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Study of Natural Nonionic and Cationic Surfactants Interactions in the Presence of Divalent Ions and their Effects on Enhancing Oil Recovery

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Abstract

Although water and gas injection within enhanced oil recovery has garnered considerable attention in the contemporary era, some engineers argue that alternative and cost-effective methods could play a pivotal role in this field. The utilization of substances like surfactants and polymers is seen as an innovative technique that has substantially impacted the oil sector; however, the large-scale production of such materials is financially burdensome. Furthermore, manufacturing these substances results in hazardous wastes, posing risks to both human health and the environment, ultimately leading to extensive and irreversible pollution. Adopting natural surfactants emerges as a viable solution with relatively high efficacy. These plant-derived surfactants, extracted from indigenous plant leaves, are cost-effective, biodegradable, and pose no threat to human health or the environment. Incorporating these natural surfactants in oil-related experiments has yielded satisfactory outcomes, showcasing their effectiveness in reducing the interfacial tension between water and oil, modifying crude oil viscosity, and isolating heavy components of crude oil. Using conventional methods in the industry, such as water and gas injection, only leads to small exploitations, and in general, a considerable amount of oil remains in place. However, using modern methods, such as surfactant injection, these efficiencies increase to 30-40%. According to the micromodel injection tests, maximum recovery factors are achieved when natural surfactants interact well with divalent ions; the cedar non-ionic and Rosemary cationic surfactants have improved efficiency by 33% and 46%, respectively.

Keywords: Natural Surfactants, Zizyphus Spina Christi, Salvia Rosmarinus, Interfacial Tension, Biodegradable.

Introduction

Using conventional oil production processes, relying on natural driving forces leads to the recovery of only 30% of the total oil reserves, which is insufficient to meet the increasing energy demand. Therefore, modern technologies have been developed to increase reservoir production. Surfactant injection has developed as a method for enhanced oil recovery, such that reducing the interfacial tension between water and oil leads to increased displacement efficiency. Surfactants, or surface-active agents, are substances that, when used in minimal amounts, alter the surface tension of fluids [1]. The implications of surface active agents in petroleum upstream have attracted significant attention in recent decades, specifically in the chemical stimulation of petroleum reservoirs and enhanced oil recovery (EOR) ends, such as surfactant flooding. However, one of the biggest challenges of implementing surfactants for EOR purposes is adsorption fate or loss to the rock surface while surfactant is injected through the porous media. The new proposed surfactant has no harmful effects on the environment because it is originally made from the Zyziphus Spina Christi, a type of tree that is easily accessible and very cheap in Middle Eastern oil-producing countries. This means that running surfactant flooding as an EOR method is economically viable. However, further studies are essential to examine the technical effectiveness of this surfactant for EOR procedures with high performance [2]. Surfactants are classified as organic materials, and they are structurally amphiphilic. It means they are made up of two groups: hydrophilic and hydrophobic. In other words, surfactants contain a part that is insoluble in water and a part that is soluble in water. By minimizing the interfacial tension at the interface between two phases, surfactants are essential in solutions Considering the effects of surfactants in reducing interfacial tension, increasing crude oil mobility, and influencing the polar fractions of crude oil, their application significantly aids recovery processes. The use of low-salinity water is one of the

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processes suitable for the industry, as the lower the number of dissolved ions, the more the reaction between substances reaches a minimum; in other words, the use of low-salinity water enhances the performance of substances such as surfactants. Therefore, using this feature can significantly enhance oil reservoir recovery using divalent ions in low-salinity water [3]. In addition, the schematic of surfactant adsorption process is shown in Fig. 1.

The droplet profiles at the moment of falling for different aqueous solutions of Seidlitzia Rosmarinus inside the oil phase are presented in Fig. 2. In addition, as the surfactant concentration increases, the resistance to drop, i.e., the interfacial tension between the drop and the surrounding phase, is decreased. Moreover, the IFT values between the aqueous solution of Seidlitzia Rosmarinus and oil were calculated, as discussed in the above method. Furthermore, the plot of IFT values for the aqueous solution of Seidlitzia Rosmarinus/oil against the aqueous solution of Seidlitzia Rosmarinus concentration. In addition, the increase in surfactant concentration causes the oil/water interfacial tension to decrease. Also, beyond a critical point called Critical Micelle Concentration (CMC) of about 8% by weight, little change in interfacial tension is observed. In addition, this is because surfactants added more than CMC participate in micelle formation and do not increase the concentration at the water/oil interface. Furthermore, the chemical flooding process improves recovery efficiency at concentrations above the CMC due to the significant reduction in IFT [4].

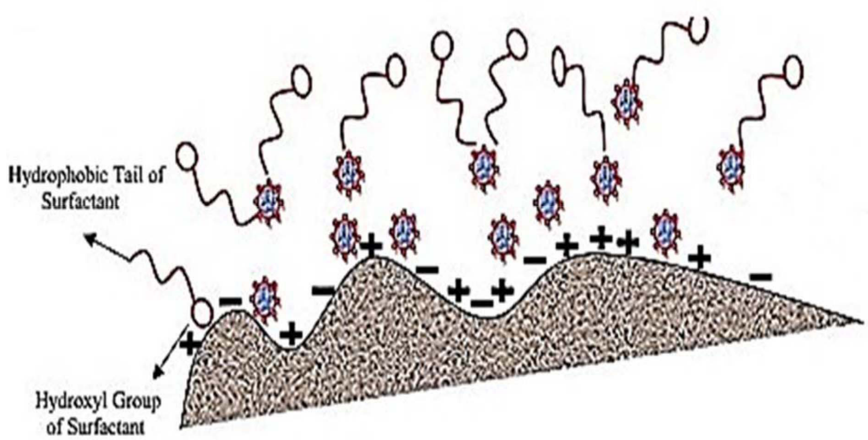


Fig. 1 Schematic of surfactant adsorption process [3].

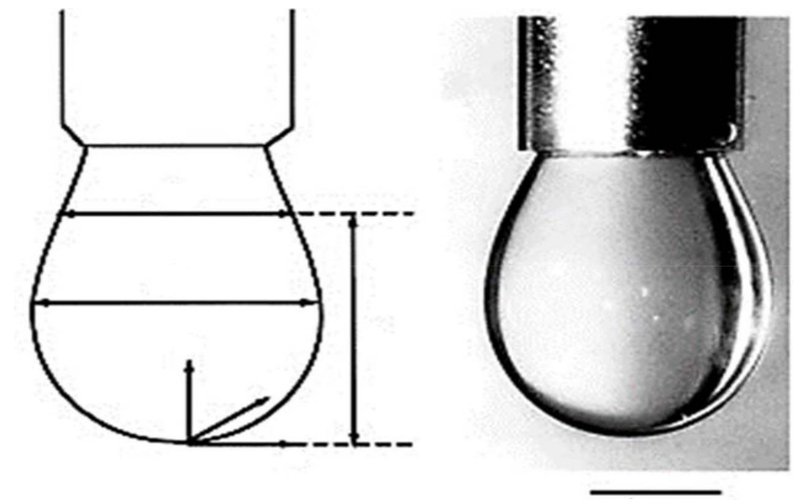


Fig. 2 Geometry and the equilibrium shape of the pendant drop with parameters [4].

$$\beta = 0.12836 - 0.7577(Ds/De) + 1.7713(Ds/De)^2 - 0.5426(Ds/De)^3 \quad (1)$$

$$De/2R0 = 0.9987 - 0.1971\beta - 0.0734\beta^2 + 0.34798\beta^3 \quad (2)$$

The maximum diameter of the drop (De) and its horizontal dimension (Ds) were measured to assess droplet behavior [4]. Moreover, the study of adhesion and cohesion between oil and water, the initial spreading coefficient, and the dispersion component of water's surface tension were also evaluated.

Additionally, the presence of ions (magnesium, calcium, and sulfate) in surfactant solutions, along with the electrical charge of ions and the chemical composition of surfactants (saponins), can enhance or diminish their effectiveness. Moreover, these plant-derived surfactants, being biodegradable and environmentally friendly, pose no threat to human health or ecosystems. Furthermore, their interaction with divalent ions does not lead to adverse effects [5]. Furthermore, the experimental results were compared with the published data of the same ternary systems (without inhibitors) to show the impact of low-concentration sulfide on formation [6].

Materials and methods

Materials

Oil sample

In this research, crude oil from the northern part of the Azadegan field was used. This crude oil, characterized by its high heavy hydrocarbon content, was analyzed using an Anton Paar device, which determined its viscosity to be 500 centipoises (Fig. 3). In addition, the interfacial tension of the crude oil was measured at 38.5 millinewtons per meter, and its asphaltene content was found to be 17.4%.

The ratios of the desired groups to specific wavelengths are shown in Fig. 4 and Table 1. These ratios can be determined by integrating and calculating the area under the diagrams. Fourier-transform infrared spectroscopy (FTIR) was employed to obtain the sample's infrared spectrum of absorption, emission, or photoconductivity. Moreover, an FTIR diagram typically shows how the sample absorbs different wavelengths of infrared light, revealing the material's molecular composition. Components of an FTIR Diagram, X-Axis (Wavenumber): The x-axis represents the wavenumber in units of cm^{1}. In addition, this is the reciprocal of the wavelength (1/λ) and is used to measure the energy of the IR radiation. Furthermore, higher wavenumbers correspond to higher energy. Y-Axis (Transmittance or Absorbance): The y-axis can represent either transmittance or absorbance. In addition, transmittance shows how much IR radiation passes through the sample (values usually range from 0 to 100%). Moreover, absorbance Indicates how much IR radiation the sample absorbs (higher values mean more absorption).

The diagram's peaks and bands feature peaks corresponding to specific molecular vibrations. Each peak relates to a functional group or bond within the molecule. Broad or sharp bands help identify the types of bonds (e.g., C-H, O-H, N-H, C=O, etc.).

Key Regions in FTIR Spectra (4000–2500 cm^{1}):

This region typically corresponds to stretching vibrations of bonds like O-H, N-H, and C-H (2500–1500 cm^{1}). The fingerprint region is complex but highly specific to individual compounds, useful for identification. Moreover, it involves bending vibrations below 1500 cm^{1}, which are often complex and unique for every substance. Scientists can identify functional groups and molecular structures in the sample by analyzing the positions, shapes, and intensities of the peaks.

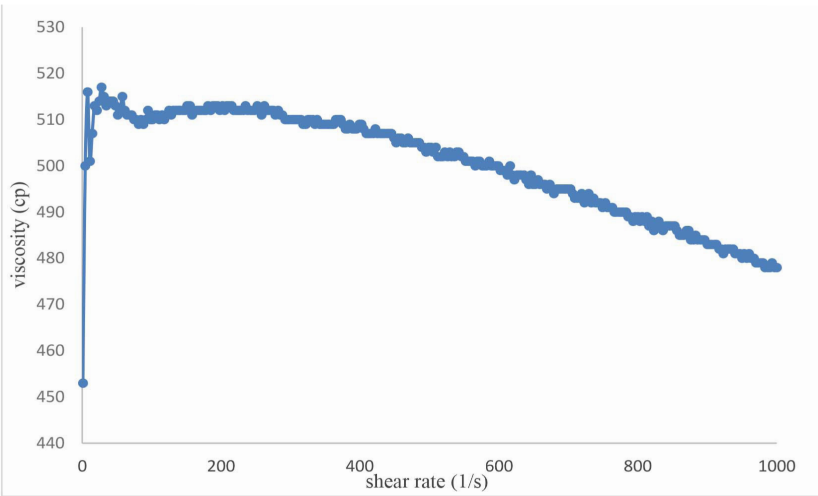


Fig. 3 Azadegan crude oil viscosity according to shear rate.

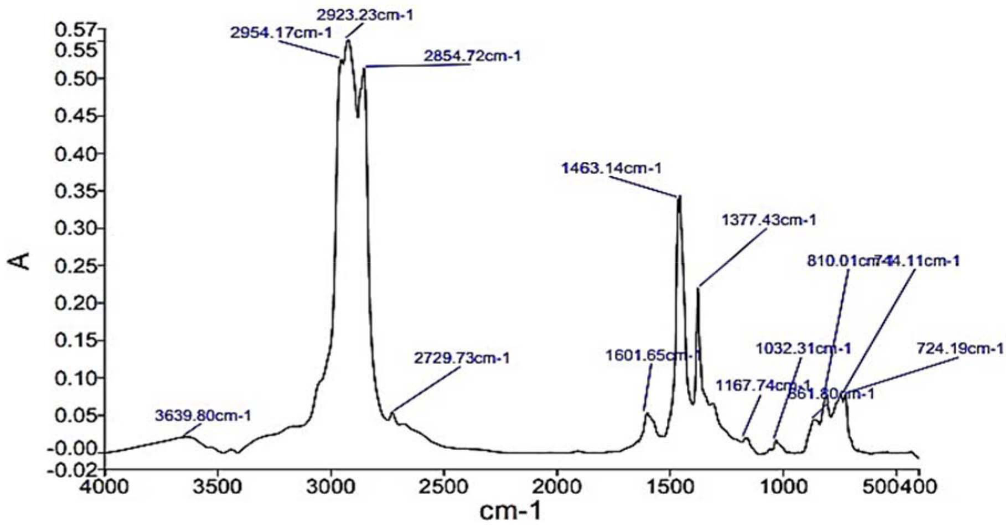


Fig. 4 FTIR of crude oil.

Table 1 Analyst of FTIR of crude oil with assignment.

Wavelength	Assignment	Area
3500-4000	N-H, O-H	5.49
3100-3500	N-H, O-H	7.67
3000-3100	C–H stretch in aromatic	9.09
2900-3000	CH_3 asymmetric stretch	47.63
2800-2900	CH_2 asymmetric stretch	28.7
2700-2800	CH_2 symmetric Stretch	5.14
1668-1800	C=O stretch in ketones, aldehydes, and carboxiclyc acids	0
1560-1668	Aromatic C-C stretch	3.58

Rate	
Carboxylic acid/Aliphatic	0
Carboxylic acid/Atromatic	0
Aromatic/ aliphatic	0.124738
Aliphatic length	1.659
Polar/Aliphatic	0.45
Polar/Aromatic	3.67597

Saponin

Saponin is a glycoside substance derived from various plants that produce foam upon agitation with water. Saponins possess a distinctive chemical structure and produce foam upon mixing with water, akin to detergents, with the ability to bind to water molecules and oil/fat compounds. In addition, a mixture was produced by using dried leaves of cedar and Rosemary mixed with distilled water and left for 20 days in a 90-degree oven. Subsequently, filtration paper was employed to refine their solutions, and upon desiccation of the residues, the primary active agents, specifically saponins, were acquired. Also, the critical micelle concentration of these substances, i.e., the cedar leaf, was determined to be 5%, while the Rosemary leaf was established at 8% by weight. Moreover, the amount of saponin in the materials is expressed in ppm, 40000 ppm Zizyphus spina-christi, and Rosemary. Moreover, The structure of Rosemary and leaf of Rozmary is shown in Fig. 5.

Within this research, for cedar and Rosemary leaves, a 4% weight solution was considered, and these concentrations were assumed to be constant to observe the effectiveness of low-salinity divalent ions on surfactant materials and their interaction with crude oil. In addition, the divalent ions used in this study included magnesium, calcium, and sulfate, and their concentrations were calculated using a stoichiometric ratio. Moreover, a constant selective concentration of 0.01% mol of these ions was chosen for the experimental assessments. The structure of Zizyphus Spina Christi and Zizyphus spina-christi leaf is shown in Fig. 6.

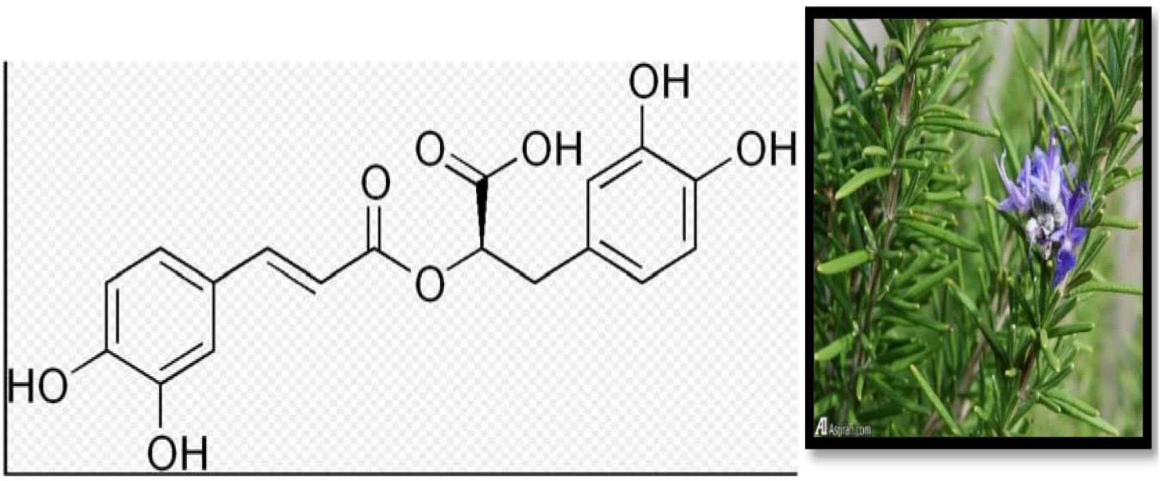


Fig. 5 The structure of Rosemary and leaf of Rozmary [1].

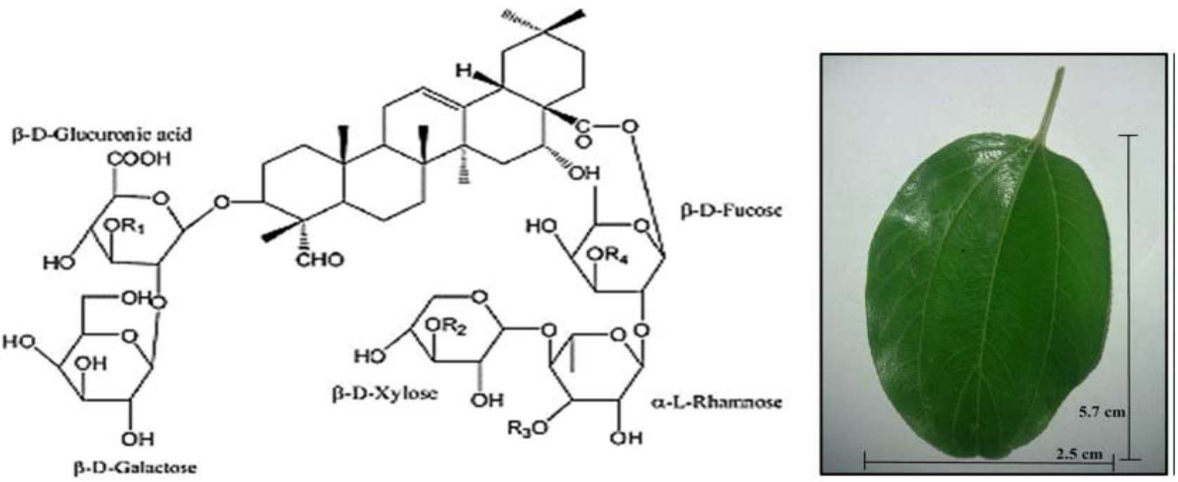


Fig. 6 The structure of Zizyphus Spina Christi and Zizyphus spina-christi leaf [4].

Divalent Ions

In this study, the synergistic effect of divalent ions in combination with natural surfactants and distilled water was investigated. Moreover, smart water was produced using this brine to examine its effect on oil recovery. Furthermore, divalent salts utilized included $MgCl_2$, $CaCl_2$, and Na_2SO_4. In industrial applications, minimizing the amount of salt is advantageous as it reduces costs. Also, the type of salts, as well as their production and purification methods, can be costly. Therefore, a single divalent concentration of 0.01% mol was used in this study, following the molarity percentage formulation.

Another crucial aspect pertains to ionic strength, manifesting when salts dissolve in water and undergo ionization, segregating into anions and cations. Moreover, this factor is more significant in divalent salts, so we used them to create smart water. In addition, the primary objective was to scrutinize the interplay between this ionization process, surfactants, and oil. To accurately interpret the results and ascertain whether the presence of salts has led to changes in crude oil or surfactants have caused these changes, it was imperative to maintain a constant concentration of surfactants throughout all experiments, especially when the brine concentration was varied. Moreover, % 0.01 mol of divalent ions which has been determined are shown in Table 2.

Materials and Methods

Viscosity

Viscosity pertains to the resistance exhibited by a fluid in response to shear stress. This kind of test is carried out using an Anton Paar (2018) device called a Rheometer, which is important for determining the effectiveness of the test. Furthermore, the equipment used for viscosity testing includes a viscometer bath, spindle, system, and computer. Also, these tests were conducted across shear rates ranging from 1 to 1000, with 300 data points collected for each viscosity measurement.

IFT

Surface tension, or interfacial tension, is the tendency of liquid surfaces to contract to the smallest possible surface area. This phenomenon occurs when a liquid surface comes into contact with another phase. Liquids tend to minimize their surface area as much as possible. The surface of a liquid behaves like an elastic membrane. Fig 7 demonstrates the elastic-like behavior of a liquid surface.

Micromodel

Researchers have focused on conducting injection tests in micromodels to understand and observe the mechanism of enhanced oil recovery. Furthermore, micromodels provide a visual representation of fluid movement and allow the assessment of pore-scale displacement mechanisms. The process begins by saturating the micromodel with sodium hydroxide and allowing it to rest for one hour. Then, the micromodel is rinsed with distilled water and dried. Finally, it is heated in a 200-degree oven for 15 minutes [16].

The next step involves preparing a mixture of 2% trichloromethyl silane and 98% toluene, which is then used to saturate the micromodel. Subsequently, the micromodel is left to rest for 5 minutes. Finally, the micromodel is rinsed with methanol and placed in a 100-degree oven for 24 hours. Oil is injected into the micromodel, followed by the injection of various materials. The displacement efficiency is then calculated [16]. Schematic of micromodel and the dimensions of micromodel are seen in Fig. 8.

Table 2 Determine % 0.01 mol of divalent ions.

component	Surfactant Solution	0.01 mol of component	ppm of component
$MgCl_2$	per 25 cc of solution	0.06 g	2033 ppm
$CaCl_2$	per 25 cc of solution	0.027 g	1110 ppm
Na_2SO_4	per 25 cc of solution	0.035 g	1420.4 ppm

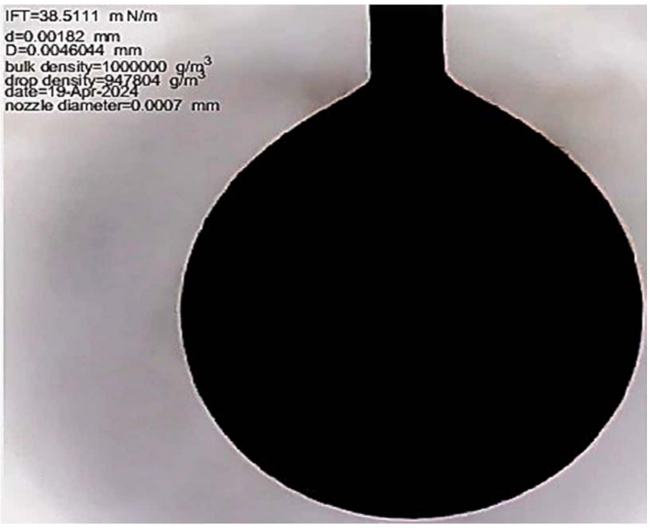


Fig. 7 Schematic of pendant drop.

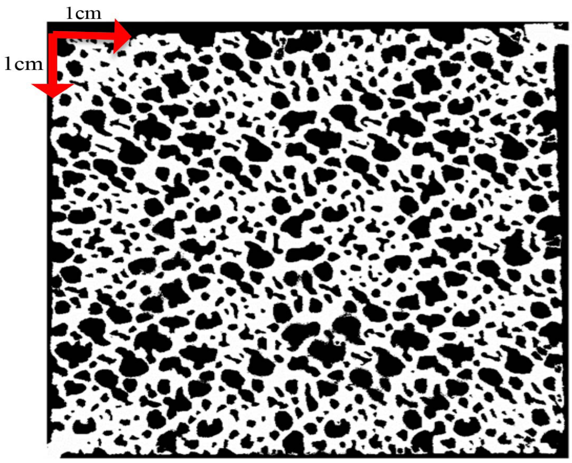


Fig. 8 Schematic of micromodel, The dimensions of micromodel are (6 x 6) cm^2.

Results and Discussions

Improvement Viscosity

The addition of divalent ions to surfactants and their interaction with oil results in a reduction in the viscosity of the crude oil. This observation indicates the compatibility of divalent ions with surfactants and enhances their effectiveness. When the viscosity of Newtonian fluids like oil decreases, the shear viscosity rate also diminishes. This correlation implies that the fluid can flow more quickly through the pores and fractures in rocks, leading to increased oil extraction from reservoirs. The reduction in the viscosity of crude oil due to the interaction between surfactants and ions signifies that the asphaltic heavy components of crude oil have transitioned into the emulsion phase. In contrast, the lighter components persist in the bulk of the oil. The purple line shows that a large part of the heavy and light components of crude oil during their interaction with the solution of surfactants and magnesium ions has caused them to become a suspension in the bulk phase of the oil, and this incompatibility has caused the up and down on this line. In addition, Changes in oil viscosity in the vicinity of cedar and Rosemary leaf solutions with 0.01 determining ions by paying attention to viscosity according to shear rate, is shown in Fig. 9.

Viscosity, which measures a fluid's resistance to flow, depends on several factors:

Temperature: as temperature increases, viscosity decreases because the increased thermal energy allows molecules to move more freely. Gases: As temperature increases, viscosity increases because higher temperatures lead to more frequent molecular collisions. However, this factor has been considered constant. Pressure, for most liquids, increasing pressure slightly increases viscosity since molecules are forced closer together. For gases, pressure typically has less effect on viscosity under normal conditions. However, this factor has been considered constant. Composition and Molecular Structure: Fluids with larger or more complex molecules (like polymers) tend to have higher viscosity because their molecular structures create more resistance to flow. Intermolecular forces (e.g., hydrogen bonding) also affect viscosity. Stronger intermolecular forces generally increase viscosity. This is an important agent of change in this research. Concentration (for mixtures or solutions): In solutions, higher solute concentrations generally increase viscosity due to more interaction between the solute and solvent molecules. External forces, shear forces (applied external forces), can influence the viscosity in non-Newtonian fluids, which change their viscosity when a force is applied (e.g., shear-thickening or shear-thinning fluids). These factors can vary significantly depending on whether the fluid is a liquid or gas and the nature of the fluid's molecular composition.

IFT

The interfacial tension graph demonstrates that cedar leaf surfactants exhibit optimal compatibility with calcium ions, while Rosemary leaf surfactants show the best compatibility with magnesium ions. Moreover, these combinations decrease interfacial tension and showcase superior compatibility compared to other pairs under examination. In many cases, reducing interfacial tension between two phases results in the separation of solutions. As the interfacial tension between water and oil decreases, displacement efficiency increases. This is evident in processes involving surfactants or ions and injection experiments conducted in micromodels. A higher oil recovery factor indicates better compatibility between surfactants, ions, and oil, improving the interaction between the solutions and crude oil. Furthermore, the images of interfacial tension with reports results are shown in Table 3.

Enhancement of Recovery Factor

Using Photoshop software, we calculated the number of oil pixels. We designated this value as the total number of pixels, subsequently iterating this procedure for the remaining micromodels to determine the ultimate recovery volume. This includes the micromodel examinations validating the tensile tests, evaluating the interfacial properties, and indicating the compatibility between ions and surfactants. Images of micromodels with reports results are shown in Fig. 10. In addition, recovery factor of surfactants is shown in Fig. 11.

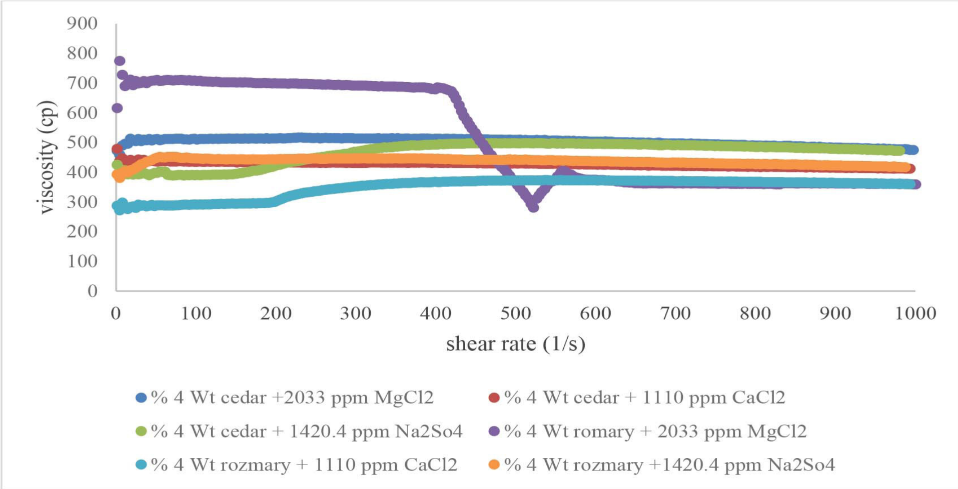
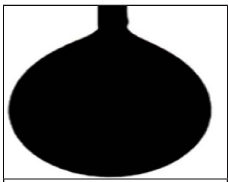
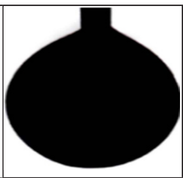
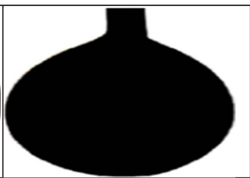
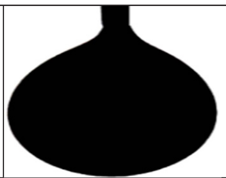
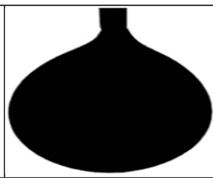
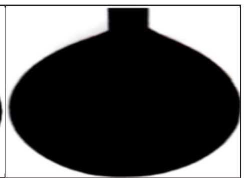


Fig. 9 Viscosity according to shear rate, Changes in oil viscosity in the vicinity of cedar and Rosemary leaf solutions with 0.01 determining ions.

Table 3 Images of interfacial tension with reports results.

					
4 Wt% Cedar + 2033 ppm $MgCl_2$	4 Wt% Cedar + 1110 ppm $CaCl_2$	4 Wt% Cedar + 1420.4 ppm $Na2SO_4$	4 Wt% Rozemary + 2033 ppm $MgCl_2$	4 Wt% Rozemary + 1110 ppm $CaCl_2$	4 Wt% Rozemary + 1420.4 ppm $Na2SO_4$
38.8 mN/m	15.7 mN/m	27.2 mN/m	18.9 mN/m	34.3 mN/m	25.4 mN/m

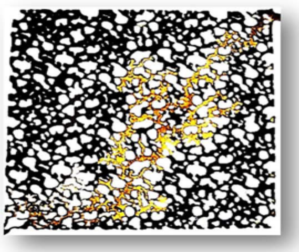
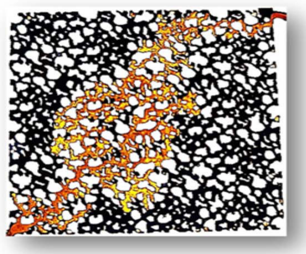
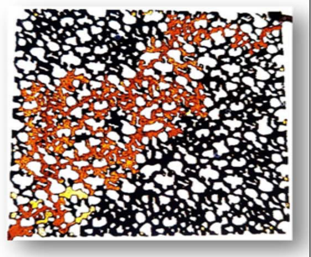
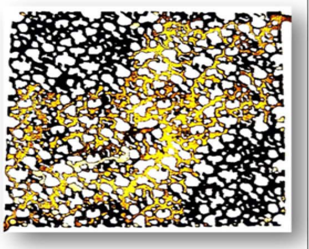
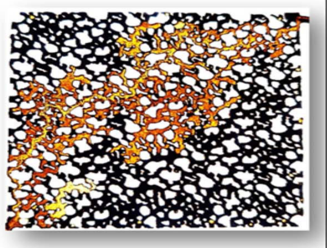
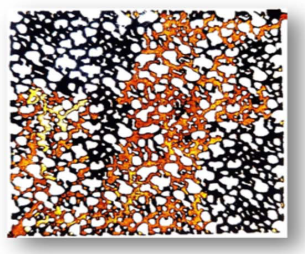
 a)	 b)	 c)
a) 4 Wt% Cedar + 2033 ppm $MgCl_2$	b) 4 Wt% Cedar + 1110 ppm $CaCl_2$	c) 4 Wt% Cedar + 1420.4 ppm Na_2SO_4
 d)	 e)	 f)
d) 4 Wt% Rozmary + 2033 ppm $MgCl_2$	e) 4 Wt% Rozmary + 1110 ppm $CaCl_2$	f) 4 Wt% Rozmary + 1420.4 ppm Na_2SO_4

Fig. 10 Images of micromodels with reports results.

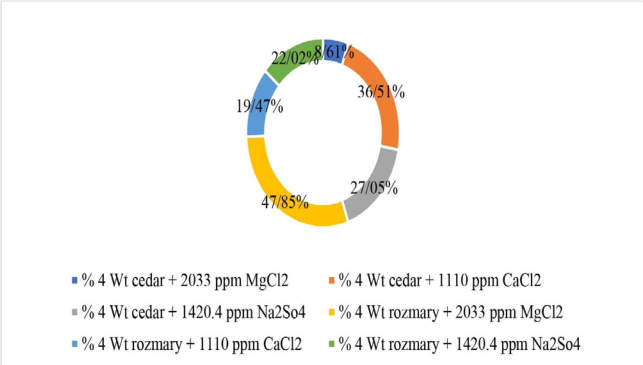


Fig. 11 Recovery factor of surfactants.

Adding divalent ions to surfactants and establishing ionic bonds between them, along with their interaction with oil, have reduced the viscosity of crude oil in certain instances. In the sequence of interfacial tension evaluations, it was observed that cedar surfactant exhibited the most optimal performance when paired with calcium ions. Conversely, rosemary surfactant demonstrated its highest efficiency when combined with magnesium ions. In the series of micromodel tests, adding divalent ions to the surfactants and injecting them into the micromodel increased the final recovery. Moreover, this outcome was consistent with the predictions from the interfacial tension assessments, where the micromodel tests displayed the highest displacement efficiency. The recovery rate of calcium ion with the cedar surfactant was about 36%, while the magnesium ion with the rosemary surfactant reached approximately 47% final recovery. In conclusion, the most significant effects can be observed if the best performance is achieved between natural surfactants and desired ions.

According to Fig 12, the beginning of the injections in the micromodels was steady, and then the injections were evaluated at times of 20 minutes, 1 hour, 2 hours, 3 hours, and finally, 4 hours. Based on the final recovery factor and considering the volume of the injection pore, it can be concluded that the reduction of interfacial tension of crude oil during the interaction with divalent ions resulted in the cedar solution with calcium ion and the rosemary solution with magnesium ion demonstrating the best performance. According to the results in Fig 12 these effects can be directly correlated with the size of the recovered pores. As it is clear from the size of the pores, the most prominent circle among the circles in the diagrams corresponds to the highest percentage of the recovery factor by the desired ions, i.e., magnesium and calcium, which is located at the breaking point of the diagrams and has the highest percentage.

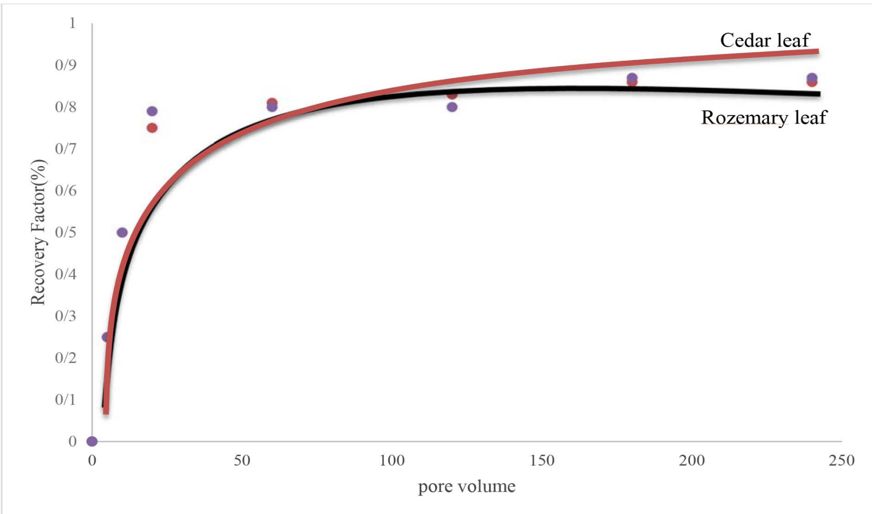


Fig. 12 Diagram of recovery factor according to pore volume for Cedar and Rozemary surfactant.

Conclusions

1. The addition of divalent ions to surfactants is attributed to their inherent nature and mutual compatibility, which reduces crude oil's interfacial tension.
2. The decrease in crude oil viscosity results from the interaction between surfactant solutions, ions, and the oil itself.
3. The specific charges of ions and surfactants and their interaction with oil separate heavier components within crude oil, retaining them in the emulsion phase. Consequently, the viscosity of the upper oil layer is lighter, accompanied by a reduction in surface tension.
4. Reducing crude oil's surface tension and injecting solutions resulted in the highest productivity in micromodel tests, and the recovery rate increased.
5. The optimal interaction between divalent ions and surfactants depends on the type of surfactant utilized, with the effectiveness of their interaction yielding significant outcomes in various tests.

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