Comprehensive investigation of low salinity waterflooding in carbonate reservoirs

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Abstract

Waterflooding has been practiced as a secondary recovery mechanism for many years with no regard to the composition of the injected brine. However, in the last decade, there has been an interest to understand the impact of the injected water composition and the low salinity waterflooding (LSWF) in oil recovery. LSWF has been investigated through various laboratory tests as a promising method for improving oil recovery in carbonate reservoirs. These experiments showed diverse mechanisms and results. In this study, a comprehensive review and analysis for results of more than 300 carbonate core flood experiments from published work were performed to investigate the effects of several parameters (injected water, oil, and rock properties along with the temperature) on oil recovery from carbonate rock. The analysis of the results showed that the water composition is the key parameter for successful waterflooding (WF) projects in the carbonate rocks. However, the salinity value of the injected water seems to have a negligible effect on oil recovery in both secondary and tertiary recovery stages. The study indicated that waterflooding with optimum water composition can improve oil recovery up to 30% of the original oil in place. In addition, the investigation showed that changing water salinity from LSWF to high salinity waterflooding can lead to an incremental oil recovery of up to 18% in the tertiary recovery stage. It was evident that applying the optimum composition in the secondary recovery stage is more effective than applying it in the tertiary recovery stage. Furthermore, the key parameters of the injected water and rock properties in secondary and tertiary recovery stages were studied using Fractional Factorial Design. The results revealed that the concentrations of Mg^{2+} , Na^+ , K^+ , and Cl^- in the injected water are the greatest influence parameters in the secondary recovery stage. However, the most dominant parameters in the tertiary recovery stage are the rock minerals and the concentration of K^+ , HCO_3^- , and SO_4^{2-} in the injected water. In addition, it appears that the anhydrite percentage in the carbonate reservoirs may be an effective parameter in the tertiary WF. Also, there are no clear relations between the incremental oil recovery and the oil properties (total acid number or total base number) in both secondary and tertiary recovery stages. In addition, the results of the analysis showed an incremental oil recovery in all ranges of the studied flooding temperatures. The findings of this study can help to establish guidelines for screening and designing optimum salinity and composition for WF projects in carbonate reservoirs.

Keywords Low salinity waterflooding · Carbonate reservoirs · Smart waterflooding · Enhanced oil recovery

Abbreviations

EOR	Enhanced oil recovery	IFT	Interfacial tension
FFD	Fractional factorial design	Incr.	Incremental
HS	High salinity water	LS	Low salinity water
HSWF	High salinity waterflooding	LSWF	Low salinity waterflooding
		MIE	Multi-component ion exchange
		NA	Not available
		OOIP	Original oil in place
		Sec.	Secondary recovery stage
		TAN	Total acid number
		TBN	Total base number
		TDS	Total dissolved salts
		Ter.	Tertiary recovery stage
		WF	Waterflooding

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Symbols

k	Permeability, md
K_a	Air permeability, md
PV	Pore volume, CC
T	Temperature, °C
α	Significance level, fraction
Φ	Porosity, fraction

Introduction

LSWF is defined as an enhanced oil recovery method that uses water with a low concentration of dissolved salts and/or modifying the ionic content of injected brines as a flooding medium (Bartels et al. 2019). The LSWF technique is also known in the literature as low salinity trademark, smart waterflood, designer waterflood, advanced ion management, modified salinity flooding, ion tuning, and engineered water injection.

The LSWF applications revealed several successful results in laboratory experiments for core plugs extracted from carbonate reservoirs. However, the effect of the LSWF in the carbonate reservoirs has not been well studied compared to the sandstone reservoirs (Al Shalabi and Sepehrnoori 2017). It should be highlighted that Bagci et al. (2001) conducted the first core flooding experiments on carbonate reservoirs. They reported 18.8% higher oil recovery by injecting 2% KCl plus 2% NaCl brine mixture rather than injecting other brine compositions. The LSWF showed incremental oil recovery up to 20% higher than the HSWF (Strand et al. 2008; Ligthelm et al. 2009; Yousef et al. 2011). In other studies, Shariatpanahi et al. (2011), Fathi et al. (2010), and Zekri et al. (2019) documented additional oil recovery up to 11% by injecting sea water in carbonate core plugs. They reported that sea water with high sulfate concentrations could increase oil recovery from the carbonate reservoirs.

The most acceptable mechanism of the LSWF in the carbonate reservoirs is the wettability change (Yousef et al. 2011; Al-adasani et al. (2012)). However, the explanation of the physical mechanism of changing the wettability is still debated. The proposed mechanisms include fine migration, multi-component ion exchange (MIE), calcite dissolution, interfacial tension (IFT) reduction, double-layer expansion, anhydrite dissolution, salting-in, water micro-dispersion, osmosis pressure effect, and surface roughness.

The fines migration mechanism was initially proposed by Bernard (1967) for sandstone reservoirs. Then, it was accepted by Tang and Morrow (1997). This mechanism was also suggested for the carbonate reservoirs by Yi and Sarma (2012); and Zahid et al. (2012). However, this suggestion was not accepted by Lager et al. (2008), Shaker and Skauge

(2013), Amirian et al. (2017), and others who noticed an increase in the oil recovery without fines migration.

The MIE evidence of the LSWF in the carbonate reservoirs was proposed by Austad et al. (2005). Then, this mechanism was accepted by Strand et al. (2008), Ligthelm et al. (2009), Zhang and Austad (2006), Puntervold (2008), and Myint and Firoozabadi (2015). They suggested that the MIE in the carbonate reservoirs is anion exchange (It is not a cation exchange like sandstone reservoirs).

The calcite dissolution mechanism of the LSWF in the carbonate reservoirs was proposed by Hiorth et al. (2008). Then, in other studies, this mechanism was confirmed by Evje and Hiorth (2010) and Den Ouden et al. (2015). However, Austad et al. (2010), Mahani et al. (2015), and Romanuka et al. (2012) did not accept this mechanism. They documented that no incremental oil recovery is achieved when only sulfate exists with no calcium, while the solubility should increase in the absence of calcium.

The IFT reduction mechanism of the LSWF in the carbonate reservoirs was suggested by McGuire et al. (2005), Vijapurapu and Rao (2003), Okasha and Alshiwaish (2009), and Mohsenzadeh et al. (2016). However, Yousef et al. (2011), Al Harrasi et al. (2012), and Al-Attar et al. (2013) reported that the application of the LSWF in the carbonate reservoirs has an insignificant effect on the IFT.

The water micro-dispersion mechanism of the LSWF in the carbonate reservoirs was proposed by Emadi and Sohrabi (2012). Then, it was documented by Mahzari and Sohrabi (2014), Sohrabi et al. (2017), Alvarado et al. (2014), Chávez-Miyauchi et al. (2016), Sarvestani et al. (2019), and Dordzie and Dejam (2021).

The double-layer expansion mechanism of the LSWF in the carbonate reservoirs was suggested by Ligthelm et al. (2009) and accepted by Shariatpanahi et al. (2011), Al-adasani et al. (2012), Fathi and Austad (2012), Al-Shalabi et al. (2014), Alameri et al. (2014), Mahani et al. (2015), Sohal et al. (2016), and Liu et al. (2016).

The anhydrite dissolution mechanism of the LSWF in the carbonate reservoirs was proposed by Austad et al. (2011) and Pu et al. (2008) and was agreed by Austad et al. (2015). They believed that it is a method of generating in situ SO_4^{2-} which is thought to act as a catalyst in the wettability alteration process. This mechanism was not accepted by Jiang et al. (2014) and Uetani et al. (2019). Furthermore, Romanuka et al. (2012) and Nasralla et al. (2014) reported that anhydrite dissolution is not considered the primary mechanism for LSWF.

The salting-in mechanism was proposed by Austad et al. (2007) as a result of rock dissolution. The migration of divalent ions from the rock surface toward the brine causes a change in rock wettability.

Osmosis mechanism was suggested by Sandengen and Arntzen (2013) and was agreed by Sandengen et al. (2016),

Fredriksen et al. (2016), and Fredriksen et al. (2018). However, Bartels et al. showed that the osmosis pressure could contribute to the incremental oil recovery, but it is not the primary mechanism of LSWF (2017).

The surface roughness mechanism of the LSWF in the carbonate reservoirs was investigated by Sari (2020). It was proposed that the contact angle consistently decreased (becomes less oil-wet) with increasing the surface roughness. However, they reported that the effect of surface roughness on contact angle reduces with decreasing brine salinity.

Derkani et al. (2018) supposed that, in general, it is believed that there should be more than one factor in effect for LSWF in carbonate reservoirs. Al-Shalabi et al. (2015) reported that the incremental oil recovery by LSWF in carbonates could best be explained as wettability alteration caused by dissolution, changes in pH, and double-layer expansion.

The above introduction shows that the applications of the LSWF can increase the oil recovery from carbonate reservoirs. However, there is no single opinion on the LSWF mechanisms. In general, there is more than one effective factor and mechanism for the LSWF in the carbonate reservoirs. In addition, there is no agreement on which mechanisms are important and can act under which conditions. Moreover, till now, it has been difficult to accurately predict the effects of the LSWF in the carbonate reservoirs because of the complexity of the chemical interactions between the carbonate rocks and the reservoir fluids, as well as the heterogeneity of the rock. This work is a trial to analyze the results of several core flood experiments from the literature to obtain guidelines for improving oil recovery in typical waterfloods by changing/optimizing water salinity and composition.

Most of the previous work depended on a limited number of core experiments. However, this work included data analysis for results of more than 300 core experiments from the literature. Therefore, the results and the analysis of the current work are reliable. The effects of several parameters (injected water, oil, and rock properties along with the temperature) on oil recovery were studied and investigated. Furthermore, statistical analysis using a two-level folded Plackett–Burman design was performed to identify the most significant parameters relative to incremental oil recovery for carbonate reservoirs in secondary and tertiary recovery stages. However, the dataset range is limited to extract a solid conclusion for the effect of rock minerals on the oil recovery during the secondary recovery stage.

Figure 1 shows a block diagram for the methodology of this work. Initially, the available data and results of published experimental works on carbonate cores were collected and reviewed. Secondly, the database was built, including all rock and fluid properties along with the flooding results. Then, the data and results were analyzed; and relationships between incremental oil recovery, rock, and fluid properties

Fig. 1 Block diagram of the methodology

were established. Finally, statistical analysis was performed to identify the most effective oil recovery parameters in both secondary and tertiary recovery stages.

Database of previous flooding experiments

A database with results of more than 300 core flood experiments of carbonate reservoirs was collected and analyzed from the literature. The analyzed data includes (1) about 200 tertiary low salinity waterflooding experimental runs that have been carried out by more than 100 core plugs, and (2) around 100 secondary waterflooding experimental runs that have been conducted by 50 core plugs approximately. The summary of the collected data, including the main rock characteristics and the core flooding experiments' results, is presented in Table 1.

The histograms of the rock properties (porosity and permeability) and the incremental oil recovery during the secondary and tertiary recovery stages are shown in Fig. 2a–d.

Figure 2a shows that most of the core plugs in the collected dataset cover range of porosity starting from 12 up to 35%. More than 75% of the core plugs have a porosity range of 15–25%. However, Fig. 2b illustrates that the core plugs' maximum permeability in the collected dataset was 470 md, while the minimum permeability was 0.2 md. It is clear that more than 80% of the core plugs in the collected data have a permeability of less than 120 md. It should be highlighted that less than 50% of the core plugs in the collected data have a description of the rock minerals.

Figure 2c demonstrates that the incremental oil recovery achieved from applying the LSWF in the carbonate rock during the secondary recovery stage ranged from –30 to 30%. The negative incremental oil recovery values mean that the application of the LSWF attained less oil recovery than that achieved in the HSWF experiments. The collected data indicate that the maximum additional oil recovery by applying the LSWF during the tertiary recovery stage was about 24% of the OOIP in most of the experiments. Most of the core plugs showed incremental oil recovery in the tertiary recovery stage, less than 18%, as shown in Fig. 2d.

Table 1 Summary of core flood experiments results collected from the literature

References	Type	No. of plugs	PV, CC	Φ , %	K_a , md	Rock mineral description	Incr. oil recovery, % OOIP
AlQuraishi et al. (2015)	Ter	1	9	18	206	Available	(16)
	Sec	1	9	18	206	Available	(10)
Al Hamad et al. (2017)	Ter	6	10–13	17–19	36–127	NA	(0)–(17)
Tetteh et al. (2017)	Ter	10 (2 Composite)	46–66	25	60–102	NA	(0)–(10)
Yousef et al. (2010)	Ter	2	14	17	23–80	NA	(5)–(11)
Sari et al. (2017)	Ter	2	9–13	18–23	3–6	Available	(6)–(18)
Austad et al. (2012)	Ter	6	14–33	16–42	0.2–4	Available	(0)–(5)
Zahid et al. (2012)	Ter	6	9–24	19–33	3–308	NA	(0)–(12)
Uetani et al. (2019)	Ter	6	14–21	17–24	2–60	Available	(0)–(7)
Hamouda and Maevskiy (2014)	Sec	5	41	49	4	NA	(–3)–(2)
	Ter	1	39	48	4	NA	(0)
Ali et al. (2012)	Ter	5 (1 Composite)	88	21	148	Available	(1.5)–(7)
Romanuka et al. (2012)	Ter	25	NA	12–51	2–235	Available	(0)–(21)
Gandomkar and Rahimpour. (2015)	Ter	12	15–19	17–22	2–7	Available	(6)–(35)
	Sec	12 Twins	15–19	17–22	2–7	Available	(0)
Aksulu et al. (2012)	Ter	2	17	31	53–78	NA	(0)–(45)
Almeida et al. (2019)	Sec	4 Twins	8	15	126–160	NA	(3)–(10)
	Ter	2	8	15	125–141	NA	(7)–(17)
Camargo et al. (2019)	Ter	1	13	21	61	NA	(17)
Masalmeh et al. (2019)	Sec	3	115–146	19–25	1.6–30	NA	(0)–(12.5)
	Ter	3	115–146	19–25	1.6–30	NA	(0)–(6)
Bazhanova and Pourafshary (2020)	Sec	2 Twin	15	17	19–47	NA	(5)
Abdeli and Seiden (2018)	Sec	8 Twins	14–20	19–22	0.4–1.7	NA	(–14)–(27)
	Ter	6	14–20	19–22	0.4–1.7	NA	(4)–(12)
Ghandi et al. (2019)	Sec	3	4.6	18.5	25.3	Available	(9)–(13)
Zekri et al. (2019)	Ter	9	10–16	17–24	0.2–1.7	NA	(1)–(7)
Hosseini et al. (2020)	Sec	2	13–18	13–17	0.7–1.7	Available	(–2)–(20)
	Ter	2	13–18	13–17	0.7–1.7	Available	(1)–(10)
Abdelhamid and Elnaggar. (2017)	Sec	8	NA	19–27	35–471	NA	(–7)–(6)
Al Harrasi et al. (2012)	Sec	1	13	25	5	Available	12
	Ter	5	11–14	22–26	5	Available	0–13

Table 2 presents the devices and measurement methods used in the collected database's experimental work. It is clear that mainly two methods were used to perform the flooding experiments in the secondary and tertiary recovery stages, namely core flooding and spontaneous imbibition methods. The core flooding was applied using a mainly liquid pump, piston cell, core holder, oven, glass beret, and computer-controlled system. However, the spontaneous imbibition method was applied by displacing the fluid with a sealed core holder's pump. In this method, the displaced fluid volume could be measured directly or determined by weight balance measurements. In one of the previous studies using carbonate core plugs, Almeida et al. (2019) studied the effect of the LSWF on oil recovery using a high-speed centrifuge method.

As shown in Table 2, various methods were used in the collected database's experimental work to study the wettability alteration behavior due to the change of the injected water salinity. The wettability indication methods were mainly the measurements of the contact angle, Zeta potential, pH, and interfacial tension. In addition, in some of the experiments, the wettability index was measured using the Amott Harvest method.

Discussion

Salinity range for the LSWF applications

There are different opinions in the literature regarding the optimum water salinity range that can improve oil recovery

Fig. 2 Histograms of porosity, permeability, and incremental oil recovery of the literature data

in carbonate reservoirs. Figure 3 presents the relationship between the injected water salinity and incremental oil recovery during the secondary and tertiary recovery stages.

Figure 3a shows the effect of the water salinity on oil recovery during the secondary recovery stage. When water with a salinity of less than 5000 ppm was used, some flooding experiments showed high incremental oil recovery up to 35%, and others showed lower incremental oil recovery. In addition, when the water of salinity of more than 5000 ppm was used, an incremental oil recovery up to 15% only was documented and observed in several experiments.

It should be highlighted that Gandomkar and Rahimpour (2015) and Abdeli and Seiden (2018) reported an increase of oil recovery up to 20% during the secondary WF recovery stage with different water compositions and the same total dissolved salinity (TDS). In addition, Alotaibi and Nasr-El-Din (2019) suggested that there is a critical salinity (not the lowest), at which the IFT between oil and brine is at a minimum level. Snosy et al. (2020) and Snosy et al. (2021) reported the same results from the sandstone reservoir.

Furthermore, the application of the LSWF in the tertiary recovery stage showed various results, as shown in Fig. 3b. Additional oil recovery was achieved in the tertiary recovery stage using water of salinity of more than 20,000 ppm. Moreover, no additional oil recovery was observed in other experiments even when WF of water salinity less than 1000 ppm was applied. Aksulu et al. (2012) reported that the incremental oil recovery with the application of the LSWF in the carbonate rock ranges from 0 to more than 30% using water of a salinity less than 500 ppm.

Moreover, Romanuka et al. (2012), Aksulu et al. (2012), and Zekri et al. (2019) reported additional oil recovery in the tertiary recovery stages reached 18% with applying HSWF after LSWF.

It should be highlighted that application of the WF with optimum composition achieved higher oil recovery in the secondary recovery stage up to 10% than that obtained during the tertiary recovery stage, as presented by Masalmeh et al. (2019); and Hosseini et al. (2020). However, Al Harrasi et al. (2012) reported that the WF with optimum composition

Fig. 2 (continued)

c Histogram of secondary incremental oil recovery

d Histogram of tertiary incremental oil recovery

obtains the same oil recovery in the secondary and tertiary recovery stages, but the oil rate in the secondary recovery stage is faster than in the tertiary recovery stage (accelerated recovery has occurred in the secondary recovery stage).

Effect of water composition

The significance of SO_4^{2-} , Ca^{2+} , Na^+ and Mg^{2+} concentration during the application of the LSWF in the carbonate reservoirs was suggested by Austad et al. (2005), Zhang et al. (2006), Strand et al. (2008), Romanuka et al. (2012),

Bazhanova and Pourafshary (2020), and others. They suggested that the MIE is the mechanism of the wettability alteration in the carbonate reservoirs. The MIE mechanism is attributed to the adsorption of potential anions/cations such as SO_4^{2-} , Ca^{2+} and/or Mg^{2+} onto the rock surface. Adsorption of the SO_4^{2-} anions decreases the positive charge density on the rock surface. This behavior minimizes the electrostatic repulsive force and causes the Ca^{2+} and Mg^{2+} cations' co-adsorption on the rock surface. The reaction of the Ca^{2+} cations with the carboxylic acid groups results in breaking the attractive interactions between the oil and the

Table 2 Flooding devices and measurement methods of the previous experimental studies

References	Test type			Wettability indication methods				
	Core flooding	Spontaneous imbibition	Centrifuge	pH	IFT	Contact angle	Zeta potential	Amott index
AlQuraishi et al. (2015)	✓				✓	✓		
Al Hamad et al. (2017)	✓	✓			✓	✓		
Tetteh et al. (2017)	✓				✓			✓
Yousef et al. (2010)	✓				✓	✓		
Sari et al. (2017)	✓				✓	✓	✓	
Austad et al. (2012)	✓	✓						
Zahid et al. (2012)	✓							
Uetani et al. (2019)		✓						
Hamouda and Maevskiy (2014)	✓			✓				
Ali et al. (2012)	✓				✓	✓	✓	
Romanuka et al. (2012)		✓						
Gandomkar and Rahimpour. (2015)	✓			✓				
Aksulu et al. (2012)		✓						
Almeida et al. (2019)			✓					
Camargo et al. (2019)	✓							
Masalmeh et al. (2019)	✓							
Bazhanova and Pourafshary (2020)		✓		✓		✓		
Abdeli and Seiden (2018)	✓					✓		
Ghandi et al. (2019)		✓			✓	✓		
Zekri et al. (2019)	✓			✓				
Hosseini et al. (2020)	✓			✓	✓	✓		
Abdelhamid and Elnaggar. (2017)	✓							
Al Harrasi et al. (2012)	✓	✓						

rock interface, which changes the rock surface behavior to be more water wet. Gopani et al. (2021) documented that the effect of the Mg^{2+} cations on the oil recovery during the application of the LSWF in the carbonate rock is less than that of the Ca^{2+} cations due to the low solubility of $MgSO_4$ in the brine at ambient conditions. In another study, Olayiwola and Dejam (2021) demonstrated that the concentrations of Ca^{2+} , SO_4^{2-} , Mg^{2+} ions react with the carboxylic component oil and cause a great effect on the wettability of carbonate reservoirs.

Yousef et al. (2011), Shariatpanahi et al. (2011a), and others hypothesized that injection of sea water containing a high amount of SO_4^{2-} anions could alter the wettability to be more water wet and increase oil recovery. The incremental oil recovery was documented in the tertiary recovery stage due to high sulfate concentration on chalks cores and limestone cores (Zhang and Austad 2005; Hognesen et al. 2005; Al-Attar et al. 2013; and Zekri et al. 2019). They showed that reducing the sea water's salinity slightly affected the carbonate rock wettability and could not increase oil recovery. They documented that increasing the sulfate concentration beyond some optimum concentration of 47 ppm can cause a negative effect on the flooding process. In another

study, Zhang et al. (2006) documented that increasing the concentration of SO_4^{2-} anions and decreasing NaCl concentration in the injected water cause a change in the wettability and improve the oil recovery from the carbonate reservoirs.

On the other hand, Abdeli and Seiden (2018) observed that the presence of the Mg^{2+} cations in the injecting brine would affect the ultimate oil recovery much more than the Ca^{2+} cations. They showed that the oil recovery factor was not affected significantly by removing the Ca^{2+} cation from the injection solution. In addition, Zekri et al. (2019) reported an insignificant effect of the sulfate in the injected water. They concluded that adding sulfate into the water would increase the process's overall cost without improving oil recovery effectiveness.

Furthermore, Al Hamad et al. (2017) studied the iodide effects in the water injection within the carbonate rocks. They clarified that sodium iodide has a tendency to alter the rock wettability to be stronger water wet conditions. In addition, they documented an increase of 3–12% incremental oil recovery using dolomitic core plugs by increasing NaI from 0 to 1000 ppm in the injected water and with no change in the other minerals. Moreover, they reported that using limestone core plugs gave an increase of 0–17% incremental oil

Fig. 3 Incremental oil recovery versus the injected water salinity in the secondary and tertiary recovery stages

recovery in the tertiary recovery stage by changing the sea water (the sea water was diluted to 1000 ppm saline water, including 1000 ppm of NaI minerals only). In another study, Hosseini et al. (2020) found that the H_2PO_4^- anions can act as a neutralizing agent to reduce the surface charge in the carbonated reservoirs.

Figure 4 shows the relation between the SO_4^{2-} , Ca^{2+} , Na^+ , Mg^{2+} , and K^+ contents of the injected low saline water versus the incremental oil recovery in the secondary recovery stage using core plugs of carbonate rocks. There is no clear relationship between incremental oil recovery and water composition. The highest and lowest oil recovery values were documented with each mineral's presence and absence in the injected water. In addition, the LSWF showed oil recovery up to 25% with no sulfates in the injected water.

In the collected data experiments, when the effects of the injected water minerals were studied during the secondary recovery stage, two experiments were usually carried out using either (1) twin core plugs or (2) the same core plug after cleaning and repeating all the experimental procedure. The two experiments were usually carried out using two different salinities and compositions (HS and LS). Figure 5 shows the impacts of the difference in the concentration of the injected water minerals on the oil recovery during the secondary recovery stage. The figure's negative values

mean that the concentration of the LS water components is lower than that in the HS water. However, the positive values mean that the LS water components' concentration is higher than that in the HS water. Figure 5a shows that a decrease in the $(\text{Na}^+ + \text{K}^+)$ cations concentration leads to an incremental oil recovery in most of the core plugs. However, an increase and decrease in the incremental oil recovery were reported in the experiments, which were conducted without changing the concentration of $(\text{Na}^+ + \text{K}^+)$ cations. The same observation was documented during the decrease in the $(\text{SO}_4^{2-} + \text{Ca}^{2+} + \text{Mg}^{2+})$ concentrations, as shown in Fig. 5b. Either a decrease or increase in the concentration of these components in the brine water showed an incremental oil recovery.

Figure 5c, d show that decreasing the concentration of the $(\text{Ca}^{2+} + \text{Mg}^{2+})$ cations may be important than reducing the concentration of the SO_4^{2-} anions in the injected water. Most of the core plugs showed additional incremental oil recovery with decreasing the concentration of $(\text{Ca}^{2+} + \text{Mg}^{2+})$ cations, while the change in the concentration of the sulfate did not reveal a clear effect with oil recovery.

Figure 5e showed that decreasing the divalent and monovalent concentration led to additional incremental oil

Fig. 4 Effects of the concentration of the injected water minerals on the oil recovery during the secondary recovery stage

Fig. 4 (continued)

recovery in most of the studies. However, the decrease of the divalent relative to the monovalent is better and gives better incremental oil recovery compared to the reverse cases, as shown in Fig. 5f. Abdeli and Seiden (2018) documented that the ratio of $[\text{SO}_4^{2-}]/[\text{Mg}^{2+}]$ concentrations in the injected water could be a controlling parameter in the process of changing the carbonate rock wettability. However, neither the ratio of $[\text{SO}_4^{2-}]/[\text{Mg}^{2+}]$ nor $[\text{SO}_4^{2-}]/[\text{Ca}^{2+} + \text{Mg}^{2+}]$ concentrations in the injected water revealed any clear relationship with the incremental oil recovery in the secondary recovery stage, as shown in Fig. 5g, h, respectively.

The same observations for the effects of the water minerals on the oil recovery during the tertiary WF are shown in Figs. 6 and 7. Figure 6 shows the relation between the additional oil recovery from carbonate core plugs in the tertiary

recovery stage versus the concentrations of the SO_4^{2-} , Ca^{2+} , Na^+ , Mg^{2+} , and K^+ in the injected low saline water. It is obviously clear that there is no clear relationship between the incremental oil recovery and the concentrations of the minerals in the injected water. The highest and lowest oil recovery values were documented with each mineral's presence and absence in the injected water. In addition, more oil recovery of up to 30% was observed with the absence of sulfates in the injected water.

In the collected data experiments, when the effects of the injected water minerals were studied during the tertiary recovery stage, each core plug was initially flooded with water (secondary recovery stage). The same core plug was then flooded with water of different salinity and composition at the same conditions in the tertiary recovery stage. The water in the tertiary stage may be HS or LS water. The additional oil recovery due to the change of the injected water salinity was calculated and reported.

Figure 7 shows the difference between the water minerals in the tertiary recovery stage. The negative values mean that the concentration of the secondary recovery stage components is lower than that in the tertiary recovery stage. However, the positive values mean that the concentration of the secondary recovery stage components is higher than that in the tertiary recovery stage. Figure 7a illustrates that a decrease in the $(\text{Na}^+ + \text{K}^+)$ cations concentration leads to an incremental oil recovery in most of the core plugs. However, some of the experiments reported an incremental oil recovery without changing the concentration of $(\text{Na}^+ + \text{K}^+)$ cations. The same observation was documented during the decrease in the concentration $(\text{SO}_4^{2-} + \text{Ca}^{2+} + \text{Mg}^{2+})$, as shown in Fig. 7b. Figure 7c, d demonstrate that the decrease in the concentration of the $(\text{Ca}^{2+} + \text{Mg}^{2+})$ cations or the SO_4^{2-} anions in the injected water can positively affect the tertiary WF's performance without any clear and consistent relation. However, Fig. 7e shows that the decrease in divalent and monovalent concentration leads to incremental oil recovery in most studies. However, the decrease of the divalent relative to the monovalent is better and gives better incremental oil recovery compared to the reverse cases, as shown in Fig. 7f. Neither the ratio of $[\text{SO}_4^{2-}]/[\text{Mg}^{2+}]$ nor $[\text{SO}_4^{2-}]/[\text{Ca}^{2+} + \text{Mg}^{2+}]$ concentrations in the injected water revealed any clear relationship with the incremental oil recovery in the tertiary recovery stage as shown in Fig. 7g, h, respectively.

The results of the previous studies indicate that the value of the water salinity has no effect on the oil recovery during the WF, but the composition of the injected water is the key parameter for the waterflooding in the carbonate reservoirs. In addition, defining a unique composition for all types of rocks is difficult. Moreover, decreasing the divalent cations Ca^{2+} and Mg^{2+} may increase the oil recovery in the secondary recovery stage. Moreover, it should be highlighted that

Fig. 5 Secondary incremental oil recover versus the difference in the concentration of the injected water minerals (Low salinity–High salinity)

a Effects of the difference in concentration of ($\text{Na}^+ + \text{K}^+$) on the oil recovery

b Effects of the difference in concentration of ($\text{Ca}^{2+} + \text{Mg}^{2+} + \text{SO}_4^{2-}$) on the oil recovery

c Effects of the difference in concentration of (SO_4^{2-}) on the oil recovery

increasing the sulfate concentration in the injected water may increase the project's cost. Furthermore, it was clear that the sodium iodide (NaI) role has not been sufficiently studied and needs further investigation. Also, it is obviously clear that experimental work for each specific reservoir is

necessary to determine the optimum salinity before the application of any waterflooding project (it is a specific application for a specific reservoir).

Fig. 5 (continued)

d Effects of the difference in concentration of $(\text{Ca}^{2+}+\text{Mg}^{2+})$ on the oil recovery e Effects of the difference in concentration of $d(\text{Ca}^{2+}+\text{Mg}^{2+}+\text{SO}_4^{2-}) + d(\text{Na}^++\text{K}^+)$ on the oil recovery f Effects of the difference in concentration of $d(\text{Ca}^{2+}+\text{Mg}^{2+}+\text{SO}_4^{2-}) - d(\text{Na}^++\text{K}^+)$ on the oil recovery

Effect of permeability and rock composition

Zoshchenko et al. (2019) showed that spontaneous imbibition of LSWF with a total salinity of ≈ 5 g/l could increase the recovery factor up to 4% and give additional recovery for core plugs with permeability less than 100 md. However, no direct relationship between the permeability and the incremental oil recovery was reported in the collected data, as shown in Fig. 8a, b. Additional incremental oil recovery was observed in all ranges of permeability.

The effect of the anhydrite has been studied in the literature as an important key parameter for LSWF applications. Austad et al. (2012) and Fathi et al. (2010) suggested that dissolution of the anhydrite into Ca^{2+} and SO_4^{2-} would generate in situ catalytic agent sulfate. This catalytic agent is required to release the carboxylic groups from the reservoir rock's surface.

Austad et al. (2012) and Fathi et al. (2010) did not observe any effect of applying the LSWF in chalk core plugs free of anhydrite. Austad et al. (2015) showed that the anhydrite

Fig. 5 (continued)

g Effects of the difference in concentration of $d(\text{SO}_4^{2-}/\text{Mg}^{2+})$ on the oil recovery h Effects of the difference in concentration of $d\text{SO}_4^{2-}/d(\text{Ca}^{2+}+\text{Mg}^{2+})$ on the oil recovery

(CaSO_4) appears to be the main factor for observing the low salinity EOR effect in carbonate reservoirs. However, they showed that anhydrite's effect in the LSWF is still debated and needs further investigation. Aksulu et al. (2012) indicate that the seawater would act as an EOR fluid in the limestone reservoirs, but its potential is probably smaller than the sea water's effect in the chalk rocks.

On the other hand, Gandomkar and Rahimpour (2015) conducted various WF experiments using 12 core plugs containing 10% anhydrite. These experiments did not achieve any incremental oil recovery in the tertiary recovery stage. They concluded that the limestone reservoir with anhydrite may not be suitable for the applications of the LSWF in the tertiary recovery stage. Their experiments were performed at 60 °C. It should be highlighted that their experiments in the secondary recovery stage showed opposite results. The core

plugs with 10% anhydrite showed 0 to 30% additional incremental oil recovery with HSWF and LSWF in the secondary recovery stage. Al Harrasi et al. (2012) and Hosseini et al. (2020) showed an incremental oil recovery of up to 20% using core plugs free of anhydrite with injected water free of sulfate. However, Uetani et al. (2019) achieved additional oil recovery in all the LSWF experiments regardless of the rock samples' anhydrite content. They reported that the anhydrite was not a mandatory mineral for the LSWF effect in the carbonate rock. In addition, the anhydrite dissolution was considered questionable as a key mechanism of the LSWF in the carbonate reservoirs. In another study, Jiang et al. (2014) did not observe any improvement in the oil recovery in their core flood experiments which were conducted using carbonate rock samples that contained anhydrite.

Fig. 6 (continued)

Fig. 6 Effects of the concentration of the injected water minerals on the oil recovery during the tertiary recovery stage

Figure 9a–c show the relation between the anhydrite, calcite, and dolomite concentration with the incremental oil recovery in the tertiary recovery stage. It should be highlighted that no clear relationship is observed between the rock minerals and the incremental oil recovery, as shown in Fig. 9. However, it is observed that increasing the concentration of the anhydrite may decrease the effectiveness of the LSWF in the tertiary recovery stage. It should be highlighted that the effect of anhydrite on the oil recovery in the secondary recovery stage was not plotted due to the unavailability of the data. The literature has few and limited experiments in

the available database for core plugs with rock descriptions in the secondary recovery stage.

Effect of oil composition

Den Ouden et al. (2015), Austad et al. (2012), Al-Khafaji et al. (2017), and Sari et al. (2017) documented that the higher incremental oil recovery with the application of the WF in the carbonate reservoirs is expected with the presence of the higher acidity oil due to the higher concentration of the carboxylates. The more carboxylates can be adsorbed on the carbonate rock surface, and the rock becomes more oil-wet. Decreasing the salinity shifts the polarity of zeta potential at the fluid-rock interface; accordingly, the wettability is changed. Sari et al. (2017) showed that the LSWF incremental oil recovery in the tertiary recovery stage may be attributed to shifting the rock's wettability from strongly water wet to intermediate wettability. Uetani et al. (2020) reported insignificant additional oil recovery by the application of the tertiary LSWF in core plugs containing low acidity oil. They documented that repeating the experiments with high acidity oil showed incremental oil recovery up

Fig. 7 Tertiary incremental oil recovery versus the difference in the concentration of the injected water minerals (Secondary–Tertiary)

a Effects of the difference in concentration of $d(\text{Na}^+ + \text{K}^+)$ on the oil recovery

b Effects of the difference in concentration of $d(\text{Ca}^{2+} + \text{Mg}^{2+} + \text{SO}_4^{2-})$ on the oil recovery

c Effects of the difference in concentration of $d(\text{SO}_4^{2-})$ on the oil recovery

to 23%. Then, they concluded that the total acid number (TAN) and the recovery mode appear to be the major factors for successful LSWF applications in the carbonate

system. Furthermore, Mozaffari et al. (2021) reported that the various acidic molecules in the crude oil, such as alkyl carboxylic acids, fused aromatic ring acids, alkylbenzene

Fig. 7 (continued)

d Effects of the difference in concentration of $d(\text{Ca}^{2+}+\text{Mg}^{2+})$ on the oil recovery **e** Effects of the difference in concentration of $d(\text{Ca}^{2+}+\text{Mg}^{2+}+\text{SO}_4^{2-}) + d(\text{Na}^{+}+\text{K}^{+})$ on the oil recovery **f** Effects of the difference in concentration of $d(\text{Ca}^{2+}+\text{Mg}^{2+}+\text{SO}_4^{2-}) - d(\text{Na}^{+}+\text{K}^{+})$ on the oil recovery

carboxylic acids, and naphthenic acids, could reduce the IFT to a significant degree. Shechter et al. (2020) showed that changing the IFT of oil in water emulsion could alter the configuration of liquid crystals inside the oil droplet.

On the other hand, Al-Khafaji et al. (2017) and Masalmeh et al. (2019) indicated that the applications of the LSWF in the carbonate reservoir do not appear to be an effective technique with high acidity and heavy crude oil. However, Maskari et al. (2020) showed that the wettability change

could occur in the carbonate reservoirs with crude oils containing lower polar components concentration.

Figure 10 shows the relation between the incremental oil recovery versus the total acid number and total base number in both secondary and tertiary recovery stages, respectively. It is clear that the number of available experiments which have TAN and TBN information in the database is limited. In addition, the available data indicate that there is no clear relationship between the incremental oil

Fig. 7 (continued)

g Effects of the difference in concentration of $d(\text{SO}_4^{2-}/\text{Mg}^{2+})$ on the oil recovery h Effects of the difference in concentration of $d\text{SO}_4^{2-}/d(\text{Ca}^{2+}+\text{Mg}^{2+})$ on the oil recovery

recovery and the TAN or TBN in both secondary and tertiary recovery stages.

Effect of temperature

The LSWF effect was studied at different temperatures in the literature. Zahid et al. (2012) and Fathi et al. (2010) found that the LSWF could cause a dissolution of rock material and increase the oil recovery from carbonate core plugs at 90 °C. In another study, Aksulu et al. (2012) found that the high temperatures ($T > 130$ °C) can cause Mg^{2+} cations to substitute the Ca^{2+} cations on the carbonate surface; and accordingly, this behavior increases the oil recovery. In addition, Austad et al. (2012) documented that the reservoir temperature should be higher than 70–80 °C to obtain additional oil from the application of the LSWF in the carbonate rock. However, Shariatpanahi et al. (2011a), Zahid et al. (2012), and Mahani et al. (2018) reported that the application of the

LSWF is an effective process when it is applied in carbonate core plugs at ambient temperature.

Figure 11 shows the relation between the incremental oil recovery and several experiments' temperatures in both secondary and tertiary recovery stages. It is obviously clear that there is no distinct and direct relationship between the temperature and the incremental oil recovery in the secondary and tertiary recovery stages. It should be highlighted that incremental oil recovery values were documented with experiments of LSWF in carbonate rock during the tertiary recovery stage at a temperature higher than 60 °C. However, in the secondary recovery stage, the incremental oil recovery values were observed at ambient temperature (Ghandi et al. 2019 and Abdelhamid and Elnaggar 2017).

Fig. 8 Effects of the rock permeability on the oil recovery during the secondary and tertiary recovery stages

Statistical analysis

The previous discussion revealed many different parameters and mechanisms that influence LSWF applications in carbonate reservoirs. Therefore, statistical analysis was performed using Fractional factorial design (FFD) to identify the high effective parameters for incremental oil recovery in both secondary and tertiary recovery stages. Minitab software Version 19 was used to apply these statistical analyses (Minitab 2020).

The fractional factorial design is one of the most important statistical contributions to exploring the influence of many controllable factors on interest response (Gunst and Mason 2009). It is a commonly used experimental design method that can be used to enhance and simplify the studies. Plackett–Burman design is a popular type used for the FFD analysis. It is used to study the dependence of some measured quantity on several independent variables/factors (Plackett and Burman 1946). On the other hand, a Pareto chart is often a good tool for presenting the statistical analysis results and can be used to present the results of the Plackett–Burman design. The Pareto chart clarifies the

Fig. 9 Effects of the rock mineral types on the oil recovery during the tertiary recovery stage

absolute values of the standardized effects from the largest effect to the smallest effect, and the magnitude of each factor is clarified by a column (Arnold 2014).

This study used a two-level folded Plackett–Burman design to determine the high effective parameters on the incremental oil recovery in both secondary and tertiary recovery stages. The analysis was performed using rock minerals, HS water, and LS water properties. Several experiments in the database have a complete set of all parameters, while others did not have the rock's properties as a result that the statistical analysis was carried out twice. The first statistical analysis was performed using all data without considering the parameters of the rock properties. However, the second statistical analysis was carried out using the

Fig. 10 Incremental oil versus oil acid and base numbers in the secondary and tertiary recovery stages

a Effects of the oil acid and base numbers on the oil recovery during the secondary recovery stage

b Effects of the oil acid and base number on the oil recovery during the tertiary recovery stage

experiments, which included rock properties. Furthermore, the analysis was performed for both secondary and tertiary recovery stages individually. Accordingly, four statistical analyses were carried out to identify the parameters which have the most influence on the LSWF.

Figure 12 shows the statistical analysis for the results of the secondary recovery stage experiments when using all data sets without including the rock minerals in the regression. It is clear that the greatest influence parameters in the secondary WF are the minerals concentrations of the HS injected water (Mg^{2+} , Na^+ , K^+ , and Cl^-). However, the effect of the temperature and the concentration of two water minerals (Ca^{+2} and SO_4^{2-}) in the injected water was found to be moderate. In addition, the effects of the TDS and the concentrations of the other water minerals in the oil recovery appear to be minimal. It should be highlighted that the mineral description for the core samples in the secondary recovery stage is very limited to extract a solid conclusion and a general result. The statistical analysis results are matched

with the previous discussion in the previous section of this work. The results reveal that adjusting the injected water minerals can lead to higher oil recovery of the secondary WF in the carbonate reservoir.

Figure 13a, b show the statistical analysis results for the application of the LSWF in the tertiary recovery stage without and with the rock minerals, respectively. It is obviously clear that the statistical analysis of the results of the tertiary recovery stage experiments without considering the rock minerals in the regression (Fig. 13a) indicates that the most dominant parameters in the LSWF applications in the carbonate reservoir are the rock permeability, the HCO_3^- concentration in the LS injected water, and the SO_4^{2-} concentration in the HS injected water. When the rock minerals are taken into considerations during the statistical analysis for the results of the tertiary recovery stage experiments (Fig. 13b); the rock minerals and the concentrations of several minerals (K^+ , Cl^- , Ca^{2+} , SO_4^{2-} , Na^+ , and HCO_3^-) in the

a Effects of temperature on the oil recovery during the secondary recovery stage

b Effects of temperature on the oil recovery during the tertiary recovery stage

Fig. 11 Incremental oil recovery versus temperature in the secondary and tertiary recovery stages

LS injected water appear to be the most effective parameters in the tertiary LSWF.

As shown in Fig. 13 a, b the statistical analysis without considering the rock minerals showed that the SO_4^{2-} concentration of the HS is the highest effective parameter. The

anhydrite dissolution is one of the sources for the in situ SO_4^{2-} . Therefore, while considering rock minerals, the effect of anhydrite seems to be the most dominant parameter.

It is obviously clear that the results of the statistical analysis are matched with the previous discussion in the previous sections. All results indicate that the composition of the injected water is more important than its salinity. In addition, it was found that changing the water composition regardless of its salinity can increase oil recovery in both secondary and tertiary recovery stages.

Summary and conclusions

In this work, a comprehensive review and analysis were conducted to derive new findings and screening criteria for waterflooding projects' applications in the carbonate reservoirs. Based on the results of this study, the following conclusions can be made:

- Wettability alteration by LSWF in the carbonate reservoirs is a very complicated mechanism.
- There are various factors that affect the WF process in the carbonate reservoirs, such as water composition, oil composition, reservoir rock properties, and temperature.
- The salinity value of the injected water appears to have an insignificant effect on oil recovery. However, water composition is the key parameter for successful waterflooding (WF) projects.
- Adjusting injected water composition (when cost allows) can improve oil recovery up to 30%.

Fig. 12 Pareto chart for the statistical analysis of the results of the secondary recovery stage without considering the rock minerals

Fig. 13 **a** Pareto chart for the statistical analysis of the results of the tertiary recovery stage without considering the rock minerals. **b** Pareto chart for the statistical analysis of the results of the tertiary recovery stage with considering the rock minerals

- Changing water salinity from LSWF to HSWF can also lead to incremental oil recovery up to 18% in the tertiary recovery stage.
- Applying the optimum salinity and composition WF in the secondary recovery stage is more effective than applying it in the tertiary recovery stage.
- The greatest influence parameters in the secondary recovery stage are the concentrations of Mg^{2+} , Na^+ , K^+ , and Cl^- in the injected water.
- The most dominant parameters in the tertiary recovery stage are the rock minerals and the concentration of K^+ , HCO_3^- , and SO_4^{2-} in the injected water.
- The effects of HCO_3^- and Iodide have not been well studied in the literature and need further investigation.
- There are no clear relations between the incremental oil recovery and oil properties (TAN and BAN) in both secondary and tertiary recovery stages.
- Incremental oil recovery values were documented in all ranges of flooding temperatures.

- It is technically difficult to introduce a unique composition in LSWF for wettability alteration for all types of rocks. Experimental work for each specific reservoir is essential for determining the optimum composition. “LSWF is a specific process for a specific reservoir.”

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Declarations

Conflict of interest On behalf of all authors, I state that there is no conflict of interest.

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