

Review on Experimental Investigation into Formation Damage during Geologic Carbon Sequestration: Advances and Outlook

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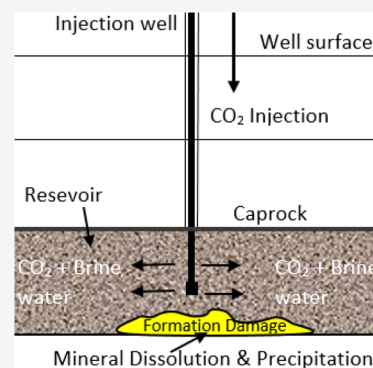
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ABSTRACT: Carbon capture and storage has been identified as an important and viable technology for climate change mitigation. The technology allows CO₂ generated from large-scale sources, such as power plants and other heavy industries, to be captured and stored in deep geological formations. However, when CO₂ is stored in geological formations, there are possibilities of formation damage, which may reduce injectivity and storage capacity. In this study, formation damage during CO₂ injection and storage is reviewed through different experimental studies. The study has shown that the interaction between CO₂, formation water, and rock minerals often results in mineral dissolution and precipitation, which affect reservoir permeability and porosity, which could possibly cause formation damage, compromising the reservoir storage capacity. This study also reveals that formation damage could be caused by the precipitation of sulfate scales, salt, and carbonate minerals. Additionally, in several studies, rock minerals were observed to dissolve and create free particles that were transported to occupy pore spaces along the fluid flow path, reducing permeability and impacting CO₂ injectivity and storage. It is worth noting that the reviewed experiments present short-term effects of formation damage on geological formations, while in reality, CO₂ storage is a long-term project, thus eliciting the need for more studies in that regard.



1. INTRODUCTION

Controlling climate change requires prompt action to mitigate the effects of anthropogenic CO₂ emissions to the atmosphere, which can lead to global warming.^{1,2} Capturing CO₂ released from massive fuel combustion³ activities, such as transportation, power, and industrial processes,^{4–11} and storing it in deep geological formations, like aquifers, depleted reservoirs, coal seams, and salt caverns, is a crucial and essential strategy for addressing global warming mitigation.^{12–17} However, CO₂ is injected into the reservoir formations at a depth more than 800 m deep to achieve this objective. These formations have high permeability and porosity and an effective caprock with low-permeability properties to act as a seal in preventing CO₂ from escaping to the surface.^{18–22} Thus, according to studies, injecting a substantial volume of CO₂ with its associated impurities into geological formations poses several challenges, including chemical changes to the formation rock that cause formation damage and reduce the injectivity capacity of reservoirs.^{23–28}

In addition, formation damage happened when a change in geochemical equilibrium occurred as a result of CO₂ impurities, brine water, and rock mineral reactions.²⁹ According to recent research by Khurshid and Afgan, the interaction of CO₂, water, and rock could lead to severe formation damage, such as plugging and blocking the porous

reservoir system and impairing the permeability of the formation reservoir.^{30,31} Additionally, the reactions may alter the grain size, wettability, and porosity of the rock formation as a result of mineral dissolution and precipitation, which consequently would affect CO₂ injection into a geological formation, caprock integrity,³² and structural injectivity of the reservoir.^{33–36} Therefore, to accurately predict the injectivity plan and storage capacity, particularly for long- and short-term storage of CO₂ in a geological formation, it is crucial to determine the influence of CO₂ and its mechanisms on brine water and rock mineral reactions.^{37–40}

Numerous studies have reported that injecting CO₂ into geological formations can disrupt the geochemical equilibrium through dissolution and precipitation processes, which affect the reservoir mineral phases.^{14,41–47} However, it has been mentioned that, by having a small number of secondary minerals present in the formation after CO₂ injection, considerable changes in permeability and porosity can be

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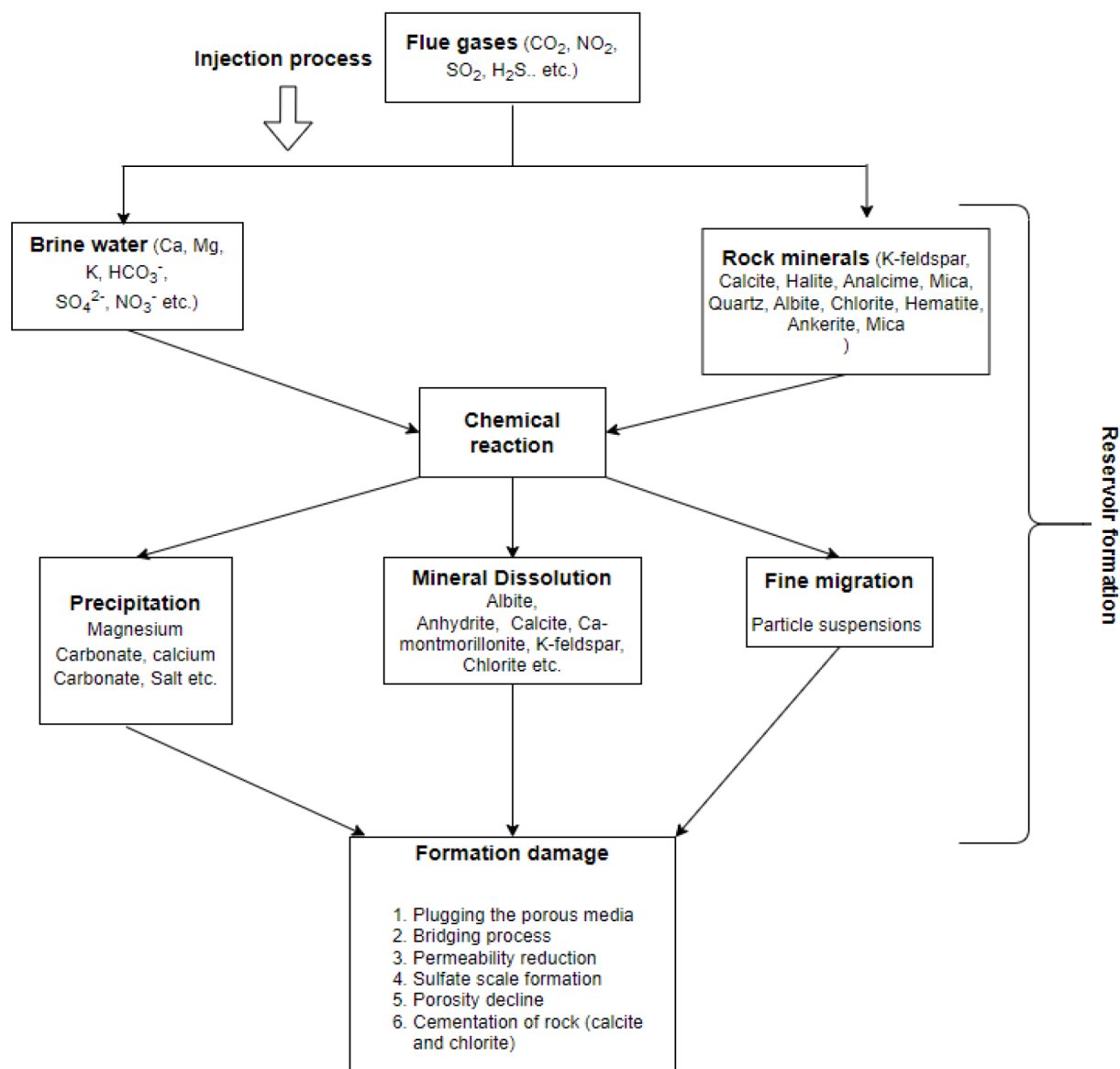


Figure 1. Conceptual flowchart of carbon dioxide injection and storage, which results in formation damage.

seen through precipitation.^{48,49} Similarly, Shao et al. reported that the permeability could decrease when the precipitation size is nearly equal to the size of the pore throats in the porous system of the formation.²⁹

According to studies by Karaei et al.,²⁰ Al-Ameri et al.,⁵⁰ and Dawson et al.,⁵¹ the injection of CO₂ into reservoir formation dissolves rock minerals by *in situ* weak and strong acids. However, reactive rock minerals, like magnesium and calcium, have also been observed to interact and react with CO₂ to generate carbonate precipitations.^{52,53} Furthermore, carbonate precipitations have been witnessed during high and low pH measurements.^{20,54,55} Different kinds of literature presented CO₂ sequestration in geological formations through solubility mechanisms. Their results have reported that solubility mechanisms are significantly affected by the temperature and pressure of injected CO₂ and the salinity of the formation reservoir.^{56–62}

On the other hand, other studies have indicated that permeability will be reduced, while others have stated that permeability will be increased during the sequestration of

CO₂.²⁰ These reports showed the critically significant challenges during CO₂ injection, which need addressing and attention for future projects. Some of the challenges presented with these reports were geo-mechanical and geochemical, such as dissolution and precipitations^{63–68} of reactive minerals that can change the injectivity of CO₂ and block pores during precipitation, consequently reducing the permeability of the formation.^{69,70}

However, the Basava-Reddi et al. investigation⁷¹ revealed the existence of various gaseous contaminants in CO₂ streams injected into geological formations, such as SO₂, O₂, N₂, and H₂S. This stream can adversely impact transport, sequestration, and storage processes. For example, SO₂ and NO₂ may react with brine water to form an acidic medium, which will then react with rock minerals to cause dissolution and precipitation, compromising caprock integrity near-wellbore zones and allowing leakage of CO₂ to the surface or overlying formations.¹⁷ Additionally, the precipitate formed may block porous media, lowering CO₂ injectivity.^{72,73}

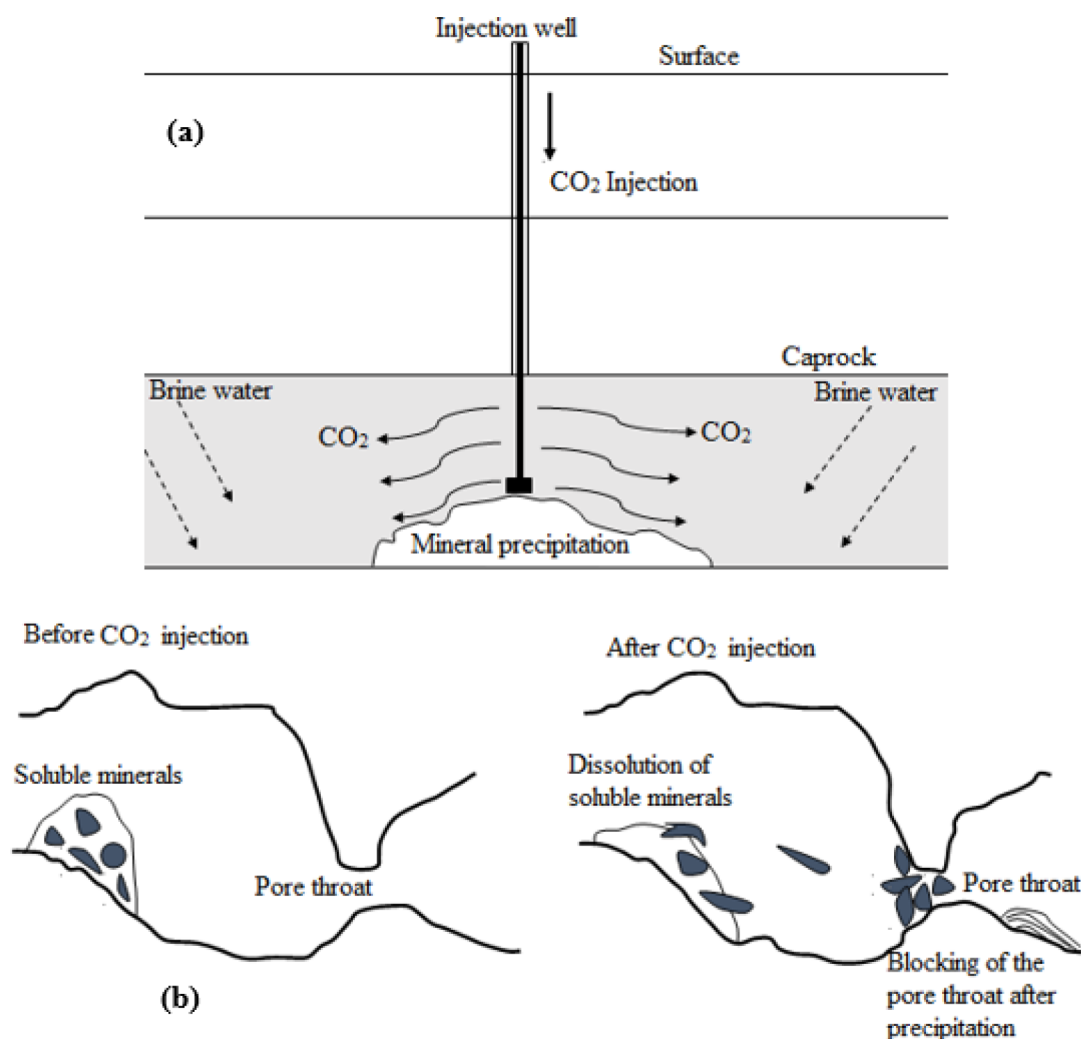


Figure 2. Conceptual models of formation damage mechanisms during CO₂ injection and storage: (a) salt precipitation and (b) dissolution and precipitation of active minerals. This figure was reproduced with permission from ref 116. Copyright 2014 Elsevier.

Most of the research did not study in detail the formation damage caused by CO₂ storage in geological formations. In contrast, Mohamed and Nasr-El-Din⁷⁴ investigated formation damage in only saline carbonate reservoirs. At the same time, other kinds of literature pay little attention to the impact of formation damage on other geological formations, such as sandstone formations. Xie et al.⁷⁵ investigated the formation damage caused by fine migration during CO₂ storage when kaolinite minerals were present. Additionally, the research performed by Luc et al. on reactions between CO₂, NO_x, and the core of CO₂ storage rock claimed that fine migrations were observed in scanning electron microscopy (SEM) images, which might result in permeability change.⁷⁶ Although injecting and storing CO₂ in geological formations can offer long-term storage capacity and security, it can also release particle suspensions at an unprecedentedly high flow rate, which may impact the formations.

Therefore, the primary objective of this research is to provide a comprehensive assessment of formation damage during CO₂ injection and storage in geological formations through experimental studies. The formation damage has been seen as the potential challenge that lowers the injectivity and storage capacity because the result the project storage capacity and economic viability might be compromised. The study

describes effects of CO₂ injection and storage on mineral dissolution and precipitation, permeability changes, and alteration of grain size and sorting properties, which significantly caused the formation damage. Additionally, the mechanisms of formation damage are elaborated in detail with vivid examples from the lab findings that will provide insight and awareness during injection and storage activities. Furthermore, the challenges and perspectives for further research were clearly described and recommended for necessary action. This study may offer essential information during the feasibility study and implementation of new CO₂ injection and storage projects in geological formations.

2. CONCEPT OF FORMATION DAMAGE DURING CO₂ SEQUESTRATION

Formation damage restricts and weakens the flow path of fluids to the porous system, affecting the injectivity and economic viability of projects.^{77–79} Understanding the mechanisms of CO₂ storage and injection into the geological formation is crucial because the properties of the formation may change, causing formation damage to the formation rock.^{80–82} Reservoir formation is altered sooner or later after CO₂ enters it.⁸³ Similarly, CO₂ can dissolve into brine water, resulting in changes in the pH of the media of the reservoir. In addition,

CO₂ can react with active minerals in the reservoir rock, causing precipitation and dissolution, which change the formation properties, resulting in abnormal reservoir performance;⁸¹ the illustration of the conceptual flowchart of CO₂ injection to the deep reservoir and the resulting formation damage is presented in Figure 1, while its mechanisms of formation damage are demonstrated in Figure 2.

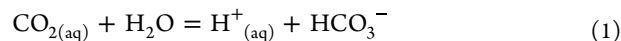
According to Sbai and Azaroual, several mechanisms have contributed to the formation damage, such as active mineral precipitation and trapped particles, causing blocking/plugging and bridging processes, particularly in porous environments. Also, clogging of the suspended particles at the pore throats during an unprecedented flow rate of CO₂ storage could result in restriction of production and a loss in permeability surrounding the injector zone.^{34,45} Research by Mohamed and Nasr-El-Din showed the reduction of CO₂ injectivity in carbonate rock as a result of the formation damage resulting from the precipitation of carbonate minerals and sulfate scales. Usually, sodium sulfate in brine water causes formation damage as a result of the precipitation of calcium sulfate.⁸⁴ However, high formation damage was reported from the dolomite rock as a result of the reaction between CO₂ and silicate minerals. Similarly, in either homogeneous or heterogeneous rocks,⁸⁵ formation damage may happen as a result of precipitation or the blockage of water,⁷⁴ making the damage more severe.⁸⁶

Furthermore, numerous researchers from the field and experimental studies have demonstrated that the dissolution of active minerals may raise permeability or porosity,^{87,88} but this is not well-suggested because the increase of permeability may result in a high flow rate of CO₂ during injection, which could potentially result in a reduction in the pressure gradient, particularly at the near wellbore zones.⁸⁹ On the other hand, the permeability reduction may significantly decline during mineral precipitation and mobilization of the physically inert minerals.⁹⁰ Similarly, permeability may be substantially reduced when secondary mineral precipitates and seals the pore throats and fractures of the reservoir formation, leading to formation damage.⁹¹ Therefore, understanding the impact of the formation damage is essential for designing and planning CO₂ injection and storage operations in geological formations.

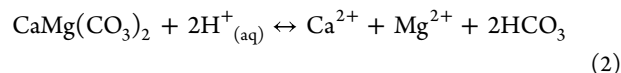
2.1. Alteration of Permeability. Permeability is a key characteristic of geological reservoirs that allows for the most effective use of the formation.⁹² It is crucial to analyze for both economic viability and appropriate storage. According to an experiment conducted by Karai et al.,²⁰ there is significant formation damage when confining or injection pressure and temperature change in carbonate aquifers with a low-permeability formation in the Iranian formation. The experiment involved three different fluids: brine water, seawater, and freshwater. These fluids were injected into core samples to identify potential changes on permeability.

When confining pressure is applied using brine water, seawater, and freshwater, a permeability reduction (60%) in brine water (injection core sample) was observed. However, the decrease in permeability (on an average of 35%) was observed in brine water injection cores when a constant pressure was applied to all three samples, and this is because of the incompatibility reactions between CO₂ and mineral composition in brine water, which resulted in precipitation and dissolution that might plug and block the porous system.⁹³ Other researchers have reported that CO₂ injection into cores having brine saturated resulted in the decline of permeability as

a result of salt precipitation that plugs the porous media, leading to an injectivity decline.^{94–97} Further analysis has shown that, when brine water was used under similar circumstances, it was shown that permeability significantly decreased by 35%. CO₂ reacts with the brine water already present in the porous systems to form carbonic acid according to eq 1.⁸²



Then, carbonic acid may dissociate into hydrogen and bicarbonate ions, changing the pH of the fluid (pH drop).⁹⁸ Hydrogen ions may react with rock and dissolve the rock in porous media, while ions of Mg²⁺ and Ca²⁺ seem to increase. Therefore, dissolution and precipitation may occur, causing formation damage and loss of the rock integrity.⁹⁹ This information is summarized in eq 2.



The experiment also examined the impact of confining pressure ranging from 5 to 15 MPa while maintaining the injection pressure constant at 1 MPa at 100 °C. The outcome has demonstrated that all forms of fluid, starting with freshwater, seawater, and brine water, have experienced a drop in permeability. However, freshwater caused a 50% drop in permeability; seawater caused a 53% reduction; and saline water caused a 60% reduction.

Additionally, it should be noted that the permeability trend in the saline formation is more severely affected by formation damage during CO₂ injection, in which the permeability was observed to drop more than in other samples. The permeability drop is due to the chemical processes that led to the formation damage from salt precipitation and active mineral dissolution.^{45,100}

A similar experiment was described by Aminu et al.³⁵ to assess the effects of injecting CO₂ impurities, such as NO₂, SO₂, and H₂S, into the saline aquifer formation at the Bunter Sandstone Formation in the Southern North Sea. The experiment was divided into four CO₂ streams: pure CO₂, CO₂ with NO₂, CO₂ with SO₂, and CO₂ with H₂S. In a test setup, the crushed rock sample was injected into each of the four streams of CO₂ independently (titanium pressure device). The findings indicated that CO₂ impurity impacted the formation damage, which significantly reduced permeability and grain size. Additionally, Figure 3 displays the findings of the experiment and the trend of permeability drop after the investigation. Observation from Figure 3 demonstrates that the permeability was raised, and the importance of the formation damage happening was modest when pure CO₂ was introduced. Furthermore, after exposure to pure CO₂, the permeability of the formation rock has risen by 5.4%. Even though pure CO₂ can react with saline water to create carbonic acid (eq 2), the dissolution is only partial and leaves behind certain solid minerals that may make carbonic acid difficult to dissolve chemically.

Similarly, Sass et al. discovered the same trend that the presence of NO₂ and SO₂ can decrease the permeability and injectivity time as a result of the formation of precipitation.¹⁰¹ Also, from Figure 3, the permeabilities of NO₂ and SO₂ were observed to decrease by 41.9 and 6.6%, respectively. The decrease in permeability is because the acid is strong and may dissolve more solid minerals than pure CO₂, leading to an

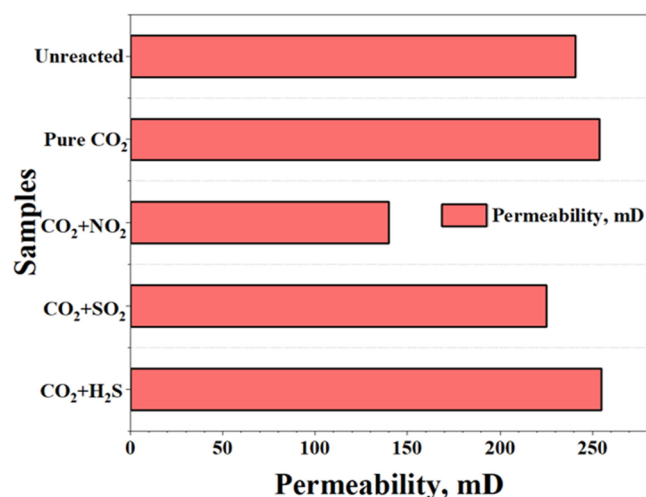
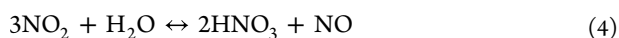
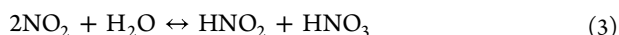


Figure 3. Permeability changes caused by CO₂ injection at the Bunter Sandstone Formation of the U.K. This figure was reproduced with permission from ref 35. Copyright 2018 Elsevier.

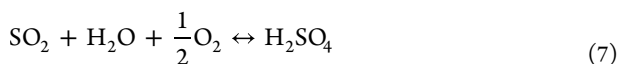
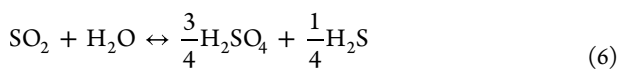
increase in mineral dissolution and precipitation that clogged and obstructed the porous networks and reduced the typical injectivity of CO₂ into the geological formation.¹⁰² Additionally, the permeability of CO₂ with H₂S increased by 6%, indicating that the formation is not significantly damaged in terms of permeability when exposed to H₂S. According to research by Zhang et al., when CO₂ and H₂S are injected into a geological formation concurrently, the solubility of CO₂ decreases, while the dissolution of H₂S may increase the dissolution of CO₂.¹⁰³ However, the following is a possible description and chemical reaction of acidic gases (NO₂ and SO₂) that influenced the formation damage.

(i) The influence of NO₂ on formation damage: The exposure of NO₂ to the formation rock may react with brine water to form nitric acid, a strong acid according to eqs 3 and 4 below.



After this reaction, the pH may show drastic changes by dropping its values, proving that nitric acid has been formed. Also, it was further observed that the presence of NO₂ resulted in the dissolution of quartz of about 8.9% of the original present and a reduction of permeability. Wang et al. report that the dissolution of quartz from NO₂ may result in salt precipitation, which influences the permeability reduction of the rock formation.

(ii) The influence of SO₂ on formation damage: SO₂ in the rock formation may react with the formation water (brine) to form sulfuric acid, a strong acid. The possible reaction can be seen in eqs 5–7.



These reactions result in weak acids H₂SO₃ and H₂S, while strong acid H₂SO₄ forms in brine water. The reduction of the

pH as a result of CO₂ and SO₂ mixtures in a sandstone saline aquifer during injectivity and storage has been confirmed by Waldmann et al.¹⁰⁴

Yu et al. experimented to evaluate the interactions between CO₂, brine water, and rock formation in a depleted reservoir at the Qing 1 Formation. The core samples were obtained at a depth of 2427.54 m and simulated under reservoir conditions at a temperature of 100 °C and pressure of 24 MPa. The experimental results revealed that the permeability was reduced because of the production of new mineral precipitation phases, like solid and kaolinite mineral phases, as listed in Table 1.

Table 1. Permeability of the Core Samples before and after the Experiment¹⁰⁵

core sample	before the experiment (mD)	after the experiment (mD)
upstream	3	2
middle	6	5
downstream	29	24

Furthermore, the permeability reduction was contributed by the dissolution of clay minerals from the dissolution process of carbonate cement, which was transported to the fluid flow path mechanisms and deposited into the porous systems, causing and restricting the normal passage of the fluid.¹⁰⁵

Also, the experimental results have shown the permeability of the core sample test to decline from 2 to 1 mD as a result of the formation damage resulting from active mineral reactions that plugged the pore throat.¹⁰⁵ Similar studies were reported with the same trend of permeability reduction, such as research by Ross et al. on the experiment of CO₂ flooding in sandstone formation, which revealed a notable decline of permeability.^{106,107} In addition, the permeability decline was observed in weak minerals, like feldspar, carbonate, etc., dissolved under reservoir conditions when the experiment was conducted.¹⁰⁵ The recent experiment by Tang et al. investigated the effect of the CO₂–brine–rock interaction during the injection and storage. They examined the impact of CO₂ injection on the gas and water zone in the saline aquifer at a gas field in China's South Sea. They have shown that the CO₂–brine–rock interaction can happen in both zones of water and gas because, during the reaction, water can evaporate to the steam phase, while CO₂ reacts with the brine water, resulting in carbonic acid. Similarly, the study revealed that the CO₂–brine–rock interaction leads to formation damage because the reaction of the rock mineral may dissolve and form free particles that can be transported and fill the pore spaces along the pass of fluid flow, leading to a severe reduction of permeability.^{105,108}

Jeddizahed and Rostami investigated the evaporation and precipitation of salt during the injection rate and the effect of brine water on the permeability of the core samples. The experiment utilized sandstone rock, which was injected with supercritical CO₂. However, the sandstone rock was saturated with NaCl to analyze the salt precipitation during evaporation. The experimental results are presented in Figure 4.¹⁰⁹

Figure 4a shows a considerable decline in permeability when the injection rate is increased as a result of salt precipitation into the porous system, which restricts the normal flow of CO₂ and lowers the storage security and practicability. The permeability decline was observed to be 43, 50, and 62% for the injection rate of 20, 10, and 5 cm³/min, respectively.

Figure 4b shows that, when the salinity increases, the permeability decreases. Similarly, the salt precipitation

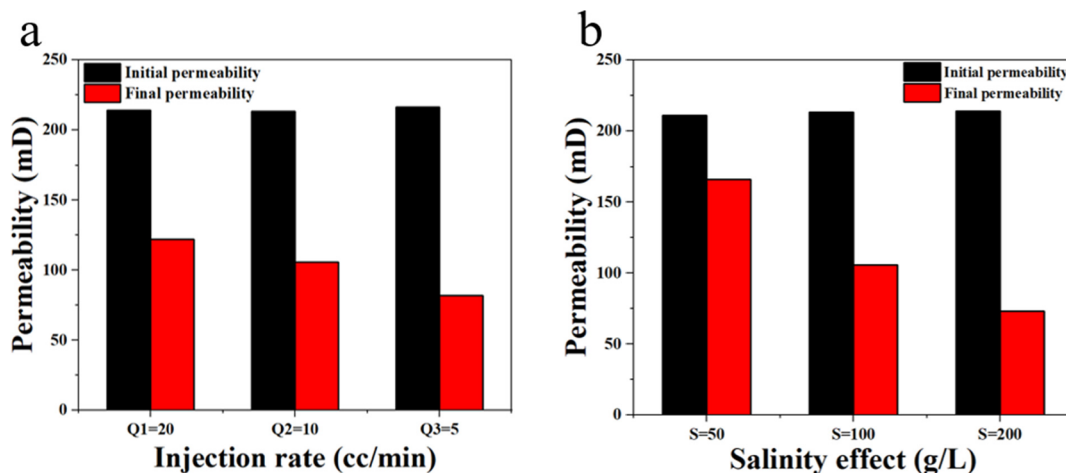


Figure 4. Description of permeability against the injection rate and salinity effect: (a) description of the injection rate and (b) salinity effect on the gas flow rate effect. This figure was reproduced with permission from ref 109. Copyright 2016 Elsevier.

increased, leading to formation damage. The reduced permeability was 21, 50, and 66% for the salinity concentration of $S = 50, 100,$ and 200 g/L, respectively.

A similar study of salt precipitation was performed by Bacci et al. They conducted laboratory work to research the alteration of permeability and porosity during the injection of CO_2 into a saline aquifer. The experiment utilized the core samples saturated with saline water from the St. Bees Sandstone Formation. The result was observed that, during the injection of CO_2 into the saline aquifer, the formation damage significantly occurred, caused by salt precipitation, affecting the permeability and porosity of the formation.¹¹⁰ Moreover, the permeability dropped from 30 to 86%, and porosity decreased from 4 to 29%. The result is presented in Figure 5.¹⁰⁷ From Figure 5, the change in permeability and porosity were observed after salt precipitation. The reduction of porosity was seen to be 22.59–16.02%, accompanied by the decline of permeability from 7.78 to 1.07 mD. In addition, the

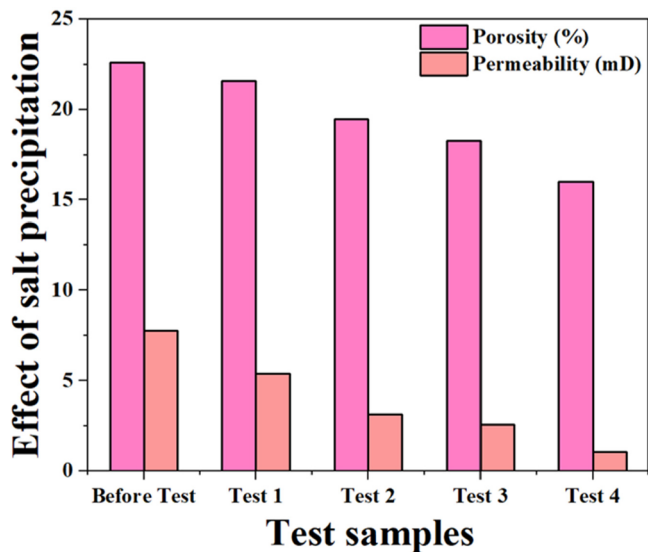


Figure 5. Effect of salt precipitation in porosity and permeability changes. This figure was reproduced with permission from ref 107. Copyright 2012 Elsevier.

overall permeability decline was 18–60%, while porosity was 1–2%.

Additionally, the experiment presented by Adebayo et al. showed the permeability reduction from 352 to 111.4 mD after CO_2 injection.¹¹¹ Figure 6 demonstrated the changes in

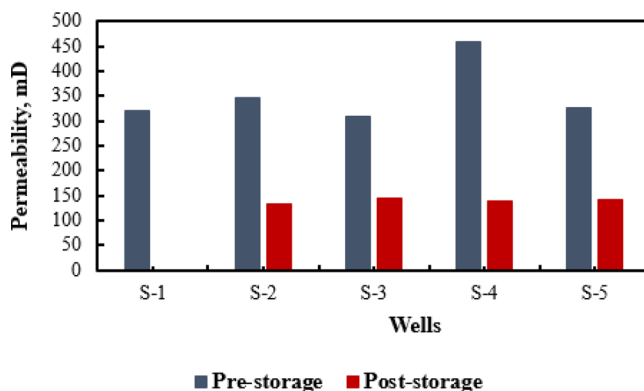


Figure 6. Illustration of the permeability decline before and after the experiment. This figure was reproduced with permission from ref 111. Copyright 2015 Elsevier.

permeability from five wells. However, Table 2 shows the summary of formation damage from the various studies undertaken in CO_2 storage in geological formations.

2.2. Dissolution/Precipitation of Minerals. The dissolution/precipitation of minerals during CO_2 injection and storage in a geological formation is an essential issue to be assessed as a result of the chemical weathering in the host storage formation. The active minerals are easily dissolved by the acidic solution,¹¹⁷ resulting in changes in the conditions of the formation rock, which affect the permeability and cause formation damage.¹²³ However, the dissolution or precipitation of active minerals during the injection and storage of CO_2 is mostly attributable to two mechanisms. The first mechanism is the reaction of CO_2 with brine water, which exchanges ions and modifies the brine water, resulting in dissolution.¹²⁴ The second is when impure CO_2 dissolves previously present minerals, resulting in precipitation.¹²⁵ These mechanisms cause variations in the pore volume, pore space,

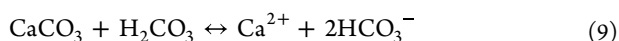
Table 2. Summary of Studies on Formation Damage during CO₂ Injection and Storage into the Geological Formations

focus point	rock formation	scope of the experiment	formation damage outcome	reference
CO ₂ -brine-rock interactions	sandstone saline aquifer	investigation of CO ₂ -brine-rock interactions to observe the dynamic and equilibrium interactions through experimental and numerical simulations using TOUGHREACT and PHREEQC	dissolution and precipitation of minerals were observed, which contributed to the formation damage; however, dolomite may dissolve very early, while another mineral may dissolve and precipitate for as long as 100 years	83 and 112
CO ₂ -brine-rock interaction	limestone formation	investigation of the effect of the CO ₂ -brine-rock interaction to analyze the cementation factor as well as saturated exponents; the experiment was conducted with and without CO ₂ using core samples from Indiana limestone formation	decrease in pore throat was observed from mercury injection capillary pressure (MICP) because new fractures were generated from the partial dissolution of grains and expansion of micro- and macropores, while small pores closed as a result of precipitation; in addition, the average permeability declined from 352 to 111.4 mD	67 and 111
CO ₂ -brine-rock interactions	sandstone formation	interaction of CO ₂ , brine water, and synthetic sandstone rock was analyzed through stainless-steel reactors; the experiment was observed at high temperatures and pressure	released clay materials as a result of the dissolution of carbonate cement facilitated the permeability decline by filling micropores and blocking the flow path of the fluid	113
CO ₂ -brine-rock interactions	reservoir carbonate	effect of permeability changes during CO ₂ -brine-rock interactions was investigated for oil recovery when CO ₂ was injected into the rock of the carbonate reservoir; however, the experiment observes the effects of the flow rate, pressure, and displacement type; different evaluation has been performed considering the water salinity and injection rate	long-term injection of CO ₂ into the carbonate rock may result in a significant decline of permeability, affect near-wellbore regions, and hence, can cause severe financial losses during the project	114
CO ₂ -H ₂ O injection and tapping mechanisms of residuals	sandstone reservoir	experiment involved the injection of supercritical CO ₂ and different cycles of water (over 6 cycles) using a high-flow rig near the wellbore zone, and the injection was performed to the specified storage rock formation (aquifer) in the North Sea, U.K.	effective permeability was observed to decrease as residual trapping was increased because it contributed to the multiple cycles of water injection; when the effective permeability was reduced, even injectivity was reduced and the effectiveness of the process of injection and storage become affected	115
impact of drying, precipitation of salt, and capillary pressure (part 2)	sandstones	study utilized numerical simulations for experimental investigations of the salt deposit, drying, and impact of capillary pressure during injection; the water, gas saturation, and salt precipitation analysis were observed from the sandstone cores, which correspond to the injection time	severe permeability decline was observed near the wellbore region as a result of the skin effect caused by salt precipitation (Figure 2)	116
impact of drying, precipitation of salt, and capillary pressure (part 1)	sandstone	study used dry gas (N ₂) and continuous injection to the sandstone cores to observe the physical properties that may affect water saturation in the rock formation	reduction of permeability was reported at the end of the experiment as a result of salt precipitation; decrease in permeability is from 74 to 22 mD (70%)	117
injection of CO ₂ into a carbonate formation	carbonate	experiment employed computed tomography (CT) to investigate the changes in permeability and porosity during the injection and storage of CO ₂ in a carbonate aquifer reservoir in Southeast Turkey; however, the experiment managed to model and analyze the effect in the near-wellbore zones, especially the flow conditions	it has been observed that the permeability was decreased as a result of blocking, bridging, or plugging of pore throats during the injection; this decrease in permeability is due to mineral precipitation	118 and 119
alteration of injectivity by the presence of salt precipitation	sandstone	experiment described the impact of evaporation and salt precipitation during the injection of supercritical CO ₂ into the sandstone rock; the effect of the injection rate and salinity of brine water was analyzed and presented	result has shown considerable formation damage, in which the permeability decreased against the change of the injection rate; the permeability decline was reported to be 43% for the injection rate of 20 cm ³ /min and 50% for the injection rate of 10 cm ³ /min, while the injection rate of 5 cm ³ /min showed a permeability decline to 62%; also, it has been reported that, when brine salinity decreased, the reduction of permeability and precipitation was high; the permeability decline was 21, 50, and 66% for brine salinity of 50, 100, and 200 g/L, respectively	109

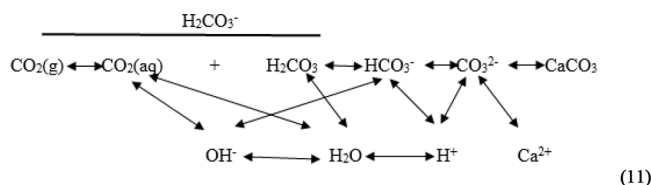
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and permeability, which can potentially restrict the route and lower the injectivity and storage capacity of the formation.¹²⁶

However, Yang et al.¹²⁷ reported that rock permeability in reservoir formation could be altered with a relatively small amount of dissolution of cementing material present. The experiment of Zou et al. demonstrated that the natural fractures and rock matrix have the potential to affect and change the behavior of hydraulic fracture growth as a result of the dissolution/precipitation of cementing materials, such as calcite minerals.¹²⁸ The chemical weathering in host storage formation can be described by the general dissolution reaction of calcite, which is reported by the research of Dreybrodt¹²⁹ and the experiment of Plummer et al. Similar to this, the dissolution/precipitation process is seen to consist of three primary phases that can all happen at once according to eqs 8–10.



In addition, the mechanisms of the dissolution/precipitation of calcite can further be described according to the CO_2 – H_2O system as presented by Usdowski¹³⁰ as the chemical reaction of the calcite surface when dissolution and precipitation happen (eq 11).



From the equation of calcite (eq 10) and the research of Usdowski, it can be observed that Ca^{2+} governs the reaction of H^+ , H_2CO_3 , and HCO_3^- , in which Ca^{2+} and H^+ participated in dissolution, while H_2CO_3 and HCO_3^- precipitated, as seen in Figure 7

In addition, other studies have reported the permeability and porosity alteration during CO_2 injection as a result of fine migration and dissolution of clay mineral reactions in sandstone formations.^{131–133} Furthermore, the experiment of Aminu et al. has shown that mineral weight fractions changed

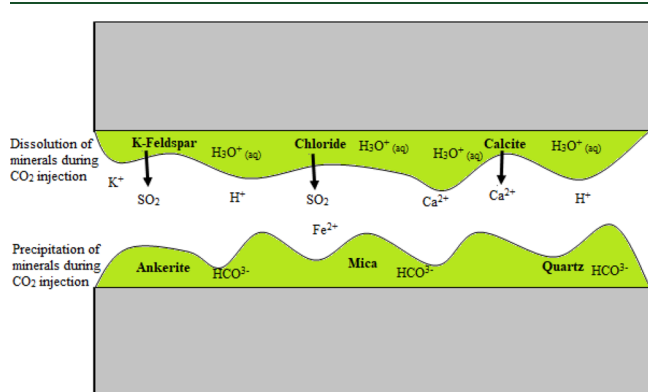
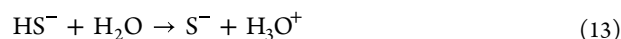
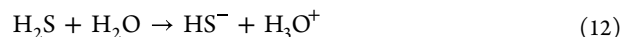
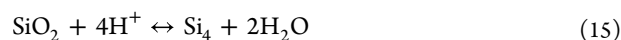
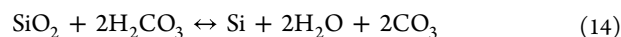


Figure 7. Description of the minerals at the Bunter sandstone formation and their dissolution and precipitation properties when CO_2 was injected into the sandstone formation. K^+ , Fe^{2+} , H^+ , and Ca^{2+} provided the reactants required during precipitation.

as a result of chemical reactions (dissolution) after CO_2 injection, Table 4. Calcite and halite disappeared during the reaction, and this is due to two facts: first, the salinity of brine water during formation (reaction) and, second, the stirring mechanisms that influenced them to disappear during the experiment. However, ankerite and quartz have shown great changes in weight fractions, as ankerite showed a substantial decrease of the weight fraction by 41.57%, while weight fractions of quartz increased by 11.44%. Also, changes can be observed in the presence of the test system of NO_2 , SO_2 , and H_2S samples. In the presence of NO_2 in the test system, ankerite and quartz minerals change from 10.4 to 15.8 μm (increase by 34.18%) and from 48.1 to 43.8 μm (decrease by 8.94%) by weight fraction, respectively. In addition, H_2S in the test system reduced the weight fraction of ankerite (8.9 μm) while increasing the weight fraction of quartz minerals (50.9 μm) as a result of the production of a weak acid and other ions in water.³⁵ However, H_2S can be slightly soluble and dissociate quickly, as explained in the following chemical equations (eqs 12 and 13):¹³⁴



Further, the dissolution and precipitation mechanisms can be observed when quartz reacted with acid, like carbonic acid, or hydrogen ions formed from water dissolution through CO_2 , which is also presented in Table 3.



The dissolution and precipitation of minerals have happened mainly as a result of attaining the equilibrium between the formation reservoir and injected CO_2 . The dissolution and precipitation degraded either the mineral matrix or the paths that CO_2 may take. Of these facts, the structural integrity of the formation is compromised,¹³⁵ especially in seal rock systems. Trapped CO_2 resulted in cations and anions released from the mineral reaction causing further mineral precipitation and weakening the caprock.¹³⁶

However, the research of Moghadasi et al. reported that the mineral precipitation is challenging in the porous system, where the permeability was observed to decline to 90%. The permeability decline can be attributed to the temperature change, injection period, flow rate, and solution composition.¹¹⁸ Yu et al. assessed mineral dissolution and precipitation by conducting experiments and numerical simulation. Similarly, different minerals were observed, such as K-feldspar, albite, quartz, calcite, kaolinite, and dolomite.¹³⁷ The result was presented in Figure 7, which shows that some minerals dissolved and others did not. However, dolomite and calcite¹²⁰ dissolved during the experiment, while quartz, albite, and K-feldspar were partially dissolved or not dissolved, like dolomite and calcite. However, Pearce et al.¹³⁸ used a core sample from the targeted zone to test the potential interactions between CO_2 , SO_2 , NO , and pure CO_2 during CO_2 storage. Throughout the experiment, the core sample was described and mineral changes, like calcite and feldspar, were observed. There have been reports of calcite minerals dissolving, and calcite minerals occupy the pore spaces in core samples, which could lower the permeability of the rock. On the other hand, quartz and kaolinite were observed to precipitate. Weibel et al.

Table 3. Summary of Major Dissolution and Precipitation Reactions

focal point	description of dissolution and precipitation
CO ₂ injection into the carbonate formations	<p>in the carbonate system, injected CO₂ or atmospheric CO₂ can dissolve in water, forming a weak acid (H₂CO₃), which can dissociate into HCO₃⁻ and CO₃²⁻ and form H⁺ ions, causing dissolution and precipitations,¹¹⁸ as observed in the following equations:</p> $\text{H}_2\text{O} + \text{CO}_2 + \text{CaCO}_3 \leftrightarrow \text{Ca}(\text{HCO}_3)_2$ $\text{CO}_2(\text{gas}) \leftrightarrow \text{CO}_2(\text{aq})$ $\text{CO}_2(\text{aq}) + \text{H}_2\text{O} \leftrightarrow \text{H}_2\text{CO}_3(\text{aq})$ $\text{H}_2\text{CO}_3(\text{aq}) \leftrightarrow \text{HCO}_3^- + \text{H}^+$ $\text{HCO}_3^- \leftrightarrow \text{CO}_3^{2-} + \text{H}^+$
precipitation of carbonate minerals	<p>carbonate minerals may precipitate through a bicarbonate reaction with divalent cations; Ca, Mg, and Fe carbonates may be observed¹¹⁹</p> $\text{HCO}_3^- + \text{Ca}^{2+} \leftrightarrow \text{CaCO}_3 + \text{H}^+$ $\text{HCO}_3^- + \text{Mg}^{2+} \leftrightarrow \text{MgCO}_3 + \text{H}^+$ $\text{HCO}_3^- + \text{Fe}^{2+} \leftrightarrow \text{FeCO}_3 + \text{H}^+$
dissolution and precipitation of minerals	<p>formation of acidic water</p> <p>acidic fluid with the calcite reaction and calcite dissolution</p> <p>acidic fluid with the dolomite reaction^{27,120}</p> $\text{CO}_2 + \text{H}_2\text{O} \rightarrow \text{H}^+ + \text{HCO}_3^-$ $\text{CaCO}_3 (\text{calcite}) + \text{H}^+ \rightarrow \text{Ca}^{2+} + \text{HCO}_3^-$ $\text{CaMg}(\text{CO}_3)_2 (\text{dolomite}) + 2\text{H}^+ \rightarrow \text{Ca}^{2+} + \text{Mg}^{2+} + 2\text{HCO}_3^-$ <p>acidic fluid with K-feldspar minerals; the precipitation from kaolinite restrains further reaction, causing K-feldspar dissolution to decline</p> $2\text{KAlSi}_3\text{O}_8 (\text{K-feldspar}) + 2\text{H}^+ + 9\text{H}_2\text{O}$ $\rightarrow \text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4 (\text{kaolinite}) + 2\text{K}^+ + 9\text{H}_4\text{SiO}_4(\text{aq})$ $\text{NaAlSi}_3\text{O}_8 (\text{albite}) + \text{CO}_2 + \text{H}_2\text{O}$ $\rightarrow \text{NaAlCO}_3(\text{OH})_2 (\text{dawsonite}) + 3\text{SiO}_2 (\text{chalcedony})$
dissolution of dolomite rock	<p>injection of CO₂ results in the dissolution of dolomite rock¹²¹</p> $\text{dolomite} + 2\text{H}^+ = \text{Ca}^{2+} + \text{Mg}^{2+} + 2\text{HCO}_3^-$
kaolinite precipitation	<p>precipitation of kaolinite was observed¹²¹</p> $\text{kaolinite} + 2\text{CO}_2(\text{g}) + 2\text{Na}^+ + \text{H}_2\text{O} = 2\text{dawsonite} + 2\text{quartz} + 2\text{H}^+$
fluid rock interactions	<p>dissolution of CO₂ to the carbonate rock</p> $\text{H}_2\text{O} + \text{CO}_2 = \text{H}_2\text{CO}_3 = \text{H}^+ + \text{HCO}_3^-$ <p>CO₂ and calcite reactions resulted in Ca²⁺ and HCO₃⁻ ions¹²²</p> $\text{H}_2\text{O} + \text{CO}_2 + \text{CaCO}_3 = \text{Ca}^{2+} + 2\text{HCO}_3^-$

Table 4. Unreacted and Reacted Mineral Samples from the Quantitative X-ray Diffraction (QXRD) Analysis of Mineral Phases (wt %)³⁵

carrier gas	impurity	albite	analcime	ankerite	calcite	chlorite	halite	hematite	K-feldspar	mica	quartz
CO ₂	SO ₂	18.5	4.0	15.6	<0.5	1.7		0.5	8.5	6.6	44.5
CO ₂	NO ₂	18.7	3.7	15.8		1.3		0.6	8.8	7.3	43.8
CO ₂	H ₂ S	20.0	3.5	8.9		1.1		<0.5	9.1	6.3	50.9
CO ₂	none	19.5	3.6	10.4		1.1	1.1	0.5	9.2	7.6	48.1

reported that precipitation of minerals, especially carbonate minerals, is usually common during CO₂ injection and storage.¹³⁹ Other experiments agreed that the precipitation of carbonate rock is inevitable during the CO₂ reaction.^{140,141}

Usually, the permeability reduction is accounted for as a result of the presence of dissolution during CO₂ injection, which releases kaolinite, clay materials, and solid-phase minerals, which plug and block the fractures and pore throat of the reservoir formation.¹³⁷ Shiraki and Dunn have reported similar results that kaolinite minerals were the main challenge to reduce the permeability after CO₂ injection.¹²³ However, the research of Luquot et al. showed that the permeability was reduced as a result of the new minerals formed during CO₂ storage.¹⁰⁷

Moreover, other studies reported the dissolution and precipitation process of the saline aquifer.^{142–145} The research

by Zhu et al. described the dissolution process during CO₂ injection into the saline aquifer. When CO₂ is injected into the saline aquifer, partial dissolution will happen to the active minerals, resulting in a pH decrease as a result of the formation of the acidic condition. During this time, the precipitation of carbon minerals may also happen, which can affect the reservoir performance or formation damage, resulting in low permeability or low integrity of the formation rock. However, the precipitation and dissolution during CO₂ injection and storage depend upon various factors, such as the formation type, mineral composition, pressure, and temperature.^{121,146} However, the acid formed in the geological formation as a result of CO₂ injection and storage has been researched by numerous researchers with agreement that precipitation and dissolution might compromise the integrity of rock formation and also cause formation damage.^{147,148} The experiment by

Cui et al. investigated the dissolution and precipitation of two formation types: sandstone and carbonate rocks. The results showed that, for the sandstone, the dissolution was observed in clay and ankerite minerals, while precipitation was observed in the plagioclase minerals, which may increase ions of Ca^{2+} and Mg^{2+} in brine water. However, considering the carbonate rock, the dissolution was seen in dolomite minerals, while the precipitation was observed in the calcite and ankerite minerals.¹⁴⁹

Various minerals were investigated by Liu et al. during CO_2 injection, and observation was performed on the chemical reaction between CO_2 , rock, and brine water. The minerals utilized in the experiment were kaolinite, quartz, Ca-montmorillonite, calcite, albite, K-feldspar, dolomite, anhydrite, illite, chlorite, and pyrite. Table 5 shows the results of the dissolution and precipitation of these minerals after CO_2 injection.⁸³

Table 5. Results of the Dissolution and Precipitation of Utilized Minerals in the Experiment⁸³

precipitated minerals	dissolved minerals	main ions	unchanged minerals
dolomite	albite	HCO_3^-	anhydrite
kaolinite	anhydrite	K^+	pyrite
quartz	calcite	Na^+	
Ca-montmorillonite	Ca-montmorillonite	Ca^{2+}	
illite	K-feldspar	Mg^{2+}	
	chlorite		

The change in mineral concentrations considerably affects the porous or fracture systems, which may cause formation damage. However, the concentrations of HCO_3^- , Mg^{2+} , K^+ , SO_4 , Na^+ , and Ca^{2+} were assessed, and the following is the discussion based on the results shown in Table 4. Because mineral dissolution may occur during CO_2 injection,¹⁵⁰ like dissolution of dolomite, K-feldspar and albite, little concentrations of Mg^{2+} , Na^+ , and K^+ were observed, possibly from

dissolution of a small amount of dolomite, K-feldspar, and albite from sandstone formation.¹⁵¹ Because sandstone contains calcite minerals, a large amount of HCO_3^- and Ca^{2+} was observed, which was believed from the dissolution of calcite minerals. Furthermore, the experiment shows that the pressure increase has little increase in the ion concentrations of Mg^{2+} and Na^+ , while SO_4 and K^+ remain constant and Ca^{2+} and HCO_3^- were observed to increase. Therefore, increased pressure led some minerals to dissolve, while some remained constant.¹⁵¹ Additionally, the concentration of Na^+ was observed to decrease when the NaCl concentration was increased as a result of the dissolution of secondary minerals,¹⁵² which contained Na^+ during the reactions.⁸³

Figure 8 shows the degree of mineral dissolution during CO_2 injection to the sandstone formation. However, albite dissolved more during the interaction with CO_2 and brine water than feldspar as a result of the low entropy and Gibbs free energy change, which is negative, while feldspar has higher entropy and Gibbs free energy. Therefore, albite spent lower energy to dissolve than feldspar. Table 3 presents the summary of the dissolution and precipitation reactions that occurred from the various experimental studies.

2.3. Changes in the Grain Size and Sorting. Mineral dissolution and precipitation led to changes in grain diameters and distributions in geological formation, resulting in a reduction of injectivity and storage capacity of CO_2 by holding grains together.¹¹⁹ Badrouchi et al. experimented to investigate the pore size distribution before and after CO_2 injection using two types of formation samples [Middle Bakken (MB) and two Three Forks (TF)].⁶⁷ They employed the nuclear magnetic resonance (NMR) to provide detailed results and concluded that the effective porosity decreased from 5.3 to 3.8% for MB samples, while for TF samples, the effective porosity was reduced from 7.6 to 6.3%, and the distribution of the pore size was presented in Figure 9 for detailed analysis. However, Figure 9 suggests that the pore distributions from all wells have shown a considerable decline in their distribution to the formation after injection as a result of dissolved CO_2 to the

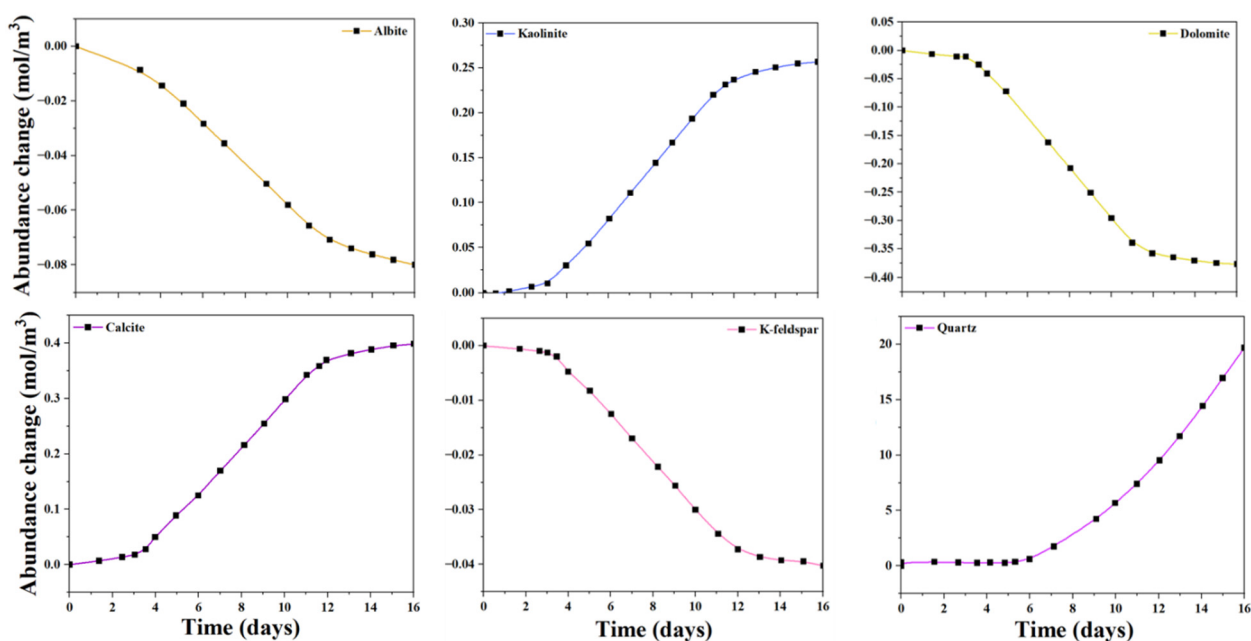


Figure 8. Change of minerals against time. This figure was reproduced with permission from ref 105. Copyright 2012 Elsevier.

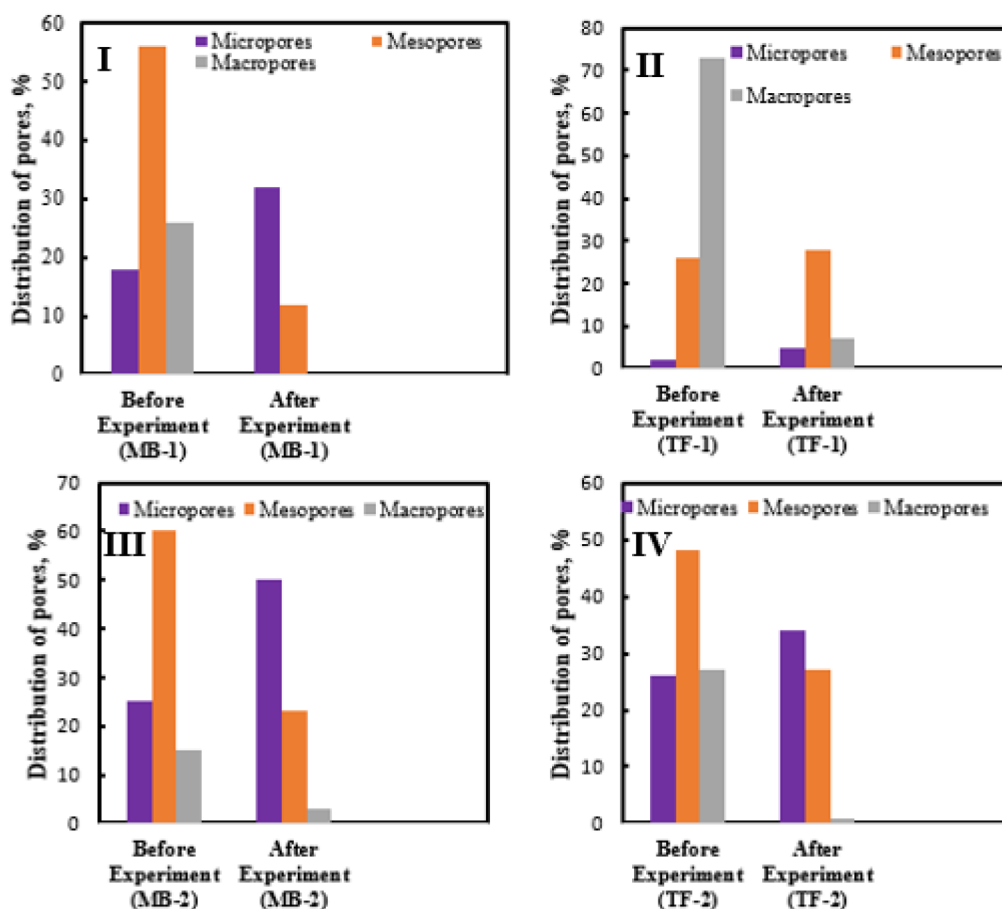


Figure 9. Distribution of pore percentages before and after the experiment. This figure was reproduced with permission from ref 67. Copyright 2022 Elsevier.

formation rock with active minerals, in which formation damage was experienced.

In this way, micro-, meso-, and macropores were blocked by mineral precipitation, leading to low injectivity by plugging and blocking micropores. Also, Adebayo et al. reported that, when CO₂ was injected, there was an alteration in grain size as a result of the chemical reaction, which affected the rock grains,¹¹¹ and the results are presented in Figure 10. However, the grain size change contributed to the solubility of grains and the development of new pores or the formation of macropores as a result of the joining of micro- and macropores.⁶⁷ Furthermore, the change in the grain size can be observed as a result of the closure of micropores or pore throats through precipitation. The pore distributions have considerable effects on permeability and porosity.¹⁵³ However, the research concerning the change of porosity and permeability before and after the experiment has been reported by other researchers.¹⁵⁴

The experiment of Zhao et al. showed that pore structures were affected during CO₂ injection, in which the precipitation of secondary minerals changed the sorting behavior of the rock formation, especially the pore throats, causing the permeability to decline.⁹⁸ The change in pore throats is because of the strong precipitation of carbonate minerals, feldspar and quartz.^{35,73,143,154–156} The results of the experiment of Zhao et al. are presented in Figure 11.

Figure 11 shows the pore throat diameter before and after the experiment. However, 0.01–0.1 μm is a small throat, while

0.44–1 μm is a relatively larger throat that decreased. The decrease was attributed to the precipitation of the active minerals, which block the small throats (0.01–0.1 μm), while the large throats decreased as a result of the shrinking of the diameter after the precipitation of the secondary minerals.¹⁵¹

The experimental studies reported by Aminu et al. also showed the average grain size and distribution changes in four samples in a test system that involved pure CO₂, NO₂, SO₂, and H₂S in the test system.³⁵ Each set of sample measurements is reported in Feret diameter, *D* (mm), using the geometric mean equations to calculate the geometric mean, *d_g* (mm), of the samples,^{157,158} and the results are plotted in Figure 12. The formation damage as a result of dissolution and mineral precipitation considerably affects the grain size and distribution within the formation. Considering the experiment,³⁵ the following can be discussed in Figure 12a: exposing CO₂ to the test system caused the average grain size to increase (geometric mean, *d_g*) from 42.6 to 48.1 μm as a result of the formation of carbonic acid and the decrease of the pH values. However, because the reaction resulted in weak acid (carbonic acid), partial dissolution of solid minerals led to increased permeability by CO₂.

In Figure 12b, the experiment has shown a considerable increase in the average grain size from 43.8 to 48.1 μm with a higher chance of pH from 7.65 to 5.23. This increase is associated with the decrease of permeability as a result of the formation of a strong acid, like nitric acid, which increases the

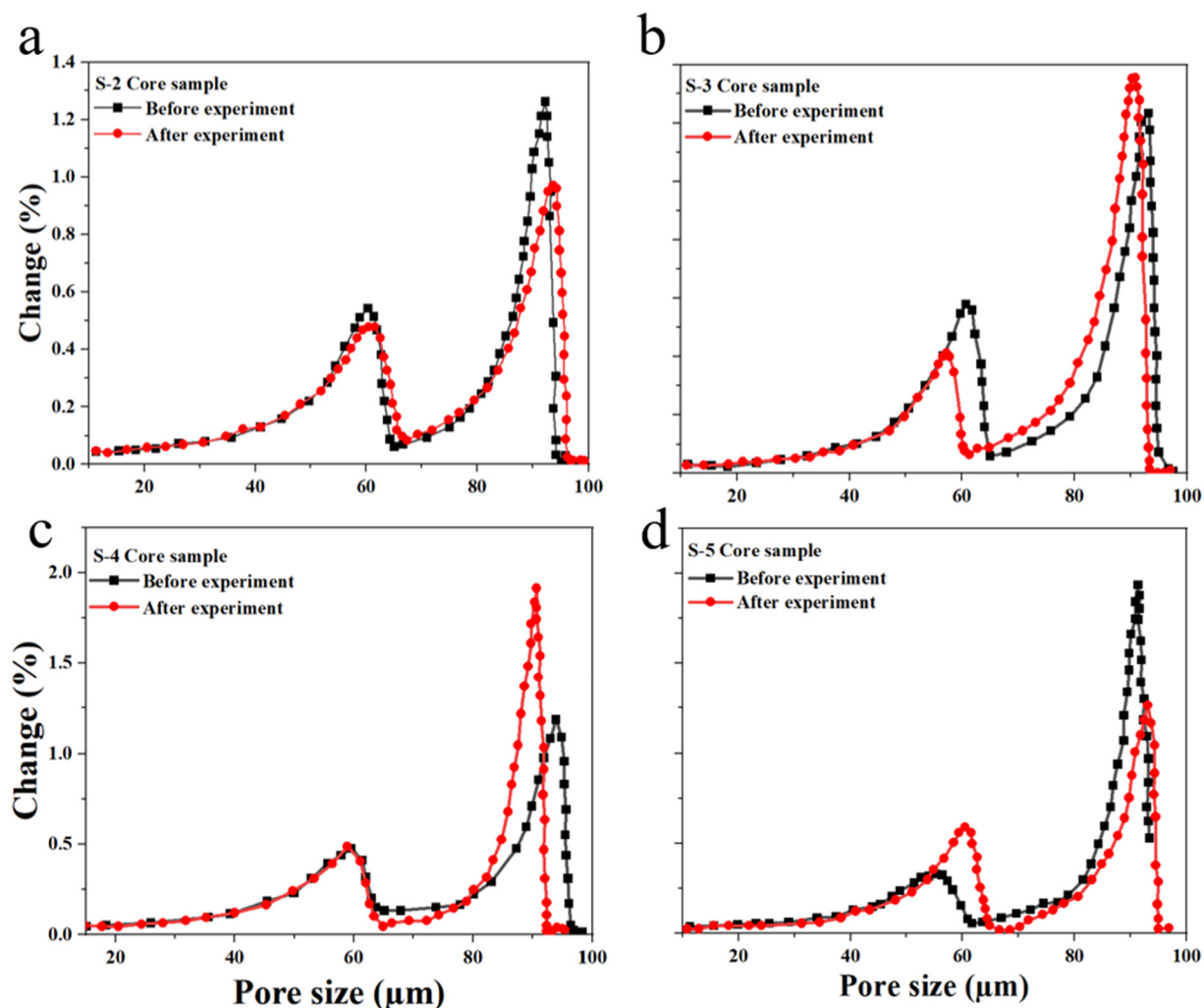


Figure 10. Pore size distribution of the core samples before and after the experiment. This figure was reproduced with permission from ref 111. Copyright 2015 Elsevier.

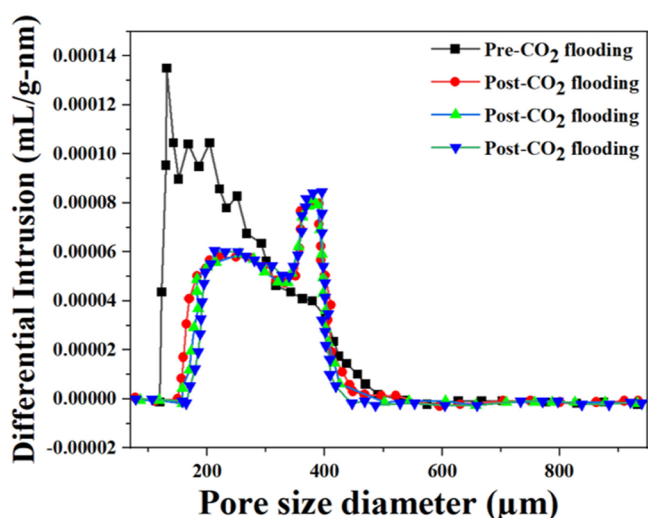


Figure 11. Pore throat distribution before and after the experiment. This figure was reproduced with permission from ref 151. Copyright 2015 Elsevier.

dissolution of minerals, leading to plugging and blocking the porous system, causing the formation damage.³³

In Figure 12c, when CO₂ and SO₂ were introduced, the average grain size was observed to change from 44.5 to 48.1 μm, as the same changes when CO₂ and NO₂ were conducted. However, there was a variation in pH and permeability. The pH value shows the change from 7.65 to 5.86 as a result of the acidic medium attributed to strong acid, known as sulfuric acid, while permeability declined as a result of the dissolution and precipitation, resulting in the blockage of the porous media. The last test exposed CO₂ and H₂S to the test system in Figure 12d, and the following was observed: the average grain size decreased from 50.9 to 48.1 μm with changes in pH values from 7.65 to 6.55. However, this is attributed to the acidic medium and dissolution of minerals.³⁵

3. CHALLENGES AND PERSPECTIVES

Formation damage can be minimized, leading to the injection and storage of CO₂ being effective for geological formations. However, laboratory experiments discussed in this study suggested some changes, especially in mineralogical and chemical reactions. These changes can affect the permeability and porosity of the formation during CO₂ injection and storage, resulting in complications of formation damage. Therefore, the following are challenges and perspectives suggested from this study: (1) Numerous experiments have

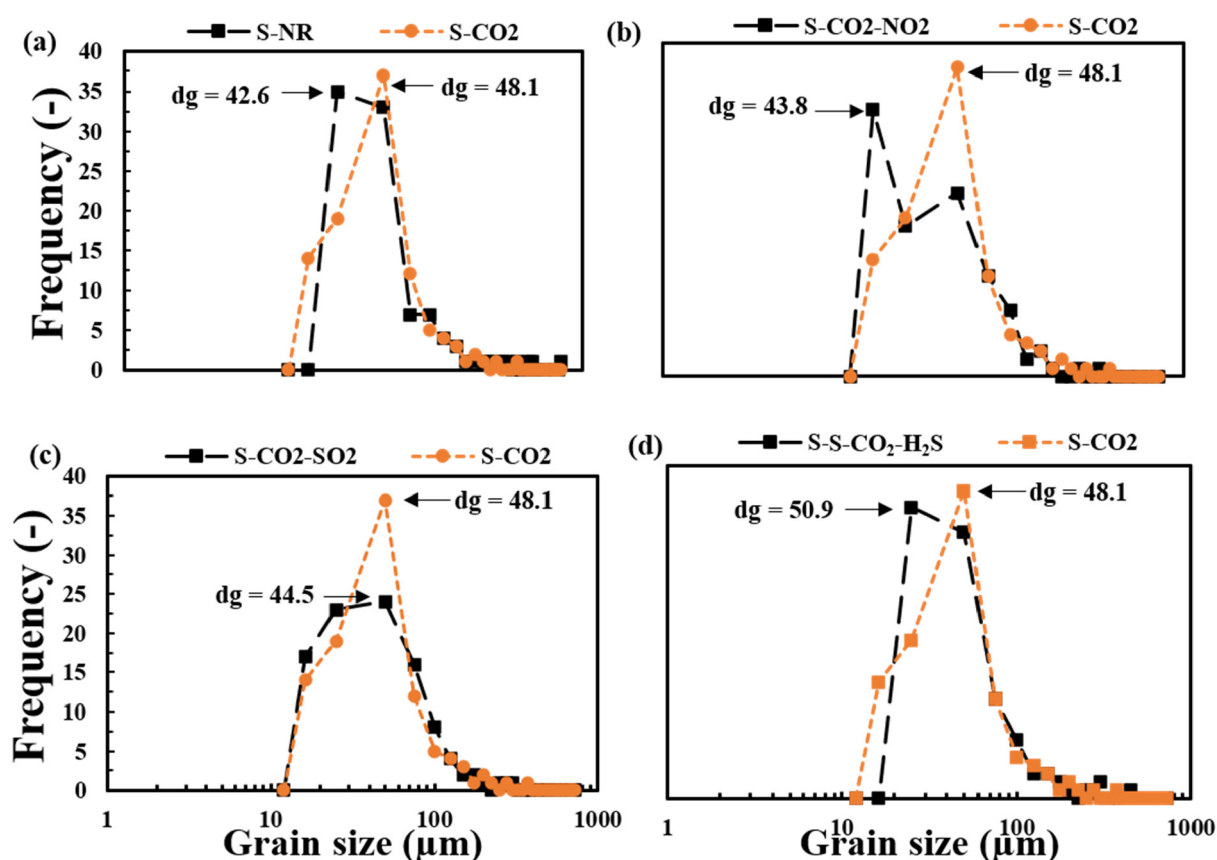


Figure 12. Changes in the average grain sizes and distributions of four samples in a test system: (a) unreacted and pure CO₂, (b) NO₂ with pure CO₂, (c) SO₂ with pure CO₂, and (d) H₂S with pure CO₂. This figure was reproduced with permission from ref 35. Copyright 2018 Elsevier.

shown the short-term effect of formation damage on geological formations, while CO₂ storage is a long-term project. More research should be conducted to determine the geochemical effect after long-term storage to determine the effect of formation damage.³⁵ (2) The extent of formation damage depends upon the type, conditions, and location of the geological formation. However, the process of mineralogical and chemical reactions can be either beneficial or sometimes very harmful. Therefore, this study recommends more research before the injection and storage of CO₂ because CO₂ is stored in the form of gas, and it is very easy to escape when leakage happens as a result of the buoyancy effect of CO₂.¹⁵⁹ To avoid escaping CO₂ to the surface, a detailed study should be performed. (3) How long CO₂ stays in the formation and its monitoring are another challenge to be addressed by researchers. The study of fault sealing capacity and petrophysical fault regions are very important. (4) The challenges of geological heterogeneity in the injection and storage of CO₂ are an important factor in the prediction of the formation damage in the reservoir because the porous medium may vary and cause inconsistencies.¹⁶⁰ (5) The study suggests that determining the initial composition of brine water is crucial because the changes of mineralogy, permeability, and porosity are generally characterized when there are reactions between them; for example, dissolution of sulfate minerals causes alteration of reservoir porosity.¹⁴⁶ More studies are recommended to have details of brine water composition.

4. CONCLUSION

This study presented and demonstrated the substantial previous and ongoing laboratory works in a wide range of potential formation damage during CO₂ injection and storage in a geological formation. The results of this review study conclude the following: The reaction between water, rock, and carbon dioxide impurities caused the formation damage. The extent of formation damage depends upon the kind of impurities contained in the stream of CO₂. It is necessary to evaluate the nature of impurities and the quality of the reservoir before the injection of CO₂. Also, it has been seen that NO₂ and SO₂ in CO₂ streams have the highest impact on the formation, which compromises the storage and injectivity efficiencies and safety of projects and may add the cost of storage, transport, and integrity of the project. When the dissolution and precipitation occurred, the mineral components shifted and plugged the porous media. Also, the study has shown that the grain sizes have been reduced on average from 50.9 to 48.1, resulting in a reduction of the permeability that leads to the occurrence of formation damage. It should be noted that storing CO₂ in a geological formation may reduce the possibility of formation damage. This study can provide knowledge that will assist in minimizing the uncertainty of storage of CO₂ in any new geological formation.

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NOMENCLATURE

ESEM = environmental scanning electron microscopy
QXRD = quantitative X-ray diffraction
SDD = silicon drift X-ray detector
XRD = X-ray diffraction
MICP = mercury injection capillary pressure
CT = computed tomography

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