

Experimental Investigation of Interactions of CO₂-Brine-Silica Sand and their Implications on CO₂ Sequestration

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ARTICLE INFO

Article history:

Received: 15th May, 2019

Received in revised form:
30th May, 2019

Accepted: 12th December, 2019

Published: 30th December, 2019

Keywords

Climate change, Saline aquifer,
Silica sand, Two-phase flow.

ABSTRACT

CO₂ sequestration into geological formations is an effective way to solve the problem of climate change. Saline aquifer is one of the attractive formations with high capacity of CO₂ storage. The injection of CO₂ into formations with brine is a two-phase flow system in a porous media. To study this process, three-pin time domain reflectometry probes (TDR probes) were used in the experiment, simulating an actual project in a lab-scale to measure geoelectrical characterizations (electrical conductivity, σ_b and bulk dielectric constant, ϵ_b) to find their relationships with formation brine saturation (S_w). Geoelectrical characterisations with corresponding formation saturation can be fitted to three-order polynomial functions which can be used for monitoring the amount of CO₂ in formations and, if the brine saturation decreases (the amount of CO₂ in the formation increases), the geoelectrical characterisation decreases. Increasing the pressure of CO₂ makes higher value of geoelectrical characterisations but this effect is not important after half of brine in the formation is displaced by CO₂.

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Introduction

In the modern world, fossil fuels are the major power source which can release CO₂ to the atmosphere and thus lead to the problem of climate change. Over the past century, the CO₂ level in atmosphere had been increased by 39% from 280ppm to 400 ppm, resulting in global temperature increasing to 0.8°C (Leung, Caramanna, and Maroto-Valer, 2014). Carbon capture and storage (CCS) is considered as the most effective way to mitigate the problem of global warming. Geological formations such as basalt rocks, saline aquifers, oil or gas reservoirs and unmineable coal bed are attractive places to store CO₂ after being captured (Altman et al., 2014; Lemieux, 2011; Leung et al., 2014; Mathias, McElwaine, and Gluyas, 2014; Suekane, Nobuso, Hirai, and Kiyota, 2008). All geological formations used for CO₂ storage must be characterised for capacity, injectivity and confinement (Bachu and Bennion 2009).

Deep aquifers at 700–1000 m underground often have high salinity formation brines as shown in figure 1 (Leung et al., 2014; Suekane et al., 2008; Zahid et al., 2011; Zhao et al., 2014; Ziabakhsh-Ganji and Kooi, 2014). Saline aquifers have an enormous potential storage capacity compared with other geological formations such as coal seam and depleted oil and gas. CO₂ injected into aquifers can move upwards because of buoyancy which is caused by the density difference between CO₂ and brine (Altman et al., 2014; Suekane et al., 2008). Carbon dioxide is usually injected as a supercritical form into saline aquifers (Altman et al., 2014; Holloway, 2007; Khudaida and Das, 2014; Leung et al., 2014; Suekane et al., 2008; Zahid

et al., 2011). It is considered that CO₂ can change from gaseous phase to supercritical phase at about 304K and 7.4MPa. Supercritical CO₂ is denser than gaseous CO₂ but has lower density than the brine in the porous space thus supercritical CO₂ tends to become buoyant and moves up to confining layer (Altman et al., 2014; Holloway, 2007; Khudaida and Das, 2014; Suekane et al., 2008). On one hand, significant amount of works have been done on supercritical CO₂, on the other hand, gas or liquid CO₂ also required more researches because CO₂ behaves differently at various temperature and pressure in the subsurface formations. During the injection of CO₂, different trapping mechanisms such as structural trapping, residual trapping, solubility trapping and mineral trapping take place in the saline aquifers (Table 1).

The most widely used porous formations for CO₂ sequestration are silica and carbonate sands. This work is limited to silica sand. Silica sand saturated with brine to store CO₂ can be considered as two-phase flow during the injection. Ahmed and Nasrabadi (2012) made a model for CO₂ storage in saline aquifer to predict storage condition with storage time. Mekala et al. (2014) carried out experiments on the effect of silica sand size on the formation kinetics during CO₂ storage. Geo-electrical characterisations such as bulk electrical conductivity (σ_b) and bulk dielectric constant (ϵ_b) with relations to water saturation can be used to monitor the condition of the formations during the CO₂-brine system in porous structure formation.

Table 1: Four major trapping mechanisms in saline aquifers and descriptions

Trapping mechanisms	Description of mechanisms	References
Structural trapping	It occurs when CO ₂ is in the porous sedimentary layers. Because of impermeable barriers, CO ₂ is trapped with the brine in the layers.	Khudaida and Das (2014); Zahid et al. (2011)
Residual trapping	CO ₂ displaces brine as it moves through the porous rock. As the CO ₂ continues to move, brine is back and replaces some of CO ₂ . Residual CO ₂ is immobile in the rock.	Altman et al. (2014); Holloway (2007); Khudaida and Das (2014); Zahid et al. (2011)
Solubility trapping	Injected CO ₂ dissolves in the resident fluid and increases the density of the brine. New denser fluid then sinks at the bottom of the rock formation.	Altman et al. (2014); Holloway (2007); Khudaida and Das (2014); Zahid et al. (2011)
Mineral trapping	Dissolved CO ₂ reacts with the brine resulting to carbonic acid and then reacts with the minerals in the surrounding rock to form solid carbonate minerals.	Altman et al. (2014); Holloway (2007); Khudaida and Das (2014); Zahid et al. (2011)

Materials and Methods

This work simulates a condition of a real CO₂ storage project with a lab-scale experiment. The stainless steel (4cm high with 10cm diameter) was filled with 500 g silica sand, and saturated with 0.2% (w/w) sodium chloride solution. The silica sand was bought from Minerals Marketing in the UK and the physical and chemical properties are shown in Table 2. CO₂ (99.9% purity) and nitrogen used in the experiments were supplied by BOC Industrial gases in the UK.

Table 2: Physical and chemical properties of the silica sand used in the experiments

Materials	Permeability, K (m ²)	Porosity, φ (-)	Density, (kgm ⁻³)	Average particle diameter, D _p (μm)	SiO ₂ content (%)
Silica Sand	3.65*10 ⁻¹⁰	0.37	2740	946.1	99

Figure 2 is a schematic diagram of the experimental rig. Three-pin time domain reflectometry probes (TDR probes) were used to measure in-situ bulk electrical conductivity (σ_b) and bulk dielectric constant (ε_b) and, then recorded on a personal computer. TDR probes and one pressure transducer (HySense PR 140, supplied by Hydrotechnik, Germany) were located at the centre of the steel cell (4cm high with 10cm diameter) at the same level. TDR probes were inserted at the

centre of the cell and connected to a multiplexer and, then connected to an electrical impulse generator which was supplied by Campbell Scientific Ltd in the UK. Datalogger (Campbell Scientific Ltd) was used for collecting and recording the data from the cell to a computer. A weigh balance was used to measure the weight of the brine coming out from the cell. It was connected directly to the computer and its data was recorded continuously by a programme (Rscm). A heating device was used to regulate and control the temperature of the cell to ensure specific constant temperature during the experiment. A supercritical pump was also used to inject CO₂ at a particular pressure.

All experiments were carried out under designed conditions. Before putting 500 g of silica sand into the cell, a small amount of brine was pumped by a hand pump into the cell to prevent air trap. Thereafter, brine was pumped into the cell to make silica sand saturated (with 130 g brine on average) and V-5 was used to release excess brine. Before pumping CO₂ into the cell, V-2 and V-3 were closed and the pressure of the pump and back pressure were set based on the designed conditions. To start running experiments, V-3 should be opened and button 'RUN' on the pump was pressed. During the experiments, the computer kept recording the data from TDR probes and the balance.

Results and Discussion

Geoelectrical characterisation can be used to track the volume of CO₂ in the storage reservoirs. Also, it can be used to detect the leakage of CO₂ after storage. The results from this work are shown in Figures 3, 4 and 5. The Figures show the relationships between geoelectrical characterizations (electrical conductivity (σ_b) and bulk dielectric constant (ε_b)) and silica sand formation's brine/water saturation. The saturation of brine/water (S_w) was calculated by using equation (1). Effects of the pressure on CO₂-water system in porous media were shown by comparing two groups of experiments under the same temperature (Figure 5).

$$S_w = \frac{(\text{saturated brine} - \text{brine out})}{\text{saturated brine}} \quad (1)$$

The relationship between geoelectrical characterisations and brine saturation

All the figures can be thought as an evidence to monitor the amount of CO₂/brine by using electrical conductivity (σ_b) and/or bulk dielectric constant (ε_b) in a storage formation under different conditions. Figure 3 and 4 show that if brine/water saturation in the formation decreases, geoelectrical characterisations (electrical conductivity (σ_b) and bulk dielectric constant (ε_b)) will be correspondingly decreased.

From figure 5, it can be found that increasing CO₂ pressure can result into increase in geoelectrical characterisations. However, the difference between the two pressures are not significant when brine saturation is decreased to 0.5. After 50% of brine displaced by CO₂, the influence of pressure is not important and thus the geoelectrical characterisations at final stage are similar. The initial geoelectrical characterisations are similar because the brine concentration and silica sand amount are same, so significant difference appears when CO₂ with high pressure injected into the cell. Abidoye (2014) showed a similar result for the effects of pressure on CO₂ storage into silica sand formation. However, in their work, two different phases of CO₂ (gaseous and fluid) with a large pressure difference were used. They also concluded that higher pressure influences the geoelectrical characterisation in formations and the same trend was observed after 50% brine displaced by CO₂. Therefore, CO₂ pressure can influence the geoelectrical characterisations of silica sand formations.

Conclusions

The geoelectrical characterisations (electrical conductivity (σ_b) and bulk dielectric constant (ϵ_b)) were measured during the injection of CO₂ into the cell filled with silica sand saturated with brine. Simultaneously, the weight of the brine displaced by CO₂ was recorded and calculated (S_w). During the injection of CO₂ into saline aquifer, the geoelectrical characterisations decreases and can be a function of formation's brine saturation. Their relationships can be used to track the amount of CO₂ in the storage reservoir and at the same time can serve as risk assessment during the CCS process. The effect of carbon dioxide pressure is not significant when 50% brine is displaced.

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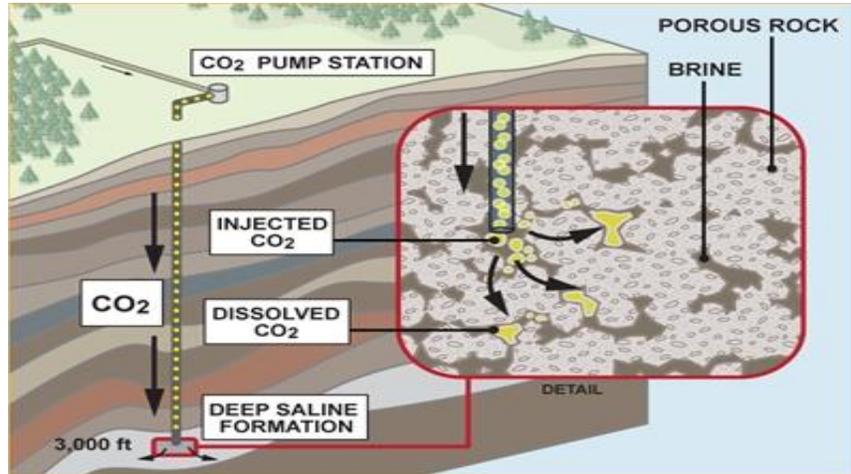


Fig. 1: Model of geological storage of carbon dioxide in saline aquifers (SoCalCarb, 2010)

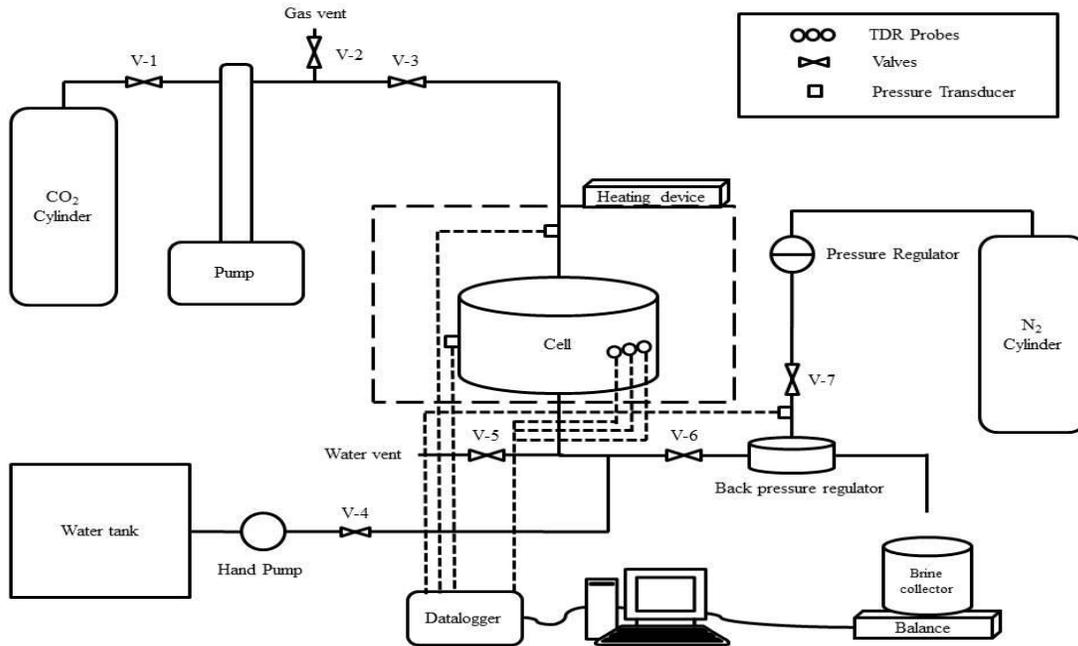


Fig. 2: A schematic diagram of the experiment rig

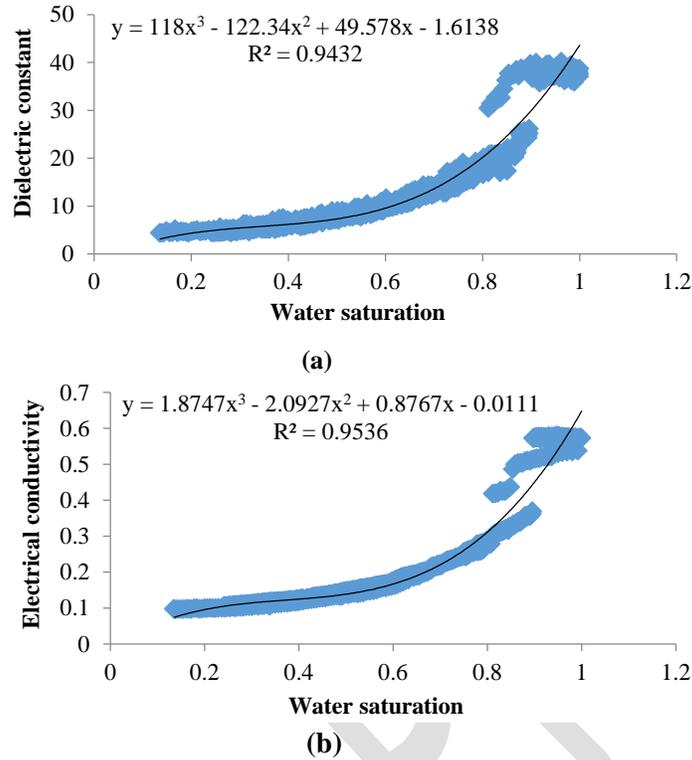


Fig. 3: (a) Dielectric constant (b) Electrical conductivity as a function of silica sand formation's brine saturation from experiments 1 and 2

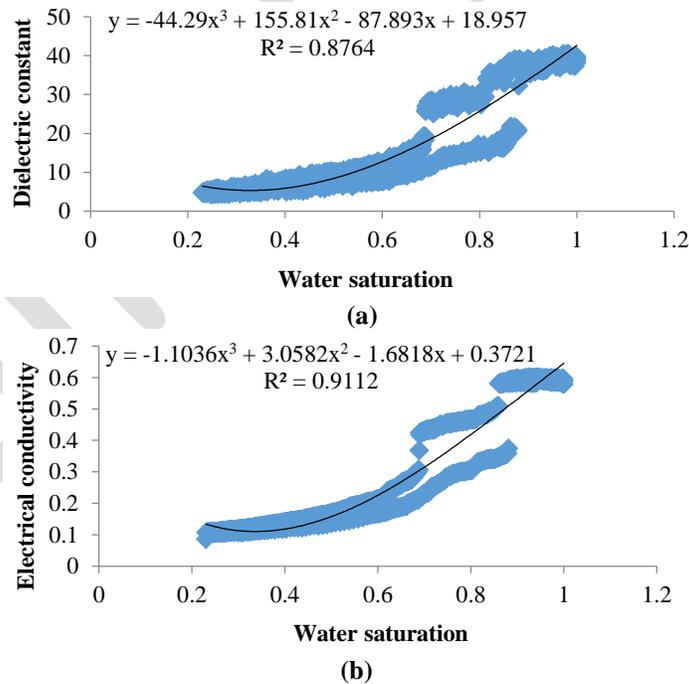
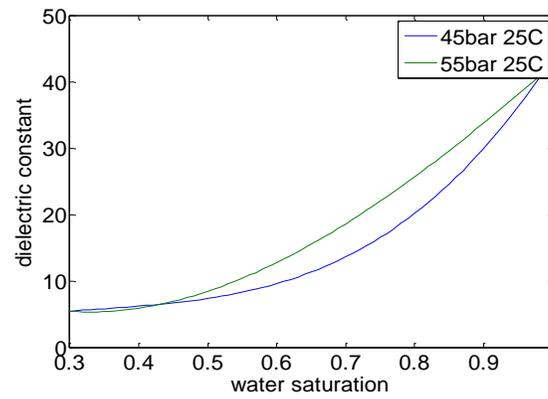
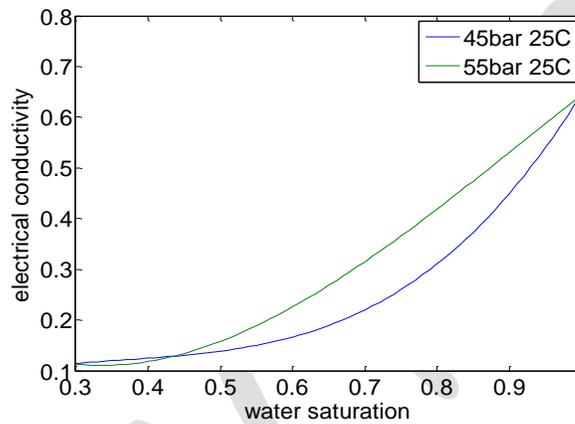


Fig. 4: (a) Dielectric constant (b) Electrical conductivity as a function of silica sand formation's brine saturation from experiments 3 and 4



(a)



(b)

Fig. 5: Comparison of (a) Dielectric constant (b) Electrical conductivity as a function of saturation under two different conditions