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Enhancement of Oil Recovery (EOR) by Ionic Liquid Assisted Low Salinity Water in Carbonate Reservoir

Maraim Kh. Uoda *, Hussein Q. Hussein ** , Rana R. Jalil ***
*Chemical Engineering Department, College of Engineering, University of Baghdad, Baghdad, Iraq
E-mail: eng.maraim89alkhleel@gmail.com
**Thi.Qar Oil Company, Nasiriyah. Iraq.
E-mail: husseinqassab@coeng.uobagdad.edu.iq
***Petroleum Research and Development Center. Baghdad, Iraq
E-mail: rana_rasool@prdc.oil.gov.iq

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Abstract

Carbonate rocks hold approximately 40% of the world's oil. Due to their low waterflood recovery rates, these reservoirs are ideal candidates for enhanced oil recovery (EOR) methods. Chemically enhanced oil recovery (CEOR) techniques, such as surfactant flooding, can increase these reservoirs' output by modifying wettability and decreasing interfacial tension (IFT). This study explored ionic liquids (ILs), a new type of surfactant, and their potential effects on wettability and IFT reduction. The study initially involved the experimental assessment of the interfacial tension and wettability, using the pendant drop and contact angle. The results reveal that this surfactant changes the wettability from 98◦ to 28◦ and the interfacial tension (IFT) from 3.13 to 1.25 dynes/cm at 250 ppm. Core flooding tests were conducted to examine the effect of ionic liquid concentration on the ultimate oil recovery from the carbonate oil reservoir's core plug. The outcomes reflected extra oil recoveries of 17% and 19.7% OOIP for 50 and 250 ppm, respectively at a flow rate of 0.667 cm^3/min, respectively. Core flooding tests achieved an additional recovery of 10.5% OOIP at a flow rate of 1 cm^3/min, using the highest ionic liquid concentration.

Keywords: Contact angle, Wettability alteration, Chemical Enhanced oil recovery (CEOR), Interfacial tension (IFT), Ionic liquid (IL).

1.Introduction

The US Department of Energy reports that world oil production accounts for barely one-third of total availability. Applying EOR techniques can boost oil output because, when need increases, supply becomes constricted, and the demand rises in response [1]. When all oil has been extracted by the main oil recovery process, the best way to boost reservoir oil production is to inject water. Another reason water injection is used is because it is inexpensive and has beneficial qualities that allow for the efficient release of stored oil. Water injection has the potential to spread via loading oil production and efficient oil replacement when the dosage is just right [2]. The method of secondary recovery most commonly used is flooding. The secondary recovery technology leaves a lot of oil behind because it only sweeps partially through the homogeneous reservoirs and because leftover oil is saturated in porous rock. Finding alternative methods of extracting the residual oil became critical following secondary recovery. Improved oil recovery, or EOR, was developed to achieve this objective [3].

When primary and secondary oil recovery methods no longer yield a sufficient amount of oil, enhanced oil recovery (EOR) is employed to increase oil production. The oil remains contained within the reservoir's rock pores due to its high viscosity and capillary forces. Thanks to injectable surfactant compositions, we can disrupt oil-rock contacts or increase sweep efficiency. This tertiary EOR method employs surfactants or microemulsion floods to reduce water-oil interfacial tension (IFT) or convert the rock's wettability from oil-wet to water-wet [4].

Ionic liquids have gained a lot of interest as a possible technological improvement tool in the last few decades. Organic salts, ionic liquids include both inorganic and organic cations, as well as organic and inorganic anions, and they melt at temperatures below 100 °C. When different types of cations and anions are mixed, Plechkova and Seddon (2008) estimate that between 1012 and 1018 synthetically accessible pairings are conceivable [5].

Solvents can be tailored to suit specific applications thanks to the large variety of solvent compositions. Many physical and chemical processes currently use volatile solvents and catalysts; however, ionic liquids have the potential to be a "greener" and "designer" alternative. To improve oil recovery, chemical-enhanced oil recovery (CEOR) makes use of ionic liquids as surface active agents to alter wettability and reduce IFT, among other features (Bera & Belhaj, 2016)[6].

By changing the length of the hydrophobic chain, ionic liquids have the power to affect the water-oil interface tension. A longer hydrophobic chain does assist ILs cling to two different phases. The reason behind this is that by extending the distance between the hydrophilic and hydrophobic heads, emulsions are made more stable. According to Hezave et al. (2013), this makes the trapped oil easier to transport. Recovering oil using ionic solutions [7].

According to earlier studies (Yousefi et al. 2017 [8]; Sakthivel et al. 2017 [9]), ionic liquids have the ability to decrease IFT, regardless of the temperature or presence of salt. We provide here the findings of an investigation into the effects of a novel set of critical parameters on oil recovery by ionic liquid injection. An interfacial tension (IFT) reading was taken using a spinning drop tensiometer in order to identify the impact of brine salinity and ionic liquid concentration. Using contact angle measurements, researchers investigated how different amounts of ionic liquid affected the wettability of the modified rock.

To determine how changing the ionic liquid concentration and flow rate affected the increased oil recovery, multiple core flooding experiments were conducted.

2. Experimental Section

2.1. Materials

To measure pore volume (PV) , porosity, and permeability, core plugs were saturated with formation water brine. Water for injection as secondary recovery came from the third river in southern Iraq after treatment. See Table 1 for the brine's chemical composition (1). Table (2) lists the qualities of the low-salinity injection water used in these tests. Macklin Company, China, supplied dodecyl pyridinium chloride. The general properties are given in Table (3). Fig.1 shows the surfactant chemical structure. The Thi-qar Oil Company supplied crude oil for these experiments. Crude oil's dynamic viscosity is 21.10 ^{o}C at 24-25 cp. Crude oil was diluted and improved for displacement by adding gas oil (75% by volume). This study used Nassiriyah Oil Field's Mishrif formation carbonate reservoir rock. The practical work flowchart is in Fig.2.

Table (1): Chemical Analysis of Formation Water.

Element	Concentration, (ppm)
	Brine
Chloride	**127498.655-96205**
Sulphate	**777.139-650**
Bromide	**863.741**
Total Carbonate	**72.928 -97.6**
Hydroxide	**0**
Butyrate	**< 5**
Cl :Br	**168.646**
Lithium	**3.988**
Barium	**< 2**
Strontium	**379.453**
Calcium	**14119.5 – 22500**
Magnesium	**4257.172-33000**
Sodium	**57076.4155 – 30920**
Potassium	**1366.26 – 320**
Sulphur	**249.55**
Total Iron	**48.998**
TDS	***206480.8185***

Table (2): Characteristics and Composition of Injection Water.

Element	Concentration, (ppm)
	Brine
CL^{-1}	**1952.5**
$SO4^{-2}$	**2012.12**
Na^{+1}	**1529**
Ca^{+2}	**1225**
Mg^{+2}	**1180**
TDS	**7898.62**

Table (3): Ionic liquid surfactant characteristics.

Surfactant name	Dodecyl Pyridinium Chloride
Chemical formula	C17H30CIN Or (C12Py)(Cl)
Melting point, (C^0)	>80
TGA, (C^0)	250
Appearance	White or faint red-yellow solid
purity	>98%
Molecular weight(g/mol)	283.88

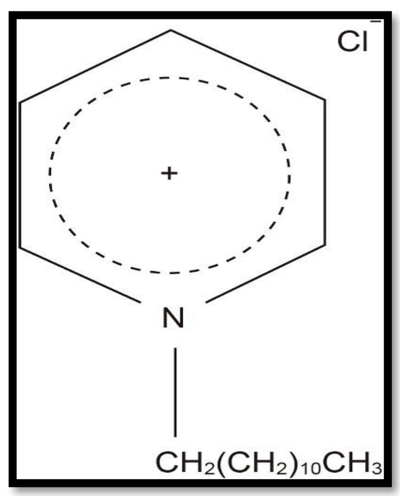

Figure 1. the surfactant chemical structure.

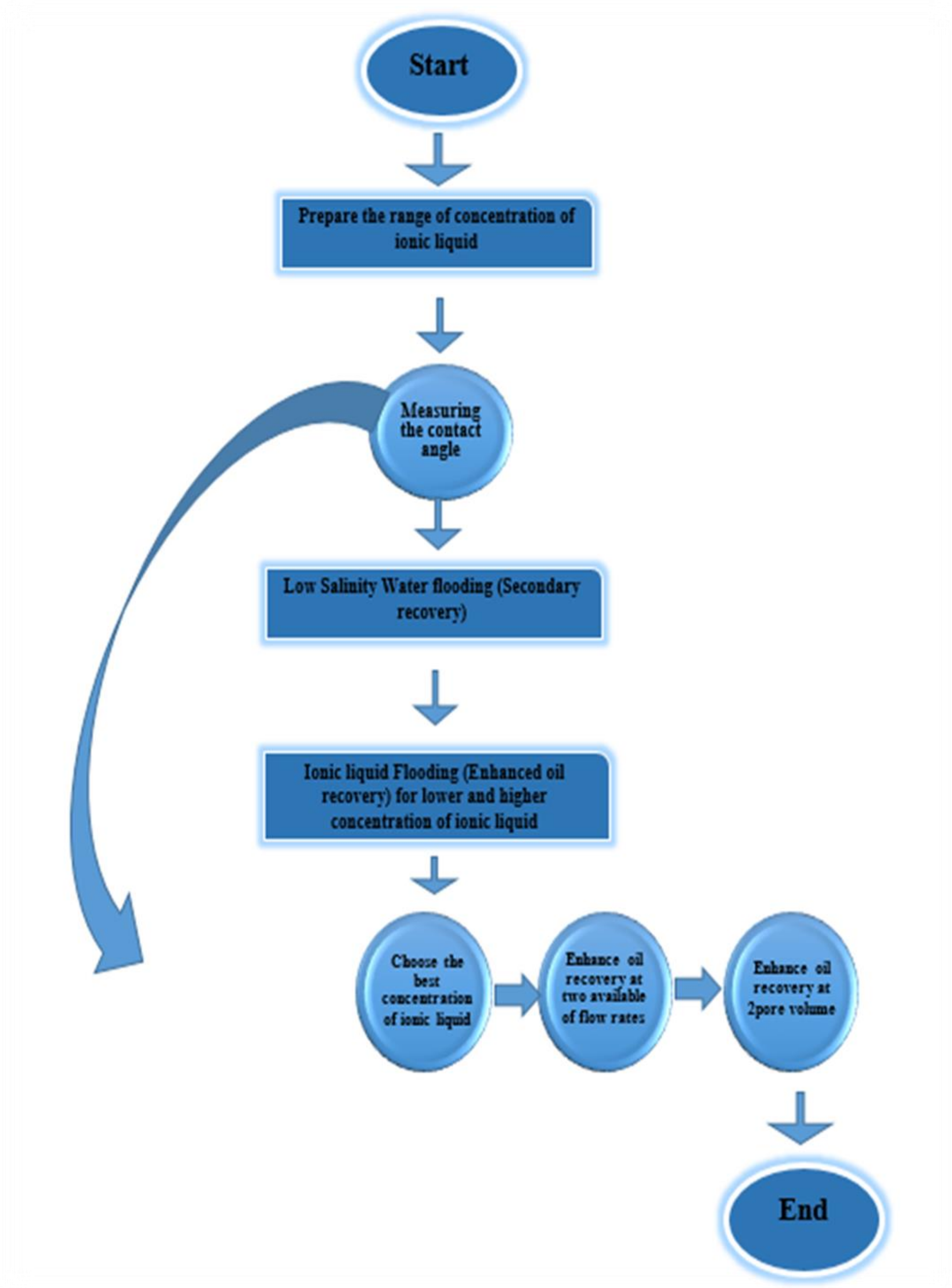

Figure 2. Flowchart of the procedure

2.2 Experimental Steps

2.2.1 Measurement of contact angle (CA) and interfacial tension

Carbonate rocks were split into thin bits so that the contact angle could be studied. The last step was to give the finished rock samples a good polish. The effect of ionic liquid concentration on wettability was investigated by immersing carbonate rock samples in solutions of 50, 100, 150, and 250 ppm for 10, 20, and 30 minutes, respectively, at 25°C [10][11].

The contact angle tests were conducted using an optical tensiometer called the Theta Lite. Between the needle's point and the cell's base, a single drop of distilled water trickled. The device's software can use a user-defined baseline to create tangent lines on either side of the drop. These lines will indicate the interface between the rock surface and the operation, and the software will also provide the average and magnitude of the contact angles on either side of the drop. Fig. 3 shows the process of determining contact angles using an optical tensiometer. Light came via a lens, and as seen in Fig.4, the interfacial tension was measured with a needle attached to the syringe piston via a steel line.

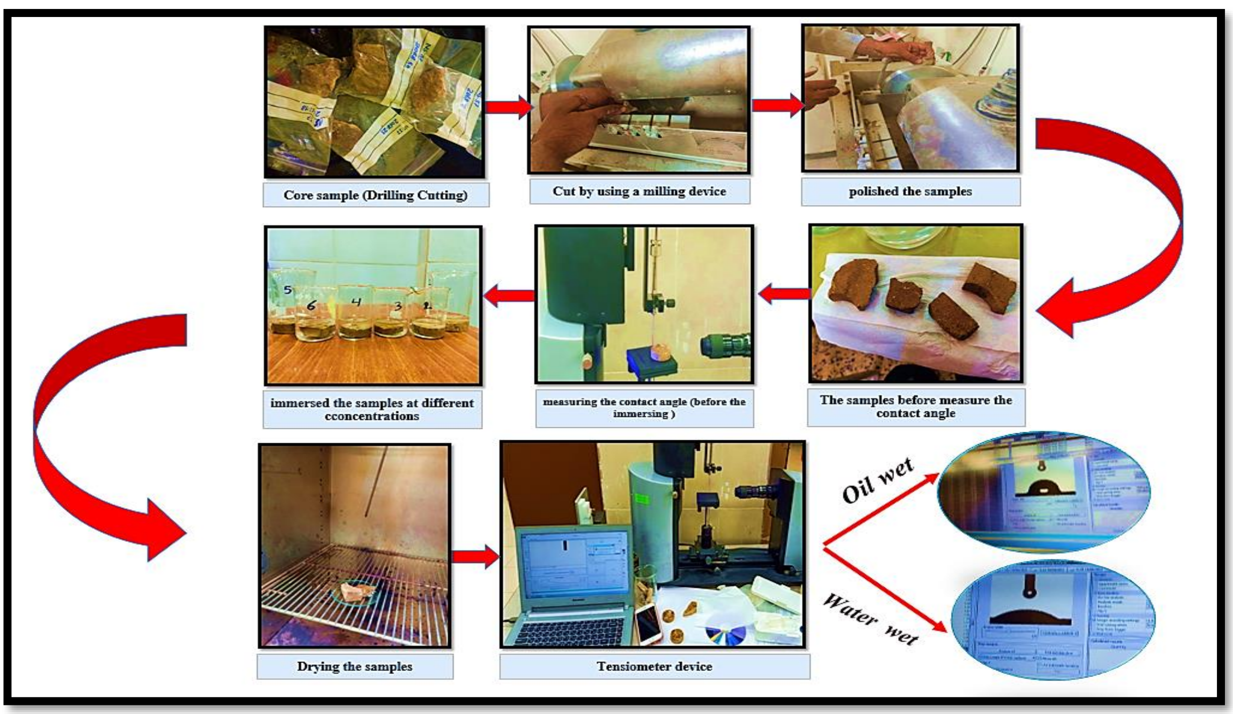

Figure 3. Shows the steps for measuring the contact angles.

Figure 4. (A) present the tensiometer apparatus. (B) When the interfacial tension is measured.

2.2.3 Core-Flooding Procedure

As seen in Fig. 5 [10] [12], a core flooding system diagram is provided. The core holder is equipped with pressure gauges to monitor variations in confining pressure and pressure over the core plug. The system is connected upon filling the accumulator with ionic liquids and brine (provided by the ISCO pump).

To put the core-flooding technique to the test, we applied the data collection approach. The process was completed by vacuum-assisted saturation of the core and then flooding it with formation water of different pore volumes. Formation water was constantly injected until the injection pressure was stable, which took some time [13][14][15].

Diluted crude oil was pumped into the water-filled deposit core to stop the production of further brine. Following its completion and the recording of the corresponding water and oil output figures, Secondary recovery employing low salinity water flooding was applied to carbonate core samples to comprehend the impact of LSW on these rocks. The LSW was injected to boost oil production. To evaluate the effectiveness of the injection procedure, the utilization of the ionic liquid known as Dodecyl pyridinium chloride was employed for territorial recovery. This was carried out at two different concentrations, along with various flow rates (1 ,0.667) cm^3/min, until two pore volumes (2PVs) were completed within the core plug.

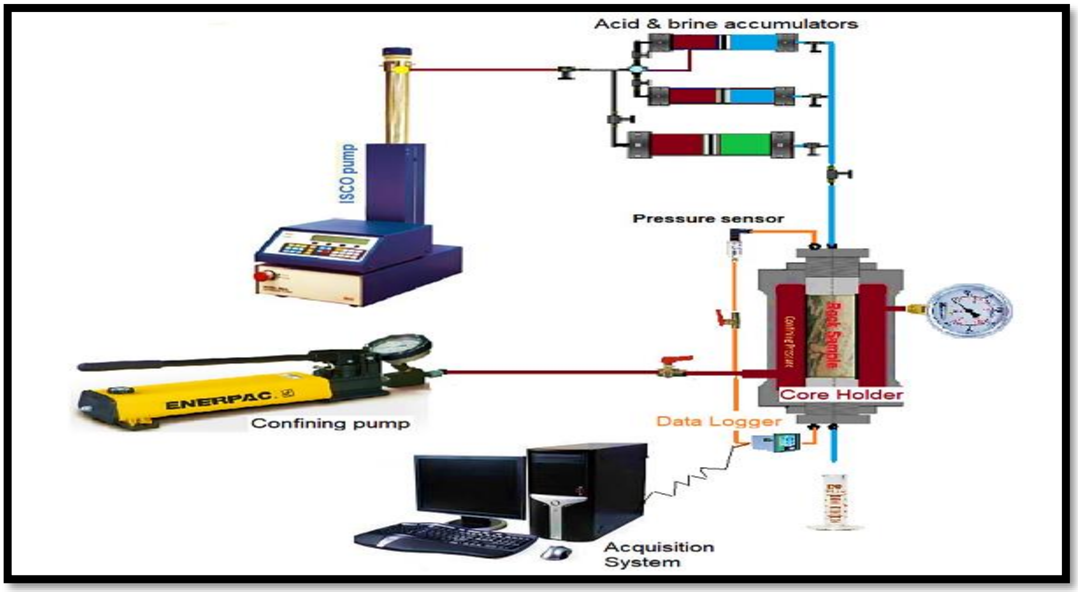

Figure 5. Schematic of the core flooding apparatus [10] [12].

3. Results and Discussion

3.1 Contact Angle for Ionic Liquid

Because of their surface-active properties, ionic liquids can alter a reservoir's capacity to retain moisture. When varying amounts of ionic liquids were introduced, the contact angle shrank, indicating a transition from a relatively oil-wet to a water-wet wettability. A decrease in contact angle from (98° to 40°), (101° to 38°), (103° to 32°), and (98° to 28°) when these chemicals (Ionic Liquids) is evidence that they are helpful for oil recovery [14]. Whenever a material's wettability changes, it's important to transition from an oil-wet to a water-wet state. Modifications to wettability are often facilitated by oil, rock, and ionic liquid processes. By studying the effects of ionic liquids on the wettability of oil-wet limestone and water-wet sandstone samples, Mohammed and Babadagli [15] aimed to better evaluate the efficacy of ionic liquids. Compared to surfactants and other agents, ionic liquids are superior in changing the wettability of oil-wet sandstone and limestone. This approach can be used to recover thermal oil, but it requires ionic liquids that are both stable and active, cooled to extremely high temperatures. Cao et al. [15] investigate how ionic liquids influence the behavior of contact angles in carbonate rock that has been wetted with oil.

Scientific research has shown that ionic liquids have the ability to reduce the oil-water contact angle, therefore changing the wettability of rocks from oil to water. As shown in Figure 3, the contact angle can be affected by ionic liquids and other chemicals that change wettability.

The varied charges of ionic liquids cause them to interact with rock surfaces in unique ways. When working with carbonate reservoirs, ionic liquids containing cations are preferable, whereas sandstone reservoirs are better suited to anions. As a qualitative measure of rock wettability, we measured and recorded the contact angles between surfaces of rocks and droplets of distilled water in various concentrations of ionic liquid. Figure 6 shows that after forty minutes of immersion in brine, a 45-degree contact angle between distilled water droplets on a polished surface of rock samples became obvious. This value decreased for various ionic liquid concentrations in brine due to variations in ageing durations. Fig. 6 shows that increasing the ionic liquid concentration from 50 ppm to 250 ppm resulted in more pronounced wettability changes. The wettability remains unchanged when the ionic liquid concentration is increased from 50 to 150 ppm, as compared to a higher concentration of 250 ppm, when tested and aged at the same conditions. Based on the results of the contact angle, it was demonstrated that ionic liquid solutions have the potential to transform oil-wet circumstances into neutral-wet and water-wet ones.

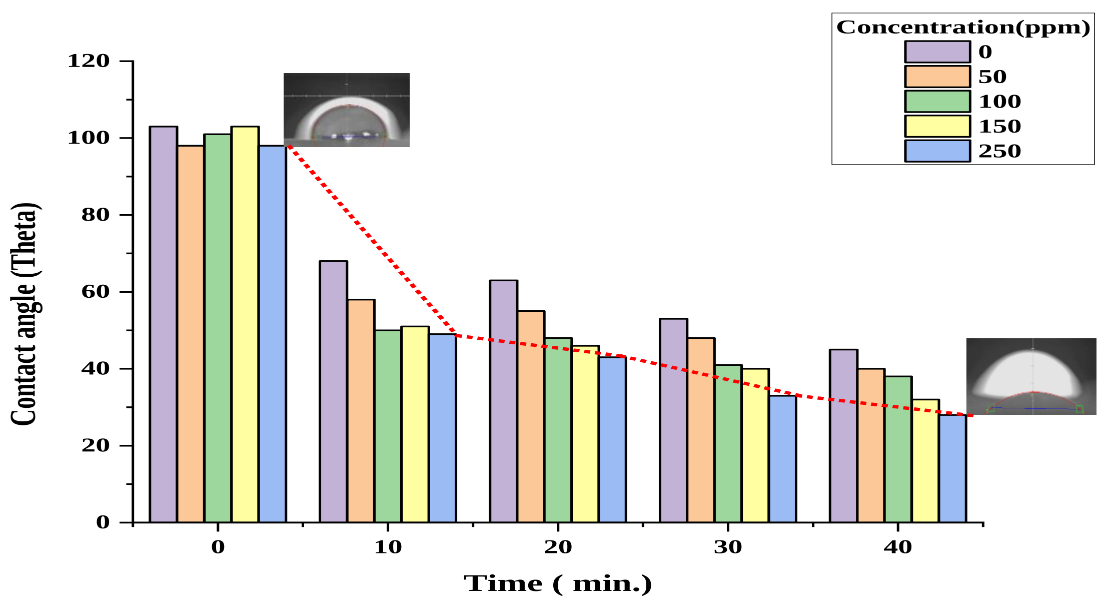

Figure 6. Variation in contact angle with time and concentration.

3.4 The effect on interfacial tension of an ionic liquid

Interfacial tension is a characteristic of interfaces between immiscible phases. Experiments on interfacial tension are shown in Fig. 7. According to this Figure,at a concentration of 250 ppm, the surface tension graph reveals a gradual flattening of the steep slope. The double hydrophilic-hydrophobic configuration of surfactant molecules causes them to disperse across the surface of a liquid. This arrangement permits the molecule's hydrophilic half to dissolve in water while its hydrophobic half stays out of it . The alteration of the interfacial tension between water and oil is caused by ionic liquids' extension of the hydrophobic chain. A lengthier hydrophobic chain corresponds to an increased propensity of ILs to adhere to two distinct phases. This is because a more stable emulsion facilitates the more complimentary circulation of confined oil when there is a greater distance between the hydrophilic and hydrophobic heads. As shown in Fig. 8, the results of this study warrant the consideration of ILs as a potential substitute surfactant for lowering the IFT between crude oil and water.

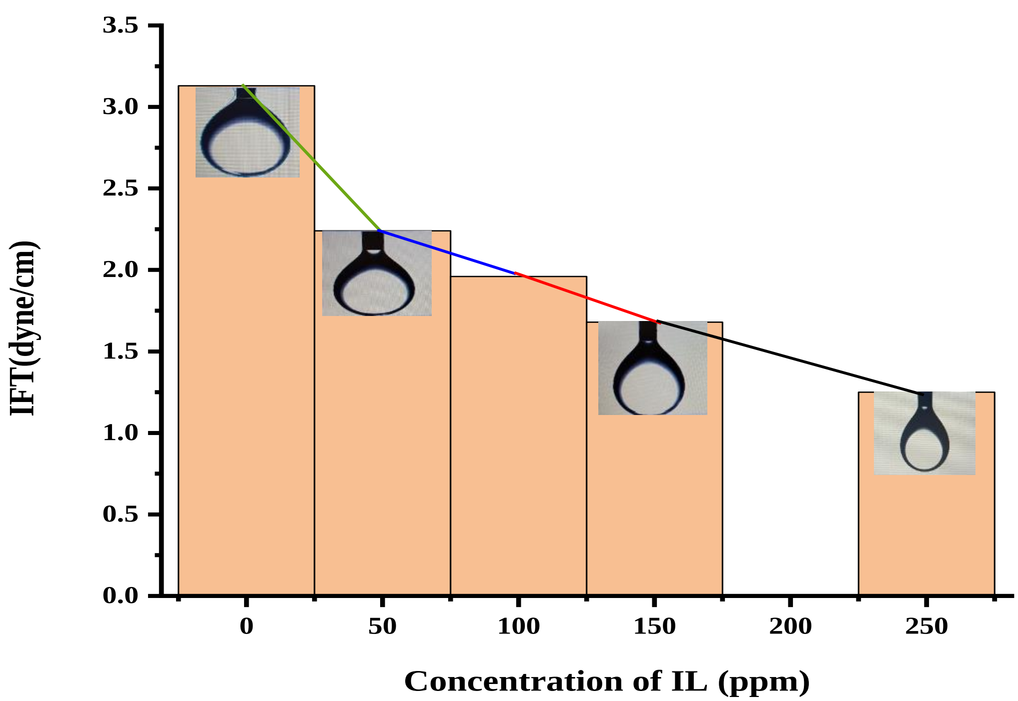

Figure 7. Variation in interface tension with respect to various concentrations.

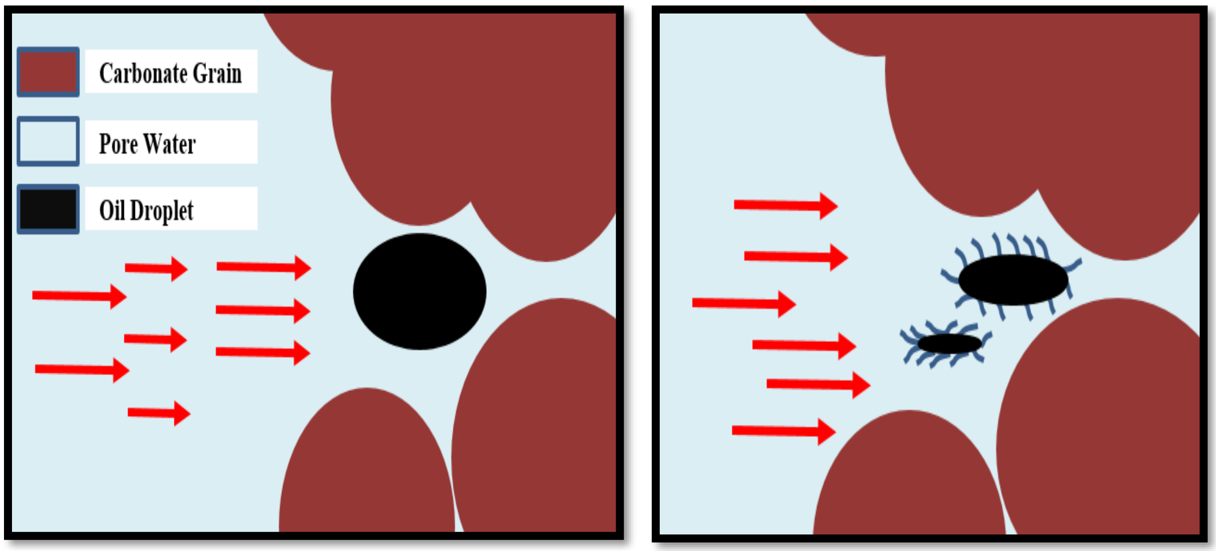

Figure 8: (A)Without Surfactant (High interfacial tension) (B)With Surfactant (Low interfacial tension)

3.5 Oil Recovery of Surfactant (Ionic Liquid) Flooding

3.5.1 Concentration's Influence on Oil Recovery Efficiency

Through laboratory flooding and displacement experiments using a production from the Nasiriyah Oil Field, this study explores the improved oil recovery performance of mixtures of cationic surfactant (dodecyl pyridinium chloride) and low salinity water. Evidence suggests that ionic liquid can improve oil recovery from carbonate reservoirs. The results show that one of the elements leading to better oil recovery is a shift in rock wettability towards more water-wet conditions. Carbonate surfaces, which are positively charged, are repelled by cationic surfactants, particularly in polar areas. The result is that the rock's surface comes into touch with the nonpolar components. The result is an increase in the already high humidity levels in the polar areas due to the water's contact with the rock-fluid interface. What Castro Dantas et al. (2014) found is in line with this theory [16].

After analysing how different concentrations of ionic liquid in hybrid solutions affected the contact angle, it was found that injecting 250 ppm was the sweet spot for the core plug, resulting in a 71% reduction in contact angle compared to a low concentration of 50 ppm. With an initial oil saturation of 67% for plug #8 and 61% for plug #10, the pore volumes (PVs) were 21.81 cm^3 and 21.93 cm^3, respectively.Carbonate core plugs 8 and 10 had porosity and permeability values of 26.2% and 4.17 mD, respectively, whereas plug 10 had 26.34% and 6.84 mD. It was necessary to inject two PVs of LSW into the core in order to start flooding. This study made use of tertiary and secondary recovery methods. Fig.9 shows the different recovery profiles through the carbonate rock plug. Injecting the low-salinity brine resulted in an increase of the oil recovery factor to 0.7 PVs for plugs 8 and 10, respectively, yielding 48% and 63% recovery, as shown in the figure. After injecting an additional 1.65 PVs of brine, oil recovery showed a small improvement. Tertiary recovery was started by injecting two PVs of the hybrid solutions as a chemical flooding after two PVs of LSW were completed. Plug #8 recovery increased from 61% to 64.5% OOIP and plug #10 recovery increased from 63% to 73.2% OOIP after 0.8 PVs of the chemical solution were injected. Oil production then began. Two PVs of the solution made with 50 ppm ionic liquid and LSW added 17.1% OOIP to the final product, whereas the optimal concentration made with 250 ppm ionic liquid and LSW added 19.7% OOIP to the final product.

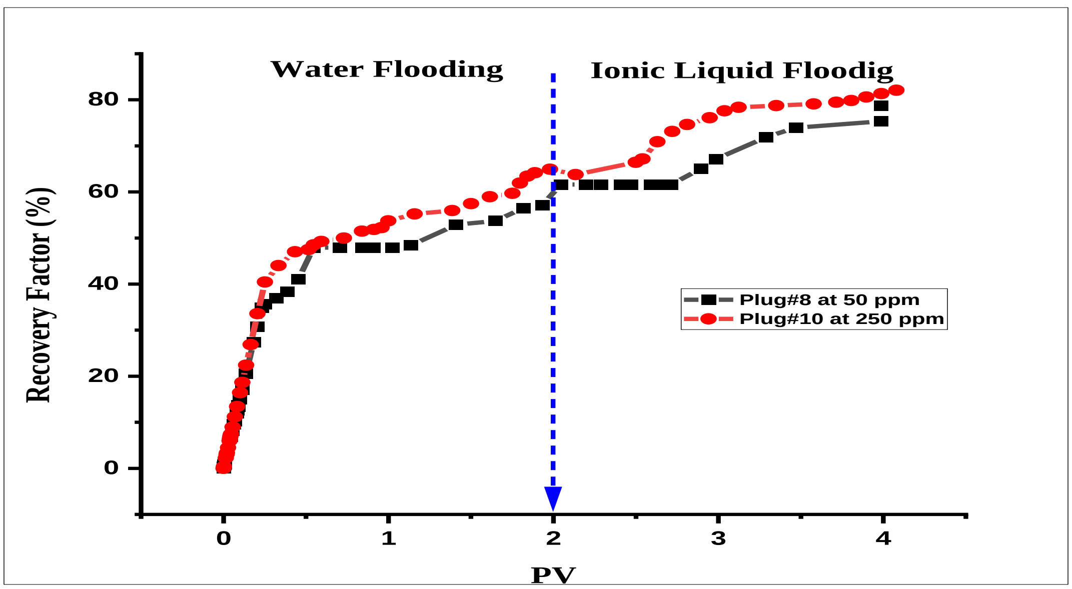

Figure 9. Oil recovery from the core for solutions at a range of concentrations [10]

3.5.2 Flow Rate's Influence on Oil Recovery Efficiency

Diluted crude oil had a measured viscosity of 3.9 cp at an external temperature of 23 °C, utilising core samples with permeability values ranging from 4 to 20 md. The recovery factor for carbonate rocks, which are usually oil-wet and heterogeneous, increases somewhat with small changes in flow rate. Figure 10 and Table 4 show that the expected migration of particles kept oil recovery low even at the maximum flow rate of 1 cm^3/min.

Degradation of reservoir formations is frequently accompanied by fines migration. As a result of LSW flooding into a reservoir, high injection or production rates, natural reservoir particles migrate, or when EOR is mixed with any of these processes, natural gas or oil is produced with LSW. If the flow rate is too high, particles like kaolinite, illite, or non-clay fines might migrate when the flow rate exceeds the critical velocity. Because fines migration can drastically reduce the formation's permeability, it is an ongoing problem that needs fixing. Increasing injection rates to 1 cm^3/min enhances oil recovery at the best concentration, as seen in Fig. 10. For 0.667 cm^3/min and 1 cm^3/min flow rates, respectively, the oil recovery rates were around 19.7 % and 10.5%. Figure 10 shows that Q = 0.667 cm^3/min is the best flow rate for carbonate core plugs, leading to a greater oil recovery than 1 cm^3/min with the same volume of surfactant input.

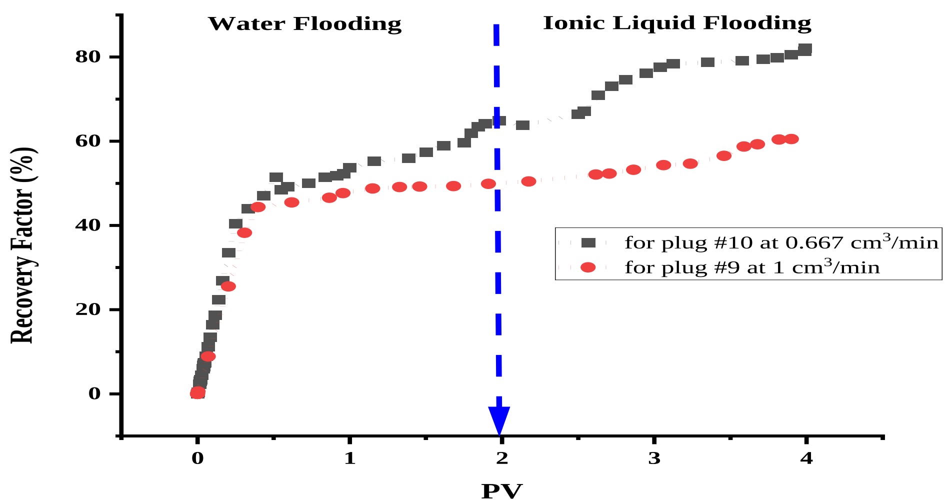

Figure 10. Oil recovery from the cores for the best concentration of ionic liquid at two available flow rates.

Table (4). Summary of Core Flooding Experiments.

Plug Number	φ (%)	K (MD)	S_{WI} (%)	Secondary recovery (%)	Ionic-liquid concentration (ppm)	Flow rate (cm^3/min)	PV Injection	Tertiary recovery (%)	Final recovery (Total) (%)
8	26.2	4.17	33	61	50	0.667	2	17	78
10	26.34	6.84	39	63	250	0.667		19.7	82.7
9	13.79	13.46	20	50.4	250	1		10.5	60.9

3.6 The previous Work

This study obtained a good value for the enhanced oil recovery using the ionic liquid solution prepared from combining (low salinity water with C12PYCL) if compared with previous research. Table (5) Illustrates the comparison between the previous work on ionic liquids flooding and the present research: -

Table (5): Compare the previous work on ionic liquid as territory recovery with present research.

	Author	Ionic Liquid / Concentration (ppm)	Brine	Secondary Extraction (% OOIP)	Flow rate (cm^3/min)	PVs injection	Additional Oil Recovery (%OOIP)
1	Hezave et al. (2013)[17]	$[C_{12}mim]Cl$	Formation Brine	50	0.3	/	13
		4000					
2	Manshad et al. (2017)[18]	$[C_{18}mim]Cl$	Formation Brine	38	0.5	2.7	13
		170					
3	Dahbag et al. (2015)[19]	Ammoeng 102	20 wt%,(83%NaCl, 17% CaCl2)	43	/		5
		500					
4	Ouda et al. (2023) [10]	C12 PYCL	Low Salinity	63	0.667	2	19.7
		50		61	0.667	2	17.3
5	**The present Study**	**C_{12} PYCL**	**Low Salinity**	**50.4**	**1**	**2**	**10.5**
		250					

4. Conclusions

The following inferences were made based on the study's experimental findings:

- The best concentration of ionic liquid (dodecyl pyridinium chloride) at 250 ppm was reduced by 71% and 60% in contact angle and interfacial tension, so it is a suitable surfactant in wettability alteration and EOR.

- At concentrations of 50 ppm and 250 ppm, respectively, the ionic liquid dodecyl pyridinium chloride enhanced oil recovery by 17.1% and 19.7%. This means it plays a major role in improving oil recovery efficiency.

- At the best concentration of ionic liquid (250ppm), additional oil recovery of (19.7%, 10.5%) from the OOIP at flow rates (0.667 ,1)cm^3/min after injection 2PVs.

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تعزيز استخلاص النفط بواسطة السائل الأيوني بمساعدة ماء منخفض الملوحة في مكمن الكربونات

الخلاصـــة : تحتوي الصخور الكربونية على حوالي 40% من نفط العالم . نظرًا لانخفاض معدلات استردادها من فيضانات المياه، تعد هذه الخزانات مرشحًا مثاليًا لطرق الاستخلاص المعزز للنفط (EOR). يمكن لتقنيات الاستخلاص المعزز للنفط الكيميائية(CEOR) ، مثل المواد الخافضة للتوتر السطحي أن تزيد من إنتاج هذه المكامن عن طريق تعديل قابلية البلل وتقليل التوتر السطحي (IFT).استكشفت هذه الدراسة السوائل الأيونية (ILs) ، وهو نوع جديد من المواد الخافضة للتوتر السطحي، وتأثيراتها المحتملة على قابلية البلل وتقليل.IFT . تضمنت الدراسة في البداية إجراء تقييم تجريبي للتوتر السطحي وقابلية التبلل باستخدام قطرة القلادة وزاوية التلامس. كشفت النتائج ان المادة الخافضة للتوتر السطحي تغير قابلية التبلل من 98° إلى 28° والتوتر السطحي (IFT) من 3.13 إلى 1.25 داين/سم عند 250 جزء في المليون. تم بعد ذلك تم اجراء اختبارات الغمر الأساسية لفحص تأثير تركيز السائل الأيوني على الاستخلاص النهائي للنفط من السدادة الأساسية لمكمن النفط الكربوني .اوضحت النتائج استرداد النفط الإضافي بنسبة 17% و19.7OOIP % عند 50 و 250 جزء في المليون على التوالي، بمعدل تدفق قدره 0.667 سم^3/دقيقة، على التوالي.حققت اختبارات الفيضانات الأساسية استردادًا إضافيًا بنسبة 10.5% OOIP بمعدل تدفق 1 سم^3/دقيقة باستخدام تركيز السائل الأيوني الاعلى .

الكلمات المفتاحية : زاوية التلامس، تغيير قابلية البلل، الاستخلاص الكيميائي المعزز للنفط (CEOR)، التوتر السطحي (IFT)، السائل الأيوني (IL).