



Carbonated water injection: an efficient EOR approach. A review of fundamentals and prospects

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Abstract

Carbonated water injection (CWI) might be an efficient alternate to CO₂ injection technique. In CWI, CO₂ exists as a dissolved phase and not as a free phase; thus, it eliminates some challenges encountered in CO₂ injection such as poor sweep efficiency and gravity segregation. In CWI, the density and viscosity of water become higher than normal due to the CO₂ dissolution, thereby reducing the gravity segregation and channeling effect. This article is a comprehensive review on how carbonated water flooding has evolved over the time and captured salient features on the mechanisms involved in its role in enhanced oil recovery. The aspects reviewed in this article include a brief comparison of conventional CO₂ injection and carbonated water injection and the benefits thereof. Solubility of CO₂ in water, brine and oil phases is discussed in detail with valid correlations. A brief history of the development of CWI in the laboratory and field information is captured from 1905s to the present followed by the possible mechanisms and principle of CWI reported by various authors. This article also captured the latest findings on the beneficial effect of hybridizing CWI with smart water technologies.

Keywords Enhanced oil recovery (EOR) · Carbonated water injection (CWI) · Carbonated smart water injection (CSWI)

Introduction; CO₂ injection and its limits

From the information available in public domain, it is unambiguous that tertiary or enhanced oil recovery (EOR) through CO₂ flooding is a proven techno-economically efficient method of recovering additional oil from conventional light, medium as well as heavy oil reservoirs (Gao et al. 2010). The factors that contribute to the oil recovery are mainly related to lowering interfacial tension, swelling oil droplet volumes, reducing oil viscosity, and by mobilizing the lighter components of the oil, which are elaborated as follows.

CO₂ coexists as gas and liquid in a single phase at its critical pressure (P_c) and temperature (T_c) (1070 psia and

87.9 °F, respectively). Above P_c and T_c (supercritical state) though its density is close to that of the liquid, its viscosity remains close to the viscosity of its gaseous phase (0.05–0.08 cp). These properties help in oil recovery by reducing the overall viscosity of oil and also reducing gravity override problem, compared to gaseous CO₂ injection (Jarrell et al. 2002).

CO₂ flooding is categorized mainly into two methods, miscible and immiscible flood. This classification depends on the reservoir rock and fluids properties at reservoir temperature and pressure conditions. When the reservoir is deep enough and the reservoir pressure exceeds the minimum miscibility pressure (MMP), CO₂ and reservoir oil reach miscibility condition through a mechanism classified as multiple-contact miscibility (MCM) which is a dynamic and time-dependent process. This consists of vaporization gas-drive (in which intermediate hydrocarbon molecules vaporize into the CO₂) and condensation gas-drive process (in which a portion of the injected CO₂ dissolves into the oil (Merchant 2010). This combined mass transfer process helps CO₂ and oil to become a single phase and drive the oil very efficiently (Diaz et al. 1996; Ju et al. 2012). If the oil is extremely light and of ultra-low viscosity, the first contact miscibility is also possible. Thus, miscibility mechanism

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depends predominantly on the crude oil composition (Kantzas et al. 2012). Though direct relationship between reservoir rock properties and CO₂ miscibility is not yet established, reservoir rock properties can have a significant impact on overall performance of CO₂ flooding performance, primarily attributed to the heterogeneity, permeability and overall porosity of the rock. Takahashi et al. (2003) demonstrated that CO₂ breakthrough can occur much earlier in carbonates than in sandstones due to greater microscopic heterogeneity of carbonate rocks. This statement is supported by Bikina et al. (2016) through a series of miscible CO₂ coreflood on oil wet and water wet (whole and fractured core plugs). They observed that the miscible CO₂ flooding recovered nearly 100% of the oil, in oil-wet homogeneous cores, while insignificant amount of oil was recovered from water-wet fractured cores. Under similar poro-perm conditions, they observed that miscible CO₂ flooding performed significantly better in the oil-wet cores compared to the water-wet cores. During a field simulation and implementation study, Bhatti et al. (2019) have emphasized on the importance of reservoir wettability, heterogeneity and permeability properties and strongly suggested for inclusion as the screening criteria for miscible CO₂ flooding.

There are also after effects of miscible CO₂ flood which needs to be considered too. Reduction in porosity was observed in both secondary and tertiary CO₂ flood modes, while permeability was seen to improve in tight carbonate rocks. Rock dissolution at high CO₂ concentration in water phase impacts not only the poro-perm characteristics but also the wettability characteristics. The injection of super critical CO₂ in tight carbonate rocks not only improved overall rock permeability but also changed the wettability toward a more water-wet state which favored improved recovery (Zekri et al. 2013). On the contrary, a recent work of Wang et al. (2019) evidenced that the CO₂ flooding in sandstone rock may result in reduction in permeability with almost no change in porosity. The degree of permeability damage is found to be more after CO₂-WAG flooding than that after only CO₂ flooding. The damage analysis through PSD (pore size distribution) studies revealed that fines migration and blocking of small pore throats are responsible for the permeability damage.

Numerous laboratory and field studies proved that miscible gas flooding is way more efficient in terms of incremental oil recovery than immiscible flood (Agustssen and Grinestaf 2004; Cobanoglu 2001; Gao and Towler 2012; Sharma and Clements 1996). During the last two decades, miscible CO₂ displacements have been well developed and applied, particularly in the US and some Chinese fields (Jishun et al. 2015; Liu 2013). The first ever CO₂-miscible flood field pilot in Permian basin (Western Texas fields) was conducted in 1976. Since then, many large-scale CO₂-miscible flood projects have been implemented with appreciable economic success

(Verma 2015). Based on the published documents, the current “best practices” CO₂-EOR technology generally recovers CO₂ incremental oil of around 5–15% of OOIP from North American fields (Gao and Towler 2012), depending mainly on the reservoir rock and fluid characteristics and the flood pattern. Healy et al. (1994) reported about 9% incremental oil recovery from miscible flood recovery from Sacroc field, whereas incremental recovery resulting from the miscible CO₂ flood is reported to be more than 15% of the OOIP in San Andres Unit (Stiles and Magruder 1992). This incremental recovery is below the industries expectation of > 80% of overall recovery (Merchant 2010), and thus leaves behind a large volume of oil in the reservoir. Analysis show that the major causes of below expectation recovery are due to (a) insufficient injection of CO₂, (b) poor sweep efficiency, (c) poor displacement efficiency, (d) lack of CO₂ contact with remaining oil resources and (e) inadequate management control (Verma 2015).

As reported by Summapo et al. (2013), reservoir heterogeneity is the major cause of poor CO₂-miscible flood performance due mainly to higher unstable flood front and an early breakthrough of injected gas. This may result either from formation depositional sequence which defines the path preference or from natural fractures which facilitate CO₂ channeling due to the high mobility of supercritical CO₂ fluid. Injecting CO₂ alone is found to cause early breakthrough of CO₂ along fracture direction (Figuera et al. 2014), which necessitates corresponding modeling of injection/production adjustment strategies in advance (Luis et al. 2016). It is suggested that flood breakthrough in multilayered, multi-well CO₂ flood system can be identified through pulse-neutron production logs combined with permeability from the magnetic resonance (Aryani et al. 2011). Unlike miscible flooding, gas breakthrough is more predominant in immiscible flooding in which gas moves through high permeable channels as evidenced in Yaoyingtai oil field (Yuncong et al. 2014). The conclusions were drawn from the changes in daily oil production, gas production rate, CO₂ content and GOR from 40 oil data.

Because of the reasons described above, direct gas injection and water-alternating-gas (WAG) injection might lead to commercial failure in heterogeneous layered and fractured formations to meet the targeted incremental oil within the projected economics. In addition, poor sweep implies lower storage capacity of CO₂, which is an additional objective of CO₂ injection, popularly known as carbon storage and sequestration (CCS) aiming at reducing the greenhouse effect.

Alternative method: Carbonated Water Injection (CWI)

To counter the issues mentioned above, carbonated water injection or CWI, that is injecting water saturated with CO₂ instead of direct injection of CO₂, is gaining rapid attention.

Though this technique was first conceptualized in the 1930s, serious investigation began in the 1970s. It is found that because of lesser difference of viscosity and density with the crude oil, CWI has better sweep efficiency than supercritical CO₂ (Sohrabi et al. 2008). Moreover, in reservoirs that have been water flooded, CWI can mitigate the negative effects of water shielding due to mixing with the resident water (Riazi et al. 2009). In the case of direct CO₂ injection, diffusion of CO₂ would take longer time because of low sweep efficiency and gravity segregation effects (Solomon 2007).

Technically, two major differences can be cited between CWI and conventional CO₂ injection or water-alternating CO₂ gas (WAG) injection. Firstly, the amount of CO₂ that might be injected to the reservoir at certain temperature and pressure will be solubility limited, which implies no separate CO₂-rich phase in the reservoir. The other point is that the displacement efficiency would not depend on the minimum miscibility pressure (MMP), because it is controlled by CO₂ mass transfer between oil and CW resulting in no-transition zone in CWI (Dong et al. 2011).

CWI can be a very attractive method for CO₂ sequestration too, which is becoming a hot topic in terms of environmental issues and can bring benefits in the form of reduction in greenhouse gases. It is reported by IPCC (Metz 2007), that to avoid the fast climate change and its side effects, global CO₂ emissions should be cut by 50–80% in 30 years. Therefore, CO₂ capturing and storage or CO₂ sequestration (CCS) has great importance that should be considered.

In conventional CO₂ sequestration processes, CO₂ floats under the cap rock and there are possibilities of leakage through the micro-pores of the cap rock. This sometimes limits the number of reservoirs available for CO₂ sequestration (Herzog 2000). On the contrary, CWI sequestration can be implemented without volume limitations with lesser risk of gas leakage through cap rock. Having a higher density and viscosity than resident water due to CO₂ dissolution (Hebach et al. 2004), carbonated water will sink into the bottom of the reservoir, eliminating the risk of buoyancy-driven leakage which is usually caused by bulk phase gas injection (Burton and Bryant 2009). In addition, CO₂ exists as a dissolved phase rather than a free phase, thus reducing the issues which are caused by the poor sweep efficiency and gravity segregation which are the drawbacks of typical direct gas injection. As a result, CWI provides a safer and better method of CCS compared to direct injection of CO₂ (Anchliya et al. 2012).

Basic theory and fundamentals for CWI

CO₂ phase behavior

The CO₂ phase behavior is highly dependent on temperature and pressure of the reservoir. Figure 1 represents a phase

diagram illustrating that CO₂ injection can be sustained under different forms such as liquid, gas or supercritical fluid, all of which depend on the two factors P and T. Other properties that are affected by P and T include viscosity, where rising temperatures can reduce it significantly. Additional properties include gas compressibility factor and density. It is recommended for CO₂ to be in the gaseous or supercritical form while injected. To specify, in supercritical condition, useful characteristics of gas and liquid phases coexist and its behaviors are similar to gaseous CO₂ and liquid CO₂ under certain circumstances. A supercritical CO₂ provides the characteristics of liquid because of its similarity in density to the liquid state.

Solubility of CO₂ in water, brine and oil phases

The amount of dissolved CO₂ is one of the critical factors to every application including the CWI because it directly affects the data variation, resulting in its application efficiency. Therefore, the solubility of CO₂ in water, brine and oil must be ascertained at the condition of its application. Several CO₂ solubility studies in water and high- and low-salinity brines have been conducted for an extensive range of temperatures, pressures and ionic concentration in conjunction with different reservoir properties by many researchers (Bamberger et al. 2000; Chang et al. 1998; Chapoy et al. 2004; Gui et al. 2017; Liu et al. 2011; Valtz et al. 2004).

Solubility of CO₂ was investigated both in pure water and in diverse brine solutions (Mg²⁺, K⁺, Na⁺, Ca²⁺, Cl⁻, and SO₄²⁻) at several different temperatures and at pressures up to 200 MPa (Duan et al. 2006). Solubility of CO₂ and other injection gases in water and NaCl solutions is modeled at varying temperatures (0–350 °C), pressures (0.1–150 MPa) and ionic concentrations (0–4.5 mol/kg) by Mao et al. (2010) using Helmholtz free energy model, which can be extended to different ranges of the variables parameters.

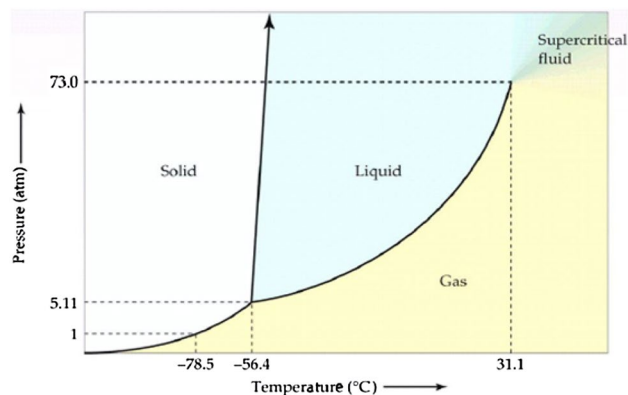


Fig. 1 Carbon dioxide (CO₂) pressure–temperature phase diagram (Whitson and Brulé 2000)

The correlation proposed by Enick and Klara (1990) on the CO₂ solubility in brine at surface condition is found to be applicable at subsurface formation conditions too, including consideration of the dissolved solids in the brine. The major conclusion drawn from this work is that solubility is solely dependent on total dissolved solids, regardless of the salt type. Overall conclusion drawn from the above studies is that there could be a significant influence of brine salinity on CWI flooding performance; hence, it is necessary to execute solubility test with reservoir brine at reservoir temperature and pressure prior to performing injection.

Solubility Correlations

One of the most important parameters that affect gas solubility is Henry's constant. It is defined as the limit of carbon dioxide's fugacity to CO₂ water ratio. K_{H,CO_2} has the dimension of pressure (Eq. 1) (Diamond and Akinfiev 2003).

$$K_{H,CO_2}(T, p) = \lim_{x_{CO_2} \rightarrow 0} \left(\frac{f_{CO_2}}{x_{CO_2}} \right), \quad (1)$$

where x_{CO_2} —the molar fraction of CO₂ in water, f_{CO_2} —fugacity of CO₂, K_{H,CO_2} —Henry's constant.

To verify the above correlation experimentally, Chang et al. (1998) studied the properties of carbonated water binary system including the CO₂ solubility in water and brine and observed that the viscosity of CO₂-saturated water remained unchanged. For solubility measurements, they used Eq. 2. given as follows for the estimation of CO₂ solubility in distilled water and later adjusted to the salinity effect of the brine (Kechut et al. 2011) and found positive agreement between the measured values and the calculated values of solubility of CO₂, implying that the correlation Eq. 2 is a reliable one.

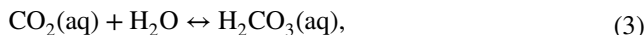
$$\log \left(\frac{R_{sb}}{R_{sw}} \right) = -0.028 \times S \times T^{-0.12}, \quad (2)$$

where R_{sb} : solubility of CO₂ in brine of salinity S (scf/STB), R_{sw} : solubility of CO₂ of water (scf/STB), S : salinity of brine in weight % of solid and T : temperature (°F).

Although it was thought that CO₂ solubility increases as pressure increases, however, below 65 °C, solubility decreases as temperature increases (Fig. 3). It can also be seen that the solubility goes up with increasing temperature when $P > 30$ MPa, while the solubility decreases with increasing temperature when $P < 30$ MPa (Perkins 2003).

Carbonate (CO₃²⁻), bicarbonate (HCO₃⁻) and carbonic acid (H₂CO₃) are the main three ions that exist once CO₂ is made soluble in water. Additionally, there are other possible ions which can contain inorganic carbon as well, but these concentrations are much less than that of the main

three ions, so they are usually ignored during calculation (NaCO₃⁻, NaHCO₃, Na₂CO₃, MgCO₃, MgHCO₃⁺, etc.). For example, the reaction of an equilibrium state established by carbonic acid (H₂CO₃) can be expressed as (Eq. 3):



The relationships which demonstrate the relative carbonate ions concentrations are two mass balance equations and mass action equation given as follows:

$$\text{Total Dissolved Carbon} = m(H_2CO_3) + m(HCO_3^-) + m(CO_3^{2-}), \quad (4)$$

$$\log K_1 = \log a(H_2CO_3) - \log a(H^+) - \log m(HCO_3^-), \quad (5)$$

$$\log K_2 = \log a(HCO_3^-) - \log a(H^+) - \log m(CO_3^{2-}), \quad (6)$$

Where the total dissolved carbon is obtained in analytical approach, log K_1 and log K_2 are relevant log equilibrium constants for the equilibrium and m and a are molality and activity of aqueous species, respectively.

The partial pressure of carbon dioxide and dissolved carbonate ions in solution can be related to each other as the following relationship:

$$\log K_3 = \log p(CO_2) + \log a(H_2O) - \log a(H_2CO_3), \quad (7)$$

With an increasing CO₂ gas partial pressure, CO₂ will be dissolved into the fluid, while with decreasing CO₂ partial pressure in the gas CO₂ will be evolved from the fluid. If the fluid turns more alkaline, CO₂ will immediately start to evolve from it. The more the CO₂ dissolves in the fluid, the more acidic it will become. Thus, more acidic solution might result in the dissolution of the rock, especially carbonate minerals from the rock surfaces (Perkins 2003).

Another important solubility parameter which may govern the success of CWI as regards phase transfer is the CO₂ solubility in crude oil. The important properties identified are crude oil composition, temperature and saturation pressure (Emera and Sarma 2006; Jamaluddin et al. 1991; Srivastava et al. 1995).

Some mathematical correlations were established for CO₂ solubility estimation in crude oil. However, these relations are restricted to certain ranges of fluid properties and conditions. Therefore, additional research on CO₂ solubility and CO₂-associated factors that affect oil swelling is required to find out the processes and mechanisms related to CO₂-based EOR methods.

History of CWI laboratory work

In the late 1940s, initial research on CWI was performed by Montclair Research from the Oil Recovery Corporation (Adiputra et al. 2018). They reported that it was possible to reduce the residual oil saturation up to 15% of pore volume

when carbonated water was flooded after conventional water flooding. From 1948 to 1952, Earlougher Engineering conducted carbonated water flood experiments with freshly sampled cores at low pressure ranges (800–1000 psig) with oil API gravity ranged from 28 to 50 API. Results indicated that CWI delivered a range of residual oil saturation depending on oil gravity. Additional recovery on tertiary mode after waterflood was achieved for light oil (up to 26%), whereas for heavier oil the additional recovery was as low as 2% of the pore volume (Lake et al. 1984). The mechanism behind oil recovery was accounted for the changes in both rock and fluid properties. Some experiments suggested that the result of additional oil recovery is due to a series of alterations in both the rock and fluid characteristics, not only relying on the oil swelling effect (Lake et al. 1984).

Martin (1951) reported that 12% oil recovery enhancement was resulted through carbonated water injection. He earlier pointed out that the recovery improvement might be correlated with the amount of carbonation in fluids (Martin 1950). Based on the work of Saxon Jr et al. (1951), Johnson et al. (1952) examined the effect of CWI on oil recovery and compared with brine flood. Using two different oil viscosities (1.42 mPa.s and 2.86 mPa.s), they concluded that CWI method could recover 15–25% of residual oil, while no recovery was observed using brine injection. The results also pointed out that oil recovery using CWI method is temperature dependent. At lower temperatures, the recovery was higher, due to higher solubility of CO₂ (Fig. 2). High solubility of CO₂ would cause further expansion in oil volume, thus resulting in better oil recovery. This observation was supported by coreflood experiments conducted by Holm (1959). The results show that keeping all parameters same, whereas 21% incremental oil recovery was achieved by CWI at 21.1 °C, the incremental recovery was limited to 19% at 37.8 °C, establishing a relation between reservoir temperature, CO₂ solubility and incremental recovery.

Several flooding experiments with sand pack were conducted for CWI by Falls (1986); Gorell and Falls (1986) and earlier by Van Dijk (1965). Conclusion drawn from these articles is that CW could bring about 12% to 23% of additional oil recovery after conventional water flooding, depending on oil viscosity. Panteleev and Tumasyan (1972) conducted a novel study on wettability changes in porous media. They observed faster water imbibition and higher oil recovery for CW compared to fresh water. CW imbibition produced 37.3% of oil compared with 26.5% oil recovery by fresh water imbibition. They also emphasized that higher CO₂ concentration in water would result in further increase in oil recovery and imbibition rate.

Flumerfelt et al. (1993) continued the oil recovery studies from Perez et al. (1992). Kerosene oil, crude oil and low-permeability dolomite rock cores were used. They also studied the influence of surfactants during carbonated water

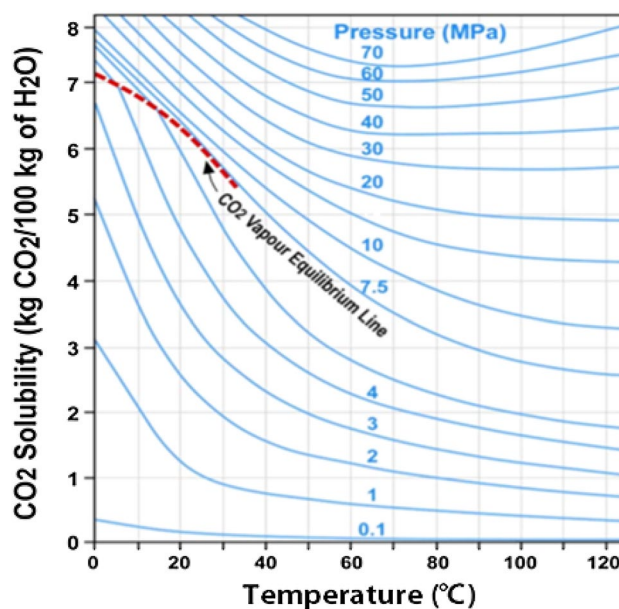


Fig. 2 CO₂ solubility in water depends on temperature and CO₂ pressure (Perkins 2003)

injection. The results showed that CW with surfactant led to the recovery of additional 50% of residual oil, while 20% incremental oil was recovered with CW without surfactant, comparing with conventional WF. During the CWI experiment by Asghari et al. (2009), 16.9% of additional original oil in place was recovered from the consolidated core samples and around 14% of OOIP while using sand packs.

Since 2006, extensive research was conducted on CWI for oil recovery by the Herriot Watt Institute of Petroleum Engineering Centre. These experimental and numerical studies were carried out by different techniques and methods including the following: micro-model, core flooding, mathematical modeling and numerical simulations. They reported that the improved oil recovery through CWI was due to higher sweep efficiency, viscosity reduction, oil swelling and thus reconnection of the isolated oil droplet which all are attributed to better CO₂ diffusion. Two samples tested resulted in 23.8% and 8.8% increment of oil recovery. They also suggested that the fundamental process is related to interactions between the fluid/fluid and fluid/solid during CWI. The studies suggested that significant improvement in oil recovery could be possible in both secondary and tertiary recoveries by using CWI (Sohrabi et al. 2011, 2015). Riazi et al. (2011) set up a mathematical model to investigate pore-scale mechanisms during flooding, which revealed that CWI might be an attractive method to store CO₂ in certain reservoirs as well. Kechut et al. (2010) focused on maximizing CO₂ concentration in CW and was able to enhance the concentration up to around 46%, which was significant in view of sequestration. Al Mesmari et al. (2016) conducted a

series of direct visualization experiments using glass micro-models and also coreflood experiments, and the data were used for history matching and simulating the performance of CWI and identified the key parameters controlling the phase behavior of crude oil and CW. To account for the formation of the new phase, three-phase flow functions and relative permeability were incorporated in the model and measured the mass transfer and multi-phase flow behavior during CWI.

More recently, Zou et al. (2018) established that CWI was more effective than water flooding in tight reservoirs. Coreflood experiments conducted on tight core samples from Ordos Basin showed that CWI promoted higher oil recovery in comparison with WF in both secondary and tertiary injection modes by 20.3% and 11.3%, respectively. Qu et al. (2018) also conducted recovery experiments on tight sandstone core samples with a permeability range of 0.1 to 0.2 mD, and compared CWI recoveries with CO₂-WAG and surfactant flooding. They claimed lesser incremental recovery for surfactant (2.05%) and WAG (4.53%) compared to CWI which resulted in 7.22% incremental oil recovery.

History of CWI Field work

In 1957, the first field test for CWI was conducted in Allegany County, New York. The outcome of using CWI was reported as successful. The production rate changed significantly from 92 to 1260 barrels/acre/year. This was followed by several other successful implementations of CWI field trials (Christensen 1961; Hickok and Ramsay 1962). In 1958, injection of CO₂ and water in the same tubing was begun. Production increased in 1959 to 123,000 STB, higher than the total oil produced from 1905 to 1934. Additionally, CWI resulted in an estimated 37% increase in net oil production compared to conventional water flooding in the same reservoir.

Another large-scale CWI implementation in the Domes Unit on 90 wells reported techno-economic success. During the simultaneous CO₂ and water injection, about 30% of pore volume equivalent of CW was injected, followed by water flooding. The reported incremental oil increment was about 9% of OOIP (Riazi et al. 2011).

Possible mechanisms and principle of CWI

Since 1950, carbonated water flooding has been considered as a promising flooding technique, because of enhanced mobility of oil when high-concentration CO₂ is dissolved in the flood water. Mobility (M) is explained as the ratio of permeability of a porous medium (k_{eff}) to that of fluid's viscosity (μ). If there is decrease in oil viscosity, the oil mobility will be higher and vice versa. It is established that the mechanisms of CWI for EOR are on the basis of changing

the physical properties of oil. The proposed mechanisms so far are as follows: (1) CO₂ dissolution in oil leading to oil swelling and subsequent viscosity reduction. This in turn leads to reconnection of the isolated oil droplets and fluid flow diversion (sweep efficiency improvement), (2) evolution of solution gas from the oil caused by CO₂ dissolution, (3) wettability alteration due to CO₂ mass transfer (Sohrabi et al. 2015), and (4) oil swelling also leads to improved relative permeability to the oil, thus providing a better mobility (Riazi et al. 2009).

CO₂ solubility in oil phase is one of the main parameters that affect the EOR performance. This is because it directly affects the oil viscosity and swelling, which enhances oil production (Abedini and Torabi 2013; Mosavat 2014). In addition, one of the key trapping mechanisms which govern CO₂ storability in high-salinity environment is its solubility, where the majority of CO₂ is trapped by dissolving in the formation brine (Ennis-King and Paterson 2005; Lindeberg and Wessel-Berg 1997).

Perez et al. (1992) carried out studies on CO₂-saturated water imbibition at different pressures and temperature. In these experiments, CO₂ was considered as the prime factor that causes an increase in the recovery of oil, compared to other factors normally considered as the major reason for oil recovery in conventional water flooding process. The authors proposed several possible mechanisms such as an increase in oil mobility, increase in carbonate core permeability, oil swelling and a gas-drive mechanism.

In general, it is found that all possible contributions of CWI have favorable impact on oil recovery. For instance, oil swelling has two effects. One of them is the swelling factor of oil which is inversely proportional to residual oil left in the formation. The second is that more volume will be occupied by swollen oil droplets, which means higher relative permeability because of enhanced oil saturation and further reduction in mobility ratio. In CWI, CO₂ is spread in the reservoir more evenly and thereby avoids the CO₂ breakthrough, thereby enhancing the sweep efficiency throughout the formation. Wettability alteration is also a possible contribution by CO₂-saturated water flooding which can shift the surface to a more water-wet state (Dong et al. 2011).

Oil swelling and CO₂ mass transfer

Several researchers have conducted experiments on CO₂-saturated water flooding to study the impact of CO₂ diffusion and the recovery process, including the studies on the performance of certain parameters that affect CWI efficacies. In addition, the effect on mass transfer during carbonated water injection is also studied. There are several proposed mechanisms in terms of CWI that will be discussed in the following chapters.

An experimental study on sand packs using medium-viscosity crude oil and CWI as flooding fluid was performed by Dong et al. (2011). Results indicated that injecting CO₂-saturated brine performs better as a displacing fluid both in secondary and tertiary modes with a reduction in residual oil saturation from 0.03 PV to 0.35 PV, compared to conventional water flooding. The mechanism of such successful outcome is explained by CO₂ migration into the oil phase from the water phase without forming a separate CO₂-rich phase. The mass transfer of CO₂ into oil phase is substantial since it is more soluble in the oil than water (3 to 7 times higher solubility) under the same pressure and temperature conditions. Due to high CO₂ mass transfer into the oil phase, oil becomes less viscous and thus enhances the oil–water mobility ratio. The relative permeability of oil might also improve due to the oil swelling. All these subsequent processes together might have resulted in greater or improved oil recovery than conventional water flooding.

In 1981, extensive PVT studies were performed by Miller and Jones in order to determine how oil physical characteristics might be changed with CO₂ saturation. They concluded that whereas the viscosity of oil without CO₂ saturation increases with increasing pressure, both density and viscosity of CO₂-saturated oil significantly decrease with increasing pressure (Miller and Jones 1981). In order to investigate the effect of CWI as a method of EOR and storing CO₂, Sohrabi et al. (2011) performed laboratory core flooding experiments focusing on oil viscosity, rock wettability and brine salinity. This study reported that in both secondary and tertiary recovery modes, oil recovery was increased. When compared with tertiary recovery (31% reduction in S_{oi}), the secondary recovery (35% reduction in S_{oi}) gave more and earlier incremental oil recovery. Through this study, they explained that the main mechanism is mass transfer of CO₂ into oil, since CO₂ has higher solubility in oil than in water. Thus, the viscosity of oil would be decreased bringing the oil mobility to a more favorable range.

Earlier, Nevers (1964) developed a mathematical model to examine the influence of changing oil phases during CWI, which revealed that CO₂ dissolution and resulting reduction in oil viscosity are the main parameters that can explain the oil recovery mechanism effectively. Recently, Riazi et al. (2011) developed another mathematical model where it demonstrated the dynamic process during oil swelling and its redistribution into oil at pore-scale system. Their results pointed out that the solubility and molar density of CO₂ in water are the most important factors influencing oil swelling during CWI (Mosavat 2014). The investigation of CWI performance on micro-model showed that the oil recovery is optimized due to viscosity reduction, while oil swelling is caused by CO₂ mass transfer between brine and the oil-in-place phase.

Riazi et al. (2009) performed experimental studies on the CO₂-enriched water flooding as an EOR method. The study consisted of direct flow visualization by high-pressure transparent porous media followed by mathematical modeling. They noticed that during CWI, the oil started to swell due to CO₂ mass transfer into oil through CW. The CO₂ diffusion into oil caused reconnection of trapped oil ganglia which started to mobilize subsequently. After performing the second WF, the residual oil saturation was 33.39%, indicating a 15.74% additional oil recovery after CWI. The data showed that CWI could add up to 16% additional oil recoveries. They assumed that the main mechanisms by which CWI recovers residual oil are enhanced sweep efficiency due to oil swelling and reconnection of the isolated oil droplet and the subsequent redistribution of fluid in consequence of CO₂ diffusion. Mosavat (2014) presented that CWI could improve the conventional water flooding by recovering around 19.0% of OOIP during the secondary stage and 12.5% of OOIP in the tertiary stage. This result shows that the CWI might be more effective over conventional water flooding for residual oil recovery from oil reservoirs.

Studies performed by Sohrabi et al. (2008) on glass micro-models show that oil can be swelled even by 105% with CWI (Fig. 3). Figure 4 shows the time dependency of oil swelling, shown in three different time steps, strongly indicating occurrence of oil swelling during CWI. The number of areal oil phase pixels according to time is plotted in Fig. 5, where the estimated increase in swelling was found to be around 22.4%. These visual evidences supported the earlier theories of CO₂ mass transfer from CW to oil leading to oil swelling and viscosity reduction resulting in: (a) increasing oil relative permeability by reconnection of isolated oil ganglia and (b) improving sweep by diversion of flood water. Viscosity reduction mechanisms also play an important role in intermediate and viscous oil systems due to improved mobility ratio.

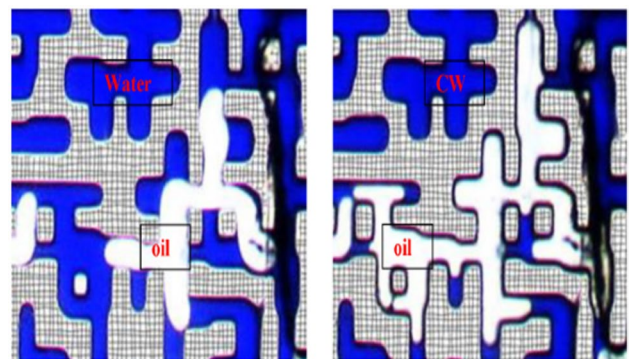


Fig. 3 Comparison of the oil volume before and after CWI. Image analysis observed that a massive 105% oil swelling was captured by image analysis (Sohrabi et al. 2008)

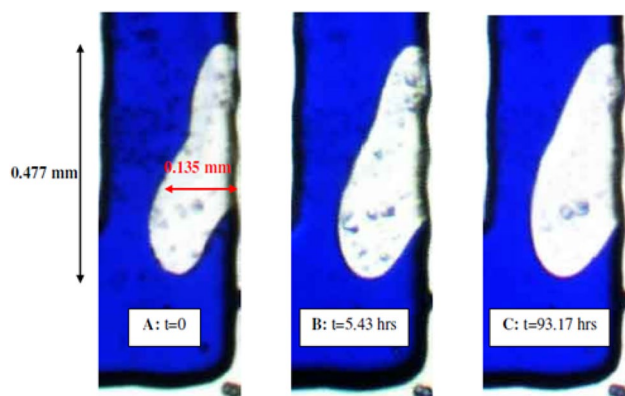


Fig. 4 Oil swelling due to CO₂ diffusion through CW into oil phase (Sohrabi et al. 2008)

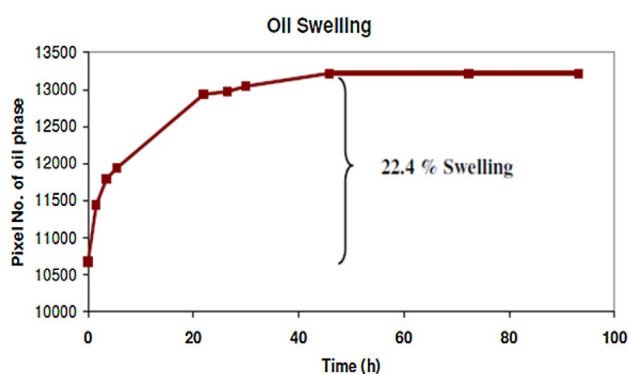


Fig. 5 The oil droplet volume vs time (Sohrabi et al. 2008)

Wettability Alteration

Sohrabi et al. (2008) performed fluid flow studies at high-pressure condition with two-dimensional glass micro-model and observed that as more CW was injected, more amount of CO₂ diffused into trapped oil that was left in pores after normal waterflood displacement. The micro-model was used to investigate the wettability effect. As the surface became more water-wet, the water thickness on the pore surface was seen to increase. The capillary forces changed the fluid interfaces shape in the porous medium, and based on this, they proposed that the change was due to decreasing interfacial tension at oil/brine interface and the wettability alteration toward water wet condition (Sohrabi et al. 2009).

Sohrabi et al. (2015) also studied the fluid interface shape to examine the change in wettability of rock surface. Figure 6a shows that the oil phase has been broken apart and spread out as the water film surrounding the oil ganglia, which means more oil-wet phase after several hours of normal WI. However, after CWI (Fig. 6b), the oil/water interfaces demonstrated a more spherical shape, which

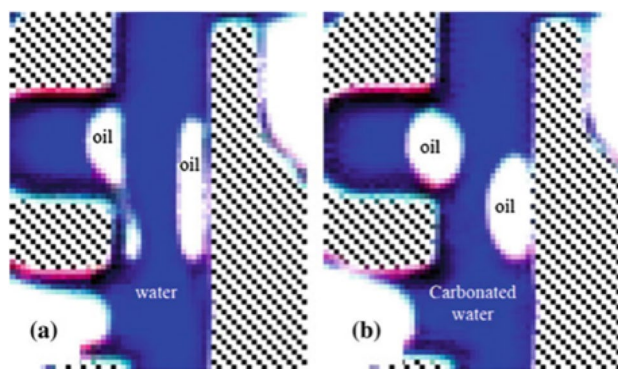


Fig. 6 A magnified image of the micro-model demonstrates two different conditions of wettability: **a** more oil wet after WI and **b** less oil wet after CWI (Sohrabi et al. 2015)

implies more water-wetness (less oil-wet). The capillary forces altered the fluids' interface shape. The fluid interface is determined through wettability and interfacial tension between oil and water. Since the IFT is not so prominent due to CO₂ dissolution, wettability alteration ranks higher as the possible cause of capillary forces alteration. The explanation for wettability alteration is given as: Due to decreases in the aqueous phase pH (in CW), changes in surface charges on the water/oil and water/rock interfaces occur leading to subsequent changes in the wettability of the system. Also, this alteration might be caused by dissolution of CO₂ in oil and destabilizing the polar components of oil. The destabilized polar components can disperse through the water layers and adsorb onto the rock surface which increases the tendency of the pore system to become water-wet.

Evolution of the New Phase

Creation and growth of new gas phase from solution gas within the oil was first seen in the micro-model tests when CW was injected for an extended period of time in dead-oil experiments (Mosavat 2014). Its mechanisms for additional oil recovery are similar to that of oil swelling but to a much larger extent by: (a) oil displacement and reconnection of trapped oil and (b) restriction of water flow path and its diversion toward un-swept parts of the porous medium which results in improved oil recovery. Figure 7 demonstrates micro-model images of the primary WI and tertiary CWI. The CWI was able to promote higher oil recovery and reduces residual oil saturation after primary WF. Results indicate about 7.7% of residual oil saturation has been reduced in the tertiary mode with CWI when compared with the primary water flooding.

Figure 8 demonstrates the ability to capture the trapped oil ganglia in tertiary CWI for recovering isolated oil after primary WI. In Fig. 8a, the residual oil sticks to the oil-wet surface and becomes trapped oil, and after tertiary



Fig. 7 Chronological progress of the primary WI followed by tertiary CWI (Mosavat 2014)

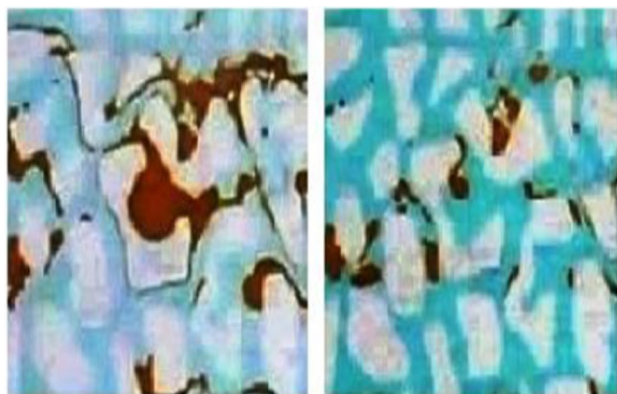


Fig. 8 Trapped oil production during CWI. **a** Before tertiary CWI and **b** after tertiary CWI (Mosavat 2014)

CWI (Fig. 8b) less residual oil in grains is seen, which means improvement in sweep. This is because, injection of carbonated water and hence the mass transfer of CO_2 from brine to oil brings the changes from an oil-wet to water-wet wettability, which helps in the enhancement of oil recovery. The process behind this is the dissolution of CO_2 into the oil system which leads to reduction in viscosity and oil swelling, thus improving the sweep efficiency and finally increasing the oil recovery.

The above images are clear evidence of the expansion of isolated oil ganglia and their reconnection which could be attributed to be the main principle for improvement in oil recovery with CWI. When the system is first water flooded, the oil saturation reaches S_{or} conditions. During the subsequent injection of CW, mass transfer occurs

between CO_2 and residual oil in place, causing an expansion of oil particles resulting in enhanced oil recovery.

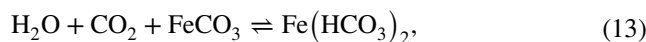
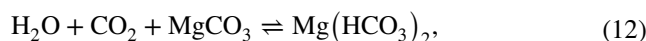
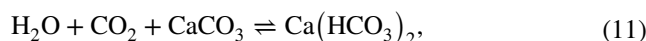
Reaction between rock and CW: Rock dissolution process

The presence of CO_2 in the formation leads to various chemical reactions between rock, aquifer and CO_2 which might affect the petro-physical properties of the reservoir formation. The effect of injected CO_2 depends on several factors such as the rock chemistry, injected fluid type, injection strategy and the physical conditions of the reservoir. It is known that when CO_2 is injected into carbonate formation, it leads to the dissolution of carbonate minerals because of the acidic nature of CW. Dissolution of the rock leads to an initial increase in formation permeability; subsequently, transportation of these minerals and later precipitation lead to decrease in permeability and effective porosity (Bowker and Shuler 1991; Grigg and Svec 2006; Shiraki and Dunn 2000; Wellman et al. 2003).

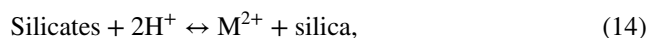
Izgec et al. (2008) observed that with the injection of CO_2 , porosity of core plug is changed with corresponding changes in permeability. CaCO_3 and MgCO_3 are the carbonate minerals that can easily react with carbonated waters. The reactions that can occur with carbonated water are:

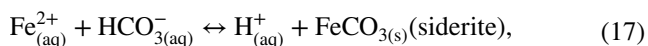
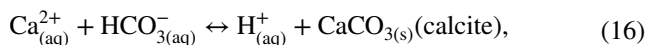
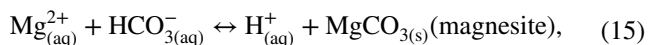


When CaCO_3 , MgCO_3 or FeCO_3 exists in the rock, the water-soluble bicarbonates might form by the following reactions:



The aforementioned reactions might be the reason for rock dissolution in the carbonate reservoir matrix. The rock dissolution will definitely result in the change in petro-physical properties of the rock by creating new flow paths and increasing the rock permeability. However, the presence of CO_2 in sandstone rocks might lead to the reduction in permeability because of dissolution of released cementing particles and later precipitation. If the released particle sizes are more than the pore throat size, they might obstruct the pore throats, thus reducing the rock permeability (Sayegh et al. 1990). The other possible reactions are:





where M^{2+} = generic carbon, l = liquid, aq = aqueous, s = solid.

Several experimental studies have been performed to examine the changes in the rock petro-physical properties by the injection of CO_2 -saturated water (Sayegh et al. 1990). Kono et al. (2014) conducted experimental studies to show how the carbonated water can change the carbonate rock properties in the reservoirs of Middle East, through core flooding at reservoir conditions, measuring porosities and permeabilities. The carbonate mineral dissolution studies were conducted on scanning electron microscope (SEM) and liquid chromatography also. Results obtained (without the compaction effects) showed an increase in porosity by a 3.6% in the first 50 pore volume injected, and a further increase in up to 6.0% from 50 to 100 pore volume injected.

Further Development; Hybrid CWI

With the advent of low-salinity water and smart water flooding techniques, CO_2 -saturated water flooding seems to be an even more efficient EOR option. The synergic effect of these two EOR techniques could be even more promising. Alizadeh et al. (2011) investigated the benefits of carbonated smart water injection (CSWI) in Berea sandstone core plugs above miscible pressure. Distilled water was used to make the aqueous phase where it contained 2 wt% of CaCl_2 , 12 wt% of NaI. In addition, 0.01 wt% of NaN_3 was used as biocides. The density of brine was measured as 1.116 g/ml at 20 °C and atmospheric pressure. The CO_2 -saturated brine contacted with all sizes of isolated oil blobs in the core. Figure 9 shows the comparison between the CT images of core slices and their respective oil saturation after brine imbibition to those at the end of the first and second carbonated smart brine flooding processes. Results indicated that the first CSWI could recover around 51% of the trapped oil and reduce the oil saturation from 41% to about 20%. This is a noticeable reduction in the residual oil in place. The third row in Fig. 9 demonstrates the oil saturation at the end of the second CSWI process, and not much difference in oil saturation could be seen. The average residual oil saturation was about 17.3%, leading to an additional recovery of about 4% OOIP compared to the first stage. Therefore, it was not recommended to reach high differential pressure drop

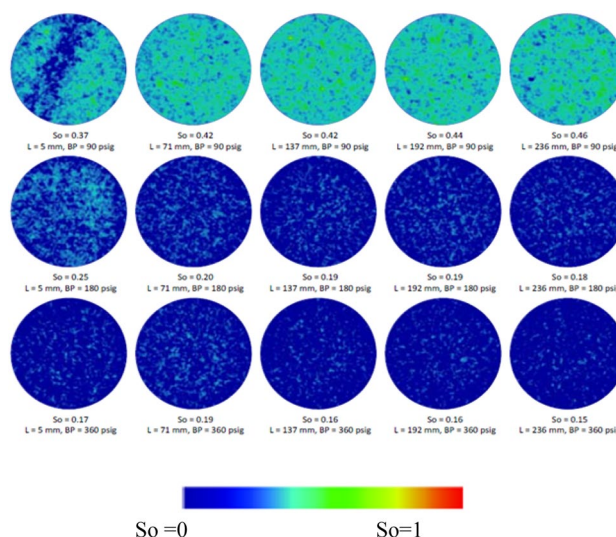


Fig. 9 CT images showing oil saturation distribution. First row: oil saturation distribution after imbibition; second row: first saturated brine flooding; third row: second saturated brine flooding (Alizadeh et al. 2011)

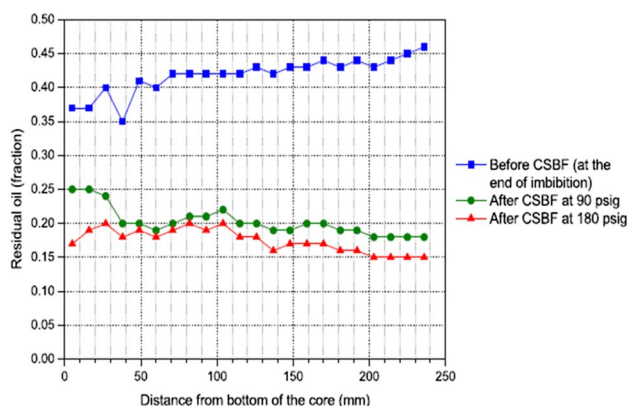


Fig. 10 Slice-averaged residual oil saturation (Alizadeh et al. 2011)

as it will not reduce oil saturation further. Another point to consider is the economical and operational difficulties that would increase by reaching high levels of CO_2 saturation in brine without significant advantage as the bulk of oil recovery takes place in the minor saturation levels of CO_2 in brine.

The slice-averaged oil saturation along the length of the core at the end of fresh brine imbibition and CO_2 -saturated brine flooding at 90 and 180 psig inlet pressures are shown in Fig. 10. The difference between the two top distributions again indicates the success of the first CSWI process in promoting higher oil recovery (Alizadeh et al. 2011).

In a recent work by (Kilybay et al. 2016), CSWI was performed on three carbonate core plugs from Abu Dhabi. The core plugs were flooded with three fluids in the following order: (1) seawater, (2) seawater with four times

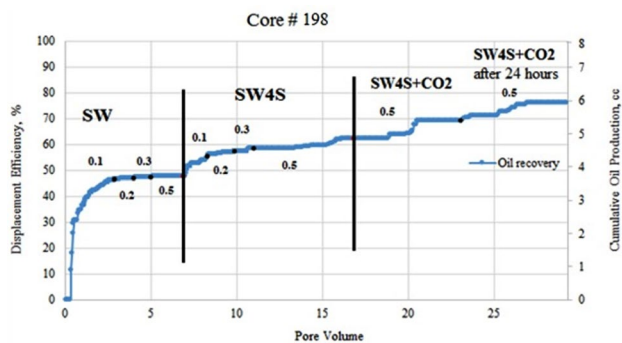


Fig. 11 Coreflood recovery results on secondary, tertiary and quaternary modes (Kilybay et al. 2016)

sulfate (SW4S) and (3) SW4S saturated with CO_2 (carbonated smart water). Results indicated that smart water saturated with CO_2 can recover a significant amount of immobile oil left after tertiary recovery with SW4S smart water. Whereas tertiary recovery with smart water injection resulted in 4.8–9.5% additional recovery, carbonated smart water injection in quaternary mode resulted in 5.7% to 13.6% additional oil recovery. This is attributed to the impact of CO_2 mass transfer from brine to oil causing viscosity drop, local flow diversion and trapped oil swelling. Carbonate dissolution and pore enlargement were also proven through NMR porosity and ICP-MS studies. Figure 11 exhibits the plots of displacement efficiency of brine flooding versus pore volume (PV) injected for three different core flooding experiments conducted. From this study, it is established that CSWI could be a novel and promising EOR technique among the latest EOR methods. It reduces the requirement for high-pressure system, the problems of gravity segregation and poor sweep efficiency.

Conclusion

This paper comprehensively reviewed CO_2 EOR techniques in its primitive and modern form including immiscible, miscible and dissolved forms. The salient features are:

- CO_2 injection in heterogeneous layered and fractured formations might lead to techno-economical failure due to poor sweep and increasing OPEX.
- CWI promoted higher oil recovery in the majority of experiments due to CO_2 mass transfer to oil which consequently resulted in lowering oil viscosity, swelling of trapped oil, enhanced flow diversion and improved sweep efficiency.

- CWI in high concentration did not provide a significant advantage over low concentration.
- In certain oil fields, CWI injection improved the well's water intake rate and was successfully deployed as a well stimulation technique.
- Results from carbonated smart water injection proved that an initial injection provided up to 51% additional recovery of residual oil in place, while a second injection only recovered 4%.
- A novel method of injecting carbonated water saturated with seawater that contains 4 times sulfate proved to be successful and provided up to 13% additional recovery in quaternary mode, emphatically proving the impact of CO_2 in reducing residual oil saturation even at low oil saturation.

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Compliance with ethical standards

Conflict of interest The authors declare no conflict of interest.

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