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Impact of cation type and concentration in injected brine on oil recovery in sandstone reservoirs

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ABSTRACT

Low salinity waterflood has a high potential to improve oil recovery. However, the optimum water chemistry for waterflooding is still not well known, and the dominant mechanisms are debated. In the present work, the effect of cation type and concentration in the injected water on the oil recovery to identify the optimum water salinity and composition was studied by performing coreflood experiments. In addition, zeta potentials at oil/brine and rock/brine interfaces were measured to examine the impact of brine composition on electrical double layer expansion and investigate if it is one of the dominant mechanisms. Furthermore, ionic exchange tests were conducted to improve the understanding of the rock/brines interactions.

Berea sandstone cores were used for waterflooding and ion exchange experiments; all coreflood experiments were performed at high-pressure/high-temperature (HP/HT) conditions. Different concentrations of NaCl, CaCl₂, and MgCl₂ solutions were tested with two crude oils of different compositions and properties. Ion concentrations in the core effluent fluids were analyzed for both waterflooding and ionic exchange tests. The zeta potential was measured for solutions of oil/brine, and solid/brine; the solids were Berea sandstone, quartz, feldspar, and four types of clays.

This work contributes to the understandings of the impacts of double layer expansion on oil recovery during waterflooding. Zeta potential results showed that Na⁺ changes the electrical charge at both oil/brine and rock/brine interfaces to highly negative, which results in higher repulsive forces between the two interfaces, and hence wettability alteration. Moreover, waterflood experiments with NaCl solutions improved oil recovery significantly compared to CaCl₂ and MgCl₂. These results showed that there seems to be a correlation between zeta-potential and oil recovery which implies that double-layer expansion could be a primary mechanism of oil recovery by low salinity waterflood. This study demonstrates that cation type has a significant impact on oil recovery, and it could be more dominant than the effect of total salinity of the injected brines. These findings can help in screening the brines that have higher potentials for oil recovery improvement.

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1. Introduction

Waterflooding is applied intensively worldwide to improve oil recovery. It was demonstrated that water chemistry has a significant impact on oil recovery in sandstone reservoirs on coreflood and fields scales (Morrow and Buckley, 2011). Several field trials proved the efficiency of low salinity water in improving oil recovery (Webb et al., 2004; Robertson, 2007; Lager et al., 2008). Vledder et al. (2010) observed a reduction in water cut due to wettability change when low salinity water was injected in Omar field, Syria. In addition, a

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comprehensive experimental work showed the success of low salinity water in both secondary and tertiary recovery modes (Zhang et al., 2007; Agbalaka et al., 2009) but sometimes for only one or the other (Zhang and Morrow, 2006). In some other studies, tertiary oil recovery improvement was never observed, but low salinity water improved secondary oil recovery significantly (Rivet et al., 2010; Nasralla et al., 2011). However, Skrettingland et al. (2011) proved that low salinity water might not be efficient in certain cases when the rock wetting conditions are optimal, such that seawater injection is already efficient. There is less evidence of the low salinity water effect available in literature on carbonate reservoirs. However, significant responses of diluted seawater during secondary and tertiary corefloods were presented by Yousef et al. (2011), and the response on field scale was confirmed by single well chemical tracer tests (Yousef et al., 2012).

Nomenclature

CBM	coalbed methane
Cp	centipoise
FB	formation brine
HP/HT	high-pressure/high-temperature
ICP-OES	inductively coupled plasma optical emission spectrometry
OOIP	original oil in place

PV	pore volume
RF	recovery factor
S_{wi}	irreducible water saturation
TDS	total dissolved solids, mg/l

Greek symbols

ζ	zeta potential (mV)
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The possible risk of low salinity waterflood is the potential to cause formation damage in sandstone because of the presence of clays. Therefore, it is important to take into consideration formation damage when designing low salinity brines. [Scheuerman and Bergersen \(1990\)](#), [\(1989\)](#) developed diagrams that defined formation damage regions based on the ionic strengths of the brines which can be used to design brines that are safe to inject. However, these charts were developed based on single phase waterflood experiments, while on field scale oil saturation cannot go below residual saturation which can reduce the risk of formation damage as oil can coat some of the clays present in the reservoir.

Although, low salinity waterflooding showed a high potential to increase oil recovery in most of the lab work and field trials, the success of low salinity waterflooding application on a field scale is still uncertain due to the lack of understanding the underlying mechanisms, which means the optimum salinity and conditions are unknown. Most researchers agree that injecting low salinity water creates a wetting state more favorable for oil recovery. The remaining oil saturation decreases by low salinity waterflooding due to the wettability alteration that affects the microscopic distribution and flow of fluids in porous media. Different mechanisms have been proposed for wettability alteration by low salinity water; however, the primary mechanisms are still debated.

[McGuire et al. \(2005\)](#) attributed the oil recovery increase to the in-situ surfactant generation (saponification) by flooding with low salinity water. [Lager et al. \(2006\)](#) suggested that multi-component ionic exchange between the mineral surface and the invading brine is the primary mechanism underlying the improved recovery. [Tang and Morrow \(1999\)](#) explained the wettability alteration with the assumption that heavy polar components of crude oil adsorb onto particles at pore walls to give mixed-wet fines, and brine chemistry has an effect on the forces needed to strip these particles from the pore walls. [Pu et al. \(2008\)](#) suggested that interstitial dolomite crystals play a role in low salinity recovery mechanisms; there was no tertiary oil recovery by coalbed methane (CBM) water in dolomite-free cores. Anhydrite dissolution was suggested as the primary mechanism for recovery of residual oil by low salinity water from reservoir cores, which are rich in anhydrite cement ([Pu et al., 2010](#)). [Ligthelm et al. \(2009\)](#) suggested that the main mechanisms of wettability alteration were cation exchange and expansion of the electrical double layer.

Rock wettability depends on the stability of water film between rock surface and crude oil ([Hirasaki, 1991](#)). The stability of water films is a function of the electrical double-layer repulsion that results from surface charges at the solid/water and water/oil interfaces. If these two interfaces have similar charges, a repulsive electrostatic force will occur that keeps the disjoining pressure high and maintains a thick water film; this produces a water-wet rock surface ([Dubey and Doe, 1993](#)). Sandstone is negatively charged above the pH of 2 ([Menezes et al., 1989](#)). Crude oils are positively charged at a low pH range (2–6) and negatively charged at higher pH values ([Takamura and Chow, 1985](#); [Buckley et al.,](#)

[1989](#)). [Nasralla et al. \(2013\)](#) investigated the effect of electrical charges at the oil/brine interfaces on the wettability of mica surfaces. Low salinity water altered the wettability towards more water-wet at different pressures and temperatures. Lowering the water salinity changed the charges at the oil/brine and rock/brine interfaces to more negative, which resulted in increasing the repulsive forces between the oil/brine and solid/brine interfaces, and hence wettability alteration. The authors used the contact angle technique for wettability evaluation, and zeta potential to estimate the surface charge at oil/brine interface. [Hassenkam et al. \(2012\)](#) performed an atomic force microscopy study to measure the adhesion forces between carboxylic acid and sandstone; they noticed reduction in the adhesion forces with low salinity brines. The authors attributed this effect mainly to the DLVO theory.

Zeta potential is the potential at the shear plane of the electrical double-layer. The magnitude of the zeta potential is related to the surface charge at the oil/brine and mineral/brine interfaces, and the thickness of the double layer. The ionic strength of water affects the surface charge of sandstone and clay particles ([Kia et al., 1987](#); [Alotaibi et al., 2011](#)). Zeta potentials of three different clays (kaolinite, illite, and chlorite) were determined in distilled water by [Hussain et al. \(1996\)](#); kaolinite was the most negative clay for the whole pH range. Furthermore, the surface charge of solids is affected by the cation type. It was found that Ca^{2+} and Mg^{2+} reduced the magnitude of zeta potential of Berea sandstone and made it close to zero more effectively than Na^+ ions ([Farooq et al., 2011](#)). The zeta potential of kaolinite with monovalent cations was more negative at 10^{-2} M concentrations at almost all pH values compared to the values obtained with fresh water, whereas, the zeta potential of kaolinite decreased (became less negative) with an increase in divalent cation concentration ([Yukselen and Kaya, 2003](#)). Moreover, the effect of brine salinity at the interface between oil and brine was reported by [Buckley et al. \(1989\)](#) using different concentrations of NaCl. Low ionic strength of NaCl solutions resulted in a stronger negative charge of the brine/oil interface compared to high ionic strength. It is evident that electrokinetic charges of both oil/brine and rock/brine interfaces are significantly affected by the ionic strength of the water.

The chemistry of water used for water flooding is a dominant parameter in determining the oil recovery factor. Most of the experimental work in the literature focused on reducing the brine salinity, and not much attention was paid to the role of cation types in the injected brine. Thus, the main objectives of this study are studying the effect of cation type, and concentration in the injected water on oil recovery, in addition to understanding the working mechanisms for improving oil recovery by manipulating the water chemistry. Therefore, several sets of experiments were conducted. First, coreflood experiments were run on different concentrations of NaCl, CaCl_2 , and MgCl_2 solutions to confirm that there is a significant effect of the cation type on oil recovery improvement. Second, zeta potential measurements were conducted at oil/brine and solid/brine interfaces by testing several single-cation solutions against crude oils, Berea sandstone, and

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