

Article

Geochemical Effects on Storage Gases and Reservoir Rock during Underground Hydrogen Storage: A Depleted North Sea Oil Reservoir Case Study

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Abstract: In this work, geochemical modelling using PhreeqC was carried out to evaluate the effects of geochemical reactions on the performance of underground hydrogen storage (UHS). Equilibrium, exchange, and mineral reactions were considered in the model. Moreover, reaction kinetics were considered to evaluate the geochemical effect on underground hydrogen storage over an extended period of 30 years. The developed model was first validated against experimental data adopted from the published literature by comparing the modelling and literature values of H₂ and CO₂ solubility in water at varying conditions. Furthermore, the effects of pressure, temperature, salinity, and CO₂% on the H₂ and CO₂ inventory and rock properties in a typical sandstone reservoir were evaluated over 30 years. Results show that H₂ loss over 30 years is negligible (maximum 2%) through the studied range of conditions. The relative loss of CO₂ is much more pronounced compared to H₂ gas, with losses of up to 72%. Therefore, the role of CO₂ as a cushion gas will be affected by the CO₂ gas losses as time passes. Hence, remedial CO₂ gas injections should be considered to maintain the reservoir pressure throughout the injection and withdrawal processes. Moreover, the relative volume of CO₂ increases with the increase in temperature and decrease in pressure. Furthermore, the reservoir rock properties, porosity, and permeability, are affected by the underground hydrogen storage process and, more specifically, by the presence of CO₂ gas. CO₂ dissolves carbonate minerals inside the reservoir rock, causing an increase in the rock's porosity and permeability. Consequently, the rock's gas storage capacity and flow properties are enhanced.

Keywords: underground hydrogen storage; geochemical modelling; energy transition

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1. Background

The current energy transition is defined as the process of moving from conventional fossil fuels to sustainable and renewable energy resources, such as solar and wind energies. This energy transition is motivated by different factors, the most important of which are climate change and energy sustainability. It is expected that hydrogen energy will play a vital role in this transition [1–4].

Hydrogen is a clean burning fuel that can be used to generate energy for vehicles, heating, and electricity. One of the best characteristics of using hydrogen as a fuel is that when it is burnt, it does not generate any carbon dioxide, and the only by-product is water vapour. Hydrogen can be produced through steam–methane reforming, which is the most common hydrogen production technique. This is carried out by reacting natural gas with steam to produce hydrogen and carbon dioxide. However, this technique is not sustainable as it requires natural gas to generate hydrogen. Another technique to produce hydrogen is electrolysis, in which electricity is used to separate hydrogen and oxygen molecules in water. This technique can be powered by the excess energy produced from renewable energy sources, which makes it more sustainable compared to steam–methane reforming [5–7].

To achieve a sustainable future, the deployment of a hydrogen economy is essential, and for that, efficient and cost-effective hydrogen storage solutions are necessary. The hydrogen gas produced from the techniques mentioned above can be stored during high-supply periods and reused during high-demand periods, either above-ground or underground. However, above-ground hydrogen storage in vessels is limited in capacity and safety, making underground hydrogen storage an attractive alternative. Large-scale hydrogen storage is possible with underground storage in aquifers, salt caverns, and depleted hydrocarbon reservoirs, making it a feasible option for supporting widespread hydrogen adoption as an energy source [8–10]. Additionally, underground storage mitigates the risk of hydrogen gas leaks, reducing associated risks compared to above-ground storage options.

The use of appropriate geological formations, such as aquifers, salt caverns, or depleted natural gas or oil reservoirs, can enable cost-effective storage of hydrogen. However, UHS techniques have some challenges as experience in this technology is scarce [9,11]. In addition to geological, technical, and economic difficulties, legal and societal interplay contributes to its overall success [12,13]. The availability of suitable geological sites is crucial to the feasibility of underground storage. Furthermore, information on the depth and storage capacity of the potential site is vital as it can affect the UHS operation [10]. The understanding of the fluid–fluid and solid–fluid interactions inside the storage formation helps to plan for an efficient UHS process. Seismic activities are also an important safety concern relating to UHS because they could compromise the integrity of storage structures [10]. Economically, the efficient and effective application of underground hydrogen storage is believed to be able to unlock hydrogen energy’s competitiveness in the energy market in the near future [14]. Therefore, it is essential to ensure all technical, economic, and legal aspects are put into perspective for an effective adaptation of UHS operations.

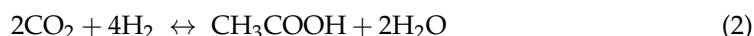
Several research studies have been carried out to investigate the technical aspects of storing hydrogen gas underground. These aspects include hydrodynamics, geochemistry and geomechanics. Hydrodynamics in underground hydrogen storage (UHS) is concerned with studying how the hydrogen gas and fluids flow inside the porous media within the underground storage site. It also studies the geological formation’s storage capacity of hydrogen gas during its injection and its recovery efficiency during its withdrawal [8,15,16]. Several parameters can affect this hydrodynamic aspect, which can be categorized into two categories: reservoir-related parameters, and operational parameters. The reservoir-related parameters can be reservoir porosity, permeability, and pressure, among other parameters. As for the operational parameters, hydrogen gas injection and withdrawal rates, storage cycles, and several operation wells affect the underground hydrogen storage operation [17]. The injection of CO₂ in the subsurface geoformations has been practised in various forms, such as CO₂-enhanced oil and gas recovery [18] and CO₂ sequestration as gas hydrates, alone or combined with methane–CO₂ replacement, in natural gas or methane hydrate reservoirs [19,20] in an effort to meet the decarbonization targets of the leading industrial sectors. In our scheme, CO₂ injections also contribute, to some extent, to these efforts while working as a cushion gas.

During UHS, geochemical interactions involving the hydrogen gas that has been stored, cushion gas, reservoir fluids, and reservoir minerals may take place [21]. Due to the conversion of hydrogen to other gases, such as methane and hydride sulphide, these processes may result in losses in the stored hydrogen. Additionally, due to interaction with other gases in the reservoir, geochemical processes have the potential to reduce the purity of hydrogen gas [22,23]. A change in the porosity and permeability of the geological structure as a result of mineral dissolution or precipitation is another significant potential consequence of the geochemical activity of hydrogen gas during UHS. As a result, the geomechanical characteristics of the geological structure can change, increasing the likelihood of structural collapse or faulting and decreasing the integrity of the cap rock [10,24]. Abiotic or biotic processes might result in geochemical reactions during UHS. Abiotic processes take place when hydrogen interacts with non-living objects such as rock minerals, oil, gas, and brine. Aqueous reactions and mineral dissolution/precipitation

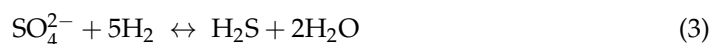
reactions are examples of abiotic processes [21,24]. One of the important factors that must be taken into account during UHS is hydrogen biotic reactions brought by microbial activity. The quantity and kind of live microorganisms (such as bacteria) present within the storage location controls these responses. Methanogenesis, acetogenesis, sulphate reduction, and iron reduction are some of the known biotic activities that use hydrogen. Methanogenesis is the conversion of hydrogen to methane and water when reacting with CO₂ in the presence of methanogens. It is also known as the Sabatier reaction and is as follows [25]



Acetogenesis is another type of biotic reaction that takes place between H₂ and CO₂ in the presence of acetogen bacteria. This reaction produces acetic acid and water as follows [26]



When sulphate-reducing bacteria are present, the sulphate reduction processes convert H₂ and sulphate to hydrogen sulphide (H₂S). Although they have been observed to be active in temperatures as high as 110 °C, sulphate-reducing bacteria are most active when the temperature is approximately 38 °C [27–29]. It is anticipated, however, that in acidic aqueous and depleted hydrocarbon reserves, particularly in the presence of high salinity brine, its action will be slower [30].



Amid et al. [31] and Hemme et al. [24] investigated the effects of geochemical and biochemical reactions on hydrogen storage in depleted natural gas and sandstone reservoirs, respectively. Their results suggested that sulphate reduction poses a greater challenge to the UHS process than methanation, but the overall hydrogen loss due to dissolution was less than 0.1%. The modelled UHS case was found to be capable of providing 42% of the energy capacity supply compared to the Rough Gas Storage Facility (UK), indicating that UHS can be a viable option for energy storage.

Laban [32], Bo et al. [22], and Zeng et al. [33,34], also studied the effects of various factors, such as temperature, pressure, mineralogy, and brine properties, on hydrogen storage and loss. Laban [32] found that bacteria growth rate, sulphate concentration, and cavern pressure and temperature are the most influential factors on hydrogen sulphide production through sulphate-reducing bacteria. Bo et al. [22] suggested that sandstone reservoirs are better for hydrogen storage than carbonate reservoirs because the water uptake of hydrogen does not react with quartz and clay minerals. Zeng et al. [34] studied hydrogen loss, mineral dissolution, and water chemistry over a long period of time and found that the Majiagou formation is a good candidate for UHS. Zeng et al. [33] evaluated the effects of various reservoir parameters on the wettability of a hydrogen–brine–calcite system and found that increasing the temperature and decreasing the salinity increase the disjoining pressure within the system, resulting in higher hydrogen gas repulsion from the calcite surface and increasing water wettability.

The results of Amid et al. [31] and Hemme et al. [24] suggested that hydrogen loss due to geochemical reactions and diffusion during UHS is negligible; however, Bo et al. [22] and Zeng et al. [34] found that hydrogen loss can be significant, up to 9.5% and 81.1%, respectively, depending on the storage period and reservoir mineralogy and conditions. The difference in predicted hydrogen losses might be a result of considering hydrogen-redox-coupled pair reactions, which accelerate hydrogen solubility and loss, as is the case in the work of Bo et al. [22] and Zeng et al. [34], in contrast to Amid et al. [31] and Hemme et al. [24]. The presence of calcite in the geological structure was found to lead to changes in the rock's porosity and permeability, affecting the reservoir's storage capacity and flow properties. Furthermore, hydrogen loss due to geochemical reactions and diffusion is directly related to both pressure

and temperature, where an increase in pressure and temperature causes a rise in the hydrogen lost during UHS [24].

In this study, we investigate the effects of geochemical activities during underground hydrogen storage in a depleted North Sea oil reservoir on the stored hydrogen gas as well as the reservoir rock properties. The impact of the geochemical reactions on both reservoir's porosity and permeability is evaluated. These evaluations will be carried out under varying pressures, temperatures and salinities to reflect potential conditional changes during the underground hydrogen storage in the targeted sandstone formation in a depleted North Sea oil reservoir. Outcomes from this study should provide valuable information for understanding the geochemical effects of underground hydrogen storage in a depleted oil reservoir to be considered during the design process.

2. Methodology

The investigations in this study are carried out using the geochemical software PhreeqC [35]. It has been previously used to quantify geochemical reactions inside oil reservoirs for different enhanced oil recovery applications [36–39]. The developed model considers equilibrium reactions, mineral dissolution/precipitation reactions, and exchange reactions. Equilibrium and exchange reactions are assumed to be fast and instant reactions and are not affected by the time of H₂ and CO₂ storage. The mineral dissolution and precipitation reactions are dependent on reaction kinetics and the speed of reaction. These reactions usually occur over a prolonged period time and result in changes in both the mineral and aqueous phases over time. Methanisation and sulphate reduction reactions are both biotic reactions, which are dictated by certain types of bacteria. The methanisation bacteria was reported to be active at temperatures up to 122 °C and salinities up to 3.4 mol/L. Sulphate-reducing bacteria can operate in temperatures up to 113 °C and 1.7 M NaCl [30]. These limiting values of temperature and salinity for both methanisation and sulphate reduction reactions are close to the conditions encountered inside the targeted depleted oil reservoirs. Hence, both methanisation and sulphate reduction reactions are expected to have minimal effects on the underground hydrogen storage process and are, therefore, ignored in our modelling. The mineral reactions adopted in this work are detailed in Table 1.

Table 1. Adopted mineral geochemical reactions in the model [24].

Mineral Phases Reaction	Log K @ 25 °C
$KAlSi_3O_8 + 8H_2O = K^+ + Al(OH)_4^- + 3H_4SiO_4$	−20.573
$NaAlSi_3O_8 + 8H_2O = Na^+ + Al(OH)_4^- + 3H_4SiO_4$	−18
$Al_2Si_2O_5(OH)_4 + 6H^+ = H_2O + 2H_4SiO_4 + 2Al^{3+}$	7.435
$SiO_2 + 2H_2O = H_4SiO_4$	3.95
$CaCO_3 = CO_3^{2-} + Ca^{2+}$	8.48
$FeS_2 + 2H^+ + 2e^- = Fe^{2+} + 2HS^-$	−18.479
$K_{0.6}Mg_{0.25}Al_{2.3}Si_{3.5}O_{10}(OH)_2 + 11.2H_2O$ $= 0.6K^+ + 0.25Mg^{2+} + 2.3Al(OH)_4^- + 3.5H_4SiO_4$ $+ 1.2H^+$	−40.267
$CaMg(CO_3)_2 = Ca^{2+} + Mg^{2+} + 2CO_3^{2-}$	−17.09

Mineral dissolution and precipitation reactions cause changes in the mineral phases present inside the reservoir and accordingly affect the overall rock porosity and permeability [10]. Changes in porosity as a result of rock dissolution and precipitation are determined using solid and pore volume calculations. Porosity of the rock \varnothing , can be calculated as follows:

$$\varnothing = \frac{V_p}{V_s + V_p} \quad (4)$$

where V_p is the pore volume, and V_s is the solid (rock) volume. The changes in porosity arise as a result of the rock dissolution and precipitation due to mineral reactions as modelled in PhreeqC. The porosity change affects the reservoir's storage capacity of the reservoir rock. These changes in porosity consequently alter the permeability of the reservoir rock influencing the rock's flow properties, and the gas injection and withdrawal process. The changes in permeability are determined using the widely used Carman–Kozeny equation as a function of the reservoir's porosity:

$$k_x = \frac{d_p^2 \varnothing^3}{300(1 - \varnothing)^2} \quad (5)$$

where k_x is the permeability (cm), \varnothing is the porosity, and d_p is the average diameter of sand grains (cm). Although the Carman–Kozeny equation assumes a uniform and isotropic pore geometry, we use the equation to quantify the change in permeability as a function of the original and altered porosity in the form of equation (6):

$$\Delta k\% = \frac{k_2 - k_1}{k_1} = 100 \times \left(\frac{\varnothing_2^3}{(1 - \varnothing_2)^2} \times \frac{(1 - \varnothing_1)^2}{\varnothing_1^3} - 1 \right) \% \quad (6)$$

where k_1 and k_2 are the original and altered permeability (cm), respectively. \varnothing_1 and \varnothing_2 are the original and altered porosity, respectively.

2.1. Base Scenario

The simulated scenario is based on storing hydrogen gas in a depleted North Sea oil reservoir and, more specifically, in the Etive sandstone formation. Hydrogen gas is assumed to be stored with 95% of the reservoir's working capacity and the remaining 5% is filled with CO₂ as a cushion gas. The mineralogy of Etive formation plays an important role in dictating the geochemical reactions that take place during the UHS process and is detailed in Table 2. The brine salinity from the formation is shown in Table 3. In this work, we investigate the geochemical effects during underground hydrogen storage in the depleted oil reservoir on the storage gases and reservoir rock properties over 30 years. The reservoir was initially at 400 atm and 100 °C temperature. However, after depletion, the reservoir pressure dropped to 135 atm.

Table 2. Reservoir rock mineralogy [40].

Mineral	Weight%
Quartz	78
Feldspar	11
Muscovite	2
Carbonate	1
Pyrite	2
Kaolinite + Illite	5

Table 3. Formation water chemistry.

Ion	Concentration (mg/l)
Si ²⁺	46
Ba ²⁺	67
Cl ⁻	14,740
SO ₄ ²⁻	18

Table 3. *Cont.*

Ion	Concentration (mg/l)
Mg ²⁺	62
Ca ²⁺	290
K ⁺	190
Na ⁺	9210
Fe ³⁺	<0.10

2.2. Sensitivity Analysis

The sandstone reservoir is assumed to have an initial pressure around 400 atm, 100 °C, and 0.5 M NaCl salinity. These conditions represent the base case values of the carried-out analysis. The sensitivity range of pressure is between 100 atm, just below the current depleted reservoir pressure, and 500 atm, the formation's fracture pressure. The temperature range is between 80 and 120 °C to reflect any possible temperature changes inside the reservoir due to gas injection and production. The current formation's water salinity is approximately 0.5 M, which is also the minimum in the salinity range in the sensitivity analysis. The maximum investigated salinity was 5 M. In this study, the effects of possibly varying conditions, i.e., temperature, pressure, salinity, and CO₂% as a cushion gas on H₂ and CO₂ solubility, and rock properties (porosity and permeability) over 30 years. A summary of the base case values and range of parameters investigated are detailed in Table 4.

Table 4. Range of parameters in geochemical investigations.

Parameter	Base Case	Minimum	Maximum
Pressure (atm)	400	100	500
Temperature (°C)	100	80	120
Salinity (M NaCl)	0.5	0.5	5

3. Results and Discussion

3.1. Model Validation

The developed model was validated using the H₂ and CO₂ solubility in water predicted from the model against experimental values reported in the literature. The validation was performed using gas solubilities at a varying range of conditions, i.e., 0–200 MPa pressure, 50–100 °C temperature, and 0–5 M NaCl salinity. The model validation is limited to gas solubility in bulk water. The gases solubility in water inside porous media is affected by the presence of rock minerals because geochemical reactions affect gas solubility.

The model-predicted H₂ solubility in brine was validated against the experimental data from Chabab et al. [41]. Figure 1 shows a comparison between the modelling results and literature data for H₂ solubility in pure water at 50 °C and 100 °C. The comparison shows that the model gave accurate predictions over changing pressure and temperature. It also reflects that the increase in pressure increases the amount of hydrogen dissolved in water at both 50 °C and 100 °C. For example, the hydrogen solubility increased from 0.000485 mol fraction at 37 MPa and 50 °C to 0.001544 mol fraction at 123 MPa and 100 °C. This same trend can be observed in Figures 2 and 3 where higher brine salinities of 1 and 5 M NaCl were used. Similar experimental observations were made in other studies [42]. It can also be seen that increasing the temperature from 50 to 100 °C slightly decreased the dissolved hydrogen.

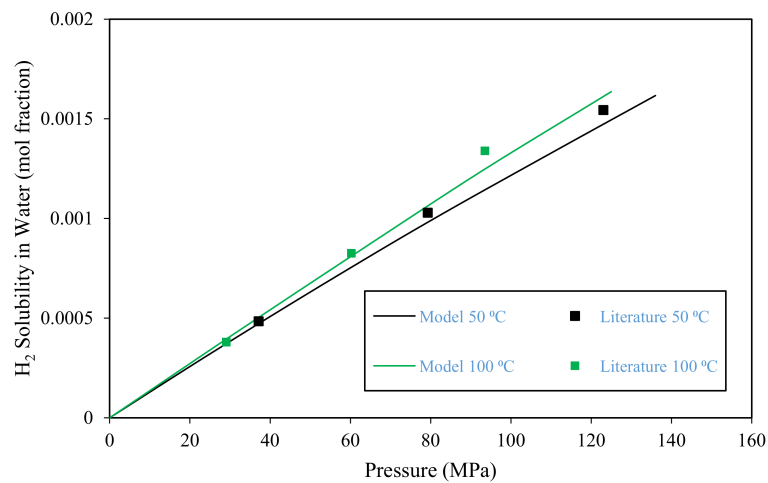


Figure 1. Comparison between modelling results and literature data [41] for H₂ solubility in pure water.

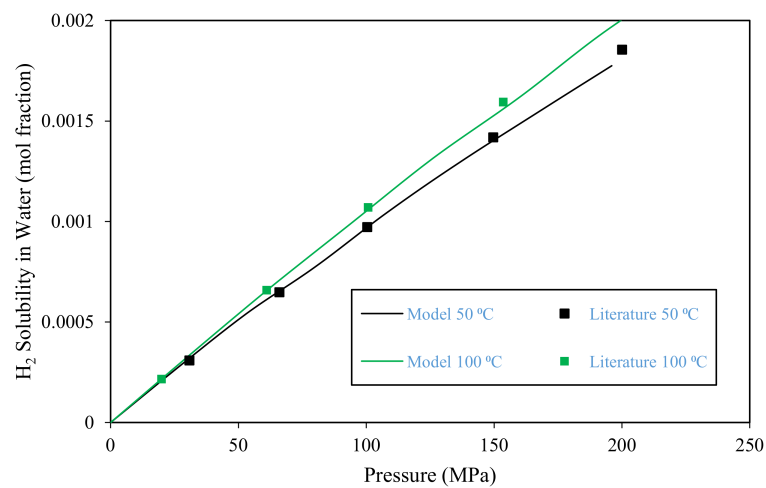


Figure 2. Comparison between modelling results and literature data [41] for H₂ solubility in 1 M NaCl water.

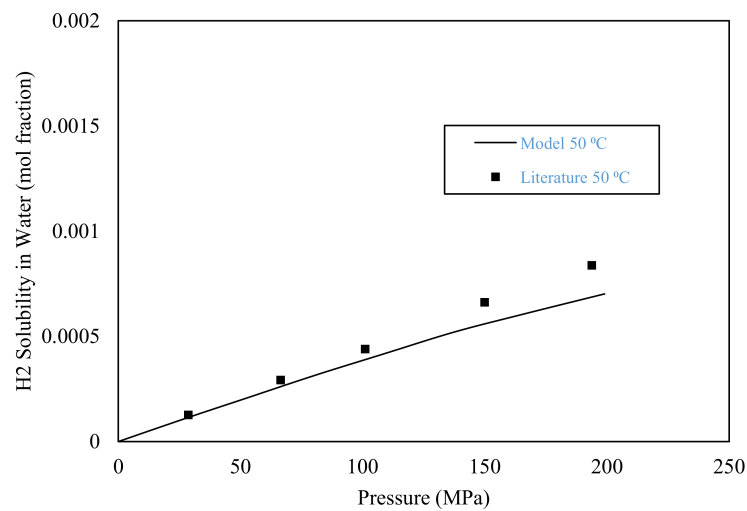


Figure 3. Comparison between modelling results and literature data [41] for H₂ solubility in 5 M NaCl water.

The developed model was also validated by comparing the model-predicted CO₂ solubility in both pure and saline water and experimental values from [43,44] at varying pres-

sure and temperature. Figure 4 presents a comparison between the modelling results and literature data in pure water at pressures between 28 atm and 2000 atm, and at 50 and 100 °C temperatures. Figure 4 shows that the model predicted the CO₂ solubility in pure water with high accuracy. The increase in pressure at a certain temperature causes an increase in CO₂ solubility in water. The effect of temperature on CO₂ solubility in water is dependent on the exerted pressure. At pressure below 450 atm, the increase in temperature decreases with the increasing temperature; however, this trend reverses above 450 atm, where an increase in temperature increases the solubility value. Similar observations were found in other experimental studies [45]. The model was also validated for CO₂ solubility in 4 M NaCl salinity brine against experimental data as shown in Figure 5. The comparison also demonstrates the model's ability to predict the CO₂ solubility in saline water with high accuracy. Having validated the model against experimental data from the literature, the model was then expanded to include mineral reactions, exchange reactions, methanisation and kinetics. The results of the sensitivity analysis of H₂ and CO₂ solubility and rock properties to change in pressure, temperature, salinity, and CO₂% as a cushion gas are presented.

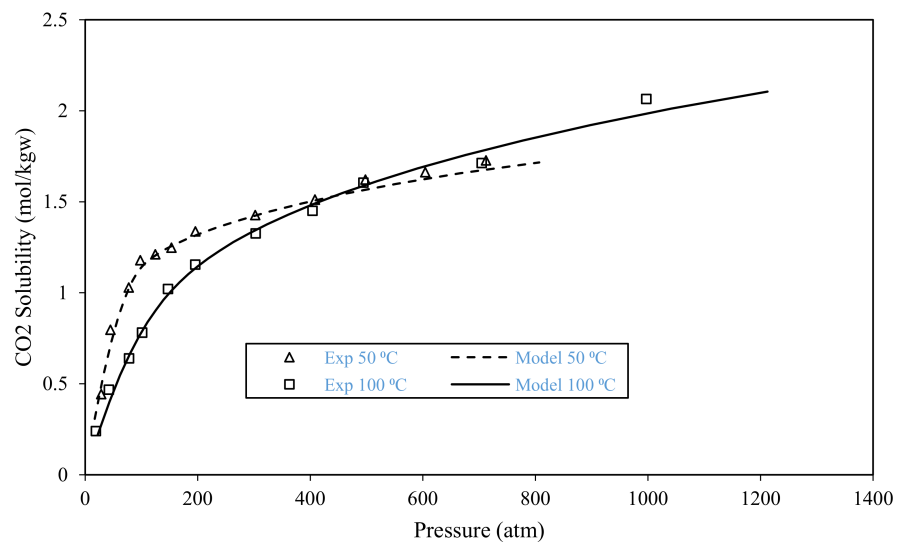


Figure 4. Comparison between modelling results and literature data [43,44] for CO₂ solubility in pure water.

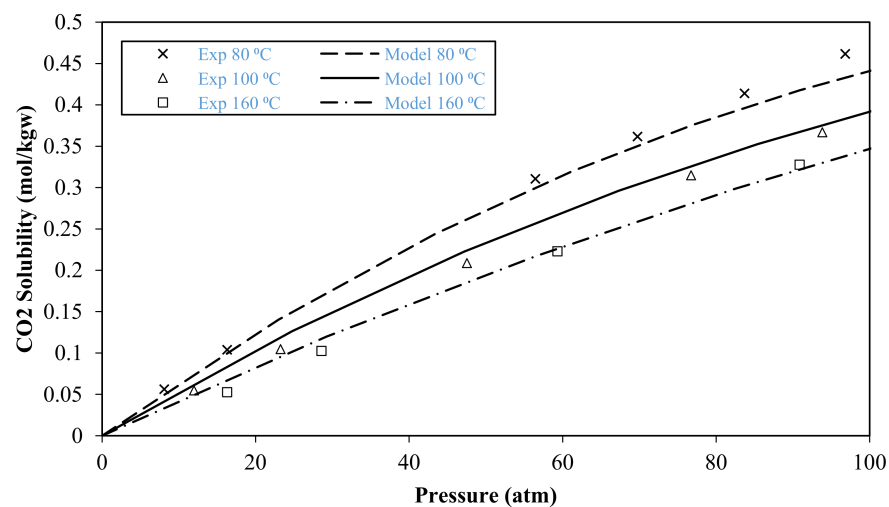


Figure 5. Comparison between modelling results and literature data [46] for CO₂ solubility in 4 M NaCl water.

3.2. Effect of Pressure

The effect of pressure on H₂ and CO₂ inventory and rock properties over 30 years at pressures between 100 and 500 atm have been determined. H₂ and CO₂ gas inventory decrease with time due to geochemical reactions (see Figure 6a). It can also be seen that the percentage of H₂ and CO₂ gas loss decreases with the increase in pressure. However, the increase in pressure reflects a higher initial amount of gas inside the reservoir as depicted in Figure 6b; hence, in absolute moles units, the amount of gas lost increases with pressure. For example, after 30 years of storage, the amount of CO₂ lost at 100 atm is 0.0945 mol/L of gas, which is less than that lost at 500 atm, which is 0.298 mol/L of gas. In the current targeted underground hydrogen storage application, the reservoir pressure is around 400–500 atm; hence, a high amount of CO₂ gas is expected to be stored inside the reservoir rock. H₂ loss inside the reservoir is less than 2% over 30 years, which is negligible compared to CO₂ gas loss, which can be up to 72% over the same period. This observation indicates that the H₂ loss due to geochemical reactions is not expected to be pronounced; however, the CO₂ loss is expected to be more pronounced, especially on the H₂ withdrawal process. Because CO₂ is intended to play the role of a cushion gas and maintain reservoir pressure to stabilise the H₂ withdrawal processes [47]. Based on this remark, more CO₂ gas can be injected as a cushion gas over the years to help maintain the cushion gas requirements.

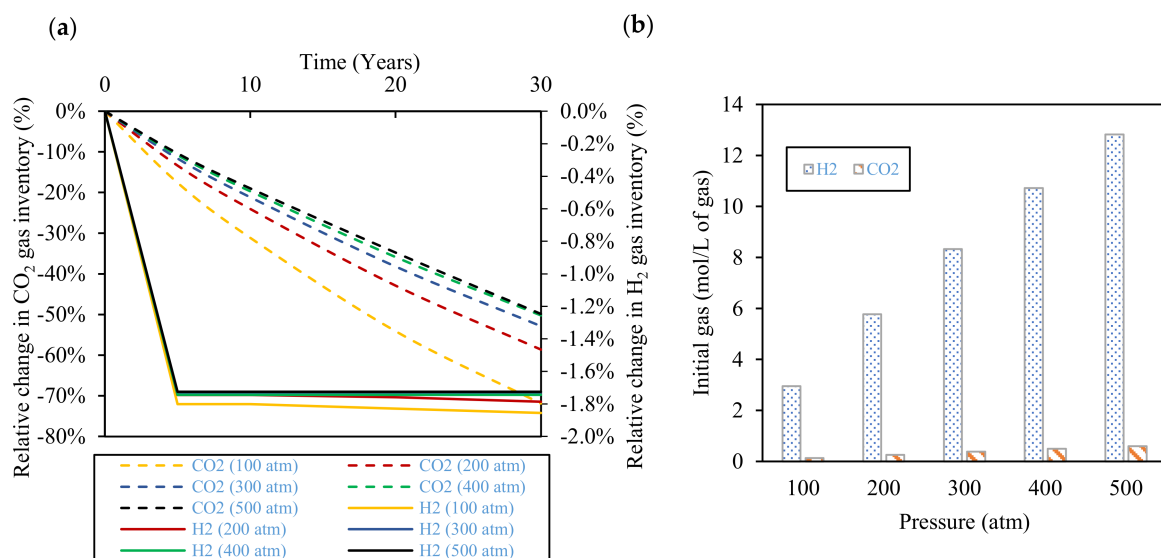


Figure 6. (a) Relative change in H₂ and CO₂ gas inventory over 30 years at various pressure values. (b) Initial H₂ and CO₂ gas present at various pressure values.

The effect of pressure on reservoir rock porosity and permeability is also evaluated and results are shown in Figure 7. The increase in pressure increases the reservoir mineral dissolution, specifically, K-feldspar and calcite, and consequently increases the porosity of the reservoir. These results agree with the experimental observations of [48], where the increased pressure boosted the mineral dissolution process. The porosity of the reservoir (Figure 7a) increases by 3.7% over the term of 30 years at 500 atm due to mineral dissolution reactions. This indicates that a higher amount of gas can be stored inside the reservoir after the initial 30-year period. The increase in porosity consequently causes an increase in the reservoir's permeability (Figure 7b). At 500 atm, the permeability can be increased by up to 12.3% relative to the initial permeability. This would enhance the fluid flow in the reservoir rock for both the injection and withdrawal processes; however, an important aspect is the effect of this change in reservoir minerals on the geomechanics of the reservoir rock and cap rock integrity. This aspect will be further investigated in the project in the next steps.

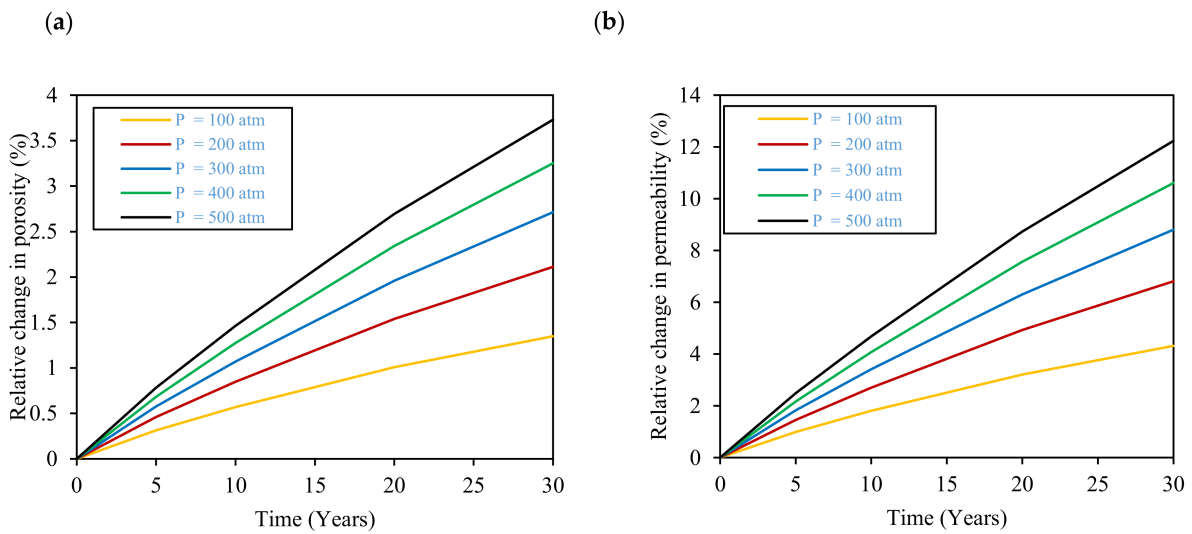


Figure 7. Kinetic effect of pressure on (a) porosity and (b) permeability over a 30-year period.

3.3. Effect of Temperature

Temperature affects both the gas solubility in water and the geochemical reactions' equilibrium constants. The increase in temperature increases the percentage of initial gases lost from geochemical reactions as depicted in Figure 8a. The increase in temperature from 80 °C to 120 °C resulted in the CO₂ losses inside the reservoir increasing from 20.8% to 67% over 30 years. The H₂ losses were less than 2% over the same period. It is also important to make the distinction that the change in CO₂ loss due to geochemical reactions in water in the presence of reservoir rock minerals is different to that in the absence of reservoir rock minerals, i.e., brine. CO₂ loss in water in the presence of reservoir rock minerals increases with temperature opposite to that in the absence of reservoir rock minerals as shown in Figure 5. Hydrogen kinetic loss due to the increase in temperature is not as pronounced compared to the CO₂ loss where the maximum hydrogen loss was less than 2% at 120 °C.

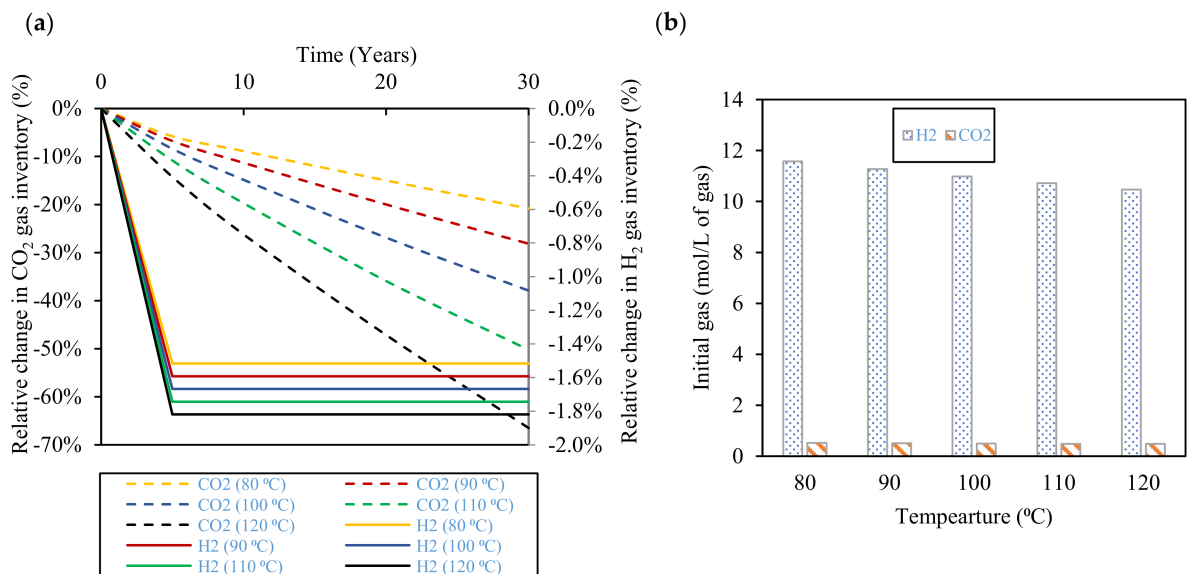


Figure 8. (a) Relative change in H₂ and CO₂ gas inventory over 30 years at various temperature values. (b) Initial H₂ and CO₂ gas present at various temperature values.

The effect of temperature on the rock properties, porosity, and permeability, was also investigated, and the results are shown in Figure 9. These modelling results agree with the

experimental results of [48], where the temperature rise enhanced the mineral dissolution. Figure 9a shows that at 120 °C, porosity can rise by up to 4% of the original porosity over 30 years, and Figure 9b indicates that permeability can increase by up to 13% over the same period. This consequently results in an enhancement in gas storage capacity and reservoir flow properties. Hence, the temperature effects need to be taken into effect when planning underground hydrogen storage operations in the Etive formation of the targeted depleted oil reservoir.

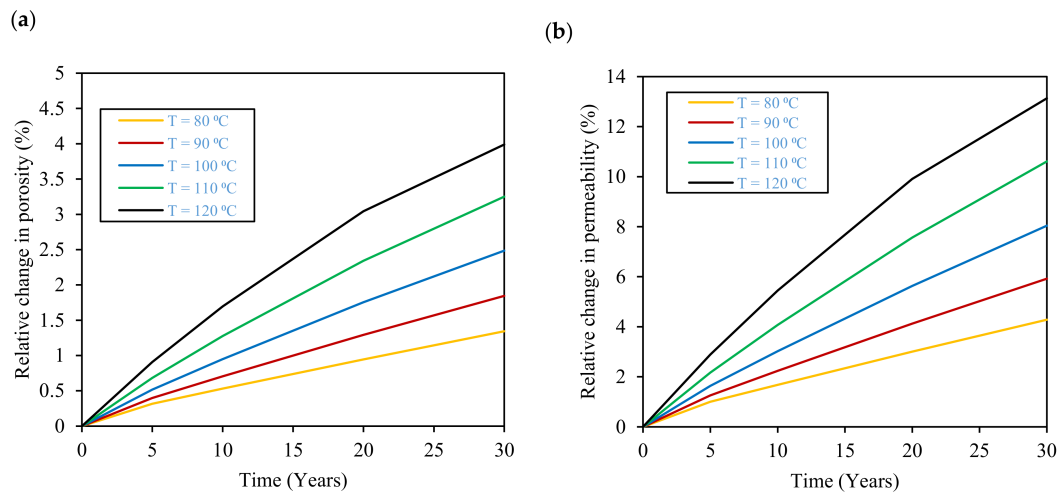


Figure 9. Kinetic effect of temperature on (a) porosity and (b) permeability over a 30-year period.

3.4. Effect of Salinity

Salinity affects the geochemical reactions occurring inside the reservoir. The increase in salinity decreases the amounts of gases lost due to geochemical activity inside the reservoir, as demonstrated in Figure 10. The effect of salinity on the gas inventory overall is not very pronounced as increasing the salinity from 0.5 to 5 M NaCl causes the relative loss of H₂ to decrease from 1.3% to 0.5% over the 30 years. However, the salinity effect would be more pronounced in the case that the reservoir temperature was lower, which would then enhance the activity of the bacteria responsible for methanisation and sulphate reduction reactions, especially at lower salinities.

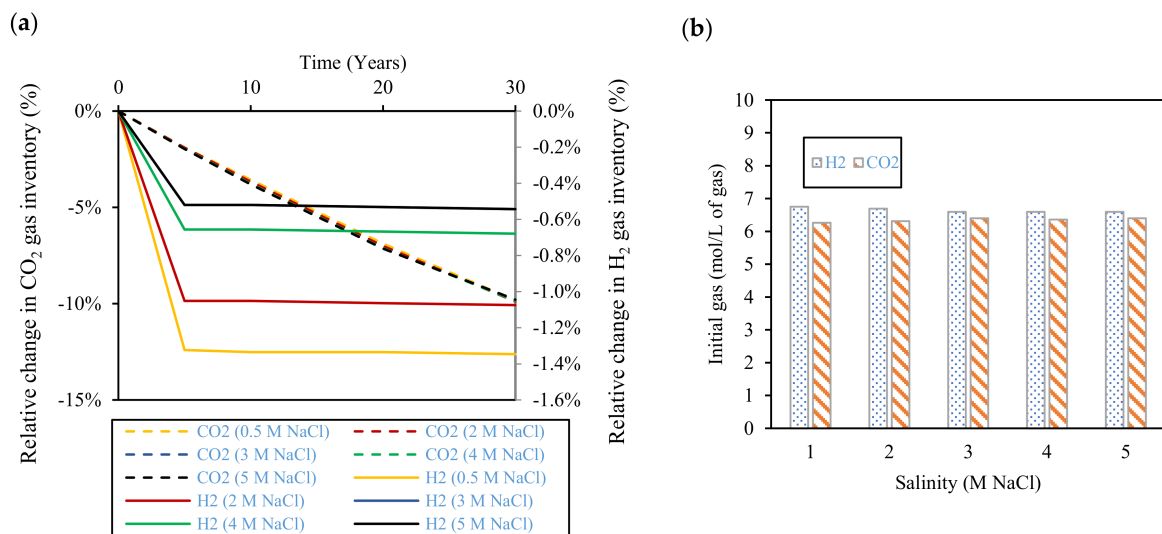


Figure 10. (a) Relative change in H₂ and CO₂ gas inventory over 30 years at various salinity values. (b) Initial H₂ and CO₂ gas present at various salinity values.

Salinity also has a minimal effect on reservoir porosity and permeability. Changing reservoir salinity in the model from 0.5 to 5 M NaCl, leads to the relative change in porosity in 30 years to decrease from 4.8% to 4.3% as shown in Figure 11a. Similarly, it can be seen from Figure 11b that salinity changes have a negligible effect on permeability. The overall reduction of porosity and permeability with salinity suggests a reduced dissolution of the reservoir minerals. Stillings and Brantley [49] found that increasing the salinity reduced the dissolution of feldspar at 25 °C and pH = 3.0. This can partially be attributed to the reduced CO₂ dissolved in brine at higher salinities, which consequently limits the mineral dissolution process.

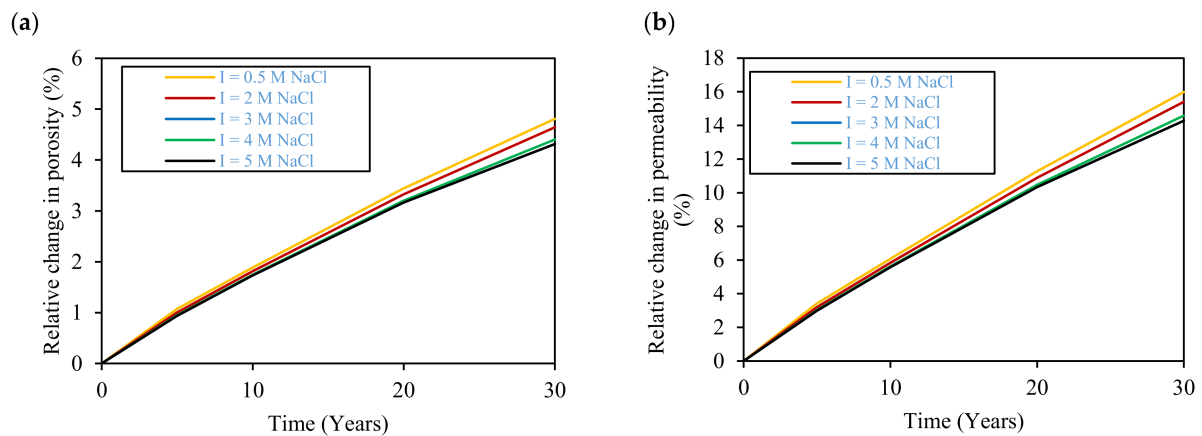


Figure 11. Kinetic effect of salinity on (a) porosity and (b) permeability over a 30-year period.

4. Conclusions

In this work, we investigated the effects of geochemical activities on the performance of underground hydrogen storage in the Etive sandstone formation in a depleted oil reservoir in the North Sea. The geochemical modelling was carried out using the geochemical code PhreeqC. The effects of varying pressure, temperature, and salinity on the injected gas inventory and the reservoir rock properties, porosity, and permeability, were evaluated over a period of 30 years. Results obtained from the geochemical investigations revealed the following:

- The increase in reservoir pressure from 100 atm to 500 atm results in an increase in the dissolved hydrogen and carbon dioxide. Moreover, as a result of mineral dissolution reactions, the higher pressures result in an increase in the reservoir rock's porosity and permeability. These porosity and permeability increases suggest that the reservoir's storage capacity and ease of storage and withdrawal will be improved with time during storage. Nonetheless, due to the expected pressure changes during the injection and production stage of underground hydrogen storage, it would be important to understand the effects of pressure combined with the expected geochemical and geomechanical impacts on the reservoir rock's strength during the underground hydrogen storage process.
- Temperature increase enhances hydrogen gas and carbon dioxide gas dissolution and, consequently, gas losses, where losses of 2% and 67% in the stored hydrogen and carbon dioxide gases are expected at 120 °C after 30 years of storage, respectively. Moreover, higher temperatures and salinities limit microbial activity inside the reservoir and restrain the rate of biotic reactions that consume stored hydrogen. Therefore, the evaluation of the temperature and salinity effects on any UHS application should consider both biotic and abiotic reactions. However, there is a lack of experimental data on biotic reactions that must be properly addressed in future research.
- In addition to the changes in porosity and permeability, this study shows that the losses of carbon dioxide gas inside the reservoir during underground hydrogen storage as

a result of aqueous and mineral reactions are significant and may require remedial carbon dioxide gas to be injected to maintain cushion gas requirements.

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