

Insights into Low Salinity Water Flooding

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ABSTRACT

In our previous work [1], we examined the potential of divalent cations (Mg^{2+} and Ca^{2+}) in formation water (FW) for low-salinity (LS) EOR effect, where the increase in divalent cations in FW lowered the impact of LS water EOR.

In this paper, we demonstrate the importance of the same divalent cations in the injected water (both FW and LS water). We also try to relate the percentage of the divalent cations in the injected water to that in the FW to engineer the optimum concentration of the injected water and obtain the maximum oil recovery from sandstone reservoirs.

Berea sandstone cores were successfully flooded with FW and LS water at 90°C. While injecting both brines, samples of the effluent were analyzed for pH. Oil recovery experiments with a double Ca^{2+} and Mg^{2+} concentration showed a lower LS water effect, inferring that the cores became more water-wet; however, the LS water effect was much greater when the amount of Ca^{2+} and Mg^{2+} in the HS water was decreased by half. The results of this work relate oil recovery with LS water chemical compositions, temperature, ion exchange, and pH.

INTRODUCTION

After 20 years of researching the mechanisms of LS water flooding, the mechanism of improved recovery using LS waters is still a topic of debate. However, the LS water flooding was used as an EOR technique and the improved oil recovery was 2-40% of the original oil in place (OOIP) [2].

The experimental observations of Tang and Morrow [3] for LS water flooding set out conditions for how LS water works. The conditions were: (1) the crude oil must contain acid and base numbers and (2) sandstone should contain clay such as illite and kaolinite. After several years, McGuire [4] and Lager and Webb [5] added another condition, which was that divalent cations must be present in the FW.

The second condition of Tang and Morrow was debated after the investigations of Al-Saedi and Brady [6] and Sohrabi [7]. The observations from chromatographic columns of quartz showed an increase in the acetate detachment from the quartz

surface [6]. The oil recovery observations from the quartz column supported the proposed mechanism (the clay is not essential) [8].

Lager and Webb [5] examined the effect of LS water during brine injection into a sandstone oil reservoir that had an identical amount of Mg^{2+} in the injected brine and formation water. The observation from the experiment was that less Mg^{2+} was produced in the effluent than the Mg^{2+} in the formation water due to the chemical reaction. Ca^{2+} has the same behavior. When Ca^{2+} and Mg^{2+} are hydrated in water, the reactivity of both increases with temperature and decreasing the desorption rate [9].

Many attempts and efforts have been presented in previous years to understand the mechanisms of the LS water flooding and design an optimum recipe for the injected LS water.

Fines migration was one such mechanism [3]. Austad [10] attributed the effect of LS water to the organic materials desorption from the clay surfaces. We investigated the desorption of organic materials for free-clay sandstone and rich-clay sandstone and found that the clay is not essential for observing LS EOR effect [6-8].

The quartz surfaces and carboxylate are both negatively charged, so carboxylate should be repelled from the quartz surface unless it forms a positively charged $-COOCa^+$ group, is able to bridge with the quartz surface, and the reaction $>SiO^- + Ca^{+2} + COO^- \leftrightarrow >SiOCaCOO$. The results suggest that when LS water invaded the quartz, the reaction above moved from right to left because of decreased Ca^{+2} levels [6]. The other mechanism was suggested by Lager and Webb [2], which was multicomponent ion exchange between the injected LS water and the porous media such as Ca^{2+} on the minerals surface exchanging with H^+ from the injected water. Many other mechanisms were proposed in the literature, such as mineral dissolution [11], interfacial tension reduction [4], double-layer expansion [12], mixed-wet particle release [3], and salt-in effect [13].

In this paper, series of Berea sandstone cores were flooded with different ionic concentrations of LS water at reservoir temperature to find an optimum design for LS water and in turn to attain a maximum oil recovery.

Methodology

Materials. Reagent-grade salts were received for this study. The brines were prepared by dissolving the salts in deionized water. The brine compositions are listed in Table 1. A crude oil from a Kansas oil field was used. The crude oil was diluted in the volume ratio 40/60 heptane/crude oil. The crude oil was then filtered through a 4.5 μm Millipore filter. No precipitation of asphaltenes was observed after diluting with heptane. The viscosity of the oil was 14 cP. at 20°C, the density is 0.815 gm/cc at 20°C TBN is 1.14 mg KOH/g, TAN is 0.66 mg KOH/g. Heptane was delivered by Fisher Scientific with purity 99%, density 0.6838 g/cm³ at 20°C, and dynamic viscosity 0.42 mPa.s at 20°C. The experimental setup is shown in Figure 1.

Core Handling. The cores were taken from a Berea sandstone block, which had identical petrophysical properties. The cores were dried overnight at 90°C. The cores were then evacuated for a day and saturated with FW under vacuum for another day. The porosity was calculated from the weight difference. The cores were mounted in the core holder for permeability measurements. The cores were flooded with 2 PV of crude oil (both directions) to initiate S_{wi} . The cores were pre-aged in the crude oil for three weeks at 90°C. The S_{wi} were 36.34%, 34.48%, 31.37%, and 35.58% for Core1-1, Core1-2, Core2-1, and Core2-2, respectively.

Core Flooding. The cores were mounted in the core holder and left overnight for thermal equilibrium purpose. The Berea sandstone cores were flooded with 2 PV FW (96,100 ppm) as a secondary flooding, and then 2 PV LS water (4000 ppm) was injected for the tertiary stage at a constant rate. The flow rate was 0.5 ml/min. While injecting brines, samples of the effluent were analyzed for pH. The experiments were conducted as follows:

1. Core1-1 was saturated with FW containing 90 mmole Mg^{2+} , and then flooded with the same FW followed by LS water containing 3 times diluted Mg^{2+} of the Mg^{2+} in FW.
2. Core1-2 was saturated with FW containing 90 mmole Mg^{2+} , and then flooded with the same FW followed by LS water containing 10 times diluted Mg^{2+} of the Mg^{2+} in FW while keeping the salinity of the LS water the same as in Core1-1 by adding NaCl.
3. Core2-1 was saturated with FW containing 90 mmole Ca^{2+} , and then flooded with the same FW followed by LS water containing 3 times diluted Ca^{2+} of the Ca^{2+} in FW.
4. Core2-2 was saturated with FW containing 90 mmole Ca^{2+} , and then flooded with the same FW followed by LS water containing 10 times diluted Ca^{2+} of the Ca^{2+} in FW while keeping the salinity of the LS water the same as in Core2-1 by adding NaCl.

The salinity of LS water was the same for all LS water used in these experiments (4000 PPM). The FW salinity was also the same for all experiments (96100 PPM).

Results and Discussion

Numerous field pilots and laboratory works have been conducted and provided optimistic oil recovery improvement when injecting LS water into reservoirs and outcrop sandstone [2, 14, 15, and 16]. The incremental oil recovery ascribed to the mechanisms was described earlier in the introduction. The chemical composition of the injection brines was carefully dealt with by Morrow et al. [3, 17, 18, 19, and 20].

In previous work [1], we investigated the role of the divalent cations (Ca^{2+} and Mg^{2+}) in the FW on the LS EOR and found that the role of the Mg^{2+} in FW has a greater impact than the Ca^{2+} even at high concentrations. As the concentration of the divalent cations increases in the FW, the sandstone turned into more water-wet and less LS EOR effect was observed.

In the present study, the focus was on the divalent cations in the injected LS water. The oil recovery results have been discussed in relation to the concentrations of the injected divalent cations.

1. Oil Recovery by LS water containing Mg^{2+} into a core saturated with Mg^{2+}

The outcrop core1-1 was sequentially flooded with FW and LS water at 90°C. No increased oil recovery was observed during LS water flooding ($d_{3Mg^{2+}}$) after core1-1 was flooded in secondary stage with FW. The ultimate oil recovery remained constant at 52.5% OOIP (figure 2).

The measurements of the pH were logged for the FW and LS water injections. The pH reading for FW effluent was 6.63, which must be sufficiently low to promote adsorption of polar components onto the sandstone surface [21].

The injection pressure was 37 psi during the FW flood. The LS water injection pressure decreased to 32 psi (figure 2).

When switching from FW to LS water, the pH of the LS water effluent increased to 7.15, which was small pH increment due to the high concentration of Mg^{2+} in the injected LS brine demonstrating very low wettability alteration. According to Lager and Webb [5] and Brady and Morrow [22], the difference in upward shift in effluent pH between HS and LS water is traditionally ascribed to the exchange of H^+ for divalent cations on clay surfaces.

Our previous work showed a similar attitude on both free-clay sandstone and rich-clay sandstone [6]. More water-wet sandstone would be expected due to that pH jump [10]. It seems the core wettability has not been altered by the injected LS water because of the high concentration of the Mg^{2+} . Mg^{2+} was responsible for the low pH in the LS water effluent, an in turn, no additional oil recovery was obtained.

Core1-2 was flooded the same way as in core1-1 but with $d_{10Mg^{2+}}$ LS water. As pointed previously, core1-1 and core1-2 were both saturated with FW containing 90 mmole Mg^{2+} .

The oil recovery during FW forced imbibition reached a plateau at 52.2% OOIP. The oil recovery was similar to core1-1 because the cores were similar petrophysically, and identical procedures were used. Upon switching to LS water, the incremental oil recovery was 1.5% of OOIP. Diluting the Mg^{2+} 10 times in the injected LS water improved the oil recovery from 0% to 1.5%. The Mg^{2+} was depleted in the injected LS water, and an additional 4% of OOIP was observed (figure 2). The initial pH of the FW was 6.83, and the pH increased to 8 when switching to LS water, which was significantly higher than for the core1-2. The pH during FW flooding providing a favorable environment for creating mixed-wet media [23]. The pressure profile had similar behavior to that in core1-1.

2. Oil Recovery by LS water containing Ca^{2+} into a core saturated with Ca^{2+}

The systematic study performed by Aghaeifar et al. [23] developed the relationship between the formation salinity and the LS EOR effect. When the reservoir core was pre-aged and flooded with the FW (salinity was 200,000 ppm, 640 mmole Ca^{2+}), no LS EOR effect was observed. When the FW salinity was reduced to 22,000 ppm (3.5 mmole Ca^{2+}), a significant LS water EOR effect was observed.

Our observations [14] were in line with Aghaeifar et al. [23] when we kept the salinity at the same level (~100,000 ppm), but when the concentration of Ca^{2+} was doubled (from 89 to 178 μmole), more water wet conditions was observed, and we observed a lower LS EOR effect. It therefore appears that the more Ca^{2+} is present in the FW, the less secondary and tertiary oil recovery was observed [1]. In this section, we examined the concentration of the injected Ca^{2+} into sandstone on LS EOR effect. As pointed out previously, core2-1 and core2-2 were saturated in FW containing 90 mmole Ca^{2+} , and the rest was NaCl providing a 96,100 ppm salinity. The FW salinity is consistent for all FW used in this work.

The oil recovery during secondary flooding with FW reached the ultimate recovery plateau of 43.25 % OOIP, which was less than the ultimate recovery of core1-1 (aged in 90 mmole Mg^{2+}), which was 52.5% of OOIP, indicating that abundance of Mg^{2+} in the injected FW is more favorable than the Ca^{2+} . Upon switching to LS water flooding, the improved oil recovery was 2.7% of OOIP, which was greater than Core1-1 (0% OOIP) and Core1-2 (1.5% OOIP), indicating that the abundance of Ca^{2+} in the injected LS water is more favorable than the Mg^{2+} .

The FW effluent pH was around 7, while it jumped up to 8.27 after injecting 2 PV LS water, indicating increased cation exchange occurred between Ca^{2+} and H^+ , and in turn, more polar component desorbed from the minerals surface. As a result, the wettability altered towards more water-wet condition and more oil recovery occurred during LS water flooding.

The injection pressure during FW flooding jumped to 134 psi. This high-pressure behavior is explained by our previous work with kaolinite [6].

Kaolinite is not a swelling clay; the high Ca^{2+} concentration led to “edge to face” agglomeration of the kaolinite plates to form higher volume assemblages. This has led to reduced permeability and increase in pressure. The more LS water flooded, the more agglomerated Ca^{2+} flushed out the core until the pressure stabilized at 87 psi during LS water flooding (figure 3).

The injection pressure for core2-2 was much less than in core1-1 because the Ca^{2+} was diluted 10 times, but it still doubled the pressure in both core1-1 and core1-2 due to the agglomeration Ca^{2+} effect under the kaolinite layers. The ultimate oil recovery was similar to core 2-1, which was 44.7% of OOIP after injecting 2 PV of FW.

The improved oil recovery from flooding with low salinity water was higher than the other cores at 5% of OOIP. It appears decreasing the amount of Ca^{2+} in the LS injection water has more significant impact than decreasing the Mg^{2+} concentration; Reducing Mg^{2+} (10 times) in the LS water provided a 1.5% improved oil recovery, while it improved to 5% of OOIP when reducing the Ca^{2+} 10 times.

Literature has shown that it is better to deplete both Ca^{2+} and Mg^{2+} in the injected LS water, as we observed in previous work [1], but it seems difficult to have a cost effective solution, especially offshore since natural sources of LS water flooding are typically rivers, lakes or aquifers and offshore solutions have to be engineered. For that reason, we should pay attention to the concentrations of divalent cations in the injected FW and LS water.

The observations of this work indicated that the Mg^{2+} could be preferred for secondary oil recovery and should be lowered as much as the technology can for the LS water. For example, when core2-1 was pre-aged in Ca^{2+} only, the secondary oil recovery was lower than that in the core containing Mg^{2+} only (core1-1).

In the previous work [1], the observations were that the existence of Mg^{2+} in the FW is favorable for more secondary and tertiary oil recovery. The core was pre-aged in FW containing 89 mmole Mg^{2+} and flooded with the same FW for the secondary stage. The oil recovery was 50% of OOIP by FW flooding, and it was 17.5 % of OOIP after flooding the core with 0 mmole divalent cations (1182 ppm salinity, NaCl only) [1].

In the present study, the same FW was used, but the LS water contained 10 times diluted Mg^{2+} of the Mg^{2+} in the FW and the secondary recovery was 52.5%, while the tertiary recovery was 0%. It is worth mentioning that the same materials and same experimental procedures were conducted for this work and the previous work [1].

The Ca^{2+} concentration, on the other hand, was observed to be favorable for the LS water flooding, but at lower concentrations. In the same study [1], we observed the presence of Ca^{2+} in the FW reduce the LS EOR effect. The LS water contained no divalent cations. Doubling the concentration of Ca^{2+} from 89 mmole to 178 mmole in the FW reduced the secondary oil recovery by 10% of OOIP and reduced the recovery by LS from 5% to 1% of OOIP. That means the abundance of Ca^{2+} in the FW could affect the oil recovery.

In the current study, the concentration of the Ca^{2+} was constant to 90 mmole in the FW, and the LS water contained 3 times and 10 times Ca^{2+} diluted of the Ca^{2+} in the FW of the core2-1 and core2-2. The results showed that diluting the Ca^{2+} 10 times in LS water improved the recovery from 2.7% to 5% of OOIP, meaning that the oil recovery was increased by approximately twice. Even diluting the Ca^{2+} 3 times in the LS water was better than diluting the Mg^{2+} 10 times, showing the important role of Ca^{2+} in the LS water. It is obvious that the ultimate oil recovery for both core1-1 and core1-2 was greater than for core2-1 and core2-1, confirming that the Mg^{2+} is preferable for secondary oil recovery when it is present in the FW. It was also

observed that as the concentration of the divalent cations decreases in the injected LS water as the pH of the LS water effluent increases. It may allow for an appropriate environment for cation exchange.

It is worth mentioning that it is complicated in the oil field to see a big jump of the pH as can be observed in the laboratory experiment because of many factors such as rock, dissolved gases and oil component buffering.

CONCLUSION

In this paper, we investigate the role of divalent cations in FW and LS water flooding in sandstone. The general conclusion can be drawn as follows:

1. When Mg^{2+} exists in the LS water, there is no oil recovery improvement during LS water flooding. There is no pH jump. It seems Mg^{2+} disrupts LS water EOR effect. Abundance of Mg^{2+} in the injected LS water could provide 0% in oil recovery improvement, but diluting the Mg^{2+} to 10 times could improve the oil recovery.
2. The experiments showed that Mg^{2+} is favorable for secondary oil recovery when the Mg^{2+} is presenting the FW and the injected FW during secondary flooding, while Ca^{2+} is favorable for LS water flooding even though the Ca^{2+} is presented in the FW, which is considered unfavorable to present in the FW for LS EOR affect.
3. The abundance of Ca^{2+} in the injected LS water could improve the oil recovery; however, diluting the Ca^{2+} in the injected LS water is required for extra oil recovery improvement.

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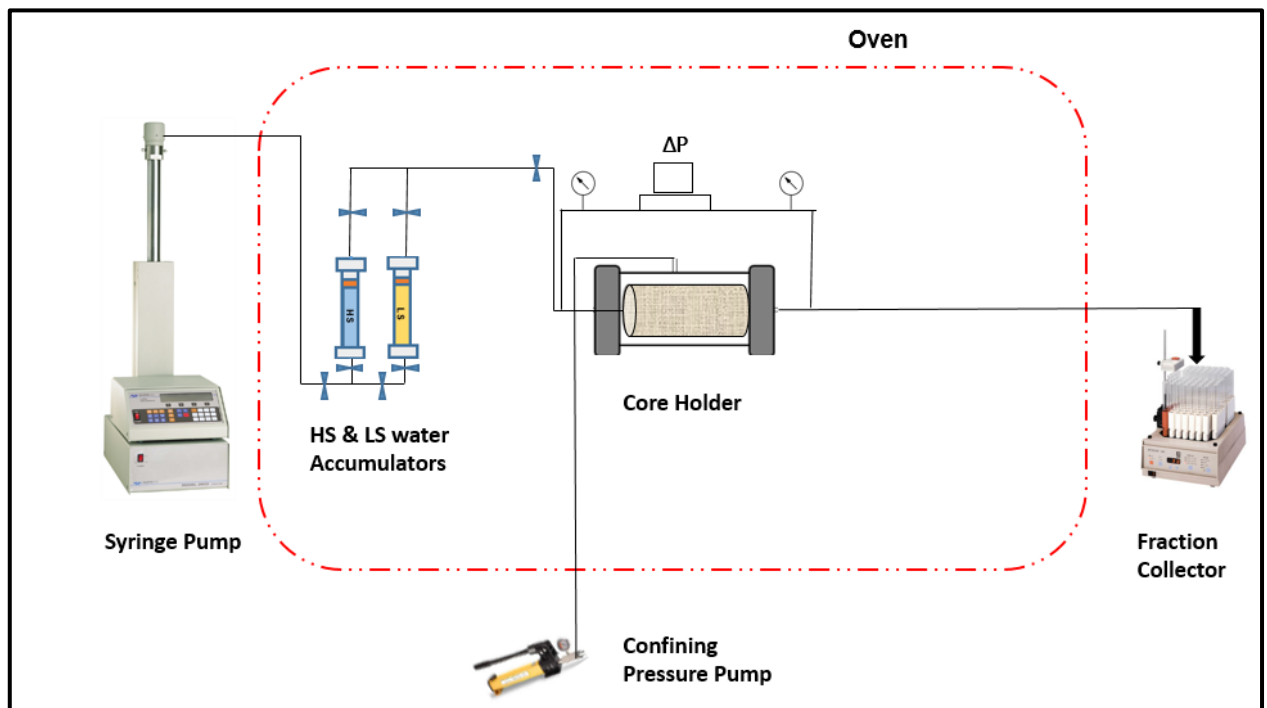


Figure 1. CoreFlood setup

Table 1. Core properties and water description

| Core | Quartz, % | Kaolinite, % | Diameter, cm | Length, cm | K, md | ϕ , % | Ca ²⁺ in FW (mmole) | Mg ²⁺ in FW (mmole) | Ca ²⁺ in LSW (mmole) | Mg ²⁺ in LSW (mmole) | NaCl in FW (mmole) | NaCl in LSW (mmole) |
|----------|-----------|--------------|--------------|------------|-------|------------|--------------------------------|--------------------------------|---------------------------------|---------------------------------|--------------------|---------------------|
| Core#1-1 | 95 | 5 | 2.54 | 14.77 | ~100 | ~21 | 0 | 90 | 0 | 30 | 3080 | 39 |
| Core#1-2 | | | | 14.67 | | | 0 | 90 | 0 | 9 | | 107.5 |
| Core#2-1 | | | | 14.78 | | | 90 | 0 | 30 | 0 | 2950 | 23 |
| Core#2-2 | | | | 14.67 | | | 90 | 0 | 9 | 0 | | 102.6 |

Table 2. Oil recovery results for both FW and LS water flooding

| Core | Ca ²⁺ in FW (mmole) | Mg ²⁺ in FW (mmole) | Ca ²⁺ in LSW (mmole) | Mg ²⁺ in LSW (mmole) | Secondary Oil Recovery by FW, % | Tertiary Oil Recovery by LSW, % |
|----------|--------------------------------|--------------------------------|---------------------------------|---------------------------------|---------------------------------|---------------------------------|
| Core#1-1 | 0 | 90 | 0 | 30 | 52.5 | 0 |
| Core#1-2 | 0 | 90 | 0 | 9 | 52.2 | 1.5 |
| Core#2-1 | 90 | 0 | 30 | 0 | 43.25 | 2.7 |
| Core#2-2 | 90 | 0 | 9 | 0 | 44.7 | 5 |

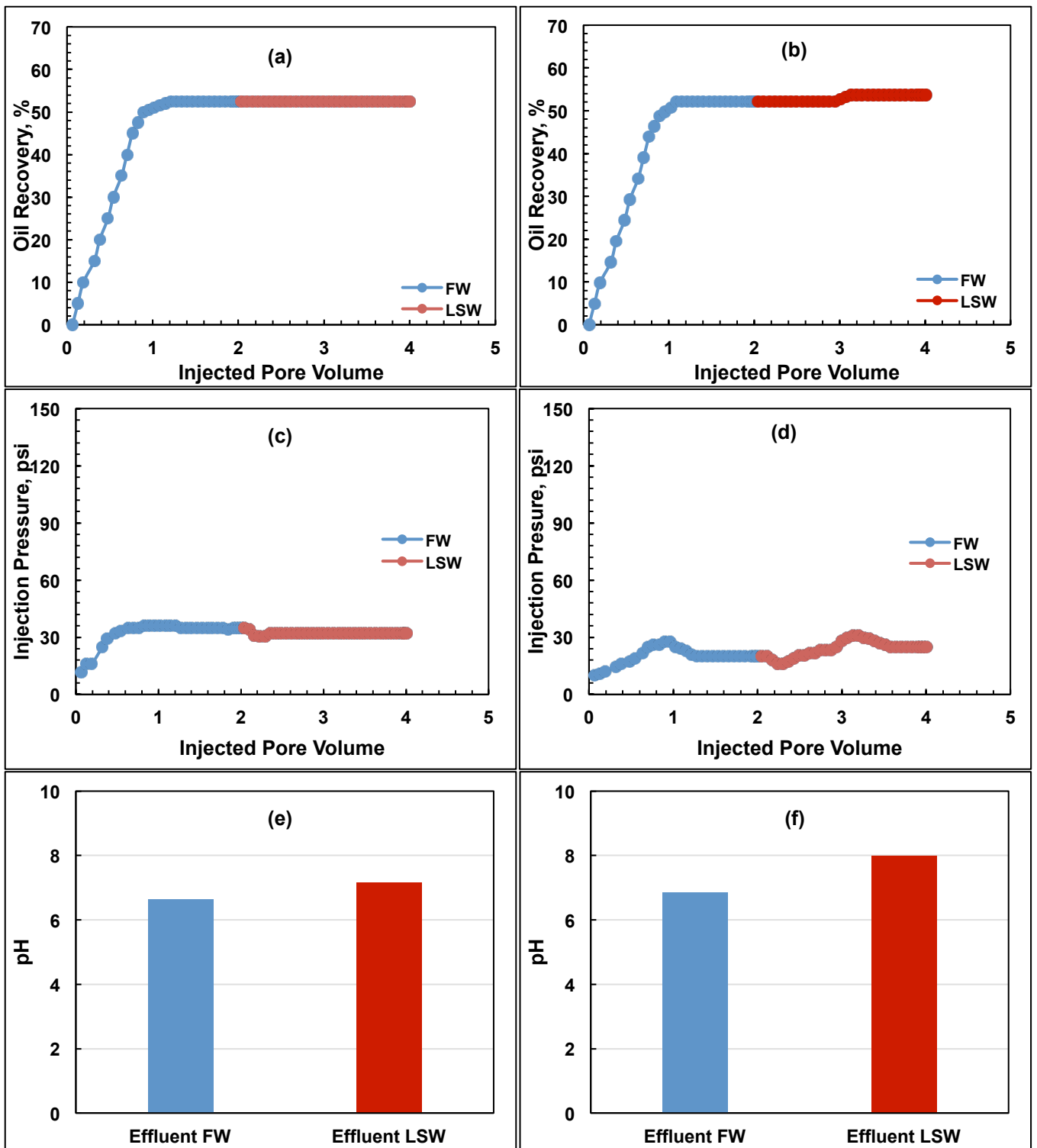


Figure 2. (a) Core1-1 oil recovery (b) Core1-2 oil recovery, (c) Core1-1 injection pressure, (d) Core1-2 injection pressure, (e) Core1-1 effluent pH, and (f) Core1-2 effluent pH. Core1-1 and Core1-2 saturated in FW containing 90mmole Mg^{2+} and the rest NaCl. Core1-1 and Core1-2 flooded with FW followed by LSW containing d_{3Mg}^{2+} and d_{10Mg}^{2+} . The FW salinity is 96100 ppm and the LSW salinity is 4000 ppm.

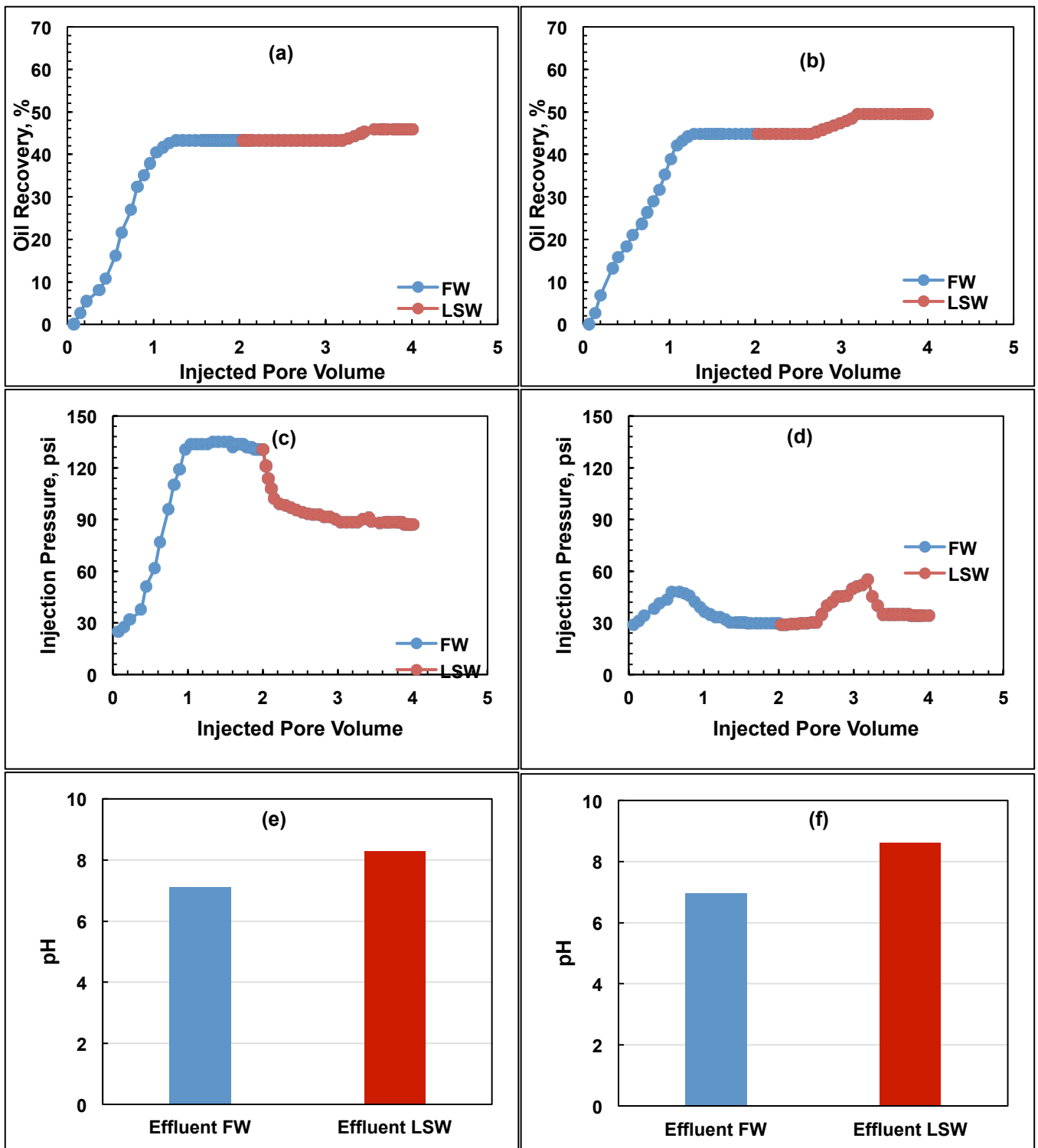


Figure3. (a) Core2-1 oil recovery (b) Core2-2 oil recovery, (c) Core2-1 injection pressure, (d) Core2-2 injection pressure, (e) Core2-1 effluent pH, and (f) Core2-2 effluent pH. Core2-1 and core2-2 saturated in FW containing 90mmole Ca^{2+} and the rest NaCl. Core2-1 and Core2-2 flooded with FW followed by LSW containing $\text{d}_{3\text{Ca}}^{2+}$ and $\text{d}_{10\text{Ca}}^{2+}$. The FW salinity is 96100 ppm and the LSW salinity is 4000 ppm.