ORIGINAL PAPER-PRODUCTION GEOLOGY



Experimental study of CO₂ injectivity impairment in sandstone due to salt precipitation and fines migration

Muhammad Aslam Md Yusof¹ · Yen Adams Sokama Neuyam² · Mohamad Arif Ibrahim³ · Ismail M. Saaid¹ · Ahmad Kamal Idris³ · Muhammad Azfar Mohamed¹

Received: 15 July 2021 / Accepted: 1 January 2022 / Published online: 13 January 2022 © The Author(s) 2022

Abstract

Re-injection of carbon dioxide (CO₂) in deep saline formation is a promising approach to allow high CO₂ gas fields to be developed in the Southeast Asia region. However, the solubility between CO_2 and formation water could cause injectivity problems such as salt precipitation and fines migration. Although both mechanisms have been widely investigated individually, the coupled effect of both mechanisms has not been studied experimentally. This research work aims to quantify CO_2 injectivity alteration induced by both mechanisms through core-flooding experiments. The quantification injectivity impairment induced by both mechanisms were achieved by varying parameters such as brine salinity (6000–100,000 ppm) and size of fine particles (0–0.015 μ m) while keeping other parameters constant, flow rate (2 cm³/min), fines concentration (0.3 wt%) and salt type (Sodium chloride). The core-flooding experiments were carried out on quartz-rich sister sandstone cores under a two-step sequence. In order to simulate the actual sequestration process while also controlling the amount and sizes of fines, mono-dispersed silicon dioxide in CO_2 -saturated brine was first injected prior to supercritical CO_2 (sc CO_2) injection. The CO_2 injectivity alteration was calculated using the ratio between the permeability change and the initial permeability. Results showed that there is a direct correlation between salinity and severity of injectivity alteration due to salt precipitation. CO_2 injectivity impairment increased from 6 to 26.7% when the salinity of brine was raised from 6000 to 100,000 ppm. The findings also suggest that fines migration during CO_2 injection would escalate the injectivity impairment. The addition of 0.3 wt% of 0.005 µm fine particles in the CO₂-saturated brine augmented the injectivity alteration by 1% to 10%, increasing with salt concentration. Furthermore, at similar fines concentration and brine salinity, larger fines size of 0.015 µm in the pore fluid further induced up to three-fold injectivity alteration compared to the damage induced by salt precipitation. At high brine salinity, injectivity reduction was highest as more precipitated salts reduced the pore spaces, increasing the jamming ratio. Therefore, more particles were blocked and plugged at the slimmer pore throats. The findings are the first experimental work conducted to validate theoretical modelling results reported on the combined effect of salt precipitation and fines mobilisation on CO2 injectivity. These pioneering results could improve understanding of CO2 injectivity impairment in deep saline reservoirs and serve as a foundation to develop a more robust numerical study in field scale.

Keywords CO₂ injectivity · Fines migration · Salt precipitation

Muhammad Aslam Md Yusof aslam.myusof@utp.edu.my

- ¹ Block 12-03-30, Department of Petroleum Engineering, Faculty of Engineering, Universiti Teknologi PETRONAS, 32610 Seri Iskandar, Perak, Malaysia
- ² Department of Petroleum Engineering, Kwame Nkrumah University of Science and Technology, Kumasi, Ghana
- ³ School of Chemical and Energy Engineering, Universiti Teknologi Malaysia, 81310 Skudai, Johor, Malaysia

Introduction

The rising demand for oil and gas led to the search for complicated sources including shale gas, tight reservoir and highly contaminated gas & oil fields (Ibrahim and Saleh, 2020; Tewari and Sedaralit, 2021). In Malaysia, there has been an emphasis to reduce CO_2 emission from high CO_2 offshore gas fields by re-injecting the produced CO_2 after its separation from the gas stream into available saline aquifers (Jalil et al., 2012; Md Yusof et al., 2021; Zaidin et al., 2018). An estimated 37 Tscf of natural gas



remains undeveloped in Malaysia's gas fields, in which the CO_2 contents exceed 10% by volume of produced acid gas (Sukor et al., 2020). Most of these gas fields were not economically viable in the past due to the presence of high CO_2 and are always associated with potentially high corrosion risks to the surface facilities and pipelines. Presently, due to rapid development of Carbon capture and storage (CCS) technology, there is a possibility to develop up to 80% of the amount of high CO_2 gas fields in the region of South-East Asia (Hassan, Omar, Jalil, Salihuddin, & Shah, 2018; Kartikasurja, Viratno, & Sukahar, 2008).

However, because CO₂ sequestration in saline aquifers is challenged with economic constraints, its injection is usually done at the highest possible flow rate with minimum number of wells (Mathias et al., 2013). Even so, actual injection rates often fall below expectation and keeps declining over time. Field reports from CCS operations such as the Ketzin formation show that operators may experience a higher reduction in injectivity compared to the results obtained from modelling and simulations (Baumann et al., 2014; Zemke et al., 2010; Zettlitzer et al., 2010). Although reduction of injectivity is a major operational and financial challenge, less attention has been dedicated to this area. Initial research work has vaguely attributed multiphase flow to be the main cause of CO₂ injectivity impairment (Benson et al., 2011; Burton et al., 2009; Cinar & Riaz, 2014; Krevor et al., 2012; Pruess & García, 2002). Further research work has mainly concentrated on the effect of salt precipitation (Bacci et al., 2011; Guyant et al., 2015; Jeddizahed & Rostami, 2016; Miri & Hellevang, 2016; Miri et al., 2015; Muller et al., 2009; Pruess & Müller, 2009; Zeidouni, Pooladi-Darvish, & Keith, 2009) until recently where fines mobilisation, migration and entrapment were found to be one of the major contributors (Sokama-Neuyam et al., 2017a, b; Yusof et al., 2021).

Injection of CO₂ in gaseous or supercritical forms (scCO₂) may disturb the initial geochemistry of the formation fluid. At the onset of CO₂ injection, scCO₂ will dissolve and interact with the nearest reactive material which is formation brine (De Silva et al., 2015). As injection continues, two initial processes may occur; while the prevailing scCO₂ push the formation brine further into the formation leaving dry salts behind, scCO₂ nearest to the formation brine front will slowly dissolve in the brine and dissociate into carbonic acid (Andre et al., 2007; Gaus, 2010). Dissociation of CO₂ into carbonic acid could cause reactive minerals in the host rock to be dissolved into the aqueous phase. These reactive minerals are usually carbonates which makes up the cementing materials around less reactive rock grains such as quartz (Kaszuba et al., 2013; Faisal Othman et al., 2019; Faisal Othman et al., 2018a, b). Under dynamic condition, the flow of fluid at high flow rates is likely to modify the porous medium microstructure by mobilising and redepositing fine particles at the pore throats. These fast-moving particles can



clog the porous medium and may have a considerable impact on the intrinsic permeability of the rock (Md Yusof, et al., 2020a, b; Faisal Othman et al., 2018a, b).

In addition, during the injection of scCO₂ into saline aquifers, there is a mutual solubility between the CO₂ stream and formation water where formation water eventually evaporates and the molar fraction of the water in the CO₂ stream increases (Giorgis et al., 2007; Miri & Hellevang, 2016; Muller et al., 2009; Wang et al., 2018). As vaporisation of the formation water progresses, the concentration of dissolved salt in the brine builds up. When the salt concentration exceeds its solubility limit under the thermodynamic state of a given reservoir, the excess salt will precipitate out of the aqueous phase (salting-out) and alter the porosity and permeability of the formation (Cinar & Riaz, 2014; Hurter et al., 2007). Salt precipitation due to brine vaporisation during scCO₂ injection into saline aquifers is a distinct phenomenon. Microscopically, salt precipitation in porous media will reduce its porosity and permeability while macroscopically, reduction in porosity and permeability will affect further injection of scCO₂ into the aquifer (Grude et al., 2014; Md Yusof et al., 2021; Miri & Hellevang, 2016; Muller et al., 2009; Holger Ott et al., 2014; Peysson et al., 2014). Additionally, pore size reduction as a result of salt precipitation will further enhance particle entrapment, coupling its effect on CO₂ injectivity alteration (Muhammad A. et al., 2021; Sokama-Neuyam & Ursin, 2018). A schematic diagram to present the mineral dissolution, salt precipitation and fines migration mechanisms during the CO₂ injection into saline aquifer is depicted in Fig. 1.

Although CO_2 injectivity alteration by salt precipitation and fines migration has been widely accepted, the coupled effect of both mechanisms has not received the deserved attention. Currently, the most widely used porosity–permeability model to predict CO2 injectivity in current numerical simulations is Kozeny-Carman model, Hagen-Poiseuille



Fig. 1 Schematic diagram of mineral dissolution, salt precipitation and fines migration mechanisms during CO_2 injection into saline aquifer

model and Power law model (Bensinger & Beckingham, 2020; Hommel et al., 2018). The models are ascribed by porosity change due to pore modification and several parameter values were derived from the fitting of experimental data (Bacci et al., 2011; Bensinger & Beckingham, 2020; Giorgis et al., 2007; Guyant et al., 2015; H. Ott et al., 2015). In many cases, the salt precipitation has completely dominated the modelling of CO2 injectivity without considering the alteration contributed by the migration of particles. On the other hand, Sokama-Neuyam et al. (2017a, b) attempted to model the coupled effect of salt precipitation and fines migration on CO_2 injectivity alteration. Even though the results show that salt precipitation seems to augment injectivity alteration induced by fines migration, their work is completely based only on theoretical modelling without any experimental verification.

The main objective of the present work is to experimentally quantify CO_2 injectivity alteration induced by the coupled effect of salt precipitation and fines mobilisation. The present work is the pioneer to experimentally verify the idea of the coupled effect of salt precipitation and fines migration on CO_2 injectivity alteration. The findings of this work can be used to enhance understanding of the synergistical effect of salt precipitation and fines mobilisation on CO_2 injectivity. The results can also be implemented to improve future modelling work.

Materials and methods

Experimental method for evaluating injectivity of CO₂ mainly include core-flooding, using a variety of fluids during which fluid flow data are collected followed by a series of core analysis. Core-flooding experiments are conducted to simulate the actual process that took place in the reservoir as close as possible and examine the sensitivity to various types of fluids while analysing the extent of alteration. While some researchers such as Sokama-Neuyam, et al. (2017a, b); Sokama-Neuyam, et al. (2017a, b) used core-flooding experiment similar to conventional oil drainage tests to quantify permeability, F Othman, Yu, Kamali and Hussain (2018a, b) argued that permeability study through coreflooding is deemed inaccurate due to extreme fluctuation of the pressure drop during $scCO_2$ injection. This is often due to the gas-like viscosity of scCO2 as well as straining and release of fines particles. Core analysis is conducted to provide vital information on the conditions of the porous media. Common analytical methods consist of mineral compositional analysis such as X-ray diffraction (XRD) and pore analysis such as porosity and permeability test. These analytical methods are used to collect vital quantitative and qualitative data used to understand reservoir formation characteristics, reactive transport mechanisms and identify parameters affecting injectivity alteration.

Main assumptions of the work

The main objective of the present work is to investigate the physical aspects of the mechanisms of salt precipitation and fines mobilisation during CO_2 injection into deep saline reservoirs. To keep the problem simple and understand the mechanisms thoroughly, the chemical aspects have been decoupled. In view of this, the following are the main underlying assumptions of the work:

- 1. The sandstone rocks used in the study are adequately homogenous, and the pore sizes are only altered by mineral deposition during the flooding
- The immediate wellbore area where most of the injectivity impairment occurs can be adequately studied by laboratory core-flood experiments

Although mineral dissolution during CO_2 storage is a chemical process, the resulting fines mobilisation and entrapment is mainly a physical process. The dissolution mechanisms and CO_2 -brine-rock interactions have been widely studied. Salt precipitation is also a fairly physical process involving the vaporisation of formation water by dry supercritical CO_2 . Therefore, the present study provides adequate basis for investigating the coupled effect of these two mechanisms and their estimating their quantitative impact on CO_2 injectivity.

Rock and fluid samples

High quartz Berea sandstone rock was selected as the main reservoir rock because of their suitable range of permeability and porosity. Prior to the test, the core samples were cut into 7.62 cm length and diameter of 3.81 cm using dry cutting techniques to ensure no reactive clay come into contact with water. In addition, several small spotted samples were prepared for compositional and pore analysis. The cores were then cleaned using an air gun and kept in an oven at 60 °C for 24 h. The core used in this research work has an average porosity and permeability of 20% and 185 mD as per results obtained using a Poroperm unit in Core Analysis Laboratory, Universiti Teknologi PETRONAS (UTP). An XRD analysis carried out at Centralised analytical laboratory (CAL) of UTP showed that the core sample is composed of mainly quartz (90.27%), muscovite (3.84%), potassium feldspar (3.44%), kaolinite (1.65%), biotite (0.69%) and zirconia (0.11%) as shown in Fig. 2. Mercury injection capillary pressure (MICP) test was also carried out on the core samples, and summary of the results as shown in Fig. 3 show that the core sample has an average, median and modal pore diameter of 1.4, 33.8 and 36.6 µm, respectively.

To represent a simple system of formation water, three (3) brine salinities containing 6000, 30,000 and





Fig. 2 XRD mineral point matching for the Berea sandstone core used in this research work showing the composition of quartz (90.27%), muscovite (3.84%), potassium feldspar (3.44%), kaolinite (1.65%), biotite (0.69%) and zirconia (0.11%)

Fig. 3 Pore size distribution of 120 Berea sandstone used in this Cumulative pore volume Pore volume distribution research work showing average, median and mode pore diam-100 1500 eter of 1.4, 33.8 and 36.6 µm. Pore volume distribution (mm /g Cumulative pore volume (%) 80 1000 60 40 500 20 0 0 100 10000 1000 Pore diameter (nm)

100,000 ppm of sodium chloride (NaCl), respectively, and ultrapure deionised (DI) water with a resistivity $\geq 9.6 \text{ M}\Omega$ were used. CO₂ of about 99.8% purity was used as the non-wetting phase. The CO_2 is a soluble fluid in brine with varying solubility depending on temperature and pressure. Preparation of CO2-saturated brines was done using a gas-tight piston accumulator by filling 800 mL of the accumulator with NaCl brine and pressurising it with CO_2 to 800 psi (5.51 MPa). To control the amount and size of fine particles, artificially introduced monodispersed colloid suspension was prepared at 0.3 wt%



using silicon dioxide of 0.005 µm and 0.015 µm. Silicon dioxide was selected for this study because of its similar physical and chemical properties to quartz (Table 1). In addition, silicon dioxide in its raw state is highly waterwet, which would form a very stable dispersion in the suspension fluid. Typical formation fines have an average particle diameter between 0.01 and 1 µm (Khilar and Fogler, 1998). The main limitation would be the shape of the particles which is constantly spherical and are not quite similar to actual fines generated in-situ which may have irregular shapes.

respectively

Ppm

μm

Quartz	Silica oxide particles
SiO ₂	SiO ₂
Solid	Solid/powder
1650 °C	1610 °C
2230 °C	2230 °C
0.3	%wt
Insoluble	Insoluble
	Quartz SiO ₂ Solid 1650 °C 2230 °C 0.3 Insoluble

 Table 1
 Summary of physical and chemical properties of quartz and silica oxide particles

Experimental setup and methodology

CO₂ injection was done using core-flooding unit in the core analysis laboratory, UTP. Schematics of the simplified experimental setup of the equipment are shown in Fig. 4. The core-flooding unit is equipped with Hastelloy tubings and hassler type core holder allowing high-pressure, high-temperature test as well as resistance to corrosion that may arise from CO₂ exposure. The cylindrical core sample wrapped in a tight fluoroelastomer rubber sleeve was loaded into the core holder. Confining pressure of 17.2 MPa was applied in the annular space between the rubber sleeve and the core holder. The pressure at the inlet/outlet of the core sample and the overburden (confining) pressure are all measured using individual pressure transducers. Likewise, the differential-pressure across the core is measured with a differential-pressure transmitter. A continuous-flow high precision twin syringe pump was used to deliver either CO₂-saturated brine or scCO₂ from dedicated piston accumulators.

The core samples were initially dried in an oven at 60 $^{\circ}$ C for at least 24 h to remove any moisture. The experiment consists of the following steps with variables and conditions as listed in Table 2:

1. The gas permeability of each core sample was measured using Poroperm unit.

work Variables Conditions Units Confining pressure 17.2 MPa Temperature 60 °C Fines concentration 0.3 %wt Flow rate 2 cm³/min

0-100,000

0.005-0.015

Brine salinity

Average Fine particle size

Table 2 Variables and physical conditions set for the experimental

- 2. Static saturation of core with designated brine samples without fines particles.
- 3. The confining pressure and temperature of the core-flood experiment were set to 17.2 MPa and 60 °C, respectively. The following injection series was performed in the experiment.
- a. The core was saturated with designated salinity of NaCl brine (between 0 and 100,000 ppm) for 30 Pore volumes (PV) to remove the gas trapped inside the core samples, and the initial liquid permeability, k_i was measured.
- b. 65 PV CO_2 saturated brine with or without fines particles was injected into the core sample at constant injection flow rate of 2 cm³/min.
- c. scCO₂ was injected into core samples for about 70 PV at injection flow rate of 2 cm³/min. The high flow rates were selected to represent the viscous effect and approximate fluid flow velocity at the vicinity of the near wellbore.
- d. Final liquid permeability, k_f was measured with designated NaCl brine sample for about 30 PV.
- 4. The core sample was dismounted from the core holder and dried in an oven at 60 °C for 24 h.
- 5. The gas permeability of the core sample was measured



Fig. 4 Schematics of the experimental setup used for CO₂ core-flooding experiments



Estimation of injectivity alteration

Under typical field injection conditions, a significant volume of CO_2 would be injected into the storage at an acceptable rate through a minimum number of wells due to economic reasons. The attainable rate of CO_2 injection into the formation without fracturing the formation, for a homogeneous reservoir, can be expressed in terms of injectivity index, (I), defined as the ratio of volumetric injection flow rate, (q) to the pressure drop (Civan, 2015; Schembre-McCabe, Kamath, & Gurton, 2007).

$$I = \frac{q}{\Delta P} \tag{1}$$

At the laboratory scale, the fluid injectivity is normally conducted by injecting CO₂ at a certain flow rate into a core sample in a core-flooding unit. The pressure drop profiles before and after the experiment are measured and used to calculate the injectivity. In order to estimate the injectivity alteration during the injection process, a relative injectivity change (RIC) index, (β) has been used by some researchers (Sokama-Neuyam, et al., 2017a, b; Sokama-Neuyam & Ursin, 2018a, b). It is defined as the ratio of injectivity index of initial injection condition (I_i), to final injection condition (I_f), at a constant injection flow rate ($q_i = q_f$), assuming that the viscosity of the fluid (μ), core area (A) and length (L) used in the measurement is considered constant, $C = A'_{\mu L}$. Thus, RIC can be defined as





$$I_i = \frac{q}{\Delta P_i} = k_i.C \tag{2}$$

$$I_i = \frac{q}{\Delta P_f} = k_f . C \tag{3}$$

$$RIC = 1 - \left(\frac{\Delta P_i}{\Delta P_f}\right) = 1 - \left(\frac{k_f}{k_i}\right) \tag{4}$$

Any pore plugging would reduce the total flow area (*A*) and increase the core pressure drop, ΔP . Therefore, it would give $\Delta P_f > \Delta P_i$ and $k_i > k_f$ because of permeability alteration. A positive RIC value indicates injectivity impairment and vice versa. The RIC value is usually viewed as a percentage, and it is an indirect way of calculating injectivity damage that occurred in the core sample during injection, irrespective of the chemical properties.

Results

Drying and salt precipitation

Figure 5 presents the relative injectivity changes of three Berea sandstone rocks after $scCO_2$ flooding. These sandstone core samples were saturated with fresh water, 6000 ppm, 30,000 ppm and 100,000 ppm to investigate the trend and severity of salt precipitation. The results suggests that the relationship between RIC and brine salinity is approximately linear. It can be seen that there is a direct correlation between salt precipitation and RIC as the RIC increased from 5.3 to 27.3% as the brine salinity increases.

Figure 6 shows the FESEM image comparison of the sandstone core saturated with 30,000 ppm of NaCl before





Fig. 6 Comparison of FESEM image before and after CO_2 injection shows the existence of salt precipitation that covers the grain and pore spaces. EDX analysis confirmed the halite precipitation

and after the CO_2 injection series. The EDX analysis indicated that the red circles area was covered by the halite (NaCl) precipitation. The formerly existing pores were reduced in size after the CO_2 exposure. Then, the images of the injected face of the sandstone core samples saturated with different brine salinity are shown in Fig. 7. The increasing growth of salt precipitation (white marks) was observed on each sample as the brine salinity increased.

As Fig. 7 illustrates, minimal change was observed on the face of the core saturated with fresh water. However, a small amount of salt started to precipitate when 6000 ppm NaCl brine was used. For the core saturated with 30,000 ppm NaCl, almost 50% of the injection face was covered with the precipitated salt, and the amount is doubled when 100,000 ppm NaCl was used. Clearly, the precipitated salt filled in the pore spaces, and the crossflow area within the pore that is available for flow decreases. This explains why the RIC value decreased at higher brine salinity. These findings are consistent with that of Pruess and Müller (2009) who explained the linear relationship between the amount of salt precipitated into the pores, S_s and brine salinity, X_s , as;

$$S_s = (1 - S_{g,d}) \frac{\rho_{aq} X_s}{\rho_s} \tag{5}$$

As shown in Fig. 8, the effective pore spaces for fluid flow in natural reservoir rocks could be represented by a bundle of parallel cylindrical tubes with various diameters between non-porous masses which indicate the rock matrix (Liu et al., 2013; Verma and Pruess, 1988). At higher brine salinity, the thickness of the deposited salt in each capillary tube, Δr_s increased according to Eq. (6) by Sokama-Neuyam and Ursin (2018a, b). Therefore, the presence of more precipitated salt reduced the presence of the crossflow area within the pore flow. As a result, lower injectivity value was formed at higher brine salinity.

$$\Delta r_s = \frac{2}{3} \frac{S_s r_i}{l_d} \tag{6}$$

It is therefore likely that such connections exist between brine salinity and the damage contributed to the CO_2 injectivity. Moreover, the results broadly agree with the experimental work of other studies in this area linking salt





Fig. 7 Compared images of Berea sandstone core samples after salt precipitation experiments. Red arrows and circles indicated the newly formed salt precipitation. The amount of salt deposited on the injection face is directly proportional to the initial brine salinity, with the 100000 ppm NaCl brine accumulating the highest salt deposits

precipitation with injectivity alteration (Bacci et al., 2011; Jeddizahed & Rostami, 2016; Faisal Othman et al., 2019). Thus, salt precipitation could induce significant effect on CO_2 injectivity, and the extent of impairment is mainly dependent on the initial brine salinity. The injectivity impairment increases with increasing brine salinity.

Fines entrapment

Fine particles are defined as mobile particles with an average diameter smaller than 40 μ m (Torsæter & Cerasi, 2018).

Under typical continuous injection of $scCO_2$ into sandstone rocks, previous findings have reported that fine particles are mobilised due to dissolution of reactive minerals, and they are migrated together with the flowing $scCO_2$ (Md Yusof, et al., 2020a, b; F Othman et al., 2018a, b). These mobile particles could clog the rock when moving through slimmer pore throats. However, the amount and sizes of fines particles detached, migrated and entrapped are very much unknown and uncontrollable. Thus, as a method of controlling the variables, artificial fines are introduced into the system during brine injection (Fig. 9).

Initial analysis using MICP (Fig. 3) shows that the Berea core sample has an average pore diameter of 1.418 µm, which agrees with previous measurements conducted by Nelson (2009). Silicon dioxide particles used in this experiment have particle sizes ranging from 0.005 to 0.015 μ m, which yields an average jamming ratio (particle size/pore size) of 0.004 to 0.01. According to the jamming ratio classifications by Khilar and Fogler (1998), these particles are classified to exhibit deposition and multi-particle blocking. However, the goal of this experiment is to show that although classified in similar category of plugging mechanism, difference in fines sizes could result in different injectivity alteration. Besides, the selected range of particle sizes would yield more realistic results because the dissolution of sandstone minerals under typical CO₂ storage conditions may yield fines of varying sizes over a wide range depending on the composition of the host rock and reaction conditions.

The effect of introducing a constant 0.3 wt% of varying size of silicon dioxide on the RIC at different brine salinity is shown in Fig. 7. It is observed that salt precipitation does in fact, augment the injectivity alteration induced by fines mobilisation. The results suggests that, while keeping the size and concentration of fine particles constant, increasing brine salinity, which gives a higher amount of salt precipitation will induce higher injectivity impairment. The underlying theory is that salt precipitates on the wall of pores will render the pore sizes smaller. A higher amount of precipitated salts increases the jamming ratio, and more fines will





Fig. 9 Relative injectivity change for the combined effect of salt precipitation and fines migration at varying particle sizes. Injectivity imairment (RIC) increases with increasing particle size with 15 nm nanosilica particles inducing the highest injectivity impairment



be prone to entrapment. The jamming ratio (β) is the ratio of particle size to pore throat diameter (Chang & Civan, 1997; King & Adegbesan, 1997). The highest injectivity impairment occurs at the highest brine salinity, as more precipitated salts reduce the pore spaces, leading to higher jamming ratio. Therefore, more particles will be blocked and plugged at the slimmer pore throats. This is in agreement with results from theoretical modelling presented by Sokama-Neuyam, et al. (2017a, b) in which they attempted to model the combined effect of salt precipitation and fines migration on CO₂ injectivity alteration. However, their work is completely based only on theoretical modelling without any experimental verification.

The results also suggests that the introduction of very small fines particles (0.005 μ m) gives very little effect on the injectivity alteration. This might be so because most of the particles are free to pass through the smallest pore throat. The RIC values show a slight increase of about 2–10% compared to injectivity alteration contributed by salt precipitation at different brine salinities. On the other hand, a significant effect on the RIC was observed when 0.015 μ m fines particles were used in the experiment. The RIC measured after addition of 0.015 μ m particles is three times higher than salt precipitation over the whole brine salinity range investigated. The increase in observed RIC with increasing fines sizes is probably caused by the jamming ratio.

Different size of particles would give different occurrence of pore plugging/piping at the pore throat due to different jamming ratios. Taking a constant brine salinity of 30,000 ppm as an example, the trends for the influence of fines sizes on RIC is shown in Fig. 10. It can be observed that the RIC value increased from 13.6% to a slightly higher value of 14.7% when for colloid dispersion with 0.005 μ m silica particles. The RIC value then jumped to 29% when 0.015 μ m fines particles was injected into the core. According to Khilar and Fogler (1998), the use of 0.005 μ m particle would result in piping as the jamming ratio is less than 0.01, leading to very low injectivity changes. For the 0.015 μ m particles, although the jamming ratio is close to 0.01, larger fines sizes at similar concentrations promote greater surface deposition and multi-particle blocking in the porous media during the scCO₂ injection, that leads to a higher injectivity impairment.

The results obtained is sufficient to establish that fines sizes could contribute significantly to the injectivity alteration by the salt precipitation during CO_2 injection. In addition, it is possible to increase the particle size and concentration to evaluate their limitation to the total CO_2 injectivity changes. However, further work would be needed to assess the variables thoroughly.

Conclusions

This work presents the results of scCO₂ core-flooding experiments conducted to assess the combined effect of salt precipitation and fines migration on CO₂ injectivity changes. It was found that brine salinity had a significant influence on injectivity change of sandstone rock after scCO₂ injection. Injectivity impairment up to about 13.6% could be induced by salt precipitation alone. The injectivity decreased with increase in brine salinity. The precipitated solid salt saturation in the core increases with increasing brine salinity as was supported by images of the injected face of the core samples after CO₂ injection. Moreover, the results showed that fines migration further increased the injectivity impairment as compared to salt precipitation alone as the brine salinity increases. Injectivity impairment up to about 29% was recorded for the combined effect of salt precipitation and fine mobilisation



Fig. 10 Effect of fines sizes on the relative injectivity change of sandstone saturated with 30,000 ppm NaCl. Again, the RIC increases with increasing particle size



induced by as low as about 0.3% wt of particles in the injection fluid. Larger particle sizes can induce up to threefold injectivity alteration, possibly due to particle piping/ plugging at a different jamming ratio. The present work serve as a preliminary evaluation to quantify the effect of fines migration on injectivity change as compared to salt precipitation. Although further work is needed to investigate and establish the effect of various range of injection flow rates, temperature, rock permeability to describe the combined impact of salt precipitation and fines migration on CO₂ injectivity changes, the present work gives adequate insight and confirms the reported findings in the literature from analytical studies. This validatory studies are important for improving large-scale modelling and properly planning CO₂ injection operations to incorporate the effect of both salt precipitation and fines mobilisation.

Acknowledgements The authors acknowledge the following partners for their contributions: PETRONAS Carigali Sdn. Bhd. and Shahrul Rizzal Md Yusof and Iswadi Radzali from Core Analysis Lab, UTP.

Funding This work is funded by the Yayasan Universiti Teknologi PETRONAS (YUTP) (0153AA-E78) project "Mechanistic study of pore-scale particle invasion, formation abatement and clean-up efficiency during well completion" and Yayasan Universiti Teknologi PETRONAS (YUTP) (0153LC0-009) project "Mechanistic study of CO_2 injectivity impairment in saline reservoir by fines mobilisation", which is supported by Institute of Hydrocarbon Recovery, UTP.

Declarations

Conflicts of interest On behalf of all the co-authors, the corresponding author states that there is no conflict of interest. The funders had no role in the design of the study; in the collection, analyses or interpre-



tation of data; in the writing of the manuscript, or in the decision to publish the results.

Open Access This article is licensed under a Creative Commons Attribution 4.0 International License, which permits use, sharing, adaptation, distribution and reproduction in any medium or format, as long as you give appropriate credit to the original author(s) and the source, provide a link to the Creative Commons licence, and indicate if changes were made. The images or other third party material in this article are included in the article's Creative Commons licence, unless indicated otherwise in a credit line to the material. If material is not included in the article's Creative Commons licence and your intended use is not permitted by statutory regulation or exceeds the permitted use, you will need to obtain permission directly from the copyright holder. To view a copy of this licence, visit http://creativecommons.org/licenses/by/4.0/.

References

- Andre L, Audigane P, Azaroual M, Menjoz A (2007) Numerical modeling of fluid–rock chemical interactions at the supercritical CO₂–liquid interface during CO₂ injection into a carbonate reservoir. Energy Convers Manage 48:1782–1797
- Bacci G, Korre A, Durucan S (2011) Experimental investigation into salt precipitation during CO₂ injection in saline aquifers. Energy Procedia 4:4450–4456. https://doi.org/10.1016/j.egypro. 2011.02.399
- Baumann G, Henninges J, De Lucia M (2014) Monitoring of saturation changes and salt precipitation during CO₂ injection using pulsed neutron-gamma logging at the Ketzin pilot site. Int J Greenhouse Gas Control 28:134–146
- Bensinger J, Beckingham LE (2020) CO₂ storage in the Paluxy formation at the Kemper County CO2 storage complex: Pore network properties and simulated reactive permeability evolution. Int J Greenhouse Gas Control 93:102887
- Benson SM, Hingerl F, Li B, Pini R, Tchelepi H, & Zuo L (2011) Investigations in Geologic Carbon Sequestration : Multiphase Flow of CO₂ and Water in Reservoir Rocks. *Annual report 2011.*

- Burton M, Kumar N, Bryant SL (2009) CO_2 injectivity into brine aquifers: why relative permeability matters as much as absolute permeability. Energy Procedia 1(1):3091–3098
- Chang F, Civan F (1997) Practical model for chemically induced formation damage. J Petrol Sci Eng 17(1–2):123–137
- Cinar Y, Riaz A (2014) Carbon dioxide sequestration in saline formations: Part 2—Review of multiphase flow modeling. J Petrol Sci Eng 124:381–398
- Civan F.(2015) *Reservoir formation damage*: Gulf Professional Publishing.
- De Silva G, Ranjith PG, Perera M (2015) Geochemical aspects of CO₂ sequestration in deep saline aquifers: a review. Fuel 155:128–143
- Gaus I (2010) Role and impact of CO2–rock interactions during CO₂ storage in sedimentary rocks. Int J Greenhouse Gas Control 4(1):73–89
- Giorgis T, Carpita M, Battistelli A (2007) 2D modeling of salt precipitation during the injection of dry CO2 in a depleted gas reservoir. Energy Convers Manage 48:1816–1826. https://doi.org/10.1016/j. enconman.2007.01.012
- Grude S, Landrø M, Dvorkin J (2014) Pressure effects caused by CO₂ injection in the Tubåen Fm., the Snøhvit field. Int J Greenhouse Gas Control 27:178–187
- Guyant E, Han WS, Kim KY, Park MH, Kim BY (2015) Salt precipitation and CO₂/brine flow distribution under different injection well completions. Int J Greenhouse Gas Control 37:299–310. https:// doi.org/10.1016/j.ijggc.2015.03.020
- Hassan H, Omar NFN, Jalil A A-MM, Salihuddin RS, & Shah SSM (2018) Gearing Toward CCUS for CO₂ Reduction in Malaysia.
 Paper presented at the Offshore Technology Conference Asia
- Hommel J, Coltman E, Class H (2018) Porosity–permeability relations for evolving pore space: a review with a focus on (bio-) geochemically altered porous media. Transp Porous Media 124(2):589–629
- Hurter S, Labregere D, Berge J (2007) Simulations for CO₂ injection projects with compositional simulator. Offshore Eur Con - Proceed. https://doi.org/10.2523/108540-ms
- Ibrahim MA, Saleh TA (2020) Partially aminated acrylic acid grafted activated carbon as inexpensive shale hydration inhibitor. Carbohy Res 491:107960
- Jalil M, Masoudi R, Darman NB, Othman M (2012) Study of the CO₂ injection, storage, and sequestration in depleted M4 carbonate gas condensate reservoir. Paper presented at the Carbon management technology conference, Malaysia
- Jeddizahed J, Rostami B (2016) Experimental investigation of injectivity alteration due to salt precipitation during CO₂ sequestration in saline aquifers. Adv Water Resour 96:23–33. https://doi.org/10. 1016/j.advwatres.2016.06.014
- Kartikasurja DO, Viratno B, & Sukahar M (2008) Produced CO₂ Storage into Aquifer in an Offshore Field, Malaysia. Paper presented at the SPE Asia Pacific Oil and Gas Conference and Exhibition
- Kaszuba J, Yardley B, Andreani M (2013) Experimental perspectives of mineral dissolution and precipitation due to carbon dioxidewater-rock interactions. Rev Mineral Geochem 77(1):153–188
- Khilar KC, & Fogler HS (1998) *Migrations of fines in porous media* (Vol. 12): Springer Science & Business Media
- King R & Adegbesan K (1997) Resolution of the principal formation damage mechanisms causing injectivity and productivity impairment in the Pembina Cardium reservoir. Paper presented at the SPE Annual Technical Conference and Exhibition
- Krevor SC, Pini R, Zuo L, Benson SM (2012) Relative permeability and trapping of CO₂ and water in sandstone rocks at reservoir conditions. Water Res Res. https://doi.org/10.1029/2011WR010859
- Liu H-H, Zhang G, Yi Z, Wang Y (2013) A permeability-change relationship in the dryout zone for CO2 injection into saline aquifers. Int J Greenhouse Gas Control 15:42–47. https://doi.org/10.1016/j. ijggc.2013.01.034

- Mathias SA, Gluyas JG, de Miguel GJGM, Bryant SL, Wilson D (2013) On relative permeability data uncertainty and CO₂ injectivity estimation for brine aquifers. Int J Greenhouse Gas Control 12:200–212
- MdYusof MA, Arif Ibrahim M, Idress M, Idris AK, Saaid IM, Rosdi NM, MataliA. A. AA (2020a) Effects of CO2/Rock/Formation Brine Parameters on CO₂ Injectivity for Sequestration. SPE J. https://doi.org/10.2118/203843-PA
- MdYusof MA, Zainal MZ, Idris AK, Ibrahim MA, Yusof SRM, Ismail SN, Mohshim DF (2020b) Petrophysical Changes of Sandstone Due to Salt Precipitation and Fines Migration During Carbon Dioxide Injection. J Comput Theor Nanosci 17(2–3):1207–1213. https://doi.org/10.1166/jctn.2020.8791
- Md Yusof MA, Mohamed MA, Md Akhir NA, Ibrahim MA, Saaid IM, Idris AK, . . . Awangku Matali AAA (2021) Influence of Brine–Rock Parameters on Rock Physical Changes During CO₂ Sequestration in Saline Aquifer. *Arabian Journal for Science and Engineering*, 1–15
- Miri R, Hellevang H (2016) Salt precipitation during CO₂ storage—A review. Int J Greenhouse Gas Control 51:136–147
- Miri R, van Noort R, Aagaard P, Hellevang H (2015) New insights on the physics of salt precipitation during injection of CO₂ into saline aquifers. Int J Greenhouse Gas Control 43:10–21. https://doi.org/ 10.1016/j.ijggc.2015.10.004
- Mohamed I, He J & Nasr-El-Din HA (2012) Carbon Dioxide Sequestration in Sandstone Aquifers: How Does It Affect the Permeability? Paper presented at the Carbon Management Technology Conference
- Muhammad A, MY, Ibrahim MA, Mohamed MA, Md Akhir NA, M Saaid I, Ziaudin Ahamed MN, Awangku Matali AA (2021) Predictive Modelling of CO₂ Injectivity Impairment due to Salt Precipitation and Fines Migration During Sequestration. Paper presented at the International Petroleum Technology Conference
- Muller N, Qi R, Mackie E, Pruess K, Blunt MJ (2009) CO2 injection impairment due to halite precipitation. Energy Procedia 1(1):3507–3514
- Nelson PH (2009) Pore-throat sizes in sandstones, tight sandstones, and shales. AAPG Bull 93(3):329–340
- Othman F, Yu M, Kamali F, Hussain F (2018b) Fines migration during supercritical CO₂ injection in sandstone. J Nat Gas Sci Eng 56:344–357
- Othman F, Naufaliansyah MA, Hussain F (2019) Effect of water salinity on permeability alteration during CO2 sequestration. Adv Water Resour 127:237–251
- Othman F, Wang Y, Hussain F (2018) The Effect of Fines Migration During CO_2 Injection Using Pore Scale Characterization. Paper presented at the SPE Asia Pacific Oil and Gas Conference and Exhibition
- Ott H, Andrew M, Snippe J, Blunt MJ (2014) Microscale solute transport and precipitation in complex rock during drying. Geophys Res Lett 41:8369–8376. https://doi.org/10.1002/2014GL062266
- Ott H, Roels SM, de Kloe K (2015) Salt precipitation due to supercritical gas injection: I. Capillary-driven flow in unimodal sandstone. Int J Greenhouse Gas Control 43:247–255. https://doi.org/10. 1016/j.ijggc.2015.01.005
- Peysson Y, André L, Azaroual M (2014) Well injectivity during CO₂ storage operations in deep saline aquifers—Part 1: Experimental investigation of drying effects, salt precipitation and capillary forces. Int J Greenhouse Gas Control 22:291–300
- Pruess K, Müller N (2009) Formation dry-out from CO_2 injection into saline aquifers: 1. Effects of solids precipitation and their mitigation. *Water Resources Research* 45(3)
- Pruess K, García J (2002) Multiphase flow dynamics during CO2 disposal into saline aquifers. Environ Geol 42:282–295. https://doi. org/10.1007/s00254-001-0498-3



- Schembre-McCabe JM, Kamath J, Gurton RM (2007) *Mechanistic* studies of CO₂ sequestration. Paper presented at the International Petroleum Technology Conference
- Sokama-Neuyam YA, Ursin JR (2018a) The coupled effect of salt precipitation and fines mobilization on CO₂ injectivity in sandstone. Greenhouse Gases: Sci Technol 8:1066–1078. https://doi.org/10. 1002/ghg.1817
- Sokama-Neuyam YA, Ursin JR (2018b) The coupled effect of salt precipitation and fines mobilization on CO2 injectivity in sandstone. Greenhouse Gases: Sci Technol 8(6):1066–1078
- Sokama-Neuyam YA, Forsetløkken SL, Lien J-E, Ursin JR (2017a) The Coupled Effect of Fines Mobilization and Salt Precipitation on CO2 Injectivity. Energies 10(8):1125
- Sokama-Neuyam YA, Ginting PUR, Timilsina B, Ursin JR (2017b) The impact of fines mobilization on CO₂ injectivity: An experimental study. Int J Greenhouse Gas Control 65:195–202. https://doi.org/ 10.1016/j.ijggc.2017.08.019
- Sukor NR, Shamsuddin AH, Mahlia TMI, Mat Isa MF (2020) Techno-Economic Analysis of CO₂ Capture Technologies in Offshore Natural Gas Field: Implications to Carbon Capture and Storage in Malaysia. Processes 8(3):350
- Tewari RD, Sedaralit MF (2021) Making Things Right in Development and Management of Highly Contaminated Giant Carbonate Gas Field and Returning the CO₂ to Subsurface Sequestration. Paper presented at the International Petroleum Technology Conference
- Torsæter M, Cerasi P (2018) Geological and geomechanical factors impacting loss of near-well permeability during CO₂ injection. Int J Greenhouse Gas Control 76:193–199. https://doi.org/10.1016/j. ijggc.2018.07.006
- Verma A, Pruess K (1988) Thermohydrological conditions and silica redistribution near high-level nuclear wastes emplaced in saturated geological formations. J Geophy Res: Solid Earth 93(B2):1159– 1173. https://doi.org/10.1029/JB093iB02p01159

- Wang Z, Zhou Q, Guo H, Yang P, Lu W (2018) Determination of water solubility in supercritical CO2 from 313.15 to 473.15 K and from 10 to 50 MPa by in-situ quantitative Raman spectroscopy. Fluid Phase Equilib 476:170–178. https://doi.org/10.1016/j.fluid.2018. 08.006
- Yusof MAM, Mohamed MA, Akhir NAM, Ibrahim MA, Mardhatillah MK (2021) Combined Impact of Salt Precipitation and Fines Migration on CO₂ Injectivity Impairment. Int J Greenhouse Gas Control 110:103422
- Zaidin MF, Chapoy A, Coquelet C, Raub A, Razib M, Salihuddin RS, ... Kashim MZ (2018) Impact of H2s in Predicting the Storage Efficiency of CO₂ Injection in a High Pressure High Temperature (Hpht) Carbonate Aquifer-a Case Study in a Sarawak Offshore High CO₂ Gas Field, Malaysia. Paper presented at the 14th Greenhouse Gas Control Technologies Conference Melbourne
- Zeidouni M, Pooladi-Darvish M, & Keith D (2009) Sensitivity analysis of salt precipitation and CO₂-brine displacement in saline aquifers. SPE International Conference on CO₂ Capture Storage and Utilization. https://doi.org/10.2118/126690-ms
- Zemke K, Liebscher A, Wandrey M (2010) Petrophysical analysis to investigate the effects of carbon dioxide storage in a subsurface saline aquifer at Ketzin, Germany (CO₂SINK). Int J Greenhouse Gas Control 4(6):990–999
- Zettlitzer M, Moeller F, Morozova D, Lokay P (2010) Re-establishment of the proper injectivity of the CO₂ -injection well Ktzi 201 in Ketzin. Germany 4:952–959. https://doi.org/10.1016/j.ijggc.2010. 05.006

Publisher's Note Springer Nature remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.

