Reliability of chemical osmosis as a practical method to extract water out of shale and promote shale’s mechanical stability

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Abstract
Wellbore instability in shale formations is one of the most bothersome problems leading to significant changes in shale’s petrophysical, mechanical, and chemical properties, and could cost billions of dollars annually. Drilling fluid’s ionic composition is one of the factors affecting wellbore stability. This paper investigates the impact of chemical osmosis, ionic diffusion, and diffusion osmosis on the stability of shale, identify the critical salt concentration, and examine the impact of ionic type and concentration on shale stability. Experimental methods include utilization of shale I and II through linear expansion and gravimetric measurement tests using different salts. Results show that the critical salt concentration in shale I and II is 8 w/w%. Chemical osmosis is found to be a reliable method for water extraction out of shale if critical concentration value is not surpassed. Beyond this value, the ionic diffusion and diffusion osmosis in shale I and II were adversely affecting shale’s mechanical and chemical stability. The swelling and gravimetric ions and water uptake tests showed that shale is a leaky semi-permeable membrane, and that chemical osmosis could be jeopardized by ionic transport into shale. However, the water transport impact on changes in the thickness of diffuse double layer of clay needs to be investigated. The impact of temperature on water and ions transportation into shale needs to be examined to reflect in-situ conditions. In this study, the cation type that makes up the salt was varied while the anion type was fixed. Since both cations and anions tend to diffuse into shale in the presence of a concentration gradient, it is recommended to vary the anion type to study the combined impact of cation and anion type on shale stability.

Introduction
According to the Energy Information Administration (EIA), shale formations make up to 42% of the recoverable hydrocarbon resources globally, making it of great importance to understand the mechanism and nature of shale formations to mitigate major problems that drillers and engineers might face during drilling and production. Interestingly, shale formation takes about 75% of total drilled formations and 90% of borehole instability problems that are encountered during drilling operations [1-6]. Wellbore instability in shale formations is one of the most bothersome problems facing drilling engineers. Besides its technical hurdle, it could add billions of dollars annually to the overall drilling cost if left unresolved. The imbalance that occurs between shale formations and drilling fluids can lead to wellbore instability and hence some serious drilling problems as a result [7]. Wellbore instability is primarily a function of how rocks respond to the induced stress concentration around the wellbore during various drilling activities. Before creating a wellbore, the rock formation is at equilibrium with its rocks surroundings and the stress distribution is at equilibrium. When a hole is made, this equilibrium is disturbed and stresses around the wellbore are changed. To reduce the impact of stresses modification, drilling fluids are utilized to compensate for the removed rocks and re-balance the altered stresses in an attempt to bring them back to their original state. There are many factors which affect wellbore stability including but not limited to far-field principal stresses, wellbore trajectory rock, mechanical properties, pore pressure, temperature, drilling fluid’s mud weight and drilling fluid’s chemistry (ionic composition).

Drilling fluid’s ionic composition is one of the main factors affecting wellbore stability particularly in shale formations. The presence of various types and concentrations of chemicals between the drilling fluid and the drilled shale can produce a chemical potential gradient which can lead to the exchange of water between the shale and the drilling fluid. The transport of water in or out of shale could change its mechanical properties and pore pressure which could cause shale failure and collapse. For low permeability shales, this issue can produce high pore pressure since the pressure dissipation will be exceptionally low; whereas, for high permeability formations such as sandstones and carbonates, the pore pressure increases are minimal and can be neglected.

The process that governs the exchange of water between shale and drilling fluids is called chemical osmosis. By definition, chemical osmosis is a process in which water molecules tend to pass through a semi-permeable membrane from a less concentrated solution to a more concentrated one in response to the existence of a chemical potential gradient (Figure 1). As stated
above, the chemical potential is highly related to the water activity (ionic concentration) of the aqueous solution. Thus, when low salinity (high water activity) causes shale shrinkage and pore pressure reduction, and this could enhance the effective drilling fluids interact with shale of higher salinity (low water activity), water will move into the higher salinity shale thereby increasing its pore pressure, decreasing the effective stresses, causing shale swelling, and ultimately reducing borehole stability. On the other hand, when high salinity drilling fluids interact with shale of lower salinity, water will flow out of shale and stresses around the wellbore and strengthen the wellbore. Thus, it could aid in sidestepping excessive problems, such as kicks, blow outs, lost circulations, and stuck pipe, and reduce time waste.

Drilling fluids’ chemistry is often manipulated to produce the most optimum chemical environment when shale interacts with drilling fluids. The main goal is to prevent water flow into shale and if reasonably possible, to induce water flow out of shale. Water extraction out of shale requires making the water activity of the drilling fluid less than that of the shale. The addition of salts such as KCl, NaCl and CaCl2 to the make-up of the drilling fluid lowers its water activity. In fact, the higher the amount of salts in the drilling fluid, the lower its water activity becomes until the saturation limit is reached. At the saturation limit, the water activities of KCl, NaCl and CaCl2 remains constant. There is only one caveat to this procedure. The addition of salt to the drilling fluid will promote ionic diffusion in the direction of shale as per Fick’s law if the ionic concentration in the drilling fluid is higher than that in the shale. In addition, water clouds will flow along with their respective ions into shale by diffusion osmosis. The transport of ions and their associated water “water cloud” into shale may negate the beneficial impact of water extraction out of shale. Namely, the flow of ions and their associated water will increase the shale’s pore pressure, change its pore fluid composition, alter its mechanical properties, and reduce its strength. Therefore, one must be careful not to reduce the water activity of drilling fluid, via addition of salt, to the point where more damage is done than good. It is believed that there exists a critical salt concentration, depending on salt type, where the full benefit of chemical osmosis can be exploited without jeopardizing the shale mechanical and chemical stability. Above this critical salt concentration, ionic diffusion and diffusion osmosis may adversely affect the shale’s mechanical and chemical stability and thus compromising the positive effect (water extraction out of shale) of chemical osmosis.

This project investigates the impact of chemical osmosis, triggered by water activity differences between drilling fluids and shale, on wellbore stability. Moreover, the impact of ionic diffusion and diffusion osmosis on shale stability will be addressed. This work also plans to experimentally identify the critical salt concentration beyond which chemical osmosis becomes ineffective. The following tasks will be conducted in order to satisfy the objectives of this proposed project:

1. Investigate the impact of chemical osmosis on shale swelling when interacting with salt solutions (KCl, NaCl and CaCl2) of different concentrations.
2. Confirm the reliability of osmosis as a practical method to extract water out of shale and promote shale’s mechanical stability.
3. Investigate the impact of ionic diffusion and diffusion osmosis on shale swelling when interacting with salt solutions (KCl, NaCl and CaCl2) of different concentrations.
4. Identify the critical concentrations for KCl, NaCl and CaCl2 beyond which chemical osmosis becomes ineffective for the purpose of shale’s stability.
5. Inspect the relationship between water and ions uptake and the swelling behaviour of shale as it interacts with KCl, NaCl and CaCl2 of different concentrations.

![Figure 1: The concept of chemical osmosis of solution molecules.](image-url)
Causes of wellbore instabilities must be identified in order to offer ways to mitigate it [8]. pointed out that the biggest contributor to wellbore instability in shale formations is the adsorption of water instigating swelling of the wellbore in shale. Water adsorption occurs during the interaction between drilling fluids and shale formation, causing a reduction in elasticity modulus and strength of the formation [9,10]. Changes in the pressure, elastic modulus, and strength of shale in addition to swelling mainly due to water adsorption lead to wellbore instability [11]. Several authors discussed the causes and their mechanisms. The water and ions movement mechanisms were investigated by [12-14]. Understanding the movement of water and ions in and out of shale and thus minimizing the effects of such movements is one of the solutions to mitigate the problem mentioned that the more inclined the borehole is, the higher the risk of encountering wellbore instability. They also did a case study on Nahr Umr Shale in Halfaya Oilfield, Iraq, and concluded that augmentation of mud’s ionic concentration and enhancement of its sealing capacity are important to maintain wellbore stability. The reason behind increasing the ionic concentration is that it prevents the flow of the mud’s free water into the formation as it can hydrate the shale leading to wellbore collapse [15,16], indicated that the extraction of water out of shale through chemical osmosis would enhance the compressive strength of shale if the water activity was marginally higher than the test solution. He also stated that the negative effects of diffusion osmosis outweigh the positive effects of chemical osmosis. Moreover, presented a case study on the wellbore stability in Zubair Shale in Kuwait [17]. They emphasized on the role of pore pressure of shale in causing rock failure. The Mohr-Coulomb and Drucker-Prager criteria are most frequently used criteria utilized for rock failure in wellbore stability analysis [18]. An explanation and comparison of both criteria are summarized by [19]. Also stated that deviated wells may require side-tracks to mitigate instability which means higher well costs [20]. studied effects of deviation angle and pore pressures in wellbore instability and found that it is insufficient to rely only on pore pressure to design the stable mud weight window [21].

Proposed two models of collapse pressure calculation and well-cycle instability area. The condition of underbalanced seepage flow and the rock strength anisotropy were taken into consideration in both models [22]. Presented a non-linear model that is ‘thermo-chemo-poroelastic’ to study the effect of distinct types of gradients on the distribution of stress and pore pressure in the wellbore. Pore pressure changes may occur due to cation exchange process presented, through a case study, an approach to incorporate the dual-porosity and permeability proelasticity, and the effects of chemical/thermal gradients into a shale wellbore stability model to get an accurate composition of drilling fluids [23-25].

One of the ways to mitigate the instability of wellbore in shale formations is to reduce the invasion of fluids into shale [26]. Tried adding silica nanoparticles in water-based muds as a seal to prevent the water invasion into shale, and it was effective [27]. Discussed the impact of mud infiltration in fracture rocks and their contribution to wellbore stability. They found that mud infiltration contributes to reduction of friction angle of fractures and as a result, affects the stability of wellbore [28]. Concluded that the following wellbore stability models were commonly used by several researchers in order to mitigate the wellbore stability problem: a) elastic model b) poro-elastic model c) thermo-poro-elastic model, and d) chemo-thermo-poro-elastic model [29]. Experimentally proved that destabilization the ionic diffusion effect on shale is more beneficial than transferring water out of shale osmotically. He stated that sodium and calcium ions in addition to their associated water can cause shale failure while potassium ions can improve shale's compressive strength. These clay swelling inhibitors are particularly important in the stabilization of shales and enhancing their inhibition properties [30]. Recommended the use of an intelligently designed KCl water-based mud in order to curb shale swelling. Investigated the efficiency of KCl water-based mud as an inhibitor and came up with the conclusion that increasing the concentration of KCl would decrease the swelling potential. However, they noticed the longer the KCl is immersed in shale, the more prone the shale is for a compressive strength reduction over time [31-33]. KCl water-based muds (WBM) can inhibit the swelling of smectite. They tried to optimize the composition of KCl and KCl+NaCl salts in WBM to stabilize shale and found that a mixture of 5% KCl and 0% NaCl, or 0% KCl and 5% NaCl are the optimum compositions with inhibition properties. Alternatively, they concluded that a mixture of 2% KCl and 3% NaCl could serve the same purpose, i.e. having a drilling fluid with top inhibition.

Materials and experimental methods
Shale and salt solutions: In this work, shales (I and II) were utilized to study the impact of chemical osmosis, ionic diffusion, and diffusion osmosis on the stability of shale and identify the critical salt concentration beyond which chemical osmosis becomes ineffective. This was accomplished through a series of swelling and gravimetric measurements. Numerous samples were carefully cut from each shale core and were cautiously handled to alleviate shale communication and exposure to air. This communication of shale to the atmosphere could alter the shale’s native water activity and water content [34-37]. Initially, the native water activities of shales I and shale II were 0.98 and 0.96, respectively, but we have intentionally conditioned shale I and II to have 0.98 water activities, prior to exposing them to salt solutions, by placing the samples in a desiccator having a saturated K2SO4 solution to retain a 0.98 water activity. Table 1 shows the minerals make up of shales I and II while Table 2 shows the petrophysical and chemical properties of shales I and II. Comprehensive explanation of the experimental procedures used to obtain the mineralogical and petrophysical features of shales I and II could be found elsewhere [38]. As part of experimental methods, sodium chloride (NaCl), potassium chloride (KCl) and calcium chloride (CaCl2) of different concentration were utilized to serve as the test solutions. As seen in the test matrix (Table 3), two salt properties will be under investigation; cation type and concentration. Figure 2 shows the corresponding water activity for NaCl, KCI and CaCl2 solutions at each concentration.
Experimental methods
Linear expansion test: A standard linear expansion device, shown in Figure 3, was used to measure the amount of swelling shale experiences when exposed to different salt solutions. Shale samples were sited in a small plastic bag and securely placed between the movable anvil and the stationary anvil of the swelling device. A 30 ml of salt solution is poured into the plastic bag holding the shale sample and air is squeezed out and the bag is tightly closed. During the linear expansion test, the shale samples are left immersed in salt solutions for interaction for a total 1500 minutes (25 hours) in which the volumetric displacement reading is taken periodically using a standard stopwatch. This displacement reading represents an increase or decrease in shale length as a result of its direct contact with the salt solutions. A positive displacement reading (increase in length) indicates shale swelling while a negative displacement reading (decrease in length) points to shale shrinkage. The original length of shale (L0) was taken prior to placing the shale sample in the plastic bag. The amount of swelling or shrinkage of shale as it interacts with salt solutions was estimated by dividing the increase or decrease in sample length, displacement reading, by the original sample length. A graph of swelling or shrinkage (%) of shale versus time (minutes) is produced as a function of salt type and concentration. It is vital to point out that this method allows us to measure shale swelling and simultaneously estimate the amount of water and ions exchanged between the shale sample and the salt solutions. This technique makes shale swelling investigation more applicable especially since shale’s swelling is argued to be extremely connected with water and ions transport in or out of shale [39-41].

Gravimetric measurement test
The quantity of water and ions movement when shale communicates with salt solutions of dissimilar concentrations was measured gravimetrically through weighing the shale sample before and after exposure to salt solutions. Precisely, this technique relies on measuring the shale’s weight modifications under various situations. A saturated shale sample with a diameter of 0.75” is first weighed. Afterwards, the sample is placed in the oven at 180 for 24 hours and then weighed again. We believe 24 hours is sufficient for the elimination of free water found within the native sample pores. These weights are set to be the preserved native sample in air, and weight of preserved native sample after drying, respectively. Shale samples are then immersed in salt solutions of various concentrations and weighed. Finally, the altered sample is positioned in the oven to measure dry altered sample. By using the subsequent equations, it was possible to quantify and separate the amount of water and ions that may have exchanged as shale interacted with salt solutions. These equations help in analysing the main source of shale swelling since it clearly quantifies both ions and water movement into or out of shale.

\[
M_{\text{water}}\% = \left(\frac{M_{\text{imm}} - M_{\text{imm}(d)} - M_{\text{na}} - M_{\text{nd}}}{M_{\text{na}}}\right) \times 100\% \quad (1)
\]
\[
M_{\text{ions}}\% = \left(\frac{M_{\text{imm}(d)} - M_{\text{nd}}}{M_{\text{na}}}\right) \times 100\% \quad (2)
\]

Where:
- \(M_{\text{na}}\): Weight of preserved native sample in air.
- \(M_{\text{nd}}\): Weight of preserved native sample after drying.
- \(M_{\text{imm}}\): Altered weight of sample after immersion in salt solutions.
- \(M_{\text{imm}(d)}\): Dry weight of altered sample after immersion in salt solutions.
- \(M_{\text{water}}\%\): Weight of water gained or lost from shale.
- \(M_{\text{ions}}\%\): Weight of ions gained or lost from shale.

<table>
<thead>
<tr>
<th>X-Ray</th>
<th>Shale (I)</th>
<th>Shale (II)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Quartz</td>
<td>13.8</td>
<td>15.2</td>
</tr>
<tr>
<td>Feldspar</td>
<td>1.8</td>
<td>4.1</td>
</tr>
<tr>
<td>Calcite</td>
<td>0</td>
<td>1.7</td>
</tr>
<tr>
<td>Dolomite</td>
<td>0</td>
<td>1.8</td>
</tr>
<tr>
<td>Pyrite</td>
<td>0.9</td>
<td>1.4</td>
</tr>
<tr>
<td>Siderite</td>
<td>0</td>
<td>1.5</td>
</tr>
<tr>
<td>Total Clay</td>
<td>77</td>
<td>74.3</td>
</tr>
<tr>
<td>Chlorite</td>
<td>n/a</td>
<td>3.5</td>
</tr>
<tr>
<td>Kaolinite</td>
<td>n/a</td>
<td>6.8</td>
</tr>
<tr>
<td>Illite</td>
<td>n/a</td>
<td>7.49</td>
</tr>
<tr>
<td>Smectite</td>
<td>15.9</td>
<td>22.4</td>
</tr>
<tr>
<td>Mixed Layer</td>
<td>21.6</td>
<td>34.18</td>
</tr>
</tbody>
</table>

Table 1: Minerals make up of shales I and II.
Table 2: Petrophysical & chemical properties of shales I and II.

<table>
<thead>
<tr>
<th>Property</th>
<th>Shale I</th>
<th>Shale II</th>
</tr>
</thead>
<tbody>
<tr>
<td>Quartz</td>
<td>13.8</td>
<td>15.2</td>
</tr>
<tr>
<td>Water Content (%)</td>
<td>10.2</td>
<td>10.09</td>
</tr>
<tr>
<td>Water Activity</td>
<td>0.98</td>
<td>0.96</td>
</tr>
<tr>
<td>Porosity (%)</td>
<td>17.3</td>
<td>12.8</td>
</tr>
<tr>
<td>Permeability (mD)</td>
<td>6.1</td>
<td>3.7</td>
</tr>
<tr>
<td>CEC (ml. eq/100 gm)</td>
<td>14.8</td>
<td>24.7</td>
</tr>
<tr>
<td>Burial Depth (ft)</td>
<td>6,320</td>
<td>8,310</td>
</tr>
</tbody>
</table>

Table 3: Salt solutions type and concentration test matrix.

<table>
<thead>
<tr>
<th>Salt Type</th>
<th>Concentration w/w %</th>
</tr>
</thead>
<tbody>
<tr>
<td>NaCl</td>
<td>1  4  8  12  16  20  24</td>
</tr>
<tr>
<td>KCl</td>
<td>1  4  8  12  16  20  24</td>
</tr>
<tr>
<td>CaCl₂</td>
<td>1  4  8  12  16  20  24</td>
</tr>
</tbody>
</table>

Figure 2: Corresponding water activity for NaCl, KCl and CaCl₂ solutions at each concentration.
Results and discussion

Interaction of KCl of multiple concentrations with shale I and II

An illustration of the swelling/shrinkage profile for shale I during interaction with 1%, 4%, 8%, 12%, 16%, 20%, and 24% w/w KCl solutions is shown in Figure 4. Construction of such profiles is made after conducting the linear expansion test. It is observed that the exposure of shale I to 1%, 12%, 16%, 20%, and 24% w/w KCl solutions caused shale I expansion. On the other hand, exposure of shale I to 4% and 8% KCl solution resulted in shrinkage of shale I. The water activity of 4% and 8% KCl solution is 0.982 and 0.963, respectively. Indeed, the flow of water is in the direction of lower activity regions i.e., the KCl solutions, resulting in a shrinkage of shale I. Hence, shale I shrinkage is expected since it has a water activity higher than the 8% KCl solutions while it is approximately equal to 4% w/w KCl solution. The exposure of the shale I to 1 w/w% KCl solution resulted in a slight swelling of the shale with a volumetric change of 0.25%. Shale I has a water activity of 0.98 whilst the 1% KCl solution had a 0.996 water activity. Such behaviour, however, is expected since the salinity of shale I is higher than that of the 1% KCl solution causing the water to flow from a low concentration solution (KCl solution) to the higher one (shale I). Conversely, it was observed that a 4% KCl solution (2) caused the shale to shrink slightly, as water activities are almost identical, with a volumetric change of -0.075%. Similarly, the shrinkage of shale I occurred during interaction with 8% KCl solution with a volumetric change of -0.51% of the original volume of shale I am resulting in the greatest shrinkage. This indicates that the shrinkage volume is due to higher water activities difference between the shale sample and KCl solution which could result in higher swelling/shrinkage volumes. Shale I shrinkage when interacted with 4% and 8% KCl solutions is due to chemical osmosis and may indicate that the amount of water extracted out of shale I exceeds the amount that has flown into shale I am resulting in the domination of chemical osmosis as it is largely dependent on pore fluid’s water activity.

However, the 12% (aw = 0.942), 16% (aw = 0.918), and 20% (aw = 0.892), 24% (aw = 0.862), KCl solutions resulted in the swelling of shale I with a volumetric change of 0.85%, 1.05%, 1.33%, and 1.71%, respectively. At this point, it is observed that the addition of salts to KCl solution exceeding 8 w/w% did not cause shale I to shrink but rather expand although the water activities of these solutions are much lower than that of shale I. This swelling could be attributed to the effect of ionic diffusion flow from KCl solution to shale I. Additionally, the imbalance of the ionic concentrations causes the infiltration of associated water that surrounds ions as a result of solutes diffusion, i.e., diffusion osmosis. Diffusion osmosis is common owing to

Figure 3: A standard linear expansion device.
the excessive addition of salts which seems to be the case here. When the net impact of ionic diffusion overcomes the impact of chemical osmosis, shale expansion occurs. Figure 5 shows the water and ions uptake into Shale I when interacted with KCl solutions of variable concentrations. This is used to identify the main source of shale I’s swelling. Results show that as we add more salts to the KCl solution, the ions uptake percentage gets higher while the water uptake percentage gets lower until we reach a salt concentration of 8%, in which any more addition of salts exceeding this value will have an adverse effect on the process of water extraction out of shale I.

Demonstration of swelling/shrinkage profile for shale II as a function of 1%, 4%, 8%, 12%, 16%, 20%, and 24% w/w KCl solutions is shown in Figure 6. The interaction between shale II and 1 w/w% KCl solution resulted in a higher swelling of shale II when compared to shale I, with a volumetric change of 0.45%. Conversely, it was observed that a 4% KCl solution caused the shale to shrink with a volumetric change of -0.2%. Similarly, the greatest shrinkage of shale II occurred during interaction with 8% KCl solution with a volumetric change of -0.85% of the original volume of shale II. However, the interactions of the 12%, 16%, 20%, and 24% KCl solutions with shale II show the same trend seen in shale I, and they resulted in the swelling of shale II with a volumetric change (swelling) of 1.05%, 1.27%, 1.64%, and 1.85%, respectively. Here, we see that more addition of salts to KCl solution did not cause shale II to shrink but rather expand although the water activities of these solutions are much lower than that of shale II. This swelling could be attributed to the imbalance of the ionic concentrations causes the infiltration of associated water that surrounds ions as a result of diffusion osmosis as the net impact of ionic diffusion is overcoming the impact of chemical osmosis in shale II. Notably, the observed behaviour of shale II when interacted with KCl solutions is similar to that of shale I. Shale II, however, has higher swelling rates than that of shale I as water and ions uptake (%) of shale II is higher as shown in Figure 7. Differences in volumetric swelling profile between shale I and II when interacted with KCl solutions of the same concentrations can be justified since the total active clay content in shale II is higher than shale I’s. Shale I have lower active clays than shale II. The total sum of mixed layer and smectite in shale I and II is 37.5 and 56.58, respectively. These active layers are swelling clays that include montmorillonite which is a subclass of smectite clay that lead to water adsorption and hence swelling of shale. This sheds light on the effect of active clay content in addition to the cation exchange capacity on how effective chemical osmosis can get in terms of shale’s wellbore stability. It is also observed for all interactions of KCl solutions of different concentrations with shale I and II that the concentration gradient (linear swelling profile) is almost flat after 180 minutes. This trend is triggered by the balance between chemical potential and concentration gradient of shale that resulted in the equalization of water and ions exchange between the shale sample and the KCl solution and hence a semi-constant swelling profile is detected.

Figure 4: Swelling profile for shale I as function of KCl concentrations.
Figure 5: Water and ions uptake into shale I when interacted with KCl solutions of variable concentrations.

Figure 6: Swelling profile for shale II as a function of KCl concentrations.

Figure 7: Water and ions uptake into shale II when interacted with KCl solutions of variable concentrations.
Interaction of NaCl of multiple concentrations with shale I and II

Figure 8 shows the swelling/shrinkage profile for shale I during interaction with 1%, 4%, 8%, 12%, 16%, 20%, and 24% w/w NaCl solutions. We observe that the exposure of shale I to 1%, 12%, 16%, 20%, and 24% w/w NaCl solutions result in shale I swelling. Conversely, exposure of shale I to 4% and 8% NaCl solution resulted in shrinkage of shale I as the flow of water seems to be in the direction of lower activity regions i.e., the NaCl solutions, resulting in a shrinkage of shale I. The water activity of 4% and 8% NaCl solution is 0.977 and 0.95, respectively. Hence, shale I shrinkage is expected since it has a water activity higher than the 4% and 8% NaCl solutions. The exposure of the shale I to 1 w/w% NaCl solution resulted in a slight swelling of the shale with a volumetric change of 0.31%. Shale I have a water activity of 0.98 whilst the 1% NaCl solution had a 0.994 water activity. This behaviour is expected since the salinity of shale I is higher than that of the 1% NaCl solution causing the water to flow from a low concentration solution (NaCl solution) to the higher one (shale I). Conversely, it was observed that a 4% NaCl solution (\(a_w = 0.977\)) caused the shale to shrink slightly with a volumetric change of -0.098%. Similarly, the shrinkage of shale I occurred during interaction with 8% NaCl solution (\(a_w = 0.95\)) with a volumetric change of -1.1% of the original volume of shale I am resulting in the greatest shrinkage. This indicates that the shrinkage volume is due to higher water activities difference between the shale sample and NaCl solution which could result in higher swelling/shrinkage volumes. Shale I shrinkage when interacted with 4% and 8% NaCl solutions is due to the fact that the net amount of water extracted out of shale I overcomes the amount that has flown into shale I am resulting in the domination of chemical osmosis. However, the 12% (\(a_w = 0.919\)), 16% (\(a_w = 0.883\)), 20% (\(a_w = 0.839\)), and 24% (\(a_w = 0.788\)) NaCl solutions resulted in the swelling of shale I with a volumetric change of 0.75%, 1.1%, 1.85%, and 2.45%, respectively. At this stage, it is observed that salts addition to NaCl solution exceeding 8 w/w% did not caused shale I to shrink although the water activities of these solutions are much lower than that of shale I. This swelling could be due to the effect of ionic diffusion flow from NaCl solution to shale I. Furthermore, the ionic concentrations’ imbalance results in penetration of associated water that surrounds ions into shale because of diffusion osmosis. The impact of ionic diffusion seems to be overcoming the impact of chemical osmosis; hence shale expansion occurs. Figure 9 shows the water and ions uptake into shale I when interacted with NaCl solutions of variable concentrations. Results show that as we add more salts to the NaCl solution, the ions uptake percentage is increasing while the water uptake percentage is decreasing until we reach a salt concentration of 8%, in which any more addition of salts exceeding this value will have an adverse effect on the process of water extraction out of shale I while the ions uptake is still getting higher.

Swelling/shrinkage profile for shale II as a function of 1%, 4%, 8%, 12%, 16%, 20%, and 24% w/w NaCl solutions is displayed in Figure 10. The interaction between shale II and 1 w/w% NaCl solution resulted in a higher swelling of shale II when compared to shale I, with a volumetric change of 0.65%. Conversely, it was observed that a 4% NaCl solution caused the shale to shrink with a volumetric change of -0.23%. Similarly, the greatest shrinkage of shale II occurred during interaction with 8% NaCl solution with a volumetric change of -1.49% of the original volume of shale II.

However, the interactions of the 12%, 16%, 20%, and 24% NaCl solutions with shale II show the same trend seen in shale I, and they resulted in the swelling of shale II with a volumetric change (swelling) of 1.1%, 1.65%, 2.25%, and 3.1%, respectively. Although the water activities of these solutions are much lower than that of shale II, more addition of salts to the NaCl solution at this point did not cause shale II to shrink but rather expand. Apparently, the impact of ionic diffusion exceeds the impact of chemical osmosis in shale II.

Remarkably, the interaction of shale II with NaCl solutions is similar to that of shale I. Shale II, nevertheless, has higher swelling rates than that of shale I as water and ions uptake (%) of shale II is higher as shown in Figure 11. As previously explained, distinctions in volumetric swelling rates between shale I and II during interaction with NaCl solutions of the same concentrations is expected since the total active clay content in shale II is higher than that of shale I. It is observed for all interactions of NaCl solutions of different concentrations with shale I and II, as seen in Figure 8 and 10, that the rate of swelling/shrinkage of shale I is largely increasing in the first 80 minutes. The concentration gradient (linear swelling profile) is almost flat after 180 minutes. This trend is likely generated by the balance between chemical potential and concentration gradient of shale that resulted in the of water and ions exchange between the shale sample and the NaCl solution to slow down and hence a flat swelling profile is observed.
Figure 8: Swelling profile for shale I as function of NaCl concentrations.

Figure 9: Water and ions uptake into shale I when interacted with NaCl solutions of variable concentrations.
Figure 10: Swelling profile for shale II as function of NaCl concentrations.

Figure 11: Water and ions uptake into shale II when interacted with NaCl solutions of variable concentrations.
Interaction of CaCl₂ of multiple concentrations with shale I & II

Figure 12 illustrates the swelling/shrinkage profile for shale I during interaction with 1%, 4%, 8%, 12%, 16%, 20%, and 24% w/w CaCl₂ solutions. Shale I interaction with 1%, 4%, 12%, 16%, 20%, and 24% w/w CaCl₂ solutions has caused shale I to swell while its exposure to 8% CaCl₂ solution has resulted in shrinkage of shale I. The water activity of 8% CaCl₂ solution is 0.96, and consequently, shale I is expected to shrink since it has a water activity higher than the 8% CaCl₂ solution. Subjecting shale I to 1 w/w% CaCl₂ solution resulted in a volumetric swelling of the shale with a volumetric change of 0.74%. Shale I have a water activity of 0.98 whilst the 1% CaCl₂ solution had a 0.994 water activity. Since the salinity of shale, I is higher than that of the 1% CaCl₂ solution, the water will flow from a CaCl₂ solution to shale I. Furthermore, we notice that the 4% CaCl₂ solution has caused the shale to swell slightly with a volumetric change of -0.15%, which is opposite to what happened to shale I when interacted with 4% w/w KCl and NaCl solutions. On the other hand, the shrinkage of shale I was seen during its interaction with 8% CaCl₂ solution (a_w = 0.96) with a shrinkage change of -1.78% which was the only shrinkage that occurred to shale I. Moreover, the 12% (a_w = 0.932), 16% (a_w = 0.892), 20% (a_w = 0.837), and 24% (a_w = 0.767) CaCl₂ solutions resulted in the swelling of shale I with a volumetric change of 1.15%, 1.67%, 2.84%, and 2.94%, respectively. This swelling could be attributed to the ionic diffusion flow from CaCl₂ solution to shale I. Furthermore, the ionic concentrations’ imbalance results in penetration of associated water surrounding ions into shale because of diffusion osmosis that lead to shale swelling. Figure 13 represents the water and ions uptake into shale I when interacted with CaCl₂ solutions of different concentrations. It illustrates the fact that more addition of salts to the CaCl₂ solution lead to an increase in ions uptake percentage. As for the water uptake percentage, it is decreasing until we reach a salt concentration of 8%, in which any more addition of salts will have a negative effect on the process of water extraction out of shale I while the ions uptake is still getting higher. Figure 14 shows the swelling/shrinkage profile for shale II as a function of 1%, 4%, 8%, 12%, 16%, 20%, and 24% w/w CaCl₂ solutions. The interaction between shale II and 1 and 4 w/w% CaCl₂ solution resulted in a higher swelling of shale II when compared to shale I, with a volumetric change of 0.95% and 0.33%, respectively. Conversely, the shrinkage of shale II occurred during interaction with 8% CaCl₂ solution with a volumetric change of -1.51% which is a lower shrinkage rate when compared to shale I’s interaction with the same CaCl₂ concentration.

On the other side, the interactions of the 12%, 16%, 20%, and 24% CaCl₂ solutions with shale II resulted in the swelling of shale II with a volumetric change of 1.64%, 2%, 2.65%, and 3.45%, respectively. The rate of swelling/shrinkage of shale II is fast in the first 100 minutes as seen in Figure 14. After 180 minutes, the linear swelling profile is flat. This trend is likely generated by the balance between chemical potential and concentration gradient of shale that resulted in the of water and ions exchange between the shale sample and the CaCl₂ solution to slow down resulting in a flat linear swelling profile. It is noted that the interaction of shale II with CaCl₂ solutions is similar to that of shale I but, nevertheless, shale II has higher swelling rates than that of shale I as water and ions uptake (%) of shale II is higher as shown in Figure 15. Final swelling profile for shale I & II as a function of different salt solutions of different concentrations is summarized in Figure 16, clearly showing the critical salt concentration.
Figure 13: Water and ions uptake into shale I when interacted with CaCl2 solutions of variable concentrations.

Figure 14: Swelling profile for shale II as function of CaCl2 concentrations.
Figure 15: Water and ions uptake into shale II when interacted with CaCl₂ solutions of variable concentrations.

Figure 16: Final swelling profile for shale I & II as a function of different salt solutions with different concentrations in w/w%.

Conclusions and recommendation
It is concluded from this work that:
1) The critical salt concentration of KCl, NaCl, and CaCl₂ when interacted with shale I and II is 8 w/w% concentration. Beyond this value, the chemical osmosis has become ineffective, and expansion of shale will be attributed to the ionic diffusion into shale.
2) Chemical osmosis is a reliable method for water extraction out of shale and improving its mechanical stability, as long as we do not exceed the critical saturation concentration value.
3) It is concluded that when salt concentration of KCl, NaCl, and CaCl₂ exceeds 8 w/w%, ionic diffusion and diffusion osmosis will adversely affect the shale’s mechanical and chemical stability.
4) The highest shrinkage rates of shale I and II occurred when it interacted with 8 w/w% CaCl₂ solution while the highest swelling rates occurred during interaction with 24 w/w% CaCl₂ solution.
5) The higher the active clay content, the higher the rate of swelling/shrinkage. In this work, shale II has higher active clay content.
6) The relationship between the ions uptake percentage and the...
linear swelling rate of shale I and II is directly proportional. 7) The water uptake percentage is inversely proportional to the linear swelling rate of shale I and II until the critical saturation point is reached. Beyond this point, the water uptake becomes directly proportional to the linear swelling rate of shale I and II. 8) The impact of cation type on the stability of shale was clear as the cation type that makes up the salt was varied (Na+, K+, Ca+2) and the anion type was fixed. Since both cations and anions tend to diffuse into shale in the presence of a concentration gradient, it is recommended to vary the anion type to study the combined impact of cation and anion type on shale stability. 9) Higher water activity (lower salinity) of shale with respect to the drilling fluids can improve be used to improve shale stability though extracting water out of shale. 10) There is a strong relationship between the chemical potential and ionic concentration of the salt solutions. 11) When the net impact of ionic diffusion exceeds the impact of chemical osmosis, shale swelling occurs. On the contrary, when the net impact of chemical osmosis overcomes the impact of ionic diffusion, shrinkage occurs. 12) Extreme reduction of the water activity of drilling fluid by adding salt without proper design can be detrimental to shale stability. So proper design of salt solution is recommended to mitigate shale’s strength reduction with time.

Limitations and future work
This work experimentally presented that chemical osmosis is an operative mechanism for averting shale swelling and following wellbore instability problems. This mechanism depends on removing water out of shale particularly when the chemical potential (water activity) of shale is less than that of the aqueous solution. The transport of water out of shale, due to the existence a water activity gradient, may reduce the shale’s pore pressure, increase its strength and cause shale’s shrinkage, all of which leads to shale instability.

In spite of the advantageous role of chemical osmosis, another problem is created specifically when shale contacts water-based drilling fluids. Ions and their associated water clouds could enter shale when the ionic concentration of shale is less than that of the water-based drilling fluid. The movement of ions and their associated water into shale refutes the benefits that might be obtained from extracting water out of shale by chemical osmosis. The transfer of ions and water into shale could cause swelling, strength reduction, pore fluid composition changes and over all mechanical instability. In other words, manipulating the water activity of drilling fluids to extract water out of shale for wellbore stability purposes may not produce the anticipated results due to the invasion of ions along with their associated water. This was shown experimentally in our work by two well-designed tests: linear swelling test and gravimetric ions and water uptake test.

Although the swelling and gravimetric ions and water uptake tests showed that shale is a leaky semi-permeable membrane and that chemical osmosis could be jeopardized by ionic transport into shale, more work needs to be conducted to completely comprehend the influence of ions and their associated water on shale swelling and subsequent stability. Specifically, separating the impact of osmotic water (due to chemical osmosis) and associated water (due to diffusion osmosis) might help us quantify the impact of each water type of shale swelling. Furthermore, the impact of water transport on changes in the thickness of diffuse double layer of clay needs to be investigated. We believe that swelling is a direct result of the thickness of diffuse double layer. The impact of temperature on the transport of water and ions into shale needs to be examined to reflect in situ conditions. Lastly, the effect of anion type and concentration on shale swelling should be explored since both cations and anions tend to diffuse into shale in the presence of a concentration gradient.

Data Availability Statement
Some or all data, models, or code that support the findings of this study are available from the corresponding author upon reasonable request.

Compliance with Ethical Standards
The authors declare that there was no funding, grants, or other support was received for conducting this study. Moreover, the authors have no relevant financial or non-financial interests to disclose.

Reference


