

## Chapter

# CO<sub>2</sub> Injectivity in Deep Saline Formations: The Impact of Salt Precipitation and Fines Mobilization

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## Abstract

Climate change is now considered the greatest threat to global health and security. Greenhouse effect, which results in global warming, is considered the main driver of climate change. Carbon dioxide (CO<sub>2</sub>) emission has been identified as the largest contributor to global warming. The Paris Agreement, which is the biggest international treaty on Climate Change, has an ambitious goal to reach Net Zero CO<sub>2</sub> emission by 2050. Carbon Capture, Utilization and Storage (CCUS) is the most promising approach in the portfolio of options to reduce CO<sub>2</sub> emission. A good geological CCUS facility must have a high storage potential and robust containment efficiency. Storage potential depends on the storage capacity and well injectivity. The major target geological facilities for CO<sub>2</sub> storage include deep saline reservoirs, depleted oil and gas reservoirs, Enhanced Oil Recovery (EOR) wells, and unmineable coal seams. Deep saline formations have the highest storage potential but challenging well injectivity. Mineral dissolution, salt precipitation, and fines mobilization are the main mechanisms responsible for CO<sub>2</sub> injectivity impairment in saline reservoirs. This chapter reviews literature spanning several decades of work on CO<sub>2</sub> injectivity impairment mechanisms especially in deep saline formations and their technical and economic impact on CCUS projects.

**Keywords:** CO<sub>2</sub> injectivity, mineral dissolution, salt precipitation, fines mobilization

## 1. Introduction

Since last decade, there has been a growing concern of the negative impacts of global climate change. In the last century, scientists believe that carbon dioxide (CO<sub>2</sub>) emission is has been the main component responsible for approximately three-quarters of global greenhouse gas emission. A roadmap developed to combat

climate change has outlined 10 scalable solutions clustered into categories of social transformative, governance improvement, market and regulation-based solutions, technological innovation and transformation, and lastly natural and ecosystem management. While some proposed mitigation techniques focus on reduction of CO<sub>2</sub> emission, Carbon Capture, Utilization and Storage (CCUS) technology can aim at achieving lower CO<sub>2</sub> amount in the atmosphere by capturing and storing the anthropogenic gas in a geological storage. Geological storage of CO<sub>2</sub> in depleted oil and gas reservoirs, deep saline reservoirs, unmineable coal seams or injected into active oil and gas reservoirs for Enhanced Oil Recovery (EOR) are currently a well-accepted method of storing CO<sub>2</sub>. For economic reasons, CO<sub>2</sub> is being injected at the highest possible rates through limited number of wells. This could trigger injectivity-related issues due to complex interactions between CO<sub>2</sub>, brine and rock initiated in the aquifer. This makes CO<sub>2</sub> injectivity not only a technical challenge but also an economic consideration.

This chapter presents a comprehensive discussion on how the various mechanisms contributed by the fluid-rock interactions during CO<sub>2</sub> sequestration affect CO<sub>2</sub> injectivity. The chapter begins by laying out the theoretical dimensions of CO<sub>2</sub> sequestration. This is followed by a brief overview of different CO<sub>2</sub> injectivity impairment mechanisms, focusing on the two main themes: salt precipitation and fines migration. The experimental findings from previous researchers have also been discussed and some findings remarks made. The insights gained from this study may be valuable to the rapidly expanding field of carbon sequestration.

## **2. Climate change, a global challenge**

Climate Change is now considered the greatest threat to global health and security. Greenhouse effect, which propels global warming, has been identified as the main driver of Climate Change. The rising of global temperature is intricately linked with many other environmental concerns such as fragile ecosystem, melting glaciers, increasing sea level, acidification of sea water and increased flooding and droughts [1]. This climate challenge is also affecting the social community which can lead to immigration and conflicts over borders and natural resources such as water. More importantly, it could severely threaten food security that may affect about 3 billion of poor people in terms of access to food supply. These series of concerns are recognized as climate change and it is well accepted that to prevent its occurrence, greenhouse gas emission has to be reduced significantly over the twenty-first century [2]. The gases which are mainly responsible for the greenhouse effect include methane, carbon dioxide (CO<sub>2</sub>), nitrous oxide, water vapor, and fluorinated gases.

CO<sub>2</sub> generated mainly from anthropogenic activities is the largest contributor to global warming. By 2020, the concentration of CO<sub>2</sub> in the atmosphere had risen to 48% above its pre-industrial level [3, 4]. An increase of 2°C above pre-industrial average temperature could induce serious negative impacts on the natural environment and human health. The Paris Agreement, which is the biggest international treaty on Climate Change is determined to limit global warming to about 1.5°C, compared to pre-industrial levels, with an ambitious goal to reach NetZero CO<sub>2</sub> emission by 2050 [5-7].

The anthropogenic activities with highest carbon footprint include the burning of fossil fuel for power generation and the production of materials. Power generation from fossil fuels is responsible for over 70% of the global CO<sub>2</sub> emissions [1]. Cumulatively, fossil fuel contributed about 84% of the World's primary energy consumption by 2019 and the world is expected to rely heavily on fossil fuels for its energy needs, at least within the short to medium term. Another major source of CO<sub>2</sub> emission is the production of materials with high carbon and energy footprint. The notable of such materials is Portland cement which is the main building material used in most countries in the world. Portland cement production is responsible for about 5% of global CO<sub>2</sub> emission [8]. About 2% of the total global energy consumption is used to produce Portland cement. Widespread use of energy efficient power generators, investing in renewable energy, Carbon Capture, Utilization and Storage (CCUS) remains the best options for reducing CO<sub>2</sub> emission from burning of fossil fuels and achieving a faster transition to green energy.

### 3. Geological storage of CO<sub>2</sub>

CCUS is considered a viable option to reduce CO<sub>2</sub> emission, sustain exploration and production of fossil fuel for the short to medium term and eventually transition to a full green energy in the long term [9, 10]. Among the proposed CO<sub>2</sub> emission reduction strategies, CCUS provides the highest emission reduction potential [11]. Generally, CCUS involves the (1) capture of CO<sub>2</sub> from large industrial emission sources and direct air capture points (2) the transportation of the captured gas to utilization, conversion, or storage facilities and (3) the utilization of the gas as feed-stock in industrial processes, conversion to other products or the injection of the gas into geological storage facilities. In terms of geological storage, the injected CO<sub>2</sub> may be stored in depleted oil and gas reservoirs, deep saline reservoirs, unmineable coal seams or injected into active oil and gas reservoirs for Enhanced Oil Recovery (EOR) [12, 13].

CCUS system, although simple in concept, would require significant investment of capital, new technology and time [14]. Besides, many current policies also need to be revised and new legal and regulations framework has to be introduced that require support from local authorities, governments and international bodies [15]. Investigation by the International Energy Agency (IEA) have shown that CCUS can contribute up to about 14% reduction in global greenhouse gas emissions required to limit global warming to 2°C by 2050 [16].

#### 3.1 Storage in saline aquifers

Saline aquifer refers to a deep, large geological formation consisting permeable sedimentary or carbonate rock types that are saturated with formation water or brines, non-potable water, containing high concentration of dissolved salts [17]. It is buried under a layer of non- or low-permeability rocks that serve as a cap rock to prohibit the fluid flowing upwards to the surface. The saline aquifer can be located both onshore and offshore and normally found at depth greater (more than 800 metres) than aquifers that contain potable water [18]. Deep saline formations have enormous potential for CO<sub>2</sub> storage in terms of volumetric storage capacity [19, 20].

On a global scale, deep saline reservoirs have the capacity to hold between 20 and 500% of the projected CO<sub>2</sub> emissions by 2050 [9, 21, 22]. Thus, worldwide CO<sub>2</sub> storage potential of deep saline reservoirs ranges from 400 to 10,000 Gt CO<sub>2</sub>. Deep saline aquifers, usually at depths between 700 and 1000 m, hold large quantities of high salinity formation brines [23].

Although the natural content of these reservoirs has no direct commercial value, the chemical composition of the formation brine makes them suitable for CO<sub>2</sub> mineralization. In deep saline aquifers, the injected CO<sub>2</sub> could be sequestered through hydrodynamic trapping where the gas is trapped beneath a caprock, residual trapping where the rock contains residual saturation of CO<sub>2</sub>, solubility trapping where the gas dissolves in the formation brine and mineral trapping where CO<sub>2</sub> reacts with Ca, Fe or Mg to form stable carbonate precipitates [24, 25]. Lack of additional economic benefits except carbon tax incentives in some countries, makes CO<sub>2</sub> storage in saline aquifers less attractive to the oil and gas industry.

### **3.2 CO<sub>2</sub> enhanced oil recovery**

CO<sub>2</sub>-EOR is a tertiary oil recovery technique where CO<sub>2</sub> and usually other fluids such as water or brine is injected into the reservoir to achieve miscibility with the oil and recover residual oil. In addition to extraction of residual oil, the injected gas provides pressure support and could remain stored permanently after the recovery process. Under subsurface conditions, CO<sub>2</sub> mixes with oil above a certain minimum miscibility pressure (MMP), reducing the capillary effect that retain the oil in place [26–28]. There are four main underlying mechanisms of CO<sub>2</sub>-EOR as outlined by Rojas and Ali [29] and Tunio et al. [30] which include (1) oil swelling; (2) reduction of oil viscosity; (3) reduction of oil and water density; and (4) extraction of oil components.

Alternative forms of CO<sub>2</sub>-EOR have been developed over the past years, including continuous CO<sub>2</sub> injection, continuous CO<sub>2</sub> injection followed by water, water-alternating gas (WAG) and WAG followed by gas or water [31–33]. To improve sweep efficiency, carbonated water injection has also been used as a viable alternative [34, 35]. Other emerging injection schemes include CO<sub>2</sub> low salinity water alternating gas (CO<sub>2</sub>-LSWAG) injection under miscible CO<sub>2</sub> displacement conditions [36–38]. Depleted oil and gas reservoirs are also attractive candidates for CO<sub>2</sub> storage due to the potential to reuse some of the production equipment and geological data collected over the producing life of the reservoirs to lower exploration cost and reduce the risk associated with CO<sub>2</sub> storage. It has been reported that depleted oil and gas reservoirs could hold about 45% of the projected CO<sub>2</sub> emissions by 2050 [9].

### **3.3 CO<sub>2</sub> injection into unconventional reservoirs**

The two main unconventional formations where CO<sub>2</sub> injection is promising are coal seams and shale gas reservoirs. CO<sub>2</sub> enhanced coalbed methane recovery (CO<sub>2</sub>-ECBM) has the potential to store large volumes of CO<sub>2</sub> in deep unmineable coal seams while improving the efficiency of coal bed methane recovery [39, 40]. The injected CO<sub>2</sub> displaces methane and remain sequestered in the coal seams as CO<sub>2</sub> is preferentially adsorbed onto coal seams, thus releasing the coal bed methane which can then be produced as free gas [41]. Based on the simple assumption that, for

every CH<sub>4</sub> molecule, two molecules of CO<sub>2</sub> can be stored, IEA-GHG [42] estimated that about 220 GT of CO<sub>2</sub> could be stored in deep unmineable coal formations worldwide.

The potential to store CO<sub>2</sub> in organic-rich gas shales is attracting increasing interest, especially in countries that have extensive shale deposits [43–45]. Although still in early-stage research, CO<sub>2</sub> injection into organic-rich gas shales could provide dual benefits: an economic benefit from the incremental recovery of adsorbed methane, and an environmental benefit of secure CO<sub>2</sub> storage.

## 4. The prerequisites of CCUS

A viable candidate for CCUS must meet a threshold well injectivity required to inject large volumes of CO<sub>2</sub> at high injection rates through a minimum number of wells, adequate storage capacity to hold large volumes of CO<sub>2</sub> and robust containment to permanently isolate the sequestered gas from the environment [16]. Storage capacity and well injectivity defines the storage potential of a geological storage facility [11, 46, 47].

### 4.1 Storage capacity

Implementation of CCUS technology require accurate estimation of the pore space available in the reservoir rock to hold the injected CO<sub>2</sub> [48–51]. The storage capacity estimated can be of different levels of certainty and cost depending on the scale and resolution. The various CO<sub>2</sub> trapping mechanisms in deep saline aquifers, namely structural and stratigraphic trapping, residual gas trapping, solubility trapping, mineral trapping and hydrodynamic trapping, which occur at different times during the storage, must be considered in the estimation to obtain a representative estimate [51]. Other parameters that affect the storage capacity include in situ pressure, injectivity, temperature, permeability, and rock compressibility.

The volume of CO<sub>2</sub> that can be commercially sequestered in a reservoir within a specific period, using available technology, under current economic conditions, operating methods and governmental regulations has been termed the CO<sub>2</sub> storage reserve [11, 48]. The USDOE [52] has developed a simplified model to quantify the storage capacity of deep saline formations which is given by:

$$M_{CO_2} = V_A \phi_T \rho_{CO_2} E_s \quad (1)$$

In Eq. (1),  $M_{CO_2}$  is the mass of CO<sub>2</sub> that can be stored,  $V_A$  is the bulk volume of the aquifer,  $\phi_T$  is the effective porosity of the aquifer,  $\rho_{CO_2}$  is the density of CO<sub>2</sub> at reservoir conditions and  $E_s$  is the storage efficiency. The storage efficiency expresses the degree of filling the reservoir [11], also defined as the ratio of the volume occupied by CO<sub>2</sub> to the total accessible pore volume of the reservoir [53]:

$$E_s = \frac{V_{CO_2}}{V_{pore}} \quad (2)$$

In Eq. (2),  $V_{CO_2}$  is the volume of injected CO<sub>2</sub> and  $V_{pore}$  is the accessible reservoir pore volume available for CO<sub>2</sub> storage. Eq. (1) and (2) can be coupled to estimate the volumetric CO<sub>2</sub> storage capacity of a given deep saline reservoir. CO<sub>2</sub> storage



efficiency in deep saline formations depends on the reservoir rock properties (porosity, permeability, net to gross, thickness and area), the efficiency of water displacement by injected CO<sub>2</sub> and the degree of conformance of the aquifer [11].

Bachu et al. [54] have also proposed a model to estimate the theoretical CO<sub>2</sub> storage capacity of depleted oil and gas reservoirs, based on the assumption that the entire pore space originally occupied by hydrocarbons can be filled by CO<sub>2</sub> and that CO<sub>2</sub> can be injected until the reservoir pressure reaches the original pressure of the virgin reservoir. These assumptions can be valid if the reservoir is not in contact with an aquifer or already flooded during secondary and tertiary recovery. For practical purposes, an effective storage capacity could be defined to incorporate other important parameters such as displacement efficiency, gravity effects, residual oil and water saturation, reservoir heterogeneity, rock-fluid interactions, and formation damage.

## 4.2 Well injectivity

The injectivity of a reservoir measures the amount of CO<sub>2</sub> an injection well can receive without fracturing the formation [11]. Well injectivity can be expressed with an injectivity index,  $I$ , often defined as the ratio of volumetric injection flow rate to the pressure drop [55, 56]. For a homogeneous and isotropic reservoir, the steady-state CO<sub>2</sub> well injectivity index can be expressed as:

$$I = \frac{q}{\Delta p} = \frac{\rho_{CO_2, res}}{\rho_{CO_2, sc}} \frac{2\pi kh}{\left[ \ln \left( \frac{r_e}{r_w} \right) + s \right] \mu_{CO_2}} \quad (3)$$

In Eq. (3),  $q$  is the volumetric injection flow rate,  $\Delta p$  is the pressure drop,  $\rho_{CO_2, res}$  is the density of CO<sub>2</sub> under reservoir conditions,  $\rho_{CO_2, sc}$  is the density of CO<sub>2</sub> under standard conditions,  $kh$  is the permeability-thickness product,  $r_e$  is the radius of the reservoir boundary,  $r_w$  is the well radius,  $s$  is the skin factor and  $\mu_{CO_2}$  is the viscosity of CO<sub>2</sub> under reservoir conditions. Well injectivity determines the number of wells required to inject a specific quantity of CO<sub>2</sub> into the reservoir. This makes injectivity an important factor for both technical and economic evaluation of CO<sub>2</sub> storage projects [56, 57].

## 4.3 Containment efficiency

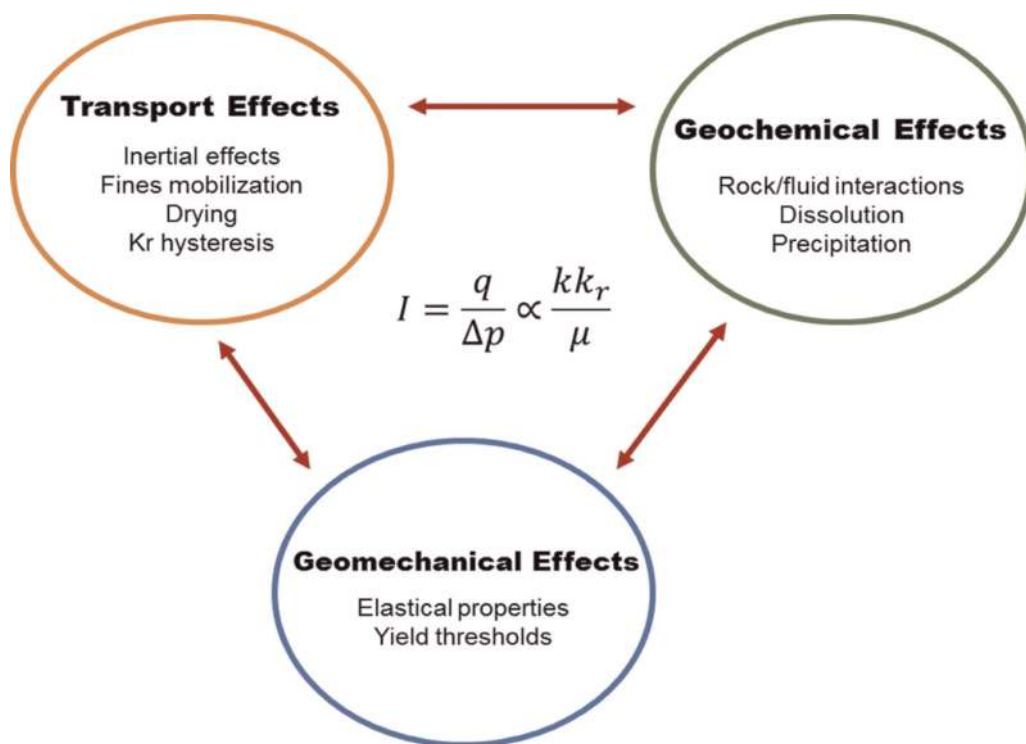
Containment efficiency characterizes the assurance of containment of the injected CO<sub>2</sub>. The ultimate objective of a CCUS project is to permanently isolate the sequestered CO<sub>2</sub> from the environment. Since formation water is denser than supercritical CO<sub>2</sub>, the CO<sub>2</sub> plume tends to rise to the top of the reservoir, where it accumulates beneath the caprock. The containment efficiency of a geological trap is therefore strongly dependent on the seal potential or the ability of the caprock to confine the injected gas and prevent leakage into overlying formations and eventually back into the atmosphere [58]. The caprock must have the lateral extent and geomechanical strength to retain the full CO<sub>2</sub> column height.

The integrity of the caprock could be compromised by mechanical deformation induced by pressure from CO<sub>2</sub> injection or through geochemical CO<sub>2</sub>-rock-brine interactions which may dissolve or precipitate minerals to increase the permeability of

the caprock [59]. Wells have also been identified as probable leakage pathways. Therefore, robust wellbore integrity is important to prevent leakage through wells.

## 5. CO<sub>2</sub> well injectivity impairment mechanisms

CO<sub>2</sub> injectivity impairment is a technical and economic constraint on geological storage of CO<sub>2</sub> [12, 60, 61]. Under typical storage conditions, several factors could influence CO<sub>2</sub> injectivity because of the complex interplay of chemical and physical phenomena in the reservoir, especially in the injection area of the wellbore [46, 62, 63]. The injection area of the wellbore has the highest fluctuations of temperature, pressure and flux, making it one of the most important sections of the formation for well injectivity impairment analysis. Lombard et al. [64] identified three main mechanisms responsible for CO<sub>2</sub> injectivity impairment: Geochemical, geomechanical and transport phenomena (**Figure 1**). Later Torsaeter et al. [65] did an extensive review on other CO<sub>2</sub> injectivity impairment mechanisms that they recommend should be given close attention in addition to salt precipitation effects. The geochemical mechanisms involve CO<sub>2</sub>-brine-rock interactions, mineral dissolution, and precipitation. The transport effects include drying of the reservoir rock and fines mobilization. The geomechanical effects, which include borehole deformation has not been given enough research attention compared to the geochemical and transport mechanisms. These phenomena depend on the physical and chemical properties of injected CO<sub>2</sub> which are in turn driven by reservoir conditions and rock properties.

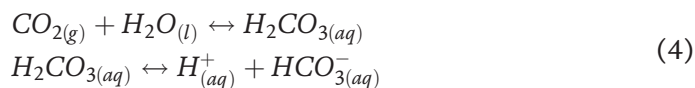


**Figure 1.**  
CO<sub>2</sub> injectivity impairment mechanisms (after Lombard et al., [64]).

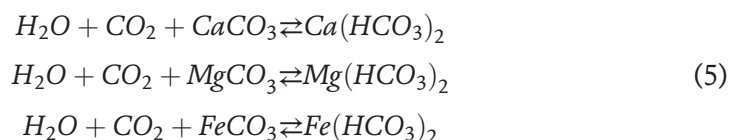
## 5.1 Geochemical effects

### 5.1.1 Effect of mineral dissolution

Mineral dissolution and salt precipitation are the two main geochemical CO<sub>2</sub> injectivity impairment mechanisms [64]. The injected CO<sub>2</sub> could dissolve in formation brine at the CO<sub>2</sub>-brine interface, altering the concentration of aquifer fluid, thus leading to precipitation. Regardless of rock composition, the progressive dissolution of CO<sub>2</sub> in the brine (formation water) forms carbonic acid that leads to a reduction in pH to about 3–5 [66, 67]. The following reactions occur at the interface between both media



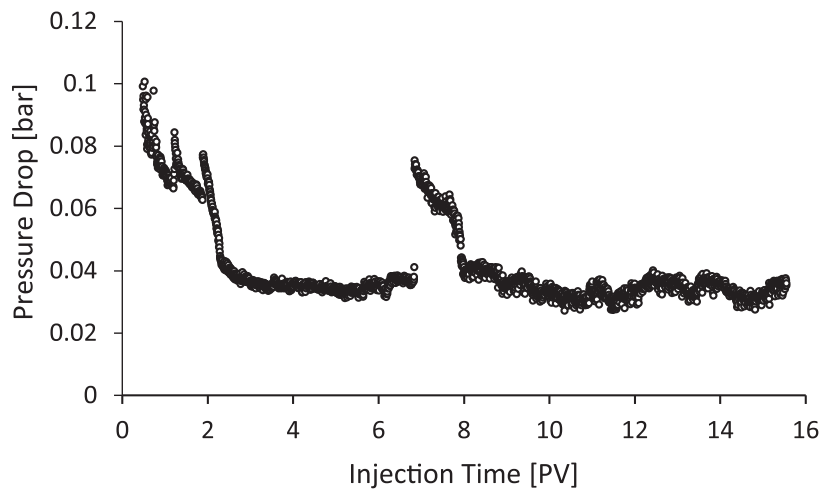
Interactions between CO<sub>2</sub> and brine forms carbonic acid and then bicarbonates. In the presence of CaCO<sub>3</sub>, MgCO<sub>3</sub> and FeCO<sub>3</sub>, the following reactions would lead to formation of water-soluble bicarbonates.



Moreover, the bicarbonates could react with cations in the rock and formation water to form stable carbonates which later could aggregate into small particles or form a scale on the pore walls [68–70]. CO<sub>2</sub>-brine-rock batch reaction under typical storage conditions have shown various amounts of dissolved minerals in solution [70–73].

Wang et al. [74] investigated mineral dissolution and precipitation in dolomite samples saturated with carbonated water at 93°C and 34.5 Mpa under static conditions. They found that mineral dissolution is predominant in highly permeable pathways in the reservoir rock. Zou et al. [75] conducted CO<sub>2</sub>-brine-rock reactions under different conditions to investigate the changes of porosity and permeability during sCO<sub>2</sub> – fracturing in shale reservoir. They reported that CO<sub>2</sub>-brine-rock reaction could occur rapidly (in less than 0.5 h), precipitating minerals such as calcite, dolomite, K-feldspar, and albite into solution. Zhang et al. [76] investigated CO<sub>2</sub>-brine-rock reaction in a tight sandstone reservoir under conditions of 75°C and 32MPa. They found that the dissolution of supercritical CO<sub>2</sub> in brine created an acidic environment which induced the dissolution of minerals into the brine, leading to precipitation of iron minerals and kaolinite. Tang et al. [77] studied the impact of CO<sub>2</sub>-brine-rock interaction on formation properties in gas zone and water zones of a reservoir under static and dynamic conditions. They found that CO<sub>2</sub>-brine-rock interaction occurs in both gas zones and water zones. Aminu et al. [78] investigated the effect of CO<sub>2</sub> and the presence of impurities (NO<sub>2</sub>, SO<sub>2</sub>, H<sub>2</sub>S) on reservoir permeability in static CO<sub>2</sub>-brine-rock reactions. They found that the presence of impurities affects the impact of mineral dissolution on rock permeability. Okhovat et al. [79] investigated the effect of CO<sub>2</sub>-water-rock interaction on rock permeability and oil recovery during CO<sub>2</sub> injection into a carbonate rock. They found that the extent of damage induced was a function of the injection rate.





**Figure 2.** Pressure drop profile recorded during injection of carbonated water into a Bentheimer core at flow rate of 0.25 mL/min at 80 bar and 60°C [80].

Under static conditions, mineral dissolution could increase rock permeability temporarily as new pore spaces are etched and old pore channels could be widened [64]. However, mineral precipitates could aggregate into fine particles in the pore fluid which could form a scale on the pore walls and reduce the flow area.

Sokama-Neuyam et al. [80] conducted experiments to investigate the effect of mineral dissolution on CO<sub>2</sub> injectivity using clay-rich Bentheimer Sandstone cores. The cores were flooded with carbonated water at 80 bar and 60°C with about 25 pore volumes (PV) of carbonated water at 0.25 mL/min. Pressure drop profiles recorded during the flooding is shown in **Figure 2** and SEM-EDS analysis of effluent samples collected are shown in **Table 1**.

**Figure 2** shows a period of immiscible displacement, where pressure drop decreased sharply from about 0.1 bar to about 0.04 bar after the core was flooded with

Element	wt. %
O	33.56
Fe	7.78
Ni	5.02
Na	17.53
Mg	0.74
Al	2.53
Si	0.35
Cl	29.79
Ca	2.52
Co	0.17
Total	100.00

**Table 1.** EDS elemental analysis of effluent samples collected during carbonated water flooding into Bentheimer core.

about 3 PV of carbonated water. As the displaced FW is replaced with the less dense carbonated water, the pressure drop falls sharply until carbonated water breakthrough at the effluent end of the core. Pressure drop was stabilized from about 3 PV to about 7 PV. In this period, the core is fully saturated with carbonated water, leading to stable flow. Unstable flow sets in from about 7 PV to the end of the test. In this period, the pressure drop is seen to rise sharply and fall to a rather haphazard behavior towards the end of the test. The EDS results in **Table 1** shows the effluent sample is composed predominantly of Na (17.53%) and Cl (29.79) which are probably the components of the effluent brine. The results also reveal the presence of minerals such as Fe, Si, Al, Ni, and Co which were not present in the saturating brine and the injected carbonated water. Therefore, Fe, Si, Al, Ni, and Co were most likely dissolved from the Bentheimer core through the interaction of the carbonated water and the rock minerals. The EDS analysis show low amounts of Fe, Si, Al, Ni, and Co, because only few particles were likely to be washed out of the core.

### *5.1.2 Effect of salt precipitation*

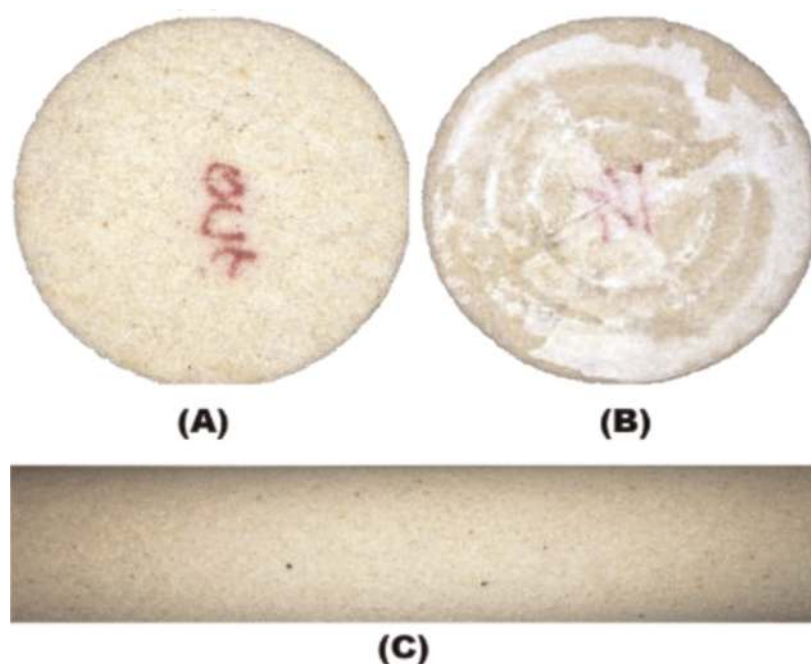
Salt precipitation is an existing injectivity challenge in natural gas wells. Kleinitz et al. [81] reported field observation of severe halite-scaling during natural gas production. Similar field experiences have been reported during injection, storage and production of natural gas [82–84]. In the context of field CO<sub>2</sub> injection, Baumann et al. [85] and Grude et al. [86] have reported evidence of salt precipitation effects in the Ketzin pilot reservoir and the Snøhvit field, respectively. More recently, Talman et al. [87] investigated drying and salt precipitation effects in a CO<sub>2</sub> injection well at the Aquistore site. Downhole images taken from the injection well, together with recovered samples revealed that scales of salts have formed on the inside of the injection well.

Miri and Hellevang [12] identified the processes leading to salt precipitation as: (1) immiscible two-phase CO<sub>2</sub>-brine displacement; (2) vaporization of brine into the flowing CO<sub>2</sub> stream; (3) capillary back-flow of brine towards the inlet; (4) diffusion of dissolved salt in the porewater; (5) gravity override of CO<sub>2</sub>; and (6) salt self-enhancing. While results from numerical modeling work reported by Roels et al. [88] suggested that precipitated salt could accumulate far from the wellbore, several research works [89–91] show that precipitated salt accumulates near the wellbore. Permeability impairment between 13 and 83% and porosity reduction between 2 and 15% have been reported from laboratory core-flood experiments [92–97]. These experimental findings have been found to be consistent with theoretical and numerical simulations [61, 98–100].

Pruess and Muller [89] suggested that pre-flush of the injection region with freshwater could reduce salt precipitation. However, Kleinitz et al., [81] have shown that freshwater injection could not mitigate salt precipitation if the flow area is completely plugged by solid salt. Fresh water also has a high tendency to react with rock minerals, leading to other injectivity impairment challenges such as clay swelling.

Sokama-Neuyam [101] grouped the mechanisms of salt precipitation into two successive processes: salt cake development at the injection inlet and drying effects. Salt cake forms on the surface of the core inlet during early stages of brine vaporization prior to drying. As drying commences, salt precipitates into pore spaces in the dry-out zone.

Sokama-Neuyam [101] investigated the development of salt cake on the surface of the injection inlet. They flooded a Bentheimer sandstone saturated with about 120 g/L

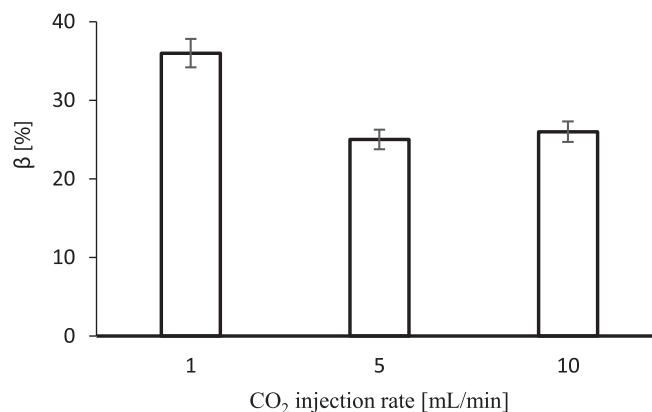


**Figure 3.** Photographs of Bentheimer core after CO<sub>2</sub> was injected at 1 mL/min into the core initially saturated with 120 g/L NaCl brine. (A) No salt cake observed at the core outlet. (B) Massive salt cake found at the injection inlet. (C) the entire core remains wet [101].

NaCl brine with about 100 PV of dry supercritical CO<sub>2</sub> at a rate of 1 mL/min. **Figure 3** shows photographs of the rock taken during inspection. In **Figure 3A**, we observe that no salt was formed at the core outlet. **Figure 3B** shows massive salt cake deposition at the core inlet and **Figure 3C** shows that the entire length of the core was still wet. They reported that salt cake formation at the injection inlet was caused by (1) High brine salinity and (2) Poor brine displacement at the injection inlet.

Furthermore, there are similarities between the increasing trend of injectivity impairment expressed by salt precipitation in Sokama-Neuyam [101] and those described by Yusof et al. [102, 103]. In their study, they examined the effects of brine salinity and brine type on CO<sub>2</sub> injectivity changes. They found that the injectivity reduction increased almost linearly between 6 and 27.3% as the brine salinity increases from zero to 100,000 ppm. The increasing growth of salt precipitation which reduced the porosity and effective flow area was identified as the main cause of the downtrend of CO<sub>2</sub> injectivity. It was also reported that the sandstone core saturated with monovalent salt such as NaCl and KCl was heavily impaired by salt precipitation as compared to the sandstone core filled by the divalent salt system (CaCl<sub>2</sub>). However, these findings may be somewhat limited by constant brine salinity of 30,000 ppm.

Moreover, to investigate the effect of drying, Sokama-Neuyam [101] flooded a Berea sandstone core initially saturated with NaCl brine with about 300 PV of supercritical CO<sub>2</sub> at a rate of 1 mL/min until the core was completely dried. The permeability of the core after drying was measured and a relative injectivity index  $\beta$  which measures the injectivity of the core before and after impairment was calculated. They then repeated the test at injection flow rate of 5 mL/min and 10 mL/min, keeping all other parameters constant, to study the effect of injection flow rate. **Figure 4** shows



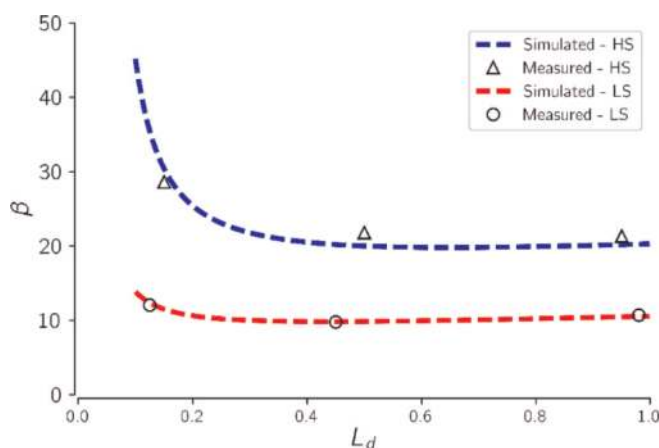
**Figure 4.** Effect of drying and salt precipitation on CO<sub>2</sub> injectivity. Injectivity impairment,  $\beta$  increased with decreasing CO<sub>2</sub> injection rate.

results of injectivity impairment induced by drying and salt precipitation at varying CO<sub>2</sub> injection flow rates.

From **Figure 4**, it was observed that CO<sub>2</sub> injectivity was impaired by about 36% for drying rate of 1 mL/min. Injectivity impairment decreased from 36% to about 25% when drying rate was increased to 5 mL/min and remained practically unchanged when the drying rate was further increased to 10 mL/min. Several researchers [94, 96, 104, 105] have earlier reported CO<sub>2</sub> injectivity impairment within a range (13–83%) that is in agreement with these figures. During drying and brine vaporization, when the concentration of brine exceeded supersaturation, salt precipitates into the pores in the dry-out region as also observed by Zuluaga et al. [106]. The deposited salts reduce the CO<sub>2</sub> flow area, impairing permeability and injectivity.

As drying progresses, a saturation gradient is established which draws more brine into the dry-out region through capillary backflow. Capillary backflow of brine leads to more salt deposition in the dry-out region. The capillary backflow of brine increases with decreasing drying rate because at high CO<sub>2</sub> injection flow rates, viscous forces overcome capillary forces. Therefore, less salts are precipitated in the dry-out region at high injection flow rates, inducing low injectivity impairment as observed in **Figure 4**. Injectivity impairment did not change when drying rate was further increased from 5 mL/min to 10 mL/min because at these injection flow rates, the resident brine is quickly swept out of the core, leaving out only immobile brine for salt precipitation.

During injection of dry supercritical CO<sub>2</sub> into brine-saturated sandstone cores, the dry-out region close to the injection inlet, extends into the core as more CO<sub>2</sub> is injected. The effect of extension of the dry-out zone on CO<sub>2</sub> injectivity is vital for understanding the underlying mechanisms of brine vaporization and salt precipitation. Sokama-Neuyam et al. [107] conducted experimental and theoretical study to investigate the development of the dry-out zone and estimate the impact of extension of the dry-out region on CO<sub>2</sub> injectivity. **Figure 5** shows the impact of the advancing dry-out zone quantified by a dimensionless dry-out length,  $l_d$  on CO<sub>2</sub> injectivity impairment  $\beta$ . From **Figure 5**, CO<sub>2</sub> injectivity impairment peaked at the onset of drying. Injectivity impairment decreased to a minimum at  $l_d$  of about 0.45 and then rose slightly as the dry-out zone approached the core effluent end. At the



**Figure 5.** The impact of the dry-out length ( $L_d$ ) on CO<sub>2</sub> injectivity impairment ( $\beta$ ) induced by salt precipitation. Magnitude of injectivity impairment increased when brine salinity was doubled from 75 g/L to 150 g/L but successive changes in injectivity impairment was not influenced by change in brine salinity [107].

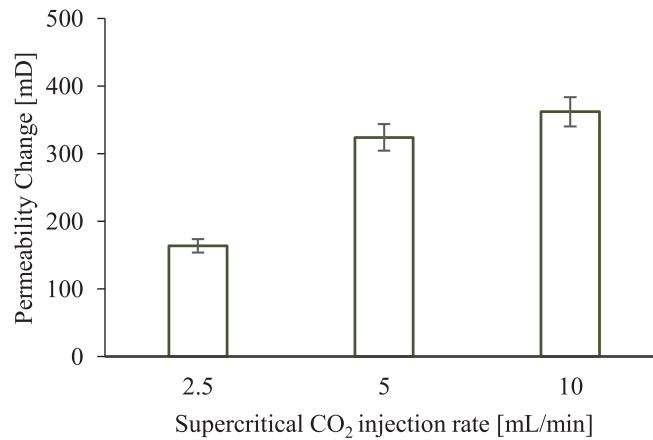
start-up of drying, two mechanisms could impair CO<sub>2</sub> injectivity: salt precipitation and relative permeability effects. Brine vaporization rate is at its highest close to the inlet region because of the high capillary driven back-fluxes. As more brine is vaporized, more salt is precipitated into the pores which in turn increased the resistance to flow in this region. As the drying front advances into the core, brine vaporization and salt precipitation decreased as most of the brine were drawn into the inlet region by capillary backflow, leaving the remaining section of the core with less amount of brine available for salt precipitation. When the core is almost completely dried, brine vaporization and salt precipitation in around the effluent end of the core are almost negligible.

To meet the global CO<sub>2</sub> emission reduction target, large injection rates will be required. Thus, after salt precipitation, there is continuous injection of CO<sub>2</sub> into the reservoir. The effect of drag forces on the deposited salt was studied by Sokama-Neuyam et al. [108]. A Berea core sample was initially vacuum saturated with FW and vaporized to complete dryness to precipitate salt into the pores. The liquid CO<sub>2</sub> permeability of the core and pressure drop across two sections of the core were measured with a pressure-tapped core holder. The core was then flooded with about 150 PV of supercritical CO<sub>2</sub> at a constant injection rate of 2.5 mL/min. During this period of CO<sub>2</sub> injection, drag forces were expected to act on the precipitated salts. Permeability and pressure drop across the same sections of the core was measured after CO<sub>2</sub> flooding. **Figure 6** shows permeability change induced by the effect of drag forces on precipitated salts at varying injection rates.

The net drag force exerted by supercritical CO<sub>2</sub> on precipitated salts depends strongly on the volumetric injection flow rate ( $v$ ), increasing with increase in flow rate. The precipitated salts are held to the pore walls mostly by gravitational and electrostatic forces offered by the complex pore structure and rock minerals. If drag forces overcome these attractive forces, the accumulated salts could be dislodged or redistributed in the pores, altering the permeability as a result. The magnitude of permeability change should therefore be proportional to the drag force which in turn depends on the injection flow rate.

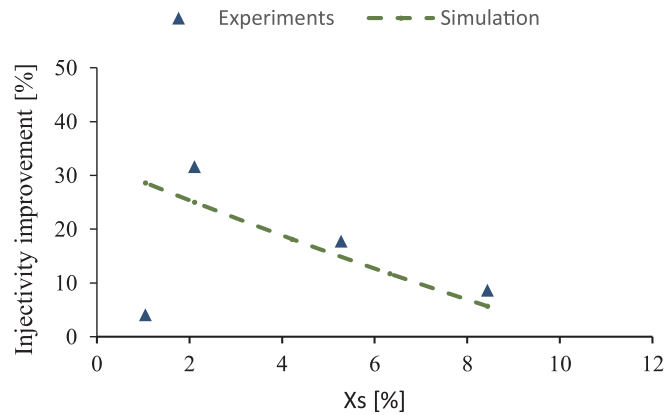
CO<sub>2</sub> alternating Low Salinity Water Flooding (CO<sub>2</sub>-LSWAG) is a promising EOR technique [37, 38, 109, 110]. Sokama-Neuyam et al. [97] investigated CO<sub>2</sub> alternating





**Figure 6.**

The impact of CO<sub>2</sub> injection flow rate on the effect of drag on permeability after salt precipitation. Permeability change is the difference between the core permeability after salt precipitation and before drag test and the permeability after drag test [108].



**Figure 7.**

Effect of diluent brine salinity on CO<sub>2</sub> injectivity change induced by alternate injection of supercritical CO<sub>2</sub> and LSW [111].

low salinity water injection as a potential technique to mitigate salt precipitation effects on CO<sub>2</sub> injectivity. After salt precipitation, a slug of diluent was injected to dissolve the precipitated salts, thus temporarily improving CO<sub>2</sub> injectivity. The diluent used was low salinity water (LSW) which was prepared by diluting FW to lower brine salinity. **Figure 7** shows injectivity improvement obtained as a function of mass fraction of salt ( $X_s$ ) in the diluent brine.

In general, CO<sub>2</sub> injectivity improved from 8.66 to 31.62% when the mass fraction of salt in the diluent,  $X_s$ , was decreased stepwise from 8.44 to about 2.11. The solubility of precipitated minerals in the diluent LSW increased with decreasing brine salinity, because, as the brine is further diluted, more free water molecules become available to interact with the precipitated salts. At  $X_s = 1.06$ , injectivity dropped significantly and the experimental data deviates dramatically from the simulation results, signifying additional injectivity impairment other than salt precipitation. At this point, the diluent starts to interact chemically with the rock minerals. Interaction between rock minerals and the diluent could induce clay swelling and colloidal transport, which have the tendency to aggravate CO<sub>2</sub> injectivity impairment.

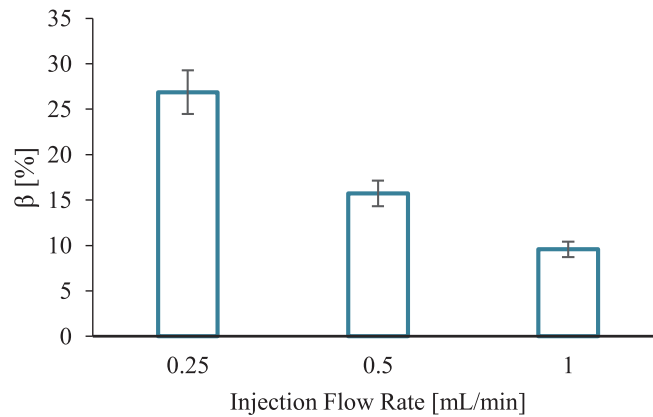
## 5.2 Transport effects

The mechanisms of fines migration and the impact of particle transport on the petrophysical properties of reservoir rocks have been previously researched. Khilar and Fogler [112] presented the mechanisms of colloidal and hydrodynamic induced release of fine particles in porous media. Muecke [113] investigated parameters controlling the movement of fine particles within the pore spaces. They identified the pH and salinity of formation brine, flow rate and temperature as some of the underlying parameters. Khilar and Fogler [114] asserted the existence of a critical salt concentration below which the pore fluid could weaken the Van der Waal's forces holding fine particles to the pore walls. Gruesbeck and Collins, [115] investigated the effect of hydrodynamic forces on the release and transport of fines. They identified a minimum interstitial velocity for fines entrainment. The effect of two-phase flow and rock wettability on fines entrainment has been experimentally investigated by Sarkar and Sharma [116]. They found that, the wettability of the core could affect the extent and rate of permeability impairment induced by migratory fines. Analytical models have been developed by Sharma and Yortsos [117] to investigate the mechanisms of size exclusion and quantify the effect of particle entrapment on rock permeability. Many other studies of fines migration in porous media under various conditions have been reported [118–120]. A thorough analysis of formation damage induced by migratory fines can be found in Civan [121].

We have already discussed that geochemical CO<sub>2</sub>-brine-rock reaction could generate secondary minerals into the pore fluid [73, 122, 123]. In addition, CO<sub>2</sub>-brine interactions could alter the pH of formation fluid which could induce the release of formation fines from the pore walls [115, 124]. While flowing with the injected fluid, the mineral particles could clog pore channels and impair injectivity. Whether entrapment or piping of fines will dominate the flow depends on characteristics of the generated fine particles, the porous medium and the permeating fluid in which the particles are suspended [125–127]. Pore structure, the size and concentration of the minerals and the hydrodynamic and colloidal conditions of the suspending medium could also affect their impact on CO<sub>2</sub> injectivity. Under radial flow conditions, plugging effects could be limited to the near well region where fluxes are highest.

The general mechanisms of fines mobilization in porous media have been well researched and understood. However, the unique properties of supercritical CO<sub>2</sub> including its gas-like viscosity and liquid-like density [128] coupled with the expected high CO<sub>2</sub> injection rates required to meet global emission reduction targets and the drying effect of supercritical CO<sub>2</sub> makes fines mobilization under CO<sub>2</sub> injection conditions a unique challenge that must be investigated separately. Adaptation and extension of previous general findings on fines migration is required to understand the mechanisms and impact of fines migration within the context of CO<sub>2</sub> injection.

Sokama-Neuyam [80] conducted core-flood experiments to measure the effect of dissolution on injectivity. A Berea sandstone core sample with known permeability was initially saturated with FW, and then flooded with about 25 PV of carbonated water at 80 bar and 60°C at constant injection rate of 0.25 mL/min to release and mobilize fine particles in the rock. The permeability of the core after carbonated water flooding was measured, and injectivity impairment index,  $\beta$ , was calculated. The experiment was then repeated at injection flow rates of 0.5 mL/min and 1.0 mL/min. **Figure 8** shows injectivity impairment decreased with increasing carbonated water injection flow rate.



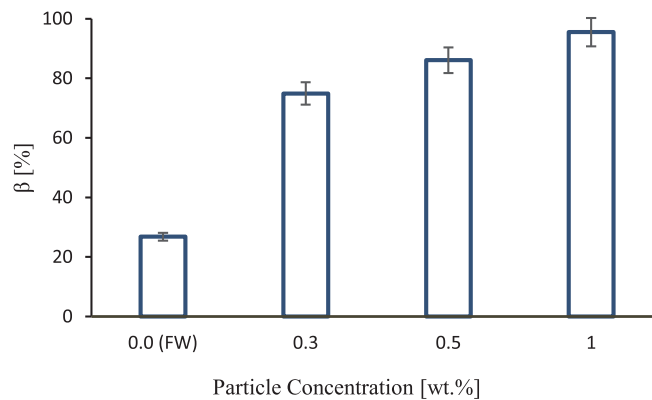
**Figure 8.**

*Effect of injection flow rate on injectivity impairment induced by fines plugging. Injectivity impairment decreased with increasing carbonated water injection flow rate [80].*

As injection flow rate is increased, the resident time of carbonated water in the rock is shortened. The number of fines generated will then decrease as the injection flow rate is increased. In addition, at high injection flow rate, hydrodynamic forces could lift smaller particles out of the core with the effluent fluid. The number of fines available to plug the rock, and therefore the chances of injectivity impairment, will reduce as carbonated water injection flow rate is increased. Up to 26% injectivity impairment was induced by mineral dissolution and fines mobilization during carbonated water injection into the Berea sandstone cores. Injectivity impairment decreased as injection flow rate was increased.

Sokama-Neuyam et al. [129] attempted to quantify and compare the individual effects of fines mobilization and salt precipitation on CO<sub>2</sub> injectivity. Mono-disperse colloid solutions were used to represent the pore fluid containing particles after mineral dissolution. A Berea sandstone core sample was initially saturated with mono-disperse colloid solution with average particle size of 0.08  $\mu\text{m}$  and particle concentration of 0.3 wt.% and flooded with about 40 PV of supercritical CO<sub>2</sub> at 5 mL/min to complete dryness. The relative injectivity change,  $\beta$ , was calculated from the permeability of the core measured before and after it was exposed to mineral impairment. The experiment was repeated for particle concentrations of 0.5 and 1.0 wt.%. **Figure 9** shows injectivity impairment induced as a function of composition of the pore fluid.

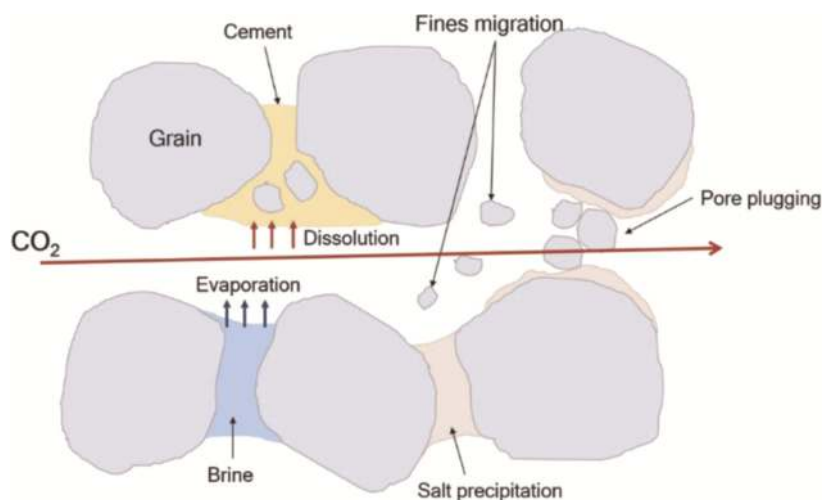
In **Figure 9**, while salt precipitation reduced injectivity by about 26.8%, particle concentration of 0.3 wt.% impaired injectivity by 74.9% through fines mobilization. About 1.0 wt.% of particles in the pore fluid almost plugged the rock. When CO<sub>2</sub> invades the pores, the mono-disperse particles could plug the narrow pore channels through bridging, surface deposition, or multi-particle blocking. As particle concentration increases, the distance between suspended particles shortens, enhancing multi-particle blocking of the invaded pores. On the other hand, precipitated salts coat the pore walls to reduce the flow area. While salt precipitation reduces the flow area, fines entrapment could plug and isolate the flow path, making them inaccessible to fluid flow. The results suggest that, under linear flow conditions, fines mobilization could induce severe CO<sub>2</sub> injectivity impairment comparable to the impact of salt precipitation.



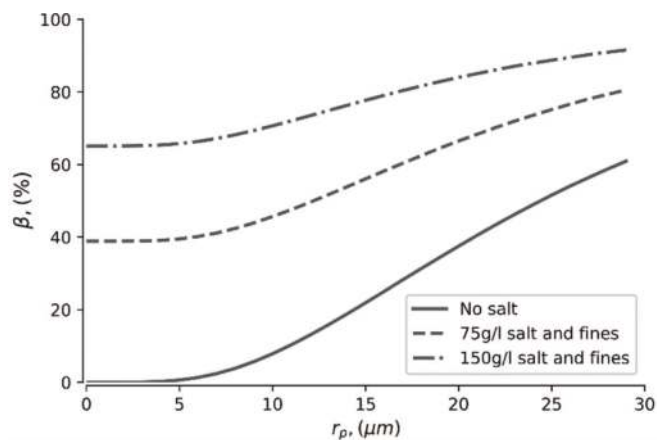
**Figure 9.** The relative impact of fines mobilization and salt precipitation on CO<sub>2</sub> injectivity. Fines migration had a more severe impact on injectivity compared to salt precipitation [129].

### 5.3 The coupled effect of salt precipitation and fines mobilization

Preliminary theoretical studies suggests that fines mobilization could compound CO<sub>2</sub> injectivity impairment induced by salt precipitation [80, 101]. After salt precipitation, the deposited salt reduces the pore spaces, increasing the jamming ratio of particles being transported in the flowing stream. A schematic diagram that summarizes the role of mineral dissolution, salt precipitation and fines migration mechanisms on CO<sub>2</sub> injectivity impairment is shown in **Figure 10**. Salt precipitation increases the susceptibility of the rock to fines entrapment. Sokama-Neuyam et al. [131], developed a dynamic core-scale model based on experimental observations to investigate the coupled effect of fines mobilization and salt precipitation on CO<sub>2</sub> injectivity. The effect of brine salinity, initial core permeability and the order of coupling were studied. **Figure 11** shows injectivity impairment induced by the combined effect of particle entrapment and salt precipitation compared to the effect of only fines mobilization.



**Figure 10.** Schematic diagram of mineral dissolution, salt precipitation and fines migration mechanisms during CO<sub>2</sub> injection into saline aquifer [130].



**Figure 11.** Effect of fines mobilization compared to coupled effect of salt precipitation and particle entrapment [131].

Generally, injectivity impairment induced by particle entrapment increases with average particle size in the inlet fluid,  $r_p$ . For the base case (No salt), particles less than the average pore radius (6  $\mu\text{m}$ ) did not induce significant injectivity impairment. These particles generally piped through the pores as they have a jamming ratio less than 1.0. As  $r_p$  increases, more pores attain a jamming ratio greater than 1.0 and are plugged and isolated. As more pores are plugged and isolated, the flow area available to injected  $\text{CO}_2$  decreases, impairing the injectivity. **Figure 11** also shows that coupling particles entrapment and salt precipitation increases  $\text{CO}_2$  injectivity impairment significantly with the severity of impairment increasing with initial brine salinity. The deposited salt reduces the pore spaces and increases the jamming ratio. Particles that could pipe through the clean pores would be entrapped after salt precipitation. Thus, salt precipitation could compound the effect of fines mobilization on  $\text{CO}_2$  injectivity.

## 6. Challenges and opportunities

Reviewing the previous works on  $\text{CO}_2$  injectivity has thrown up many questions in need of further investigation. Some of the highlighted concerns are as follows.

- Most of previous studies that have been made in the context of  $\text{CO}_2$  storage have focused on salt precipitation. Little experimental work has been performed to better understand near-well rock compaction, impact of temperature and operational parameters (drilling mud, residual hydrocarbons in pores). To give research-based advice on injectivity loss it is necessary to take geology and geomechanics into account. These cannot be reliably assessed without studying radial flow of  $\text{CO}_2$  (both injection and backflow) under true subsurface stress conditions. Temperature issues should also be considered for the cases such as injection of cold  $\text{CO}_2$  with potential to fracture the near-well rock and thus increase the permeability.
- Further work is required to enable direct pore-scale, real-time visualization of fluid-solid interactions with representative pore-geometry and realistic surface interactions between injectant, reservoir fluids and the formation rock.



- Current research approach to understand the reaction mechanism of CO<sub>2</sub>-brine-rock for CO<sub>2</sub> sequestration are mainly limited to pure CO<sub>2</sub>. However, due to high cost of gas purification, industrially sourced injection stream CO<sub>2</sub> contain impurities such as SO<sub>2</sub> which when dissolved in formation water will increase acidity beyond that of carbonic acid formed through CO<sub>2</sub> dissolution alone and may also have different reaction mechanism towards rock minerals present in subsurface CO<sub>2</sub> storage. Therefore, there is a need for a study to evaluate the effect of CO<sub>2</sub> and impurities in brine solution on chemical and physical rock properties.
- Majority of the dynamic CO<sub>2</sub> injection experiments are conducted within a limited time considering the limitation of available equipment. Therefore, there is an argument among the researchers on the sufficient CO<sub>2</sub> solubility into the brine and establishment of acidic environment that is critical in geochemistry study. Any future work to extend the CO<sub>2</sub> exposure time is highly recommended to identify the extended scale accumulation, mineral dissolution, and fines migration problems.
- Recent work on wettability during CO<sub>2</sub> injection has focused on the wettability changes of glass micromodel and shows that with increasing ionic strength, contact angle increases with increasing residence time. However, with understanding that fine particles released during CO<sub>2</sub> injection, may have not been exposed if the surface of the rock, its wettability may be slightly different or remain unaffected. Investigating the wettability of the released fines particles may provide insight on the behavior in which the fines particles migrate and accumulate.
- When it comes to mechanical stability, initial studies have shown that cyclic injection and shut-in may influence borehole deformation. This is probably exacerbated if some drawdown is experienced upon shut in. Shut-in cycle frequency effect on the amount of borehole deformation should be studied in more detail.
- Available field data and observation reports indicate the occurrence of injectivity loss. However, detail investigation analysis on the main issues that caused the problem is limited to certain fields. While there are more than 20 active fields worldwide, it should not be challenging to share their findings since the CO<sub>2</sub> mitigation would require collaboration.

## **7. Conclusion**

In this chapter, a review of the current literature indicates that dissolution, precipitation, and fines mobilization are the main mechanisms that cause CO<sub>2</sub> injectivity impairments especially in deep saline reservoirs. Dissolution of carbonate minerals due to CO<sub>2</sub>-brine-rock reaction is dominant and could increase the porosity and permeability of sandstone core samples. On the other hand, detachment, precipitation of salt and clay minerals and deposition of fines particles would decrease the permeability and even clog the flow paths despite net dissolution. The effect of these two seemingly opposing processes on CO<sub>2</sub> injectivity has been clearly demonstrated

through numerous experimental studies supported by some field reports. However, the results are case dependent and lack generality in terms of quantifying the petrophysical damage.

There are many underlying parameters with positive and negative impacts on CO<sub>2</sub> injectivity. It has been highlighted that injection scheme (flow rate, time frame), mineral composition (clay content, sensitive minerals), particulate process in porous media (pore geometry, particle, and carrier fluid properties), and thermodynamic conditions (pressure, temperature, salinity, CO<sub>2</sub>, and brine composition) have substantial effect on fines migration during CO<sub>2</sub> injection. However, there is abundant room for further progress in determining the impact of different fluid-rock mechanisms on CO<sub>2</sub> injectivity.

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