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CO_2 Cushion Gas Effects on H_2–Brine Interfacial Dynamics in Underground Hydrogen Storage

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KEYWORDS

H2, CO2, Interfacial tension (IFT), Underground Hydrogen Storage, Cushion gas

ABSTRACT

Hydrogen (H_2) is an essential energy vector for the global shift to cleaner power yet storing it at scale above ground remains both technically and environmentally challenging. Underground hydrogen storage (UHS) offers a viable alternative, but gas mobility and trapping behaviour are strongly controlled by interfacial processes. This study focuses on interfacial tension variations in H_2-CO_2-brine systems, under subsurface conditions, focusing on the effects of pressure (10–100 bar), temperature (20–80 °C), salinity, and gas composition.

Gas-liquid IFT (γLG) was measured experimentally in distilled water and formation brine. Results show that IFT in pure H_2–water systems increase with temperature, whereas in H_2–brine systems it decreases due to salt-driven modifications to intermolecular forces. CO_2-containing mixtures exhibit stronger pressure dependence, with increasing pressure lowering IFT through enhanced CO_2 solubility and molecular interactions. Additionally, CO_2 as a cushion gas played a crucial role in improving hydrogen mobility and storage efficiency by reducing IFT, thereby minimizing capillary trapping and enhancing gas injectivity. The observed nonlinear pressure dependence at higher CO_2 concentrations underscores the complexity of multiphase interactions in subsurface environments, emphasizing the need for accurate modelling in UHS. The findings contribute to the fundamental understanding of gas-fluid interactions under reservoir conditions and support the development of more efficient hydrogen storage and carbon sequestration strategies. ..

1. INTRODUCTION

Addressing the escalating issue of greenhouse gas emissions, especially those produced through fossil fuel use, is vital for slowing global warming and mitigating climate change [1,2]. In this context, renewable energy technologies have emerged as key alternatives, with hydrogen increasingly recognised as a central component of future low-carbon energy systems. [3,4]. Rising global energy needs, fueled by population growth and industrial development, highlight the urgency of shifting toward sustainable energy options such as solar, wind, and tidal power [5]. As a clean energy carrier, hydrogen offers a compelling replacement for traditional fossil fuels [6], with forecasts suggesting that it could supply a significant portion of the world's energy demand by 2050. The urgency of reducing greenhouse gas emissions, as reinforced by the Paris Agreement, highlights the importance of transitioning to low-carbon energy systems such as hydrogen [7]. Consequently, hydrogen is gaining prominence as a clean energy vector with the potential to displace fossil fuels and support a sustainable energy transition [8]. However, above-ground storage technologies struggle to accommodate large-scale or long-duration hydrogen storage, positioning UHS in saline aquifers and depleted hydrocarbon fields as a more viable option [9]. The efficiency of UHS is strongly governed by subsurface petrophysical properties, including mineral composition and surface behaviour [10,11]. In particular, interfacial tension and wettability are key parameters that dictate hydrogen injectivity, mobility, and recovery within geological formations [12].

In UHS, introducing a cushion gas alongside hydrogen is essential for maintaining reservoir pressure and enhancing overall storage performance [4,13]. CH_4 and CO_2 are commonly employed for this purpose, each offering specific advantages and operational constraints [14]. The selection and proportion of cushion gas relative to hydrogen are influenced by key reservoir characteristics, including permeability, wettability, and depth [4]. Among these options, CO_2 is particularly advantageous due to its widespread availability, cost-effectiveness, and compatibility with subsurface storage conditions [4,6]. Its higher density and compressibility compared with hydrogen enable more effective pressure support and smoother hydrogen withdrawal during production cycles [15,16]. Additionally, the use of CO_2 as a cushion gas supports global initiatives aimed at reducing greenhouse

gas emissions, further reinforcing its suitability for UHS applications [4,17]. Beyond its role in hydrogen storage, investigating the interactions between CO_2, H_2, and mineral surfaces has implications for underground bio-methanation.

This approach involves the controlled injection of hydrogen and carbon dioxide into a subsurface reservoir during periods of surplus energy production. Hydrogenotrophic methanogenic archaea present in the formation water can convert a portion of the stored gases into methane [4]. During periods of high energy demand, this methane-enriched gas mixture can be produced and used as an energy source. This process parallels the operation of engineered bioreactors, which are increasingly being integrated into existing gas infrastructure to enhance energy storage and distribution efficiency [18].

While extensive data exist on CO_2-brine interfacial tension [19,20], and some studies have examined H_2-brine interfacial properties [21], the influence of interfacial tension in subsurface storage systems involving mixed gases, particularly H_2 and CO_2, remains insufficiently investigated. Limited experimental research has explored the impact of gas mixtures on the interfacial properties of gas-brine systems [22] Previous studies have reported IFT values for pure H_2 and water under geological storage conditions (e.g., [8,23]). However, data on the IFT of mixed gas-water systems remain scarce, with only a few recent investigations addressing this topic. For instance, [24] measured the IFT of an ($H_2O + CO_2 + H_2$) system across a range of pressures and temperatures using the pendant-drop method. Their findings indicated that increasing pressure and temperature led to a reduction in IFT. Similarly, [25] conducted pendant-drop method experiments to assess the influence of temperature and methane (CH_4) content, as a cushion gas, on the IFT of an H_2-CH_4/brine system at 1000 psi. Their results demonstrated that IFT decreased with increasing temperature and methane concentration, underscoring the role of CH_4 as a cushion gas and its potential implications for caprock integrity and leakage risk.

This study examines the IFT of H_2+CO_2/brine systems under physio-thermal conditions relevant to geological storage. By analysing the variations in interfacial tension across gas compositions, specifically H_2 and CO_2, this research provides a detailed assessment of the influence of gas mixtures on gas-brine interfacial behaviour.

To conduct this investigation, brine samples were prepared and subjected to interfacial tension measurements using a pendant-drop device under a range of reservoir conditions. This study is distinct in its focus on the interfacial properties of mixed gases in brine, an area that has yet to be thoroughly explored. By evaluating interfacial tension across varying pressures, temperatures, and salinity levels, this research offers valuable insights into the role of CO_2 as a cushion gas in UHS.

Our study provides new experimental IFT data for H_2-CO_2 mixtures under various geo-storage conditions, offering critical insights for industrial-scale hydrogen storage. These findings support the development of simulation models to predict storage performance and flow behaviour, aiding in the effective implementation of underground hydrogen storage projects with CO_2 as a cushion gas.

2. EXPERIMENTAL METHODOLOGY

2.1. Materials

High-purity hydrogen (H_2) and carbon dioxide (CO_2) gases (≥99.999 wt%) were utilized to create gas mixtures with different mole fractions, formulated as [$(1-x)\ H_2 + x\ CO_2$], where x was set at 0.25, 0.50, and 0.75. To assess the impact of salinity, experiments were used both distilled water and formation brine to replicate realistic subsurface conditions [9,13].

2.2. Methodology

IFT measurements were measured using the captive drop technique, where the interaction between H_2+CO_2 and brine was analyzed under subsurface conditions. Three key parameters were considered: the inner needle diameter (used as a reference scale for image analysis), the density of the brine, and the density of gas at each experimental pressure and temperature. The IFT values were determined using the Young-Laplace capillarity equation and analyzed with ImageJ software. Each experiment was repeated three times, with the mean values reported, ensuring a maximum error margin of approximately ±3% [4,26]. Prior to conducting the primary experiments with H_2 and CO_2, nitrogen (N_2) was used to calibrate the experimental setup. The measured rock/N_2/brine contact angle data closely aligned with previously reported values under comparable conditions [13].

3. RESULTS AND DISCUSSION

This section presents a detailed evaluation of how the presence of CO_2 in an H_2 mixture affects interfacial tension (IFT) under conditions representative of subsurface storage. Gas compositions with CO_2 fractions of 25%, 50%, and 75% were analyzed across a range of conditions relevant to geological storage systems. Understanding these effects is essential, as reservoir depth and petrophysical characteristics can alter the composition of stored gas mixtures [27]. The selected pressure range (10–100 bar) and temperature range (20°C–80°C) were determined based on preliminary experiments and validation studies to ensure the findings accurately represent CO_2 and H_2 behavior in underground hydrogen storage. IFT measurements were conducted using both distilled water and formation brine to assess the role of salinity in gas-liquid interactions.

3.1. The impact of pressure, temperature, and salinity on IFT within 100 % H_2/water systems

IFT of H_2 with distilled water and formation brine is affected by temperature, salinity, and pressure. As can be seen in the Figure 1, the results indicate that temperature plays a significant role in altering the IFT trends for these systems. In the H_2/distilled water system, IFT increases with temperature, rising from 64.53 mN/m at 20°C to 73.34 mN/m at 80°C at 10 bar, with a similar trend observed at higher pressures. Conversely, in the H_2/formation brine system, IFT decreases with temperature, dropping from 67.11 mN/m at 20°C to 61.61 mN/m at 80°C at 10 bar. This difference is likely due to the interactions between dissolved salts and the gas-liquid interface, which alter the system's interfacial properties.

Pressure, on the other hand, has a minor effect on IFT compared to temperature. At 80°C, the IFT of the H_2/distilled water system slightly decreases from 73.34 mN/m at 10 bar to 69.12 mN/m at 100 bar, while for the H_2/formation brine system, it decreases from 61.61 mN/m at 10 bar to 58.54 mN/m at 100 bar. This marginal pressure dependency is attributed to the relatively small change in H_2 density with increasing pressure at constant temperature. Meanwhile, salinity significantly reduces IFT, as seen in the lower IFT values of H_2/formation brine compared to H_2/distilled water under the same conditions. At 10 bar and 80°C, for instance, the IFT of H_2/distilled water is 73.34 mN/m, whereas it is 61.61 mN/m for H_2/formation brine. These findings highlight that temperature has the most substantial impact on IFT, increasing it in distilled water but decreasing it in formation brine, while salinity plays a crucial role in lowering IFT, which could influence gas mobility in subsurface conditions.

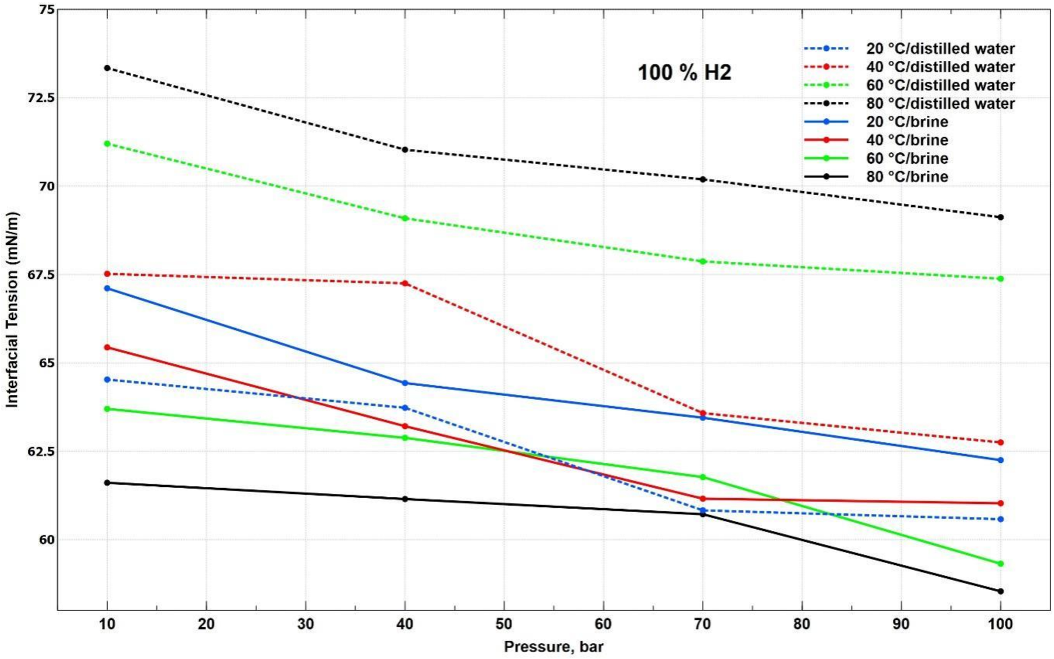

Figure 1: Interfacial tension (IFT) between 100% hydrogen (H_2) and distilled water/brine as a function of pressure and temperature.

3.2. The impact of pressure, temperature, and salinity on IFT within 75 % H_2+ 25% CO_2/water systems

IFT of a 75% H_2 + 25% CO_2 mixture with distilled water and formation brine is influenced by temperature, salinity, and pressure. As shown in Figure 2, the results indicate that temperature plays a significant role in altering IFT trends for these systems. In the $H_2 + CO_2$/distilled water system, IFT increases with temperature, rising from 60.55 mN/m at 20°C to 70.36 mN/m at 80°C at 10 bar, with a similar trend observed at higher pressures. Conversely, in the $H_2 + CO_2$/formation brine system, IFT decreases with temperature, dropping from 62.21 mN/m at 20°C to 64.82 mN/m at 80°C at 10 bar. This difference can be attributed to the interactions between dissolved salts and the gas-liquid interface, which alter the system's interfacial properties.

Pressure has a relatively minor effect on IFT compared to temperature. At 80°C, the IFT of the $H_2 + CO_2$/distilled water system slightly decreases from 70.36 mN/m at 10 bar to 60.31 mN/m at 100 bar, while for the $H_2 + CO_2$/formation brine system, it decreases from 64.82 mN/m at 10 bar to 52.73 mN/m at 100 bar. This marginal pressure dependency is due to the relatively small change in the $H_2 + CO_2$ mixture's density with increasing pressure at constant temperature.

Salinity plays a significant role in reducing IFT, as seen in the lower IFT values for $H_2 + CO_2$/formation brine compared to $H_2 + CO_2$/distilled water under the same conditions. For example, at 80°C and 10 bar, the IFT of $H_2 + CO_2$/distilled water is 70.36 mN/m, whereas it is 64.82 mN/m for $H_2 + CO_2$/formation brine. These findings emphasize that temperature has the most substantial impact on IFT, increasing it in distilled water but decreasing it in formation brine, while salinity significantly lowers IFT. This could have implications for gas mobility in subsurface conditions.

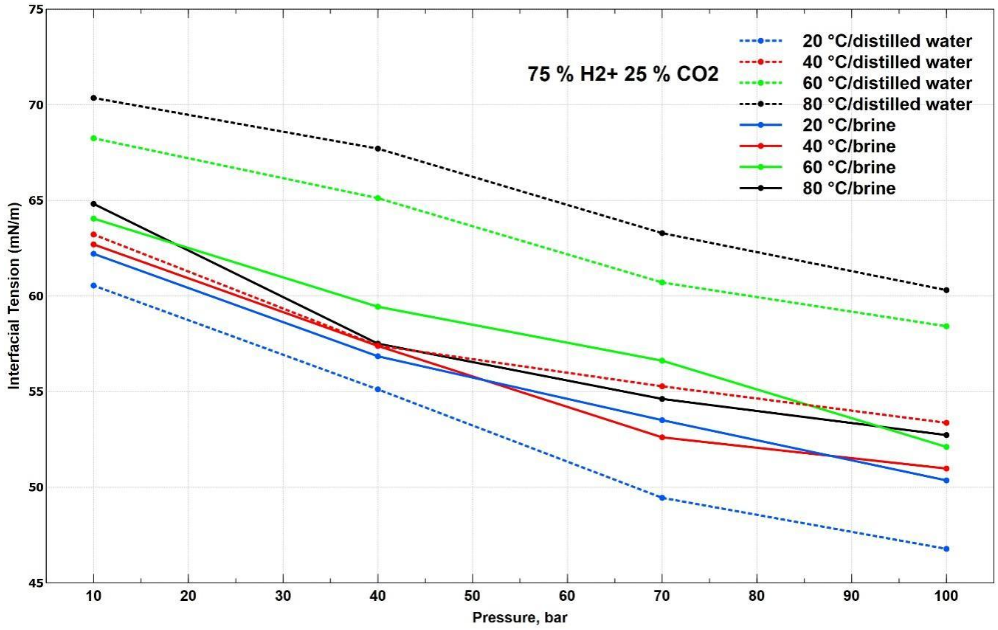

Figure 2: Interfacial tension (IFT) between 75% H_2 + 25% CO_2 mixture and distilled water/brine as a function of pressure and temperature.

3.3. The impact of pressure, temperature, and salinity on IFT within 50 % H_2+ 50% CO_2/water systems

The interfacial tension (IFT) of a 50% H_2 + 50% CO_2 mixture with distilled water and formation brine is influenced by temperature, salinity, and pressure. As shown in **Figure 3**, the results indicate that temperature plays a key role in modifying IFT trends for these systems. In the $H_2 + CO_2$/distilled water system, IFT increases with temperature, rising from 56.21 mN/m at 20°C to 67.15 mN/m at 80°C at 10 bar, with similar trends observed at higher pressures. In contrast, in the $H_2 + CO_2$/formation brine system, IFT initially increases but then decreases with temperature, rising from 59.12 mN/m at 20°C to 66.87 mN/m at 80°C at 10 bar.

Pressure shows a relatively minor effect on IFT compared to temperature. At 80°C, the IFT of the $H_2 + CO_2$/distilled water system decreases from 67.15 mN/m at 10 bar to 51.71 mN/m at 100 bar, while for the $H_2 + CO_2$/formation brine system, it decreases from 66.87 mN/m at 10 bar to 55.65 mN/m at 100 bar. This modest pressure dependence is primarily due to the small change in the $H_2 + CO_2$ mixture's density with increasing pressure at constant temperature.

Salinity has a moderate effect in reducing IFT, as observed from the lower IFT values for $H_2 + CO_2$/formation brine compared to $H_2 + CO_2$/distilled water under the same conditions. For example, at 80°C and 10 bar, the IFT of $H_2 + CO_2$/distilled water is 67.15 mN/m, whereas it is 66.87 mN/m for $H_2 + CO_2$/formation brine. These findings suggest that temperature has the most profound impact on IFT, increasing it in distilled water but altering it in formation brine, while salinity plays an important role in reducing IFT, which could influence gas mobility in subsurface environments.

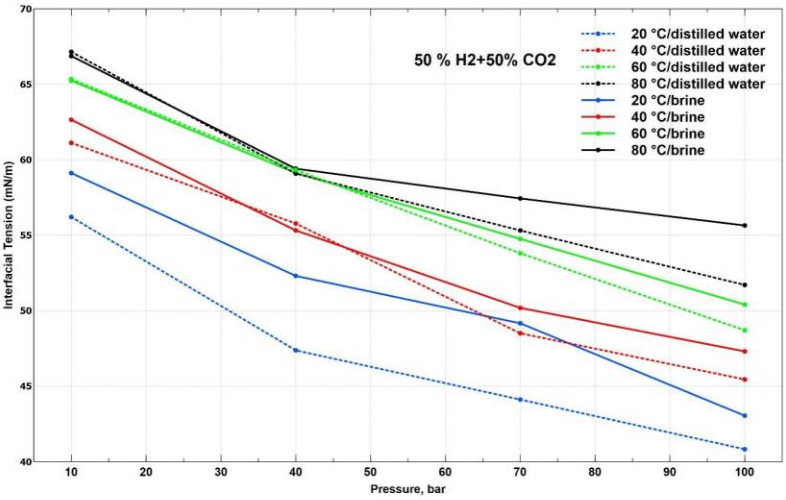

Figure 3: Interfacial tension (IFT) between 50% H_2 + 50% CO_2 mixture and distilled water/brine as a function of pressure and temperature.

3.4. The impact of pressure, temperature, and salinity on IFT within 25 % H_2+ 75% CO_2/water systems

The interfacial tension (IFT) of a 25% H_2 + 75% CO_2 mixture with distilled water and formation brine is significantly influenced by temperature, salinity, and pressure. As shown in **Figure 4**, both temperature and pressure have substantial effects on IFT, but pressure appears to have a more pronounced impact, particularly in the H_2 + CO_2/distilled water system.

In the H_2 + CO_2/distilled water system, IFT increases with temperature, rising from 53.1 mN/m at 20°C to 63.15 mN/m at 80°C at 10 bar. However, at higher pressures, the IFT decreases significantly. For instance, at 100 bar, IFT decreases from 63.15 mN/m at 80°C to 42.48 mN/m, indicating that pressure has a greater influence than temperature in this system. Similarly, in the H_2 + CO_2/formation brine system, IFT shows a similar increase with temperature, rising from 56.33 mN/m at 20°C to 65.7 mN/m at 80°C at 10 bar. Yet, pressure exerts a more substantial effect, as the IFT decreases from 65.7 mN/m at 10 bar to 45.65 mN/m at 100 bar (both at 80°C).

These results highlight the dominant role of pressure in reducing IFT in both systems, as the changes in IFT due to pressure are larger than those observed with temperature. The increase in pressure leads to a denser gas mixture, which affects the gas-liquid interface, thus lowering the IFT. On the other hand, temperature increases IFT in distilled water but influences it differently in brine.

Salinity also plays an important role in reducing IFT, as observed in the H_2 + CO_2/formation brine system, where the IFT values are consistently higher than in the distilled water system at the same temperature and pressure. For example, at 80°C and 10 bar, the IFT of H_2 + CO_2/formation brine is 65.7 mN/m, compared to 63.15 mN/m in distilled water.

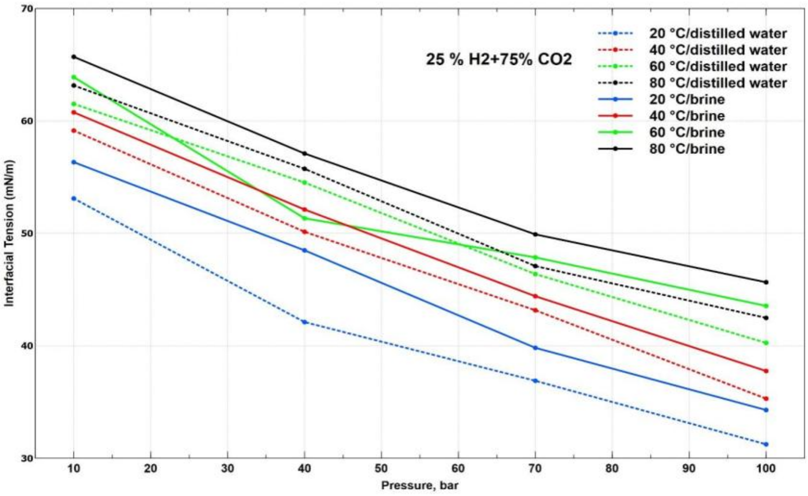

Figure 4: Interfacial tension (IFT) between 25% H_2 + 75% CO_2 mixture and distilled water/brine as a function of pressure and temperature.

3.5. The impact of pressure, temperature, and salinity on IFT within 100 % CO_2/water systems

The interfacial tension (IFT) of a 100% CO_2 mixture with distilled water and formation brine is significantly influenced by temperature, salinity, and pressure. As shown in **Figure 5**, both temperature and pressure affect IFT, with pressure having a more pronounced impact, particularly in the CO_2/distilled water system.

In the CO_2/distilled water system, IFT increases with temperature, rising from 51.41 mN/m at 20°C to 61.24 mN/m at 80°C at 10 bar. However, as pressure increases, IFT decreases substantially. For example, at 100 bar, IFT decreases from 61.24 mN/m at 80°C to 37.81 mN/m, showing that pressure has a greater influence than temperature in this system. Similarly, in the CO_2/formation brine system, IFT increases with temperature, rising from 54.49 mN/m at 20°C to 63.29 mN/m at 80°C at 10 bar. Yet, pressure exerts a more significant effect, with IFT decreasing from 63.29 mN/m at 10 bar to 40.78 mN/m at 100 bar.

These results underline the dominant role of pressure in reducing IFT in both systems. The decrease in IFT with increasing pressure can be attributed to the increase in the density of the CO_2 gas mixture, which modifies the gas-liquid interface. In contrast, temperature increases IFT in distilled water but has a different effect in the brine system, where the interaction with dissolved salts may influence the interfacial characteristics differently.

Salinity also plays a critical role in reducing IFT, as observed in the CO_2/formation brine system, where the IFT values are consistently higher than in the distilled water system at the same temperature and pressure. For instance, at 80°C and 10 bar, the IFT in the CO_2/formation brine system is 63.29 mN/m, compared to 61.24 mN/m in distilled water.

In summary, pressure has the most substantial impact on IFT in both systems, with temperature and salinity also contributing to variations in IFT. This knowledge is essential for understanding gas behaviour and mobility in subsurface environments, particularly in high-pressure CO_2 storage scenarios.

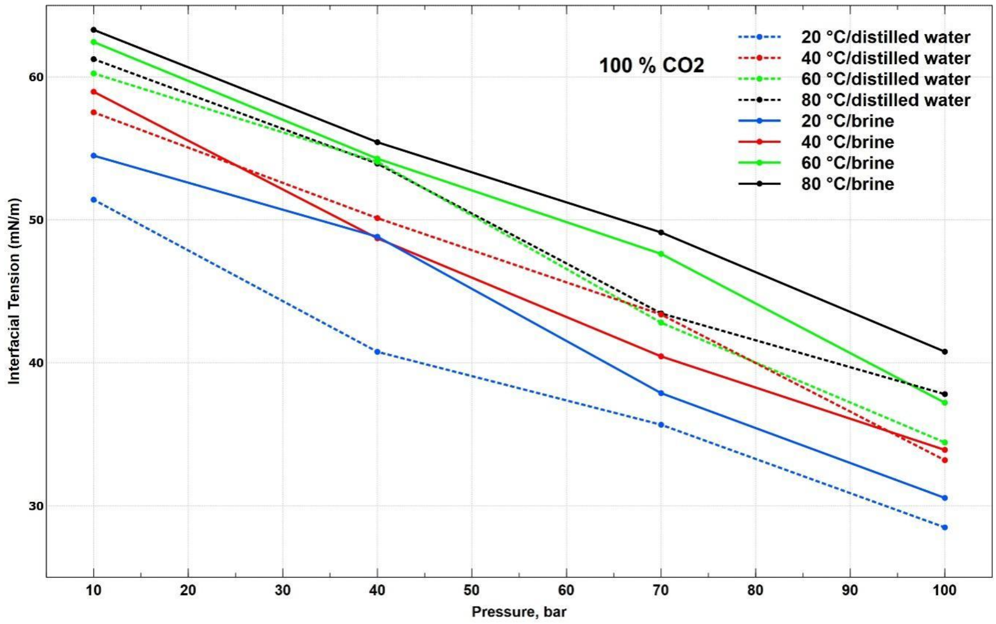

Figure 5: Interfacial tension (IFT) between 100% CO_2 mixture and distilled water/brine as a function of pressure and temperature.

3.6. Discussion

The interfacial tension (IFT) between gas and liquid phases plays a fundamental role in subsurface gas storage, influencing gas mobility, capillary trapping, and overall reservoir behaviour. This study demonstrates the intricate effects of pressure, temperature, and salinity on IFT for various H_2 and CO_2 gas mixtures under realistic subsurface conditions, aligning with established physicochemical principles governing gas-liquid interactions.

Temperature exhibited a dual effect on IFT, depending on the salinity of the aqueous phase. In pure H_2/distilled water systems, IFT increased with temperature, rising from 64.53 mN/m at 20°C to 73.34 mN/m at 80°C under 10 bar, consistent with previous findings that attribute this trend to enhanced molecular kinetic energy and reduced intermolecular cohesion in the liquid phase. However, in H_2/formation brine systems, the opposite trend was observed, with IFT decreasing from 67.11 mN/m at 20°C to 61.61 mN/m at 80°C, likely due to the solubility effects of dissolved salts that weaken intermolecular forces at higher temperatures. A similar trend was evident in CO_2-rich systems, where IFT increased with temperature in distilled water but exhibited a less pronounced increase or even a decrease in formation brine.

Pressure effects on IFT were relatively minor for pure H_2 systems but more significant in CO_2-containing mixtures. For H_2/distilled water at 80°C, IFT decreased slightly from 73.34 mN/m at 10 bar to 69.12 mN/m at 100 bar, reinforcing the notion that H_2 compressibility effects are minimal due to its low molecular weight and weak intermolecular interactions. Conversely, in the 25% H_2 + 75% CO_2/distilled water system, IFT dropped substantially from 63.15 mN/m at 10 bar to 42.48 mN/m at 100 bar, driven by CO_2's higher solubility at elevated pressures, which enhances gas dissolution and promotes stronger molecular interactions with the liquid phase.

Salinity exerted a pronounced influence on IFT across all gas mixtures, reducing interfacial tension in both H_2 and CO_2-rich systems. In the 100% H_2 system at 80°C and 10 bar, IFT was 73.34 mN/m in distilled water but decreased to 61.61 mN/m in formation brine, consistent with the "salting-out effect," where dissolved salts elevate the ionic strength of the solution, decrease gas solubility, and consequently lower IFT. This trend persisted in CO_2-containing mixtures, with formation brine consistently yielding lower IFT values compared to distilled water under similar conditions.

These findings provide valuable insights into the interfacial behaviour of H_2 and CO_2 mixtures in subsurface environments, reinforcing the complex interplay of temperature, pressure, and salinity in determining IFT. Understanding these dynamics is crucial for optimizing gas storage strategies, enhancing reservoir performance, and mitigating potential risks associated with subsurface hydrogen and CO_2 injection.

4. CONCLUSIONS

This study provides a comprehensive evaluation of interfacial tension (IFT) variations in H_2-CO_2-brine systems under reservoir conditions, offering valuable insights into the physicochemical interactions that govern subsurface hydrogen storage. The results confirm that temperature, pressure, and salinity significantly influence IFT, with distinct trends observed depending on the gas composition and aqueous phase properties. Temperature effects varied with salinity, leading to either an increase or decrease in IFT, while pressure had a more pronounced impact in CO_2-containing mixtures due to enhanced gas solubility. Salinity consistently reduced IFT, reinforcing the "salting-out effect" that alters gas-water interactions.

Furthermore, the study highlights the critical role of CO_2 as a cushion gas in underground hydrogen storage. Increasing the proportion of CO_2 led to a decline in IFT, improving hydrogen mobility and potentially enhancing storage efficiency. The observed nonlinear pressure dependence at higher CO_2 concentrations underscores the complexity of multiphase interactions in subsurface environments. These findings provide essential experimental data for modelling and simulating hydrogen storage in geological formations, contributing to the optimization of cushion gas strategies for maintaining reservoir pressure and maximizing hydrogen recovery.

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