

An Investigation of Oil Spreading Coefficient in Carbonated Water+ Gas + Oil System: an Experimental Study in an Iranian Asphaltenic Oil Reservoir

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Abstract

To provide supplementary oil recovery after the primary and secondary processes, enhanced oil recovery (EOR) techniques are introduced. Carbonated water injection (CWI) as an EOR method can improve sweep efficiency and the risk of gas leakage. On the other hand, the interfacial tension (IFT) is one of the key factors which can affect fluid displacement during the process of CWI greatly. Therefore, the analysis of the IFT on an oil-carbonated water-CO₂ system is vital. In this paper, the interfacial interactions of binary systems of asphaltenic crude oil (ACO), carbon dioxide (CO₂), and carbonated water (CW) at different pressures and at two temperatures of 40 °C and 50 °C and their effects on the oil spreading in the water phase in the presence of gas are experimentally investigated. The IFT measurements were performed by axisymmetric drop shape analysis (ADSA) technique for the pendant/rising oil drop case. It is found out that the equilibrium interfacial tension (EIFT) of the two systems of crude oil-CO₂ and water-CO₂ is reduced almost linearly with pressure but increased with temperature. Moreover, the pressure has an increasing and decreasing effect on the water-oil and CW-oil IFT's respectively. However, temperature has a reverse effect for the both systems. Spreading coefficient (SC) concept would help better understand the oil recovery mechanisms and potential. The results show that SC curve has a minimum point value as a specific pressure, which increases with temperature. The presence of CO₂ in the water phase could strongly affect the oil spreading phenomenon through which oil recovery could be significantly enhanced.

Keywords: Interfacial Tension (IFT), Oil Spreading Coefficient (SC), Carbonated Water (CW) Enhanced Oil Recovery (EOR), Axisymmetric Drop Shape Analysis (ADSA) Technique, Iranian Asphaltenic Oil Reservoir

1. Introduction

Many of oil reservoirs in Iran are toward the end of their natural production age. This important topic means that the petroleum industry is required to use EOR methods. In recent years, it is well-known that interfacial properties play a main role in the efficiency of enhanced oil recovery (EOR) process

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(Bayat et al., 2016; Honarvar et al., 2017; Lashkarbolooki et al., 2014). The distribution of the reservoir fluids is also important in displacement efficiency and in the recovery of oil reservoirs using EOR techniques (Mani and Mohanty, 1997). To displace the trapped oil from reservoir rock, EOR methods are proposed and employed (Green and Willhite, 1998; Tzimas et al., 2005). Experimental studies and field applications of water alternating gas injection (WAGI), cyclic injection of carbonated water (CW) and CO₂, and CO₂-EOR processes to examine the potential of carbonated water injection as an injection process for EOR and CO₂ storage have been accomplished (Ahmadi et al., 2013; Riazi, 2011; Riazi, Sohrabi, et al., 2011; Sohrabi et al., 2012; Sohrabi et al., 2008a, 2008b). Nowadays, the impact of greenhouse gases emissions on the environment has been a global concern which can be decreased using storage in the oil reservoir. The interfacial properties between reservoir fluids vary significantly during CO₂ flooding in reservoir conditions (Yang et al., 2005). Thus, there is a basic and operational importance to exactly study the IFT in the crude oil–water–CO₂ systems in elevated pressure and temperature conditions. The oil-water IFT is an important factor that determines how easily the trapped oil can be released. To increase capillary number (N_c) by orders of magnitude, the viscosity of the fluid should be increased, or the water-oil IFT should be decreased. Common methods for reducing the IFT are use of surfactants and alkalis. The injection of surfactants into the reservoir can also change the mechanisms of wettability and emulsification in the porous media and reduce the viscosity of heavy crude oil (Bera et al., 2013; Kumar and Mandal, 2016; Kumar et al., 2016). As a result, these lead to the improvement of the mobility of crude oil in the reservoir and to the enhancement of crude oil recovery.

CWI has established important attention recently for increasing the efficiency of oil recovery (Riazi and Golkari, 2016). CWI diminishes the negative impact of water shielding. This problem takes place in water alternating gas injection (WAGI) where the water phase acts as a barrier against CO₂ mass transfer into the oil phase (Riazi et al., 2009; Sohrabi et al., 2008a; Sohrabi et al., 2009). Therefore, CWI is improved the mobility of oil and sweep efficiency. In this process, because of CO₂ diffusion, the other mechanisms, including the reduction of oil viscosity, the reduction of IFT, the alteration of wettability, oil swelling, and coalescence of the trapped oil ganglia occur (Foroozesh et al., 2016; Riazi, 2011; Riazi et al., 2011; Shakiba et al., 2016; Sohrabi et al., 2011; Zeinali Hasanvand et al., 2013).

In recent experimental works to measure the interfacial tension (IFT) for the case of pendant/rising drop, the axisymmetric drop shape analysis (ADSA) method has been applied in reservoir conditions (Hjelmeland and Larrondo, 1986; Yang et al., 2005). In many of the experimental studies, the ADSA technique has also been applied to IFT measurement for different systems: the IFT's of n-alkane-CO₂ (Gasem et al., 1989; Hsu et al., 1985; Nagarajan et al., 1990; Nagarajan and Robinson Jr., 1986, 1987; Zolghadr, Escrochi et al., 2013), the synthetic oil–CO₂ systems, the crude oil–CO₂ systems (Nobakht et al., 2008a, 2008b; Saini and Rao, 2010), the n-paraffin-water systems (Al-Sahhaf et al., 2005; Cai et al., 1996), the crude oil–brine systems (Moeini et al., 2014), and the systems of water–CO₂ (Chun and Wilkinson, 1995; Georgiadis et al., 2010; Ren et al., 2000). In this paper, by determining the spreading coefficient (SC) of the crude oil–water/CW-CO₂ systems, an optimum condition for the WAGI and CWAGI methods is concluded.

Researchers (Riazi, 2011; Riazi, Sohrabi et al., 2011) studied the influence of carbonated water injection (CWI) instead of water flooding (WF) prior to carbon dioxide injection (CO₂I) on light and heavy oil displacement mechanisms and on recovery at high pressures up to 5000 psia and at a temperature of 38 °C by a high-pressure micromodel device. According to the micromodel experiments, they discovered that wettability alteration during CWI becomes more water-wet conditions. They also indicated that CWI, compared to conventional water injection, improves oil

recovery in both secondary and tertiary injection techniques. They showed that the oil spreading as a continuous film at the interface of water and CO₂ phases was the main mechanism for the higher efficiency of CO₂ injection in the previous technique.

The purpose of the current work is to explain how the IFT's of the carbonated water (CW)-CO₂ system, the water/CW-oil system, and the oil-CO₂ system could be applied to the optimization of the cyclic injection of carbonated water and CO₂ technique. Based on the experimental consequences, the capability of the oleic phase to spread on the water phase in the presence of CO₂ phase was studied. These consequences were also used to estimate the best injection pressure conditions for original oil in place recovery after the cyclic injection of water and gas in an oil reservoir in the south of Iran.

The ability of the oil phase to spread as a stable thin film between the water and gas phases is determined by spreading coefficient (SC). This phenomenon takes place in the water-wet porous media when SC value is positive. The oil spreading coefficient is given by the following equation (Rowlinson and Widom, 1982):

$$SC = \gamma_{CO_2/W} - (\gamma_{O/W} + \gamma_{CO_2/O}) \quad (1)$$

where, SC is the spreading coefficient of oil, and γ_{CO_2-W} is the CO₂/water IFT; γ_{CO_2-O} is the CO₂/oil IFT, and γ_{O-W} is the oil/water IFT. The concept of oil spreading between water and gas, which allows a flow of oil by film, has been utilized to describe the very low oil saturations. However, little information is available on the physicochemical properties of the fluids used for better understanding of SC. Because of the dependence of SC upon the physical characteristics of reservoir fluids and the reservoir temperature and pressure conditions, it can be either positive or negative (Oren and Pinczewski, 1994). SC and wettability are two important factors that influence the oil recovery potential and the efficiency of an EOR process.

Mani and Mohanty (1997), using numerical simulations, investigated the influence of the spreading coefficient on a three-phase flow in porous media. They showed that the residual oil saturation is relatively low (about 8%) when SC is positive. In addition, the residual oil saturation increases in the range of 11 to 22% with the magnitude of the spreading coefficient when SC is negative.

Grattoni and Dawe (2003), using core experiments, investigated the impacts of wettability and oil spreading characteristics on gas and oil production from waterflood residual oil. They showed that, for water-wet media and positive SC, both the gas flow rate and gas saturation increase as more gas was released from the solution until the gas saturation reached the value of 30%. Furthermore, the non-volatile oil produced at the end of the experiment of the core was about 32% of the total liquid collected. However, for water-wet media and a negative SC, when the gas saturation extended about 26%, water production practically stopped; the gas saturation was increased to 31%. At the end of the experiment, the oil produced was less than 12% of the total liquid collected, which is obviously less than for the spreading oil.

Moreover, reservoir fluid displacement experiments stated by Oren et al. (Oren and Pinczewski, 1994; Øren and Pinczewski, 1991) and (Oren, 1994; Oren et al., 1992) were performed in two-dimensional glass micromodels in different conditions of wettability (i.e. water-wet and oil-wet) with fluids forming positive and negative spreading systems.

In the positive SC systems, the interfacial energy between reservoir fluids can be decreased by creating a continuity film of oil between the gas and water phases, so thin oil films will spread between water and gas. In addition, the connection of oil film improves the hydraulic constancy which causes higher oil recovery and diminishes the residual oil saturation by gas injection (Blunt et al.,

1994; Chatzis et al., 1988; Kalaydjian, 1992; Kalaydjian et al., 1993).

When the SC is negative, no oil connection exists between the water phase and the gas phase in the porous media. In these conditions, large oil drops may trap and lead to high residuals. Moreover, the water phase may move and circumvent the oil, so eventually a greater volume of oil saturation may be left behind (Amin and Smith, 1998).

It is noteworthy that, in our previous work, the SC phenomena for the CB/brine 10000 ppm-crude oil-CO₂ system was investigated (Riazi and Golkari, 2016). To better understand the SC effect on the operating conditions in CWAGI, the investigation of oil SC in the absence of salt in the aqueous phase is essential. Therefore, in this paper, the EIFT value of the binary systems of water/carbonated water (CW)-crude oil-CO₂ is investigated. Finally, the SC value is calculated in order to find out the best injection pressure for the cyclic injection of carbonated water and CO₂ in an Iranian oil reservoir.

2. Experimental

2.1. Properties of fluids

Crude oil used in this work was supplied from one of the asphaltenic oil reservoirs in the south of Iran. Its properties are presented in Table 1; the composition of the oil is also shown in Table 2. The physical and chemical specifications stated in Table 1 and 2 are reported by National Iranian South Oil Company (NISOC). The water used in all the IFT tests was purified deionized water (DW). The CO₂ having purity higher than 0.9995 was also supplied from a local company.

Table 1

The properties of asphaltenic crude oil (ACO) used in this work ("Analysis of SARA Test," 2014).

Properties	Gravity of dead oil	C ₁₂ ⁺ MW	C ₁₂ ⁺ density at 15 °C	Saturated	Aromatic	Resin	Asphaltene
Unit	°API	gr/mol	g/cm ³	mass %	mass %	mass %	mass %
Value	24.46	326	0.9355	42.68	40.69	7.63	9

Table 2

Compositional analysis of the ACO sample used in this work ("Analysis of Reservoir Fluid Composition", 2014).

Component	H ₂ S	N ₂	CO ₂	C ₁	C ₂	C ₃	iC ₄	nC ₄	iC ₅	nC ₅	C ₆	C ₇	C ₈	C ₉	C ₁₀	C ₁₁	C ₁₂ ⁺
Mole %	1.39	0.88	5.18	22.57	6.94	5.91	0.98	2.97	0.93	1.03	3.07	4.06	4.14	3.69	3.45	2.11	30.68

2.2. High pressure apparatus and experimental procedure

In this study, the dynamic IFT between fluids was measured using a high pressure apparatus. Figure 1 shows the schematic diagram of the experimental device. This apparatus works at a maximum operating pressure of 69 MPa and at a temperature of 175 °C ("IFT 700 For InterFacial Tension Determination, Operating Manual," 2014). The system includes five main parts, namely a high pressure cell (cell's volume is 20 cm³), bulk and drop fluid tanks, a CCD camera, a light source, and a personal computer. Basically, a drop fluid is injected from a calibrated capillary into a bulk fluid in a cell in high pressure and temperature conditions. A CCD camera (1.4 Mega Pixel) and a panel light connected to a computer record the shape of the drop for IFT measurement by pendant/rising drop state. In this experimental study, to create the pendant and rising drops into the cell, a steel needle

with the outer diameter of about 1.5 mm (for the oil–CO₂ and water–CO₂ systems) and 0.79 mm (for the system of water/CW–oil) with an accuracy of ± 0.01 mm was used.

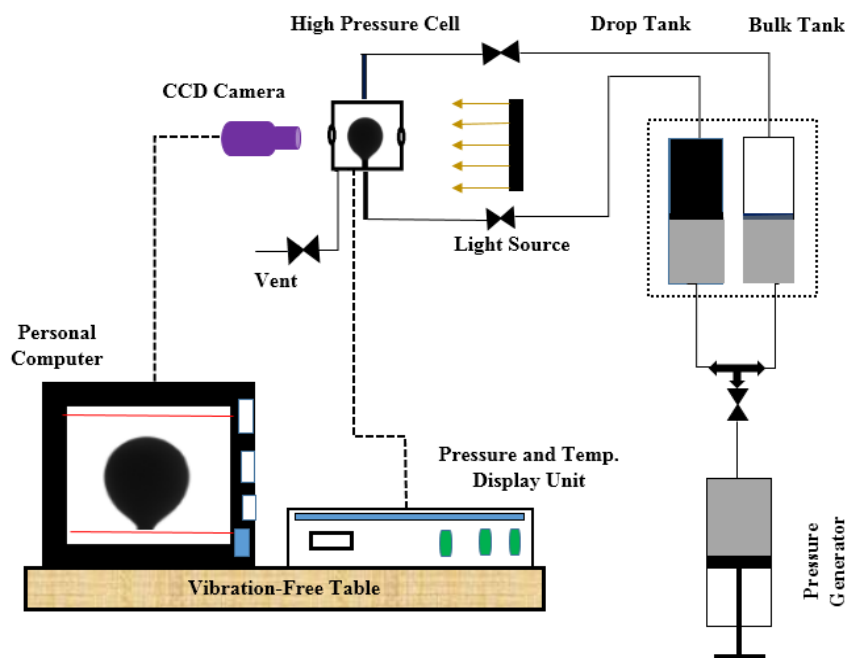


Figure 1

A schematic diagram of the experimental setup used for measuring the EIFT for CW-ACO system at various equilibrium pressures and at temperatures of 40 and 50 °C.

To accurately measure the IFT, the total device was initially tested for leakage with deionized water. Then, it was thoroughly cleaned with toluene and acetone before use to remove any contaminations remaining from formerly performed experiments. Finally, it was drained, flushed with N₂, and vacuumed at a high temperature. This confirmed that any residual solvent was entirely evaporated. Before introducing the drop fluid into the high pressure cell, the temperature and pressure of bulk fluid were retained as in the steady state conditions of the experiment. For both systems of the crude oil/CO₂ and water/CO₂, the bulk fluid (i.e. CO₂) is fully injected into the high pressure view cell initially until it reached the suitable pressure. Then, some of the drop fluid (i.e. water or oil) is injected inside the view cell, and after a certain time allowing the CO₂ bulk to be saturated with water; drop fluid was introduced as a pendant drop at the CO₂ phase. This is useful to simulate the process of CO₂I and CO₂ storing within the oil reservoir. Finally, both systems reached the equilibrium time after about 45 min to 1 hour, and using the image analyzer software, the dynamic IFT was recorded. For the water-oil system, water as the bulk fluid is injected within the view cell at each test pressure, and oil is then formed as the rising drop on the apex of the needle. The experimental process of the oil-CW system is similar to the oil-water system with the exception that the bulk fluid (i.e. water) is pre-equilibrated by CO₂, and a CO₂ cap is created at the above of the CW bulk in order to approach the equilibrium conditions in EOR-CO₂ process. The work procedure is stated as the following steps: initially about 15 cm³ DW is transferred inside the high pressure cell. Secondly, the CO₂ is sluggishly injected within the water phase until it is saturated from CO₂ at the desired pressure, and a CO₂ cap is developed as a gas phase at the contact with CW solution. Finally, the system was given a time of 6-8 hours in equilibrium conditions to reach the steady state. However, to exactly measure the dynamic IFT, the density data in each test conditions are required. In this paper, densities of crude oil and water were measured and presented using density meter DMA HPM (Anton paar, Austria) ("DMA HPM Density Measuring Cell for High Pressure and High Temperatures, Anton Paar Instruction Manual

GmbH," 2005). The CO₂ density was obtained from National Institute of Standards and Technology Chemistry Webbook ("NIST, Chemistry WebBook, NIST Standard Reference Database," 2014). CW density was determined using the equation explained by Hebach et al. (2004). A detailed description of the used densitometer device can be referred to in our previous works (Golkari and Riazi, 2016; Riazi and Golkari, 2016). Finally, for the dynamic IFT, the software is used to analysis the images recorded at two-second intervals at a special temperature and pressure. The accuracy of both devices, i.e. the IFT measurement device and density measurement device, has been confirmed and proved in each desired temperature and pressure conditions (Golkari and Riazi, 2016; Riazi and Golkari, 2016; Zolghadr, Riazi et al., 2013). The temperature and pressure control was monitored at an accuracy of 0.1 °C and 0.05 MPa respectively. The equilibrium IFT (EIFT) was determined using the average of last 100-second of the dynamic IFT. The IFT experiments for each system were performed and repeated three times, and their averages were reported here to ensure acceptable under defined pressures at each desired temperature. The errors in the ACO-CO₂ IFT, the ACO-water IFT, the ACO-CW IFT, and the water-CO₂ IFT systems at each desired temperature and pressure were less than 0.10 mN/m, 0.27 mN/m, 0.25 mN/m and 0.32 mN/m respectively. The error analysis also indicated that the maximum standard deviations of CO₂-ACO, water-ACO, CW-ACO, and water-CO₂ IFT measurements in the mentioned systems were ±0.18 mN/m, ±0.56 mN/m, ± 0.50 mN/m, and ±0.60 mN/m respectively. These were resulted from a lot of experimental data collected for each test point.

3. Results and discussion

3.1. Pressure effect on the water/CW–crude oil–CO₂ EIFT

In this paper, the EIFT for the two systems of (water–crude oil–CO₂) and (CW–crude oil–CO₂) at different pressures from (2.76 to 13.79) MPa and at two temperatures of (40 and 50) °C has been measured. The EIFT data were determined by averaging the dynamic IFT in the last hundred seconds in thermodynamic equilibrium conditions. The results of EIFTs of the water–crude oil–CO₂ system at various pressures and at two temperatures of 40 and 50 °C are given in Table 3. According to Table 3, the EIFT of the systems of water–CO₂ and crude oil–CO₂ declines with pressure, while it rises as the temperature increases. For the water–CO₂ system, both temperatures indicate a fast change of slope linearly at low pressures and a gradual change of slope at high pressures. This means that $(|dy/dp|)$ significantly decreases at low pressures. Nevertheless, the similar behavior has also been reported by several authors for the water–CO₂ systems (Bachu and Bennion, 2009; Chun and Wilkinson, 1995; Hebach et al., 2002; Jho et al., 1978; Tewes and Boury, 2004). For both systems, a decrease in EIFT versus pressure is due to an increase in CO₂ solubility in the water and crude oil phases. Since the CO₂ solubility decreases versus temperature, the EIFT value increases in both systems. The data listed in Table 3 show that the influence of pressure on the EIFT is dominant compared to the effect of temperature in the water–CO₂ system, especially at pressures lower than 8.27 MPa and 9.65 MPa at 40 °C and 50 °C respectively. According to Table 3, at pressures greater than 9.65 MPa and at a temperature of 40 °C, a constant IFT of oil–CO₂ as low as 1–2 mN/m is attained. Moreover, the EIFT approaches 2.87 mN/m at pressures more than 11.03 MPa and at a temperature of 50 °C. The decline in IFT is inconsequential at high pressures. The EIFTs of the crude oil–water system at different pressures and at two temperatures of 40 °C and 50 ° are tabulated in Table 3. It can be seen that the EIFT value increases a little at a mild slope as the pressure rises; however, it drops against temperature. These results show that the influence of pressure on the IFT data of the crude oil–water system is negligible, while the temperature is more effective on the IFT reduction (Cai et al., 1996; Jennings Jr, 1967; Matubayasi et al., 1977; Moeini et al., 2014; Yang et al., 2005). The contradictory behaviors in IFT have also been stated by other authors. For example, the IFT's of oil–water system

indicated a decreasing behavior versus pressure (Hassan et al., 1953), and it displays an increasing behavior versus temperature the system of n-decane–water (Jennings and Newman, 1971). The IFT reduction when temperature rises could be due to an increase in the kinetic energy and in the mobility of the molecules at the interface between crude oil and water, which clearly increases the total entropy of the two phase surface; as a result, its free energy (ΔG) is reduced (Gibbs, 1928; Hirasaki, 1991; Moeini et al., 2014; Myers and Surfaces, 1999). An increase in the IFT value against pressure is attributed to the strengthened intermolecular forces or the decreased distance between molecules at the interface of crude oil and water at high pressures (Moeini et al., 2014) (see Table 3).

Table 3

Experimental results of EIFT and fluids density at different pressures and two temperatures of 40 and 50 °C for the ACO-water/CW-CO₂ systems.

P (MPa)	T (°C)	CO ₂ density (gr/cm ³)	Water density (gr/cm ³)	CW density (gr/cm ³)	Oil density(gr/cm ³)	γ_{CO_2-w} (mN/m)	γ_{O-w} (mN/m)	γ_{O-CO_2} (mN/m)	γ_{O-CW} (mN/m)
2.76	40	0.0534	0.9934	0.9983	0.8994	46.457	28.833	16.325	24.250
3.45	40	0.0696	0.9937	0.9998	0.8999	43.974	28.896	14.656	23.232
4.83	40	0.1079	0.9943	1.0025	0.9009	38.440	29.084	11.2625	22.971
6.89	40	0.1920	0.9952	1.0056	0.9024	29.598	29.357	6.208	22.563
8.27	40	0.3138	0.9958	1.0072	0.9034	23.932	-----	4.186	22.534
9.65	40	0.5977	0.9964	1.0080	0.9044	23.923	29.598	2.269	22.510
11.03	40	0.6848	0.9970	1.0087	0.9054	23.699	29.692	1.897	22.465
13.79	40	0.7593	0.9982	1.0102	0.9074	24.949	30.009	1.280	22.385
2.76	50	0.0509	0.9893	0.9927	0.8957	46.930	28.065	16.185	23.135
3.45	50	0.0659	0.9896	0.9939	0.8962	45.117	28.106	14.889	22.719
4.83	50	0.1001	0.9902	0.9962	0.8972	39.740	28.236	12.238	22.517
6.89	50	0.1677	0.9911	1.9990	0.8988	33.110	28.476	8.123	22.299
8.27	50	0.2348	0.9917	1.0016	0.8996	28.710	-----	5.699	22.203
9.65	50	0.3453	0.9923	1.0023	0.9008	25.810	28.700	4.105	21.979
11.03	50	0.5059	0.9929	1.0031	0.9016	25.212	-----	2.879	21.969
13.79	50	0.6654	0.9941	1.0045	0.9039	25.712	29.017	1.725	21.916

The presented results are similar to those exhibited for the crude oil–brine/water systems (Amin and Smith, 1998; Flock et al., 1986) and for the hydrocarbon–water systems (Cai et al., 1996; Goebel and Lunkenheimer, 1997; Hauser and Michaels, 1948; Hough et al., 1951; Jennings Jr, 1967; Michaels and Hauser, 1951). The EIFTs of carbonated water–crude oil system are shown in Table 3. As can be seen from Table 3, the CW-crude oil IFT drops versus increasing pressure and temperature; the greatest value of IFT was observed at 2.76 MPa, while the lowest value of IFT was recorded at 13.79 at both temperatures. This behavior is due to the solubility of CO₂ in both phases of crude oil and water, which is also enhanced when pressure rises. The impact of CO₂ on EIFT was observed by measuring the CW/oil IFT. The IFT data of CW and water against the oil is shown in Table 3. The water/oil IFT is 29.02 mN/m, whereas the value is decreased to 21.96 mN/m for CW and oil. The governing mechanism for controlling the CW/oil IFT is the solubility of CO₂. Oil swelling phenomena is one of the most important mechanisms that has also been observed in CW injection prior to the CO₂ injection in the micromodel experiments for the light and heavy oil samples (Riazi et

al., 2011). A decrease in the CW-oil EIFT against temperature is attributed to the entropy increasing between oil drop and CW solution boundary. In fact, few researchers have been working in this area. Hence, in this paper, the IFT tests for the asphaltenic crude oil-CW-CO₂ systems were performed, and the role of interfacial properties in the efficiency of EOR processes were well understood. In the experimental work, it was also found out that at pressures higher than 6.89 MPa and at a temperature of 40 °C as well as at pressures higher than 9.65 MPa and at a temperature of 50 °C the impact of temperature on EIFT is dominant compared to the influence of pressure on EIFT. In order to investigate the effect of salinity on IFT, the data presented in this research are compared with the data reported in our previous study (Riazi and Golkari, 2016). The carbonated brine 10000 ppm/oil IFT has changed from 21.67 mN/m to 21.46 mN/m with increasing temperature from 40 °C to 50 °C at a constant pressure of 13.79 MPa respectively, whereas, in the same conditions, the CW/oil IFT is obtained 22.39 mN/m and 21.92 mN/m respectively at temperatures of 40 and 50 °C and at a pressure of 13.79 MPa. Due to the mechanisms of salting-in and salting-out, the presence of salt can change the distribution of surface active components existent in the oil phase and the brine phase. The salting-out effect mechanism leads to an increase in preference for the oil phase or the solid surface with increasing brine salinity (Standal et al., 1999). In addition, the salting-in effect is the reverse trend if the solubility of polar organic species increases in the aqueous phase (Rezaei Doust et al., 2009). Because of salting-in effect, inorganic ions can accelerate the diffusion of surface active components from bulk solution to the interface at low concentrations (Bai et al., 2010). On the other hand, the aqueous solubility of oil species decreases with increasing salinity at high salt concentrations (i.e. salting-out effect) (Armenante and Karlsson, 1982; Standal et al., 1999; Yi et al., 2014). As a result, use of carbonated brine with low salinity creates a satisfactory condition for reducing the IFT.

3.2. Spreading coefficient (SC) estimation

In this study, spreading coefficient (SC) value was determined using the CW/water-ACO-CO₂ EIFTs reported (see Table 3) and Equation 1. It was confirmed that the SC curve for the two systems of water-ACO-CO₂ and CW-ACO-CO₂ increased with respect to pressure from 2.76 to 13.79 MPa at two temperatures of 40 and 50 °C (see Figure 2). Furthermore, this figure shows that the plot of SC versus pressure has a minimum point. The decrease in SC is because of the solubility of CO₂, which is greatly increased in the water and the crude oil phases as pressure rises; subsequently, the oil-CO₂ and water-CO₂ IFTs are reduced. Afterwards, SC is limited to a minimum value, which indicates the worst conditions for the CO₂-EOR process. Finally, due to the decrease of oil-CO₂ IFT at high pressures in the other two systems, SC value is slightly increased. According to Figure 2, the SC value of CW-ACO-CO₂ system is larger and more positive compared to that of water-ACO-CO₂ system. This means that CWI is useful in an EOR-CO₂ process in which, in a system of CW-oil-gas, oil is easily spread between water and gas phases in water-wet porous media and retains continuity through thin films which lead to very low oil saturations.

Moreover, Figure 2 indicates that the SC value slightly increases when temperature rises from 40 to 50 °C, especially at lower pressures. As it was described earlier, the CO₂ solubility and total entropy are two governing mechanisms in controlling the SC. At a consent pressure, the solubility of CO₂ influences the water/oil-CO₂ IFT against temperature; the higher the temperature is, the lower the solubility becomes, thereby increasing the IFT. Moreover, the total entropy rises against temperature, which causes an effective reduction in the water/CW-oil IFT. The both mechanisms mentioned above have positive results on SC. For the CW-ACO-CO₂ system, a negative SC was also detected at 40 °C and in a pressure range of 6.89–11.03 MPa as well as at 50 °C and in a pressure range of 8.27–11.03 MPa (see Figure 2). The results indicate that for the maximum recovery efficiency the appropriate

pressure of the CWI process for this particular reservoir is at pressures higher than 11.03 MPa and lower than 6.89 MPa; indeed, at other pressure values, the value of SC is almost negative.

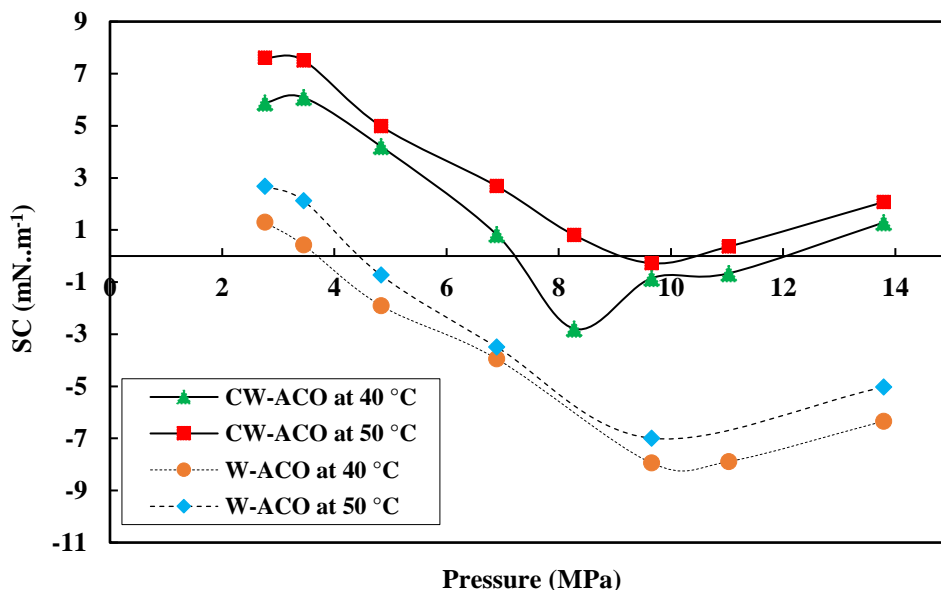


Figure 2

SC of systems defined as a function of pressure at two temperatures.

Furthermore, for the water–ACO–CO₂ system, a negative SC was seen at 40 °C and at pressures higher than 3.45 MPa as well as at 50 °C and at pressures higher than 4 MPa. As a consequent, for the maximum recovery, the best pressure for EOR–CO₂ process in this given reservoir should be below 3.45 MPa. At a constant pressure, the SC is affected by the temperature. The higher the temperature is, the lower the solubility of CO₂ becomes, and as a result, the higher the SC is. Moreover, according to the reasons presented earlier for the impact of salinity on water/oil IFT, the SC is greatly affected by brine salinity; the higher the salinity is, the higher the SC becomes. For example, at 3.45 MPa and 50 °C, the SC of CB/oil/CO₂ system (Riazi and Golkari, 2016) and CW/oil/CO₂ are 9.47 mN/m and 7.61 mN/m respectively.

According to the results reported and from an operational point of view, the SC's of both systems are positive for an oil reservoir at low pressures. Thus, there might not be such necessity to substitute water injection (WI) with CW injection before CO₂ injection although the SC value of CW system is greater than that of the water system. This means that at 3.45 MPa and 50 °C, the SC of water–oil–CO₂ system and CW–oil–CO₂ system are 2.12 and 7.51 respectively. On the other hand, since the majority of reservoirs are in high pressure conditions, the SC of CW–oil–CO₂ system is positive, whereas the SC of water–oil–CO₂ system is negative. The SC of CW/oil/CO₂ at 13.79 MPa and 50 °C was 2.07 mN/m, while that of the water/oil/CO₂ system in the same conditions was reduced to -5.03 mN/m. Therefore, the presented results propose the cyclic injection of CW prior to CO₂ injection, principally at high pressures.

4. Conclusions

In this study, the effects of temperature and pressure on the EIFT of the systems of water–CO₂, ACO–CO₂, ACO–water, and ACO–CW were investigated. The exact analysis of the achieved results directed the findings toward the following conclusions:

- 1- The effects of pressure and temperature on the IFT of the defined systems:

- For both of water/CO₂ and ACO/CO₂ systems, the EIFT decreases against pressure, whereas it increases versus increasing temperature. This is attributed to increasing the solubility of CO₂ versus pressure and its reduction as a function of temperature.
 - The EIFT of the water-ACO system slightly increases versus pressure, whereas it drops for the CW-ACO system at low pressures until the IFT reaches a constant value in high pressure conditions. A decrease in the IFT is because of the CO₂ movement from CW solution and its diffusion inside the oil drop at low pressures. In addition, the CO₂ solubility reaches its maximum value at high pressures. Also, the presence of CO₂ in water was observed to be in favor of decreasing the oil-water IFT, where CW had lower IFTs compared to water.
 - According to the results of this study and our previous work (Riazi and Golkari, 2016), the oil/water IFT is affected by brine with low salinity; the higher the salinity is, the lower the IFT becomes, thereby raising SC.
- 2- In the case of CW/water, CO₂, and an oil type having a high asphaltene content, the SC curve versus pressure displays two different trends:
- A decreasing trend related to the solubility of CO₂, which is greatly increased in the water and crude oil phases as pressure rises and leads to a reduction in the IFT of oil-CO₂, water-CO₂, and CW-oil systems.
 - In a specific pressure range, SC reaches a minimum point value that indicates the worst conditions for EOR-CO₂ operations.
 - The last increasing trend of the SC is due to the fact that the sum of IFT values of ACO-water and ACO-CO₂ is lower than the IFT of the water-CO₂.
- 3- The results show that SC slightly increases versus temperature, especially at lower pressures. This is because of the role of temperature in the solubility of CO₂ in the water phase; in fact, the higher the temperature is, the lower the solubility becomes, which increases the brine-CO₂ IFT. Additionally, the total entropy as another governing mechanism is affected by temperature, which mainly reduces the water/CW-oil IFT.
- 4- It was found out that the SC of CW-ACO-CO₂ is positive and higher compared to that of the water-ACO-CO₂ system. The SC of CW/oil/CO₂ at 13.79 MPa and 50 °C was 2.07 mN/m, while that of water/oil/CO₂ system in the same conditions was reduced to -5.03 mN/m. This represents that the cyclic injection of carbonated water and CO₂ could result to greater recovery efficiency compared to the cyclic injection of water and CO₂.

Nomenclature

ADSA	: Axisymmetric drop shape analysis
API	: oil API gravity (American Petroleum Institute)
CW	: Carbonated water
EOR	: Enhanced oil recovery
IFT (γ)	: Interfacial tension (mN. m ⁻¹)
P	: Pressure (MPa)
SC	: Spreading coefficient (mN. m ⁻¹)
T	: Temperature (°C)

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