using SEM image analysis. Interestingly, the injection of CO_2 at higher pressures (>6 MPa) caused the pressure development in the sample to be held for a significant time due to the blocking of CO_2 flow by the accumulation of transported clay particles in pores. This pressure holding period lasts until sufficient pressure development occurs at the upstream side of the barrier to initiating a fluid flow by breaking that barrier. The findings of the study will be very useful for advances in numerical modelling and analytical equations and worldwide CO_2 geo-sequestration projects in fine-rich sandstone aquifers.

1. Introduction

Anthropogenic carbon dioxide has become the primary greenhouse gas, with its rapid increase in the atmosphere over the past several decades creating severe issues for human beings, such as rising sea levels, unexpected climate changes, melting of snow covers and lowering of ground water levels [1]. According to current records, CO_2 accounts for more than 60% of the world's total greenhouse gas emissions [2], indicating the importance of finding appropriate CO_2 emission control techniques. Geo-sequestration of carbon dioxide, the storage of carbon dioxide in deep geological formations, is considered to be one of the most feasible methods due to the widely available CO_2 sinks underground and their substantial storage capacities [3,4]. According to current research, the most preferable geological conditions for safe CO_2 storage are at around 600 m–3000 m below the earth's surface [5], where the pressure and temperature increase at gradients of about 22–27 MPa/km and 20–30 °C/km [6–8] respectively from the

* Corresponding author.
E-mail address: ranjith.pg@monash.edu (P.G. Ranjith).

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earth's surface. The physical state of CO_2 depends on pressure and temperature and therefore may vary with changing depth. For example, CO_2 density may increase from 150 kg/m^3 to 900 kg/m^3 at depths between 500 m and 1500 m [9,10], mainly caused by the change of its physical state from sub-critical gas to super-critical CO_2 (when the temperature goes beyond 31.8 °C and pressure goes beyond 7.35 MPa) or gas to liquid under sub-critical temperatures (<31.8 °C). Below super-critical conditions, CO_2 may exist as either a gas or a liquid, depending on the relative position in the P-T phase diagram [10]. Since high density is essential for optimum CO_2 storage in sedimentary formations, the injection of high-pressure CO_2 (either liquid or super-critical) is preferred in geo-sequestration processes due to the high density values compared to gaseous CO_2 [11]. Injected CO_2 may exist either as a sub-critical gas, super-critical gas or a sub-critical liquid, depending on the reservoir conditions [8].

CO_2 flow behaviour in reservoir rock after injection is very important for successful CO_2 geo-sequestration, and the term "permeability" or "intrinsic permeability" can be used to quantify it. Permeability of a porous media is largely controlled by its pore dimensions, principally the pore aperture or width. According to current research, the permeability of deep reservoir rock may vary from few micro-darcies to several thousands of milli-darcies, depending on the rock type, the depth of the reservoir and the geological position of the reservoir [12,13]. A number of empirical correlations can be used to predict permeability using sample porosity. One such basic correlation can be generally expressed as [14]:

$$k = \frac{m^2}{k_0}\phi^3 \tag{1}$$

where, k is permeability, ϕ is porosity, m is hydraulic radius and k_0 is a dimensionless constant, which may vary between 2 and 3 [15]. According to Eq. (1), porosity is dependent on the microstructure of the rock matrix and greatly affects the rock mass permeability. Rock porosity more likely changes with confining stress and temperature due to the associated compaction and expansion of mineral grains and the corresponding pore structure modification [16]. According to the Kozeny equation [17], not only porosity (ϕ) but permeability (k) also has a direct relationship with specific surface (S), the ratio between the internal surface area of pores and rock, as follows:

$$k = \frac{c}{S^2}\phi^3 \tag{2}$$

where, c is a geometry-dependent function that can be expressed as follows [18]:

$$c = \left(4\cos\left(\frac{1}{3}arccos\left(\phi\frac{8^2}{\pi^3} - 1\right) + \frac{4}{3}\pi\right) + 4\right)^{-1} \tag{3}$$

According to Eq. (2), a greater surface area causes permeability reductions in rock. This is one of the main reasons for clay-rich sandstones having quite low permeability values compared to other sandstones (clay minerals generally have high surface areas). Some researchers have modified Eq. (2) and obtained Eq. (4) [19] by considering the effect of tortuosity (τ): non-straight flow paths due to uneven distribution of pores, on rock mass permeability.

$$k = \frac{\phi^3}{2\tau(1-\phi)^2 S^2} \tag{4}$$

However, most of permeability predicting equations have been derived for homogeneous material and need to be modified when employing permeability prediction in heterogeneous materials. Moreover, interactions between rock-grains and injection fluid also significantly alter the pore structure of reservoir rock in various ways, such as moisture-adsorbing swelling in clay minerals creating pore reduction [20] and possible chemical interactions between rock minerals and acidic gases like CO_2 [17]. The prediction of reservoir rock permeability has become more difficult due to the widely existing heterogeneities in these rocks including the presence of micro-cracks and fractures [21], the existence of authigenic quartz and authigenic clay depositions [22], various pore throat size distributions and low permeable strata (well-compacted clay layers) at different lithification stages [23]. As a result, low permeability may exist in highly porous sandstones [24], because porosity is simply a measure of the pore volume and does not explain pore sizes or their distribution along the sample. However, permeability measures the ability of fluids to pass through the pore structure, and therefore directly relates to the pore characteristics of the medium. For this reason, the existence of any obstacle to the flow, such as aforementioned heterogeneities, may have a significant influence on reservoir flow behaviour or permeability, and as a result permeability may unexpectedly change, even in a highly porous medium. On the other hand, different fluids behave differently when they traverse through rock pores. For example, the permeability obtained using sub-critical CO_2 may differ from that obtained for liquid or super-critical CO_2, as the movement of gas may be affected by molecular phenomena such as gas slippage [25]. The precise understanding of pore structure and, its behaviour under various pressures and temperatures for various injection fluids are therefore important if an effective injection of gas in many actual field applications is to be achieved. As a result, the estimation of flow behaviour in a selected reservoir based on laboratory experiments/numerical modelling is necessary, in the light of the extensive time and costs associated with field tests.

Current experimental studies of flow behaviour through reservoir rocks can be divided into three major categories: tri-axial tests [24,26–29], core flooding tests [30,31] and batch autoclave tests [32]. Of these, core flooding tests have been widely used by researchers due to their unique advantages over other testing techniques [30,31], including the ability to measure the pore pressure development over the sample, which is very important in understanding the fluid flow migration patterns along a core sample or in a reservoir. Laboratory core flooding tests conducted to date on gas injection in sandstone have used samples of limited size, with lengths up to 150 mm and an average sample length around 90 mm, and a mean core diameter around 35 mm [33], and only minor consideration has been given to the temperature effect on sandstone permeability in high confinement environments [24]. There is an increasing awareness of the need to experimentally understand rock heterogeneities and their influence on gas permeability, in order to improve the confidence of flow behaviour predictions using numerical simulations. According to drained tri-axial tests conducted by Shukla, et al. [27] in low CO_2 injection pressure conditions (<5 MPa), sandstone permeability reduces with increasing confining pressure and injection pressure. However, high injection pressures may cause the sandstone pore structure to be altered, having a direct influence on sandstone permeability. Nasvi, et al. [26] observed reductions in sandstone permeability for a wide range of CO_2 injection pressures (6–20 MPa), with increasing injection pressure, and the reduction was more significant when CO_2 became denser. According to these researchers, this is related to Klinkenberg's slip flow effect [34]. Contradictory results have also been observed by some researchers [35] where higher permeability for liquid (water) compared to gas (nitrogen and methane) in sandstone. According to their test results, gas permeability in sandstone decreases with increasing