The effect of CO_2 injection on caprock permeability in deep saline aquifers

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Abstract. During CO_2 injection into deep saline aquifers, the overlying caprock may be subjected to geochemical reactions which can alter the leakage pathways for injected CO_2. Thus, it is crucial to identify the supercritical CO_2 ($scCO_2$) flow behaviour via fractures in caprock and its permeability to estimate the permanence of injected CO_2. The objective of this study is to find the effect of $scCO_2$ flow on fractured caprock permeability. A fractured siltstone sample was saturated in deionized water and conducted $scCO_2$ permeability tests using a high-precision advanced core flooding apparatus under different injection pressures and confinements. Next, the siltstone sample was saturated in 10% w/w NaCl brine and conduced $scCO_2$ permeability tests as described earlier. The results show that the brine-saturated sample has low permeability compared to water-saturated siltstone sample. The reason would be the deposition of evaporites during $scCO_2$ flow through the fractured sample. This is known as CO_2 dry-out phenomenon or absorbing moisture into the $scCO_2$, making the remaining brine saturated with salts. Thus, the CO_2 back-migration through the caprock discontinuities becomes minimized due to CO_2 dry-out phenomenon, which is an advantage for the caprock integrity in deep saline aquifers. In addition, aquifers with high salinity contents show significant dry-out phenomenon because pore fluid easily becomes supersaturated with salts due to evaporation of moisture into the $scCO_2$.

Keywords: CO_2 sequestration, caprock, fracture, $scCO_2$, permeability

1 Introduction

Since the industrial revolution, the atmospheric concentration of CO_2 and other greenhouse gases has been risen, leading to global warming and climatic changes, affecting weather patterns, flora and fauna [1-5]. The new Victorian state temperature record (in Mildura) in September 2017 with a top of 37.7°C, provides the most recent evidence to prove the adverse effects caused by the atmospheric CO_2. Accordingly, the modern world is experiencing calamitous side effects such as rising of sea levels, polar ice melting, changes in global water and carbon cycles [4]. For instance, the Great Barrier Reef, which is the world's largest reef system, has currently been subjected to a significant threat by being a victim of climatic change caused ocean acidification and the rise of sea temperature. The Great Barrier Reef Marine Park Authority (GBRMPA), Australia, reveals that the sea temperature rise in 2016 caused the worst destruction of coral reefs, with the highest recorded coral bleaching in the documented history of the Great Barrier Reef. In addition, the dry condition and high temperatures due to climate change can create devastating catastrophes such as British Columbia's wildfire, which destroyed 1,079,569 acres of land on July 6, 2017. It is evident that the rise of anthropogenic CO_2 levels in the atmosphere is not only a threat to the human lifestyle but also for the survival of flora and fauna. The atmospheric CO_2 concentration will exceed 50 billion tonnes per year by 2050 if it is not controlled by any means [6]. Therefore, it is essential to yield mitigatory measures to reduce climate change and to achieve a sustainable living environment. In other words, it is essential to implement appropriate CO_2 emission control techniques to create a better place for living beings.

As a solution, carbon capture and storage (CCS) techniques have been gained special attention due to its high efficiency and long-term stability [7]. Among, sequestrating CO_2 in deep saline aquifers is one of the most promising CCS techniques in the world due to its many advantages such as extensive availability, high efficiency, long-term stability, quick and simple capture process, huge storage capacity and advantageous locations close to major CO_2 sources [8]. The injected CO_2 in deep saline aquifers lie in supercritical phase due to the temperature and pressure of the aquifer, which are greater than the critical point of CO_2. Caprock, one of the major components of deep saline aquifers plays important roles in CO_2 sequestration process such as preventing back-migration of CO_2, limiting seismic events, inhibiting contamination of CO_2 with fresh underground water, restraining ground heave, and contributing for mineral trapping [9]. However, one of

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the major concerns of underground CO_2 sequestration in deep saline aquifers is the back-migration of CO_2 through the caprock (mainly depends on caprock permeability) due to its degradation as a result of the dissolution of minerals. Thus, it is crucial to investigate the caprock permeability that highly depends on mineral reactivity due to carbonic acid created by injected CO_2. Hence, the objective of this study is to investigate the caprock permeability due to CO_2 injection in deep saline aquifers.

2 Methodology

Siltstone samples obtained from Eidsvold basin in Queensland were used as representative of caprock. The composition of the rock is 35% quartz, 53% kaolinite, 8% muscovite, 3% alunite and 1% other minerals [10]. The cylindrical siltstone sample (38 mm diameter and 76 mm long) was cored, cut and ground in Deep Earth Energy Research Laboratory (DEERL) at Monash University, according to ASTM standards (D2664). The prepared siltstone sample was oven-dried at 40 °C for 5 days to remove all moisture trapped in the rock pores and such kind of low temperature was used to avoid appearing thermal cracks. After that, the sample was subjected to create a Brazilian test as shown in Fig. 1 to make a fracture along the cylindrical sample. The fractured sample was subjected to computed tomography (CT) scanning to identify the fracture volume using the Australian Synchrotron radiation facility. The digital image processing was conducted using Avizo 9.4.0 software and the extracted fracture is shown in Fig. 2. Firstly, the median filter and then unsharp mask filter were applied to enhance the quality of the CT images. After that, the image segmentation was conducted to separate rock matrix, pores and rock fracture.

After that, the fractured sample was saturated in water until it reaches 100% degree of saturation using a desiccator under vacuum conditions as shown in Fig.3. Usually, within one month the sample becomes saturated with water, but in order to make sure its full saturation, the sample was saturated for 3 months (until the mass of the saturated sample did not change with time). After that, the permeability tests were conducted using a high-precision advanced core flooding apparatus as shown in Fig. 4. It can simulate the complex underground conditions by applying different temperatures, confining and injection pressures. The temperature is applied using an electric oven which can be heated from 20 to 150 °C and the sample is confined using water. In addition, different kinds of fluids (liquids and gases) such as water, brine, CO_2 and N_2 can be injected into the samples.

In order to inject CO_2 in the supercritical state, the gas was injected at different pressures and temperature which are greater than the critical point of CO_2 such that injection pressures were 8, 9 and 10 MPa and the temperature was maintained at 35ºC. The permeability tests were conducted under confining pressures of 15, 20, 25 and 30 MPa. After that, the sample was saturated in 10% w/w NaCl brine sample for 3 months in a desiccator and conducted $scCO_2$ permeability tests as described earlier. The permeability was calculated using a transient method called pore pressure transmission method as shown in Eq. (1) and (2) [11]. The novelty of this method is the consideration of porosity to calculate permeability since siltstone has high permeability compared to granite. Thus, the pore pressure transmission method was used rather than calculating using conventional pulse decay method. Here, infinite storage was created in the upstream by injecting $scCO_2$ at a constant flow rate while measuring the downstream pressure development by closing the downstream pressure release valve.

$$K=\alpha\varphi\mu L^2\beta_f/-\theta^2 \quad (1)$$

$$\tan\theta=\gamma/\theta \quad (2)$$

where, β_f is the fluid compressibility; K is the permeability; μ is the dynamic viscosity, L is sample length φ is the porosity (fracture volume) and γ is the ratio of pore space volume of the sample to downstream volume.

Fig. 1. Brazilian test conducted to make a fracture in the sample

Fig. 2. The extracted fracture using CT scanning

3 Results and discussion

The calculated transient permeability values for water-saturated siltstone sample under different confining pressures are shown in Fig. 5. Accordingly, the permeability increases with increasing injection pressures because the effective stresses reduce at high pore pressures. Moreover, the permeability values reduce at high confinement due to closure of flow paths as a result of high stress created on the rock sample. Fig. 6 shows the $scCO_2$ permeability values for brine saturated sample under different confinements and injection pressures. Accordingly, the $scCO_2$ permeability of brine-saturated sample is lower than water-saturated permeability values. For instance, the CO_2 permeability for water-saturated sample under 15 MPa confinement at 8 MPa injection is 0.52 µD while for brine-saturated sample is 0.03 µD. Therefore, there is 94 % CO_2 permeability reduction. The reason is subflorescence phenomenon due to CO_2 dry-out effect or the deposition of evaporites in the flow paths as a result of absorption of moisture available in rock pores into flowing $scCO_2$ [12]. Thus, the permeability of the fractures in caprock reduces due to $scCO_2$ flow and eventually it can reduce the risk of back-migration of injected CO_2 which is favourable for maintaining the caprock integrity. Moreover, the caprock integrity can be maintained very well in deep saline aquifers if the pore fluid salinity is high since it can create a significant CO_2 dry-out phenomenon [10] which eventually reduce the flow ability of injected CO_2 through caprock discontinuities.

Fig. 3. Desiccator under vacuum condition used to saturate the siltstone sample

Fig. 4. The core flooding apparatus in Monash University

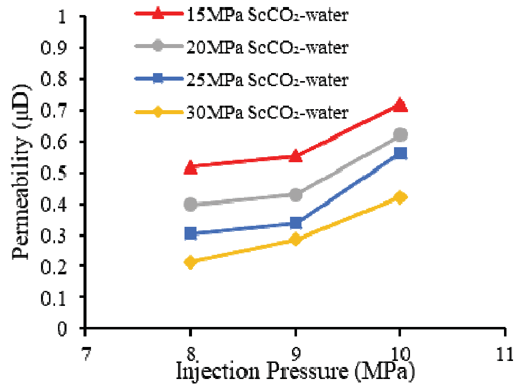

Fig. 5. The $scCO_2$ permeability of the fractured siltstone sample which is saturated with water

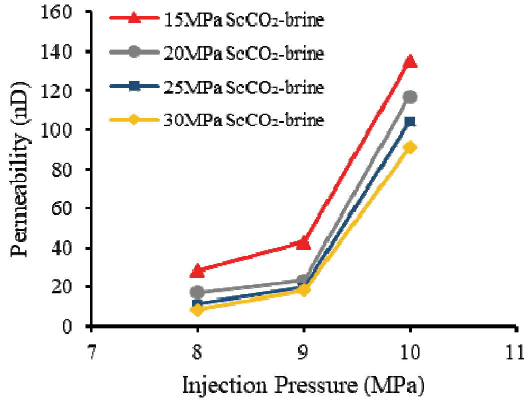

Fig. 6. The $scCO_2$ permeability of the fractured siltstone sample saturated with brine

4 Conclusion

The caprock in deep saline aquifers can be subjected to chemico-mechanical changes due to the injected CO_2. As a result, the caprocks with fractures can be appeared (due to mineral dissolution and pressure changes) which can induce CO_2 back-migration risks. Thus, the CO_2 permeability in fractured caprock was investigated in the current study using CT scanning and high-precision advanced core flooding apparatus. For that, CO_2 permeability tests for both water-saturated and brine-saturated samples were conducted to compare the flow ability.

According to the results, the $scCO_2$ permeability of the fractured caprocks in deep saline aquifers is low due to CO_2 dry-out phenomenon, which is an advantage for the caprock. In other words, CO_2 dry-out phenomenon is the evaporation of moisture into the $scCO_2$ making the remaining brine super-saturated with salts. Thus, salt crystallization occurs during $scCO_2$ flow through the fractures in caprock. Moreover, the dry-out phenomenon becomes very significant if the salinity of the pore fluid is high. A conclusion from the results is that the caprock integrity can be maintained very well in an aquifer which contains high salinity pore fluids.

However, there are different types of ions in pore fluid other than Na^+ and Cl^- in brine. Therefore, it is crucial to find the influence of those ions on salt crystallization during scCO_2 flow. Therefore, further permeability studies considering this variable are recommended as a future work along with chemical analysis to predict the influence of ions on CO_2 dry-out phenomenon.

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