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Research Article

Enhanced Wettability Modification and CO_2 Solubility Effect by Carbonated Low Salinity Water Injection in Carbonate Reservoirs

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Carbonated water injection (CWI) induces oil swelling and viscosity reduction. Another advantage of this technique is that CO_2 can be stored via solubility trapping. The CO_2 solubility of brine is a key factor that determines the extent of these effects. The solubility is sensitive to pressure, temperature, and salinity. The salting-out phenomenon makes low saline brine a favorable condition for solubilizing CO_2 into brine, thus enabling the brine to deliver more CO_2 into reservoirs. In addition, low saline water injection (LSWI) can modify wettability and enhance oil recovery in carbonate reservoirs. The high CO_2 solubility potential and wettability modification effect motivate the deployment of hybrid carbonated low salinity water injection (CLSWI). Reliable evaluation should consider geochemical reactions, which determine CO_2 solubility and wettability modification, in brine/oil/rock systems. In this study, CLSWI was modeled with geochemical reactions, and oil production and CO_2 storage were evaluated. In core and pilot systems, CLSWI increased oil recovery by up to 9% and 15%, respectively, and CO_2 storage until oil recovery by up to 24% and 45%, respectively, compared to CWI. The CLSWI also improved injectivity by up to 31% in a pilot system. This study demonstrates that CLSWI is a promising water-based hybrid EOR (enhanced oil recovery).

1. Introduction

For decades, waterflooding has been deployed to pressurize depleted reservoirs for oil recovery when primary recovery becomes inefficient. After waterflooding, general recovery of oil is limited to approximately 30% because of factors such as heterogeneity, wettability, and unfavorable mobility. Therefore, economic and practical enhanced oil recovery (EOR) technologies have been developed to recover the oil remaining after waterflooding.

One such technique is carbonated water injection (CWI), which is CO_2-enriched waterflooding. When carbonated water (CW) comes into contact with oil, the CO_2 dissolved in CW moves into the oil, resulting in oil viscosity reduction and swelling [1]. These effects depend on the CO_2 solubility of brine. The effects of CWI have been investigated for decades. In the 1960s, commercial applications of CWI in Oklahoma, Texas, and Kansas demonstrated that CWI resulted in increased water injectivity and oil recovery [2]. In more recent studies, the CWI phenomenon has been visualized in glass micromodels [1, 3, 4]. These studies confirmed oil swelling, oil viscosity reduction, and the generation of CO_2-enriched gas during CWI. Moreover, Sohrabi et al. [1] and Kechut et al. [4] have conducted coreflooding experiments to observe whether oil production is enhanced and performed numerical simulations.

Another attractive EOR process is low salinity water injection (LSWI). Because conventional waterflood injects seawater or produced formation brine and LSWI uses diluted seawater, low saline brine indicates diluted seawater hereafter. Various laboratory studies have demonstrated enhanced oil recovery by LSWI through spontaneous imbibition and coreflooding tests for carbonate reservoir rocks [5–7]. This enhanced oil production was attributed to wettability modification of the rock surface. Many experimental techniques (interfacial tension measurement, contact angle measurement, NMR, ζ-potential measurement, imbibition testing, and coreflooding testing) have suggested mechanisms responsible for wettability modifications, such as calcite dissolution [7, 8], anhydrite dissolution [9], and surface charge change [10–12]. Although the underlying mechanism is still unclear, Yousef et al. [13] confirmed the reduction of residual oil saturation through the single well tracer test in field trials.

Both CWI and LSWI are advanced waterflooding processes that modify the ionic composition of brine through CO_2 dissolution or dilution. Since both methods are waterflooding-based EOR processes, it is relatively straightforward to combine CWI with LSWI. Recently, Kilybay et al. [14] have performed such a study in which they evaluated carbonated smart water flooding. They selected smart water as the sulfate ion-enriched brine through surface charge evaluation, rather than diluted seawater. However, only one of the three coreflooding experiments showed enhanced oil production, while the last two experiments showed negligible oil production [14]. Moreover, this approach did not account for any synergetic effects between smart water and carbonated brine.

While various studies have focused on single low salinity water injection or carbonated water injection in carbonate, hybrid EOR has not yet been investigated as a carbonated low salinity water injection (CLSWI) technique. Moreover, hybrid EOR potentially has additional synergetic effects because of the relationship between CO_2 solubility in brine and the low salinity of brine. This promising hybrid CLSWI technique is expected to yield oil viscosity reduction, oil swelling, and wettability modification effects, with the additional advantage of storing CO_2 through the solubility mechanism. While the hybrid technique is expected to enhance oil recovery and CO_2 storage, complex geochemical phenomena in the brine-rock-oil system underlie the CLSWI mechanism.

The objective of this study was to evaluate oil production in CLSWI considering the geochemistry of the brine-rock-oil system. The performance of CLSWI in oil viscosity reduction, oil swelling, and wettability modification was investigated and compared to those of seawater injection (SWI), carbonated seawater injection (CWI or CSWI), and LSWI. Additionally, CO_2 storage by the solubility trapping mechanism was investigated.

2. Basic Theories

In CLSWI, CO_2 moves into the oleic/gas phases to reach equilibrium at a specific temperature and pressure. This mass transfer across the interphase proceeds until the fugacity of CO_2 becomes equivalent in all existing phases. Li and Nghiem [15] have calculated the composition in each phase through the equation of state (EoS), considering Henry's law. The CO_2 solubility in the aqueous phase is determined from Henry's law and the CO_2 fugacity in the gas phase. The CO_2 fugacity in the gas phase is calculated from the EoS. Henry's law takes into account salinity by introducing a salting-out coefficient. This coefficient depends on temperature and salinity [16].

$$
\begin{aligned}
f_{CO_2,g} &= f_{CO_2,aq}, \\
f_{CO_2,g} &= H_{salt,CO_2} x_{CO_2,aq}, \\
\ln\left(\frac{H_{salt,CO_2}}{H_{CO_2}}\right) &= k_{salt,CO_2} m_{salt}, \\
k_{salt,CO_2} &= 0.11572 - 0.00060293T + 3.5817 \\
&\quad \times 10^{-6}T^2 - 3.7772 \times 10^{-9}T^3,
\end{aligned} \tag{1}
$$

where $f_{CO_2,j}$ indicates the CO_2 fugacity in phase j, j represents the gas and aqueous phases, H_{salt,CO_2} is Henry's constant of CO_2 in brine, $x_{CO_2,aq}$ is the molar fraction of CO_2 in the aqueous phase, H_{CO_2} is Henry's constant of CO_2 at zero salinity, k_{salt,CO_2} is the salting-out coefficient of CO_2, and m_{salt} is the molality of the dissolved salt.

CLSWI interrupts geochemical equilibrium in the formation of brine/rock system and establishes a new equilibrium state. In compositional simulations, Nghiem et al. [17] have introduced geochemical reactions and captured aqueous phase behavior. The first reaction associated with CLSWI is an aqueous reaction. The ionic concentration in the reaction is calculated when the ion activity product (IAP) is equivalent to the equilibrium constant at a specific temperature.

$$
\begin{aligned}
Q_\alpha - K_{eq,\alpha} &= 0, \\
Q_\alpha &= \prod_{k=1}^{n_{aq}} a_k^{\nu_{k,\alpha}},
\end{aligned} \tag{2}
$$

where α indicates the aqueous reaction, $K_{eq,\alpha}$ is the temperature-dependent equilibrium constant, Q_α is the IAP, k stands for the component, n_{aq} indicates the number of aqueous reactions, a_k is the ionic activity, and $\nu_{k,\alpha}$ are the stoichiometric coefficients of an aqueous reaction.

Geochemical reactions are determined by the effective ionic concentration, which is equivalent to ion activity, considering electrostatic interactions among ions in the aqueous phase. Ion activity is a function of activity coefficient and molality. The activity coefficient is determined by ionic strength, ion size, and temperature. Here, a modified Debye-Hückel model was used to calculate the ion activity coefficient at a specific temperature, as shown below:

$$
\begin{aligned}
a_k &= \gamma_k m_k, \\
\log \gamma_k &= -\frac{A_\gamma z_k^2 \sqrt{I}}{1 + \dot{a}_k B_\gamma \sqrt{I}} + \dot{B} I, \\
I &= \frac{1}{2}\sum_{k=1}^{n_{aq}} m_k z_k^2,
\end{aligned} \tag{3}
$$

where γ_k indicates the activity coefficient; m_k is the molality; A_γ, B_γ, and $\dot{B}$ are temperature-dependent coefficients; I indicates the ionic strength; $\dot{a}_k$ is the ion size parameter; and z_k represents the ionic charge.

Another geochemical reaction is mineral dissolution, otherwise known as precipitation. This reaction is characterized by nonequilibrium and slow kinetics, in contrast to aqueous reactions. Mineral dissolution is a rate-dependent

TABLE 1: Brine compositions [18].

Ion	FW (mg/L)	SW (mg/L)	LSW (mg/L)
Na^+	9,614.97	11,429.38	1,142.93
Ca^{2+}	320.36	429.60	42.96
Mg^{2+}	218.94	1,361.60	136.16
K^+		351.10	35.11
Ba^{2+}		0.01	
Sr^{2+}		8.37	0.83
Cl^-	15,117.25	20,040.00	2,004.00
SO_4^{2-}	550.63	3,500.00	350.00
HCO_3^-	1,135.90	47.58	4.75
pH	8.01	7.80	7.20

kinetic reaction, represented by the rate law, by which ion species are generated or consumed in brine. In mineral dissolution/precipitation, the generation/consumption rates of minerals are determined by the reaction rates, mineral reactive surface areas, and stoichiometric coefficients, as shown below:

$$r_\beta = \widehat{A}_\beta k_\beta \left(1 - \frac{Q_\beta}{K_{\mathrm{eq},\beta}}\right),$$
$$Q_\beta = \prod_{k=1}^{n_{\mathrm{aq}}} a_k^{v_{k,\beta}}, \tag{4}$$

where β defines the mineral, r_β is the reaction rate, k_β is the reaction rate constant, $\widehat{A}_\beta$ is the reactive surface area of a mineral, $K_{\mathrm{eq},\beta}$ is the solubility product constant at a specific temperature, Q_β is the IAP, and $v_{k,\beta}$ are the stoichiometric coefficients of a given mineral reaction.

Since mineral dissolution or precipitation provides or consumes ionic species in the aqueous phase, the rate of generation/consumption of a given ionic species is closely related to the mineral reaction, as shown in the following equation:

$$\gamma_{k,\beta} = v_{k,\beta} r_\beta, \tag{5}$$

where $\gamma_{k,\beta}$ indicates the consumption or production rate of a given ionic species in brine due to the mineral reaction.

3. Numerical Modeling

Numerical modeling of hybrid CLSWI has been conducted with GEM simulator, which was developed by CMG (Computer Modeling Group Ltd.). This simulation study was based on a coreflooding experiment by Gachuz-Muro and Sohrabi [18]. Specifically, the numerical 1-dimensional model describes the limestone core, which was discretized into 20 grid blocks. The total pore volume and oil volume were 15.55 and 10.55 cm^3, respectively, and the permeability and temperature were set to 19.4 md and 92°C, respectively. The target oil had an API gravity and viscosity of 14.12°API and 53,484.31 cp, respectively, at 20°C; the oil viscosity was approximately 111.4 cp at the reservoir conditions. The compositions of the formation brine, injected SW, and injected LSW are shown in Table 1. In SW, the low concentration of Ba^{2+} was neglected to improve numerical convergence. Geochemical reactions, including aqueous reactions and mineral reactions, were implemented into the flow simulation and are listed in Table 2.

First, the wettability modification effect was extracted from the LSWI experiment. Using the oil production data, history-matching was used with CMOST simulator to evaluate the modification of relative permeability curves and residual oil saturation. Next, CLSWI modeling was performed. The performance of CLSWI was assessed by comparing its results with those of continuous SWI, LSWI, and CSWI. To construct different saline CWI techniques (CSW and CLSW), geochemical analysis was used to estimate CO_2 solubility at specific temperature and pressure. The oil production from CLSWI was assessed in terms of wettability modification, oil swelling, and oil viscosity. Moreover, since CLSWI has the potential to capture CO_2 in the reservoir, the storage potential was evaluated by considering the solubility trapping mechanism.

4. Results

4.1. History-Matching Process: SWI and LSWI. History-matching was performed through CMOST to assess experimental oil production [18]. The injection scenario was designed with successive SWI following LSWI. The best history-matched model, shown in the red line, was used in the experiment (Figure 1). Regular seawater injection for the first cycle recovered up to 37% of the oil. Successive LSWI after SWI retrieved an additional 15% of the oil due to the wettability modification effect. This wettability modification effect is characterized by relative permeability change and residual oil saturation reduction, as shown in Figure 2. Residual oil saturation was modified with 4% reduction, after which the water endpoint was reduced by up to 20%, and the oil endpoint was increased by up to 4%. After LSWI, 48% of the OOIP (Original Oil in Place) remained in the reservoir due to unfavorable oil mobility. Based on this model, numerical modeling of CSWI and CLSWI was conducted, and the performances of both processes were compared.

Table 2: Geochemical reactions.

Aqueous reactions	$H^+ + OH^- \leftrightarrows H_2O$
	CO_2 (aq) $+ H_2O \leftrightarrows H^+ + HCO_3^-$
	$H^+ + CaCO_3 \leftrightarrows Ca^{2+} + HCO_3^-$
	$H^+ + MgCO_3 \leftrightarrows HCO_3^- + Mg^{2+}$
	$H^+ + NaCO_3^- \leftrightarrows HCO_3^- + Na^+$
	$CaHCO_3^+ \leftrightarrows Ca^{2+} + HCO_3^-$
	$MgHCO_3^+ \leftrightarrows HCO_3^- + Mg^{2+}$
	$NaHCO_3 \leftrightarrows HCO_3^- + Na^+$
	$H^+ + MgOH^+ \leftrightarrows H_2O + Mg^{2+}$
	$H^+ + NaOH \leftrightarrows H_2O + Na^+$
	$CaSO_4 \leftrightarrows Ca^{2+} + SO_4^{2}$
	$NaSO_4^- \leftrightarrows Na^+ + SO_4^{2-}$
	$MgSO_4 \leftrightarrows Mg^{2+} + SO_4^{2-}$
	$SrHCO_3^+ \leftrightarrows Sr^{2+} + HCO_3^-$
	$SrSO_4 \leftrightarrows Sr^{2+} + SO_4^{2-}$
Mineral dissolution/precipitation	Calcite $+ H^+ \leftrightarrows Ca^{2+} + HCO_3^-$
	Dolomite $+ 2(H^+) \leftrightarrows Ca^{2+} + 2(HCO_3^-) + Mg^{2+}$

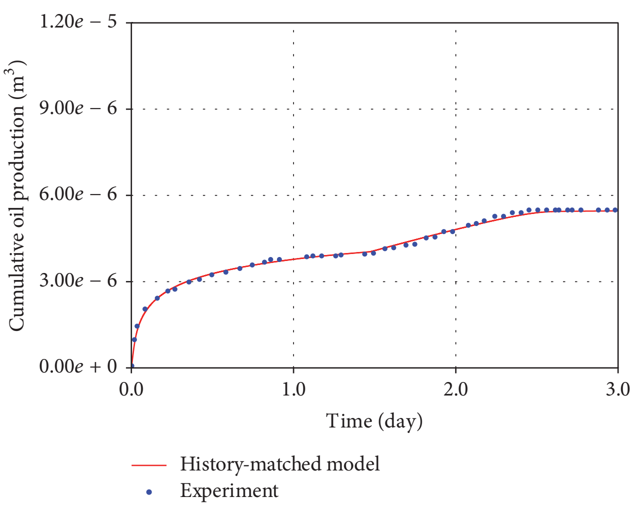


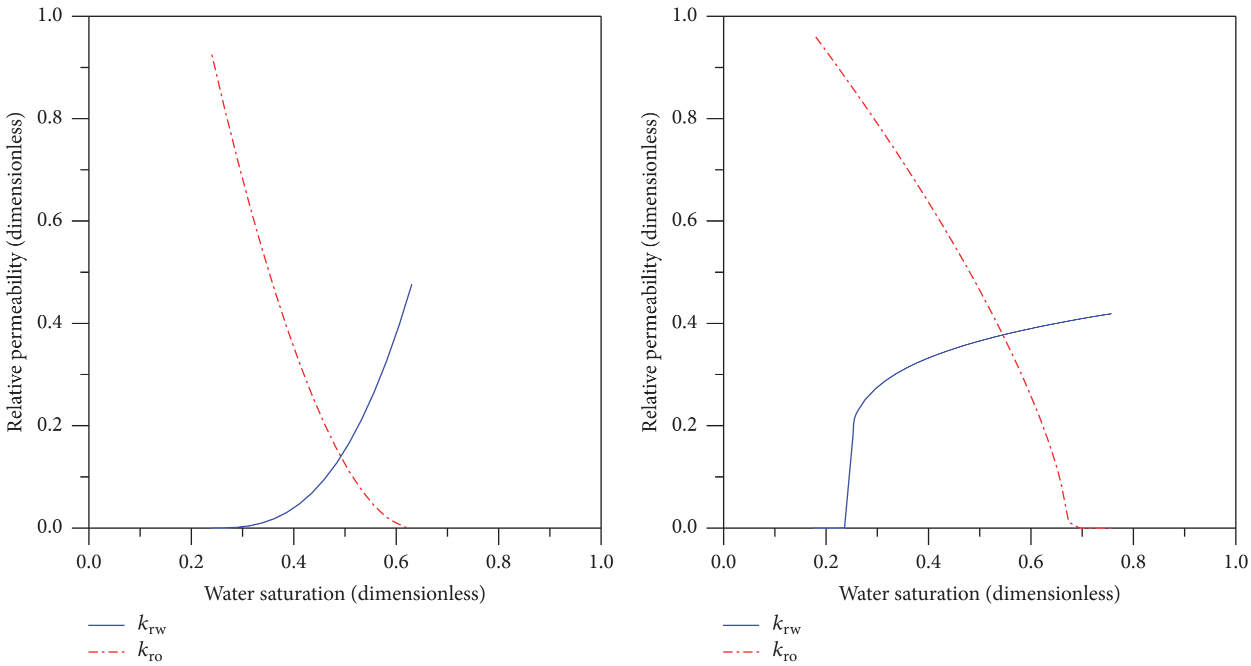
Figure 1: History-matched oil production in successive seawater injection and low salinity water injection.

4.2. Carbonated Water Injection: CSW and CLSWI. Geochemical analysis with PHREEQC (PH-Redox-EQuilibrium C programming language) simulator was used to evaluate CO_2 solubility in different brines. When the pressure increases, more CO_2 can be dissolved in the brine. Figure 3 illustrates the CO_2 solubility at the reservoir temperature and reveals salinity-dependent and pressure-dependent solubility. Both brines had higher solubility as the pressure increased. Of note, solubility was higher in LSW than in SW. SW and LSW have ionic strength as 0.4765 and 0.04934 mol/kg. At regular atmospheric conditions, as many as 9.74×10^{-3} and 1.07×10^{-2} moles of CO_2 could be dissolved into the SW and LSW, respectively. In high-pressure conditions (up to 10 atm), the dissolution increased up to 0.08913 and 0.1037 moles, respectively. The increased CO_2 solubility in low saline brine is a manifestation of the salting-out phenomenon, which predicts that low saline water is the optimum condition for constructing CWI.

To compare the performance of CLSWI to that of other techniques, continuous SWI, LSWI, and CSWI were also modeled. Figure 4 displays the oil recovery history for SWI, LSWI, CSWI, and CLSWI. SWI recovered about 40% of the oil after 30 PV (pore volume), while LSWI recovered up to an additional 11% due to the wettability modification effect. Mineral dissolution of Ca^{2+}, Mg^{2+}, and SO_4^{2-} were the major factors responsible for this wettability modification. Figure 5 depicts the ionic concentrations in the reservoir and the mineral dissolution. The ionic concentrations of Ca^{2+}, Mg^{2+}, and SO_4^{2-} after LSWI were less than 10% of the concentrations after SWI. Both SWI and LSWI dissolved calcite minerals, with LSWI dissolving 16% more calcite minerals than SWI.

In CSWI, 5% more oil was recovered compared to SWI (Figure 4). Interestingly, CSWI and SWI led to different Ca^{2+}, Mg^{2+}, and SO_4^{2-} concentrations in the reservoir. Specifically, CSWI resulted in slightly increased (2 to 3%) concentrations. The main reason for this difference is the 11% increase in mineral dissolution. These increases in the ionic concentrations of Ca^{2+}, Mg^{2+}, and SO_4^{2-} mean that wettability could potentially be modified, albeit by a relatively small increment. However, the oil swelling and viscosity reduction changes were significant. Figure 6 presents the CO_2 dissolution in oil, oil density, and oil viscosity at the center of the core. In CSWI, the CO_2 dissolved in brine was transferred into the oil to reach an equilibrium state. This dissolution involved oil swelling and reduced oil density by as much as 2.8%, while also reducing oil viscosity by as much as 75%.

CLSWI recovered 56.5% of the oil. This yield was higher than that achieved with SWI, LSWI, or CSWI (up to 16%, 5%, and 9%, resp.) (Figure 4). First, the performance of CLSWI was evaluated in terms of oil swelling, density, and viscosity. This evaluation was performed by comparing CSWI with CLSWI. As illustrated in Figure 6, CLSWI involves



(a) Seawater injection

(b) Low salinity water injection

FIGURE 2: Modification of relative permeability and residual oil saturation in successive seawater injection and low salinity water injection.

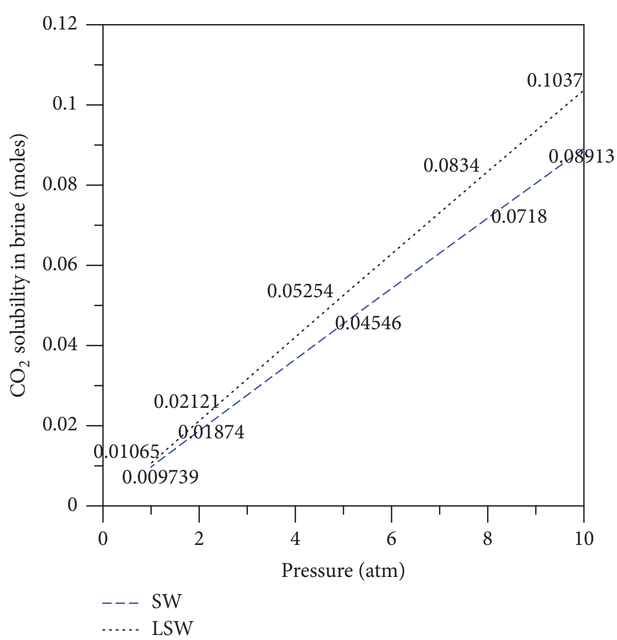


FIGURE 3: Pressure-dependent CO_2 solubility in brine.

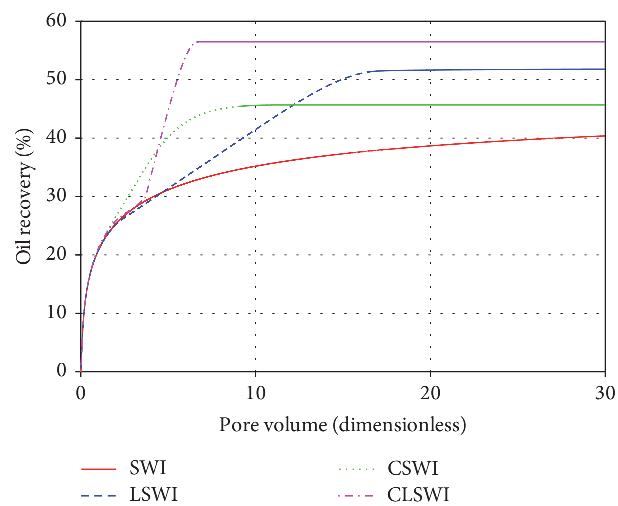


FIGURE 4: Cumulative oil recovery after seawater injection, low salinity water injection, carbonated seawater injection, and carbonated low salinity water injection.

mass transfer of CO_2 from brine into oil. This transport increases the CO_2 molar fraction in oil up to 0.148, which is as much as 8% higher than that in CSWI. Low salinity brine has CO_2 solubility by up to 15% higher compared to seawater due to the salting-out phenomenon (Figure 3). In these reservoir conditions, more dissolved CO_2 came out of the brine in CLSWI than in CSWI; this CO_2 dissolved into the oil. This enhanced dissolution resulted in greater reductions of oil density and viscosity, as shown in Figure 6. Next, the potential of CLSWI to modify wettability was evaluated and compared to that of LSWI. The slight changes in Ca^{2+}, Mg^{2+}, and SO_4^{2-} concentrations resulted from aqueous and mineral reactions. In addition, CLSWI yielded as much as 4% greater mineral dissolution, thereby providing more Ca^{2+}, Mg^{2+}, and SO_4^{2-} ions in the brine (Figure 5). This finding implies that CLSWI has the potential to improve wettability in terms of

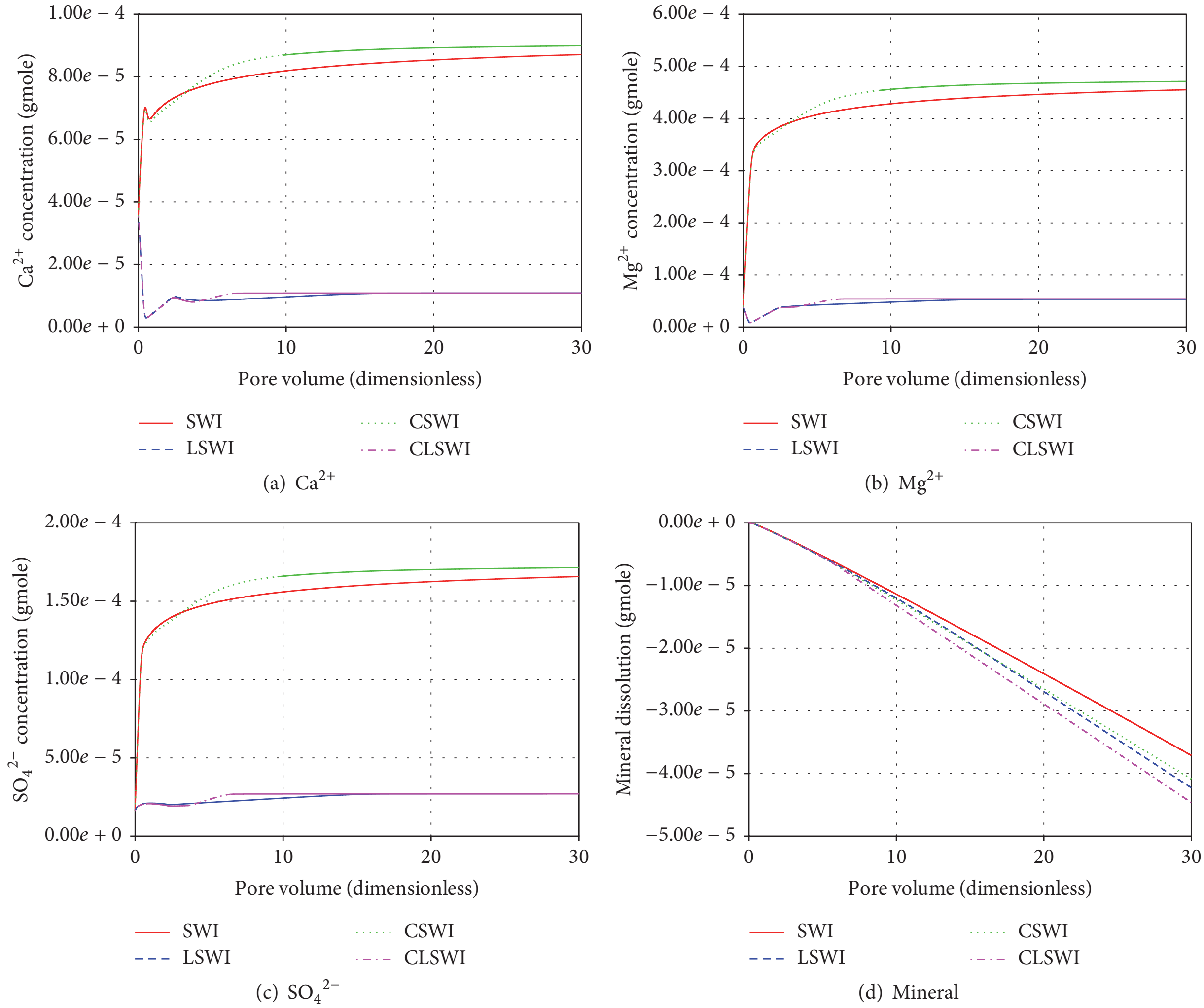


FIGURE 5: History of ionic concentrations (Ca^{2+}, Mg^{2+}, and SO_4^{2-}) and mineral dissolution in the reservoir after seawater injection, low salinity water injection, carbonated seawater injection, and carbonated low salinity water injection.

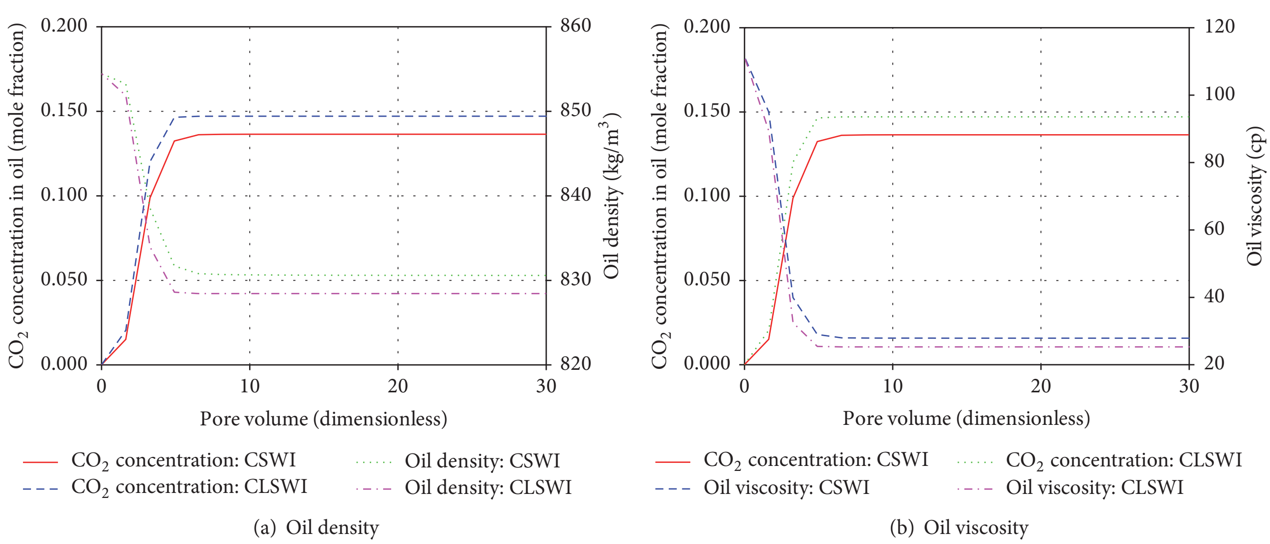
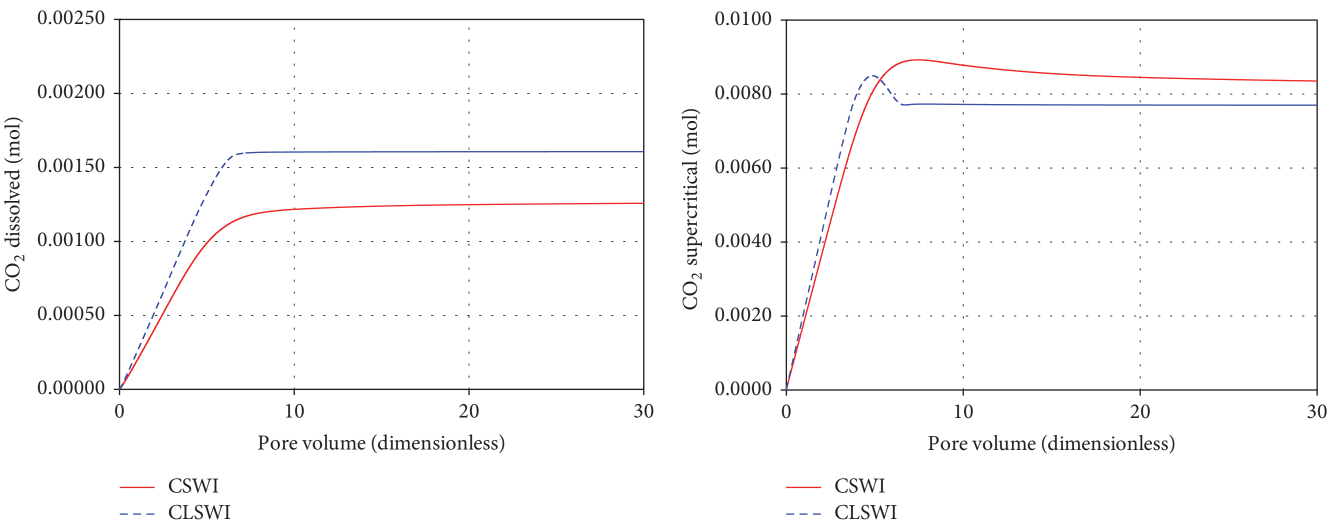


FIGURE 6: History of oil density and viscosity reduction for carbonated seawater injection and carbonated low salinity water injection.



(a) CO_2 dissolution (b) Super-critical CO_2

Figure 7: History of dissolved CO_2 concentration in brine and supercritical CO_2 after carbonated seawater injection and carbonated low salinity water injection.

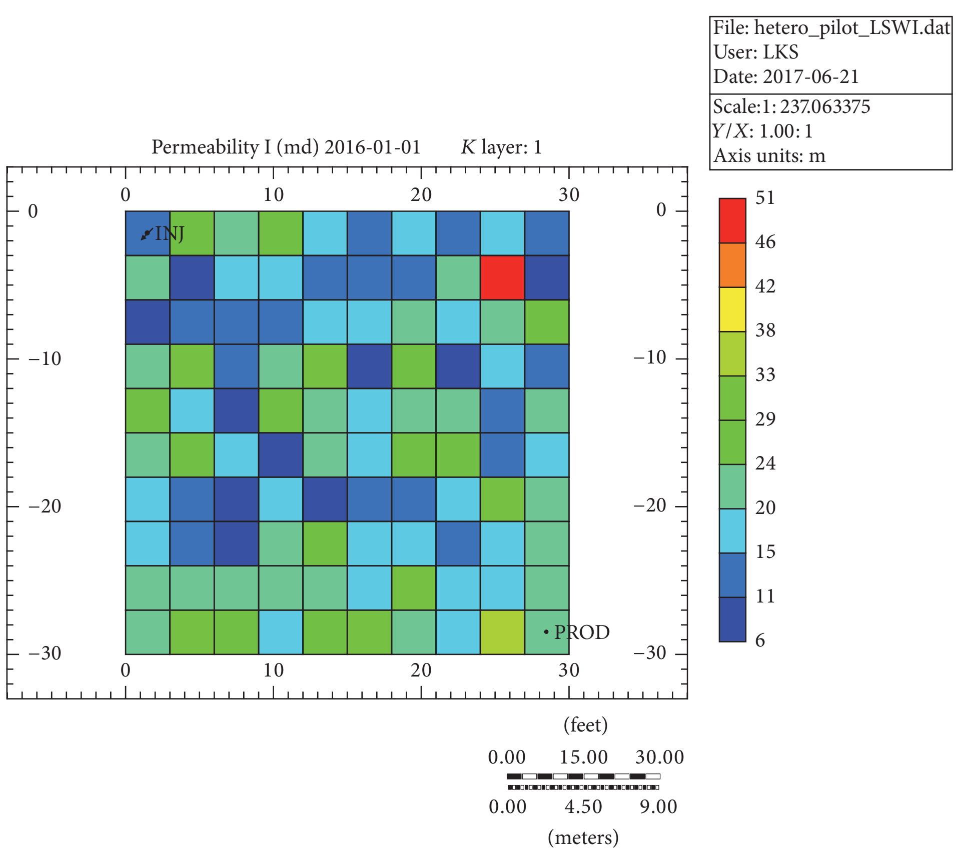
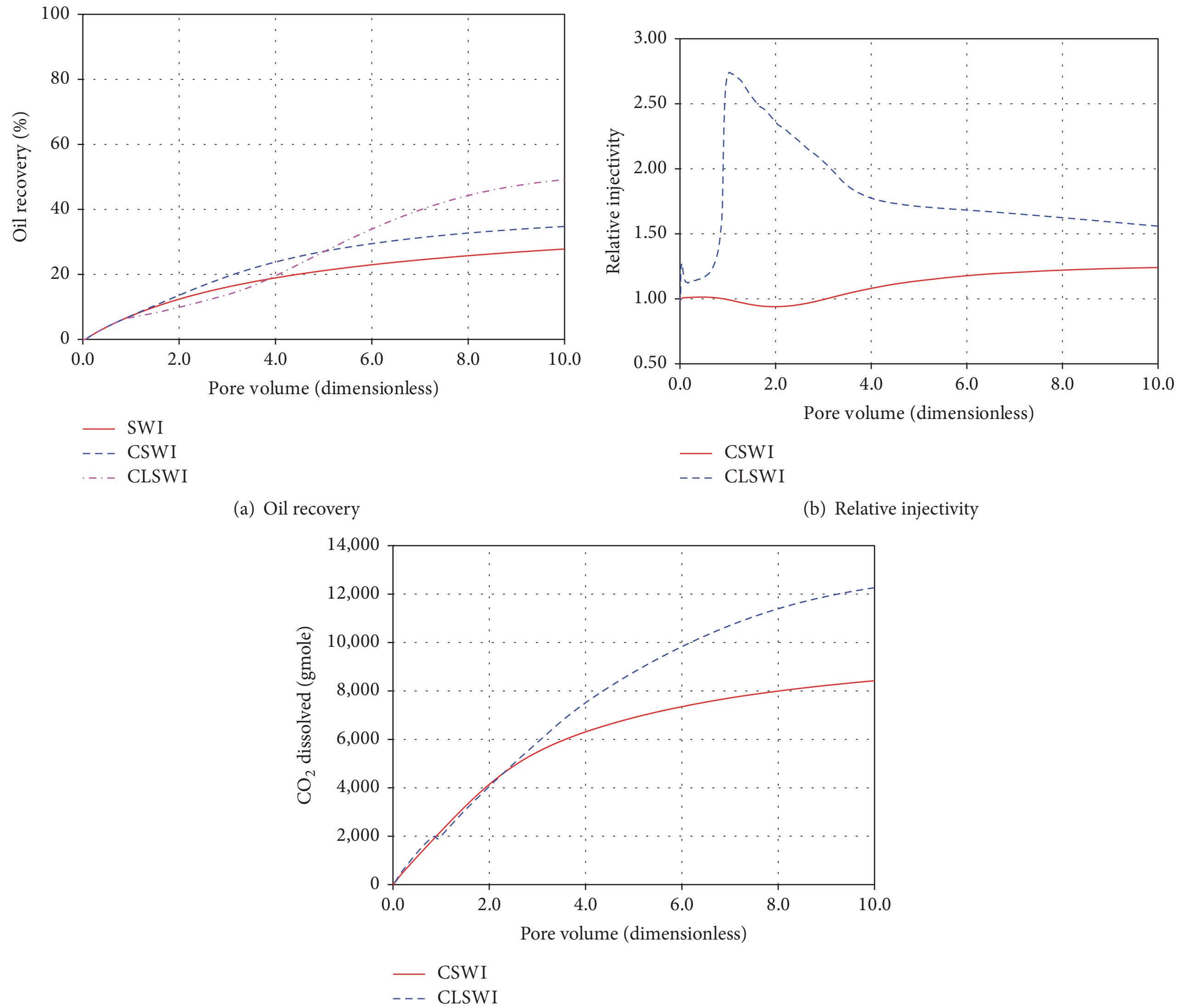


Figure 8: Heterogeneous permeability (md) of the reservoir.

geochemical evaluation. In summary, CLSWI has advantages compared to both CWI and LSWI and also improves oil recovery.

Conventional CSWI has the ability to store CO_2 by the dissolution mechanism. The promising hybrid CLSWI technique also captures stable CO_2 in the reservoir. After CLSWI, the in situ brine exists in a low saline condition, which is favorable for CO_2 solubilization due to the salting-out phenomenon. CLSWI demonstrated an enhanced capability for CO_2 storage compared to conventional CWI.



(a) Oil recovery

(b) Relative injectivity

(c) CO_2 dissolution

FIGURE 9: History of oil recovery, relative injectivity, and CO_2 storage after seawater injection, carbonated seawater injection, and carbonated low salinity water injection.

Specifically, the total amounts of CO_2 injected by CSWI and CLSWI were 6.25123×10^{-2} and 7.23759×10^{-2} moles, respectively. As shown in Figure 7(a), CSWI stably stored as many as 1.25×10^{-2} moles CO_2 in brine. Moreover, CLSWI captured up to 24% more CO_2 than CSWI. While up to 17% more CO_2 was injected in CLSWI, mass transfer of CO_2 into oil and brine was greater in CLSWI than in CSWI. Finally, CLSWI yielded as much as 13% lower supercritical CO_2, which corresponds to CO_2 not stored by the solubility mechanism, in the reservoir compared to CSWI (Figure 7(b)).

4.3. Pilot-Scaled Reservoir. The performance of CLSWI was also assessed in a 2-dimensional heterogeneous permeable reservoir (Figure 8), which had a pore volume of 1.8576×10^2 m^3. The average horizontal permeability of this reservoir was approximately 19.4 md, and the Dykstra-Parsons coefficient of the permeability was calculated to be 0.35. Figure 9(a) shows the oil recovery after injection of 10 PV, with SWI yielding 27.8% oil recovery. CSWI improved oil recovery by 7% compared to SWI due to the oil swelling effect and viscosity reduction. The viscosity reduction also contributed to the improved injectivity compared to SWI. The relative injectivity of CSWI, which is defined as the injectivity of CSWI divided by the injectivity of SWI, is shown in Figure 9(b). The injectivity of CSWI was improved by as much as 20% compared to SWI due to oil viscosity reduction. Hybrid CLSWI enhanced oil recovery by up to 22% compared to SWI; the hybrid approach showed as much as 15% more recovery than CSWI. This significant increase in oil recovery is mainly attributed to the wettability modification effect and oil viscosity reduction. In terms of injectivity, the wettability modification effect introduced a slight increase in injector BHP. However, the oil viscosity reduction by CO_2 dissolution into the oil significantly improved injectivity. At injection of 1 PV, CLSWI exhibited up to 170% greater injectivity than SWI. After injection of 10 PV, 55% improvement was observed, still as much as 31% higher than that of CSWI. Moreover, 45% more CO_2 was stored in CLSWI than in CSWI due to the solubility mechanism (Figure 9(c)).

5. Conclusions

This study assessed the performance of hybrid CLSWI in terms of oil production and CO_2 storage. The process behind hybrid CLSWI depends on geochemical reactions in the brine/oil/rock system. This study, which incorporated geochemical reaction modeling, demonstrated the promising potential of CLSWI. Compared to LSWI, CLSWI resulted in negligible changes of ionic concentrations (Ca^{2+}, Mg^{2+}, and $SO_4{}^{2-}$) and 4% greater mineral dissolution. These geochemical reaction results explain why CLSWI has the potential to introduce the wettability modification effect, similar to LSWI. Additionally, CLSWI yielded enhanced oil swelling and oil viscosity reduction compared to CSWI. Because of the salting-out phenomenon, CLSWI carried up to 15% more dissolved CO_2 into the reservoir and transferred more CO_2 from brine into oil, thereby reducing oil viscosity. These results clearly demonstrate that CLSWI yielded up to 31% improved injectivity compared to CSWI in the pilot-scale reservoir. The synergetic effects of wettability modification, oil swelling, and oil viscosity reduction in hybrid CLSWI contributed to enhanced oil recovery (up to 9% and 15% more in core-scaled and pilot-scaled systems, resp.). Along with the enhanced oil recovery, CLSWI yielded enhanced CO_2 storage by the solubility mechanism. Specifically, CLSWI captured more CO_2 than CSWI (by up to 17% and 45% more in core-scaled and pilot-scaled systems, resp.) because of the salting-out phenomenon. In summary, hybrid CLSWI is a promising EOR method for improving oil recovery, injectivity, and CO_2 storage. In addition to enhanced oil production, additional direct economic benefit via CLSWI is smaller amount of CO_2 to be required than CO_2 EOR. Future value of CO_2 emission reduction considering carbon tax is also expected via hybrid EOR technology.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

Acknowledgments

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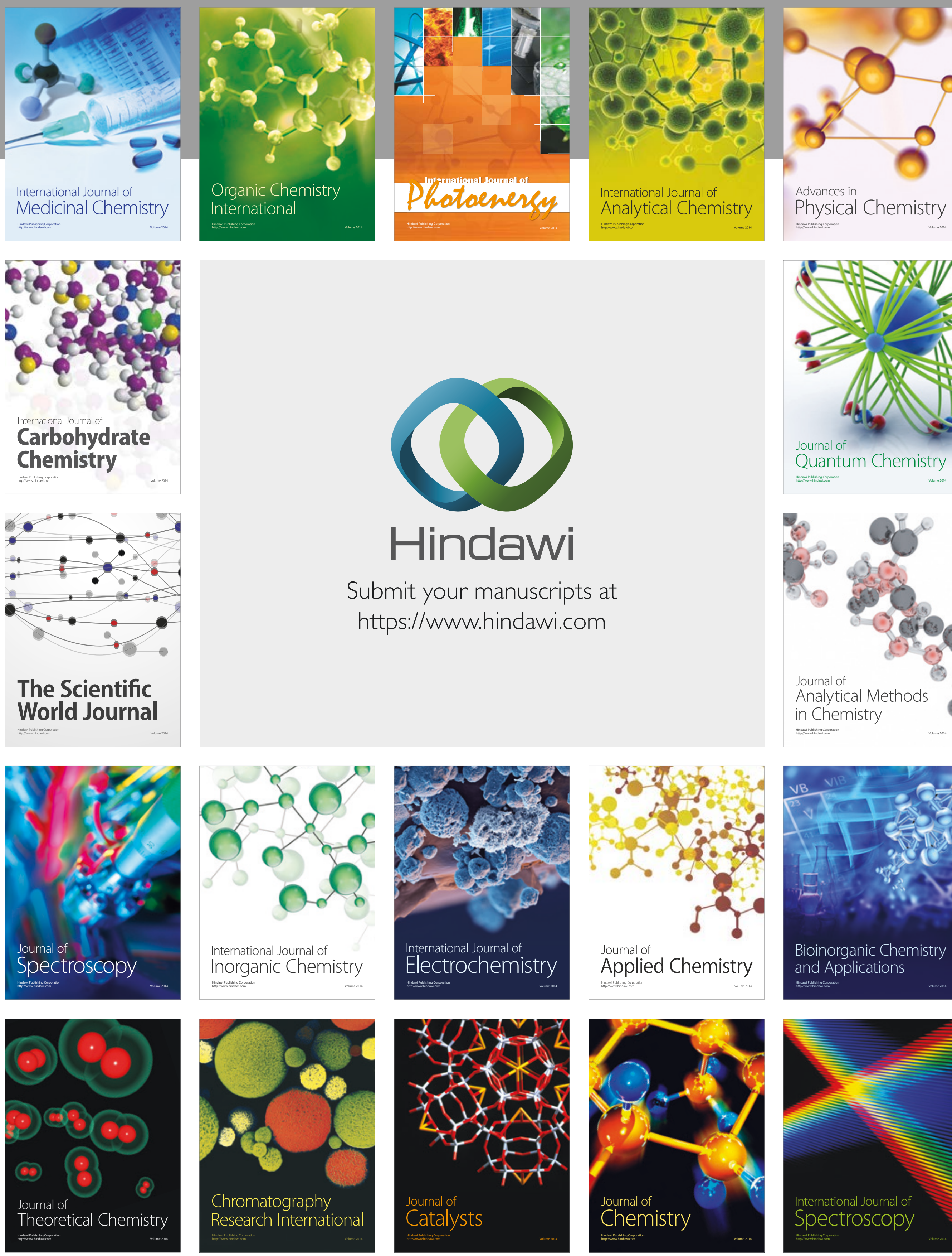
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