



Swelling behavior of heavy crude oil in carbonated water at the presence of Na_2SO_4 and Mg_2SO_4

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Abstract

Carbonated water (CW) and low salinity (LS) injection processes are among the most widely investigated method during the past decade due to their unique advantages. Although the results revealed rather efficiency of these two methods, it is reported that using the combination of these two methods can introduce new insight about the smart and newly developed enhance oil recovery processes. Respect to these facts, the current work is aimed to investigate the swelling behavior of heavy crude oil in carbonated water at the presence of Na_2SO_4 and Mg_2SO_4 . In this way, one of the main effective mechanisms through the carbonated water injection which is swelling factor is examined in the presence of two different salts, namely, Na_2SO_4 and Mg_2SO_4 , under different pressures (500–4000 psi) and temperatures (30–80 °C). The results obtained in this investigation are compared with the previously reported results regarding the carbonated brine (CB) solutions consisted of different salts such as KCl, NaCl, CaCl_2 , and MgCl_2 with the same concentration of 15,000 ppm. The results not only reveal the possible mechanism behind the swelling factor variation, but also reveal that crude oil and ion type as well as temperature besides the solubility of CO_2 in aqueous phase can introduce tremendous influence on the mobility of CO_2 molecules and their partitioning from aqueous phase towards the oil phase. Also, the obtained results reveal that the presence of divalent ions can directly affect the crossover pressure toward lower values.

Keywords Carbonated brine · Swelling · Bond number · Crude oil · EORs

Introduction

Among the different possible EOR methods, gas injection is the most common method and using carbon dioxide (CO_2) is the most widely used gas due to several features and concerns existed regarding this gas. In detail, CO_2 injection not only can lead to suitable oil recovery but also injecting CO_2 into reservoir can manage the CO_2 release into environment through CO_2 capture which consequently reduces the global warming concern raised by existence of CO_2 in atmosphere (Abedini et al. 2014; Lashkarbolooki et al. 2018a). Besides the carbonated brine/water injection, low salinity water injection is the other innovative method for EOR purposes which mostly activates the wettability alteration of the rock surface, although it introduces influences on the other aspects of solution interface and rock surface properties. More recently, due to the advantages of these two methods, a new combination of these two approaches is proposed known as carbonated brine (CB) which utilizes advantages of both methods (Ahmadi et al. 2016; Mosavat and Torabi 2014; Sohrabi et al. 2012). Injection of

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carbonated water not only can lead to higher oil recovery, but also it can retard the CO₂ movement toward the surface from the deep of the reservoir resulted in more safe CO₂ storage and sequestration (Foroozesh et al. 2016; Mosavat and Torabi 2016). Unfortunately, although this proposed combination moved the researchers toward experimental investigations, no consistent results and mechanisms are proposed (Foroozesh and Jamiolahmady 2016; Riazi 2011; Shakiba et al. 2016). On the other hand, similar to the different areas of sciences, since measuring and experimentation for all the conditions and different types of crude oil is a tedious and expensive approach, modeling and simulation are highly recommended, but, unfortunately, no reliable simulation approach existed regarding this combination due to complexities of this method. For example, Sohrabi et al. (2011b) utilized the ECLIPSE software to simulate the carbonated water injection process, but only overestimated results were obtained. Moreover, Riazi et al. (2011a) proposed a mathematical based method to model the CO₂ mass transfer from brine toward crude oil as the main phenomenon during CB injection besides the other few mathematical equations proposed by different researchers to calculate the solubility of CO₂ in crude oil and its effect on the oil swelling factor and viscosity reduction no one leads to reliable results (Barclay and Mishra 2016; Simon and Graue 1965; Welker 1963). Besides the effect of CW on swelling factor, several researchers investigated the effect of different operating and non-operating parameters on possible IFT reduction of solutions comprised of CO₂ which revealed no significant IFT reduction using this method (about 5 mN/m reduction which is insufficient for effective enhanced oil recovery purposes) (Honarvar et al. 2017; Riazi and Golkari 2016; Yang et al. 2005). Also, the performed core flooding (Ahmadi et al. 2016; Foroozesh and Jamiolahmady 2016; Mosavat and Torabi 2014; Shakiba et al. 2016; Sohrabi et al. 2011b, 2012) and micromodel experiments (Riazi et al. 2011b; Sohrabi et al. 2011a) revealed that oil swelling due to CO₂ partitioning from aqueous phase toward oil phase is an effective parameter which can, consequently, introduce a reduction in oil viscosity during CW flooding (Mosavat et al. 2014). Moreover, Abedini et al. (2014) showed that the oil swelling factor of crude oil–CO₂ systems increases with the equilibrium pressure and then reduces with further increase in equilibrium pressure. Besides, Lashkarbolooki et al. (2018b) reported no clear and consistent pattern between CO₂ solubility in water and swelling of crude oils in the crude oil/CW system. Despite the important role of crude oil swelling during CW flooding, the effects of temperature and pressure on the swelling behavior of crude oil are still not well understood but certain. Moreover, in addition to the other effective parameters currently discussed through CW injection, Lashkarbolooki et al. (2017) have reported that crude oil type is the other crucial parameter determines

the efficiency of CW injection on tertiary oil recovery. In this way, Lashkarbolooki et al. (2018b) examined the possible effects of temperature and pressure on the swelling and Bond number behavior of heavy-acidic crude oil (HACO). Their results demonstrate that two different patterns would be observed regarding the effect of temperature and pressure on the swelling factor of HACO. They also concluded that there is a crossover pressure for swelling factor which leads to a dual effect on the swelling factor behavior. Besides the effect of CO₂ existence in aqueous solution, it has been well established that the presence of salinity can introduce positive effect on the aqueous solution for higher oil recovery efficiency (Austad et al. 2010; Lashkarbolooki et al. 2014; Ligthelm et al. 2009; McGuire et al. 2005; Morrow et al. 1998; Tang and Morrow 1997). Respect to the aforementioned shortcomings, it seems that experimental approach is the best way to find the efficiency of this new approach in the current status, especially the swelling factor, since this parameter is the most effective parameter among the possible effective parameters needs to a wide, accurate, and systematic investigation as a function of pressure and temperature especially at the presence of the sulfate ions existed in the formation brine limitedly investigated.

Regarding the aforementioned facts and shortcoming existed regarding both CW injection and aqueous solution modified by different ions and since no report existed on the effect of divalent ions of MgSO₄ and Na₂SO₄ concomitant with CW, the effect of ion type on the swelling behavior of crude oil in the presence of CB as a function of temperature and pressure has investigated in the current investigation. The worth mentioning point is that the current investigation is a complementary phase of previous study performed by Lashkarbolooki et al. (2019) regarding the systems fully saturated with CO₂ consisting of 15,000 ppm of NaCl, KCl, CaCl₂ and MgCl₂ (named CNaCl, CKCl, CCaCl₂, and CMgCl₂, respectively) at temperatures (*T*) of 30, 50, and 80 °C and pressures (*P*) of 500, 1000, 2000, and 4000 psi with especially focus on the two different salts including MgSO₄ and Na₂SO₄.

Materials and experimental procedure

Materials

A heavy-acidic crude oil (HACO) type as previously used by Lashkarbolooki et al. (2019) was provided from southern Iranian oilfields (total acid number of 1.5 mg KOH/g and API° of 21.5) and used as the sample crude oil. Besides, the used aqueous solutions for carbonated water preparation were consisted of two different salts of magnesium sulfate heptahydrate (MgSO₄·7H₂O) (MW of 246.48 g/mol) and sodium sulfate (Na₂SO₄) (MW of 142.04 g/

mol) with concentration of 15000 ppm [Merck, Germany (Purity > 99%) used without any further purification]. In addition, the carbonated water was prepared by CO₂, purchased from Abughadareh Co., Iran (purity > 99.9%).

Experimental procedure

Swelling and Bond number measurements

A previously validated and established high temperature–high pressure pendant drop interfacial tension equipment (Atiyeh Pouyandegan Exir Technologies Co. (Apex technologies), Arak, Iran) (see Fig. 1) is used to measure the swelling factor through the current investigation. In brief, it is possible to inject the bulk and drop at desired pressure and temperature in pendant or rising positions. Besides, the measuring chamber (total volume of 15 cc) is equipped with two sapphire sight glasses capable of the operator to monitor the inside content of the chamber using a CCD camera equipped with macro lens (Computar, Japan) which dispatches the images to online software. Besides the main chamber, a twin accumulator block existed which separately holds the bulk and drop phases to inject them into the measuring chamber. The temperature of both measuring chamber and accumulators is controlled using elements coupled with PT-100 thermocouple with accuracy of about 0.1 °C. In addition to temperature, the pressure of the system was monitored using a digital pressure transmitter (WIKA, Germany). After tuning the pressure and temperature and forming the drop at the tip of the nozzle, the images of the formed drop which were captured using CCD camera instantly measure the volumes of the drops which are capable of the operator to calculate the swelling factor ($SF = \frac{V_t}{V_0}$) and Δ

Bond number (Δ) where V_t is the crude oil drop volume at time t , V_0 is the crude oil drop volume at initial time, BN_t is the Bond number at time t , and BN_0 is Bond number at initial time.

Results and discussion

Evaluation of swelling and Bond number behaviors of HACO/CNa₂SO₄ and CMgSO₄

In the first stage, the swelling and Δ Bond number variations of the first binary system namely HACO/CNa₂SO₄ as a function of time and pressure (500–4000 psi) were examined under different isotherms of 30, 50, and 80 °C (see Fig. 2). Before any measurements, to ensure about the effect of ions on the swelling factor, several measurements were carried out in the absence of CO₂ in the aqueous phase which revealed no swelling can be observed for the system contain no dissolved CO₂. A glance into the results depicted in Fig. 2 reveals that at a specific isotherm, an increase in the pressure may lead to an increase in the swelling factor. In more details, the results demonstrated that for isotherm of 30 °C, increasing the pressure from 500 psi to 4000 psi enhances the swelling factor from 1.03 to 1.11 which means three times larger swelling factor. In addition, from the closer examination in the results depicted in Fig. 2, one can conclude that there is a direct relation between the temperature and swelling factor. In details, increasing the temperature from 30 °C to 80 °C for pressure of 4000 psi, an increase in the swelling factor from 1.11 to 1.24 which means two times enhancement in swelling factor would be obtained. Moreover, comparing the swelling factor of two pressures including 500 and 1000 psi, one can conclude that as the temperature enhances, the swelling factor of these two isobars are getting close to each other, while for pressures of 2000 and 4000 psi, the trend is reversed.

The overall observed swelling factor enhancement regardless of temperature and pressure for all the studied systems can be related to this fact that the presence of CO₂ in the aqueous solution can lead to partitioning of CO₂ toward the crude oil which consequently leads to swelling of the oil drop. In other words, it can be concluded that the swelling of the crude oil in the presence of CW occurs caused by the partitioning of CO₂ from the CW phase by diffusion and dissolution of CO₂ into the oleic phase. Furthermore, the higher swelling factor observed for higher pressures can be related to the direct relation existed between the pressure and capability of aqueous solution to dissolve the CO₂. In detail, as the pressure enhances, the aqueous solution capability to dissolve CO₂ enhances which means that higher CO₂ content would be available to penetrate into the oil drop which

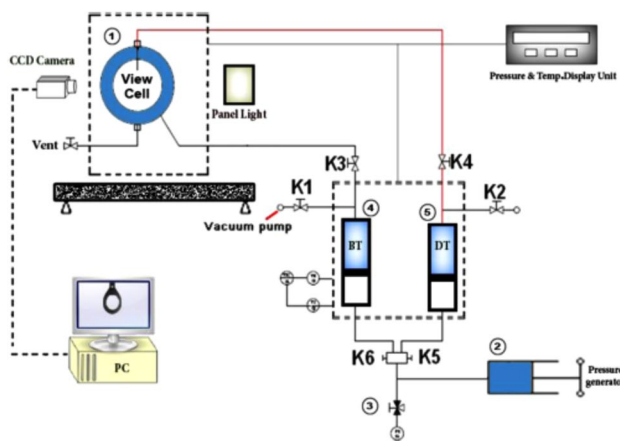
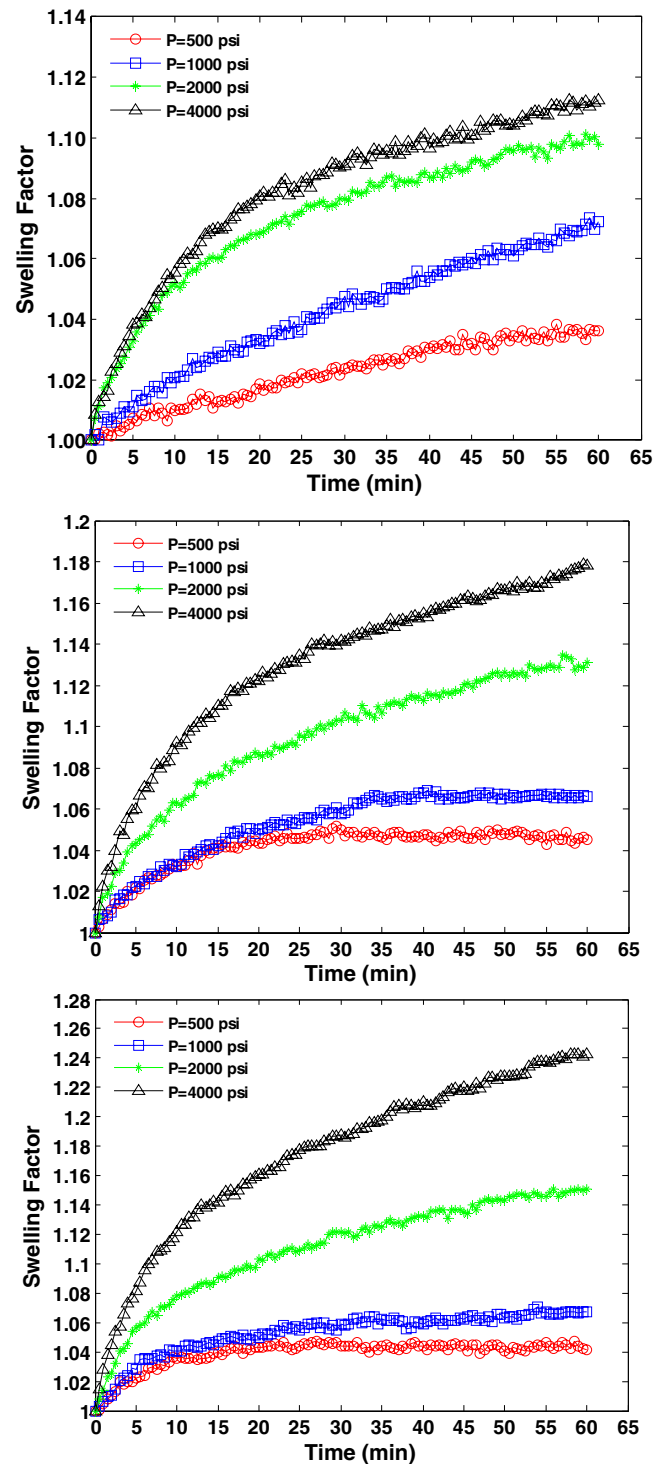


Fig. 1 The used IFT measurement Apparatus (1, view cell; 2, pressure generator; 3, pressure manometer; 4, bulk tank; 5, drop tank) (Zolghadr et al. 2013)

Fig. 2 Dynamic swelling factor of crude oil in the presence of 15,000 ppm of Na_2SO_4 at temperature of **a** $T=30^\circ\text{C}$, **b** 50°C , and **c** 80°C



directly leads to more swelling of oil drop. However, in the regard of temperature effect on the swelling factor, the situation is more complex, since temperature has a dual effect. In detail, enhancing the temperature can increase the molecular energy and movement of the molecules leads to higher probability of molecular packing into the interface, while it can lead to a reduction in CO_2 solubilization in aqueous solution,

since temperature has a reverse effect on the solubility of gases in water. Therefore, this is the net effect of these two competing factors, dictate if the temperature leads to larger swelling factor or not. Regarding this fact, it seems that for all of the studied temperatures, the first effect is dominant. However, closer examination in Fig. 2 also demonstrates that as the temperature enhances the swelling factor of two

isobars of 500 psi and 1000 psi getting closer to each other, while for pressures of 2000 psi and 4000 psi, this trend is different and temperature enhancement increases the difference existed between the swelling factors of these two isobars. In sum up, according to these findings, it can be concluded that the examined system of HACO/CNa₂SO₄ may have a cross over pressure of about 2000 psi, since the effect of temperature is different on the swelling factor variation for pressures above the 2000 psi.

In the second stage of this investigation, the effects of pressure and temperature on the Bond number ratio of the HACO/CNa₂SO₄ were examined (see Fig. 3). The obtained results revealed that for temperature of 30 °C, increasing the pressure led to a reduction in Bond number ratio for all the studied isobars. However, for the other two examined isotherms of 50 and 80 °C, the effect of pressure on the Bond number ratio is more complicated than 30 °C isotherm. In details, for isotherm of 50 °C, although for all of the examined pressures of 1000, 2000, and 4000 psi, the Bond number ratio is decreasing similar to the 30 °C isotherm; for pressure of 500 psi, two phases of variation can be observed.

In details, for this pressure and before period of 30 min, slight decreasing trend can be observed, while for period of after 30 min, the trend of variation is reversed. For a better clarification regarding the obtained results, the ultimate values for Bond number ratio and swelling factor are depicted in Fig. 4. Mining into the results depicted in Fig. 4 revealed that for CNa₂SO₄/HACO, there is a sharp crossover pressure of around 1000 psi.

In details, it is obvious that for the aforementioned system inverse swelling and Δ Bond number trends could be observed for two different regions which can be categorized as the below cross over pressure (BCP) and above the crossover pressure (ACP) regions. In the BCP, swelling of crude oil drop decreases with temperature, while in the ACP region, the trend get reversed [crossover pressure (P_{CO}) of about 1000 psi]. This observed trend can be related to the mechanism previously proposed by Lashkarbolooki et al. (2019) that in the first region (i.e., $P < P_{CO}$), CO₂ solubility in aqueous phase is the controlling factor, while in the second region (i.e., $P > P_{CO}$), mobility and loosening of the H Bond between water molecules have the dominant impact on partitioning of CO₂ (Lashkarbolooki et al. 2018b). In more details, there is a direct relation between gas dissolution in liquids and the interaction between the molecules of gas and liquid molecules. However, this is the formation of carbonic acid that led to dissolution of CO₂ in water which rise from the chemical structure and its ability to react with H₂O. The point regarding the CO₂ dissolution into the water is that no strong attraction would be obtained between these two chemicals which, as a consequence, the CO₂ molecules can easily come out of CW solution. The solubility of CO₂ in

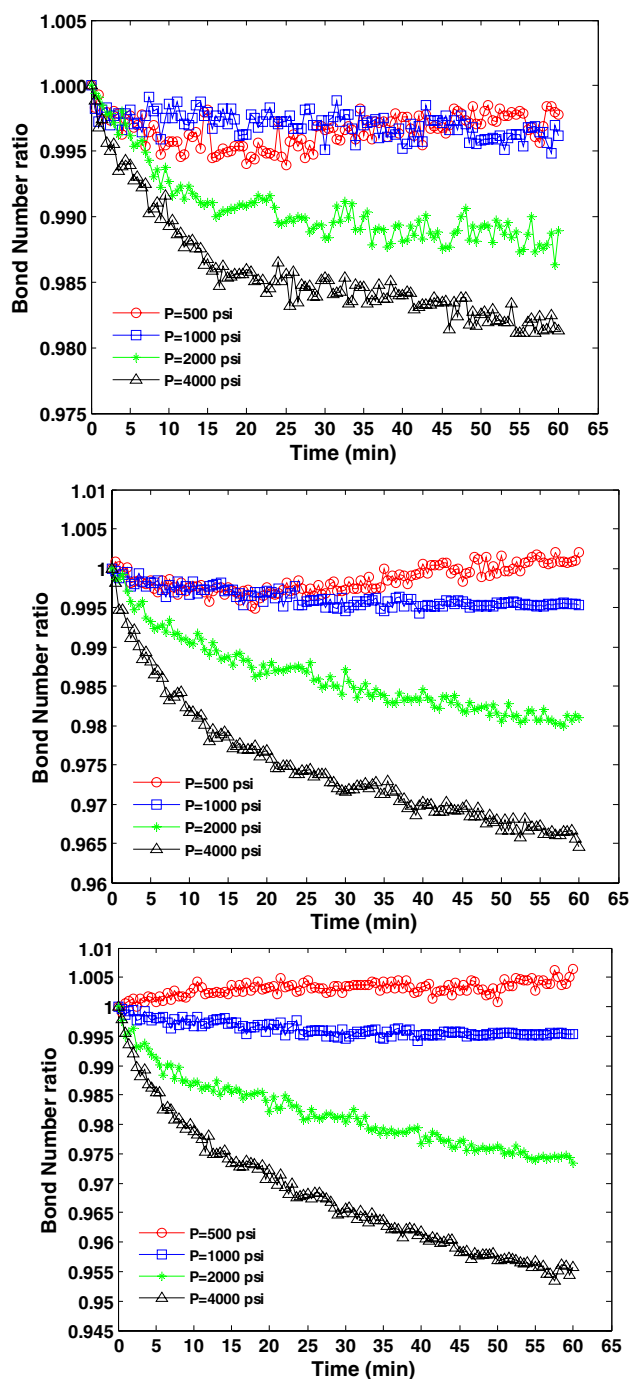


Fig. 3 Dynamic Bond number ratio of crude oil in the presence of 15,000 ppm of Na₂SO₄ at temperature of **a** $T=30$ °C, **b** 50 °C, and **c** 80 °C

DW increases when they have functional groups enables to form H Bonds with water. For CO₂ partitioning from CW phase to the crude oil phase, there is very little interaction between CO₂ particles; hence, no solute–solute interaction needs to be broken, while water–water interaction (i.e., H Bond) have to be broken in order to partition CO₂ from

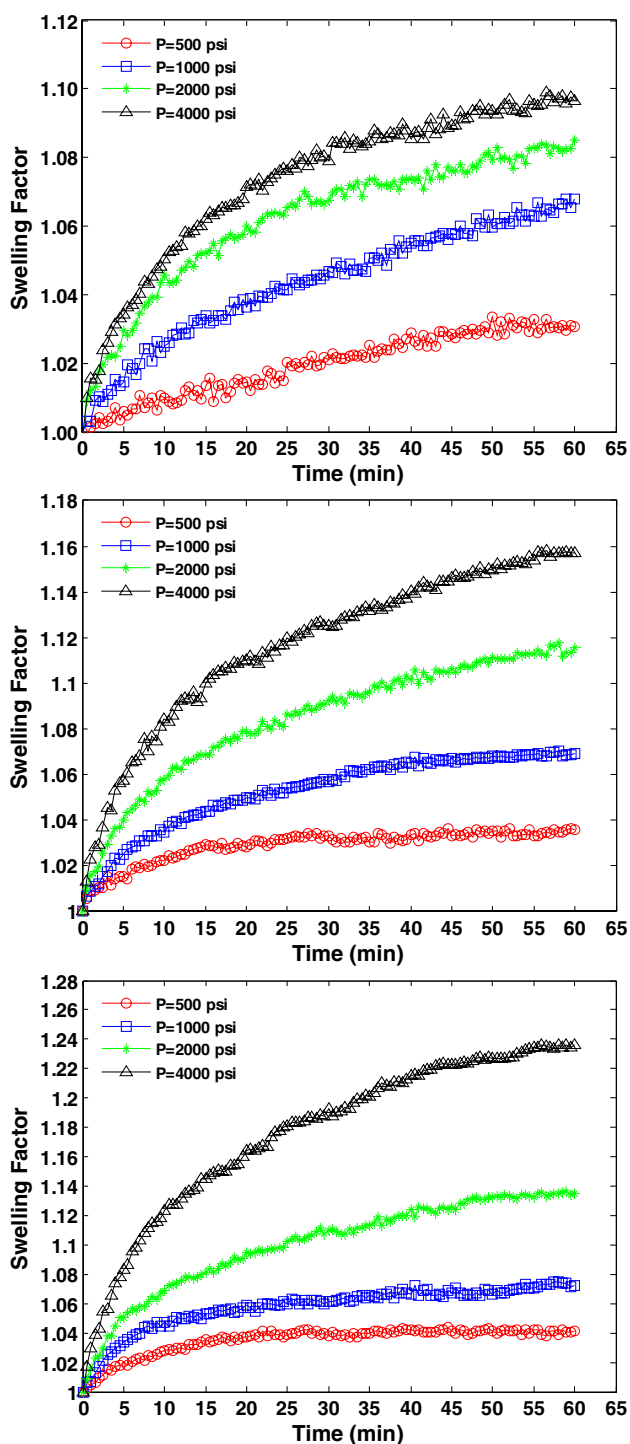


Fig. 4 Dynamic swelling factor of crude oil in the presence of 15,000 ppm of MgSO_4 at temperature of **a** $T = 30^\circ\text{C}$, **b** 50°C , and **c** 80°C

the aqueous phase to the crude oil phase (Lashkarbolooki et al. 2018b).

In the next stage of this investigation, dynamic swelling factor and Bond number ratio for carbonated water

consisted of 15,000 ppm of MgSO_4 under different pressures and temperatures were examined. Similar to the results obtained for Na_2SO_4 , as the pressure increases from 500 psi to 4000 psi, the swelling factor was increased for all the examined isotherms. However, comparing the results depicted in Figs. 2, 3, 4 and 5, demonstrated that the swelling factor as a function of pressure is lower for MgSO_4 compared with Na_2SO_4 . In addition, the results revealed that the effect of temperature for swelling factor reduction for pressure of 2000 psi is sharper than that observed for Na_2SO_4 although the effect of this parameter is less than that obtained for Na_2SO_4 . In addition, it seems that not only in the case of MgSO_4 respect to Na_2SO_4 , the swelling factor is less than that observed for Na_2SO_4 , but also the observed slope is moderate compared with the slope of Na_2SO_4 . The interesting point is that for both cases of MgSO_4 and Na_2SO_4 , even after 1 h measurement from the beginning of the test, some drop volume did not reach the equilibrium condition. This phenomenon is more considerable at higher temperature and pressure conditions (see Figs. 2 and 4). As can be seen from Figs. 2 and 4, at isobaric of 4000 psi, an increase in temperature from 30°C to 80°C increases the swelling factor tremendously with no obvious plateau which can be due to this fact that the CO_2 transfer between the aqueous phase and the oil phase is slow and equilibrium state could not be reached (Fig. 6).

In the next stage, the variation of Bond number ratio as a function of pressure at different isotherms was investigated for MgSO_4 which was rather different from that observed for Na_2SO_4 . In details, for pressures of 2000 psi and 4000 psi, the Bond number ratio variation is below 1 for all the examined isotherms similar to Na_2SO_4 , while for pressures of 1000 psi and 500 psi, the Bond number ratio shift toward 1 and even getting larger than 1 for pressure of 500 psi and temperatures higher than 30°C . For a better clarification, similar to the Na_2SO_4 , the ultimate values for swelling factors and Bond number ratio values as a function of temperature and pressure are depicted in Fig. 7. A close examination into this figure can illustrate that although no cross over pressure can be observed for swelling factor, a sharp and obvious cross over pressure of about 1000 psi can be observed considering the ultimate Bond number ratio values. Similar to those obtained for Na_2SO_4 , the observed trend can be related to the mechanism previously proposed by Lashkarbolooki et al. (2019) that in the first region (i.e., $P < P_{\text{CO}}$), CO_2 solubility in aqueous phase is the controlling factor, while in the second region (i.e., $P > P_{\text{CO}}$), mobility and loosening of the H Bond between water molecules has the dominant impact on partitioning of CO_2 (Lashkarbolooki et al. 2018b). Similarly, Zhao et al. (2015) revealed that CO_2 solubility in the aqueous phase shows different trends in

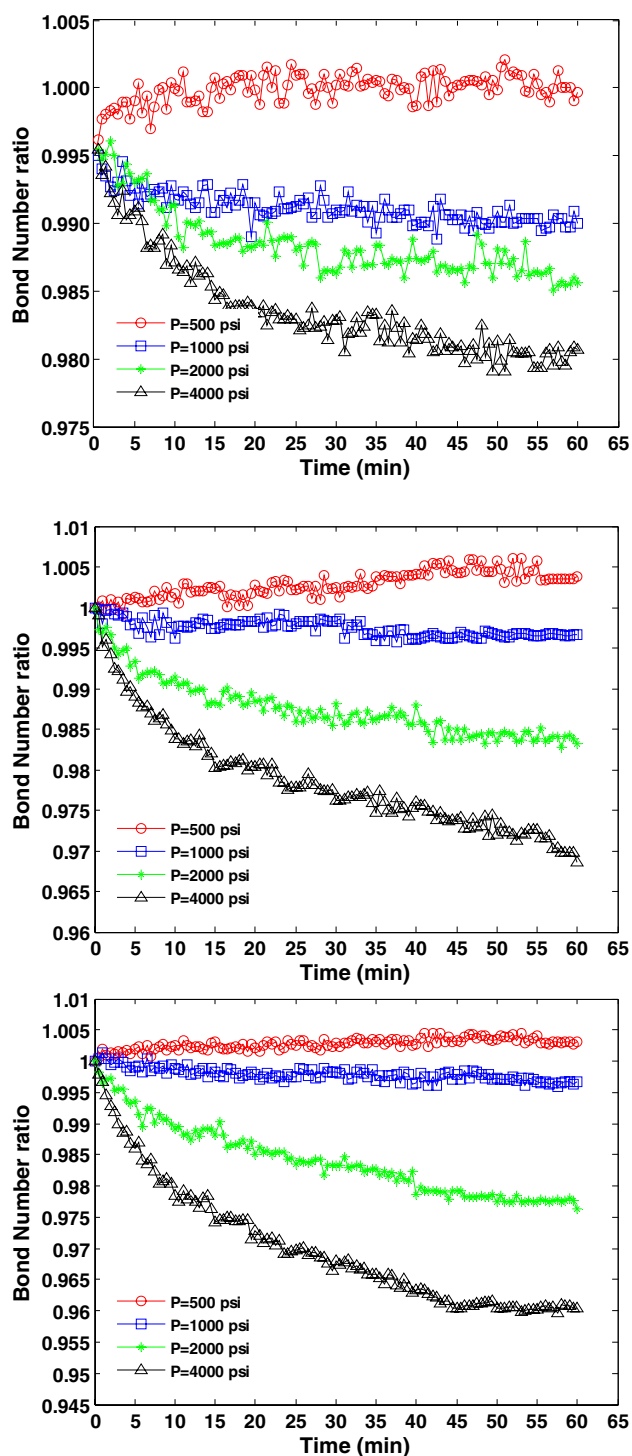


Fig. 5 Dynamic Bond number ratio of crude oil in the presence of 15,000 ppm of MgSO_4 at temperature of **a** $T=30\text{ }^\circ\text{C}$, **b** $50\text{ }^\circ\text{C}$, and **c** $80\text{ }^\circ\text{C}$

two distinct regions. In region I, the CO_2 solubility in the aqueous phase decreases monotonically as a function of temperature, reversely at a given pressure in region II, the CO_2 solubility increases (Duan et al. 2006).

Comparison between the studied carbonated brines and carbonated water

In the next stage of this investigation, the effect of salt dissolution in the carbonated water on the swelling factor and Bond number ratio was investigated. In this regard, the obtained results of these two parameters for both carbonated brine (CB) and carbonated water (CW) are depicted in Figs. 8 and 9. Mining into the results depicted in these two figures demonstrated that dissolution of salts regardless of which salt is dissolved led to two different phenomena including reduction in swelling factor and enhancement in Bond number ratio and shifting the cross over pressure toward lower values. In details, it seems that dissolution of salts into the CW had a undesired effect on the swelling factor which may be due to this fact that the presence of ions on the carbonated water may form a strong bonding to the CO_2 preventing the easier movement of CO_2 toward the oil interface to be dissolved in the oil drop.

This observed trend is in agreement with the results reported by Lashkarbolooki et al. (2019) regarding the effect of ion types on the swelling factor. They have reported that existence of monovalent ions, i.e., Na^+ and K^+ in the carbonated brine may lead to an enhancement on the crossover pressure, while the existence of divalent ions such as Ca^{2+} and Mg^{2+} can reduce the crossover pressure of CCaCl_2 and CMgCl_2 . The worth mentioning point is that Lashkarbolooki et al. (2019) have reported that the observed trend can be related to the direct effect of salts on the solubility of CO_2 in the aqueous solutions. However, in the current study, considering the solubility of CO_2 in the brine solutions using Duan model (Duan et al. 2006) (see Fig. 10), it is completely obvious that the solubility of CO_2 in the MgSO_4 is larger than the value of CO_2 solubility in Na_2SO_4 , but the swelling factor for MgSO_4 is lower than Na_2SO_4 . This observed trend can be related to this fact that although the presence of MgSO_4 may lead to higher solubility of CO_2 in the brine solution compared with the Na_2SO_4 , the bonding between the MgSO_4 and CO_2 in the aqueous solution is stronger than Na_2SO_4 prevents easy movement of CO_2 molecule into the oil drop surface and consequent swelling. In other words, stronger bonding between CO_2 and MgSO_4 which led to higher solubility of CO_2 compared with the Na_2SO_4 act as a preventing parameter reduces the swelling factor.

Conclusions

In the current experimental investigation, the effect of pressure and temperature on the swelling factor and Bond number ratio of carbonated water including 15,000 ppm of MgSO_4 and Na_2SO_4 were investigated. Considering the

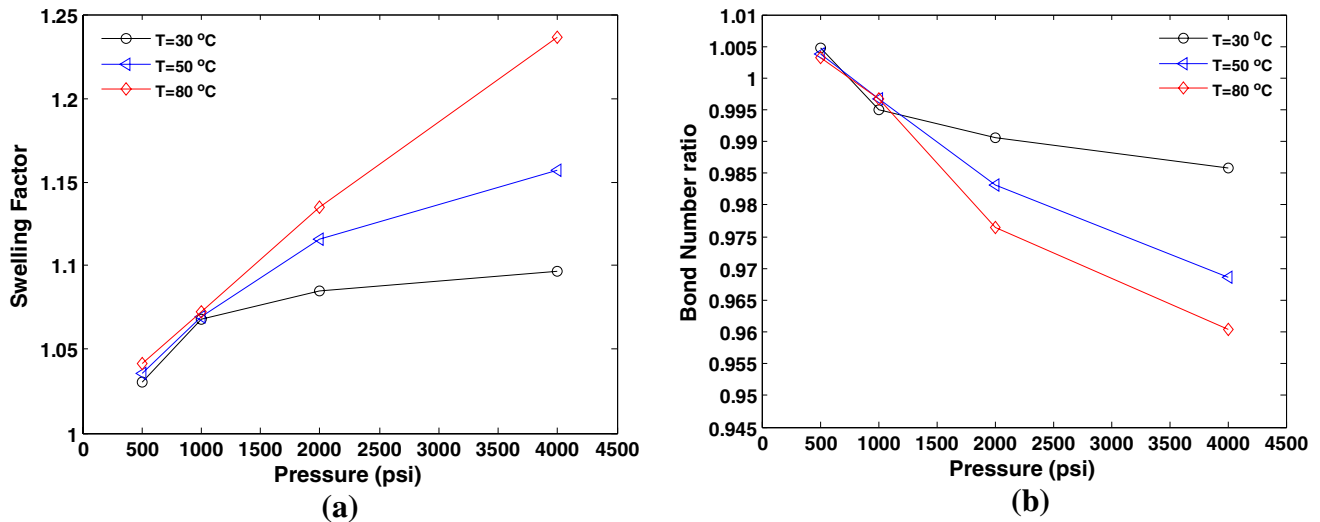


Fig. 6 a Swelling factor and b Bond number ratio after 1 h at different temperature and pressure condition for case of 15,000 ppm of $MgSO_4$

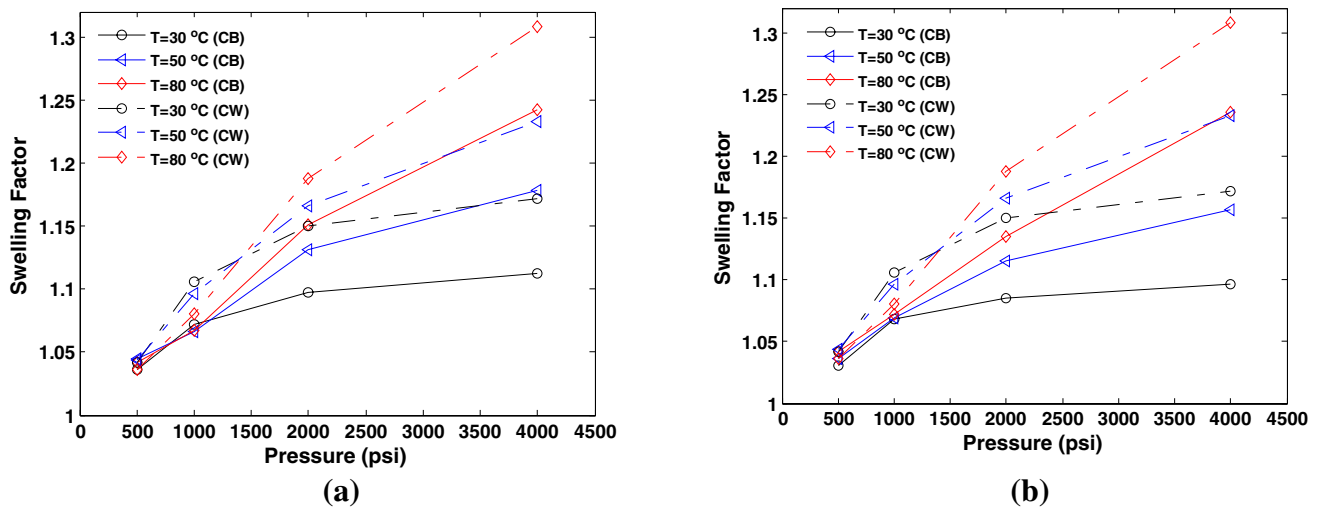


Fig. 7 Comparison of swelling factor in the presence and absence of a Na_2SO_4 and b $MgSO_4$ at different temperature and pressure condition

performed measurements, one can conclude the following outcome:

- Pressure and temperature can introduce direct effects on the swelling factor and Bond number ratio. Pressure has a simple and direct relation on the swelling factor, since it directly affects the solubility of CO_2 in the aqueous solution, while the effect of temperature was more complicated.
- Regarding the examined pressure and temperatures, two different trends were observed for the effects of these two operational parameters called cross over pressure. Crossover pressure point was defined as a point which below and above this point, two different trends

on the swelling factor and Bond number ratio can be observed regardless of the used salts. This point which was about 1000 psi revealed that for the pressures lower than this value (i.e., $P < P_{CO}$), CO_2 solubility in the aqueous phase is the dominant parameter dictates the swelling factor is increasing or decreasing, while in the region which the pressure is larger than this point (i.e., $P > P_{CO}$), mobility and loosening of H bond between water molecules are the most effective and dominant factors which tremendously affect the partitioning of CO_2 into crude oil phase.

- The obtained results demonstrated that the presence of salts can introduce direct effect on the swelling factor of HACO.

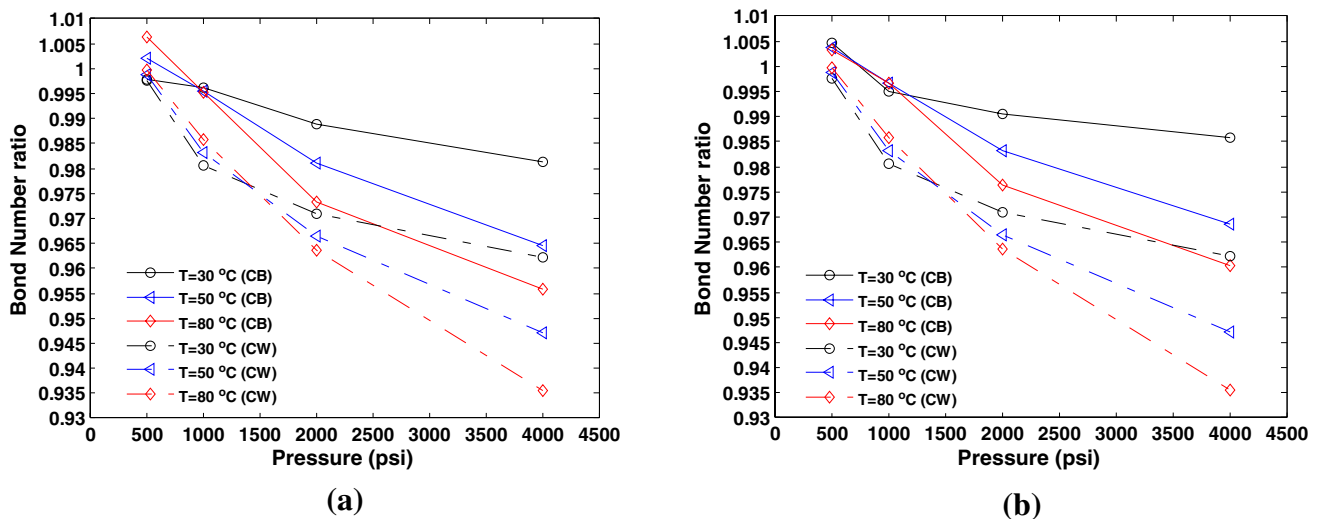


Fig. 8 Comparison of Bond number ratio in the presence and absence of **a** Na_2SO_4 and **b** MgSO_4 at different temperature and pressure condition

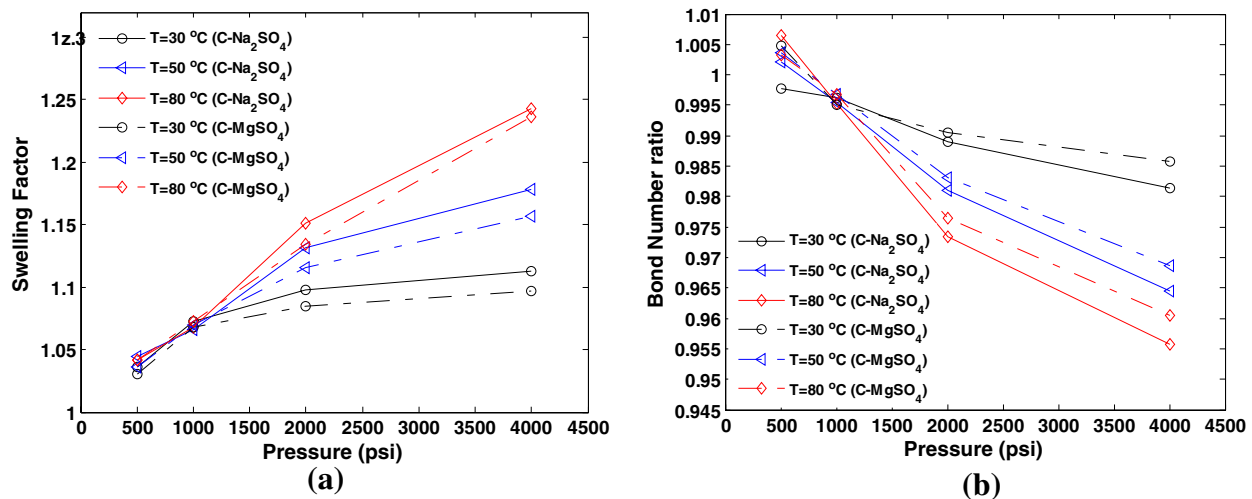


Fig. 9 Comparison of **a** swelling factor and **b** Bond number ratio in the presence of 15,000 ppm of Na_2SO_4 and 15,000 ppm of MgSO_4 at different temperature and pressure condition

- The presence of divalent ions can significantly affect the cross over pressure and shift this value toward lower values from 1500 psi to 1000 psi which means 50% reduction in crossover pressure point.
- The presence of divalent ions, especially Na_2SO_4 , may retard the solubility of CO_2 in the brine solution. However, the presence of MgSO_4 may lead to more undesired effect on the swelling factor due to stronger bonding between the MgSO_4 and CO_2 which prevent easier movement and partitioning of CO_2 on the interface and consequently reduces the swelling factor compared with the solution comprised of Na_2SO_4 .
- Ion type, temperature and CO_2 content of aqueous phase showed remarkable effect on the mobility of CO_2 molecules and their partitioning from aqueous phase towards oil phase and consequently on the swelling of crude oil during carbonated brine injection.

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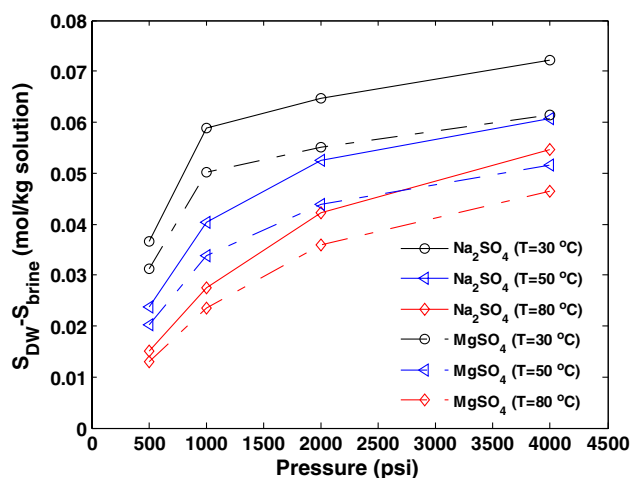


Fig. 10 Differences between solubility values of CO₂ in the presence and absence of 15,000 ppm of Na₂SO₄ and MgSO₄ at different temperature and pressure conditions obtained based on Duan model (Duan et al. 2006)

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