Experimental Investigation of Using Albizia Julibrissin Extract as a Plant Surfactant for Oil Recovery

Seyed Reza Shadizadeh[1*], Seyed Ramin Seyedi Abandankashi[2], and Siyamak Moradi[3]

[1] Professor, Department of Petroleum Engineering, Petroleum University of Technology, Abadan, Iran
[2] M.S. Student, Department of Petroleum Engineering, Petroleum University of Technology, Abadan, Iran
[3] Assistant Professor, Department of Petroleum Engineering, Petroleum University of Technology, Abadan, Iran

Highlights

- The Albizia Julibrissin extract (AE) was used as a natural plant-based surfactant;
- The AE decreased the interfacial tension between the aqueous phase and the oil phase from 34 to 10 mN/m;
- The AE reduced the wettability toward a water-wet system as it decreased the contact angle by about 46.5% at the CMC point;
- The AE solution flooding as a tertiary method conducted on a carbonate core increased the oil recovery by up to 11.6% of original oil-in-place (OOIP).

Abstract

In recent years, the use of natural surfactants, instead of chemical surfactants, as surface active agents in chemical methods of oil recovery has been under consideration due to the absence of environmental problems. In this study, a new plant, Albizia Julibrissin (Albizia), was introduced as a natural surfactant. Our novelty resides in a unified approach that deals with the introduction of Albizia Julibrissin as a new natural surfactant, the interpretation of the chemical enhanced oil recovery (EOR) objectives, the interface reactions, and the induced optimization to improve oil recovery. For this purpose, the plant was extracted using the Soxhlet extraction method in aqueous base solutions, and the interfacial tension between the natural surfactant aqueous solutions and kerosene as an oil phase was measured by the pendant drop method. The critical micelle concentration structure formed by this material was determined by the interfacial tension tests and confirmed by the electrical conductivity tests. The results show that Albizia extract begins to form micelles at a concentration of 3.5 wt %, which is the critical concentration of Albizia plant micelles. At this concentration, the interfacial tension between the deionized water and the oil phase was reduced from 34 to 10 mN/m, which indicates a significant decrease in the interfacial tension by this plant. Carbonate rock was employed for the core flooding experiments in order to investigate the effect of Albizia extract (AE) on oil recovery. Also, based on the results, by using AE, the wettability of the oil-wet carbonate rocks was altered from about 165.02° to 86.59°. Finally, the AE enhanced ultimate oil recovery by about 11.6% of original oil in place in tertiary recovery for a carbonate rock.

Keywords: Albizia Extract, Interfacial Tension, Natural Surfactant, Oil Recovery, Wettability Alteration

* Corresponding author:
     Email: shadizadeh@put.ac.ir

1. Introduction

Chemical oil recovery is one of the tertiary oil recovery methods, which includes the injection of surfactants to increase oil production from the reservoirs. Surfactants are surface active agents that, with their dual structure, are capable of forming a bridge between two immiscible fluids. This dual structure has a hydrophobic tail and a hydrophilic head, which decreases the interfacial tension by diffusing into the common interface of two immiscible fluids (Ayirala et al. 2010). A simple classification of surfactants based on their hydrophilic head nature is as follows:

- Anionic surfactants such as alkyl benzene sulfonate that have a negative charge on their surface active section;
- Cationic surfactants such as long-chain amine salts that have a positive charge on their surface active section;
- Amphoteric or zwitterionic surfactants such as long-chain amino acids that may have both positive and negative charges on their surface active section;
- Nonionic surfactants such as long-chain fatty acids that have no charge on their surface active section (Urbina-Villalba et al. 1996).

These materials reduce capillary pressure by decreasing the interfacial tension of two immiscible water–oil fluids. As a result, the oil production increases in porous media due to higher accessibility to trapped oil in small pores. Surfactants, in addition to the general method mentioned above, improve oil production in a number of ways: reducing interfacial tension between hydrocarbons and aqueous phase, altering the wettability of the reservoir rock surface, creating foam, and creating stable emulsions. Table 1 summarizes the research conducted on natural and chemical surfactants in recent years. Various types of surfactants are used worldwide to increase oil production. Almost all surfactants are expensive, which is why using the chemical process in some situations is not economically feasible; the high cost of these materials, along with their incompatibility with the environment, leads to increased operational difficulties (Manshad et al. 2017; Emadi et al. 2019).

Table 1

Summary of the studies on the application of chemical or natural surfactants in EOR.

Author	Surfactant	Results
Rostami et al. (2017)	Amino acid-based surfactant	- Interfacial tension (IFT) was reduced to 15 mN/m; - Wettability changed to water-wet; - Oil recovery increased by 12%.
Pal et al. (2018)	Sunflower oil-based Gemini surfactant	Very low IFT results
Pal et al. (2018)	Coconut oil-based anionic surfactant	- Very low IFT results; - Water-wet wettability alteration; - 20.05% oil recovery improvement.
Madani et al. (2019)	Amino acid-based surfactant	- IFT deceased to 20 mN/m; - Wettability changed to water-wet; - Oil recovery improved in secondary and tertiary methods.

Author	Surfactant	Results
Ganie et al. (2019)	Lignin-based surfactant	Ultra-reduction of IFT to 0.5–1.0 mN/m
Saxena et al. (2019)	Madhuca longifolia oil based anionic surfactant	• Very low IFT results; • Wettability changed to water-wet; • Oil recovery increased by 20%.
Nowrouzi et al. (2020)	Anabasis Setifera	• Reduction of IFT value to 1.066 mN/m; • Wettability altered the two poor water-wet; • Oil recovery of 15.4% was obtained.

Recently, different natural plants, which are cheaper and eco-friendlier than chemical surfactants, have been used in the chemical flooding process (Ahmadi et al. 2014; Deymeh et al. 2012; Pordel et al. 2012). Chhetri et al. (2009) evaluated the effects of different concentrations of soapnut on IFT reduction and effectiveness in the enhanced oil recovery process. Pordel et al. (2012) showed that adding saponins into their nonionic surfactant in distilled water with 1 g/L NaCl would reduce the amount of interfacial tension from 48 to 9 dyne/cm. Deymeh et al. (2012) used Seidlitzia rosmarinus powder prepared by the spray drying method. The interfacial tension of the aqueous base solution of Seidlitzia rosmarinus surfactant and oil phase was measured using the pendant drop method, and kerosene was used as the hydrocarbon phase in the interfacial tension measurement. Their results showed that Seidlitzia rosmarinus reduced the interfacial tension from 32 to 9 dyne/cm. Furthermore, Zendehboudi et al. (2013) reported the IFT reduction from 32 to 11 mN/m using a Zizyphus Spina-Christi extract. Ahmadi et al. (2014) used mulberry leaf extract as a natural surfactant; the mulberry leaf extract was used to prepare a microfluid formulation. Their results showed that the sample containing 1 wt % of mulberry leaf particles reduced the interfacial tension between kerosene and water from 44 to 17.9 dyne/cm. Ravi et al. (2015) experimentally investigated the effect of the mulberry leaf extract on the alteration of wettability, on the reduction of IFT, and on oil recovery. Moreover, Rahmati et al. (2015) worked on Henna extract, and Shadizadeh and Kharrat (2015) used Matricaria Chamomilla extract; Barati-Harooni et al. (2016) also investigated the influence of Trigoonella Foenum-Graceum extract on reducing IFT.

In this work, the possibility of using Albizia Julibrissin extract as a natural surfactant in the process of oil recovery is investigated. For this purpose, by using the pendant drop method, the contact angle and the interfacial tension between the surfactant and oil is measured; the critical concentration of micelle formation is determined and confirmed by the electrical conductivity test. Then, the optimum concentration of the Albizia extract solution is used to evaluate the effect of this solution on oil recovery in a carbonate rock.

2. Materials and methods

2.1. Albizia extract

This plant is known as the Persian silk tree across the globe and is a type of a tall tree with a height of 5 to 12 m. Figure 1 shows an image of the leaf of the Albizia plant. The canopy of this plant is broad, so it has a relatively good shade. It grows slowly and takes 10 to 15 years to grow. The flowers of this tree are pink, white, and cluster-like, and the trunk skin of this tree has healing properties. The traditional Chinese medicine has used its flowers to nourish the heart. This plant is abundant in the Middle East and East Asia, especially in Iran and Iraq (Gargiullo et al. 2005; Cheatham et al. 1998; Lau et al. 2007). It has been used in the northern parts of Iran for washing dishes and hands. The leaves of this plant are used to remove stains created by the walnut skin in the summer in the northern villages of Iran, and indigenous people believe that it has a cleansing ability because of its high foaming when it is mixed with water. Kokila et al. (2013) analyzed the structure of the Albizia plant and found that it contained

three triterpenoid saponins, three new oleanane-type triterpene saponins, cytotoxic compounds, and glycosides. Zhang et al. (2013) after the analysis of the oil extracts of the plant concluded that it is comprised of 23.3% of palmitic acid, 6.6% of trans-linalool oxide, 7.2% of pentacosan, 6.2% of methyl salicylate, 6.1% of eugenol, and 5.1% of octanol. Some physical properties of the Albizia Julibrissin extract are listed in Table 2.

Table 2

The properties of Albizia Julibrissin extract (Zhang et al. 2013 and Kokila et al. 2013).

Product name	Albizia Julibrissin extract
Used part	Leaves
Odor	Specific odor
Color	Light to dark brown
Solubility in alcohol	Soluble
Solubility in water	Soluble
Density (gr/cc)	1.0049
pH	7.06
Clarity	Clear liquid

Figure 1

Leaves of Albizia plant.

2.2. Aqueous and oil phases

In order to measure the interfacial tension, kerosene was utilized as the oil phase. The most important reason for substituting kerosene for crude oil is its cleanness. It has also been used to measure the interfacial tension in order to remove the effects of asphaltene and surfactants in crude oil (Ahmadi et

al. 2014). The density of kerosene used in this study was 0.79 g/cm^3, and the deionized water was used in the preparation of different solutions of the extract. For the core displacement testing, the crude oil from one of Iran's oilfield was utilized, and the density of the crude oil sample at atmospheric pressure was 0.89 gr/cm^3; also, a synthetic brine with a concentration of 10 wt % was implemented to simulate the conditions of the reservoir.

2.3. Core sample

A carbonate rock with a permeability of 6.8 mD and porosity of 12.9% taken from an outcrop in the vicinity of Shiraz was used for the oil recovery test. Also, for the contact angle test, this type of rock was prepared in thin sections.

2.4. Preparation of AE solutions

After collecting the leaves of the Albizia plant, drying them under ambient conditions, separating the sub-branches of these leaves, and crushing the main part of the leaves by mortar, the final powder was poured into a Soxhlet apparatus using deionized water, and the extraction process of the leaves of this plant was completed. The final extract was placed in a water bath to reach a temperature equilibrium, and the prepared material was finally ready for storage.

In this research, 10 solutions with a weight percentage of 0.5, 1.0, 1.5, 2.0, 2.5, 3.0, 3.5, 4.0, 4.5, and 5.0 were prepared. As shown in Figure 2, the solutions were prepared from 0.5 to 5 wt %. To prepare these solutions, a certain amount of the plant extract was mixed with 100 mL of distilled water, and a magnetic stirrer was used for a limited period of 20 minutes at 50 °C to obtain a homogeneous solution.

Figure 2

Different solutions of the Albizia extract.

2.5. Interfacial tension measurement of AE solutions

The pendant drop method was implemented to measure the interfacial tension between the fluids. The pendant drop apparatus (Figure 3) consists of several components: the fluid chamber, the injection syringe, the lighting section, and the data acquisition section, which includes a high-resolution camera for taking a picture and computer software for displaying droplets and performing calculations. The software matches the theoretical interfacial shapes-generated using the Young–Laplace equation with the experimental drop profiles at the mechanical equilibrium by a curve fitting method. The pendant drop method is based on curve fitting, and the following relationships are used to determine the IFT.

$$\gamma = \frac{\Delta \rho g b^2}{\beta} \tag{1}$$

$$S = \frac{d}{D} \tag{2}$$

$$\beta = 0:12836 - 0:7577S + 1.7713S^2 - 0.5426S^3 \tag{3}$$

$$b = \frac{d}{2(0.9987 + 0.1971\beta - 0.0734\beta^2 + 0.34708\beta^3)} \tag{4}$$

where $\Delta\rho$ is the difference between the densities of the drop and the material surrounded the drop. β is a dimensionless number, and b represents the radius of the curvature at the drop; they are calculated by Equations (3) and (4) respectively. γ indicates the interfacial tension, and g is the gravitational acceleration; D is the equatorial diameter of the drop, and d is the distance of D from the apex. A density meter apparatus (DMA) was used to measure the densities of the solutions with different concentrations (Eslahati et al. 2020).
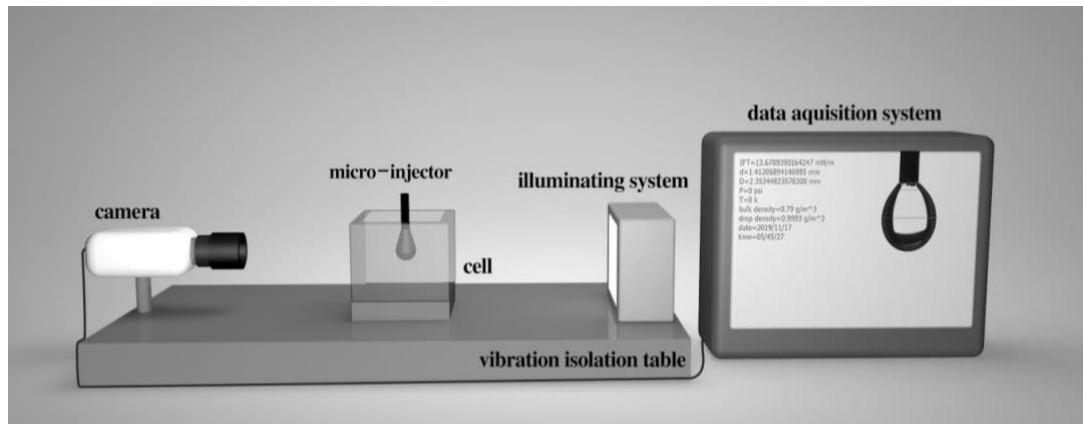

Figure 3

The pendant drop apparatus.

In this method, using an interfacial tension measuring device, the surfactant is pumped from the piston cell containing the solution, inserted into the needle, and placed as a suspension drop into a cell containing kerosene as the oil phase. The suspended drop remains in the oil phase, and the amount of the interfacial tension is recorded continuously by the device as the process is measured for a specified period of time; the dynamic interfacial tension is also measured. These values are recorded as a graph over time, and at the end of the graph, they are known as the equilibrium interfacial tension due to a stable state. Then, the equilibrium value is reported as the interfacial tension.

2.6. Contact angle measurement

The carbonate cores were cut into a bunch of pellets, and the pellets were aged for about a week into crude oil at a temperature of 80 °C. Afterward, they were washed with kerosene dried for a day at a temperature of 50 °C. Then, the pellets were placed into the prepared surfactant solutions for five days to check the changes of rock wettability. Subsequently, the pellets were removed from the solutions and dried in air. Eventually, they were placed inside the contact angle chamber filled with kerosene, and then a drop of the solution was thrown on the pellet with a needle device. The camera sends the thin section pictures to the computer, and the software measures the contact angle using image processing.

2.7. Core flooding tests

After porosity and permeability determination, the core flooding tests were conducted to evaluate the recovery factor for surfactant flooding. In order to perform the core flooding tests, the cores were primarily saturated with water and displaced with oil to reach connate water saturation. Then, water and surfactant were injected into the cores to displace the oil. In order to calculate the recovery factor, the differential pressure and the effluent volume of the displaced oil were recorded.

After the permeability measurement of the core, it was completely saturated with the brine. In order to measure the oil in-place the core, the oil was injected into the core until the brine was produced inside the core.

The oil accumulator was connected to the core holder instead of the brine accumulator, and the confining pressure was applied to the core. Then, the oil was injected into the core. The brine was produced by the oil injection into the core. The oil injection continued until the core produced the brine. According to Equation (5), the amount of oil in-place is equal to the amount of the brine production from which the dead volume before and after the core is subtracted.

$$OOIP = Brine\ Production - Dead\ Volume\ \text{(cc)} \tag{5}$$

Also, the amount of the irreducible water saturation is calculated by subtracting the pore volume from the brine production.

After the oil was injected into the core, and the brine production finished completely, the secondary recovery was performed by the brine injection. The secondary recovery was carried out by the brine injection inside the core and the measurement of the oil production. The brine was injected into the core at a concentration of 10 wt % and a flow of 0.2 cc/min. The pressure, time, and pore volume of the injection were measured during the test.

According to Equation (6), the amount of the secondary recovery is equal to the ratio of the oil production in the secondary stage to the oil in-place.

$$RF = \frac{Oil\ Production\ in\ Secondary\ Stage\ (cc)}{Oil\ in\ Place\ (cc)(\%\ OOIP)} \tag{6}$$

Afterward, the accumulator brine (with an NaCl concentration of 10 wt %) was replaced with the crude oil and allowed to flow at a constant rate of 0.2 cc/min. After injecting 2 or 3 PV of the crude oil into the core, the irreducible water saturation was measured. Again, the accumulator was replaced with the brine solution for the secondary recovery and then with the AE solution as the tertiary recovery (with a concentration of 3.5 wt %).

The tertiary recovery is done for higher recovery. In the tertiary recovery, the surfactant solution was injected into the core instead of the brine. After the preparation of the surfactant solution, the solution was added into the solution accumulator, and then the accumulator was connected to the core holder. The surfactant injection was carried out at a flow rate of 0.2 cc/min, and the amount of the oil production per pore volume in this section was measured in this stage. The tertiary recovery achieved by the oil production at the tertiary stage was determined by Equation (7).

$$RF = \frac{Oil\ Production\ in\ Tertiary\ Stage\ (cc)}{Oil\ in\ Place\ (cc)\ (\%\ OOIP)} \tag{7}$$

3. Results and discussion

In the study of the surfactant flooding, the best result will be the unique concentration at which the micelles are formed, which is known as the critical micelle concentration (CMC) (Ahmadi et al. 2014; Deymeh et al. 2012; Pordel et al. 2012). On the other hand, using a higher concentration of surfactant increases the viscosity and slightly improves the mobility ratio, while increasing the concentration of the surfactant above the CMC causes a higher cost. Moreover, it increases the IFT and the contact angle

since after the CMC, the surfactant amphiphilic molecules are no longer at the interfaces and tend to create micelles. The goal of this study is to introduce and evaluate the efficiency of the AE as a surfactant with the potential to increase oil recovery. To this end, the IFT and contact angle measurements were done to determine the CMC of the solutions used in the flooding test.

3.1. Interfacial tension

At the interface between the water and oil phases, the nonionic molecules of the Albizia surfactant orient in such a way that the lipophilic part resides in the oil phase, and the hydrophilic part lies in the aqueous phase. Before the CMC, the surfactant molecules are placed in the space between the oil and water phases, creating a strong connection between the two phases. As a result, the IFT is reduced. Increasing the amount of surfactant molecules reduces the amount of IFT, and this decrease continues until the CMC is reached (Eslahati et al. 2020).

In order to achieve the equilibrium interfacial tension of the solutions with different concentrations, the values of the dynamic interfacial tension were measured since the dynamic interfacial tension behavior was reduced in a specific trend as shown in Figure 4. At the beginning of the curves, there is a very steep slope, but they level off as the time increases, that is to say, the equilibrium area is achieved. These dynamic interfacial tension values are obtained in a way that shows the stability of the interfacial tension for each concentration from the beginning to the end.

As shown in Figure 4, the interfacial tension diagrams for 10 concentrations of the surfactant solutions are given in Figures 4a–4j. As the concentration increases from 0.5 to 3.5 wt %, the IFT is reduced from 34 to 10 mN/m, which indicates that the interfacial tension is reached a critical point. After this concentration, the IFT reaches a stable value as shown in Figures 4g–4j, which implies that the optimum amount of the surfactant is reached. This is due to the formation of the micelles and the lack of any surfactant interference within the interface. In other words, after the optimum concentration, the surfactants prefer to enter the structures formed by themselves, i.e. micelles, rather than to diffuse into the interface.

In the pendant drop method for measuring the dynamic interfacial tension, after a considerable period of time, the interfacial tension leveled off at a constant value, which is the equilibrium interfacial tension resulting from the equilibrium between the oil phase and the surfactant solution drop. The equilibrium interfacial tension diagrams at different concentrations of the AE in the oil phase are delineated in Figure 5. The results show that the extract of the AE reduces the amount of the interfacial tension between water and oil from 34 to 10 mN/m. As shown in this figure, after the critical AE concentration of 3.5 wt %, known as the optimal CMC value, the IFT changes slightly.

Figure 6 depicts the variation of the electrical conductivity with the weight percentage of the AE. It is obvious that the electrical conductivity increases linearly as the concentration rises from 0.5 to 3.5 wt %; however, at concentrations higher than 3.5 wt %, the electrical conductivity increases at a lower rate and tends to plateau. The difference in the slope of the curve happens at an AE concentration of 3.5 wt %, which is the CMC for these solutions.
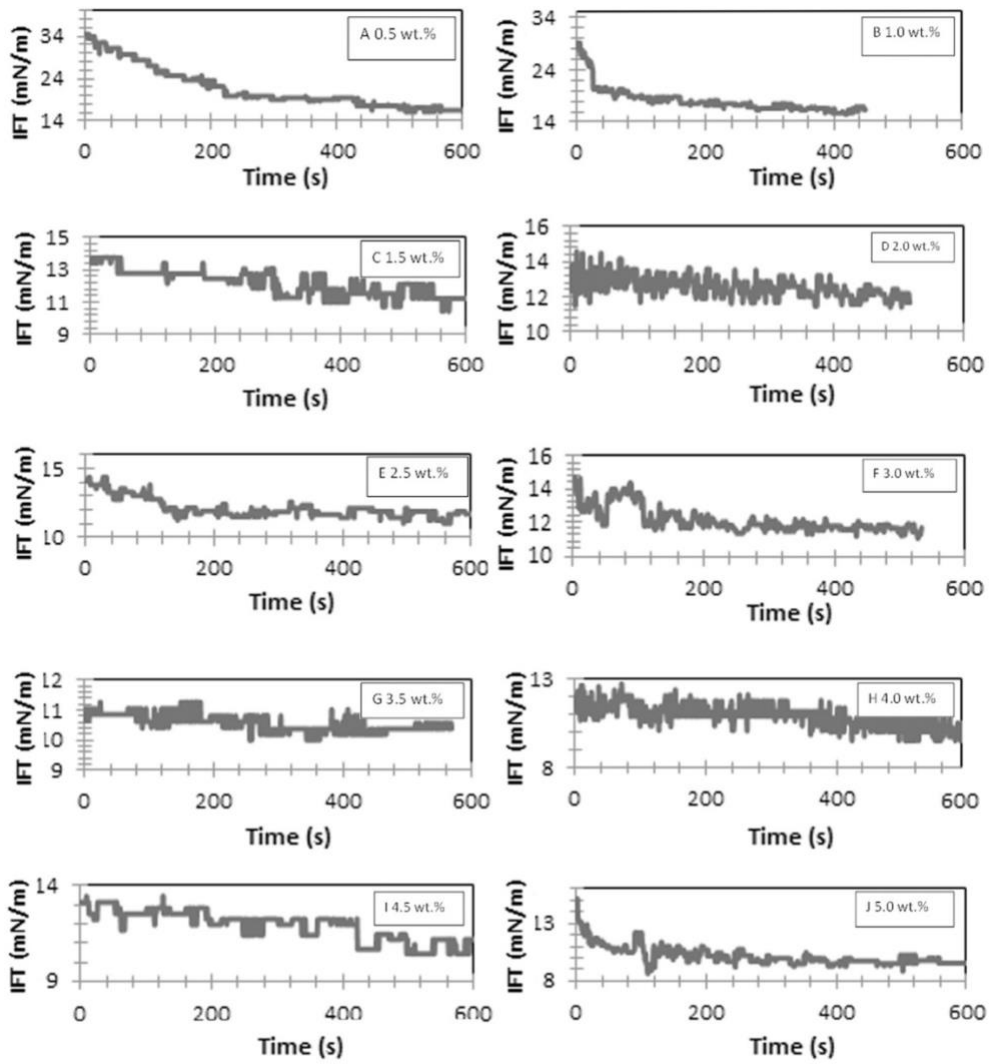
Figure 4

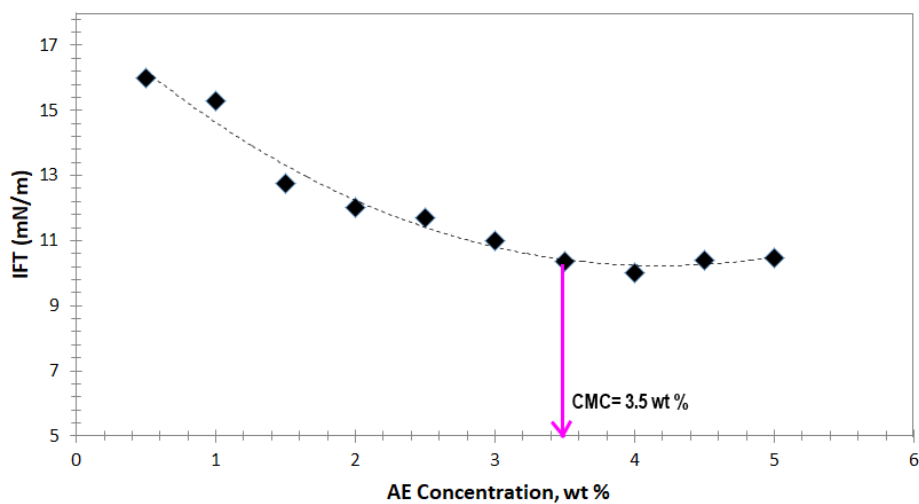Dynamic interfacial tension diagrams at different concentrations of the AE.

Figure 5

The variation of the equilibrium interfacial tension versus the surfactant concentrations.
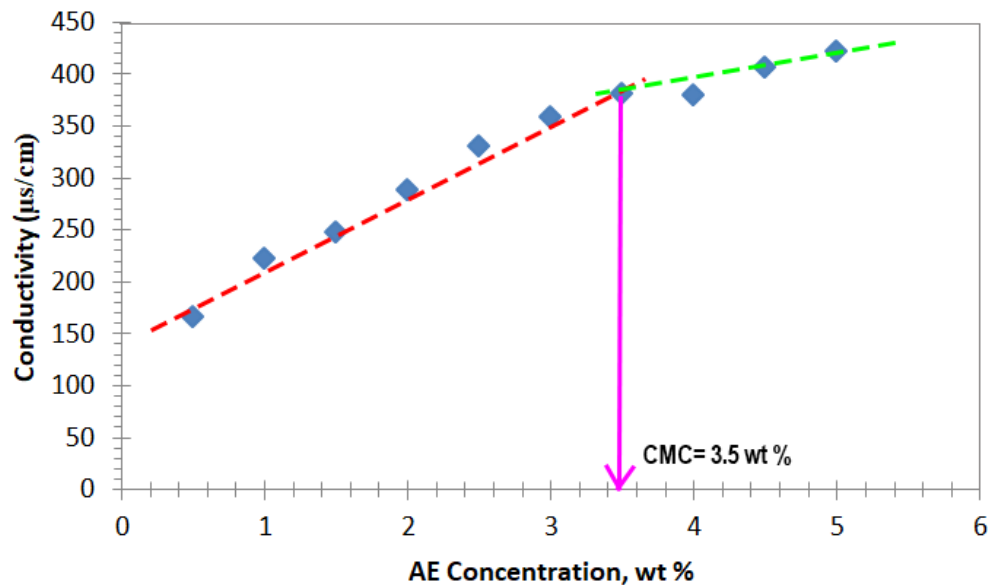
Figure 6

The variation in the electrical conductivity with the concentration of the AE surfactant.

In recent years, different plant surfactants have been used for chemical flooding due to their more environmentally friendly features in comparison to the synthetic surfactants (Deymeh et al. 2012; Pordel et al. 2012; Ahmadi et al. 2014; Ravi et al. 2015; Shadizadeh et al. 2015). These surfactants have the ability to reduce the IFT value. Figure 7 shows the IFT measurements of different plant surfactants along with that of the Albizia extract. As can be seen in this figure, the effect of the Albizia extract on the IFT is even better compared to the other plant surfactants.
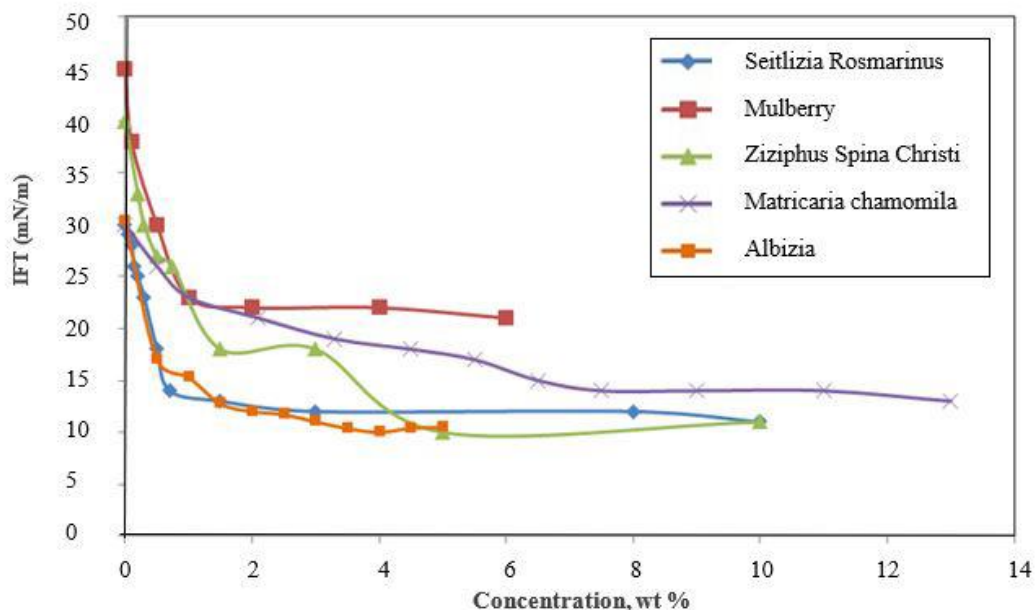

Figure 7

The variation in the interfacial tension of kerosene as the oil phase at room temperature and atmospheric pressure in the presence of different plant surfactants: Seidlitzia rosmarinus (Deymeh et al. 2012); Ziziphus Spina-Christi

(Pordel et al. 2012a); mulberry (Ahmadi et al. 2014; Ravi et al. 2015); Matricaria Chamomilla (Shadizadeh et al. 2015).

3.2. Contact angle

The below mechanisms show the reasons for the change in wettability following the use of the Albizia surfactant:

- The surfactant has two parts: hydrophilic and lipophilic. The hydrophilic part of the surfactant absorbs resin and asphaltene of the oil. A substitution between the desorbed organic compounds in the oil phase and the nonionic surfactants in the brine take places, and eventually the rock surface undergoes a wettability alteration as a result of this process;
- The positive charge of natural surfactants attracts the negative charge of oil compounds like asphaltene, and they are desorbed from the surface of the rock (Eslahati et al. 2020);

As mentioned before, kerosene was used as an oil phase in the contact angle testing. The contact angle tests were done on the carbonate pellets. The photograph of the contact angle at different concentrations of the AE is depicted in Figure 8. The contact angles of the AE solution at a concentration in the range of 0.5 to 5 wt % are shown in Figure 9. As the results show, the AE can decrease the contact angle from about 165.02° to 86.59° for carbonate rocks. The importance of this result is that the wettability alteration of this extract is desirable. The results of the change in the carbonate rock wettability demonstrate that the contact angle decreased by up to 46.5%. In other words, the reduction in the contact angle was 74.5°, which is really significant.

Figure 8

The photographs of the contact angle at different concentrations of the AE solutions.
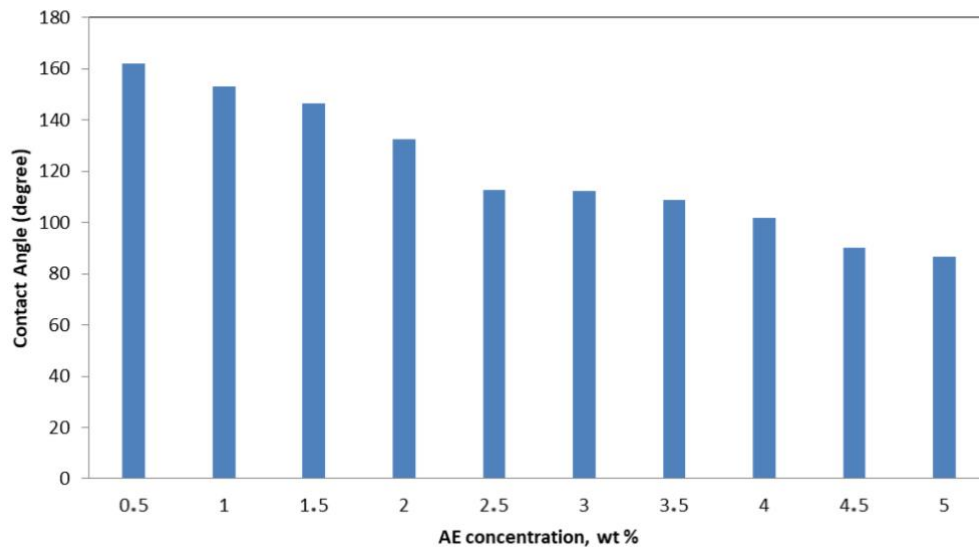
Figure 9

The variation of the contact angle at different concentrations of the AE surfactant.

3.3. Core flooding

High capillary forces cause small droplets of oil to remain in pore structures. Modifications such as wettability alteration toward a water-wet system and IFT reduction can improve oil recovery by reducing the capillary forces of the system. Capillary forces prevent the displacing of oil through the small pore throats of the reservoir, and removing these forces cause the oil to move out of the pore system. Capillary pressure is related to the contact angle, the IFT, and the pore radius as expressed in Equation (8):

$$Pc = \frac{2\sigma Cos\,\theta}{r} \tag{8}$$

where σ is the fluid–oil IFT, θ represents the angle between the wetting fluid and the solid surface, *r* stands for the effective radius of the interface, and *Pc* indicates the capillary pressure. Since the radii of the pore throats are fixed, either a reduction in the water–oil IFT or the wettability alteration toward a water-wet system is needed to reduce the capillary pressure (Bahraminejad et al. 2019).

Following the water flooding, the AE solution with a concentration of 3.5 wt % was injected into the core as the tertiary oil recovery. Figure 10a shows the results of the oil recovery of the AE solution after the water flooding. The water flooding recovery was 43.3% for the brine solution. By injecting the AE solution after the water flooding, the recovery factor reached 54.9 % and then remained constant. The oil produced during the tertiary recovery equaled 11.6%. Figure 10b displays the relation between the pressure difference and the injected pore volume for the AE solution flooding as the tertiary recovery after the water flooding. The pressure difference first increased at the breakthrough and then decreased to 168 psi; the curve then stabilized. From the relation between the pressure difference and the injected pore volume, it can be inferred that the breakthrough occurred at 0.3 PV injection.
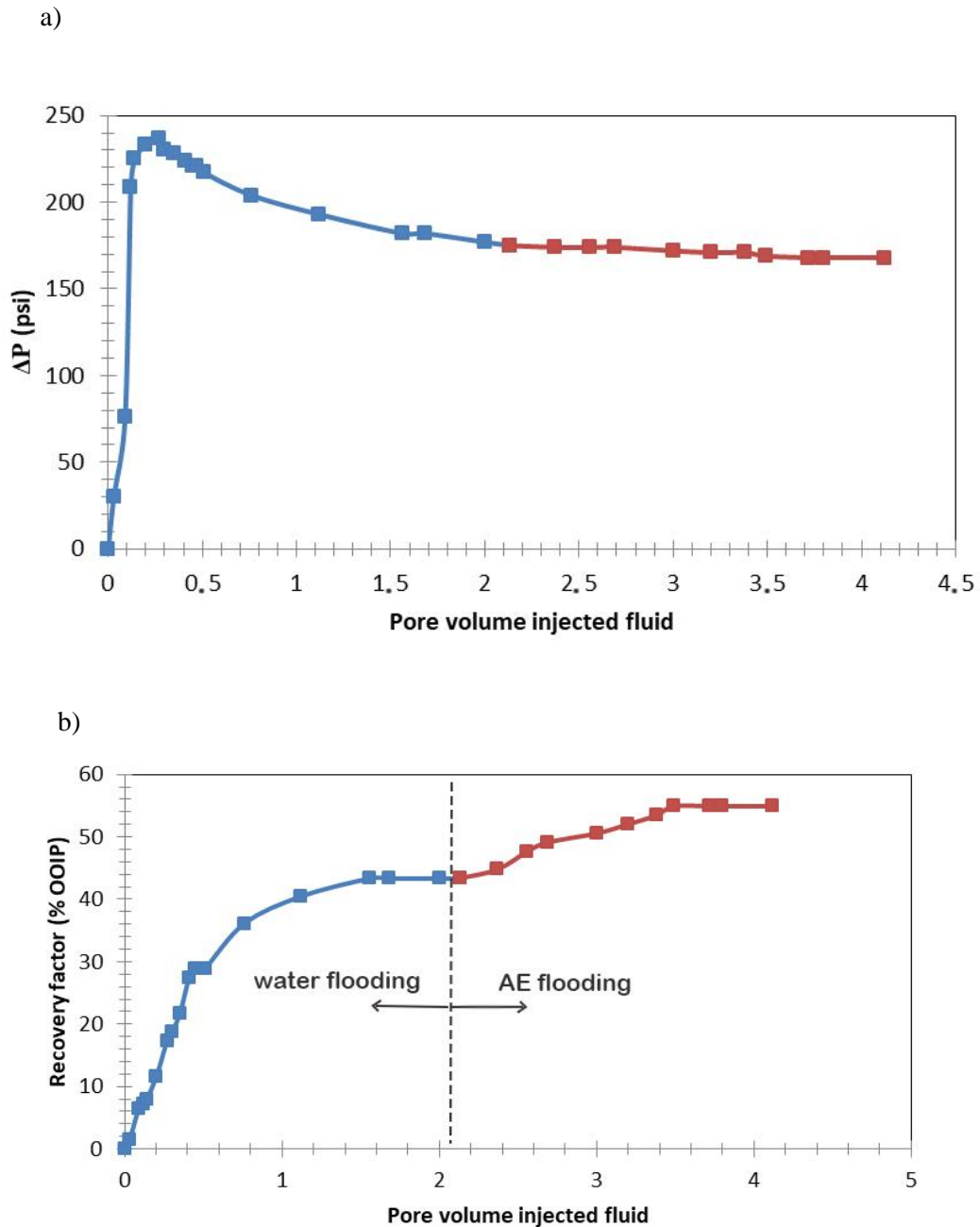
a)

b)

Figure 10

a) the oil recovery of the water flooding and the AE solution flooding as the tertiary method conducted on the carbonate core; b) the pressure drop versus the pore volume of the injected fluid of the water flooding and the AE solution flooding conducted on the carbonate core.

To evaluate the effect of the AE natural surfactant as a sole material on the improvement in the oil recovery factor, the recovery factor curves of Sumac (Faramarzpour et al. 2016), CTAB (Sofla et al. 2016), LArg (Asl et al. 2019), and Cedr (Sofla et al. 2016) obtained from the core flood displacement tests are compared with the oil recovery factor incremental graph of this study obtained from the core flooding experiment (see Figure 11).

The comparison presented in Figure 11 is conducted in a way that the lithological properties and the petrophysical characterizations of the rock, as well as the injectional conditions for the core flooding

experiments, of the used literature are 60% similar to the ones employed in the experiment of this study in order to achieve unity in the comparison. As shown in Figure 11, the oil recovery factor curves of five surfactants, including the Sumac, Cedr, CTAB, LArg, and AE demonstrate that the waterflooding conditions are approximately similar for the compared materials; therefore, the tertiary flooding of these materials can be compared successfully. The waterflooding recovery factor of the Sumac, Cedr, CTAB, LArg, and AE surfactants is 42, 45, 33, 41, and 43 respectively. Hence, the evaluation of the recovery factor of the tertiary oil recovery process conducted on the AE confirms that the value of the tertiary recovery factor is 54.9% and represents that this value is as high as the tertiary oil recovery values of the other surfactants and possesses; it leads to an 11.6% increase in the oil recovery factor regarding its recovery factor value for the second recovery.
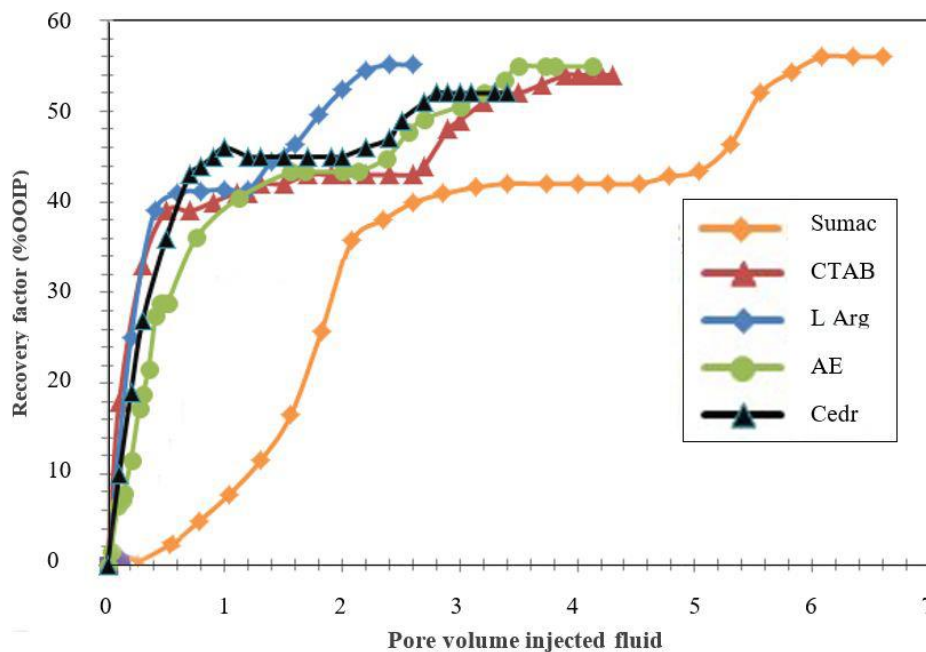

Figure 11

The oil recovery of the water flooding of Sumac (Faramarzpour et al. 2016); CTAB and Cedr (Sofla et al. 2016); LArg (Asl et al. 2019); and the AE as the tertiary oil recovery.

4. Conclusions

The goal of this study is to investigate the effect of the natural surfactant from the Albizia leaves extract on the wettability alteration, the improvement in the IFT, and the oil recovery of carbonate rocks. To this end, the IFT and wettability were measured by using the pendant drop and contact angle methods respectively, and the oil recovery was conducted by the core flooding test. The conclusions drawn from the findings of this work are as follows:

- The AE reduced the interfacial tension between the aqueous phase and the oil phase from 34 to 10 mN/;
- The critical micelle concentration of the AE was measured by the interfacial tension test, and the optimum surfactant concentration was found to be 3.5 wt %, which was also confirmed by the electrical conductivity test;
- The wettability test proved that the average contact angle of the base sample was measured at around 165.2°. Compared with the thin section samples at different concentrations of the AE,

oil wetting was reduced. The AE decreased the wettability toward a water-wet system as it reduced the contact angle by about 46.5% at the CMC.

- The amount of the water flooding recovery was 43.8% OOIP by the brine injection. Also, the AE solution flooding as the tertiary recovery method conducted on the carbonate core increased the amount of the oil recovery by up to 11.6% OOIP.

Nomenclature

A	Area [cm^2]
AT	Adhesion tension [dynes/cm or mN/m]
BV	Bulk volume [cm^3]
D	Equatorial diameter [cm]
IFT	Interfacial tension [dynes/cm or mN/m]
CA	Contact Angle [degree]
K_{abs}	Absolute Permeability [mD]
L	length [cm]
P_c	Capillary pressure [dynes/cm^2]
PV	pore volume [cm^3]
S_{or}	Residual Oil Saturation [dimensionless]
t	Time [s]
Greek Symbols	
μ	Dynamic viscosity of the fluid [cp]
μ_w	Water viscosity [cp]
μ_o	Oil viscosity [cp]
γ	Interfacial tension [dynes/cm]
θ	Contact angle measured through the water phase [degree]
ρ	Density of fluid saturating the core [g/cm^3]
$\Delta\rho$	Density difference between two fluids [g/cm^3]
Δp	Pressure drop [Psi]
$\emptyset$	Porosity [dimensionless]
Abbreviations	
CMC	Critical Micelle Concentration
EOR	Enhanced Oil Recovery
IFT	Interfacial tension
NP	Nanoparticle
PPM	Part Per Million
OOIP	Initial Oil in Place
AE	Albizia Extract

References

Alhseinat, Ahmadi, M. A., Arabsahebi, Y., Shadizadeh, S. R., and Shokrollahzadeh, S. B., Preliminary Evaluation of Mulberry Leaf-Derived Surfactant on Interfacial Tension in an Oil-Aqueous System: EOR Application, Fuel, Vol. 117, p. 749–755, 2014.

Asl, H. F., Zargar, G., Manshad, A. K., Takassi, M. A., Ali, J. A., and Keshavarz, A., Experimental Investigation into L-Arg and L-Cys Eco-Friendly Surfactants in Enhanced Oil Recovery by Considering IFT Reduction and Wettability Alteration, Petroleum Science, p. 1–13, 2019.

Ayirala, S.C. Surfactant Induced Relative Permeability Modifications for Oil Recovery Enhancement, M.S. Thesis, Baton Rouge, LA: Louisiana State University, 2010.

Bahraminejad, H., Khaksar, Manshad, A., Riazi, M., Sajadi, S.A., Keshavarz, ,A., CuO/TiO$_2$/PAM as a Novel Introduced Hybrid Agent for Water–Oil Interfacial Tension and Wettability Optimization in Chemical Enhanced Oil Recovery, The Journal of Energy Fuels. Vol. 33, No. 11, p. 10547–10560, 2019.

Eslahati, M., Mehrabianfar, P., Isari, A. A., Bahraminejad, H., Khaksar Manshad, A., Keshavarz ,A., Experimental Investigation of Alfalfa Natural Surfactant and Synergistic Effects of Ca^{2+}, Mg^{2+}, and SO_4^{2-} Ions for EOR Applications: Interfacial Tension Optimization, Wettability Alteration and Imbibition Studies CuO/TiO$_2$/PAM as a Novel Introduced Hybrid Agent for Water–Oil Interfacial Tension and Wettability Optimization in Chemical Enhanced Oil Recovery, J. Mol. Liq., Vol. 293, p. 111–573, 2020.

Barati-Harooni, A., Najafi-Marghmaleki, A., Tatar, A., Mohammadi, A.H., Experimental and Modeling Studies on Adsorption of a Nonionic Surfactant on Sandstone Minerals in Enhanced Oil Recovery Process with Surfactant Flooding, J. Mol. Liq., Vol. 220, p. 1022–1032, 2016.

Cheatham, S., Johnston, M., and Marshall, L., Useful Wild Plants of Texas, The Southeastern and Southwestern United States, the Southern Plains and Northern Mexico, Vol. I, Herbalgram, 1998.

Chhetri, A.B., Watts, K.C., Rahman, M.S., Islam, M.R., Soapnut Extract as a Natural Surfactant for Enhanced Oil Recovery, Energy Sources, Part A Recovery, Util. Environ, Eff. Vol. 31, No. 20, p. 1893–1903, 2009.

Deymeh, H., Shadizadeh, S. R., and Motafakkerfard, R., Experimental Investigation of Seidlitzia Rosmarinus Effect on Oil–Water Interfacial Tension: Usable for Chemical Enhanced Oil Recovery, Sci. Iran., Trans. C: Chem. Chem. Eng., Vol. 19, p. 1661–1664, 2012.

Emadi, S., Shadizadeh, S.R., Manshad, A.K., Rahimi, A.M., Nowrouzi, I., Mohammadi, A. H, Effect of Using Ziziphus Spina Christi or Cedr Extract (CE) as a Natural Surfactant on Oil Mobility Control by Foam Flooding, J. Mol. Liq. Vol. 293, 111573, 2019.

F. Faramarzpour, S. R. S., Experimental Investigation of Rhus Coriaria (Sumac) Extract on Wettability Alteration: Carbonate Rocks, Petroleum Engineering, Abadan Faculty of Petroleum Engineering, 2016.

Gargiullo, M. B., Invasive Plants of Asian Origin Established in the United States and Their Natural Enemies, Volume, the Journal of the Torrey Botanical Society, Vol. 132, No. 2, p. 375–375, 2005.

Ganie, K., Manan, M.A., Ibrahim, A., Idris, A.K., An Experimental Approach to Formulate Lignin-based Surfactant for Enhanced Oil Recovery, Int. J. Chem. Eng. 6. Article ID 4120859, 2019.

Kokila, K., Priyadharshini, S. D., and Sujatha, V., Phytopharmacological Properties of Albizia Species: A Review, Int J Pharm Pharm Sci, Vol. 5, No. 3, p. 70–3, 2013.

Lau, C. S., Identification and Quantification of Glycoside Flavonoids in the Energy Crop Albizia Julibrissin, Bioresource Technology, Vol. 98, No. 2, p. 429–435, 2007.

Madani, M., Zargar, G., Takassi, M.A., Daryasafar, A., Wood, D.A., Zhang, Z, Fundamental Investigation of an Environmentally-Friendly Surfactant Agent for Chemical Enhanced Oil Recovery, Fuel, Vol. 238, p. 186–197, 2019.

Manshad, A.K., Rezaei, M., Moradi, S., Nowrouzi, I., Mohammadi, A.H, Wettability Alteration and Interfacial Tension (IFT) Reduction in Enhanced Oil Recovery (EOR) Process by Ionic Liquid Flooding, J. Mol. Liq. Vol. 248, p. 153–162, 2017.

Pal, N., Kumar, N., Verma, A., Ojha, K., Mandal, A., Performance Evaluation of Novel Sunflower Oil-based Gemini Surfactant (S) with Different Spacer Lengths: Application in Enhanced Oil Recovery, Energy Fuels, Vol. 32, No. 11, p. 11344–11361, 2018.

Pal, N., Saxena, N., Laxmi, K.D., Mandal, A, Interfacial Behavior, Wettability Alteration and Emulsification Characteristics of a Novel Surfactant: Implications for Enhanced Oil Recovery, Chem. Eng. Sci., Vol. 187, p. 200–212, 2018.

Pordel, M., Shadizadeh, S. R., and Jamialahmadi, M., A New Type of Surfactant for Enhanced Oil Recovery. Pet. Sci. Technol., Vol. 30, No. 6, p. 585–593, 2012.

Rahmati, M., Mashayekhi, M., Songolzadeh, R., Daryasafar, A, Effect of Natural Leaf-Derived Surfactants on Wettability Alteration and Interfacial Tension Reduction in Water–Oil System: EOR Application. J. Jpn. Pet. Inst. Vol. 58m No. 4, p. 245–251, 2015.

Ravi, S. G., Shadizadeh, S. R., and Moghaddasi, J., Core Flooding Tests to Investigate the Effects of IFT Reduction and Wettability Alteration on Oil Recovery: Using Mulberry Leaf Extract. Petrol. Sci. Tech., Vol. 33, p. 257–264, 2015 .

Rostami, A., Hashemi, A., Takassi, M.A., Zadehnazari, A, Experimental Assessment of a Lysine Derivative Surfactant for Enhanced Oil Recovery in Carbonate Rocks: Mechanistic and Core Displacement Analysis. J. Mol. Liq. Vol. 232, p. 310–318, 2017.

Saxena, N., Saxena, A., Mandal, A., Synthesis, Characterization and Enhanced Oil Recovery Potential Analysis Through Simulation of a Natural Anionic Surfactant. J. Mol. Liq. Vol. 282, p. 545–556, 2019.

Shadizadeh S. S. and Kharrat Riyaz, Experimental Investigation of Matricaria Chamomilla Extract Effect on Oil–water Interfacial Tension: Usable for Chemical Enhanced Oil Recovery, Petroleum Science and Technology, Vol. 33, p. 901–907, 2015.

Sofla, S. J. D., Sharifi, M., and Sarapardeh, A. H., Toward Mechanistic Understanding of Natural Surfactant Flooding in Enhanced Oil Recovery Processes: The Role of Salinity, Surfactant Concentration and Rock Type, Journal of Molecular Liquids, Vol. 222, p. 632–639, 2016.

Urbina-Villalba German, Reif Isaac., Simple Theoretical Framework for a Systematic Molecular Dynamics Study of the Interfacial Properties of Oil/ Water Systems in the Presence of Surfactant Molecules, Colloids Surf A: Physicochem Eng. Aspects, Vol. 106, No. 2–3, p. 90–175, 1996.

Zendehboudi, S., Ahmadi, M.A., Rajabzadeh, A.R., Mahinpey, N., Chatzis, I, Experimental Study on Adsorption of a New Surfactant onto Carbonate Reservoir Samples—Application to EOR, Can. J. Chem. Eng. Vol. 91, No. 8, p. 1439–1449, 2013 .

Zhang. H. and Setzer, W. N., The Floral Essential Oil Composition of Albizia Julibrissin Growing in Northern Alabama, American Journal of Essential Oils and Natural Products, Vol. 1, No. 2, p. 41–42, 2013.