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An Experimental Comparison between Ionic Liquid and Nanofluid Injection in the Nassiriyah Oil Field: Application for Enhanced Oil Recovery)

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Abstract

Many oil fields worldwide are reaching maturity and have stopped producing oil economically. It is necessary to extract oil from tertiary recovery technologies in addition to primary and secondary stages to meet the growing demand for power worldwide, increasing the price of oil. For this reason, enhanced oil recovery (EOR) will become more critical. A feasible technique to increase oil recovery is to inject chemicals into reservoirs.

The present research investigated the effect of ionic liquid (Dodecyl pyridinium chloride) alone and nanofluid formulations from (50 ppm of carbon nanoparticle CNPs with a range of concentrations of ionic liquid) on the interfacial tension and then application in enhanced oil recovery.

The result shows that the dodecyl pyridinium chloride alters interfacial tension (IFT) from 3.13 to 1.25 dynes/cm at 250 ppm. In contrast, the optimal nanofluid formulation (50 ppm of nanocarbon with 250 ppm of ionic liquid) reduced the surface tension from 55.07 to 39.85 dyne/cm at 25 oC.

Finally, Core-flooding tests were carried out; additional oil recoveries of 5.58% and 20.56% of the original oil in place were achieved after injection 2PV of ionic liquid only and optimum nanofluid formulation at the same flow rate, respectively. At the same time, additional oil recovery was 21.67 % after injection of 4 PV of optimum nanofluid. According to the research, these nanofluids are very promising for EOR.

Keywords: Nanofluid; Carbon nanoparticles CNPs; Enhanced oil recovery EOR; Ionic liquid (IL); Dodecyl pyridinium chloride.

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مقارنة تجريبية بين حقن السائل الأيوني والسوائل النانوية في حقل الناصرية النفطي : تطبيق لتحسين استخلاص النفط

الخلاصة:

وصلت العديد من حقول النفط حول العالم إلى مرحلة النضج وتوقفت عن إنتاج النفط اقتصادياً. فمن الضروري استخراج النفط الخام من تقنيات الاستخلاص الثلاثي بالإضافة إلى المرحلتين الأولية والثانوية لتلبية الطلب المتزايد على الطاقة في جميع أنحاء العالم، مما يؤدي إلى ارتفاع أسعار النفط. ولهذا السبب، سيصبح الاستخلاص المعزز للنفط أكثر أهمية في السنوات القادمة. ولزيادة استخلاص النفط، فإن الأسلوب الممكن هو حقن المواد الكيميائية في المكامن.

تناول البحث الحالي تأثير السائل الأيوني (كلوريد دوديسيل بيريدينيوم) وحده والموائع النانوية المحضرة من (50) جزء في المليون من جسيمات الكربون النانوية CNPs مع مجموعة من تراكيز السائل الأيوني (على التوتر السطحي ومن ثم تطبيقه في الاستخلاص المعزز للنفط.تظهر النتيجة أن كلوريد دوديسيل بيريدينيوم يغير التوتر السطحي (IFT) من 3.13 إلى 1.25 داين/سم عند 250 جزء في المليون. في المقابل، فإن تركيبة السوائل النانوية المثالية (50 جزء في المليون من الكربون النانوي مع 250 جزء في المليون من السائل الأيوني) خفضت التوتر السطحي من 55.07 إلى 39.85 داين/سم عند 25 درجة مئوية.

وأخيرا، تم إجراء اختبارات الفيضانات الأساسية. تم تحقيق استرداد إضافي للنفط بنسبة 5.58% و20.5% من الزيت الأصلي الموجود في مكانه بعد حقن PV2 من السائل الأيوني فقط وتركيبة السائل النانوي الأمثل بنفس معدل التدفق، على التوالي.

وفي الوقت نفسه، بلغ معدل الاستخلاص الإضافي للنفط 21.67% بعد حقن PV4 من السائل النانوي الأمثل. ووفقا للبحث، فإن هذه السوائل النانوية تعد واعدة للغاية بالنسبة للاستخلاص المعزز للنفط.

1. Introduction

Enhanced oil recovery (EOR) is used to produce more oil once primary and secondary oil recovery steps stop producing oil feasibly. High viscosities and capillary forces keep the oil within the reservoir's rock pores. Improving sweep efficiency or breaking oil-rock interactions is possible by injecting surfactant formulations. The goal of this tertiary EOR technique, which is water-oil interfacial tension (IFT) reduction or rock wettability change from oil-wet to water-wet, is to use surfactants or microemulsion flooding [1]. In recent decades, ionic liquids have attracted increasing attention as a potential tool for technological advancement. Because they melt below 100 °C, ionic liquids are organic salts, including organic and inorganic anions with organic cations. There are an estimated 1012–1018 synthetically accessible pairs possible when various cations and anions are combined.[2]. The wide range of solvent compositions enables the customization of solvent properties to meet the needs of various applications. Scientists are putting much effort into studying ionic liquids as a potential "greener" and "designer" alternative to traditional volatile solvents and catalysts in various physical and chemical processes.

Chemical-enhanced oil recovery (CEOR) incorporates ionic liquids as surface active agents to promote oil recovery by decreasing IFT, changing wettability, and so on [3]. Ionic liquids can alter the water-oil interface tension by lengthening the hydrophobic chain. Indeed, ILs adhere better to two distinct phases when the hydrophobic chain is more extended. This is because the distance between the hydrophilic and hydrophobic heads is extended, creating more stable emulsions. As a result, the trapped oil is made more accessible for movement [4]. Using ionic solutions for oil

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recovery. The use of ionic liquids has been demonstrated in previous laboratory studies to decrease IFT, even under extreme heat and salt [5], [6].

Emulsion stability [7, 8] and oil recovery [9,10, and 11] are just two of the numerous applications of nanoparticles (NPs) in the last decade.

A surfactant-NP Pickering emulsion, formed when NPs are mixed with surfactant, considerably increases the surfactant's thermal stability [12]. According to multiple findings, NPs dispersed in salt water with surfactant can reduce IFT in an oil-water system [13]. This research needs More information regarding the suggested EOR combination of nanoparticles and SAILs (Surfactant C12PYCL). Recent studies show that oil recovery can be enhanced by mixing surfactants with NPs [14,15]. These chemicals may synergistically enhance oil recovery by modifying wettability, decreasing IFT, and improving sweep efficiency in carbonate reservoirs.

In this study, the ionic liquid's interfacial tension and the nanofluid suspension's stability were systematically investigated to find the optimum concentration for EOR application in carbonate reservoirs. Then, it discusses the impact of a carbonate core oil recovery method involving Dodecyl pyridinium chloride alone and nanofluid injection. The tests involved flooding cores with low-salinity water as a secondary recovery strategy, flooding them with surfactant at room temperature, and comparing the recovery factors with nanofluids recovered at the same flow rate.

2. Experimental Work

2.1. Materials

The experimental work of this study was conducted in the labs of Baghdad University. Appendices to Tables (1) and (2) detail the substances, specifications, and analytical tools utilized in the current study.

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Table (1): The materials of this study

The materials	The source	The properties
Porous media	The carbonate reservoir/ Mishrif formation in Nassiriyah oil field	Depth (m) , (2012 – 2012.5) Length (cm) , (7.5 – 7.6) Diameter (cm), (3.8)
Carbon nanoparticles (CNPs)	Synthesis by [16] [17]	Average diameter, nm (40.16). Shape, irregular shape. The purity, (Atomic % =89.5, Weight %=82.8) Crystallite size (nm), 16. Insoluble in brine
Ionic liquid (1-dodecyl pyridinium chloride) (C12Py)(Cl)	Macklin company, China	purity > 98 wt.%
Aqueous phases (The base fluid)	low salinity water	T.D.S = 7898.62 ppm [8]
Crude oil	Nassiriyah oil field	Viscosity of only (1–1.5) cp, gas oil (75 Vol %) was added to the crude oil to increase the dilution ratio and create a more favorable displacement situation in wettability measurements and flow experiments.
n-hexane	Macklin Company, China.	purity > 95mol %

Table (2): Instruments of analytical tests

The device	The location
Interfacial tension	Petroleum Research and development Center/Baghdad
Surface tension test Zeta potential test	Ministry of science and technology/ Baghdad
DDM5210 Density meter ASTM D-445 Viscometer	Thi-qar oil company (T.O.C) / Nassriyah
Core flooding system	Petroleum engineering department / University of Baghdad / Baghdad.
Ultrasonic Homogeniser (200W, 26 kHz)	University of Technology / Baghdad

2.2. Formulation of solutions

2.2.1 Surfactant Formulation

A stock surfactant ionic liquid (C12py) (Cl) with a concentration of 2000 ppm was produced by dissolving 2 g in 1 liter of brine (low salinity) while agitating. The dilution law prepared a surfactant concentration range of 50 to 250 ppm from the surfactant stock.

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2.2.2 Nano fluid Formulation

The first stage involved adding carbon nanoparticles (CNPs) to the base fluid (low salinity) at a constant concentration of 50 ppm. Nanofluid (NF) was produced in the second step by dispersing carbon nanoparticles and a range of concentrations of ionic liquid using an ultrasonic homogenizer (Model.UP200St. Hielscher) for 30 min. Nanofluid experimental formulas are shown in Table (3).

Table (3): Experimental formula of nanofluids

Sample	The concentration of Ionic liquid (ppm)	The concentration of nanocarbon solution (ppm)
1	10	
2	50	
3	150	50
4	250	
5	350	
6	500	

2.3. Surface tension and Interfacial tension measurements

"Interfacial tension" describes the relationship between two liquids, while "surface tension" describes the relationship between air and another phase.All concentrations of ionic liquid were measured using an optical tensiometer known as the Theta Lite for the interfacial tension.

A drop of pure water is expelled from the tip of a needle and the cell's cross-section from its base. The light source was found to be a lens, and the interfacial tension was measured using a needle connected to the syringe piston by a steel line, as illustrated in Figure (1, A), which provides further detail on the preceding explanation.

A lot of the time, CNP's solutions are black. The fact that crude oil is black is well-known. Consequently, the IFT calculation would provide inaccurate results due to the difficulty distinguishing between crude oil and carbon nanoparticle droplets. Instead of measuring the interfacial tension (IFT), which would have been much more accessible, the surface tension (ST) of the carbon nanoparticles was measured [18]. During the experiment, the surface tension was evaluated using a Sigma 703D, a robust digital force tensiometer that accurately detects surface and interfacial tension for nanofluid formulations. It is simple, stands alone, and is resistant to damage. This device has a massive digital screen that displays measurements as they happen, as shown in Figure (1B).

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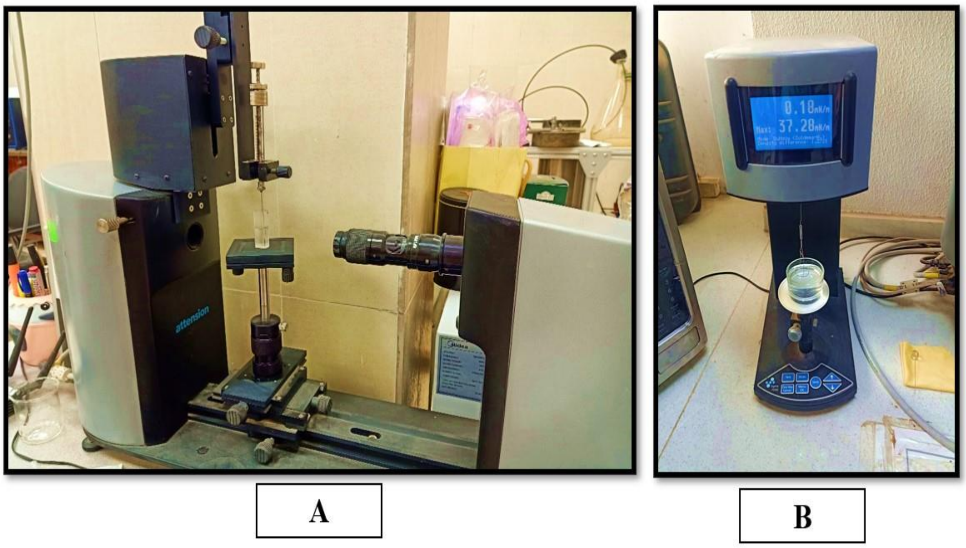

Fig. (1): A. quantifying the interfacial tension between a solution of ionic liquid at various concentrations and n-hexane, B. Sigma 703D device

2.4. Core flooding procedure

Exhibited in Figure (2) is the core-flooding procedure, which was subjected to testing and data accumulation. The core was initially saturated in a vacuum before being saturated with formation water in varying pore volumes [8]. Formation water was continuously injected at a concentration of 200,000 parts per million (ppm) until the injection pressure was steady. After that, diluted crude oil was pumped into the water-soaked deposit core until no more brine was produced. Following its completion and the recording of water and oil production data, secondary oil recovery procedures were established using the water flooding process (11). The efficacy of the injection procedure was determined by injecting separate solutions of nanofluid and ionic liquid at 250 ppm [8, 16] and [18,19].

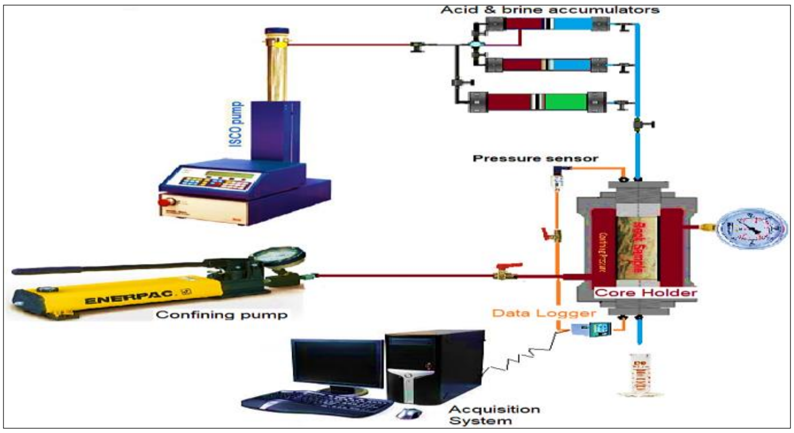

Fig. (2): Schematic of the core flooding apparatus [8][16-18][23]

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3. Results and Discussion

3.1. Stability of Nano fluid

The surfactant dodecyl pyridinium chloride [C12Py]Cl can change the electrostatic force due to surface charges on the particles. The oil development sector uses this surfactant, an essential component of several EOR agents, like chemical flooding slug. After adding 250 ppm of ionic liquid, the optimal concentration of nanofluids reached a stable state with a zeta potential of -33.88 mV. In the addition of ionic liquid, electrostatic attraction is expected to occur between the negatively charged carbon surface sites and the surfactant of ionic liquid cationic head groups.

Therefore, the mechanism by which the cationic surfactant molecules adsorb on the carbon nanoparticles surface involves electrostatic and Van der Waals interactions and depends on the surfactant charged. The results agree with Ding et al. [20] and Shi et al. [21]. The optimum concentration of nanofluid synthesized from (250 ppm of IL + 50 ppm of CNPs) is graphically shown in Figure (3).

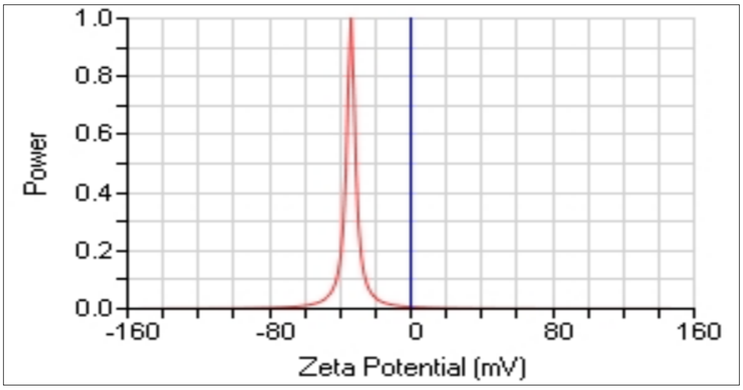

Fig. (3): Zeta potential for optimum Nano fluid

3.2. Effect of ionic liquid and nano fluid on the interfacial tension and surface tension

A range of ionic liquid solutions was used to measure the IFT in n-Hexane. In Figure (4), it is shown that adding ionic liquid up to a concentration of 250 ppm flattens down the surface tension graph's sharp slope. Since surfactant molecules have a twofold hydrophilic-hydrophobic configuration, they tend to disperse on the surface of liquids. This arrangement permits the molecule's hydrophilic half to dissolve in water while its hydrophobic half stays out. Based on the findings of this investigation, ILs may be a novel surfactant option for reducing the IFT between water and crude oil [8]. Surfactant (ionic liquid) interfacial tension mechanism is illustrated in Figure (5).

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In the meantime, the Sigma 703D gadget was used to determine the surface tension for each nanofluid formula, with air serving as the second phase. Studying the impact of CNPs on surface tension at different concentrations of an ionic liquid was the main goal of this experiment. The primary goal was to find the best nanofluid to use in the flooding experiment. The relationship between the surface tension values and nanofluid concentrations is shown in Figure (6). The surface tension value dropped precipitously for ionic liquid concentrations of 10, 50, and 150 ppm. Figure (6) shows that compared to low concentrations of ionic liquid, it gradually declined at 250, 350, and 500 ppm.

Recently developed nanofluids have a lower surface tension because they include a higher concentration of [C12Py] Cl. According to these results, the following nanofluid composition is optimum: the concentration of nanocarbon at 50 ppm and ionic liquid at 250 ppm [16].

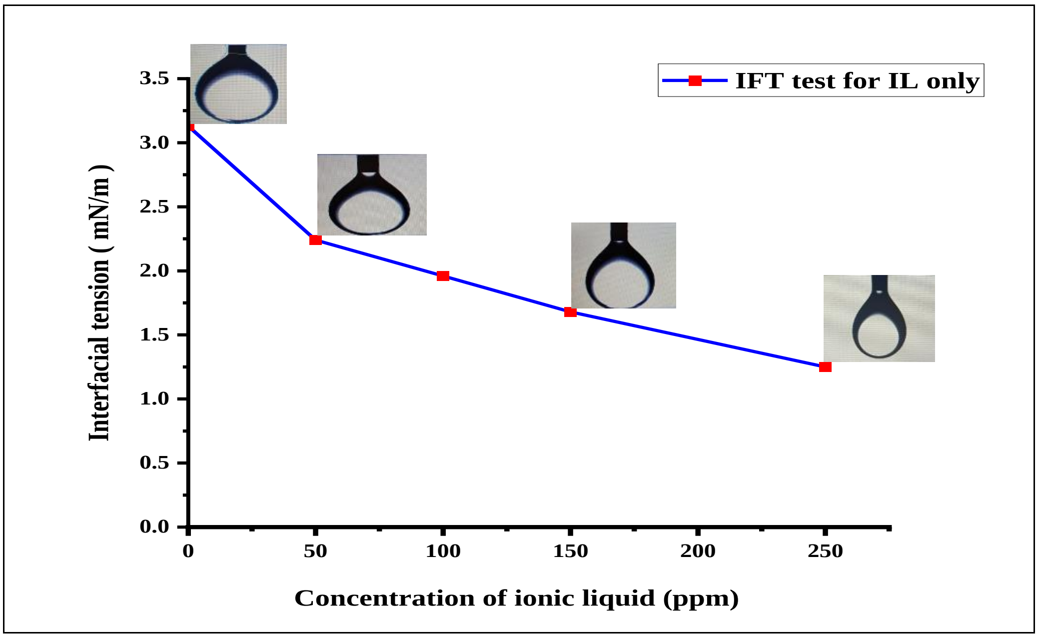

Fig. (4): Variation of interfacial tension with different concentrations

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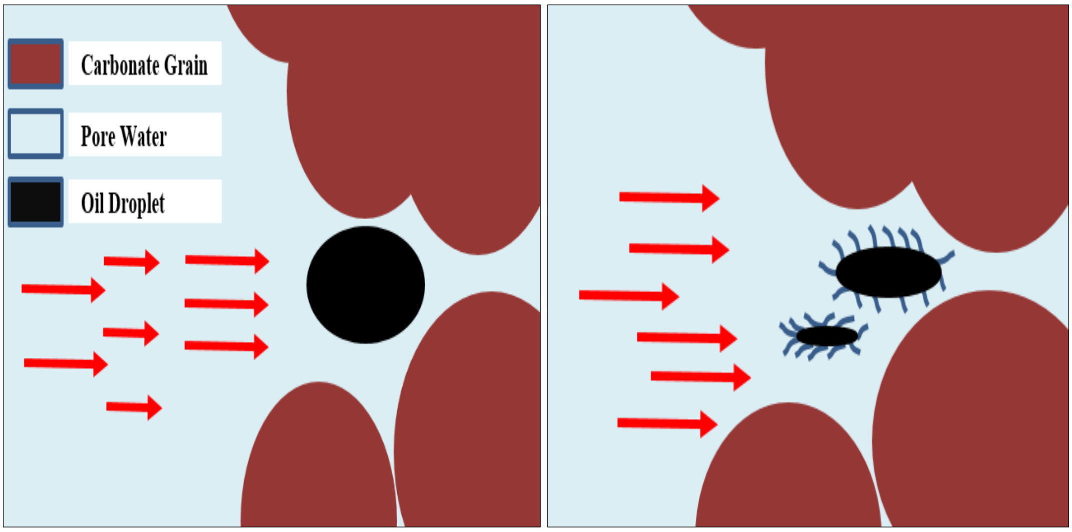

Fig. (5): A. Without Surfactant (High interfacial tension), B. With Surfactant (Low interfacial tension)

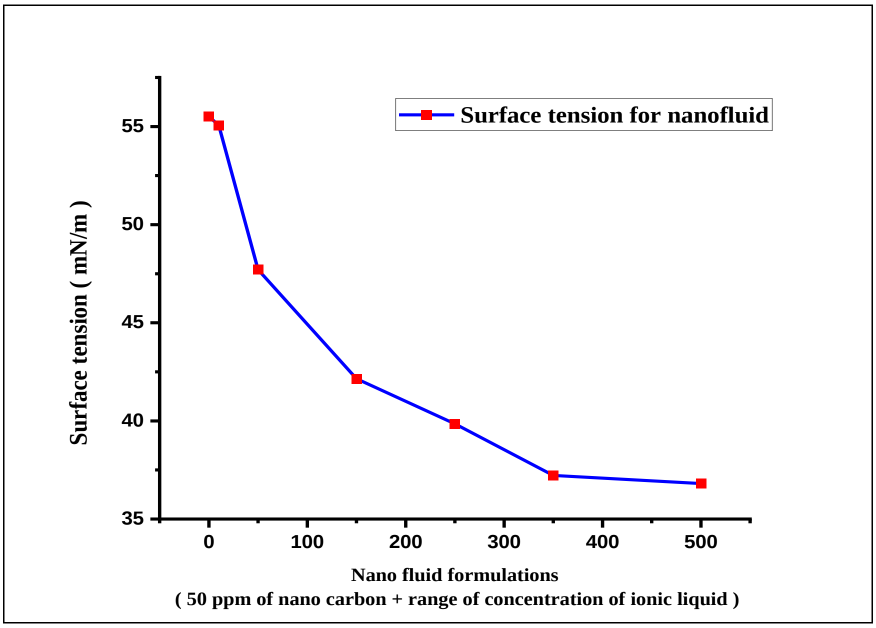

Fig. (6): The surface tension (ST) of the nanofluid formulations

3.3. Oil Recovery

The optimal concentrations for nanofluid formulation and ionic liquid alone were tested in core-flooding experiments conducted at room temperature in carbonate cores (Table 4 displays the specifics). The flooding operation included [22,23]:

• The subsequent recovery involves injecting 2 pore volumes (PVs) from water with a low salinity as secondary recovery.

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- Two different injections were performed: one using 250 ppm of ionic liquid in plug #3 and the other optimum nanofluid in plug #11.

After secondary flooding, plugs #3 and #11 (refer to Table 4) were injected with the territory recovery. Figure 7 shows that after injecting 2PV of ionic liquid and (2PV,4PV) of optimum nanofluid at the same flow rate, there was an additional oil recovery (AOR) of 5.57% OOIP and (20.56 %, 21.67%) OOIP for ionic liquid and optimum nanofluid formulation, respectively. The results from the present study and the previous study in Table 5 prove that increasing the flow rate leads to an increase in recovery factor in the case of Ionic liquid injection. The higher flow rate leads to a lower recovery factor when injecting nanofluid [16].

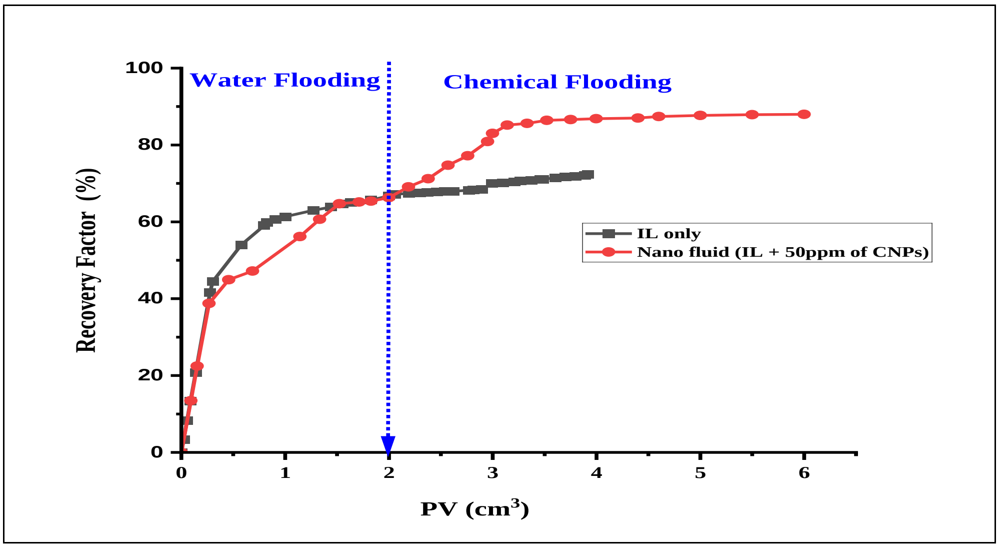

Fig. (7): The oil recovery after injection 2PV of ionic liquid and 4PV of nanofluid

4. Conclusions

Researching innovative and cost-effective methods to optimize oil recovery from existing reservoirs is of the utmost importance due to the rising global need for energy. One practical way to meet the increasing global demand for energy while simultaneously preserving oil supplies is to combine low-salinity water floods with nanofluid hybridization.

Our results show that low-salinity water, ionic liquid [C12Py]Cl, and CNPs greatly enhance oil recovery. After injecting 2PV of ionic liquid at 250 ppm and 2PV of nanofluid synthesis (50 ppm of CNPs + 250 IL), respectively, the oil recovery increased from 66.75% to 72.32% at the same flow rate, leading to an additional 5.57% of oil production; and from 66.29% to 86.85%, leading to an additional 20.56 % of oil production. For the same injection volume and flow rate, nanofluid is more effective than ionic liquid at improving oil recovery.

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Table (4): Carbonate core plug properties

	plug # 11 Nano	plug #3 IL
Depth, m	2012.2	2012.45
The porosity of the core, %	15	20.96
Absolute permeability, mD	6.115	18.24
Length, cm	7.6	7.5
Diameter, cm	3.8	3.8
Pore volume, cc	13.13	17.93
Bulk volume, cc	86.68	85.54
OOIP. Original Oil In Place, cc	8.9	11.6
Initial oil saturation, %	68	65
Initial water saturation, %	32	35
The injection volume, pv	2 PV , 4 PV	2 PV
Flow rate, cc/min	0.334	0.333
Additional Oil Recovered after chemical flooding, %	20.56 (at 2 PV) 21.67(at 4 PV)	5.57

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Table (5): Selected publications of Carbon nanoparticles (CNPs) application in EOR studies

	Type of NPs	The surfactant dispersion	The concentration of nanoparticles (ppm)	Flow rate Q (cm^3/min)	Territory Recovery	Pore volume injection	The Reference
1	carbon nanotubes (CNTs)	Sodium dodecyl sulfate (SDS)	3000	1	18.57%	2	**Hassan Soleimani et all (2018) [18]**
2	silica–graphene quantum dots (Si–GQDs)	/	1000	0.5	14.4 %	2	**Mehran Mirzavandi et all (2023) [24]**
3	graphene oxide (GO)	disodium cocoamphodiacetate (CAD)	20	/	11.80%	/	**Kaili Liao et all (2022) [25]**
4	modified carbon black nanoparticles	/	2500	0.2	27.27%	5.7	**Guang Zhao et all (2022) [2]**
5	carbon nanoparticle	ionic liquid	50	0.334	20.50%	2	**Uoda et all (2023) [16]**
				0.667	8.46%		
6	0	250 ppm ionic liquid	0	0.667	28.8%	7.75 Continuous injection	**Uoda et all (2023) [26]**
7	0	250 ppm ionic liquid	0	0.667	19.7 %	2	**Uoda et all (2023) [8]**
8	0	250 ppm ionic liquid	0	0.334	5.57 %	2	**The present study**
	carbon nanoparticle	ionic liquid	50	0.334	21.67 %	6	

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