The effect of CO₂-enriched water salinity on enhancing oil recovery and its potential formation damage: an experimental study on shaly sandstone reservoirs

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Abstract

Many experimental investigations on carbonated water injection (CWI) have shown an increase in oil recovery which CWI is defined as the process of injecting CO₂-saturated water in oil reservoirs as a displacing fluid. In every enhanced oil recovery method, the potential formation damage of the injected fluid is considered. This is due to the fact that the injection of incompatible fluids often causes clay swelling and fines migration and thus impairs the formation permeability. Permeability reduction by clay particles mostly depends on its distribution which can be pore lining, pore bridging, dispersed or combination of these causing pore blocking or pore-throat diameter reduction. Besides, fine migration is considered as an important mechanism of recovery improvement during injection of low-salinity water in sandstone oil reservoirs. The present paper investigates the impact of injection of carbonated water and brines with the different salt concentrations on oil recovery and formation damage focusing on permeability variation. The investigation has been done on 12 relatively homogeneous clay-containing sandstone cores, while the compositions of the injection water were varied from 40,000 to 1000 ppm, at 176° F and 2000 psi. The amount of recovery improvement and permeability drop recorded in all tests and the fine effluent of two experiments were analysed using XRD, one for CWI and one for WF (water flooding). In all salinities, CWI has shown more oil recovery improvement than conventional water. CWI of 40,000 ppm showed the minimum permeability reduction of 6 percent, while the highest permeability was obtained by injection of water with 1000 ppm. Maximum ultimate oil recoveries of 61.2% and 42% were achieved by 1000 ppm both for CWI and WF, respectively. In comparison with brine injection, CWI resulted in more permeability drop in salinity above critical salt concentration (CSC), while below CSC, WF has caused more formation damage than CWI. Experimental results also showed that fine migration was the main reason behind formation damage. It was also revealed that permeability was significantly reduced due to fine production in the effluent.

Keywords EOR · CWI (Carbonated Water Injection) · Formation damage · Clay fine migration

Abbreviations

CWI	Carbonated water injection
WF	Water flooding
UOR	Ultimate oil recovery
IOIP	Initial oil in place
LSW	Low-salinity water
HPHT	High pressure, high temperature
EOR	Enhanced oil recovery

CSC	Critical salt concentration
CEC	Critical exchange capacity
FB	Formation brine
TDS	Total dissolved solid
XRD	X-ray diffraction

Introduction

In today's world, inexpensive production of oil and gas is decreasing, and the side expenditures of operations are rising. The majority of sandstone reservoirs are being depleted, and effective production has been decreased. As the demand for energy grows rapidly, the need for IOR and EOR becomes more vital. In order to maintain the reservoir pressure, secondary production methods such

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as gas/water injection have been applied. But even after implementing these methods, a significant part of IOIP (initial oil in place) remains in the underground, particularly in heavy oil reservoirs. In this regard, investigating an efficient EOR method is highly recommended. Nowadays, there is a heated controversy around potential concerns of global warming throughout the world. This issue has made the utilisation of greenhouse gases globally viral in EOR projects. Among these greenhouse gases, CO₂ has the advantages of the highest solubility in water which would increase water density and decrease the oil density. On the other hand, CO₂ usually is placed in the critical state in the most reservoirs condition, which brings in advantages of chemical stability, high volume and density compared to noncritical state (Egermann et al. 2005).

One of the most promising EOR methods in both secondary and tertiary flooding is CO₂-enriched water, also known as CWI. As a matter of fact, gas injection in the reservoir usually faces problems such as gas override, low sweep efficiency and supplying a high volume of injection fluid, which makes it a noneconomic method for most cases. For avoiding these problems, CO₂ can be dissolved in water at first and then injected into the reservoirs. In other words, CWI can be considered as an immiscible injection which includes the advantage of CO₂ diffusion from carbonated water to oil (with respect to the higher solubility of CO₂ in oil compared to water); subsequently, with more CW–oil contact time, the oil phase becomes lighter as the partitioning mechanism happens. To summarise, this method leads to higher oil recovery by swelling the oil phase, decreasing the oil viscosity and oil–water IFT, changing the wettability of reservoir rock toward more water-wet (Ahmadi 2016; Ali et al. xxxx; Foroozesh and Jamiolahmady 2016; Kumar and Mandal 2017; Mosavat and Torabi 2013; Riazi 2011; Seyyedi et al. 2015; Seyyedi and Sohrabi 2017; Singh 2014; Sohrabi et al. 2012).

Nearly most of the sandstone reservoirs contain some percentages of clay content categorised as swelling and nonswelling ones. Clay minerals usually cause drastic permeability drop because of their inclination to detach from rock surface and plug the pore throats. This susceptibility turns clay fine migration to be the primary mechanism of formation damage in sandstone reservoirs. According to the literature, the critical factors of clay fine migration are interstitial velocity, rock failure, clay mineralogy and type, the concentration of exchangeable cations, wettability alteration, change in pH and temperature (Zhou et al. 1996; Valdya and Fogler 1992; Tchistiakov 2000; Schembre et al. 2006; Musharova et al. 2012; Mungan 1965). It is also essential to mention that among the whole clay minerals, *kaolinite* and *illite* play an important role in this type of formation

damage (Musharova et al. 2012; Khilar and Fogler 1983; Zhou et al. 1997).

In the literature, most of the studies have been focused on the fundamental mechanisms of recovery improvement and CO₂–rock interaction during injection of carbonated water into the reservoirs (Sohrabi et al. 2012,2011,2015; Alizadeh et al. 2014; Fathollahi and Rostami 2015; Honarvar 2017; Kechut et al. 2010,2011; Kilybay et al. 2017; Riazi et al. 2011; Seyyedi et al. 2017; Shakiba et al. 2016), while the effect of CW on formation damage in porous media has been less discussed. Several investigations in the laboratory and field scale show the promising results of CWI on EOR. The K&S project is the first economic CWI project performed in Dewey-Bartlesville Field (started in 1958). The average oil recovery was reported to be 37% more than water injection. Besides, the duration of injection diminished from 15 to 5 years. The problems of corrosion and also emulsion formation also were solved (Hickok and Ramsay 1962). Holm et al. investigated the effect of CWI on oil recovery enhancement. They reported that CO₂ extracts oil in a more selective way from porous media and increases permeabilities of sandstone and dolomite. CWI boosts the oil recovery factor up to 60–80%, which is higher than its value during WF (30–40%) (Holm 1961). Ramsay et al. reported that injection of carbonated water in different sandstone formations increases the oil recovery significantly. They also claimed that CWI surpasses WF in lowering clay swelling (Ramsay and Small 1964).

CO₂ interaction with reservoir fluid and rock is a controversial topic. Siregar et al. studied carbonated water and also water alternating gas (WAG) injection followed by surfactant in different carbonate rocks. Although the displacement front was stable through all tests, they reported that the positive dynamic interaction between oil and CO₂ could reach a maximum value by optimising slug size, CO₂ volume and surfactant concentration (Siregar et al. 1999). Fisher et al. investigated sandstone–CO₂–brine interactions in a sandstone reservoir. The dissolution of plagioclase that was rich in calcium, potassium, feldspar and anhydrite and deposition of albite were observed (Fischer 2010). Ross et al. studied CO₂ injection and declared that CO₂ injection dissolves carbonate rock and increases the local permeability (Ross et al. 1981). Oomole et al. studied the interaction between dolomite and CO₂. They concluded that an increase in pressure drop leads to precipitation of carbonate minerals and permeability reduction (Oomole and Osoba 1983). Bowker et al. stated that permeability rise related to carbonate dissolution could be neutralised by the permeability reduction of fine migration and pore blockage, and thus, no significant change in permeability may be observed (Bowker and Shuler 1989). Kaszuba et al. investigated the effect of CO₂ interaction with brine and sandstone. The dissolution of *Microlite*, *biotite* and *quartz* was observed

(Kaszuba et al. 2003). Bertier et al. studied the interaction between sandstone, brine and CO₂. Although the dissolution rate of carbonates was intense primarily, it reached a constant value at the end (Bertier et al. 2006). Wigand et al. investigated sandstone samples (Wigand et al. 2008). Their results showed that carbonate cement would be dissolved during CO₂ injection. Zemke et al. studied CO₂ injection in a sandstone formation. After 15 months, minor permeability increase was observed (Zemke et al. 2010).

CO₂ sequestration in depleted reservoirs and aquifers is an environmentally hot topic. Egermann investigated the CO₂ injection flow rate in both sandstone and carbonate cores. They reported that high flow rate ends up in further dissolution. They also stated that salt concentration reduces water solubility and rock permeability (Egermann et al. 2005). Izgec et al. studied CO₂ sequestration and reached the point that the porosity and permeability profiles had a constant trend. They also reported that CO₂–rock retention time and surface contact area have more significant effect compared to flow rate (Izgec et al. 2005). Mohamed et al. investigated CO₂ sequestration effect on permeability in a sandstone aquifer. They reported that during gas–water alternative injection, calcium carbonate precipitation and fine migration lead to permeability reduction (Mohamed et al. 2012). The same conclusion has been reported by Krumhansl et al. in a depleted sandstone reservoir (Krumhansl et al. 2002).

In the past decade, the role of CWI in changing rock properties such as porosity and permeability has been inspected in both calcite and porous sandstone media. Filho et al. studied the effect of CO₂–water alternative injection

on the porosity and permeability reduction in limestone and dolomite porous media. The results showed that fine migration causes a reduction in porosity and permeability (De 2012). Nunez et al. investigated dolomite dissolution phenomena of CWI. They reported that porosity and permeability are increased in the near-wellbore region, but they have an opposite trend far from wellbore (Nunez et al. 2017). Ross et al. investigated CO₂–brine dissolution effect on sandstone cores. Results showed that CWI increases the permeability due to the dissolution of cores cement (Ross et al. 1982). Sayegh et al. studied the sandstone interaction with CWI in low pressure and temperature. They pointed out that fine migration causes permeability reduction, and CWI dissolves carbonate cement of the core (Sayegh et al. 1990). Seyyedi et al. studied heterogeneous sandstone cores in HPHT condition. They observed low intensity of mineral dissolution and 6 percent absolute permeability reduction in tertiary CWI (Seyyedi and Sohrabi 2017).

In this study, we attempt to investigate quantification of oil recovery improvement and formation damage during CWI at low salinities in a heavy oil sandstone reservoir containing a few percent of clay minerals. To achieve this target, a series of HPHT core flood experiments have been performed at a constant operating pressure and temperature of 2000 psi and 176 °F in dirty sandstone cores.

Experimental set-up and procedure

Core flood set-up. An experimental set-up was used to conduct high-pressure, high-temperature core flood experiments. The set-up of the core flood (Fig. 1) consists of four

Fig. 1 Schematic diagram of the core flood experimental set-up

accumulators, two pumps, two backpressure regulators and a core holder which is being held in a quite horizontal position in an oven at 176 °F. All accumulators are in the oven temperature. An HPLC pump injects the prepared fluids into the core with a constant flow rate. For applying the overburden pressure on the reservoir model, a hydraulic pump has been used, and the hydraulic fluid is being injected into the annulus space between the sleeve and the core holder. A backpressure regulator is provided to keep the overburden pressure at a constant value of 2200 psi. A Siemens differential pressure transmitter monitors the pressure difference between the two sides of the core holder. The reservoir pressure is applied by providing a backpressure regulator at the end of the core at 2000 psi. Effluents accumulate inside a measuring cylinder, and the dissolved CO₂ is set free by passing through the backpressure regulator. The maximum operating pressure and temperature of the apparatus are 5800 psi and 250 °F, respectively.

Fluids' properties. In this work, crude oil from one of the Iranian oil reservoirs was used. The oil had the API of 17.9 and density and viscosity of 0.917 gr/cm³ and 18.5 cp at reservoir temperature of 176 °F. Besides, the asphaltene content of the oil was measured to be 11.32 wt% (IP-143). The formation brine with the salinity of 75,000 ppm and density of 1.055 gr/cm³ is used to saturate the cores (Table 1). The injection fluid used in the experiments was made by dissolving different concentrations of NaCl, MgCl₂·6H₂O and CaCl₂·2H₂O (at the same ratio) in pure water. Table 2 shows the composition of the injected fluid

in each test in detail. For the tests CWI 40,000 to CWI 1000, CO₂ has been fully saturated in injected brine at experimental condition (2000 psi and 176 °F).

Core properties. The core samples used in the experiments were cut from a relatively homogeneous sandstone outcrop. For each test, a separate core is used. Absolute permeability of cores was measured by core flood apparatus using formation brine. The diameter of all cores is 1.5 inch with a length of 3.5 inch. Average permeability of core samples was determined to be 78.2 mD, and the range of cores porosity was around 11–13%. Mineral composition of the outcrop is provided in Table 3, which is not clean and contains approximately 13.3 wt % of unstable clay minerals in total.

Procedure. All cores were dried and weighted before saturation with formation brine in a vacuum condition. Then, the core was set in core holder, and the formation brine was injected into the core at a low flow rate (0.2 cc/min) until the core becomes fully saturated (15 bar). The porosity of the core was measured by the weight difference between the saturated core and dry core. Then, permeability was calculated by injecting different rates of formation brine in line with Darcy equation. To avoid the non-Darcy condition, we examined several flow rates of 0.1, 0.2, 0.3 and 0.5 cc/min. After permeability calculation, the dead oil was injected into the core with the flow rate of 0.2 cc/min until the water was observed in the effluent fluid. At this step, backpressure was set at 2000 psi, and the fluid was kept injecting until no water came out of the model. Then, the amounts of IOIP and irreducible water of the core were calculated by measuring the volume of the water produced. The injection fluid (brine water or carbonated water) was provided to conduct into the core at a constant flow rate of 0.3 cc/min, the temperature of 176 °F, backpressure of 2000 psi and overburden pressure of 2200 psi in all cases. Each flooding test was performed by

Table 1 Formation brine composition (gr/litre water)

NaCl	KCl	MgCl ₂ ·6H ₂ O	CaCl ₂ ·2H ₂ O
36.81	0.31	4.48	33.40

Table 2 Compositions of injected fluid with different salinities (ppm)

Test	Salinity	NaCl	MgCl ₂ ·6H ₂ O	CaCl ₂ ·2H ₂ O	CO ₂ saturated level (%)
CWI 40,000	40,000	26,000	10,000	4000	100
WF 40,000					0
CWI 20,000	20,000	13,000	5000	2000	100
WF 20,000					0
CWI 10,000	10,000	6500	2500	1000	100
WF 10,000					0
CWI 7000	7000	4550	1750	700	100
WF 7000					0
CWI 5000	5000	3250	1250	500	100
WF 5000					0
CWI 1000	1000	650	250	100	100
WF 1000					0

Table 3 Sandstone core composition

Component	Wt%
Quartz	76.4
Kaolinite	5.6
Montmorillonite	2.2
Chlorite	3.6
Illite	1.9
Feldspar	6.4
Siderite	1
Calcite	2.1
Dolomite	0.8

Fig. 2 Ultimate oil recovery of six water flood experiments with respect to various salinities

10 pore volume (PV), as the slow process of formation damage occurs. During the process, ultimate oil recovery was measured, and for investigating the effect of CWI on porous media, the effluent samples simultaneously were collected. After collecting the effluent, the samples were centrifuged through 5000 rpm for 20 min (Mini Centrifuge Digital RST-1), and the particles were extracted from effluent by passing through a filter paper (No. 400 mesh). XRD analysis was taken from solid particles to determine the ingredients of fine particles. The residual oil in the cores was completely washed out by using Soxhlet extraction method. In this method, for measuring absolute permeability after each test, the sandstone core was cleaned up by toluene steam for several days. It is assumed that the washing method does not affect the rock surface and its permeability. Afterwards, absolute permeability has been measured with permeability apparatus (Gas Permeameter GP-R30) by injecting N₂ into the core and considering Klinkenberg effect at four different reversal mean pressures of 0.2, 0.4, 0.6 and 0.8 (1/psi).

Results and discussion

WF. As mentioned above, there is some percent of clay in our sandstone cores which are both swelling (*smectite*, *montmorillonite* and *mixed-layer illites*) and nonswelling clays (*kaolinite*, *illite*). Among these clay minerals, the swelling capacity of *montmorillonite* is extremely higher than *illite* and *chlorite* (Tchistiakov 2000). Clay swelling reduces porosity and permeability by moving off from the porous media surface and plugging pore throats in the next stage, which can often take place through salinity fluctuations. However, the water-sensitive behaviour of clay sandstones is mostly due to the fines migration rather than swelling of clays (Jones 1964). To have a yardstick, we carried out several low-salinity water injections into the cores to evaluate the amount of ultimate oil recovery improvement in each salinity in the absence of dissolved carbon dioxide. In this regard, we tried salinity concentrations below 40,000 ppm. Having indicated in the experimental section, all of the tests were performed at a constant flow rate. Thus, the hydrodynamic repulsive force term is equal for all different tests (DLVO Theory). Figure 2 illustrates the ultimate recovery factor versus salinity in six WF tests. As can be seen, oil production is almost the same value during injecting water with salinities of 20,000 ppm and 40,000 ppm (33–34%). But below the salinity of 20,000 ppm, the trend of the figure has changed dramatically with a reduction in salinity. With the further reduction in salinity, particularly below than 5000 ppm, the rate of increase in oil recovery has become steady. Injecting water with the salinity of 1000 ppm increased recovery by 9 percent compared to the salinity of 40,000 ppm. According to Fig. 2, it is clear that decreasing salt concentration leads to a significant increase in the ultimate recovery factor, particularly in salinities lower than 10,000 ppm.

One argument on this remarkable growth can be understood by Fig. 3, which indicates injection profiles of WF with different salinities from 40,000 to 1000 ppm. It can be noticed that the extra oil recovery during WF with lower salinity can be due to the role of fine migration. This phenomenon makes the injection water divert from water regions into the areas with higher oil saturation and improve mobility ratio (Dang et al. 2013; Lever and Dawe 1984; Lemon et al. 2011; Tang and Morrow 1999). It can be inferred that low-salinity water injection can enhance the ultimate recovery factor by fine migration phenomenon in clay sandstone reservoirs.

In addition to ultimate recovery, we measured the eventual absolute permeability of cores after injection of 10 PV. Figure 4 depicts the percentage of absolute permeability reduction $[(1-k/k_0) * 100]$ concerning salt concentration. The permeability loss during injecting water with the salinity of

Fig. 3 Water injection profile of core flooding in two salinities of 40,000 and 1000 ppm

Fig. 4 Reduction percent of absolute permeability during WF with the different salt concentrations

40,000 ppm is almost zero, and it is around 2 percent for the salinity of 20,000 ppm. Injecting water with the salinity of 10,000 ppm dropped off the permeability sharply, and this reduction gets faster when total dissolved solid in water is decreased. The highest permeability drop is 32.3 percent, and it belongs to 1000 ppm. Having a closer look at Fig. 2 reveals that switching from TDS content of 5000 to 1000 ppm causes a huge permeability loss, but it has an insignificant effect on increasing the ultimate oil recovery.

Until now, lots of researchers have proposed several mechanisms for low-salinity water and its effects on improving oil production, such as the expansion of electrical double layer, multi-ion exchange, pH increase (Sheng 2014). In our research, however, recovery factor experienced a

significant improvement when fine particles appeared in the effluent. As can be displayed from Fig. 3 and Fig. 4, although fine migration has a positive impact on recovery improvement during low-salinity water injection, they do not show exactly the same trends. It can be concluded that the other possible mechanisms improve the recovery in lower salinity, especially in salinity below 5000 ppm. According to DLVO theory, the low-salinity water causes the expansion of the electrical double layer and leads the total energy of interactions to move toward positive value, which increases the tendency of the fine particles to detach from the rock surface. It has been supported that fines migration is capable of oil recovery improvement as released clay particles can entirely block or partially plug pore throats facilitating microflow diversion through establishing new flow channels, proposed for the first time by Bernard et al. (Bernard 1967). This leads to more microscopic sweep efficiency and then higher oil recovery during LSW processes (Tchistiakov 2000; Khilar and Fogler 1983; Khilar et al. 1990; Kia et al. 1987a,b; Ochi and Vernoux 1998; Sharma et al. 1985). Moreover, it is worth mentioning that monovalent and divalent ions can have significantly different effects on formation damage and consequently on ultimate recovery. In other words, the expansion of electrical double layer is an underlying mechanism engaged in oil–brine and brine–rock interactions. Accordingly, when a polar surface is exposed to an electrolyte solution, an additional ionic layer is established as a result of ion adsorption (Salopek et al. 1992). According to the equation below, Debye length (DL), κ^{-1} is correlated inversely with ionic strength (I) (Myint and Firoozabadi 2015):

$$\kappa^{-1} = \left(\frac{\epsilon_r \epsilon_0 k_B T}{2 N_A e^2 I} \right)^{1/2} \quad (1)$$

where ϵ_r and ϵ_0 represent the relative permittivity of brine and free-space permittivity, respectively. k_B is Boltzmann constant, T represents temperature, N_A indicates Avogadro's number and I is determined by:

$$I = 0.5 \sum_{i=1}^n c_i z_i^2 \quad (2)$$

As can be seen, the ionic strength is formulated as the summation of the molar concentration, c_i of each ion species multiplied by the valence squared, z_i^2 . This matter highlights the greater contribution of monovalent ions rather than divalent ions in facilitating oil drop release from rock surface and consequent oil recovery improvement by expanding the electrical double layer. As the proportion of each ion species in the brine solution is constant in all experiments, quantification analysis of the effect of each monovalent ion and

divalent ion on Debye length expansion has been neglected in this study.

Tchistiakov et al. reported that in dirty sandstone with 7–15% clay content of mostly *illite*, *chlorite* and *montmorillonite*, the grain size is not a major factor in determining the permeability of the rock. The “pore-lining” clay particles reduce fluid flow path by lining on the pore throat. Weakening the brine ionic strength increases the diffuse layer, which can cause significant formation damage (Tchistiakov 2000). In another theory, it has been assumed that basic and acidic components of heavy crude oil adsorb on the fine clay particles and make them become mix-wet. By introducing low-salinity water into the porous media, fine particles move and carry oil droplets towards production well (Figs. 5, 6). In this standpoint, the presence of residual water saturation, clay fine detachment and absorption of the polar component of oil on the surface of fine particles are three essential factors (Tang and Morrow 1999). In these experiments, we introduced salinities below 10,000 ppm as low-salinity water since the results of these salinities are quite different from those with higher salinities. We also observed critical exchange capacity of our system between 10,000 and 20,000 ppm during conventional water flooding.

CWI. To have a comprehensive understanding of the advantages and disadvantages of CWI, we conducted six other core flood experiments. As a matter of fact, all parameters are just like water flooding other than carbonated level.

Fig. 6 CO₂ solubility vs. salinity in freshwater (independent of pressure and temperature) (“P1-05@ xxxx)

It allows us to compare CWI in both oil recovery factor improvement and formation damage with WF and come to a better conclusion. For having the optimum CO₂ amount in carbonated water, the brine concentrations are selected from Fig. 7. According to this figure, CO₂ solubility would increase dramatically in salinity below 50,000 ppm.

Figure 8 represents the obtained ultimate oil recovery of carbonated water injections, varying salt concentration up to 10 PV. Unlike water flooding experiments, there is a growing trend of ultimate recovery factor for salinities lower than 7000 ppm. It also shows that CWI 1000 has resulted

Fig. 5 Effect of clay fine migration on recovery improvement throughout low-salinity water flooding

Fig. 7 Ultimate oil recovery of six carbonated water flood experiments with respect to several salinities

Fig. 9 Percentage of absolute permeability reduction versus dissolved salt in carbonated water after performing six CWI experiments

Fig. 8 Oil recovery curve for CWI in 40,000 and 1000 ppm

in the maximum ultimate recovery of 61.2% and improved oil recovery of 14.7% than CWI 40,000. In addition to fine migration phenomenon and consequent microflow diversion during LSW process, when less solid particles dissolve in the water phase, much carbon dioxide can be miscible under constant pressure and temperature. As a result, carbon dioxide is transferred from the aqueous phase to the oil phase reducing oil viscosity and water–oil interfacial tension and more oil recovery is achieved through wettability alteration (Foroozesh and Jamiolahmady 2016; Seyyedi et al. 2017; Sohrabi et al. 2015,2008; Steffens 2010; Son Tran 2009; Riazi et al. 2009). Besides, by comparing with what was observed in the water flooding experiments, CWI 1000 has improved oil recovery by 19% in comparison with WF 1000. Figure 9 displays the difference between the injection

Fig. 10 Residual fine after water and carbonated water injection 40,000 ppm (no fine was observed in WF 40,000)

profiles of CWI in 40,000 and 10,000 ppm. As observed, breakthrough time of carbonated water injection is the same as water flooding. This issue is in line with what Seyyedi et al. cited (Seyyedi et al. 2018).

Figure 10 reveals the effect of CWI on formation damage and fine migration. It introduces permeability drop percentage during CWI with various total dissolved solids.

Also, migration of sand has been monitored in the effluent using XRD analysis (Fig. 11). The composition of the solid effluent of WF 1000 and CWI 1000 is reported in Table 4. For water flooding, the effluent XRD analysis shows much amount of 46 wt% of *kaolinite* in the effluent as it had formed most of the clay content of the core samples. It can

Fig. 11 Result of XRD analysis of produced fine in outlet fluid of WF_1000 and CWI_1000 **Table 4** Weight percent of mineral compositions produced by WF 1000 and CWI 1000

Component	WF (wt%)	CWI (wt%)
Quartz	3	7
Kaolinite	46	29
Montmorillonite	23	18
Chlorite	7	17
Illite	19	14
Feldspar	1	12
Dolomite	1	1
Calcite	–	2

be deduced that *kaolinite* is more responsible for fine migration during LSW (low-salinity water) flooding among *illite*, *montmorillonite* and *chlorite* and the role of other clay types is not significant. The amounts of *illite* and *montmorillonite* are close together in the effluent sample, and *chlorite* plays a small role in this process.

Permeability reduction is caused by the change in ionic strength, and it gets intense by salinity shock effect in low-permeability areas. Moreover, small radius of pore throats has made the high velocity of the injected fluid and clay particles detachment. On the other hand, dissolution of calcite and dolomite (mainly known as rock cement) accelerates detachment of other minerals such as quartz and detrital feldspar. In CWI, however, the result was a bit different.

The beginning of clay fine migration occurred at 7000 ppm, which is compatible with Fig. 8. Just as water flooding, it can be understood that fine migration is one of the recovery improvement mechanisms during low-salinity carbonated water injection. It is also clear that although carbonated water injection reduced absolute permeability of the rock, it could control fine migration by postponing initiation of clay separation from rock surface (Seyyedi et al. 2018). In the first two tests (CWI 40,000 and CWI 20,000), permeability reduction percentage was less than what had been observed in water flooding, but the status has been changed in 10,000 ppm. The argument why carbonated water

represents different interactions with porous media than conventional water flooding is as follows. The following reactions release a high amount of H^+ in carbonated water rather than brine water and drop pH of the fluid:

By increasing the amount of H^+ , dolomite and calcite minerals of the reservoir rock would be dissolved through different reactions as below:

Dolomite:

Calcite:

Although the dissolution of quartz and feldspar is higher during carbonated water injection at the salinity of 1000 ppm compared to WF with the same salinity, *kaolinite* shows the opposite behaviour. According to DLVO theory for a sphere-plate model, the double-layer repulsive force can be calculated by Eq. 8. ϵ is dielectric constant, α_p is the radius of particle and κ is Debye length:

$$V_{DLR} = \left(\epsilon \times \frac{\alpha_p}{4} \right) \times \left\{ 2\psi_{01}\psi_{02} \ln \left[\frac{1 + \exp(-\kappa h)}{1 + \exp(-\kappa h)} \right] - (\psi_{01}^2 + \psi_{02}^2) \times \ln [1 - \exp(-2\kappa h)] \right\} \quad (10)$$

ψ is considered as zeta potential (ζ), and it is related to pH in Eq. 11:

$$\zeta_i = \psi_{oi} = -2.3 \left(\frac{KT}{e} \right) (pH - pH_{oi}) \quad (11)$$

pH_{oi} is defined as the isoelectric point of the surface at which the zeta potential becomes zero. Indeed, the position of the isoelectric point has an important effect on the sensitivity of the system to the fluid conditions and temperature (Schembre et al. 2006). Schembre et al. investigated that by introducing high pH condition into clay sandstone, more significant permeability drop and intense fine migration have taken place. They deduced that with an increase in pH, the zeta potential tends to be more negative, which increases repulsive force (Schembre et al. 2006). By increasing alkalinity as well, the zeta potential of minerals would decrease (Kaya and Yukselen 2005). Musharova et al. observed that potential clay stabilising solutions such as HCl work well for sandstone reservoirs at high temperature. They reported that injection of 15 wt% HCl with 0.8 wt% corrosion inhibitor with a measured pH of 0 had preserved the original permeability. They also concluded that acid solutions could be considered to work as a clay stabilising agent (Musharova et al. 2012). Nunez et al. reported some percentage increase in permeability during CWI in dolomite cores (Nunez et al. 2017). In contrast to their investigation, permeability tends to decrease in the sandstone rock. We believe that fewer permeability drops of low-salinity carbonated water are due to its potential to control clay fine migration, especially *kaolinite* fine migration by lowering the pH of the environment and making it stable. It also has been assumed that the reason behind two stages of the intense decline in permeability value is related to the variation in CEC for each clay mineral in both WF and CWI experiments.

Fig. 12 **a** Recovery improvement of secondary CWI compared with WF respect to salinity **b** Percentage of the difference between formation damage caused by CWI and WF

To have a better assessment of CWI performance, Fig. 12 is presented. This figure shows the amount of CWI recovery improvement and its ability to control or intensify permeability drop than WF in each salinity. As it clearly indicates, CWI worked worse on the rock surface and caused more serious formation damage than WF intensifying fine migration in 40,000 and 20,000 ppm. From 15,000 ppm on, it has diminished the permeability drop, and among salinities lower than 10,000 ppm, 5000 ppm displayed the best improvement in controlling the fine migration where 7000 ppm was less efficient in hindering formation damage. In recovery improvement graph, the results show domination of CWI than WF in all salinities. In addition, there is a good improvement from 40,000 ppm to 1000 ppm, but the figure has few fluctuations. According to this figure, 40,000 ppm and 10,000 ppm were less effective in improving recovery, and 1000 ppm has the highest value. It also indicates whenever the curve of differential formation damage increases, the ultimate oil recovery decreases and vice versa. In the end, we can conclude that fully saturated carbon dioxide miscible in low-salinity water is promising to diminish fine migration and reduce permeability in the presence of the oil phase. In fact, in our case, CWI could reduce the permeability drop by 9.7% at 1000 ppm. However, it has been reported that crude oil may also prevent formation damage and permeability decline of low-salinity water (Mungan 1965; Farajzadeh et al. 2017). The presence of oil might act as a barrier to prevent direct contact of fine and aqueous phase coating pore surface by organic components.

Conclusion

This paper investigated the influence of CWI and WF on oil recovery improvement and induced formation damage in various low salinities. The result of CWI on permeability in sandstone media can be quite different from dolomite (observed by Nunez et al.) (Nunez et al. 2017). However, the following conclusion can be proposed from this study:

- (1) Generally, the oil recovery improvement of CWI is more than WF (e.g. up to 19% improvement in salinity of 1000 ppm).
- (2) In water flooding, the reduction in salinity concentration leads to recovery improvement, and it gets more effective in salinities below 20,000 ppm for WF. The maximum recovery was achieved in 1000 ppm, which was 9% more than the 40,000 ppm. In this paper, we supposed, because of clay fine migration, salinity reduction has enhanced the ultimate oil recovery.

- (3) The salinity reduction has a stronger impact on enhancing the oil recovery in CWI (14.7% higher oil recovery in 1000 ppm compared to 40000 ppm).
- (4) Salinity reduction causes formation damage in both WF and CWI. For WF, a sharp permeability reduction has occurred at 10,000 ppm simultaneously with a sharp increase in recovery improvement.
- (5) In the salinity below 10,000 ppm, CWI had a significant role in clay stabilisation (particularly *Kaolinite*) and hindering the fine migration that can be attributed to the decreasing pH by CO₂ dissolved in water.

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