

Original article

Numerical investigation of optimum ions concentration in low salinity waterflooding

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Abstract:

Injecting low saline water is one of the practices used to improve hydrocarbon production that has recently significantly grown due to its advantages over seawater and chemical flooding. Although many theories and mechanisms have been provided on how additional oil recovery has been achieved utilizing low salinity waterflooding, the principle fundamentals of the mechanism(s) are still ambiguous. This article investigates the potential use of low salinity waterflooding (LSWF) to improve oil production from a sandstone formation. A 3D field-scale model was developed using Computer Modeling Group (generalized equation-of-state model simulator) based on a mature oil field data. The developed model was validated against actual field data where only 8% deviation was observed. Simulation analysis indicated that multi-component ion exchange is a key factor to improve oil production because it alters rock wettability from oil-wet to water-wet. Simulation sensitivity studies showed that low salinity water flooding provided higher oil production than high water salinity flooding. Moreover, simulation showed early breakthrough time of low salinity water injection can provide high oil recovery up to 71%. Therefore, implementing LSWF instantly after first stage production provides recovery gains up to 75%. The determined optimal injected brine composition concentration for Ca^{2+} , Mg^{2+} and Na^{+} are 450, 221, and 60 ppm, respectively. During LSWF, a high divalent cations and low monovalent cations' concentration can be recommended for injected brine and formation aquifer for beneficial wettability alteration. Simulation also showed that reservoir temperature influenced the alteration of ion exchange wettability during LSWF as oil recovery increased with temperature. Therefore, high temperature sandstone reservoirs can be considered as a good candidate for LSWF.

1. Introduction

Reservoir fluid production is accomplished through three different stages. The primary reservoir fluid recovery refers to fluid production by natural reservoir drive mechanism(s) such as aquifer driver, gas cap drive, gravity drainage, solution-gas drive and rock and fluid expansions. This primary stage may also involve utilizing artificial pumps to improve reservoir fluid production. Through primary recovery, only 5% to 15% of oil originally in place (OOIP) is usually produced (Santa et al., 2011).

Depletion of natural reservoir energy overtime causes significant decline in oil production, therefore; an external energy

is introduced to maintain formation pressure, and therefrom, improve hydrocarbon production. The second 'conventional' stage of hydrocarbon recovery mechanism is implemented by introducing an external source of energy to elevate reservoir pressure to initial pressure condition. The secondary recovery mechanism is implemented after the primary recovery and involves either gas or water flooding to increase oil production. During this stage, the recovery factor from using waterflooding could achieve 35%-50% of OOIP (Tang, 1999). The last stage of oil recovery is identified as enhanced oil recovery (EOR), which involves different recovery approaches such as thermal, chemical, or miscible flooding to produce additional residual hydrocarbon fluids. EOR techniques may improve recovery

factor by up to 75% of OOIP by enhancing trapped fluid mobility through modified fluid properties (Dang et al., 2015).

Water flooding was implemented for the first time in 1924 in Pennsylvania's Bradford field (Austad, 2013), however, this secondary recovery mechanism has become widespread in conventional oil reservoirs due to its simplicity and reliability. Numerous studies have been conducted in order to optimize the process on compatibility between flooded water and existing reservoir/aquifer brine to avoid potential formation damage. Results reveal that flooded water composition plays a significant role in oil recovery. Many investigators including Jadhunandan and Morrow (1991, 1995), Tang and Morrow (1994, 1997) and Yildiz and Morrow (2006) studied the potential use of low salinity waterflooding (LSWF). Moreover, extensive experimental core flooding investigations were performed and showed the advantages of LSWF in improving oil recovery (Green and Willhite, 1998; Tang, 1999; Fjelde et al., 2012; Griffiths et al., 2015; Erke et al., 2016; Etemadi et al., 2017). The results generally showed that higher oil recoveries were obtained, in both secondary and tertiary stages, as flooded water salinity was less than reservoir brine and aquifer salinities. Studies on sandstone reservoirs showed that utilizing LSWF increased recovery factor by up to 40% compared to the standard high salinity waterflooding (HSWF) (Halvorsen, 2013). Thus, utilizing LSWF might lead to higher oil recovery compared with injecting chemicals such as alkaline or polymer or injecting steam.

Almeida et al. (2020) conducted core flooding experiments and investigated oil recovery mechanisms and pH effect in high and low salinity brine solutions utilizing clastic samples. They found that as the pH shifts towards alkaline conditions due to ionic exchange during LSW injection, it may approximate the isoelectric point (IEP) of pH-dependent surface charges in oil and rock minerals, weakening the electrostatic attraction between their surfaces leading to higher oil recovery. Snosy et al. (2020) conducted a comprehensive review on more than 500 core-flood published experiments to study the effects of changing water salinity on oil recovery. They found that there is an optimum composition and optimum salinity for waterflooding in secondary flooding stage. The controlling factor for tertiary flooding stage, however, may not be decreasing the salinity but modifying the salinity up and down might slightly increase oil recovery. Naeem and Dehaghani (2020) conducted experiments on capillary tubes and confirmed the role of the newly osmotic theory mechanism that leads to increase in oil recovery besides the main mechanism which is the wettability alteration towards water-wet conditions. The performance of oil, according to the newly added mechanism, as a semi-permeable membrane between connate and low-salinity waters. Water molecules move from low-salinity water through oil to connate water because of the difference in their osmotic pressure, which works as the driving force, until the system moves toward equilibrium in concentration. Thus, water molecules introduced into connate water causes the swelling of connate water, and eventually moves the oil towards pore throats. Similar to Snosy et al. (2020), Naeem and Dehaghani (2020) also reported an optimum value of low-salinity water concentration that led to the highest possible oil

recovery.

In recent years, two additional EOR techniques have been introduced; first carbonated water injection where CO₂-containing water is injected in order to decrease free CO₂ injection mobility, increase water viscosity, and store/remove produced greenhouse CO₂ gas safely. Second smart water injection where the ions in brine are adjusted to cause designed reactions with distributed ions on the rock surface to ultimately lead to additional hydrocarbon recovery. Soleimani et al. (2020) studied the combined effect of both techniques, smart carbonated water injection, as hybrid to find out the recovery factor change and effective mechanisms using experimental core flooding setup. Hybrid method was found to yield the highest recovery of 70% mostly due to wettability alteration, oil swelling, ion exchange and permeability enhancement.

Zaheri et al. (2020) studied, using flooding experiments on carbonate cores, the effect of salinity and different types of ionic contents, especially calcium ions contained in formation water and sulfate ions in injected fluid on oil recovery. They reported that reducing the salinity of injected water, which caused a decrease in ionic strength, may not lead to a greater oil recovery but higher calcium concentration in the formation water and sulfate ion concentration in the injected water alter rock wettability and therefrom affect oil recovery.

Wang et al. (2020) also conducted experimental and model studies on the effect of LSWF on the recovery of carbonate reservoirs. They reported LSWF increased recovery rate by 9% compared with the formation water flooding (FWF) as the wettability of rock surface, in the low salinity conditions, changed by the chemical reaction of divalent ions (Ca²⁺, Mg²⁺, SO₄²⁻), which showed more hydrophilic. Chávez-Miyauch et al. (2020) performed waterflooding experiments on Berea sandstone cores and reported no recovery increase in tertiary flooding mode but some increase in recovery from LSW injection in secondary mode.

The earliest attempt of LSWF modeling was conducted in 2008 by Jerauld et al. (2013) using a modified model of Buckley and Leverett. Both capillary pressure and relative permeability in the model were set as a function of salinity of flooded brine. The model also took into account the relative permeability effect of the secondary drainage water between imbibition and connate water. Wu and Bai (2009) presented a similar fractured sandstone mathematical and numerical model of LSWF using Multiphase Subsurface Flow (MSFLOW) general simulator. Obtained oil recovery results were in good agreement with experimental results. Furthermore, Sorbie and Collins (2010) developed a semi-quantitative model based on theoretical consideration of pore-scale that described LSWF multicomponent ion exchange mechanism. The model included the effect of polar organic species adsorption and electrical double layer expansion on the changes of wettability. However, there was a mismatch between predicted and experimental results. Omekeh et al. (2008) developed a mathematical model that took into account the effect of two-phase (oil-brine) flow, ion exchange, dissolution and precipitation of mineral that occurs during LSWF. Results showed that the significant impact of LSWF can be reduced by the presence of carbonate minerals, hence, improving oil recovery. Cations were effec-

Table 1. A summary of LSWF numerical modeling studies.

Investigator(s)	Numerical model	Research scope	Observation/Results
Jerauld et al. (2006)	Buckley and Leveret	Effect of water salinity injection on capillary pressure and relative permeability	<p>A model of wettability change with relative permeability can be used to describe LSWF.</p> <p>A higher recovery at breakthrough is not evidence that connate-water banking is not occurring.</p> <p>Connate water banking is important in interpreting laboratory relative permeability curves.</p> <p>Both at the laboratory and field scales, mixing is important and influences the interpretation and predictions of flood performance.</p> <p>The model is capable of producing the slug-size dependence observed in single well tracer tests as well as the behavior of coreflooding.</p>
Rueslatten et al. (1994)	PHREEQC geochemical	Change reservoir pH during LSWF	<p>High salt content in the injected water would lead crude oil to react with clay particles to form an organometallic complex, which would lead to the partial lipophilicity of the clay mineral surface.</p>
Wu and Bai (2009)	Buckley and Leveret	Correlation between injection salinity concentration and wettability	<p>The main mechanisms of IOR by low-salinity water injection were described by incorporating salinity-dependent changes in relative permeability, capillary pressure, and residual oil saturation in the model formulation.</p> <p>A model provides a general capability for quantitative evaluation of LSWF.</p>
Sorbie and Collins (2010)	PHREEQC geochemical	Investigate multicomponent ion exchange process at a pore scale	<p>Changing of divalent cation concentrations (Ca^{2+} and Mg^{2+}) leads to the development of a "self freshening" zone within the water-flooded region.</p> <p>A change in contact angle of only 10° can easily lead to a significant % incremental of oil recovery in the range 16%-60% which indicates that this is a very sensitive parameter.</p> <p>Expansion of the electrical double layers and changes in the adsorption of polar organic species resulting changes in wetting.</p>
Omekeh et al. (2012)	Mathematical and salt reaction	Numerical study of mineral solubility and ion exchange in LSWF	<p>Ion composition of the low salinity brine changes due to interaction with the reservoir rocks.</p> <p>pH increases during the interaction between low-salinity brine and reservoir rocks.</p> <p>Dissolution of carbonate minerals can occur and alter composition of the injected low-salinity brine, the concentration of divalent ions on the rock surface and hence the potential of low salinity to improve recovery.</p>
Kazemi et al. (2013)	IPHREEQC geochemical module coupled with UTCH-EM chemical-flooding reservoir simulator	Mechanism and effect of multi-component ion exchange multi-component ion exchange	<p>Explains capability of the UTCHEM-IPHREEQC for mechanistic modeling of low salinity water injection, this integrated tool can be applied to many processes such as injection of alkali and surfactant flooding for wettability alteration in carbonate reservoirs.</p>
Dang (2015)	PHREEQC geochemistry coupled with CMG's GEM	Optimize influence of clays and ion exchange process on LSWF	<p>A wide range of recovery factors from 19% to 40% indicate that the effectiveness of LSWF strongly depends on geological factors such as facies properties, clay distribution and clay proportion.</p> <p>Secondary and tertiary LSWF give about 6% and 4.1% incremental OOIP over high salinity water-flooding, respectively.</p>
Koleini et al. (2018)	Molecular dynamics simulations	Studying the characteristics of the calcite-water interface at atomic level in the presence of different ions in brine	<p>Low salinity water forms an electric double layer at the interface of calcite/low salinity water while the ions in the high saline water form several aggregates of ions that persist inside the nano-layer brine separating oil film from carbonate surface and act as anchors that hold oil components in the vicinity of the substrate. LSW breaks the ionic aggregates and release oil components in favor of more water-wet state leading to an increase in oil recovery.</p>
Tale et al. (2020)	A geochemical package (PHREEQC) used to simulate geochemical reactions	Studying brine/rock interactions for thin sections of calcite and dolomite	<p>Low salinity water increased pH of reached brine for calcite and dolomite leading to ion exchange and surface reactions and ultimately alteration of rock surface due to brine-rock interactions.</p> <p>Precipitation and dissolution decrease with increasing water dilution.</p>
Koleini et al. (2019)	Molecular dynamics simulations	Studying the thin brine film characteristics that wets carbonate reservoir rocks at molecular level	<p>Electric double layer is formed at the interface of calcite/low salinity water, the ions in the high saline water form several aggregates of ions that persist inside the nano-layer brine separating oil film from carbonate surface. Thus, LSWF breaks ionic aggregates and release oil leading to higher recovery.</p>

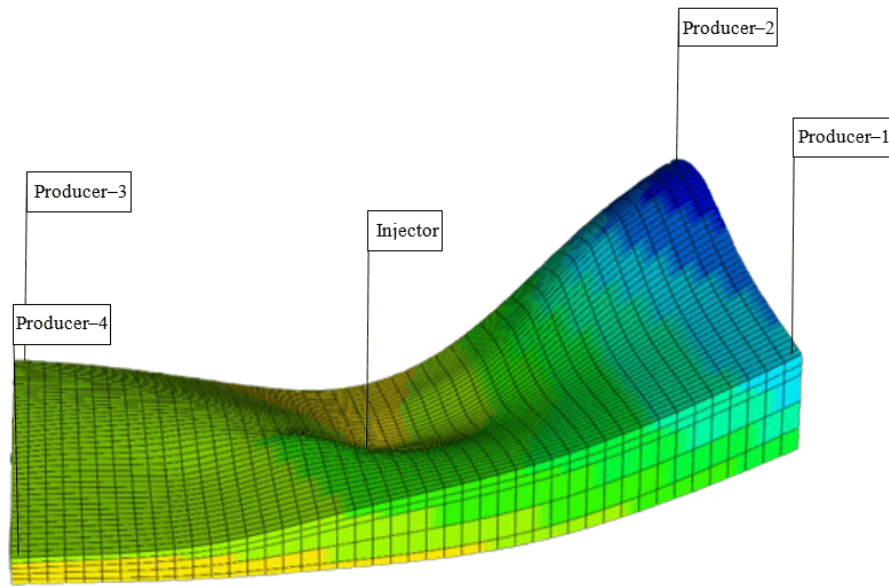


Fig. 1. Developed 3D field scale reservoir model.

Table 2. Description of 3D model volume and geometrical.

Geometry	Value
Total reservoir area (x-y direction)	$2.42 \times 10^7 \text{ ft}^2$
Total bulk volume of reservoir	$9.01 \times 10^8 \text{ ft}^3$
Total pore volume of reservoir	$1.98 \times 10^8 \text{ ft}^3$
Original oil initially in place (OIIP)	$1.48 \times 10^8 \text{ ft}^3$

tively involved in ion exchange process by exchanging negative ions charged clay surface and releasing cations from rock surface which increased oil mobility and relative permeability. Divalent ions desorption was found the main factor in LSWF process as Suijkerbuijk et al. (2012) found divalent ions adsorption taking place on clay minerals favorably altered rock wettability towards a successful LSWF.

Dang et al. (2015) studied the potential use of LSWF to improve oil production by developing mechanistic model utilizing a Computer Modeling Group's GEM software. The developed model showed a significant effect of geological clay during the LSWF process on field scale at secondary and tertiary stages. The results showed a wide range of recovery factors varying from 19% to 40% that indicated the effectiveness of LSWF strongly governed by geological factors such as facies properties, clay distribution and proportion. It also identified that the wettability alteration was the dominant effect on oil recovery and contributed approximately 58% to 73% of production. Table 1 shows previous LSWF modeling studies.

The increasing application of LSWF in oil reservoirs has made it vital to determine the most effective optimization process by modifying operating conditions to comprehend a possible approach in maximizing oil recovery. As concluded by Dang et al. (2015) that LSWF performance is strongly

governed by reservoir geology, clay minerals, ion exchange and geological reactions. However, the effect of reservoir temperature on chemical ion exchange reactions has not been previously studied. A field scale reservoir model was developed to simulate the performance and optimization of LSWF process including the effect of temperature. .

2. Material and method

2.1 Development of reservoir numerical model

The reservoir model was developed using CMG-GEM based on an onshore oil field, located in Sirt Basin, Libya. The developed reservoir model consisted of 9,600 blocks and simulated inverted 5-spot injection configuration as seen in Fig. 1. Table 2 shows the model geometrical data and Table 3 indicates the key input parameters for the reservoir and LSWF.

Geochemical reactions: Reactions between multiphase multicomponent flow equations are expressed with following equations:

For hydrocarbon (oil and gas) phases, n_h :

$$\Psi_i \equiv \sum_{\alpha=o,g,w} \Delta T_{\alpha}^u y_{i\alpha}^u (\Delta P^{n+1} + \Delta P_{c\alpha}^u - \tilde{\rho}_{\alpha}^u g \Delta d) + \sum_{\beta=g,o,w} \Delta T_{\beta}^u y_{i\beta}^u (\Delta P^{n+1} - \tilde{\rho}_{\beta}^u g \Delta d) + \sum_{q=g,w} \Delta D_{iq}^u \Delta y_{iq}^u + V \sigma_{i,ag}^{n+1} + q_i^{n+1} - \frac{V}{\Delta t} (N_i^{n+1} - N_i^n) = 0 \quad i = 1, \dots, n_h \quad (1)$$

For aqueous phase, n_a :

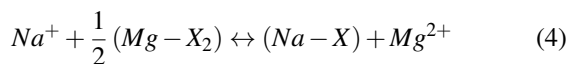
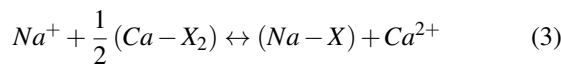
$$\Psi_j \equiv \Delta T_w^n y_j^n (\Delta P^{n+1} - \tilde{\rho}_w^n g \Delta d) + \Delta D_{iw}^u \Delta y_{iw}^u + V \sigma_{j,aq}^{n+1} + V \sigma_{j,mn}^{n+1} + q_j^{n+1} - \frac{V}{\Delta t} (N_{j,aq}^{n+1} - N_{j,aq}^n) = 0 \quad j = 1, \dots, n_a \quad (2)$$

Table 3. Input parameters of developed field scale model.

	Layer	Porosity (%)		Horizontal permeability (mD)			Oil saturation (%)		Net-to-gross ratio (%)		Water saturation (%)	
Reservoir properties	1	14		17			65		50		35	
	2	19		58			69		32		31	
	3	16		30			67		38		33	
	4	18		286			83		96		17	
	5	17		74			70		72		30	
	6	16		64			67		65		33	
Oil phase components	Component	N ₂	CO ₂	CH ₄	C ₂ H ₆	C ₃ H ₈	IC ₄	nC ₄	IC ₅	nC ₅	C ₆ H ₁₂	C ₇ ⁺
	Mole fraction (%)	0.39	0.44	8.48	2.47	4.42	1.14	3.44	2.13	2.99	5.67	68.43
Well properties	Well name			Maximum production/injection rate (bbl/d)				Minimum bottom hole pressure (psi)				
	Producer-1			1000				2000				
	Producer-2			1000				2000				
	Producer-3			1000				2000				
	Producer-4			1000				2000				
	Injector			4000				6500				
Aquifer information	Property	Thickness (ft)		Porosity (%)			Permeability (mD)		Compressibility (psi ⁻¹)			
	Value	200		17			80		5 × 10 ⁻⁶			
Aqueous phase components	Type of water	Component			H ⁺	Ca ²⁺	Mg ²⁺	Na ⁺	Cl ⁻	Total salinity (ppm)		
	Formation water	Salinity			0.005	18492	2320	68520	150060	240000		
	Injection seawater	Salinity			0.00	1550	1540	13200	23400	40000		
	Injection low saline brine	Salinity			9 × 10 ⁻¹⁰	26	77	660	1170	1873		

The superscript n and $n + 1$ represents the old and new time levels. The equations are discretized in an adaptive implicit manner. The term $V\sigma_{i,aq}^{n+1}$ and $V\sigma_{k,mn}^{n+1}$ refers to mineral dissolution/precipitation and the intra-aqueous reaction rates.

Wettability variation: The variation of wettability and multiple ion exchange during the process of LSWF were considered as the main mechanism of an extra oil recovery. Two typical reactions of ion exchange that are involved in LSWF can be written as:



Ion exchange: The multiple ion exchanges between ions in an aqueous phase and clay minerals were modeled based on the chemical equilibrium equation. The equation of ion exchange with an exchanger X in the aqueous phase can be written as:

$$\Delta T_{aq}^u y_{i,aq}^u (\Delta p^{n+1} - \tilde{\rho}_{aq}^u g \Delta \Delta) + \Delta D_{i,aq}^u \Delta_{i,aq}^u + V \sigma_{i,aq}^{n+1} + V \sigma_{i,mn}^{n+1} + q_i^{n+1} - \frac{V}{\Delta t} \left[(N_{i,aq}^{n+1} + N_{i-X}^{n+1}) - (N_{i,aq}^n + N_{i-X}^n) \right] = 0 \quad (5)$$

CO₂ presence in the oleic phase can give an initial pH value of 5.2 to connate water which presents an acidic condition. Formation water consisted of five components yielding a

salinity of 240,000 ppm considering that typical sandstone formation water salinity ranges from 50,000 to 250,000 ppm (Austad, 2013). Salinity of injected brine was set at a low default value of 1,873 ppm based on previous LSWF studies that suggested a range from 1,000 to 2,000 ppm (Esene et al., 2018).

At each corner edge of the reservoir model, a vertical producer well was introduced. At the center of the reservoir grid, a vertical injector well was positioned. This pattern was used to simulate an inverted five spot injection. Production wells were perforated from layer 1 to 5 but the vertical injector well was perforated for all 6 layers in order to achieve maximum efficiency of water flooding. Table 3 summaries reservoir and well properties, and phase component used in building the model. The reservoir's initial condition was set using the datum depth pressure and PVT data so fluid saturation in each grid block was determined according to the capillary pressure.

2.2 Boundary condition

The reservoir model was set to no-flow boundary condition. However, as oil expansion was weak, oil was energized by strong aquifer drive. To simulate this condition, an aquifer layer was implemented below the bottom layer of the reservoir. Aquifer modeling was completed using a steady-state aquifer model that assumes no change in pressure at the aquifer external boundary. The water influx rate is determined by the

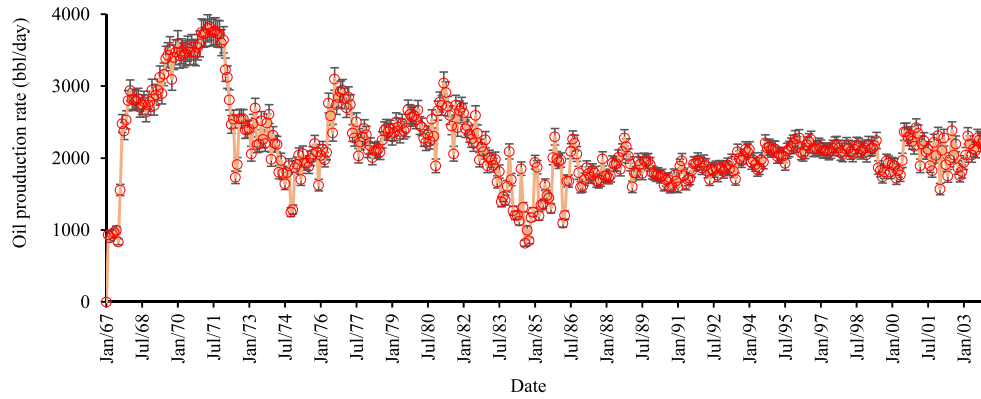


Fig. 2. Validation of field oil production rate.

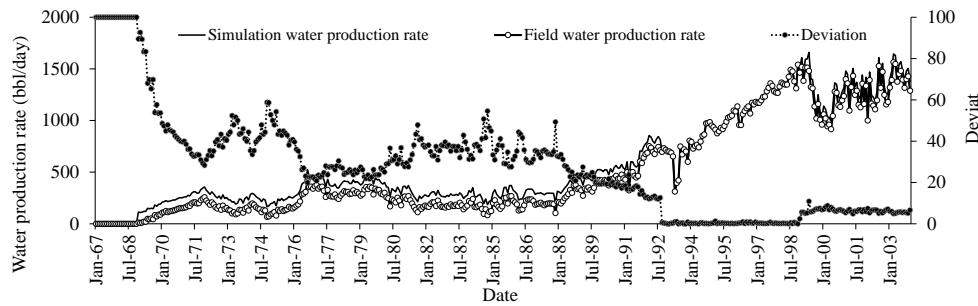


Fig. 3. Validation of field water production rate.

following equation:

$$Q_A = C_A (p_A^i - \bar{p}) \quad (6)$$

2.3 Volume constraint

GEM, as a compositional simulator, uses volume constraint equation as indicated by Eq. (7) which includes parts of Eqs. (3)-(5). Based on Newton's method, these equations are solved simultaneously in the simulator.

$$\Psi_p \equiv \Sigma_q \left(\frac{N_q^{n+1}}{\rho_q^{n+1}} \right) - \Phi^{n+1} = 0 \quad q = o, w \quad (7)$$

3. Results and discussion

3.1 Model validation

The developed model was made to produce naturally for the purpose of history matching. Field oil production began naturally, without any external energy supply, from January 1967 to end of December 2003. In order to ensure that the simulated model has the same initial condition, the reservoir model was naturally depleted before applying water flooding. Maximum production rate and minimum wellbore pressure of production wells were set at 1,000 bbl/d and 2,000 psi per well, respectively. Actual field production data were used to endorse the field liquid production rate of oil and water. Adjusting water relative permeability and aquifer thickness

were modified to match field production. The model predicted results were compared with field data for the period from Jan-1967 to Jan-2003 as shown in Figs. 2 and 3. Fig. 2 shows a very good match for oil production between simulator results and field data giving support to implementing reservoir properties. Fig. 3 shows comparison between predicted and field water cut values. The figure shows some discrepancy between 30%-50% till 1990, after that a small difference ($\leq 10\%$) was observed in water production till the end period in 2003. This further confirms that modified aquifer properties in CMG-GEM provides reliable history-matching and is capable of future predictions as well as field development. Fig. 4 shows reservoir pressure and oil recovery factor during primary production stage. Reservoir pressure declined by 24% of original pressure of 4,200 psi and oil recovery factor of around 10% was attained indicating that substantial amount of oil was still unrecovered.

3.2 Modeling waterflooding

Waterflooding simulation was initiated in January 2004 after the primary recovery that ended in December 2003. As shown in Fig. 1, a vertical injector well was positioned at the center of the developed model to simulate the inverted five-spot injection. Water was injected in the fifth layer to assist displacing the remaining oil towards production wells. Water injection was conducted distinctively through conventional high and low salinity of 40,000 and 1,873 ppm, respectively. High salinities brine was implemented, due to clay presence

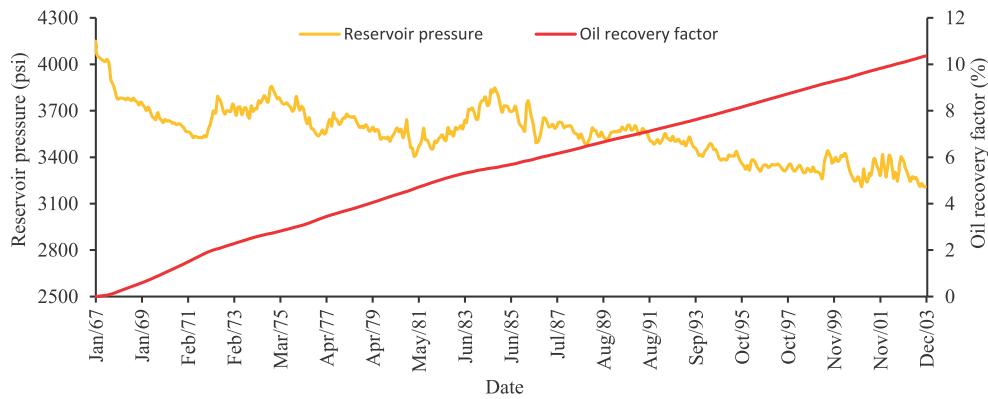


Fig. 4. Shows alteration reservoir pressure with oil recovery factor during primary recovery.

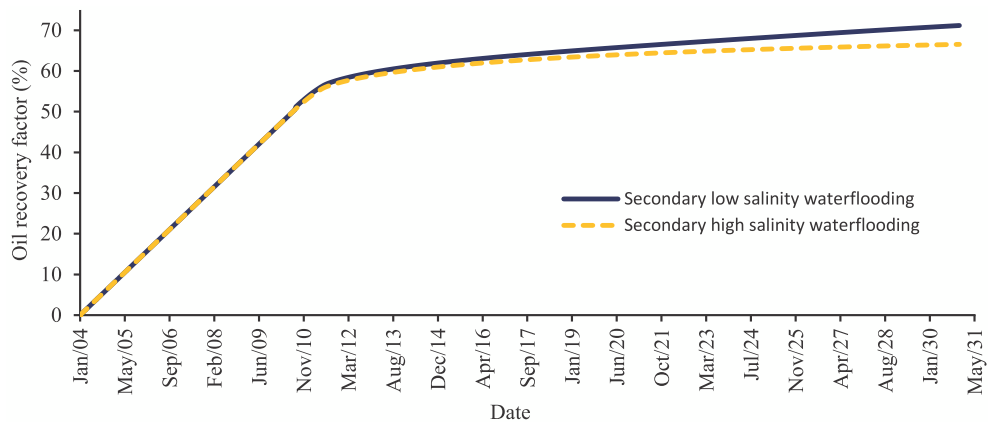


Fig. 5. Additional oil recovery at implementing low and high water salinity.

in the reservoir, in order to avoid detrimental effects on the reservoir production as a result of clay swelling (Turgazinov et al., 2018).

26-year forecast was made for oil production during secondary water flooding and at least one constraint was required by the injection well to run the simulation. Therefore, surface water injection rate was set at 6,000 bbl/d as a base case scenario. In order to prevent bottom hole pressure (BHP) increment higher than the initial reservoir pressure and possible formation damage, a maximum BHP of injection well was set at 4,200 psi. Fig. 5 shows an increase in oil recovery after implementing high and low salinity waterflooding.

Fig. 5 shows the effect of implementing low and high salinity waterflooding at the secondary stage, maintained production plateau and improved oil recovery from around 10% in December 2003 to 50% in November 2010. This outcome is consistent with Hu (2013) that indicated water injection into the reservoir provided support to reservoir pressure and improved fluid sweep efficiency.

3.3 Role of low salinity waterflooding

Based on simulation findings, the main mechanisms in LSWF that result in an increase in oil recovery are the integrated effects of higher sweep efficiency and favorable

alteration of reservoir wettability towards more water-wet by ion exchange. Vledder et al. (2010) reported similar findings; however, it is unclear if incremental oil from conventional HSW is a result of the same LSWF mechanisms. A few hypotheses were proposed in the literature, but there is still lack of understanding of the actual mechanism(s).

If the mechanisms of LSWF remains unclear, it would be difficult to implement, optimize and control the process of oil recovery. Thus, a comparative study between low and high salinity water was conducted to better understand the process of LSWF. The analysis was performed by evaluating the model cross-sectional area.

Figs. 6a and 6b show oil saturation remained, in the reservoir layer 3, after implementing LSWF and HSWF, respectively where oil saturation remained after implementing LSWF was less than that after implementing HSW and that oil saturation was significantly reduced around the injection well. Modeling was performed by using the ion exchange technique to observe the effect of wettability alteration. The amount of Na^+ ions that was still attached to clay was analyzed. Figs. 7a and 7b show ion exchange equivalent fraction of Na-X in the third layer cross the reservoir using LSWF and HSW, respectively. It was observed for LSWF case that there was significant reduction in Na^+ ions attached to clay surface. This was because of ion exchange as well as the clay's least affinity

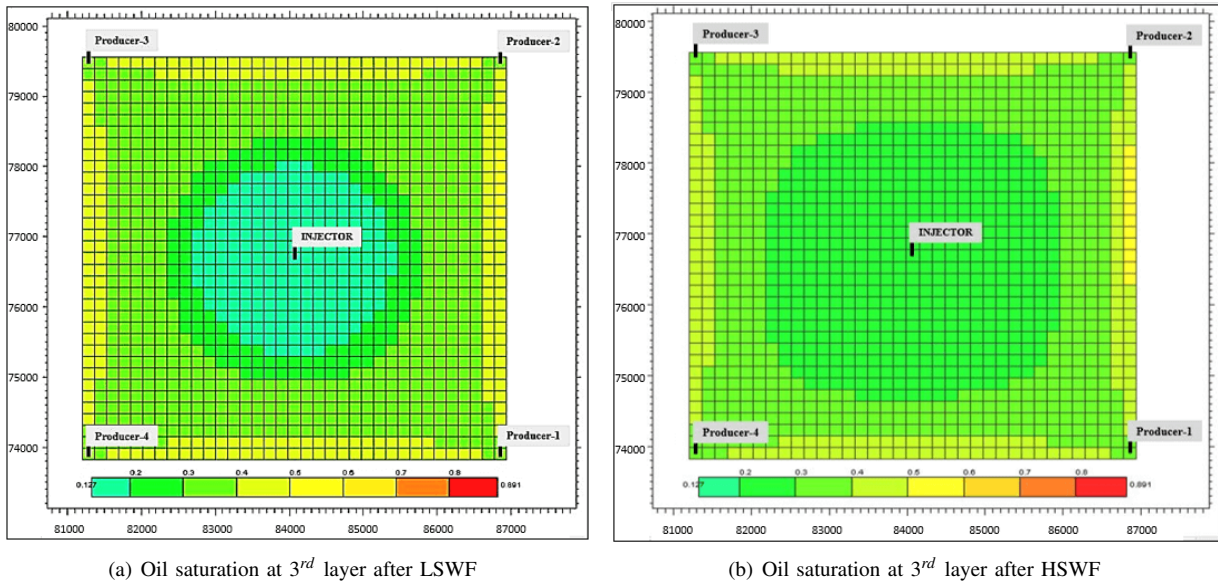


Fig. 6. Oil saturation after waterflooding.

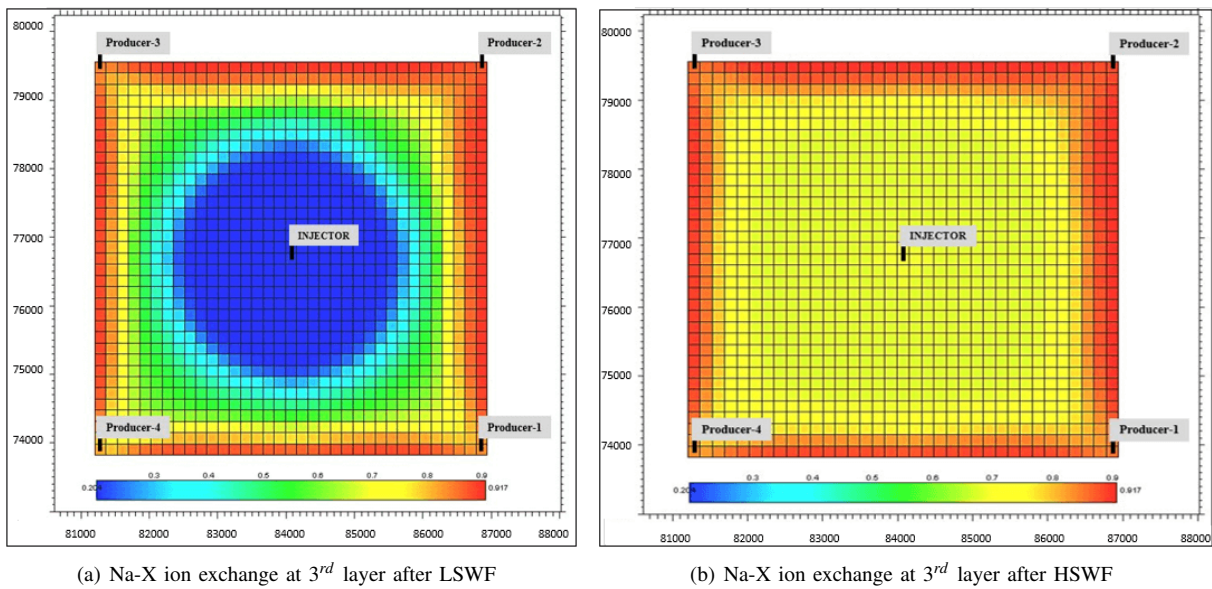


Fig. 7. Na-X ion exchange after waterflooding.

towards Na^+ . Similar observations were reported by Halvorsen (2013). Also Esene et al. (2018) noted that as the concentration of Na^+ in the injected brine was reduced, the brine-rock equilibrium was altered and Na^+ on the sandstone surface needed to be desorbed. Na^+ desorption from the sandstone surface led to an exchange with a divalent ion to attain a new ionic bridge equilibrium state. Thus, injecting low salinity water made favorable reservoir condition that promotes the process of ion exchange that takes place on the clay surface, hence, resulting in a higher recovery of oil as compared to the injection of high salinity brine. This observation is similar to the simulation study of LSWF field-scale that was performed by Dang et al. (2015) on Brugge field. They indicated that the key mechanism of using LSWF to alter the wettability

towards more water-wet condition is the multicomponent ion exchange. This mechanism caused the originally immovable oil to become mobile, hence, significantly reduced the residual oil saturation.

Clay mineral is the most governing factor that causes wetting condition in sandstone reservoirs as it is present in different brine and oil phases (Dang et al., 2015). Clay surface has the highest affinity towards an active oil polar component for a porous system so that initial oil wettability condition is caused by the adsorption process of crude oil polar components on clay surface (Strand et al., 2016). All reactive cations are adsorbed by negative charge clay surface along with other acidic and basic polar organic components. Because of its unique chemical structure, clay is an exchanger for cation.

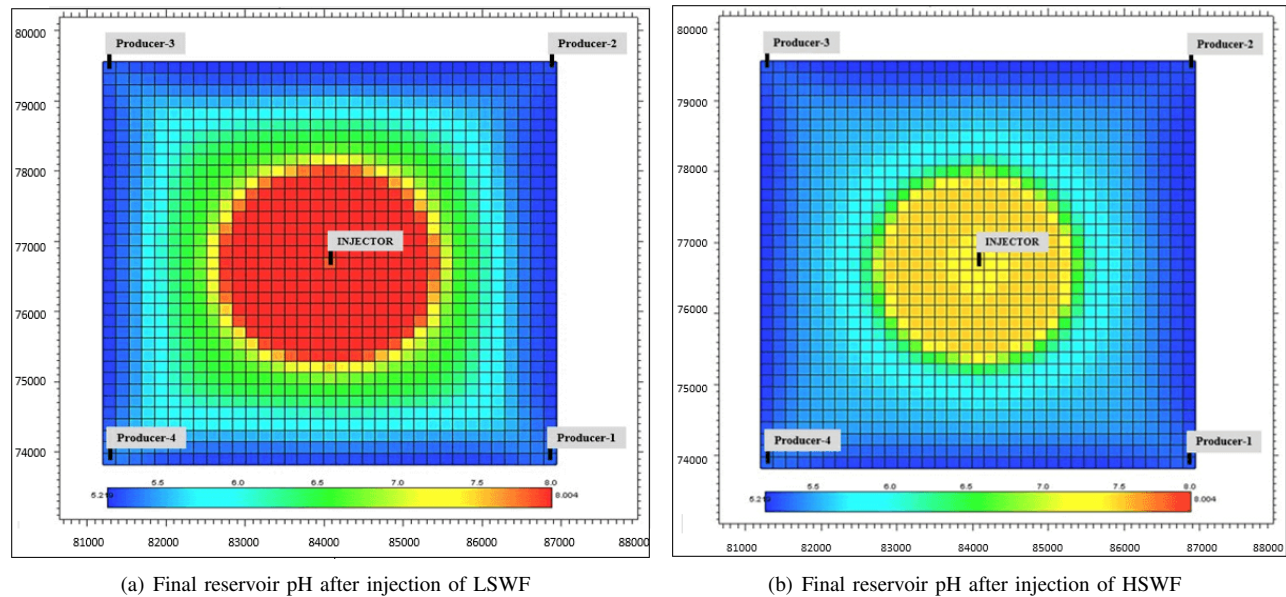


Fig. 8. Reservoir pH after waterflooding.

When cations are adsorbed, they tend to balance clay surface that was permanently negatively charged. All cations have different affinities towards clay surface. The order of affinity in an increasing manner is Na^+ , Mg^{2+} , Ca^{2+} and H^+ . Na^+ can be potentially bonded onto clay surface at the conditions of having high salinity formation water (Chakravarty et al., 2015) which is why we obtained lower oil recovery using HSWF than LSWF as depicted in Fig. 5. Vledder et al. (2010), Al-adasani et al. (2014), Dang et al. (2017) and Esene et al. (2018) indicated that as the chemical equilibrium of LSW changed, it caused an exchange in the low-affinity cation ions and changed with high-affinity cations. This may release organic components from clay surface and result in wettability alteration to a more favorable water-wetting condition.

Original field formation water pH was 5.22 and the presence of acidic CO_2 gas describes the initial reservoir acidic conditions. As LSW was injected, more reactive H^+ ions can be adsorbed through clay surface. This occurred because ion exchange led to increase in OH^- ions concentration, which further increased the aquifer pH (Mugele et al., 2018). Fig. 8a shows a similar observation as formation water pH increased from 5.22 to 8 after LSWF, while formation water pH decreased, as shown in Fig. 8b by magnitude of 0.5 when high salinity water was injected. Up to 10 pH increase has been reported (Chen et al., 2018), however, pH during LSWF in most cases was below 7, H^+ has a strong affinity towards the mineral surface. Therefore, Na^+ , K^+ , Ca^{2+} and Mg^{2+} were substituted by H^+ (Austad et al., 2010). However, pH value of 8 after LSWF was too low to produce soap. Chen et al. (2018) also noted that as pH was more than 7, the concentration of H^+ ion was dramatically declined and, as a result, less impact of pH on the exchangeable sites was observed. To produce an in-situ surfactant, pH should be higher than 9. Chemical reactions involved could lead to either an increase or decrease in the effluent formation water pH (Sheng, 2014). Therefore,

pH value alone should not be utilized to verify the LSWF mechanism of incremental oil recovery.

3.4 Response of tertiary low salinity waterflooding

To further study tertiary recovery using LSWF, sensitivity analysis on injection periods was performed after secondary seawater flooding for 5, 10 and 15 years and simulation forecast was made until year 2030. Fig. 9 compares different injection periods of brine with low salinity and showed that the secondary LSWF yields the highest oil recovery while subsequent injections of LSW yields less additional oil recovery. This is because reservoirs may induce low-resistance water channels bypassing future injected water during oil production (Derakh et al., 2011). Meanwhile, high-salinity brine injection compresses the ionic double layer and increases the clay-to-clay attraction, hence, causing the oil layer becoming strongly attracted by rock surface (Griffiths et al., 2015). Koleini et al. (2019) indicated that as an electric double layer is formed at the interface of formation/LSWF, the ions in HSW generated some aggregates of ions. These aggregates remained inside the brine nano-layer the nano-layer brine splitting oil film from carbonate surface and act as anchors that hold oil components

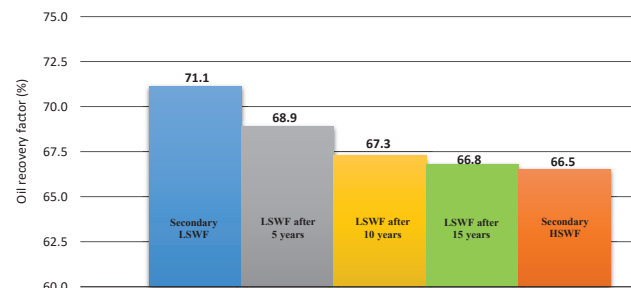


Fig. 9. Comparison of the oil recovery factor after different LSW injection periods.

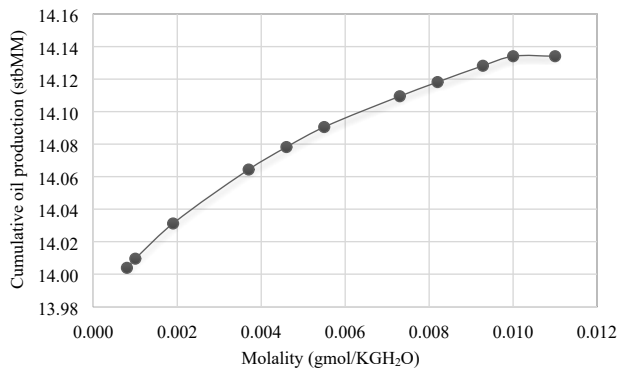


Fig. 10. Effect of Ca^{2+} concentration in LSWF.

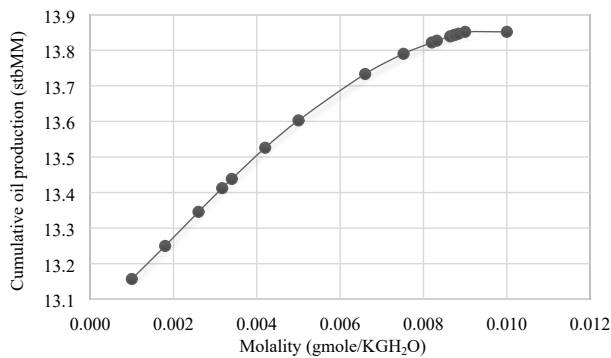


Fig. 11. Effect of Mg^{2+} concentration in LSWF.

in the substrate vicinity.

Thus, the advantages of implementing tertiary LSWF can significantly modify rock wettability when the injected brine is too dilute or ions chemical equilibrium is altered. Moreover, water injection period is one of the essential criteria. LSW is more efficient if it is implemented at the first stage of secondary recovery compared to second or third stage of LSWF.

3.5 Low salinity waterflooding optimization

3.5.1 Composition of injected low-salinity brine

Multiple ions present in low salinity brine can be classified based on the charge ions that they possess, as anion, monovalent cation and divalent cation. Injected brine in sandstone reservoirs usually contains a monovalent cation Sodium (Na^+), divalent cations Calcium (Ca^{2+}) and Magnesium (Mg^{2+}) and an anion Chloride (Cl^-) ions. The present study utilized the default composition of brine with low salinity and ion concentration was changed over a range of values, while ion concentrations were maintained to determine their sole effect on oil production.

Fig. 10 and Fig. 11 show increasing Ca^{2+} and Mg^{2+} ions concentration yields higher cumulative oil production then production remained constant as it reached an optimum concentration of Ca^{2+} or Mg^{2+} of 0.01 and 0.009 gmole/KGH₂O, respectively. The same outcome was reported after applying

LSWF on an Alaskan sandstone formation by modifying Ca^{2+} concentration from 70 to 100 ppm led to an additional 10% of oil recovery (Webb et al., 2003; McGuire et al., 2005; Webb et al., 2005a, 2005b; Griffiths et al., 2015; Wang et al., 2020). Adding divalent cations caused a 20% increase in cumulative oil production than without adding Ca^{2+} or Mg^{2+} in injected brine (Etemadi et al., 2017). Bedrikovetsky et al. (2011) also reported the same observation indicating an inappropriate design of divalent cation in low salinity brine suppresses oil recovery because reverse ion exchange maybe promoted. Meanwhile, increasing Na^+ concentration in injected brine led to decrease in cumulative production as shown in Fig. 12. This observation is in agreement with LSW coreflooding and field scale tests (Al-adasani et al., 2012; Fjelde et al., 2012; Al-adasani et al., 2014; Shehata et al., 2014; Erke et al., 2016; Qiao et al., 2016; Esene et al., 2018). Decreasing Na^+ ion concentration in injected brine led to change in brine-rock equilibrium and Na^+ ion on the rock surface needs to be desorbed by releasing the formation Na^+ to balance the equilibrium state. This desorption of Na^+ ion from sandstone surface promotes replacement by divalent ions to achieve new ionic bridge equilibrium state. Such phenomenon induces that polar components of oil which were initially attached to divalent ions that were released by brine-rock interaction (Esene et al., 2018). While increasing Na^+ concentration enhances ion adsorption on clay surface, this would displace the divalent cations and change rock wettability to more oil-wet condition, hence, decreasing oil production

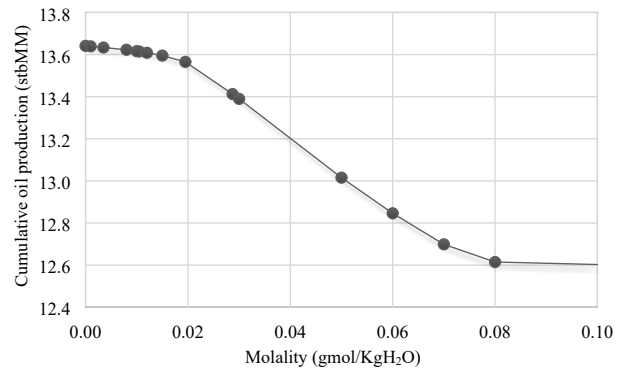


Fig. 12. Effect of Na^+ concentration in LSWF.

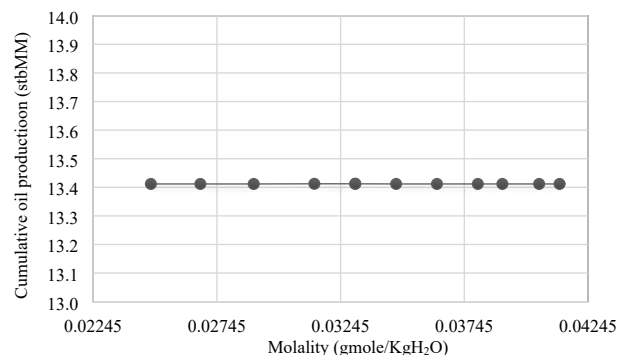


Fig. 13. Effect of Cl^- concentration in LSWF.

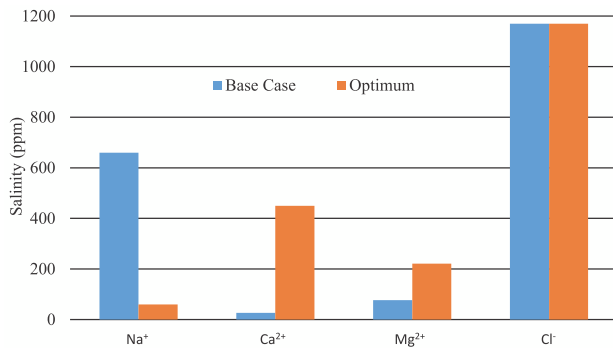


Fig. 14. Comparison of ion concentration of base and optimum salinity.

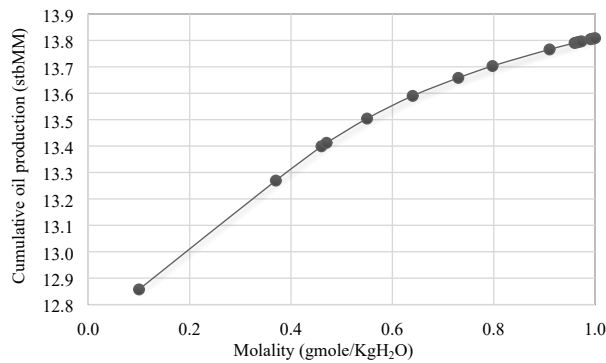


Fig. 15. Effect of aquifer Ca²⁺ ion in LSWF.

(Naem et al., 2020). Moreover, changing Cl⁻ ion concentration does not improve cumulative oil production as shown in Fig. 13.

Fig. 14 shows the most optimal ion composition of low salinity injected brine. A comparison against the initial base composition was made. The final total brine salinity with an optimum concentration of ions was 1,900 ppm, which is higher than the initial prediction of the base case composition. The prediction was made according to previous studies (Zhang et al., 2007; Patil et al., 2008; RezaeiDous et al., 2009; Singhal and Ashok, 2009; Al-adasani et al., 2012; Fjelde et al., 2012; Al-adasani et al., 2014; Erke et al., 2016; Qiao et al., 2016; Snosy et al., 2020). Although typical records of low salinity brine in core flooding and field tests range between 1,000 to 2,000 ppm, few cases reported that the optimum low salinity brine that achieved maximum oil recovery was outside this range. For instance, the optimum brine salinity of Berea sandstone formation was found 10,000 ppm, resulting in 30% increase in oil recovery (Agbalaka et al., 2009). It was also noticed that the optimum salinity might range for sandstone formations; a reduction in low water salinity of up to 100% to provide salinities as low as 100 to 2,000 ppm was possible (Tong et al., 2003; Hassenkam et al., 2012). RezaeiDoust et al. (2009) concluded that salt alteration played a significant role in explaining low salinity water effect. There is a critical ionic strength increasing the solubility of organic material, and the value usually is in the range of 2,000 to 3,000 ppm.

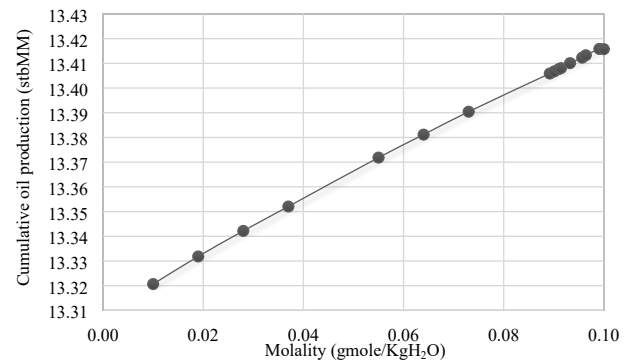


Fig. 16. Effect of aquifer Mg²⁺ ion in LSWF.

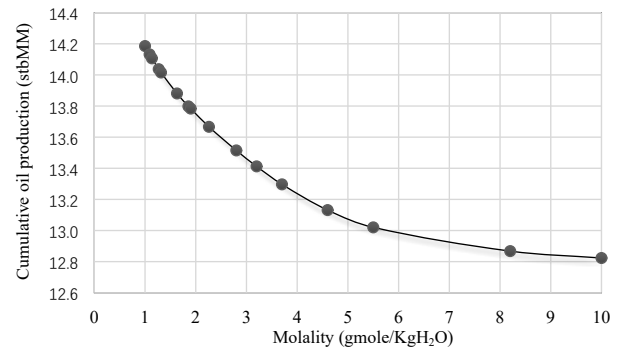


Fig. 17. Effect of aquifer Na⁺ ion in LSWF.

3.5.2 Effect of aquifer composition

Based on the sensitivity study and analysis on low salinity injection brine composition, it was found that the optimal composition of brine results in maximum oil recovery but it differs according to different reservoir scenarios. Initial fluid properties of the reservoir, such as connate water composition, are responsible in determining the success rate of LSW since the process is governed by ions in the brine. Coreflooding and field tests previously conducted on LSW (Al-adasani et al., 2012; Fjelde et al., 2012; Al-adasani et al., 2014; Erke et al., 2016) indicated that the composition of aquifer affected LSW process. Therefore, the effect of different formation water compositions on oil production was investigated in the present study. Connate water chemical composition research (Shehata, 2014) indicated that the presence of an adequate amount of divalent cations in formation water was important as primary requirement in maximizing oil production using LSW. In the absence of Ca²⁺ ion in the aquifer, no increase in oil recovery is obtained. Furthermore, our simulation results show that as the concentrations of Ca²⁺ and Mg²⁺ in the aquifer increase, the cumulative oil production also increases as shown in Figs. 15 and 16.

In contrast to the divalent cations, increase of monovalent Na⁺ results in decrease in cumulative oil production as shown in Fig. 17. As Na⁺ concentration increases, an adverse effect on oil production was observed because of unfavourable alterations of wettability. Adding excessive quantity of Na⁺

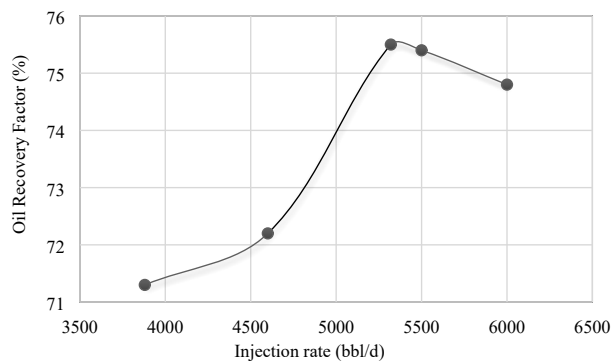


Fig. 18. Effect of different brine injection rates in LSWF.

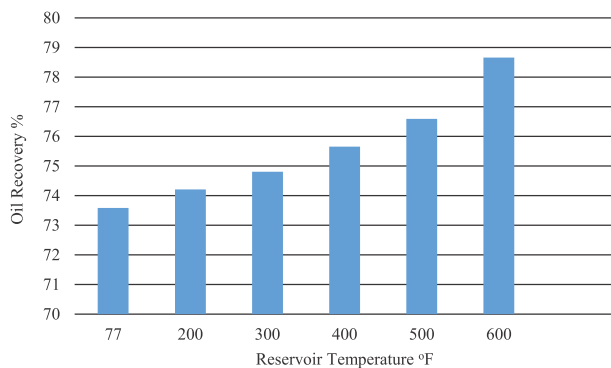


Fig. 19. Effect reservoir temperature in LSWF.

promotes the desorption of divalent cations from the clay surface, hence, changing the wettability to an oil-wet and causing decrease in cumulative oil production (Wu and Bai, 2009).

3.5.3 Effect of low-salinity injection rate

Fig. 18 shows the optimum injection rate of low-salinity brine at 5,320 bbl/d. which is lower than the base rate of 6,000 bbl/d. However, this outcome refers to the alterations in reservoir pressure and sweep efficiency during water flooding. Increasing brine injection rate causes an increase in reservoir pressure. However, high rate of injection is unnecessary because applying an injection rate of 5,320 bbl/d was enough to maintain reservoir pressure and improve oil production. During the forecast period, lower oil recovery was observed, as the rate of injection was higher than the optimal point. The effect of mobility ratio began to increase at higher rate of injection and mobile oil bypassed by water together with the initial increase in oil drainage. Thus, end point mobility ratio became higher than the one that implies an unstable flood front, hence, inducing a “viscous fingering” phenomenon and causing an inefficient oil displacement because of early breakthrough (Yildiz and Morrow, 1996). Consequently, a lesser oil recovery was obtained as the injected water flows ahead of the displacement fronts. This contrary effects of mobility ratio can be avoided at the optimal rate of injection as long as stable displacement front was well maintained proving

better sweep efficiency.

3.5.4 Effect of temperature

Temperature is another important factor that determines the success rate of LSWF procedure because it may change rock wettability. With the exception of Xie et al. (2017) and Aghaeifar et al. (2018), most studies (Tang and Morrow, 1997; Zhang and Morrow, 2006; Patil et al., 2008; Agbalaka et al., 2009; RezaeiDoust et al., 2009; Fjelde et al., 2012; Suijkerbuijk et al., 2012) investigated the effect of temperature on LSWF in sandstone reservoirs at different temperatures up to 200 °F. Alteration of rock wettability towards more water-wet occurred when a mechanism of multi-component ion exchange was prompted with chemical equilibrium at a low-salinity water injection. The rate of chemical reactions is theoretically higher at elevated temperatures. Thus, rapid ion exchange between clay surface and brine occurred at high temperature in order to achieve chemical equilibrium (Zhang and Morrow, 2006). Various runs of simulation were performed through increasing reservoir temperature from initial value of 200 °F to higher values. Simulation results shown in Fig. 19 were consistent with previous observations as small improvement in oil recovery is noticed at higher temperatures. At high temperature conditions, the reactivity of divalent cations surges because partial dehydration stimulated through the disruptions in water molecule structure (Benneker et al., 2018). Moreover, as temperature increases oil viscosity decreases and production rate increases. Apart from the hydration energies, reactivates and chemical equilibrium justification explained from the simulation above, as temperature increases oil viscosity decreases. Thus, reduced oil viscosity increases oil mobility in the reservoir and results in improved oil recovery which is the basis of thermal EOR.

4. Conclusions

Matching actual mature field oil and water production rates using CMG-GEM was successfully performed and the following conclusions can be made:

The main contribution of LSWF process is multi-component ion exchange, shown by fraction of ion exchange equivalent of Na-X, which is also a key parameter of improving oil production because it alters rock wettability from oil-wet to water-wet.

LSWF provided higher oil production than HSWF by an oil recovery up to 70%. Also implementing LSWF after primary stage production provided recovery gains up to 75%. Therefore, it is better to implement LSWF as a secondary oil recovery method.

Optimal injected ion concentrations for Ca^{2+} , Mg^{2+} and Na^+ are 450, 221, and 60 ppm, respectively.

High divalent cations and low monovalent cations concentration are recommended for injected brine and formation aquifer for beneficial alteration of wettability.

Reservoir temperature influenced the alteration of ion exchange during LSWF leading to wettability change and slight increase in oil recovery as temperature increased.

Nomenclature

p_{cwo} = Water-oil capillary pressure
 p_{cog} = Oil-gas capillary pressure
 p_{cwg} = Water-gas capillary pressure
 q = Rate of injection or production
 γ = Specific gravity
 V = Volume of grid block
 P = Pressure
 σ = Matric capillary pressure
 y = Mole fraction
 d = Hydrostatic fluid column
 $\bar{\rho}$ = Mass density of fluid
 ΔT = Transmissibility
 t = Time step
 D = Depth
 Q_A = Volumetric aquifer influx
 C_A = Total compressibility of the aquifer
 \bar{p} = Average pressure at aquifer-reservoir interface
 p_A^i = Constant pressure at external boundary of aquifer
 ψ_p = Primary component
 N_q^{n+1} = Moles of component at a new time step
 ρ_q^{n+1} = Density of phase fluid at a new time step
 ϕ^{n+1} = Porosity at a new time step

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Conflict of interest

The authors declare no competing interest.

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