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# A Comprehensive Review on Dynamics of Electrical Double Layer Expansion in Enhancing Oil Recovery

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**Abstract**
Low salinity waterflooding (LSWF) has gained prominence as an enhanced oil recovery (EOR) method by exploiting electrical double layer (EDL) expansion to alter wettability and enhance oil displacement in both sandstone and carbonate reservoirs. This review critically evaluates the role of EDL dynamics in modifying crude oil/brine/rock (COBR) interactions through electrostatic mechanisms, with a focus on the effects of brine composition, salinity, and reservoir conditions. Insights from zeta potential measurements and surface complexation modelling (SCM) indicate that EDL expansion increases electrostatic repulsion, promoting a shift toward water-wet conditions and improving oil recovery. Ion-specific effects are discussed, showing that monovalent ions (e.g., $Na^+$) are more effective in sandstones, while divalent potential-determining ions (e.g., $Ca^{2+}$, $Mg^{2+}$, $SO_4^{2-}$) play a key role in carbonates. A "thermal resilience window" is identified 50–90 °C for sandstones and above 100 °C for carbonates within which EDL-driven processes are most efficient. By integrating experimental evidence with theoretical modelling, this work offers a comprehensive framework for optimizing LSWF strategies, linking nanoscale electrokinetic behaviour with practical field implementation. The insights offered here advance the scientific understanding of EDL expansion while presenting actionable guidelines for optimizing LSWF in diverse reservoir settings.

**Keywords:** Low salinity waterflooding; wettability alteration; electrical double layer; zeta potential; surface complexation modelling; enhanced oil recovery

## 1. INTRODUCTION

EOR techniques have become indispensable in maximizing hydrocarbon extraction from mature reservoirs. Among these techniques, LSWF has emerged as a promising and environmentally sustainable method due to its ability to improve recovery efficiency in both sandstone and carbonate reservoirs. The effectiveness of LSWF stems from its unique influence on COBR interactions, which alter reservoir wettability—a critical factor in optimizing oil displacement and recovery. While multiple mechanisms have been proposed to explain the success of LSWF, including multicomponent ion exchange (MIE) and mineral dissolution, the expansion of the EDL has gained significant attention as a primary driver for wettability modification and incremental oil recovery.

The EDL is a fundamental electrochemical phenomenon that forms at charged interfaces, such as those between rock/brine and oil/brine (Figure 1) [1,2,3,4]. It consists of two layers: the Stern layer, where counterions are tightly bound, and the diffuse layer, where ions are more loosely associated due to thermal motion and electrostatic forces. The thickness of the EDL, characterized by the Debye length, is highly sensitive to brine salinity and composition. When low-salinity brine is injected into a reservoir, the EDL expands, increasing electrostatic repulsion between oil components and rock surfaces. This repulsion disrupts the adhesion of polar oil compounds, shifting wettability toward a more water-wet state and facilitating oil mobilization [1,2,5]. The role of EDL expansion in LSWF was first highlighted by British Petroleum in 2006, and since then, it has been extensively studied through laboratory experiments, zeta potential measurements, and surface complexation modelling (SCM).

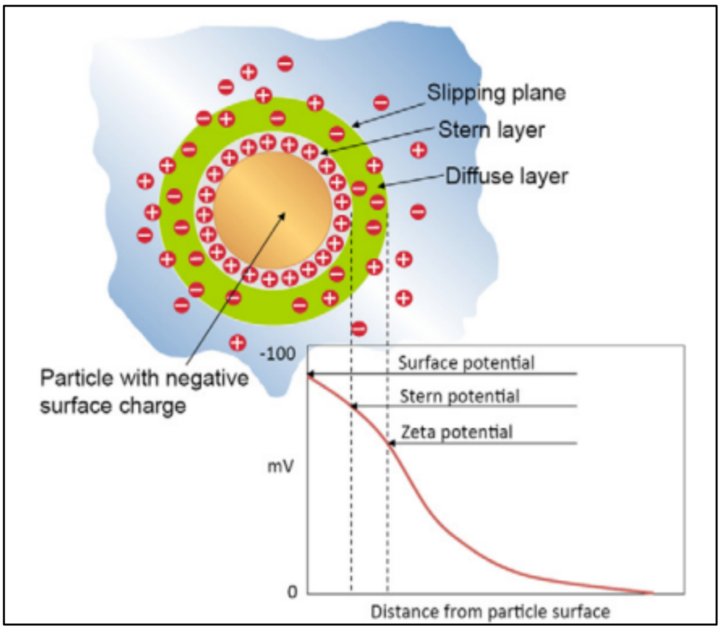


**Figure 1.** Electrically charged surface around mineral and zeta potential [6].

Despite its recognized importance, the contribution of EDL expansion to oil recovery is often conflated with other mechanisms, such as ion exchange or pH effects, in existing literature. This review aims to provide a clearer distinction by synthesizing recent advancements in understanding EDL dynamics and their direct impact on wettability alteration. For instance, studies have shown that the extent of EDL expansion is governed by factors such as brine ionic strength, rock mineralogy, and reservoir conditions (e.g., temperature and pressure). In sandstones, the presence of clay minerals like kaolinite enhances EDL expansion due to their high surface charge density, while in carbonates, potential-determining ions (PDIs) like $Ca^{2+}$, $Mg^{2+}$, and $SO_4^{2-}$ play a pivotal role. Such insights are critical for tailoring LSWF strategies to specific reservoir types.
This paper builds upon and differentiates itself from prior reviews by offering a comprehensive examination of EDL expansion as a standalone mechanism, supported by experimental and theoretical evidence. We delve into key laboratory techniques, such as zeta potential measurements, which quantify surface charge alterations, and SCM simulations, which predict electrokinetic interactions at COBR interfaces. Furthermore, we explore how variations in brine composition, salinity, and reservoir conditions influence EDL behaviour and, consequently, oil recovery efficiency. By consolidating these findings, this review not only clarifies the mechanistic role of EDL expansion but also provides practical guidelines for optimizing LSWF in diverse reservoir settings.

**Table 1:** Summary of impact of zeta potential on oil recovery from relevant published papers

| **Reference** | **Reservoir type** | **Brine Salinity** | **Zeta potential (mV)** | **pH** | **Ions present** | **Incremental recovery** | **Remarks** |
|---|---|---|---|---|---|---|---|
| **Buckley and Morrow [7]** | Sandstone | — | -50 | >4 | $Na^+$ | Yes | Increased pH at low saline condition increases repulsion between oil and rock surface increasing recovery. |
| **Ligthelm et al. [8]** | Sandstone | 2000 mg/l | — | — | $Na^+$, $Mg^{2+}$, $Ca^{2+}$ | Yes | Wettability alters as repulsion increases between oil and rock surface leading to incremental recovery. |

| | | | | | | | |
|---|---|---|---|---|---|---|---|
| **Strand et al. [9]** | Carbonate | — | — | 8.4 | $Ca^{2+}$, $Mg^{2+}$, $SO_4{}^{2-}$ | Yes | Change in surface charge properties of chalk due to increased adsorption of $SO_4{}^{2-}$ and $Ca^{2+}$ close to rock surface facilitates desorption of negatively charged carboxylic components increasing recovery rate. |
| **Mahani et al. [10]** | Carbonate | 100 times diluted seawater (43731 ppm) | <-10 | >8 | $Ca^{2+}$, $Mg^{2+}$, $SO_4{}^{2-}$ | Yes | Low salinity brine increases negative magnitude of zeta potential. |
| **Rodriguez and Araujo [11]** | Quartz, Kaolinite, Calcite | — | Zeta potential decreases at the rate of −2.3 mV/◦C for quartz, −0.96 mV/◦C for kaolinite, and −2.1 mV/◦C for calcite for pressure values less than 45 psi | 2-9 | — | Yes | At elevated temperature and pressure negativity of zeta potential increased thereby increasing thickness of EDL and stability of water film around mineral |
| **Lee et al. [12]** | Sandstone | — | — | — | $Na^{+}$, $Mg^{2+}$, $Ca^{2+}$, $K^{+}$, $Li^{+}$ | Yes | Decreasing ionic strength of brine increased thickness of water layer |
| **Nasralla et al. [13]** | Sandstone | 10% Aquifer water (5436 mg/L) and Deionised Water | At pH 6-11, zeta potential lies in the range of -40 to -60 for Crude oil A and from -40 to -50 for Crude oil B | >6 | — | Yes, 13-22% of OOIP by DIW and 8-14% of OOIP by AQ injection respectively in secondary mode. | Injection of low salinity brine (deionized water) increased negative zeta potential values significantly at oil/brine interface. |
| **Nasralla and Nasr-El-Din [14]** | Sandstone | 10% Aquifer water (544 mg/L) and 5000 mg/L Seawater | Zeta potential >-10 and >-20 for 5000mg/L NaCl brine and 10% AQ at the given pH values respectively. | 5.9 (5000 mg/L)-7.3 (10% AQ) | — | Yes, 12% of OOIP with 10% AQ and 10% of OOIP with 5000mg/L NaCl respectively in secondary mode. | With increasing pH, repulsion at oil/brine, clay/brine interface increased rendering water wet surface. |
| **Jackson et al. [15]** | Carbonate | 20 times | -8 | — | $Ca^{2+}$ or $Mg^{2+}$ | Yes | LSW yields more negative zeta potential at the mineral-brine interface |

| | | | | | | | |
|---|---|---|---|---|---|---|---|
| | | diluted SW | | | | | altering wettability and increasing recovery. |
| **Alroudhan et al. [16]** | Carbonate | 20times and 10times diluted seawater | -9 to -10 | — | $Ca^{2+}$, $Mg^{2+}$, $SO_4^{2-}$ | Yes | Increased SO4 $^{2-}$ concentration or injection of diluted brine increses recovery in carbonates by changing electric charge properties at rock/brine interface. |
| **Yang et al. [17]** | Sandstone | 1580ppm | ζ potential value of -50 and -31 at brine/crushed Berea interfaces and brine/crude oil interfaces respectively. | — | $Ca^{2+}$, $Na^{+}$ | Yes | LSW injection alters wettability by increasing diffuse layer thickness giving incremental oil recovery. |
| **Mahani et al. [18]** | Carbonate | 25 times diluted seawater (1750ppm) | At ambient conditions (25°C) ζ-potential values were negative for LS brine. At elevated temperatures (50 and 70°C) ζ-potential became more positive. | 6-8 | $Ca^{2+}$, $Mg^{2+}$, $SO_4^{2-}$ | Yes | Decreased salinity resulted in a more negative zeta potential value. Zeta potential became positive with increased pH and temperature. |
| **Wei et al. [19]** | | 500 mg $L^{-1}$ | — | — | $Ca^{2+}$, $Na^{+}$ | Yes | Thickness of diffuse layer increases in presence of monovalent cations by altering wettability. |
| **Rodrigues et al. [20]** | Carbonate | | — | — | — | No recovery | Zeta potential did not affect recovery in tertiary mode. |
| **Rahevar et al. [21]** | Sandstone | — | -10 | — | — | Yes | LSW in conjuction with surfactant yielded negative zeta potential leading to increased oil recovery. |

## 2. MAJOR FACTORS AFFECTING EDL EXPANSION

### 2.1 Composition and Salinity of Injected Brine

The composition and salinity of injected brine significantly impact rock wettability and oil recovery. Tailoring brine's ionic composition generally enhances oil recovery [22, 23]. Yildiz and Morrow [24] observed that low-salinity brine (2% $CaCl_2$) improved oil recovery by 5.5% compared to high-salinity brine (4% NaCl + 0.5% $CaCl_2$) in Berea sandstone cores aged in Moutray crude oil. However, imbibition tests revealed higher recovery with high-salinity brine due to less water-wet conditions. Tang & Morrow [25] highlighted the role of cation valency, noting that higher valency increased waterflood recovery but reduced imbibition rates, except for $AlCl_3$ due to pH effects. Increased ion valence or salinity compresses the electrical double layer, promoting polar oil adsorption onto sandstone surfaces via ion binding, reducing water wetness and recovery. Bagci et al. [26] found improved recovery with specific ionic compositions (2% KCl + 2% NaCl) over salinity alone, which stems from synergistic EDL stabilization—$K^+$ reduces clay swelling while $Na^+$ maximizes diffuse layer thickness [27]. Optimal recovery from low-salinity brines is yielded at a salinity <5000 ppm [1, 28, 29] which is found to coincide with the increasing EDL thickness at zeta potential <-25mV [10]. These findings underscore the importance of brine composition and salinity in optimizing oil recovery processes. Torrijos et al. [29] demonstrated that high-salinity brines can induce smart water effects, emphasizing the importance of brine composition alongside salinity. Khosravi et al. [30] reported a 4% increase in oil recovery using brines with 8000 ppm NaCl and 20,000 ppm $CaCl_2$. For carbonate rocks, optimal recovery is achieved with potential determining ions (PDIs) like $Ca^{2+}$, $Mg^{2+}$, and $SO_4^{2-}$ at higher temperatures (>90°C) [31, 1]. Carbonate cores perform well at salinities of 20,000-30,000 ppm, unlike sandstone cores, which favour low-salinity brines (<5000 ppm) [32]. Derkani et al. [2] attribute this carbonate rock tolerance to higher salinity conditions to the enhanced resistance of $Mg^{2+}$-$SO_4^{2-}$ ion complexes to electrical double layer compression. $SO_4^{2-}$. also enhances recovery in clay-bearing sandstones by forming aggregates with $Na^+$ near clay surfaces, reducing oil adsorption and altering wettability [33]. Ligthelm et al. [8] concluded that wettability alteration is driven by brine composition rather than ionic exchange, with $SO_4^{2-}$, $Ca^{2+}$, and $Mg^{2+}$ improving recovery in carbonates. Strand et al. [34] confirmed that anionic exchange between $SO_4^{2-}$ and divalent cations in brine enhances oil recovery and shifts wettability to a more water-wet state. PDIs are crucial for carbonate recovery, while tuning ionic composition benefits sandstone cores [1,2]. This suggests that wettability alteration in crude oil/brine/rock systems is influenced by DLVO and surface force interactions [35, 22]. Overall, brine composition, particularly PDIs, plays a key role in optimizing oil recovery for both carbonate and sandstone reservoirs.

### 2.2 Temperature and Pressure

Temperature significantly impacts low salinity waterflooding (LSWF) by altering pH, surface potential, interfacial tension (IFT) and wettability at oil-brine and rock-brine interfaces [36, 37]. Higher temperatures reduce residual oil saturation and water relative permeability, promoting water-wet conditions in addition to reducing IFT and enhancing oil recovery [37, 38]. However, the relationship is complex. Shimoyama and Johns [39] found that increasing temperature decreases crude oil acid number (AN) due to decarboxylation, while Mansi et al. [36] noted that the base-to-acid ratio influences wettability alteration at oil/brine interfaces. Temperature affects aging and displacement processes differently. Rezaeidoust et al. [31] observed optimal recovery in sandstone at 90°C, with diminished low salinity effects outside this range. Cissokho et al. [40] highlighted that displacement temperature impacts recovery more than aging temperature, with higher temperatures improving recovery. Strand et al. [41] reported that increased temperature reduces cation desorption due to collapse of the EDL leading to lower recovery, while Aghaeifar et al. [42] found that high temperatures and salinities (20,000 ppm) diminish the low salinity effect by reducing polar component adsorption. In carbonates, temperatures >100°C enhance recovery [18, 31]. Strand et al. [9] noted that elevated temperatures improve imbibition rates with adequate divalent ions ($Ca^{2+}$, $SO_4^{2-}$). Heidari et al. [43] concluded that temperature, not aging time, primarily controls wettability alteration, with faster intermediate wetting at 50°C than 25°C. Pressure also critically influences LSWF. Lower pressures reduce asphaltene solubility, causing polar components to precipitate and create oil-wet conditions resulting in supressed EDL expansion [2, 35]. This section particularly *identifies EDL's "thermal resilience window"* which is concluded as 50–90°C for sandstones; >100°C for carbonates, unifying disparate observations from Cissokho et al. [40] and Heidari et al. [43].

### 2.3 Rock Mineralogy and Surface Charge Densities

Low salinity water flooding (LSWF) enhances oil recovery differently in sandstones and carbonates due to their mineralogical differences. Sandstones, with their negatively charged clay minerals (e.g., kaolinite, chlorite), exhibit enhanced recovery primarily through EDL-driven processes [23]: kaolinite fines migration creates water-wet surfaces not merely by physical detachment as emphasized by Bernard [44], but by exposing fresh mineral faces with varying charge density [45] that modifies EDL thickness. This reconciles observation by Austad et al. [46] that even non-kaolinite clays (illite, muscovite) improve recovery—due to variable charge densities that can sustain EDLs under flow conditions [47].

In carbonates, the mineral-EDL relationship is more complex but equally decisive. While Yousef et al. [48] attributed the success of LSWF in carbonates primarily to mineral dissolution, subsequent SCM studies by

Mahani et al. [49]; Alroudhan et al. [16] have demonstrated that these improvements correlate strongly with ζ-potential modifications induced by potential determining ions ($Ca^{2+}$, $Mg^{2+}$, and $SO_4^{2-}$) adsorption at the rock-brine interface. This aligns with findings by Strand et al. [34] that $SO_4^{2-}$-$Ca^{2+}$ exchange alters surface charge, while subsequent work by Zhang et al. [50] and Alroudhan et al. [16] demonstrates this ion exchange directly expands the EDL, particularly when sulfate concentrations exceed stoichiometric balance with calcium. Mahani et al. [18] later confirmed these effects are temperature-dependent, with maximum EDL expansion occurring at 70-90°C.

However, challenges persist. Excessive dissolution of minerals like anhydrite and dolomite during successive flooding can lead to overly water-wet surfaces, reducing recovery [51]. LSWF effectiveness depends on mineral composition, surface charge dynamics, and brine chemistry, necessitating tailored approaches for optimal results in both sandstone and carbonate reservoirs.

## 3. ROLE OF EDL EXPANSION IN OIL RECOVERY AND THEIR EXPERIMENTAL INVESTIGATIONS

Numerous studies highlight wettability modification as a key mechanism in enhanced oil recovery through low-salinity water flooding (LSWF). Tailored brine injection alters rock surface wettability towards a more water-wet state, driven by factors such as the presence of potential determining ions (PDIs) and ionic exchange between brine and rock surfaces [31, 34, 12]. These interactions reduce oil adhesion, improving recovery. Additionally, electrokinetic charges play a critical role in altering surface charge properties of sandstone and carbonate rocks, influencing electrostatic forces at rock-brine and oil-brine interfaces, thereby enhancing oil displacement efficiency [10, 14]. The expansion of the electrical double layer (EDL) during LSWF further shifts wettability, facilitating pore fluid movement and incremental recovery [52]. Wettability, defined as the affinity of one phase towards the rock surface in the presence of another immiscible phase [53] is a complex interplay of rock-brine interactions and reservoir heterogeneity. This paper reviews wettability alteration through EDL expansion, supported by zeta potential measurements and surface complexation modelling (SCM) studies.

### 3.1 Zeta Potential Measurements

Zeta potential plays a critical role in understanding wettability alteration during low-salinity water flooding (LSWF). Injecting low-salinity brine disrupts the stability of the water film on mineral surfaces due to double-layer expansion, creating a more water-wet surface and increasing electrostatic repulsion between the mineral surface and oil components, thereby enhancing oil recovery [14, 18]. Studies indicate that reducing brine salinity expands the double layer, repelling oil molecules and improving recovery [54, 55, 16].

Buckley and Morrow [7] emphasized that wettability alteration during LSWF depends on pH, brine composition, and concentration. Ligthelm et al. [8] demonstrated through coreflood experiments that double-layer expansion is key to altering wettability. They found that lowering brine salinity, rather than ionic exchange, is the primary driver of wettability change. Injecting diluted NaCl brine, free of multivalent cations, increased electrostatic repulsion, promoting water-wetness and recovery. This effect was most pronounced with 100-fold diluted NaCl brine. Lee et al. [12] supported these findings, showing that reducing brine salinity from 0.1M to 0.001M increased water layer thickness more significantly for divalent cations (e.g., $MgCl_2$: 8.14Å to 14.8Å) than for monovalent cations (e.g., NaCl: 10.8Å to 11.8Å). They concluded that divalent cations in sandstone result in thinner water layers, while monovalent cations promote thicker layers, enhancing oil displacement efficiency. In summary, LSWF alters wettability through double-layer expansion, with reduced salinity and monovalent cations enhancing water-wetness and oil recovery by increasing electrostatic repulsion and water layer thickness.

The impact of potential determining ions (PDIs) like $Ca^{2+}$, $Mg^{2+}$, and $SO_4^{2-}$ on the zeta potential and wettability of carbonate surfaces has been extensively studied. Strand et al. [9] observed that $SO_4^{2-}$ adsorption, along with $Ca^{2+}$ co-adsorption, altered surface charge characteristics, leading to wettability modification and desorption of polar oil components at higher temperatures. Zhang et al. [50] further demonstrated that $Mg^{2+}$ could substitute $Ca^{2+}$ at high temperatures, increasing the positive charge on chalk surfaces. They concluded that $SO_4^{2-}$ interaction with $Ca^{2+}$ and $Mg^{2+}$ enhances water wetness, improving oil recovery at both low and

high temperatures. Alroudhan et al. [16] found that increasing $SO_4^{2-}$ concentration or reducing $Ca^{2+}/Mg^{2+}$ in brine yields more negative zeta potentials, repelling polar oil components and enhancing recovery.
Brine salinity and pH significantly influence zeta potential and wettability during low salinity water flooding (LSWF). Mahani et al. [10] reported that diluted seawater (1750 ppm) with $SO_4^{2-}$ resulted in more negative surface charges compared to high-salinity water (180,000 ppm), promoting water-wet conditions. Conversely, Nasralla and Nasr-El-Din [14] noted that lower pH in low-salinity brine increased surface charges at oil/brine and rock/brine interfaces, forming a thinner double layer and reducing recovery. In contrast, Buckley et al. [7] observed that higher pH enhances electrostatic repulsion between interfaces, improving oil recovery. Optimizing brine chemistry by increasing $SO_4^{2-}$ or reducing $Ca^{2+}/Mg^{2+}$ concentrations can enhance recovery by shifting surface charge toward more negative zeta potentials and repelling oil components.
Mahani et al. [18] found that electrical properties at rock-brine and oil-brine interfaces increase with temperature (25-70°C). At higher temperatures, lower sulfate concentrations allow more $Ca^{2+}$ and $Mg^{2+}$ adsorption, making surfaces oil-wet. Mehraban et al. [56] confirmed that tailored formation brine at elevated temperatures yielded better recovery than low-salinity brine at ambient temperatures. Sulfate increased repulsion forces, while calcium had the opposite effect. Yang et al. [17] studied wettability modification during LSWF, showing that reducing $CaCl_2$ or NaCl concentrations increased repulsion between oil and mineral surfaces, leading to more negative zeta potential values and improved recovery. They concluded that repulsive forces exceed oil-binding energy, breaking calcium bridges that adsorb carboxylate groups onto rock surfaces, shifting wettability to water-wet.
Wei et al. [27] examined the role of $Ca^{2+}$ and $Na^{+}$ in microscopic sweep efficiency. Higher concentrations of these cations increased negative zeta potential values on quartz and clay surfaces (Figure 2). $Na^{+}$ had a more pronounced effect than $Ca^{2+}$, as the latter acts as a bridging cation binding oil to rock surfaces. Overall, the authors concluded that ionic concentration in injected brine plays a key role in wettability modification during LSWF in sandstone and carbonate reservoirs.

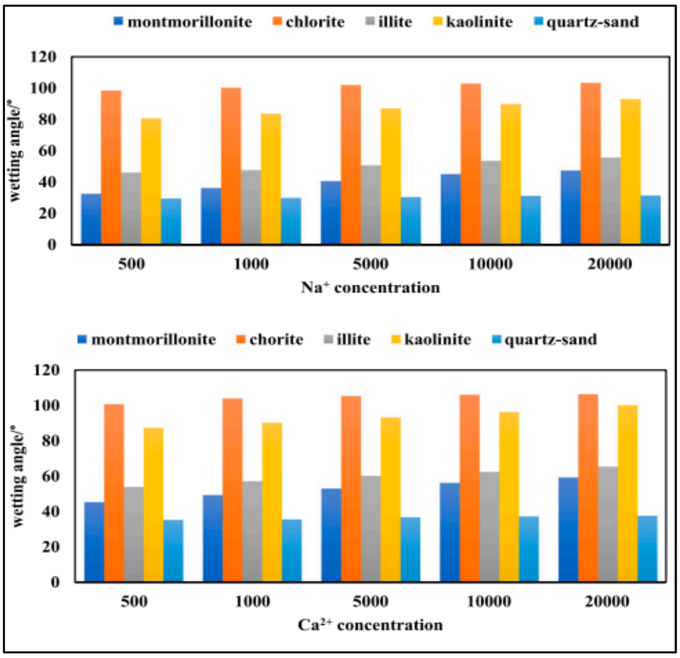


**Figure 2.** Wetting angle of different types of clay minerals at different $Na^{+}$ and $Ca^{2+}$ concentration [27].

Jackson et al. [15] showed that incremental recovery in carbonate reservoirs occurs when mineral-brine and oil-brine interfaces share the same zeta potential sign. This increases electrostatic repulsion, raising disjoining pressure and stabilizing the water film, leading to higher recovery rates. However, Rodrigues et al. [20] found conflicting results, attributing discrepancies to uncertainty in oil/water interface polarity, suggesting that zeta potential may not always be a critical factor in LSWF recovery. Rahevar et al. [21] confirmed that modified brine alters surface charge properties and interfacial tension (IFT) in sandstone reservoirs, enhancing recovery

(Figure 3). Increased electrostatic repulsion mobilized trapped oil and modified wettability. They also examined surfactants, finding that sodium dodecyl benzene sulfonate (SDBS) was more effective than cetyltrimethylammonium bromide (CTAB) due to its opposite polarity to the rock surface, resulting in a more negative zeta potential value (Figure 4). These findings highlight the significance of zeta potential in wettability alteration and optimizing LSWF for enhanced oil recovery, though some studies challenge its direct impact.

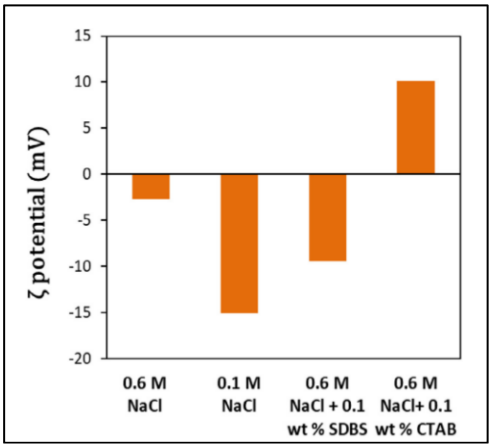


**Figure 3.** Zeta potential measurements in sandstone surface by injection different fluids [21].

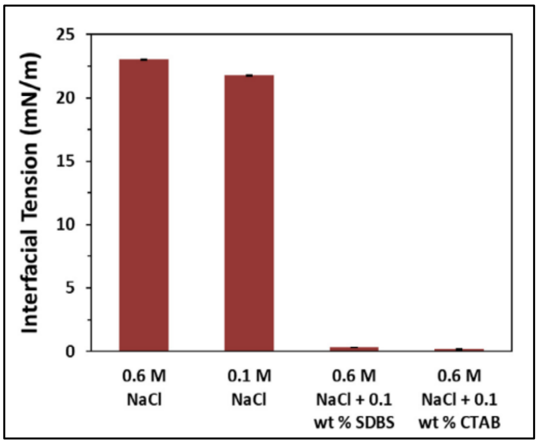


**Figure 4.** Interfacial tension between crude oil and the injection fluids [21].

### 3.2 Surface Complexation Model

SCMs have become essential for understanding electrokinetic interactions at rock/brine and oil/brine interfaces in enhanced oil recovery (EOR). Studies [57-60] demonstrate that SCMs predict charge distribution and zeta potential values, offering insights into wettability alteration. By simulating chemical equilibrium at mineral/brine interfaces, SCMs define surface complexes responsible for charge development, with equilibrium constants like bulk reactions [49].

Mahani et al. [49] developed an SCM to explain surface charge formation on carbonate rocks, validating experimental zeta potential variations with pH and ionic composition. They found that increasing pH made zeta potential more negative, while at high salinity, zeta potential remained stable due to reduced pH sensitivity. A follow-up study by Mahani et al. [10] modelled zeta potential variations with brine salinity and pH, demonstrating that sulfate interactions with calcite sites correlated well with experimental data. Brine dilution led to a more negative surface charge by increasing negatively charged species and reducing positive species, shifting wettability toward a more water-wet state.

SCMs have been combined with DLVO theory to assess repulsive and attractive forces between crude oil and rock surfaces. Elakneswaran et al. [58] integrated a triple-layer SCM with a thermodynamic equilibrium model to evaluate electrokinetic changes at the sandstone/brine interface. Their study found that seawater dilution increased the negative zeta potential, enhancing repulsion between crude oil and sandstone, leading to higher disjoining pressure and more water-wet conditions. Their results indicated that sandstone interface properties could be modelled using quartz and kaolinite, with mineral composition significantly influencing pH and wettability.

Sanaei et al. [57] developed a geochemical SCM that accurately predicted experimental zeta potential values at oil/brine and rock/brine interfaces based on pH and PDI concentrations. Their findings showed that increasing pH and sulfate concentration induced negative surface potential on calcite, enhancing oil recovery. SCMs have also been incorporated into reservoir simulators to dynamically model wettability alteration.

Erzuah et al. [61] correlated SCM results with a flotation technique to analyse electrostatic interactions at mineral/brine and oil/brine interfaces. Their study showed that divalent cations enhanced oil adhesion to quartz and kaolinite through cation bridging, making the surface oil-wet. In contrast, for calcite, direct carboxylate adhesion determined wettability. Their results concluded that oil adhesion depends primarily on the mineral's surface charge rather than oil/brine interface charge.

These methods modify surface charge, shifting zeta potential toward a more negative value, which improves wettability and enhances oil displacement. Abu-Al-Saud et al. [60] quantitatively validated SCM by demonstrating that divalent anions ($CO_3^{2-}$, $SO_4^{2-}$)—particularly from $Na_2CO_3$—are the primary drivers of negative zeta potentials, directly linking ion-specific adsorption to EDL expansion and water-wetness. Unlike prior studies focused on sandstones, this work reveals carbonate-specific mechanisms, showing that alkali synergy (e.g., $Na_2CO_3$) amplifies EDL effects more than monovalent ions, a critical insight for low-salinity EOR in carbonates. The SCM's ability to match experimental data (within 2.5 mV for calcite/brine) underscores its predictive power, though discrepancies at the crude oil interface highlight unmodeled organic-acid interactions—a key area for future refinement. By identifying $Na_2SO_4$ + $Na_2CO_3$ as the optimal formulation for maximizing negative surface charge, the study not only reinforces EDL expansion as a dominant wettability-alteration mechanism but also offers a practical framework for tuning brine chemistry in field applications. This work stands out for bridging theoretical SCM with experimental electrokinetics, providing a robust foundation to argue for EDL's centrality in carbonate EOR while pinpointing gaps (e.g., dynamic dissolution effects) for further research. Overall, SCMs provide critical insights into wettability modification by predicting zeta potential variations with pH, salinity, and PDIs. Their integration with other models and EOR techniques enhances our understanding of electrokinetic interactions, making them a valuable tool in optimizing LSWF and improving oil recovery.

## 4. DISCUSSIONS

The effectiveness of Low Salinity Waterflooding (LSWF) as an enhanced oil recovery technique fundamentally depends on its ability to modify reservoir wettability through Electrical Double Layer (EDL) expansion. This review systematically examines this phenomenon by integrating experimental evidence with theoretical modelling, demonstrating how different approaches collectively validate EDL expansion as a primary recovery mechanism.

Experimental observations in the literature showed that reduced brine salinity consistently alters rock wettability toward more water-wet conditions. Core flooding tests and zeta potential measurements reveal that low-salinity brines (<5000 ppm) generate stronger electrostatic repulsion between crude oil components and rock surfaces, particularly in clay-bearing sandstones. These experimental findings are further supported by interfacial studies demonstrating increased water film stability and reduced oil adhesion under low salinity conditions. The consistency of these results across multiple studies establishes a strong empirical foundation for EDL-driven recovery.

Transitioning from experimental observations to theoretical validation, surface complexation models (SCMs) emerge as powerful tools for validating experimental observations and advancing our understanding of EDL-driven recovery processes. These models successfully simulate the chemical equilibria at mineral/brine interfaces, reproducing the experimental trends in zeta potential measurements with remarkable accuracy. The models particularly excel in predicting how specific brine compositions (varying in salinity, pH, and ion types) modify surface charges - a capability that directly informs field applications. For instance, SCM outputs confirm why certain reservoirs respond better to $Na^+$-dominant brines while others require tailored solutions with potential determining ions ($Ca^{2+}$, $Mg^{2+}$, $SO_4^{2-}$).

The integration of SCMs with DLVO theory provides even deeper mechanistic insights. This combined approach quantitatively explains the balance between attractive (van der Waals) and repulsive (electrostatic) forces governing oil-rock interactions. The models successfully predict, for example, why monovalent cations ($Na^+$) typically generate stronger repulsive forces in sandstones compared to divalent cations ($Ca^{2+}$, $Mg^{2+}$), while the opposite holds true for carbonates where potential determining ions play a more significant role. This theoretical consistency with experimental recovery data reinforces the reliability of EDL expansion as a primary recovery mechanism.

While the current understanding of EDL expansion provides a robust framework for LSWF design, certain challenges remain. These include better characterization of oil/brine interfaces and incorporation of dynamic flow conditions into models. Nevertheless, the consistent agreement between experimental data and theoretical predictions across multiple studies leaves little doubt about EDL expansion's central role in wettability alteration.

This comprehensive analysis demonstrates how fundamental electrokinetic principles, when properly understood and applied, can significantly enhance oil recovery. The transition from experimental observations to theoretical validation and finally to practical application forms a coherent narrative that not only confirms EDL expansion as a key recovery mechanism but also provides a clear methodology for optimizing LSWF in diverse reservoir environments. As the industry seeks more efficient and sustainable recovery methods, this electrokinetic approach represents a scientifically grounded solution worthy of continued development and field implementation.

## 5. CONCLUSION

This comprehensive review establishes Electrical Double Layer (EDL) expansion as a transformative mechanism in low salinity waterflooding (LSWF), offering a scientific foundation for next-generation enhanced oil recovery strategies. Through systematic analysis of experimental data, advanced modelling, and field observations, we demonstrate that controlled manipulation of brine-rock-fluid interfaces represents a paradigm shift in reservoir engineering - one where nanoscale surface chemistry dictates macroscopic recovery efficiency.

The key insights from this review reveal:

- Ion-Specific Effects: Sandstones thrive with monovalent ions ($Na^+$) expanding EDLs, while carbonates respond to divalent players ($SO_4^{2-}$, $CO_3^{2-}$) - a paradigm shift from "one-size-fits-all" brine formulas.
- Predictive Power: Advanced modelling (SCM-DLVO integration) now replaces trial-and-error, enabling field-ready designs that optimize wettability alteration.
- Sustainable Advantage: LSWF cuts chemical use and energy demands by >30% versus conventional EOR, aligning with net-zero goals while boosting recovery.
- As the oil industry confronts dual challenges of energy security and environmental responsibility, EDL-driven LSWF emerges as a timely solution. Future research should focus on real-time monitoring of wettability alteration, machine learning-assisted brine optimization, and hybrid applications with green surfactants. The journey from laboratory discovery to field implementation, as chronicled in this review, exemplifies how fundamental scientific principles can revolutionize industrial practice. By harnessing the power of interfacial science, the industry can unlock billions of barrels of additional recovery while transitioning toward more sustainable operations - a compelling proposition for researchers and practitioners alike.

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## NOMENCLATURE

COBR - Crude Oil/Brine/Rock
LSWF- Low Salinity Waterflooding
ζ - Zeta Potential
PDIs- Potential Determining Ions
SDBS- Sodium Dodecyl Benzene Sulfonate
IFT - Interfacial Tension
EDL- Electrical Double Layer
DLVO- Derjaguin, Landau, Verwey and Overbeek
MIE- Multicomponent Ionic Exchange
LSE- Low Salinity Effect
CTAB- Cetyltrimethylammonium Bromide

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