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# Role of the injected water salinity and ion concentrations on the oil recovery in carbonate reservoirs

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#### ABSTRACT

Low salinity water flooding (LSWF) was initially considered using water with a low concentration of dissolved salts and was later extended to include modifying the ionic content of injected brines. This work investigates the effects of changing water salinity and composition along with the concentration of sulfate and iodide ions on oil recovery in carbonate reservoirs during the tertiary recovery stage. An experimental study was carried out using crude oil of 29°API, 8 core samples extracted from the Eocene carbonate reservoir (Egypt), and 6 different water salinities.

The results showed additional oil recovery up to 5% of the original oil in place (OOIP) in the tertiary recovery stage with changing water salinity and water composition. Injection of high salinity (HS) and low salinity (LS) brines with high sulfate concentrations increased the incremental oil recovery by a value ranging from 1.7 to 3.8% of the OOIP. On the contrary, injection of HS and LS brines with low sulfate concentrations showed insignificant incremental oil recovery (less than 1% of the OOIP). Furthermore, injection of water with potassium iodide (KI) after injection of water with high sulfate brines showed additional oil recovery of about 1.7% of the OOIP. On the other hand, injection of water with KI after injection of water with low sulfate concentration showed insignificant incremental oil recovery (less than 0.4% of the OOIP).

The concentration of sulfate in the injected water appeared to be key parameter to achieve effective waterflooding (WF) projects in carbonate reservoirs. Moreover, the results revealed that the multi-component ion exchange (MIE) mechanism seems to be the primary recovery mechanism for LSWF in carbonate reservoirs. The results and conclusions of this study can be used to develop guidelines for designing waterflooding projects in carbonate reservoirs with optimum salinity.

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#### 1. Introduction

The effect of LSWF on carbonate reservoirs was investigated by Bagci et al. (2001). Their experiments showed 18.8% higher oil recovery by injecting 2% KCl plus 2% NaCl brine mixture rather than injecting other brine compositions. Later, Austad et al. (2005), Strand et al. (2008a), Al Shalabi and Sepehrnoori (2017), and others concluded that the effect of injected water composition in carbonate reservoirs has a significant effect on oil recovery rather than salinity. Furthermore, Snosy et al. (2020) and Snosy et al. (2021a)

documented that there is an optimum salinity and optimum composition of the injected water for a successful WF project in sandstone reservoirs. Therefore, the definition of LSWF was considered as a 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 technique was also identified in the literature as Smart Waterflood, Designer Waterflood, Advanced Ion Management, Modified Salinity Flooding, Ion Tuning, and Engineered Water Injection.

The laboratory work of LSWF in carbonate rocks showed incremental oil recovery up to 20% higher than the HSWF in the studies by Strand et al. (2008b), Lightelm et al. (2009), and Yousef et al. (2011) and others. Various mechanisms for the effect of the LSWF on oil recovery in carbonate reservoirs were proposed with

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Nomenclature		MIE	Multi-component ion exchange	
		NA	Not available	
		OOIP	Original oil in place	
Abbreviations		PDI	Potential determining ions	
API	The American Petroleum Institute gravity	TDS	Total dissolved solids	
EOR	Enhanced oil recovery	WF	Waterflooding	
HS	High salinity brine			
HS-HSU	High salinity — high sulfate brine	Symbols		
HS-LSU	High salinity — low sulfate brine	Ka	Air permeability, md	
HSWF	High salinity waterflooding	PV	Pore volume, CC	
IFT	Interfacial tension	$S_{or}$	Residual oil saturation	
LS	Low salinity brine	Sw	Water saturation	
LS-HSU	Low salinity – high sulfate brine	$S_{wi}$	Initial water saturation, fraction	
LS-LSU	Low salinity – low sulfate brine	Φ	Porosity, fraction	
LSWF	Low salinity waterflooding		•	

no agreement on the effective primary mechanism. These proposed mechanisms included fine migration, interfacial tension (IFT) reduction, multi-component ion exchange (MIE), salting-in, double-layer effects, calcite dissolution, anhydrite dissolution, water micro-dispersions, and osmosis pressure effects. Suijkerbuijk et al. (2012) documented that several mechanisms can be working together, but the right mechanism behind the LSWF has not yet been discovered.

Two mechanisms were proposed to clarify the effects of injecting brine of sulfate anion in carbonates: MIE and anhydrite dissolution mechanisms. The MIE evidence of the LSWF in the carbonate reservoirs was proposed by Austad et al. (2005). Then, it was accepted by Strand et al. (2008a), Zhang and Austad (2006), Zhang et al. (2007), Puntervold (2008), Lager et al. (2007), Ligthelm et al. (2009), and Myint and Firoozabadi (2015). However, the mechanism of the anhydrite dissolution was proposed by Austad et al. (2011). They suggested that the anhydrite (CaSO<sub>4</sub>) dissolution into Ca<sup>2+</sup> cations and SO<sub>4</sub><sup>2-</sup> anions generates in-situ SO<sub>4</sub><sup>2-</sup> anions. The sulfate anions act as a catalyst agent in the wettability alteration process. In another study, Austad et al. (2015) concluded that the anhydrite (CaSO<sub>4</sub>) dissolution mechanism seemed to be key factor for obtaining incremental oil recovery during the LSWF in the carbonate reservoirs.

As a result of these two mechanisms, Yousef et al. (2010), Shariatpanahi et al. (2011), Fathi et al. (2010), Hognesen et al. (2005), Zekri et al. (2019), and Al-Attar et al. (2013) hypothesized that injection of sea water containing a high amount of  $SO_4^{2-}$  anions can alter the wettability to be more water-wet and increase oil recovery in carbonate reservoirs. In another study, Zhang et al. (2007) documented that increasing the concentration of  $SO4^{-2}$  anions and decreasing NaCl concentration in the injected water cause a change in the wettability and improve the oil recovery in the carbonate reservoirs. Chandrasekhar and Mohanty (2013) and Yousef et al. (2012) documented that the diluted sea water alters the carbonate rock wettability to a more water-wet state.

On the other hand, the MIE mechanism was not agreed upon by Abdeli and Seiden (2018), and Zekri et al. (2019). They concluded that adding sulfate into the water would increase the process's overall cost without improving oil recovery. In addition, the anhydrite dissolution mechanism was not accepted by Jiang et al. (2014), Romanuka et al. (2012), Nasralla et al. (2014), and Uetani et al. (2019). They documented an unclear correlation between improved oil volume due to the low salinity effect and the amount of anhydrite dissolution. In addition, Gandomkar and Rahimpour (2015) concluded that the limestone reservoir with anhydrite

may not be suitable for the applications of the LSWF in the tertiary recovery stage. Furthermore, Al Harrasi et al. (2012) and Hosseini et al. (2020) showed an incremental oil recovery of up to 20% using core samples free of anhydrite with injected water free of sulfate.

Furthermore, Al Hamad et al. (2017) clarified that sodium iodide can alter the rock wettability to a stronger water-wet in carbonate reservoirs. In addition, they documented an increase of 3–17% incremental oil recovery using dolomitic core samples by increasing NaI from 0 ppm to 1000 ppm in the injected water and with no change in the other minerals. Moreover, they reported that using limestone core samples increased oil recovery by 0–17% in the tertiary recovery stage by changing the salinity and diluting the seawater (the sea water was diluted to 1000 ppm saline water) to 1000 ppm of NaI minerals only. However, the effect of iodide is not fully understood in the literature.

This work is concerned with studying the effects of sulfate and iodide anions on the oil recovery during the applications of the LSWF in the tertiary recovery stage. The work attempts to explore the reasons behind improved LSWF tertiary recovery under some conditions and to help in designing successful LoSal applications.

#### 2. Experimental setup and procedure

Eight core samples were extracted from the carbonate-producing reservoir in the Eocene formation in Egypt. Eocene is a fractured well bedded limestone section, argillaceous in some parts and with dolomitic limestone in some intervals. The thickness of the Eocene varies rapidly over the field from 125 m to 550 m. Thickness variations are believed to be a combined result of syntectonic depositional variations and subsequent Oligocene erosion. The porosity range varies from 10% up to 30%. The core was extracted from the middle of the formation, which is almost pure limestone and consists of grain-stone and pack-stone limestone with pore storage type. The core samples have 1.5" diameter and 2" length. Table 1 shows the core samples' basic properties. The core samples were divided into four groups according to the experimental procedure.

Crude oil of 29° API and six sets of water samples with different salinities and sulfate concentration were used in this study. Tables 2 and 3 summarize the main fluid properties. The water samples are mainly: (1) Base/Reference water sample (2) high salinity- high sulfate (HS-HSU) sample, (3) high salinity- low sulfate (HS-LSU) sample, (4) low salinity- high sulfate (LS-HSU) sample, (5) low salinity- low sulfate (LS-LSU) sample, and (6) KI water sample. It

**Table 1** Core samples properties.

Sample No.	Group No.	PV (cm <sup>3</sup> )	Φ (%)	Ka (md)	Grain Density (g/cm³)	Swi (% PV)
C-1	1	13.0	24.7	37.06	2.69	18.4
C-2		10.5	17.0	75.66	2.79	16.9
C-3	2	8.5	12.0	5.73	2.81	28.6
C-4		16.5	27.3	19.63	2.69	25.3
C-5	3	15.9	25.3	21.89	2.68	23.7
C-6		15.9	25.6	21.77	2.69	24.0
C-7	4	14.8	25.6	15.41	2.68	27.1
C-8		16.3	25.2	25.88	2.70	22.6

**Table 2** Injected water properties.

Fluid Type	TDS	Na <sup>+</sup>	K <sup>+</sup>	Mg <sup>2+</sup>	Ca <sup>2+</sup>	Cl <sup>-</sup>	SO <sub>4</sub> <sup>2-</sup>	I
	ppm	Ppm	ppm	ppm	ppm	ppm	ppm	ppm
Base	39,226	14,958	299	52	120	23,391	406	0
HS-HSu	80,850	30,692	598	104	240	46,782	2,434	0
LS-LSu	3,923	1,496	30	5	12	2,339	41	0
HS-LSu	77,311	29,546	598	104	240	46,782	41	0
LS-HSu	5,532	1,904	15	3	6	1,170	2,434	0
KI	1,500	0	354	0	0	0	0	1146

should be highlighted that the Fluid-Fluid compatibility tests between the different water salinities were conducted before proceeding with core flood experiments. The results of the compatibility tests did not show any scales formation due to the injection of these different types of water samples.

The core samples were divided into four groups. Each group contained two core samples. All core samples were initially flooded with the Base water as a secondary recovery stage. After that, each group was flooded with different water samples in the tertiary recovery stage to investigate the effects of sulfate on oil recovery. Then, five core samples were injected with KI to realize any additional oil recovery due to the change in the water salinity. Fig. 1 summarizes the experimental procedure for all groups. It should be highlighted that all core samples were prepared for flooding experiments with the same procedure as follows:

- The core samples were cleaned from the hydrocarbons by the Soxhlet extraction using chloroform.
- The core samples were then dried using humidity drying in a conventional oven at 140 °F. After that, the core samples were cooled in a sealed room temperature condition.
- Porosity and grain density were measured by a double cell helium expansion gas porosimeter.
- Gas permeability was measured with air using a calibrated steady state permeameter.
- Synthetic formation brine was used to saturate the core samples with 100% brine.
- The fully saturated core samples were placed in individual hydrostatic core holders under a confining pressure of 2000 psig.

Oil was injected into the top of the core until water production ceased.

- The overburden pressure was increased gradually to the required effective value of 4,100 psi. The amount of the displaced oil by the decrease in the pore volume was determined. As the pore space changed and the water volume remained constant, the initial water saturation (S<sub>wi</sub>) as a percentage of pore space was recalculated.
- The core samples were then restored in an aging cell under reservoir temperature and pressure (200 °F and 1600 psig) for 28 days.
- The restored state core samples were placed in individual hydrostatic core holders at reservoir conditions (200 °F and 1600 psig).
- The core samples were subjected to water injection using the Base brine as secondary flooding until the oil production ceased at reservoir conditions.
- The core samples were subjected to waterflooding using different brine salinity as tertiary flooding, and the excess oil production was measured.
- After the tertiary recovery stage, five core samples were subjected to waterflooding using KI brine salinity, and the excess oil production was also measured.

#### 3. Results

Table 4 summarizes the results of the flooding experiments. It presents the oil recovery of the eight-core samples during the secondary and tertiary recovery stages in addition to the results obtained after flooding with the KI water at the end (for five core samples). The core samples of the first group (C1 and C2) were flooded with Base brine in the secondary recovery stage and followed by HS-HSU brine in the tertiary recovery stage. The oil recovery of C-1 and C-2 in the secondary recovery stage were 70.7% and 65.2 of the OOIP, respectively. When HS-HSU brine was used to flood these two core samples in the tertiary stage, the incremental oil recovery was 2.5% and 3.8% of the OOIP, respectively.

The core samples of the second group (C-3 and C-4) were flooded with Base brine in the secondary recovery stage and followed by HS-LSU brine in the tertiary recovery stage. These core samples displayed (1) oil recovery of 47% and 54% of the OOIP in the

**Table 3**Density and viscosity of oil and injected water.

Fluid Type	Density (gms/cc) @ 70°F	Density (gms/cc) @ 200°F	Viscosity (gms/cc) @ 70°F	Viscosity (gms/cc) @ 200°F
Base	1.03	1.01	1.32	0.50
HS-HSU	1.06	1.04	1.54	0.57
LS-LSU	1.01	0.89	1.22	0.44
HS-LSU	1.05	1.03	1.49	0.54
LS-HSU	1.01	0.95	1.23	0.41
Oil	0.92	0.86	219.09	9.49

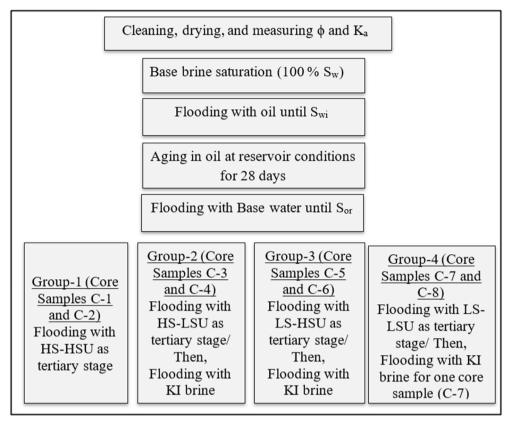


Fig. 1. Experimental procedure flow chart.

**Table 4**Oil recovery in the secondary and tertiary recovery stages.

Sampl	e Ka (md)	Injected Fluid in the Secondary Recovery Stage	Injected Fluid in the Tertiary Recovery Stage	Secondary Oil Recovery, % OOIP	Tertiary Additional Oil Recovery, % OOIP	KI Additional Oil Recovery, % OOIP
C-1	37.06	Base Brine	HS-HSU <sup>b</sup>	70.7%	2.5%	NA <sup>a</sup>
C-2	75.66			65.2%	3.8%	NA <sup>a</sup>
C-3	5.73		HS-LSU <sup>b</sup>	47.2%	0.8%	0.0%
C-4	19.63			54.0%	0.0%	0.1%
C-5	21.89		LS-HSU <sup>b</sup>	75.4%	1.7%	1.7%
C-6	21.77			75.2%	2.6%	1.7%
C-7	15.41		LS-LSU <sup>b</sup>	60.1%	1.0%	0.4%
C-8	25.88			68.8%	0.8%	NA <sup>a</sup>

LS-HSU: Low salinity — high sulfate brine LS-LSU: Low salinity — low sulfate brine.

secondary recovery stage, and (2) incremental oil recovery of 0.8% and 0.0% in the tertiary recovery stage, respectively.

The third group core samples (C-5 and C-6) were flooded with Base brine in the secondary recovery stage and followed by LS-HSU brine in the tertiary recovery stage. The oil recovery of C-5 and C-6 in the secondary recovery stage were 75.4% and 75.2% of the OOIP, respectively. After flooding these core samples with HS-LSU brine in the tertiary stage, the oil recovery increased by 1.7% and 2.6% of OOIP, respectively.

The core samples of the fourth group (C-7 and C-8) were flooded with Base brine in the secondary recovery stage and followed by HS-HSU brine in the tertiary recovery stage. The oil recovery in the secondary recovery stage were 60% and 68% of the OOIP, respectively. When the core samples were flooded by HS-HSU's brine in the tertiary stage, the incremental oil recovery was 1% and 0.9% of the OOIP, respectively.

Furthermore, waterflooding using KI brine was continued in the tertiary recovery stage using five core samples (from C-3 to C-7). In these experiments, Core samples C-3 and C-4 showed nil additional oil recovery. However, Core samples C-5, C-6 and C-7 reported an incremental oil recovery of 1.7%, 1.7% and 0.4% of the OOIP, respectively.

Figs. 2 and 3 show the oil recovery of the eight core samples during the secondary and tertiary recovery stages. Fig. 2 shows the results of the core samples when they were flooded with high sulfate brine in the tertiary recovery stage. While, Fig. 3 shows the results of the core samples when they were flooded with low sulfate brines in the tertiary recovery stage. The results reported additional oil recovery of less than 4% of the OOIP in all core samples. However, the brines with high sulfate content showed additional oil recovery of 1.7and 3.8% of the OOIP. The highest additional oil recovery (3.8% and 2.5% of the OOIP) was shown in Core samples

<sup>&</sup>lt;sup>a</sup> NA: Not available (the experiments with KI brine were not performed for these samples).

<sup>&</sup>lt;sup>b</sup> HS-HSU: High salinity – high sulfate brine HS-LSU: High salinity – low sulfate brine.

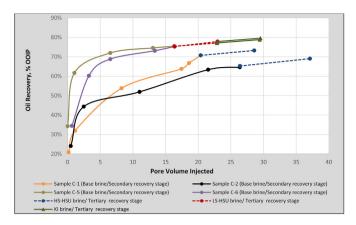


Fig. 2. Oil recovery for core samples C-1, C-2, C-5, and C-6.

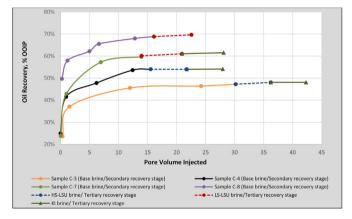


Fig. 3. Oil recovery for core samples C-3, C-4, C-7, and C-8.

C-1 and C-2. These two samples were flooded with high salinity brine in the tertiary recovery stage after LSWF was applied in the secondary recovery stage. In the contrary, flooding the core samples with LS and HS brines along with low sulfate concentration in the tertiary recovery stage showed insignificant effect on the oil recovery. The results confirmed that the sulfate concentration has the highest impact on the LSWF projects in carbonate reservoirs. Moreover, the total dissolved solids (TDS) seem to have insignificant effect on the additional oil recovery due to the tertiary recovery stage.

#### 4. Discussion

There are more than nine proposed mechanisms for the effect of LSWF in carbonates. However, only the MIE mechanism attributed the effect of LSWF to sulfate anions in carbonate reservoirs (without anhydrite). This mechanism is attributed to the adsorption of potential determining ion (PDI)  $SO_4^{2-}$ ,  $Ca^{2+}$  and/or  $Mg^{2+}$  onto the rock surface. The adsorption of  $SO_4^{2-}$  anions decreases the positive charge density on the rock surface, which minimizes the electrostatic repulsive force and causes co-adsorption of  $Ca^{2+}$  and  $Ca^{2+}$  cations on the rock surface. The  $Ca^{2+}$  cations reaction with the carboxylic acid groups breaks the attractive interactions between the oil and rock interface, which changes rock surface into more water wet.

The experimental results showed incremental oil recovery during flooding with a high sulfate concentration water and insignificant effect with low sulfate concentration water in the tertiary recovery stage. Therefore, it seems that the MIE mechanism (which is dependent on the presence of sulfate ions) is the primary mechanism for the LSWF in carbonates.

It should be highlighted that the results of this work disagree with the results of Jiang et al. (2014). They reported that increasing the sulfate concentration beyond some optimum concentration of 47 ppm can cause a negative effect on the flooding process. However, the same positive effect of the sulfate concentration on oil recovery was documented in the secondary recovery stage by Abdelhamid and Elnaggar (2017) using core samples from Egypt's Eocene reservoir. They performed lab experiments using 3 different water samples with different sulfate ion concentrations. They documented that applying WF with sea water (35000 ppm TDS and 2700 ppm Sulfate) achieved higher oil recovery (up to 6%) than applying WF with formation water (152000 ppm TDS and 1300 ppm Sulfate) or low salinity water (with NaCl 1000 ppm TDS and zero ppm Sulfate) brines. In addition, flooding the core samples with brine of high salinity and high sulfate content produced higher oil recovery in the secondary recovery stage than applying WF with brine of low salinity and zero sulfate content.

Moreover, it should be highlighted that there is no correlation or direct relation between the additional oil recovery and the core samples' permeabilities as shown in Fig. 4.

Furthermore, the Core samples C-3 to C-7 were flooded with KI after they were flooded by low and high sulfate brines in the tertiary recovery stage. Additional oil recovery of 1.7% OOIP was recorded in the core samples that were flooded with high sulfate brines followed by KI. In contrast, insignificant additional oil recovery was observed when the core samples were flooded by KI after they were flooded by the low sulfate concentration brine.

Based on the results of this work, two mechanisms for increasing oil recovery due to increasing Iodide ions in the injected water are proposed. The first mechanism is the MIE mechanism: it is proposed that the Iodide ions play the same role as the sulfate ions. Adsorption of I<sup>-</sup> and SO<sub>4</sub><sup>2</sup> anions decreases the positive charge density on the rock surface, which minimizes the electrostatic repulsive force and causes co-adsorption of Ca<sup>2+</sup> and Mg<sup>2+</sup> cations on the rock surface. The Ca<sup>2+</sup> cations reaction with the carboxylic acid groups breaks the attractive interactions between the oil and rock interface, which changes rock surface into more water wet. This mechanism is supported by Al Hamad et al. (2017) experiments which reported that adding sodium iodide to the sea water allowed the alteration of rock wettability to stronger waterwet conditions. The second mechanism is related to the iodide effect on the oil recovery. The I- anions adsorption onto the rock surface causes the desorption of Ca<sup>2+</sup> cations from the rock surface as shown by Equation (1) (Feng and Redfern, 2018).

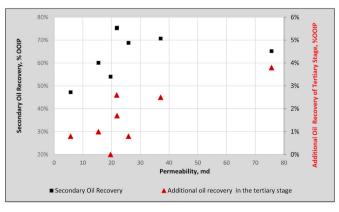


Fig. 4. Core samples rock permeability versus oil recovery.

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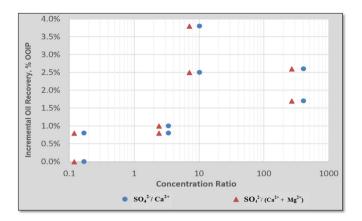


Fig. 5. Incremental oil recovery versus  $SO_4^{2-}/Ca^{2+}$  and  $SO_4^{2-}/(Ca^{2+}+Mg^{2+})$  concentrations ratio.

$$24(Ca (CO_3)) + 0.5I_2 \leftrightarrow Ca_{23} I (CO_3)_{24} + Ca^{2+}$$
 (1)

It should be emphasized that the desorption of the active cation Ca<sup>2+</sup> is associated with the desorption of organic materials. In addition, the sulfate ions increase the ability of the adsorption of iodide to the rock surface. It should be highlighted that the increase of sulfate ions in the injected water causes an increase in the total cost of the project due to its effect on the production facilities. Therefore, it is recommended to apply WF projects in carbonate reservoirs with water containing iodide and sulfate ions (with lower concentration than sea water).

Furthermore, Fig. 5 indicates that there is no clear relationship between the ratio of sulfate ion concentration and calcium ion concentration ([SO $_4^2$ ]/[Ca $^2$ +] or [SO $_4^2$ ]/[Ca $^2$ + + Mg $^2$ +]) of the injected water on the incremental oil recovery. This observation is consistent with the results of Snosy et al. (2021b) who did not observe any clear relationship between the ratio of [SO $_4^2$ ]/[Mg $^2$ +] or [SO $_4^2$ ]/[Ca $^2$ + + Mg $^2$ +] concentrations in the injected water with the incremental oil recovery.

The experimental results of this work indicate that the salinity value of the injected water appears to have an insignificant effect on oil recovery. However, the water composition is the key parameter for successful waterflooding projects in carbonate reservoirs on both secondary and tertiary recovery stages. These results are consistent with the results of another previous comprehensive analysis (Snosy et al., 2021b).

#### 5. Conclusions

- Additional oil recovery up to 5% of OOIP was documented with changing water salinity and composition in the tertiary recovery stage.
- 2. Changing water salinity (not only reducing water salinity) may achieve higher oil recovery in carbonate reservoirs. In addition, sulfate content has important consequences on the injection of both HS and LS brines in the tertiary stage as follows:
  - (a) high sulfate concentrations yielded additional oil recovery (range from 1.7% to 3.8% of the OOIP).
  - (b) low sulfate concentrations showed insignificant incremental oil recovery (less than 1% of the OOIP)
- 3. The multi-component ion exchange (MIE) mechanism seems to be the primary recovery mechanism for the LSWF in the carbonate reservoirs.
- 4. Injecting water of potassium iodide (KI) component after the injection of high sulfate brines showed 1.7% of the OOIP as an incremental oil recovery.

- 5. The KI brine showed insignificant incremental oil recovery (less than 0.4% of the OOIP) when it was injected after the low sulfate concentration brine.
- 6. Iodide ions can increase oil recovery due to (a) the MIE mechanism similar to the sulfate ions effect or (b) the desorption of the organic components associated with the desorption of calcium cation from the rock surface.
- 7. WF with water containing iodide and sulfate ions (lower concentration than sea water) can cause an increase in the oil recovery without increasing the project cost.

#### **Declaration of competing interest**

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