

**ANALYSIS OF SPONTANEOUS IMBIBITION
ENHANCEMENT IN FRACTURED RESERVOIRS
USING DIFFERENT EOR APPROACHES**

by

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ORIGINALITY STATEMENT

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ABSTRACT

Carbonate reservoirs, which host more than half of the world's oil reserves, are predominantly oil-wet and fractured, where recovery depends on spontaneous imbibition (SI). Under such conditions, conventional waterflooding provides little recovery, making chemical enhanced oil recovery (EOR) a promising strategy. This study evaluates the performance of standalone and hybrid EOR methods in fractured carbonate cores, with a focus on the role of sequencing. Surfactant, alkali, and silica nanoparticles were tested individually in seawater (SW) and in hybrid formulations with LSW, applied either after standalone chemical flooding or following LSW pretreatment.

The findings demonstrate that the reduction of interfacial tension (IFT) is the most effective approach for improving imbibition, as evidenced by the highest imbibition rates and recovery factors (RF) in the initial stages. The wettability alteration-based approaches of LSW and nanoparticles demonstrated a gradual yet steady recovery of imbibition. The findings confirm that imbibition recovery and rate are regulated by distinct mechanisms. The IFT reduction method controls how fast the imbibition happens, while changing the wettability controls recovery extent. A comparison of standalone and hybrid designs revealed that the hybridization of methods is advantageous for improving the recovery of less effective techniques. For example, NP→LSW+NP almost doubled the recovery compared to just using nanoparticles. The most effective way to improve imbibition recovery is to use LSW followed by surfactant flooding (LSW→LSW+S). This method had an imbibition rate that was almost five times higher than using LSW alone. Using LSW→LSW+NP sequence resulted in a slight increase in imbibition recovery. However, using LSW→LSW+A design did not show any significant improvement in imbibition recovery because the crude oil had a low total acid number (TAN). Additionally, ion-modified LSW was studied to improve the final surfactant flood stage. The results showed that ion modification greatly improved wettability alteration and made the next surfactant injection more effective, which improved the kinetics of imbibition and oil recovery. Overall, the results show that hybrids can make weaker standalones work better, but the order in which they are used is very important. The best approach is to change the wettability first and then lower the IFT. For oil-wet fractured carbonates, LSW pretreatment followed by surfactant flooding in LSW (LSW→LSW+S) is recommended over LSW alone. Ion-modified LSW is even better; LSW+NP hybrids are a second choice, while LSW+A is not a good choice for low-TAN crude.

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LIST OF ABBREVIATIONS

A	Alkali
AFO	All Faces Open
CA	Contact Angle
CMC	Critical Micelle Concentration
EDL	Electrical Double Layer
EOR	Enhanced Oil Recovery
FW	Formation Water
IFT	Interfacial Tension
LSW	Low Salinity Water
MIE	Multiple Ion Exchange
NMR	Nuclear Magnetic Resonance
NP	Nanoparticles
NTI	Nuclear Tracer Imaging
OOCO	One Open, One Closed
OOE	One Open End
OOIP	Original Oil in Place
PDI	Potential Determining Ion
RF	Recovery Factor
S	Surfactant
SI	Spontaneous Imbibition
SW	Seawater
TAN	Total Acid Number
TEO	Two Ends Open
XRD	X-Ray Diffraction

LIST OF SYMBOLS

A	Cross-sectional area
C	Constant depending on pore geometry
g	Gravitational acceleration
h	Height of the core/sample
k	Permeability
L	Length of the core/sample
N_B^{-1}	Inverse Bond number
P_c	Capillary pressure
q	Volumetric flow rate
r	Effective pore radius
t	Time
$\sqrt{t}$	Square root of time
w_{total}	Total weight of the sample
$w_{water-wet}$	Weight of water-wet particles
$\Delta\rho$	Density difference between the immiscible fluids
Δp	Pressure drop across the sample
μ	Dynamic viscosity of the fluid
ϕ	Porosity
θ	Contact angle
σ	Interfacial tension

1. INTRODUCTION

1.1 Background

The demand for energy resources continues to grow worldwide, and as conventional reservoirs mature, maximizing the extraction of hydrocarbons from existing fields becomes increasingly important. Among the different types of hydrocarbon reservoirs, carbonate formations represent one of the most important global georesource reservoirs and play crucial role in a worldwide energy supply. It is estimated that more than 60% of the world's oil reserves and around 40% of gas reserves are located in carbonate formations (Kaminskaite-Baranauskiene et al., 2023; Kashiri et al., 2023; Qi & Liu, 2024). Despite their vast resource potential, these reservoirs are typically characterized as naturally fractured, highly heterogeneous, and predominantly oil-wet (Dehaghani & Daneshfar, 2024; Guo & Li, 2024; Sagbana et al., 2023; Tajikmansori et al., 2021). These characteristics strongly influence fluid flow behavior and hydrocarbon recovery efficiency, making carbonate formations a major focus of research and development.

In naturally fractured carbonate systems, hydrocarbons are mainly stored within the low-permeability rock matrix, while fractures primarily serve as highly conductive pathways for fluid flow (Austad et al., 1998; Chilingar & Yen, 1983; Karimova et al., 2023). As a result, efficient oil production relies on the transfer of fluids between fractures and the matrix. However, the efficiency of this matrix-fracture fluid exchange is strongly controlled by wettability conditions. Carbonate reservoirs often exhibit oil-wet or mixed-wet wettability conditions due to the adsorption of polar organic compounds from crude oil onto positively charged carbonate rock surfaces (Deng et al., 2024; Lin et al., 2018; Nande & Patwardhan, 2022; Sagbana et al., 2023). This wettability state controls fluid distribution within the pore space and governs capillary-driven displacement within the matrix (Karimova et al., 2023; Kashiri et al., 2024).

Spontaneous imbibition (SI) is one of the main recovery mechanisms in fractured carbonate reservoirs, where the wetting phase invades the matrix and displaces oil toward the fracture network (Karimova et al., 2023; Kashiri et al., 2024). This process is governed by capillary forces, which depend on interfacial tension (IFT), wettability, and pore structure. In strongly water-wet systems, capillary forces promote efficient imbibition of water into the matrix. In contrast, in oil-wet or mixed-wet carbonate formations, capillary driving forces are

significantly reduced or may even oppose the imbibition process, making imbibition extremely slow or even negligible (Alvarez & Schechter, 2017; Austad & Standnes, 2003; Ghandi et al., 2019).

Although waterflooding is generally considered as a secondary recovery method, owing to its simplicity and the ability to maintain reservoir pressure, its efficiency in fractured carbonate reservoirs is still considered to be low (Guo & Li, 2024; Hassan et al., 2023; Nande & Patwardhan, 2022; Sohal et al., 2016; Xu et al., 2020). The permeability of the fractures is much higher than the permeability of the rock, and hence the injected water mainly flows through the fractures and bypasses the oil in the rock matrix (Karimova et al., 2023; Moldabayeva et al., 2025; Song et al., 2026). The amount of oil left unrecovered is very high, as the injected water does not enter the oil zones (Ismailova et al., 2021; Zeinijahromi et al., 2015). The recovery of the oil is then left to the imbibition, which is considered to be a very inefficient process in the case of an oil-wet medium.

To address this problem, enhanced oil recovery (EOR) techniques have an important role in increasing the lifespan of the reservoir through increased displacement efficiency and mobilizing trapped oil (Alvarado & Manrique, 2010; Shakeel et al., 2024; Yuan et al., 2024). Among all EOR techniques, chemical-based EOR methods have attracted significant attention for altering the interfacial interactions between rock and fluids (Fani et al., 2022; Jain et al., 2022). Low-salinity water (LSW), ion-modified brine, surfactant, alkali, and nanoparticles have been widely reported in literature for altering wettability, decreasing IFT, and increasing mobilization of trapped oil (Ahmadi et al., 2021; Al-Nofli et al., 2018; Druetta & Picchioni, 2020; Hincapie et al., 2022; Kamal et al., 2017; Wang et al., 2024; Zhang & Austad, 2006).

In recent years, hybrid EOR approaches, wherein multiple EOR techniques are applied sequentially or simultaneously to synergistically improve oil displacement efficiency, have gained increasing attention because they can enhance oil recovery beyond what standalone methods can achieve (Araz & Kamyabi, 2021; Jafarbeigi et al., 2022a; Sekerbayeva et al., 2020). However, the efficiency of such hybrid systems strongly depends on the sequence of application, including whether chemical agents are injected directly or after LSW pretreatment. The choice of technique and the sequence of application significantly influence SI behavior, particularly in fractured formations. Despite numerous studies have investigated individual chemical EOR methods for improving oil recovery from carbonate reservoirs, the combined influence of wettability alteration, IFT reduction, and ionic interactions on SI behavior remains

insufficiently understood. In particular, a systematic experimental evaluation of different hybrid EOR sequences is necessary to better understand the mechanisms controlling SI and oil recovery in carbonate formations.

1.2 Objectives of the Thesis

1.2.1 Main Objectives

The main objective of this thesis is to systematically investigate the effectiveness of various chemical EOR techniques in improving SI and oil recovery in carbonate formations. Emphasis is given in understanding the mechanisms of SI improvement techniques, including wettability alteration and IFT reduction, as well as the effects of hybrid EOR techniques on oil recovery performance. The main objective is to identify which EOR design is more effective in accelerating the SI process and thus offer guidelines on how to design best EOR techniques in fractured carbonate reservoirs.

1.2.2 Specific Objectives

To achieve the main objective, the following specific objectives are addressed:

- To evaluate the effectiveness of standalone chemical EOR methods - including LSW, surfactant, alkali, and nanoparticle solutions - in enhancing SI and improving oil recovery from carbonate rocks through laboratory SI experiments.
- To investigate the performance of hybrid EOR approaches by combining LSW with surfactants, alkali, and nanoparticles in order to explore potential synergistic effects between wettability alteration and IFT reduction mechanisms.
- To analyze the influence of injection sequencing on SI behavior by comparing different sequences of chemical application and evaluating their impact on oil recovery in fractured carbonate systems.
- To assess the effect of ion-modified LSW on the efficiency of the most promising hybrid EOR method in order to determine whether optimization of brine ionic composition can further enhance SI and oil recovery.

- To investigate the mechanisms governing SI enhancement by conducting contact angle (CA) measurements and IFT experiments to quantify wettability alteration and interfacial effects associated with different EOR strategies.

1.3 Scope of Work

This study focuses on the laboratory investigation of SI in carbonate rocks under different chemical EOR conditions. The experimental work is based on SI tests to evaluate the performance of standalone and hybrid EOR methods, including LSW, surfactants, alkali, and nanoparticles. The influence of injection sequencing and ion-modified LSW was also examined to determine their impact on oil recovery. In addition, CA and IFT measurements were conducted to analyze the mechanisms governing oil recovery enhancement, particularly wettability alteration and IFT reduction. The study is limited to laboratory-scale experiments aimed at improving the understanding of SI-driven recovery in carbonate formations.

2. LITERATURE REVIEW

2.1 Fundamentals of Spontaneous Imbibition

SI is a key process in porous media, in which a wetting fluid invades a porous medium and displaces a non-wetting fluid, without any external pressure gradient. This process has a vital influence on fluid transport and hydrocarbon recovery from porous media, especially when the medium is dominated by capillary forces compared to viscous forces (Ashraf et al., 2023; Ashraf & Gupta, 2025; Karimova et al., 2023; Kashiri et al., 2024; Meng et al., 2017). For hydrocarbon reservoirs, SI has a critical influence when fluid transport takes place in porous media with different permeability values, such as matrix-fractures.

The driving force behind the occurrence of SI is the capillary pressure, which is given as the difference between the wetting phase and the non-wetting phase pressures. The capillary pressure is caused by the combined effect of IFT, wettability, and pore geometry (Li et al., 2017; Mo et al., 2019). It is given mathematically by the Young-Laplace equation as shown in Equation 2.1:

$$P_c = \frac{2\sigma\cos\theta}{r} \quad (2.1)$$

where P_c is the capillary pressure, σ is the IFT between the immiscible fluids, θ is the contact angle representing wettability, and r is the effective pore radius.

This relationship also implies that capillary pressure is directly proportional to $\sigma\cos\theta$, again highlighting the significant impact of IFT and wettability. The sign and magnitude of $\cos\theta$ will define the direction and efficiency of imbibition.

In water-wet systems, where θ is less than 90° , capillary forces are positive and will drive the imbibition of the wetting phase into the porous medium. Conversely, in oil-wet systems, where θ is greater than 90° , capillary forces are diminished or even negative, thus inhibiting or completely stopping imbibition (Alvarez & Schechter, 2017; Austad & Standnes, 2003; Ghandi et al., 2019).

This is why matrix wettability is a key parameter in SI. Oil displacement is successful in systems that are strongly water-wet, but in oil-wet systems, poor capillary forces make oil recovery more difficult (Fernø, 2012; Karimova et al., 2023; Standnes, 2004; Yu et al., 2008).

Experiments have shown that wettability plays a significant role in imbibition performance. Babadagli (2003) conducted SI experiments in a system of water-wet sandstone and oil-wet carbonate rock. Figure 1 illustrates the results of these experiments. High RF was achieved in a system of water-wet sandstone (more than 50% OOIP), while lower recovery was observed in oil-wet carbonate rock (about 5% OOIP) when a brine solution was used. The use of a surfactant solution increased oil recovery in oil-wet carbonate rock up to approximately 9-10% OOIP, indicating that modifying interfacial conditions can partially overcome wettability constraints.

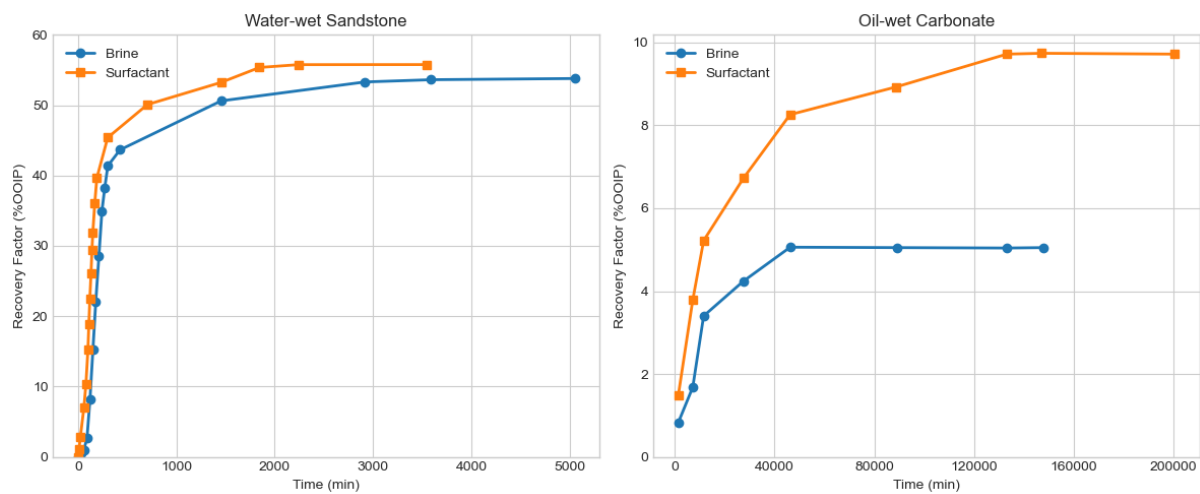


Figure 1. Comparison of SI recovery in water-wet sandstone and oil-wet carbonate systems for brine and surfactant solutions (adapted from Babadagli, 2003). Reproduced with permission from Elsevier.

Similar observations were made by Rezaveisi et al. (2012), who conducted an experimental study on the process of SI in fractured porous media for different wettability conditions. The results of the experiments are presented in Figure 2 and indicate that the recovery of oil decreases drastically for an increase in oil-wetness conditions. The recovery decreases from 37% OOIP for fully water-wet conditions to less than 5% OOIP for 100% oil-wet conditions. In addition, an increase in oil-wetness conditions by a small percentage decreases the recovery efficiency significantly, highlighting the strong sensitivity of imbibition performance to wettability state.

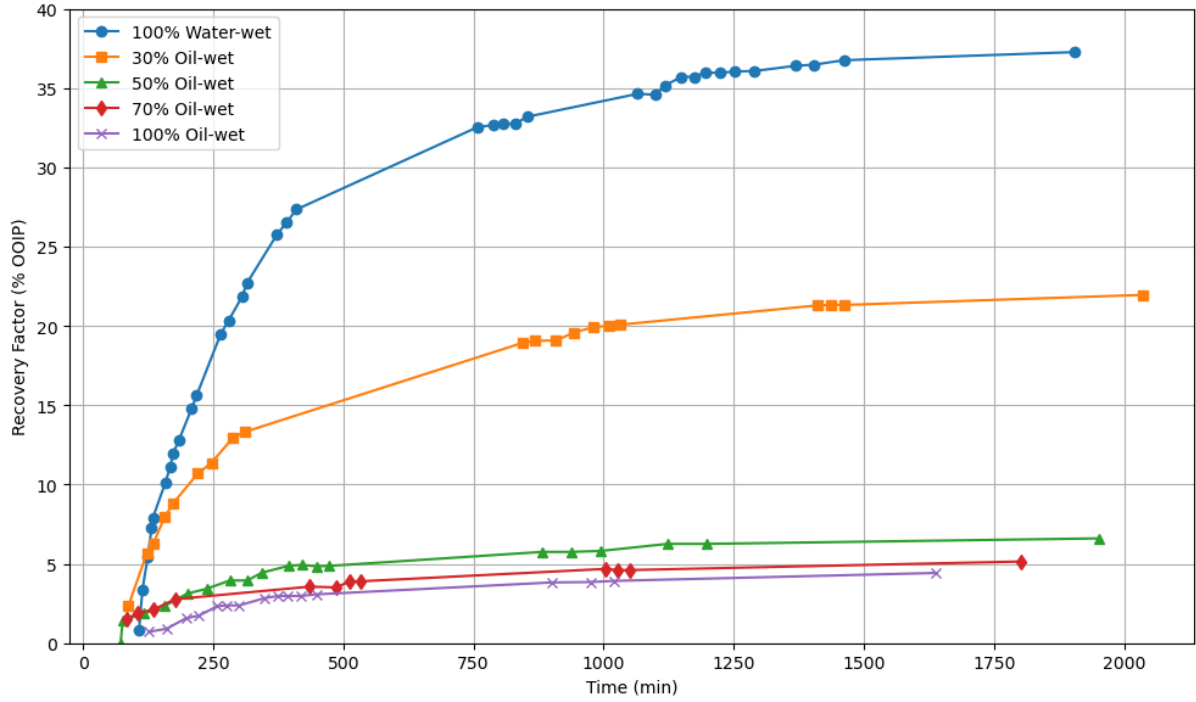


Figure 2. Influence of wettability on SI behavior in fractured porous media (adapted from Rezaveisi et al., 2012). Reproduced with permission from Elsevier.

Though wettability is the dominant factor in controlling the direction of SI, IFT is also important in controlling the magnitude of capillary forces and fluid mobilization. As indicated in the Young-Laplace relationship in Equation 2.1, capillary pressure is directly proportional to IFT. Therefore, an increase in IFT will result in an increase in capillary pressure, thus increasing the capillary driving force for imbibition (Li et al., 2017; Mo et al., 2019). However, the effect of IFT on SI is complex. On one hand, an increase in IFT will result in an increase in capillary pressure, thus increasing capillary trapping of oil. On the other hand, decreasing IFT will result in an increase in oil mobilization but will reduce the capillary driving force. Therefore, an optimal balance is necessary to maximize the efficiency of SI (Hou et al., 2015; Karimova et al., 2023; Santanna et al., 2014; Xu et al., 2019).

At ultra-low IFT, capillary forces diminish significantly, and the imbibition process may transition from capillary-dominated to gravity-influenced flow (Karimova et al., 2023; Xu et al., 2019). Under such conditions, gravity forces arising from density differences between the injected fluid and oil can contribute to fluid redistribution, particularly in fractured reservoirs. This behavior has been described using the inverse Bond number, which characterizes the relative importance of capillary and gravity forces (Schechter et al., 1994; Xu et al., 2019). The inverse Bond number was expressed as in Equation 2.2:

$$N_B^{-1} = \frac{C\sigma\sqrt{\frac{\phi}{k}}}{\Delta\rho gh} \quad (2.2)$$

where N_B^{-1} is the Bond number, σ is the IFT between the immiscible fluids, ϕ is porosity, k is permeability, $\Delta\rho$ is the density difference between the immiscible fluids, g is the gravitational acceleration, h is the height of the core and C is constant which depends on pore geometry. It has been reported that capillary forces dominate imbibition at high values of this parameter, whereas gravity-driven flow becomes significant at low values, with both mechanisms contributing under intermediate conditions (Schechter et al., 1994; Xu et al., 2019).

Some studies have proved that the reduction of IFT can enhance oil recovery during the process of SI (Hou et al., 2015; Qi et al., 2022; Santanna et al., 2014). Figure 3 is used to show the experimental data reported by Schechter et al. (1994), which proved that the reduction of IFT from 38.1 to 0.10 mN/m can enhance the ultimate recovery up to 90% OOIP during the transition from capillary-dominated to gravity-driven flow. Moreover, it is proved by Qi et al. (2022) that the reduction of IFT to the level of 10^{-2} mN/m can improve the efficiency of imbibition by enhancing the oil mobility and oil droplet transport.

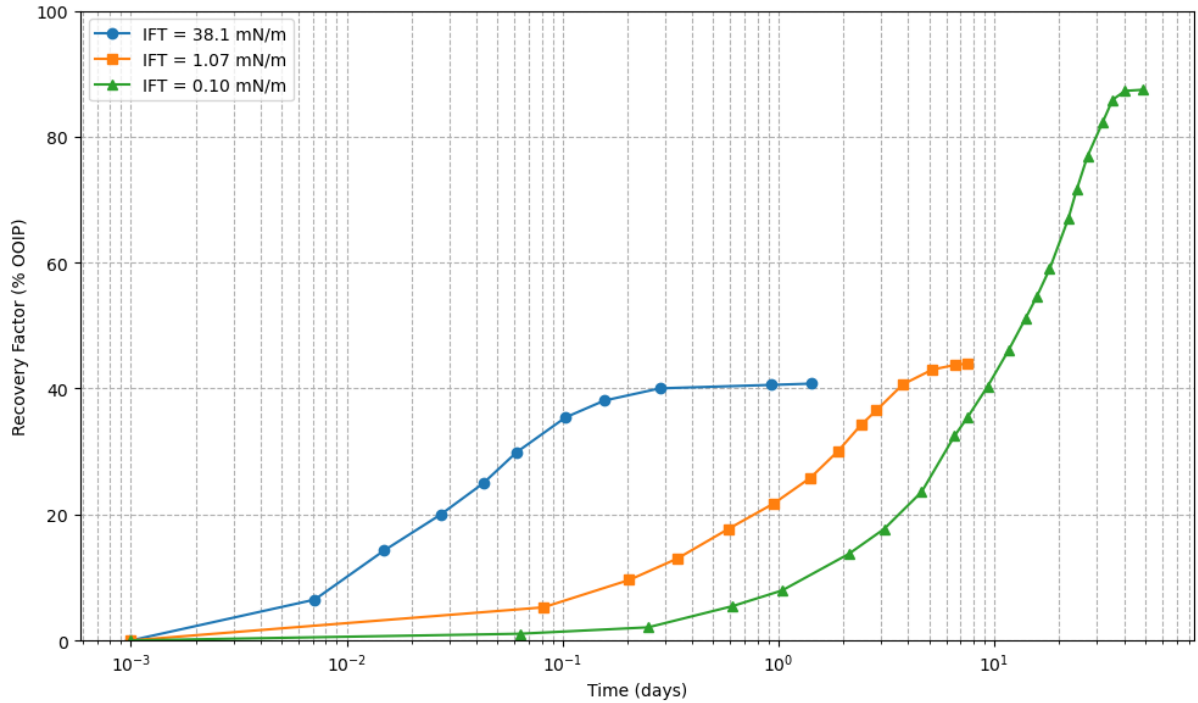


Figure 3. Effect of IFT on recovery factor during SI (adapted from Schechter et al., 1994).

Pore structure and rock properties also play an important role in the imbibition process. These properties include pore size distribution, permeability, porosity, and heterogeneity. Pores with small sizes produce high capillary pressure but limit the flow due to low permeability. On the contrary, pores with larger sizes allow more fluid flow but produce low capillary pressure. The heterogeneity of the rock also causes non-uniform imbibition fronts due to the difference in pore structure, leading to complex flow patterns in the medium (Karimova et al., 2023; Mahdaviara et al., 2022; Yang et al., 2016). Table 1 summarizes the effect of different parameters on the performance of SI in fractured carbonate formations.

Table 1. Key factors controlling spontaneous imbibition (adapted from Karimova et al., 2023)

Parameter	Effect on SI Recovery	Dominant Mechanism	Remarks
Gravity	Increases (especially at ultra- low IFT)	Gravity drainage	Becomes dominant at ultra-low IFT
IFT reduction	Increases ultimate recovery	Reduced capillary forces, increased mobility	Transition from capillary to gravity-driven flow
Wettability (oil-wet to water-wet)	Strong increase	Capillary force increase	Most critical controlling factor
Initial water saturation	Decreases recovery if high	Reduced capillary suction	Limits imbibition potential
Porosity	Increases recovery	Storage capacity	Higher pore volume available
Permeability	Increases rate and recovery	Reduced flow resistance	Enhances fluid transport
Oil viscosity	Decreases recovery	Viscous resistance	High viscosity limits mobility
Temperature	Increases recovery	Reduced viscosity, lower IFT	Enhances fluid mobility

In laboratory studies, SI is commonly investigated using four types of boundary conditions that control fluid flow behavior and the interaction between co-current and counter-current mechanisms. Co-current imbibition refers to the simultaneous flow of the wetting and non-wetting phases in the same direction, whereas counter-current imbibition occurs when the two phases flow in opposite directions (Mirzaei-Paiaman et al., 2011; Pooladi-Darvish & Firoozabadi, 2000). These flow mechanisms are strongly influenced by the imposed boundary conditions, which include all faces open (AFO), one open end (OOE), two ends open (TEO), and one open end with one isolated end (OOCO) (Karimova et al., 2023). In the most common AFO configuration, all faces of the core are exposed, allowing both co- and counter-current flow to occur simultaneously, although their individual contributions cannot be distinguished (Meng et al., 2017). In the OOE condition, the flow is restricted to one open face, thus

encouraging mostly counter-current imbibition with capillary forces as the driving force (Mason & Morrow, 2013). In the TEO configuration, both ends are open to flow, thus facilitating linear flow with co- and counter-current processes occurring simultaneously but being difficult to distinguish (Meng et al., 2017; Yu et al., 2008). The OOCO configuration facilitates better distinction of flow mechanisms due to the isolation of one end, thus being able to quantify co- and counter-current flow mechanisms independently (Ahmadi et al., 2021; Andersen & Ahmed, 2021). These boundary conditions are shown in Figure 4. They are critically important in understanding laboratory findings and the imbibition process under different reservoir conditions.

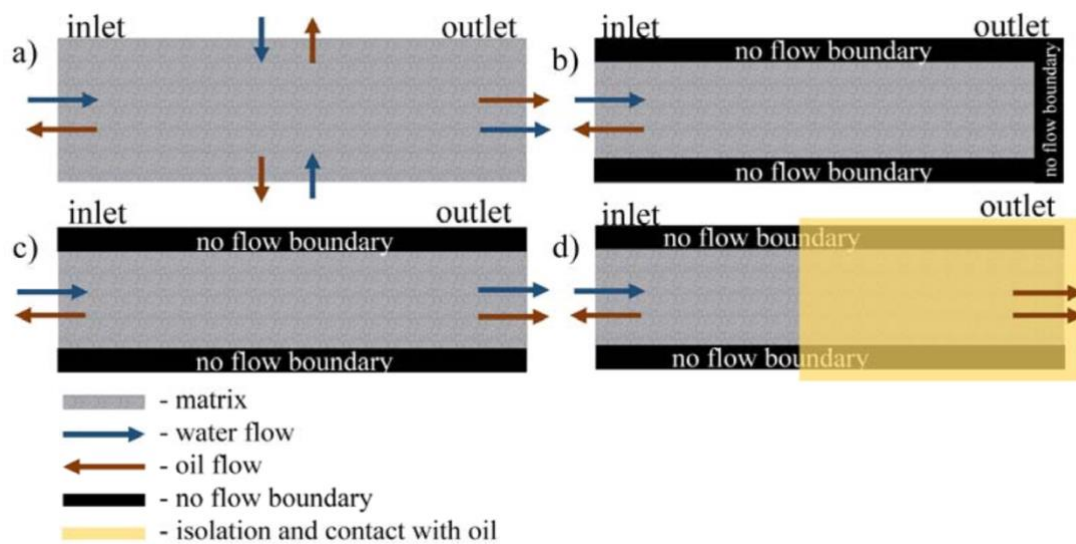


Figure 4. Schematic representation of boundary conditions during SI tests: (a) AFO, (b) OOE, (c) TEO, (d) OOCO (Karimova et al., 2023)

In addition to ultimate oil recovery, the rate of SI is a key parameter in determining the efficiency of fluid exchange between matrix and fractures in carbonate reservoirs. Although capillary pressure controls the driving force for imbibition, the rate of fluid displacement during imbibition is controlled by a dynamic balance among capillary forces, viscous resistance, and fluid-rock interactions (Ghandi et al., 2019; Morrow & Mason, 2001; Ruiz-Gutiérrez et al., 2022; Schechter et al., 1994).

SI typically exhibits two distinct stages: an early-time period characterized by rapid oil production driven by strong capillary forces, followed by a late-time stage where the rate declines as the system approaches equilibrium. The early-time imbibition rate is particularly important in practical applications, as it determines how quickly oil can be mobilized from the

matrix into the fracture network. This behavior is commonly described by a square-root-of-time ($\sqrt{t}$) scaling, reflecting that recovery increases rapidly initially and gradually slows as the imbibition front advances (Andersen, 2023; March et al., 2016; Velasco-Lozano & Balhoff, 2022; Washburn, 1921).

Both IFT and wettability are important factors that govern the imbibition rate. It is evident that a decrease in IFT, although to a moderate extent, may improve the oil mobility and subsequently the rate of imbibition. On the other hand, an ultra-low level of IFT may reduce the driving force for imbibition, resulting in a slower imbibition rate. For example, Zhang et al. (2006) reported that the injection of surfactants significantly increased the rate of imbibition, such that oil production was observed to start after an hour compared to significant delay in the brine system. Furthermore, experimental results indicate that, at low levels of IFT, the flow pattern changes from capillary-dominated counter-current flow to gravity-driven co-current flow, resulting in a slower imbibition rate but a potentially higher oil recovery at later times (Al-Attar, 2010; Schechter et al., 1994). Moreover, a change to a more water-wet condition sustains a higher rate of imbibition for a longer time (Ghandi et al., 2019; Hirasaki & Zhang, 2004; Morrow & Mason, 2001). For example, experiments on SI processes employing a modified LSW system, as performed by Karimi et al. (2016), showed that a change in wettability increased the oil recovery and consequently the rate of imbibition. The results indicate that the effect of low levels of IFT on the rate of imbibition is not monotonic, and an optimal balance is required for an increased rate of imbibition.

Experiments of SI in porous media are the primary approach used to quantify oil production during imbibition processes and to predict the rate and extent of oil recovery in fractured reservoirs. Laboratory techniques such as the weighing method and Amott cell measurements provide direct evaluation of oil production, while more advanced imaging methods, including CT scanning, nuclear tracer imaging (NTI), and nuclear magnetic resonance (NMR), enable detailed characterization of fluid distribution and saturation profiles within the porous medium (Akin & Kavscek, 2003; Baldwin & Spinler, 2002; Ersland et al., 2010; Fernø et al., 2013; Karimova et al., 2023; Meng et al., 2017). These experimental methods not only allow assessment of recovery performance but also help distinguish between co-current and counter-current flow mechanisms under different boundary conditions. Consequently, SI experiments play a critical role in understanding fluid transport mechanisms and in scaling laboratory observations to field-scale recovery predictions in fractured carbonate reservoirs.

Overall, SI is governed by a complex interplay of wettability, IFT, pore structure, and boundary conditions. Therefore, understanding the fundamental mechanisms of SI is essential for interpreting experimental observations and for designing effective strategies to enhance oil recovery.

2.2 Chemical EOR Methods

2.2.1 Surfactant Flooding

Surfactant flooding is a chemical EOR process that is extensively and successfully implemented in the petroleum industry, in which surface-active agents are injected into the reservoir to increase the effectiveness of the oil displacement process. The surfactant flooding process is very effective in the mobilization of the residual oil trapped in the pore volume of the reservoir through the modification of the interfacial and surface properties of the rock-fluid system. Apart from the surfactants, co-surfactants are added to the surfactant flooding process in order to improve the phase behavior and stability properties of the injected solution in the reservoir, considering the temperature, pressure, and salinity of the reservoir (Hirasaki et al., 2011; Iglaier et al., 2010; Shakeel et al., 2024; Sheng, 2015).

The effectiveness of surfactant flooding is mainly attributed to its capacity to substantially reduce IFT between crude oil and the aqueous phase. The reduction in IFT reduces capillary forces, thus increasing oil mobilization while reducing residual oil saturation. Furthermore, surfactants are able to alter rock wettability to water-wet, thereby increasing SI and matrix-fracture fluid exchange in fractured formations (Negin et al., 2017; Reppert et al., 1990; Shakeel et al., 2024). All these mechanisms contribute to the effectiveness of surfactant flooding, especially in carbonate formations where oil-wetness is common.

Surfactant molecules are amphiphilic in nature, having hydrophilic (polar) head and hydrophobic or lipophilic (nonpolar) tail. Due to this nature, they tend to accumulate at the interface between two liquids, such as oil and water, and decrease the interfacial energy, thereby creating an emulsion or micro-emulsion. This helps in the transport of the oil by effectively dispersing the oil droplets in the aqueous phase and facilitating its transport through the pores. The efficiency of the surfactant is largely dependent on the amount of the concentration, usually above the critical micelle concentration (CMC), to effectively function (Druetta & Picchioni, 2020; Kamal et al., 2017; Shakeel et al., 2024).

Surfactants are commonly classified based on the charge of their head groups into anionic, cationic, and nonionic types. Anionic surfactants are widely used in EOR applications due to their effectiveness in reducing IFT, relatively low cost, and good stability under a range of reservoir conditions. Cationic surfactants, while effective for wettability alteration in carbonate rocks due to electrostatic interactions with negatively charged oil components, are often limited by higher cost and potential adsorption losses. Nonionic surfactants, which carry no charge, are generally used as co-surfactants to improve formulation performance and phase behavior (Jia et al., 2019; Kamal et al., 2017; Thomas et al., 1993; Zhao et al., 2005). Figure 5 represents the main mechanisms of hydrocarbon recovery involved in surfactant flooding.

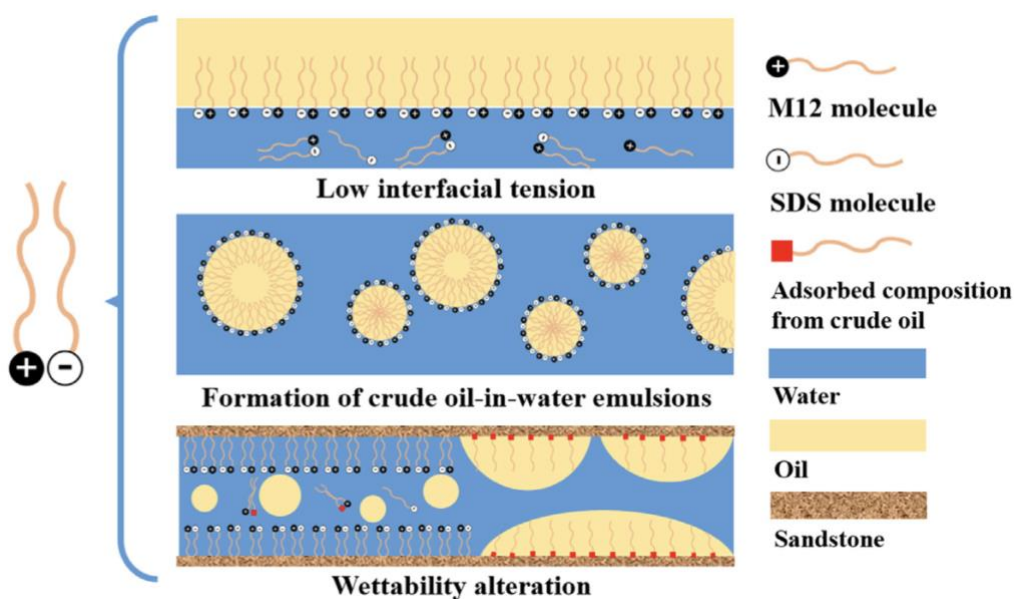


Figure 5. Schematics of oil recovery mechanisms by surfactant flooding (Jia et al., 2019)

Several experimental studies have demonstrated the effectiveness of surfactant flooding in enhancing SI in carbonate reservoirs. Representative results from the literature are summarized in Table 2. The overall effect is a significant increase in both the rate and extent of oil recovery compared to water alone, by imbibition. However, surfactant performance is strongly influenced by salinity, hardness, and adsorption, which can reduce effectiveness in high-salinity brines or carbonate formations if not properly optimized.

Table 2. Summary of surfactant performance in SI experiments in carbonates

Study	Surfactant Type	Dominant Mechanism	Recovery Improvement
(Standnes et al., 2002)	Cationic	IFT reduction with wettability alteration	Up to 40-50% OOIP

(Austad & Standnes, 2003)	Cationic	Wettability alteration	Up to ~70% OOIP in chalk cores; up to ~90% in dolomite cores
(Hirasaki & Zhang, 2004)	Anionic	IFT reduction	Up to 44% OOIP
(Seethepalli et al., 2004)	Anionic	IFT reduction with wettability alteration	Up to 56% OOIP
(Leslie Zhang et al., 2006)	Anionic	IFT reduction with wettability alteration	Imbibition started within 1 hour, indicating acceleration of imbibition rate
(Chen & Mohanty, 2013)	Cationic	IFT reduction	Up to 50-65% OOIP
(Karimi et al., 2016)	Cationic	IFT reduction with wettability alteration	Up to 82-95% OOIP
(Mohammadi et al., 2019)	Anionic	IFT reduction	Up to ~55% OOIP
(Ahmadi et al., 2020)	Anionic	IFT reduction	Up to ~65% OOIP
(Souayeh et al., 2021)	Nonionic and anionic	IFT reduction with wettability alteration	Up to 31-74% OOIP
(Xu et al., 2021)	Anionic	Wettability alteration	Up to ~38% OOIP
(Ali Akarim Al-Sayegh, 2022)	Anionic	IFT reduction with wettability alteration	Up to ~86% OOIP
(Xu et al., 2025)	Anionic	Wettability alteration	Up to ~20.5% OOIP
(Xu et al., 2025)	Anionic	IFT reduction with wettability alteration	Up to ~25.6% OOIP

2.2.2 Alkali Flooding

Alkali flooding is an EOR method that uses an injection of an alkaline solution into the reservoir to improve oil recovery using in-situ chemical reactions and physicochemical changes. Alkali flooding increases oil recovery based on the generation of in-situ surfactants using chemical reactions between injected alkalis and acidic components of crude oil (Belhaj et al., 2020; Rahimi et al., 2022). Alkalis such as sodium carbonate (Na_2CO_3) or sodium hydroxide (NaOH), when injected into the reservoir, react with the acid components of crude oil to produce natural soap, thus reducing the IFT between crude oil and water (Hincapie et al., 2022; Schumi et al., 2020; Shakeel et al., 2024). The reduction in IFT enables the mobilization of oil and increases capillary imbibition in fractured carbonate reservoirs. Figure 6 presents the schematics of IFT reduction mechanisms performed using alkali flooding. Under favorable conditions, these mechanisms can accelerate imbibition and increase recovery. However, the effectiveness of alkali flooding is greatly dependent on the crude oil composition, especially total acid number (TAN). Alkali flooding is less effective in crude oils with low TAN due to the limited amounts of acidic components, thus reducing in-situ surfactant formation (Hincapie et al., 2022). Moreover, alkalis can have negative effects, especially scaling through

precipitation of Ca^{2+} and Mg^{2+} carbonates in hard brines, consumption by reservoir minerals, and pH instability in diluted brines. Therefore, alkali flooding is less effective in fractured carbonate reservoirs unless carefully adjusted to crude chemistry and brine composition.

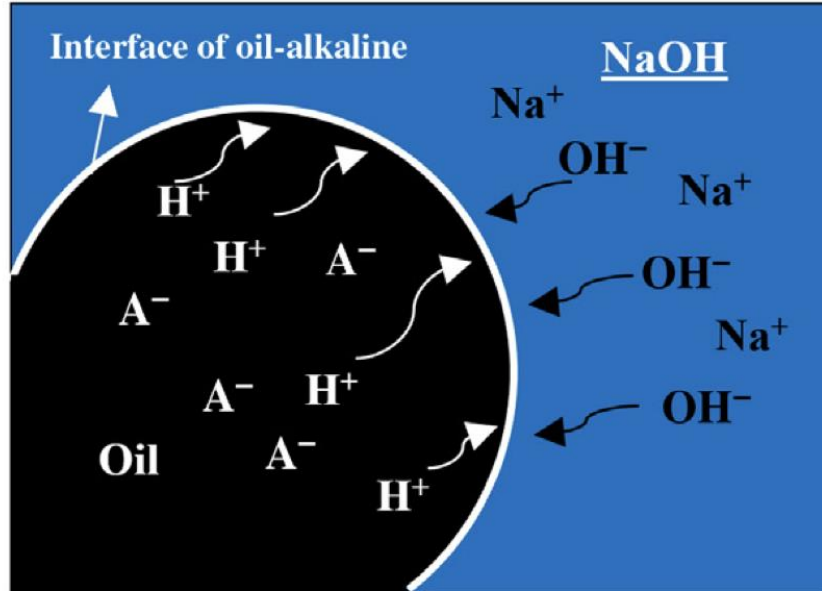


Figure 6. Schematics of IFT reduction by alkali flooding (Mahdavi & Zebarjad, 2018).

Compared to surfactant-based systems, standalone alkali flooding in carbonate SI studies generally demonstrates moderate recovery improvement, with its performance highly dependent on crude oil acidity and brine composition (Gong et al., 2016; Mahdavi & Zebarjad, 2018).

2.2.3 Nanoparticles Flooding

Nanoparticles have emerged as promising agents for enhanced oil recovery due to their unique physicochemical properties and ability to interact at both rock-fluid and fluid-fluid interfaces. One of the primary mechanisms of nanoparticle flooding is wettability alteration. In SI processes, nanoparticles can adsorb onto mineral surfaces and form a thin hydrophilic layer, thereby altering wettability from oil-wet toward more water-wet conditions, which strengthens capillary forces and promotes water entry into the matrix (Hincapie et al., 2022; Khoramian et al., 2022; Maghzi et al., 2012; Ogolo et al., 2012). The successful wettability alteration effect was reported in several research studies, which are summarized in Table 3.

Table 3. Summary of wettability alteration by nanoparticles flooding in carbonates

Study	Nanoparticle Type	Initial Contact Angle (°)	Final Contact Angle (°)
(Esfandiyari et al., 2014)	SiO ₂	90	26
	Al ₂ O ₃	90	71
	TiO ₂	90	57
(Moradi et al., 2015)	SiO ₂	134	82
(Nazari et al., 2015)	SiO ₂	140	79
	Al ₂ O ₃	140	104
	TiO ₂	140	53
(Khoramian et al., 2022)	SiO ₂	147	85

Apart from the effect of wettability, nanoparticles may also affect oil-water IFT. Even though the reduction of oil-water IFT by nanoparticles is generally less significant compared to that of surfactants, nanoparticles may also have synergistic effects when used with other chemicals. Nanoparticles may adsorb at the oil-water interface and stabilize emulsions, which can improve microscopic displacement efficiency and prevent oil droplet coalescence (Kamal et al., 2017; Lan et al., 2007; Moradi et al., 2015).

Another unique mechanism involving nanoparticles is the formation of structural disjoining pressure. Nanoparticles that accumulate in aqueous films, which are situated between the rock surface and the oil, can create a repulsive force that aids in the detachment of oil droplets that are adhered to the surface. This is especially significant for oil-wet carbonate reservoirs, where the use of a conventional capillary displacement is limited. The use of nanoparticles improves the stability of the aqueous films, ensuring that oil is continuously detached and moved (Sircar et al., 2022). Figure 7 illustrates the different oil recovery mechanisms involving nanoparticles flooding.

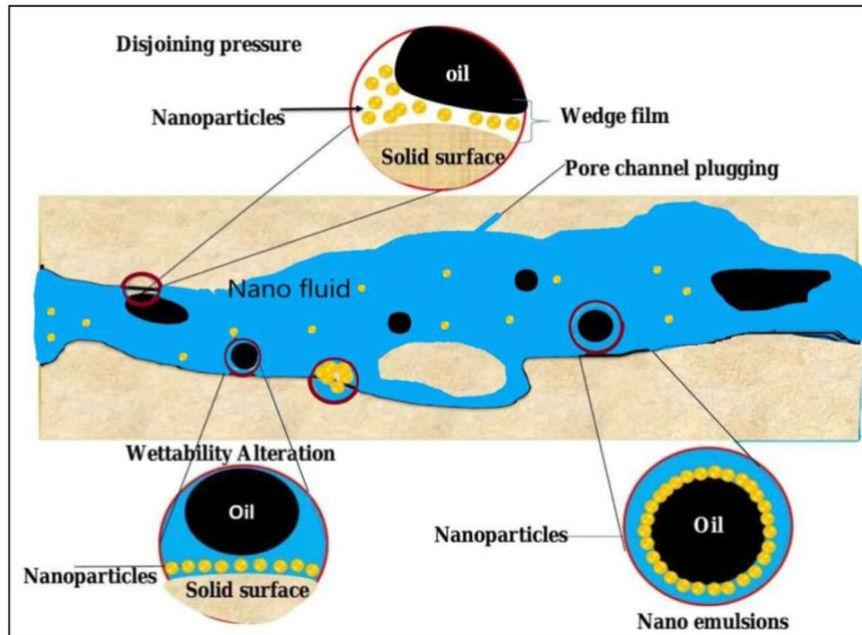


Figure 7. Schematics of nanoparticles flooding (Sircar et al., 2022)

Nanoparticles usually work more slowly than surfactants because they need time to adsorb and build up enough surface coverage. However, they can support moderate recovery over longer periods of time. The effectiveness of nanoparticles depends on factors like concentration, particle size, and the salinity of the brine. Low ionic strength brines can make nanoparticles dispersions unstable, which can cause aggregation or sedimentation. On the other hand, the right salinity levels can keep the dispersions stable and improve their EOR effect (Shakeel et al., 2024).

2.2.4 Low Salinity Water Flooding

Low salinity water (LSW) flooding has gained significant attention as an effective and relatively low-cost EOR method, particularly in carbonate reservoirs. Unlike conventional waterflooding, LSW involves the injection of brine with reduced salinity and modified ionic composition to alter rock-fluid interactions and improve oil recovery. In the context of SI, LSW plays a critical role by enhancing wettability alteration and promoting capillary-driven oil displacement (Ahmadi et al., 2021; Darvish et al., 2019; Zhang & Austad, 2006).

The change in wettability is the main and acceptable mechanism to improve oil recovery in carbonate rocks with LSW. The main mechanisms leading to alterations in wettability have been discussed in many works on the subject, and it was concluded that there can be a few possible factors influencing its successfulness. Calcite dissolution is defined as the first

possible mechanism, which is based on precipitation of calcite (Ca^{2+}) in form of anhydrite (CaSO_4) as a result of presence of excess sulfate ions (SO_4^{2-}) in LSW. This process causes ionic imbalance and further release of oil molecules from the rock surfaces due to the chemical equilibrium being reinstalled by dissolved calcite. This process potentially alters the wettability and enhances the pore connectivity (Al-Shalabi et al., 2015; Karimova et al., 2023; Sagbana et al., 2023). Schematics of calcite dissolution during LSW are presented in Figure 8.

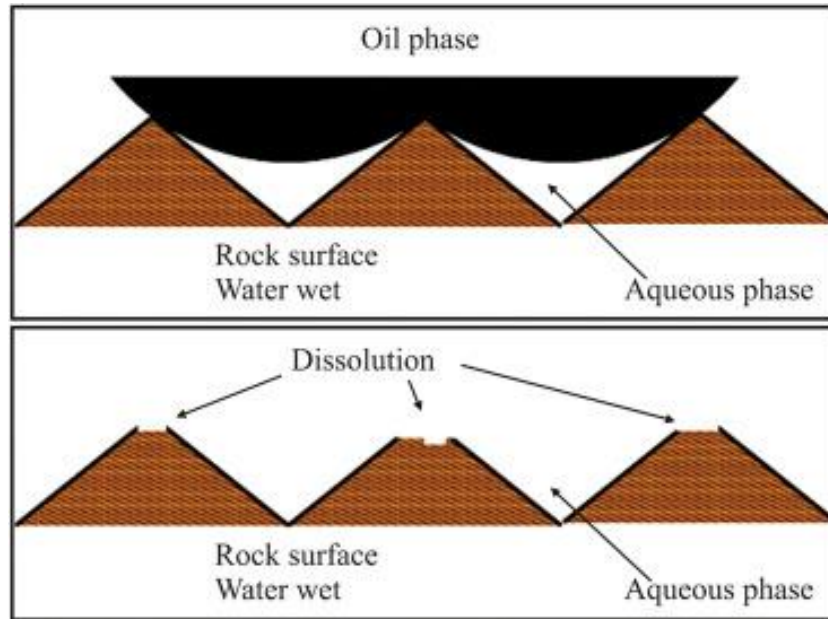


Figure 8. Calcite dissolution mechanism (Sagbana et al., 2023)

Another mechanism is the expansion of electrical double-layer (EDL), which is defined as an accumulation of the ions around the surface caused by charged rock surface and liquids containing ions, such as brine. During LSW flooding, the reduction of ionic strength and the exchange of cations, such as calcium (Ca^{2+}), lead to changes in the rock surface charge, making it less positive. This causes the EDL to expand, increasing repulsion forces between the rock and oil, stabilizing the brine film, and detaching oil from the rock surface, ultimately altering the wettability to a more water-wet state (Al-Nofli et al., 2018; Karimova et al., 2023; Lee et al., 2010; Sagbana et al., 2023). Figure 9 below shows the process of electrical double-layer expansion.

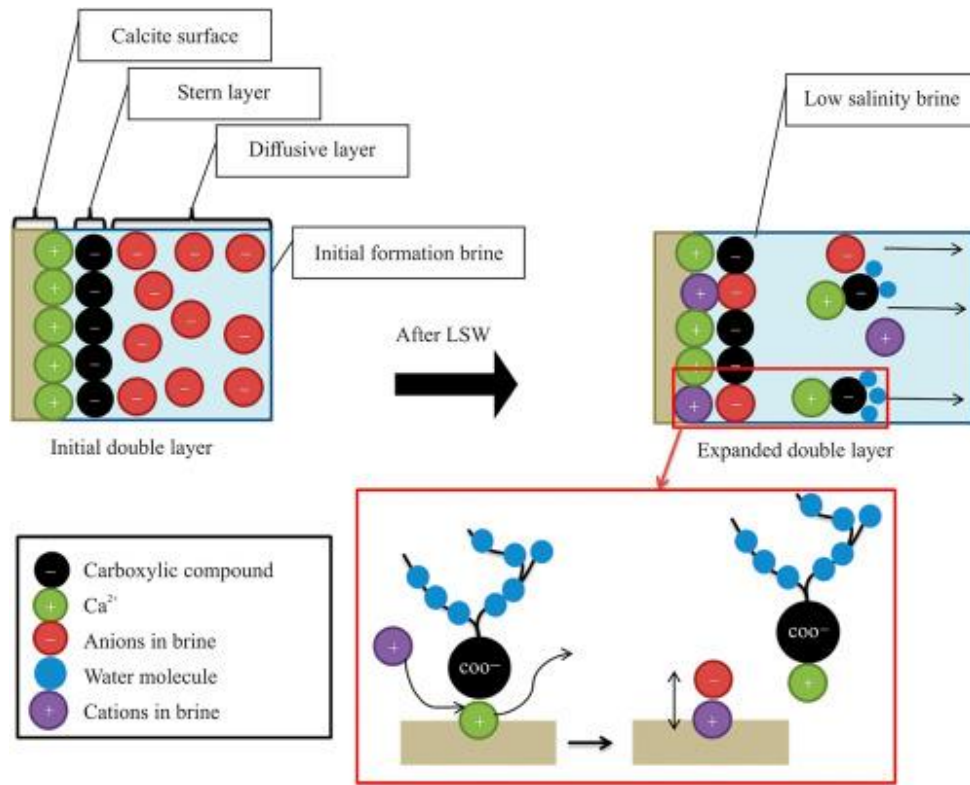


Figure 9. Electrical double-layer mechanism (Sagbana et al., 2023)

The third mechanism to be discussed is the multiple ion exchange (MIE) which occurs when Potential Determining Ions (PDIs), such as Ca^{2+} , Mg^{2+} and SO_4^{2-} , are present in sufficient amount in injected LSW. This process resulted by the adsorption of negatively charged sulfate ions (SO_4^{2-}) on the surface of the positively charged carbonate rock, leading to reduction of the total positive charge of the rock. In such conditions, positively charged PDIs from injected brine adhere more easily to the carbonate surface and interact with surface-active oil components, leading to effective release of oil from the rock surface. As the oil molecules are released, the rock surface becomes less oil-wet and more water-wet (Al-Shalabi et al., 2015; Al-Shalabi & Sepehrnoori, 2016; Fathi et al., 2011; Moradpour et al., 2020; Sagbana et al., 2023). Figure 10 shows the process of MIE in carbonate rocks.

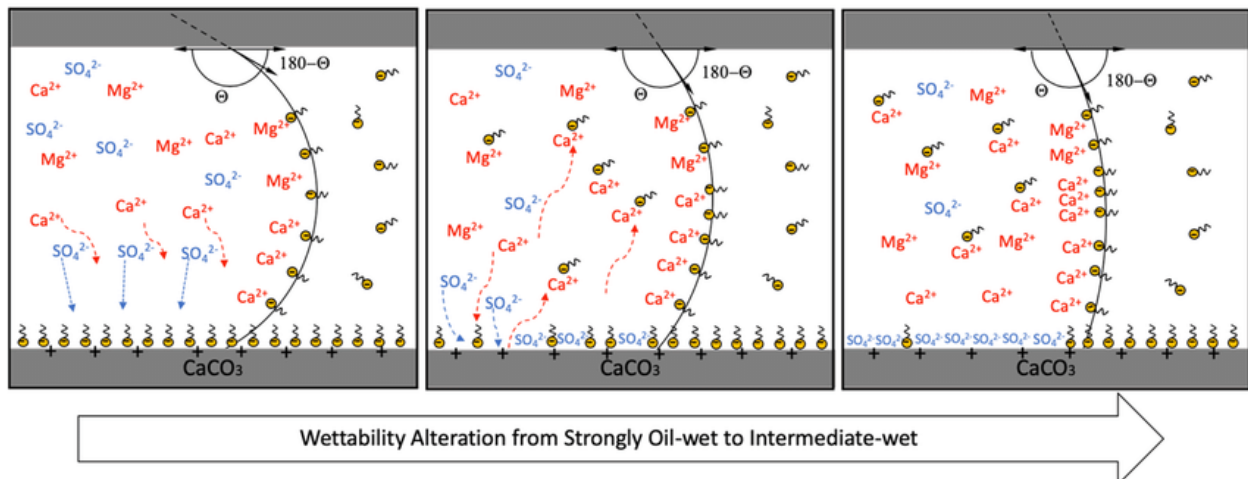


Figure 10. Multiple ion exchange mechanism in carbonate rock (Tawfik et al., 2019)

LSW can also trigger minor pH changes that further destabilize organic films and contribute to in-situ surfactant generation. Collectively, these mechanisms gradually shift wettability from oil-wet toward water-wet conditions, thereby increasing capillary forces and accelerating SI (Karimova et al., 2023; Kashiri et al., 2023). The recovery gain is usually small (a few percent of OOIP) and depends strongly on rock/mineralogy and crude composition, the underlying mechanisms remain debated despite many experimental studies. Representative results from the literature are summarized in Table 4.

Table 4. Summary of LSW performance in SI experiments in carbonates

Study	LSW TDS	Dominant Mechanism	Recovery Improvement
(Al-Harrasi et al., 2012)	~1945-38890 ppm	Wettability alteration	+16-21% OOIP
(Al-Shalabi & Sepehrnoori, 2016)	350-3500 ppm	Wettability alteration	+15-20% OOIP
(Shariatpanahi et al., 2016)	~3340 ppm	Wettability alteration	+10-15% OOIP
(Karimi et al., 2016)	~1960-12160 ppm	Wettability alteration	Up to ~75% OOIP
(Ahmadi et al., 2017)	~5× diluted SW	Wettability alteration with IFT reduction	+10-15% OOIP
(Zaeri et al., 2018)	~730-5850 ppm	Wettability alteration	+14% OOIP
(Hidayat et al., 2018)	500-10000 ppm	Wettability alteration	Incremental RF: up to 100% (10000 ppm); 68.75% (5000 ppm); lowest at 500 ppm
(Feldmann et al., 2020)	438-43800 ppm	Wettability alteration	Up to ~38.5% recovery
(Façanha et al., 2022)	1000-5000 ppm	Wettability alteration	About 2× higher recovery vs FW
(Feldmann et al., 2024)	4000 ppm	Wettability alteration	RF increased from ~0-2% (FW) to 36.1% (LSW)

(Karoussi et al., 2025)	1400 ppm	Wettability alteration	+4-6% OOIP
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In addition to salinity reduction, the ionic composition of the injected brine can also be modified to further enhance oil recovery. Ion-modified LSW flooding, also referred to as engineered water or smart water flooding, is an extension of conventional LSW flooding in which not only the total salinity but also the ionic composition of the injected brine is deliberately adjusted to maximize oil recovery. In carbonate reservoirs, this approach is particularly important because recovery is highly sensitive to the presence and concentration of PDIs, especially sulfate (SO_4^{2-}), calcium (Ca^{2+}), and magnesium (Mg^{2+}) (AlGeer et al., 2016; Ali Buriro et al., 2023; Al-Shalabi & Sepehrnoori, 2016; Kilybay et al., 2017). Unlike simple dilution, ion-modified LSW aims to optimize the balance between ionic strength reduction and ion-specific interactions at the rock-brine-oil interface, thereby promoting stronger wettability alteration and improved SI.

In the case of carbonate rocks, the surface is usually positively charged due to the presence of Ca^{2+} ions in the rocks (Ali Buriro et al., 2023; Zhang & Austad, 2006). This results in the strong adsorption of negatively charged acidic components of crude oil, which leads to oil-wet conditions and hinders water imbibition. The addition of sulfate ions acts as a key initiating step in altering this state. Sulfate ions are attracted to the positively charged carbonate surface, where they adsorb and reduce the surface charge. This process enables divalent cations such as Ca^{2+} and Mg^{2+} to approach the rock surface (Fathi et al., 2011; Standnes & Austad, 2000; Strand et al., 2006).

Calcium and magnesium ions, after the adsorption of sulfates, react with carboxylic groups present in the crude oil through MIE mechanism. The interaction reduces the bond strength between the rock and the components of oil, causing them to desorb and creating a water-wet system. The sequence of sulfate adsorption and divalent ion interaction is well accepted as the major mechanism responsible for ion-modified LSW flooding in carbonate reservoirs (Derkani et al., 2019; Zhang et al., 2007).

The effectiveness of this mechanism depends on the concentration of these ions, particularly sulfate, which has been considered the most critical component, as it acts as a catalyst, allowing the ions of Ca^{2+} and Mg^{2+} to become effective. While Ca^{2+} plays a key role in surface reactions, Mg^{2+} has also been identified as a strong PDI, especially at elevated temperatures, where it can replace Ca^{2+} in the carbonate lattice, with the extent of substitution increasing with temperature.

Thus, the interaction between SO_4^{2-} ions and either Ca^{2+} or Mg^{2+} is essential for effective wettability alteration. The efficiency of this mechanism improves with increasing temperature, indicating that it is governed by the combined and temperature-dependent action of these ions rather than their individual effects (Derkani et al., 2019; Gomari et al., 2006; Kasha et al., 2015; Zhang et al., 2006, 2007; Zhang & Austad, 2006).

Experimental studies confirm the effectiveness of these ions in increasing oil recovery compared to conventional LSW flooding operations. Sulfate ions in seawater are effective in increasing the rate of wettability alteration, while adjusting the concentrations of Ca^{2+} and Mg^{2+} ions further increases this effect, especially under high-temperature conditions. In the absence of sufficient sulfate or divalent cations, the effect of wettability alteration is very low, even when the salinity of the water is reduced (Fathi et al., 2011; Karimi et al., 2016; Shariatpanahi et al., 2016).

Another significant feature of ion-modified flooding is the interaction between PDIs and other ions in the brine solution. High concentrations of non-active ions, especially Na^+ and Cl^- , have been shown to inhibit the accessibility of sulfate and divalent cations to the rock surface (Mushabe et al., 2022). Therefore, lowering the concentrations of these dominant ions is expected to improve the effectiveness of SO_4^{2-} , Ca^{2+} and Mg^{2+} by increasing their interaction with the carbonate surface and enhancing ion exchange processes (Mushabe et al., 2022; Strand et al., 2008; Zhang & Austad, 2006). This implies that the efficiency of ion-modified flooding depends not only on the presence of active ions but also on the reduction of competing ions.

In summary, unlike other methods involving only the concept of dilution, ion-modified LSW flooding is considered to be more efficient and predictable in wettability alteration through the direct modification of surface charge, ion exchange processes, and fluid-rock interactions. A summary of relevant literature studies on ion-modified brine compositions and their impact on recovery from carbonate systems is presented in Table 5.

Table 5. Summary of ion-modified LSW imbibition studies in carbonates (Karimova et al., 2023)

Study	Brine	Temperature	Investigated Ion	PDI concentration change	Recovery Improvement
(Zhang & Austad, 2006)	Modified SW	70°C	SO_4^{2-}	From 0 to 0.096 mol/L	+40-45% OOIP
(Tweheyo et al., 2006)	Modified SW	70°C	Ca^{2+}	From 0.058 to 0.232 mol/L	+6% OOIP

(Zhang et al., 2007)	Modified SW	70°C	SO ₄ ²⁻	From 0 to 0.096 mol/L	+35% OOIP
			Ca ²⁺	From 0 to 0.045 mol/L	+30% OOIP
			Mg ²⁺	From 0 to 0.045 mol/L	+15% OOIP
(Yu et al., 2008)	Modified SW	50°C	SO ₄ ²⁻	4× increase	+9-15% OOIP
(Strand et al., 2008)	Modified SW	120°C	SO ₄ ²⁻	0 to 0.024 mol/L	+15% OOIP
(Yu et al., 2009)	FW and SW	130°C	SO ₄ ²⁻	0 to 0.024 mol/L	+7-8% OOIP
(Shariatpanahi et al., 2010)	Modified SW	110°C	SO ₄ ²⁻	0 to 0.032 mol/l	+7% OOIP
			NaCl	Na ⁺ : 2.62 to 0.068 mol/l Cl ⁻ : 3.643 to 0.136 mol/l	+13% OOIP
(Fernø et al., 2011)	Modified SW	130°C	SO ₄ ²⁻	0 to 0.094 mol/l	+6.5-35% OOIP
(Romanuka et al., 2012)	Synthetic SW	70°C	SO ₄ ²⁻	0.002 to 0.019-0.099 mol/l	+5-15% OOIP
(Shariatpanahi et al., 2016)	Modified FW	25°C	Mg ²⁺	0.054 to 0.66 mol/l	+5% OOIP
(Karimi et al., 2016)	Modified FW	75°C	SO ₄ ²⁻	0.0718 to 71.88 mol/l	+7% OOIP
			SO ₄ ²⁻ and Mg ²⁺	SO ₄ ²⁻ : 0.0718 to 71.88 mol/l Mg ²⁺ : 0.929 to 465.44 mol/l	+17% OOIP
(Ahmadi et al., 2020)	Modified SW	70°C	SO ₄ ²⁻	0.033 to 0.099 mol/l	+10% OOIP
			Mg ²⁺	0.073 to 0.219 mol/l	+4% OOIP
			Ca ²⁺	0.012 to 0.036 mol/l	+2% OOIP
			NaCl	Na ⁺ : 0.5525 to 0.068 mol/l Cl ⁻ : 0.6545 to 0.17 mol/l	+5% OOIP
(Shirazi et al., 2020)	Modified SW	60°C	SO ₄ ²⁻	3210 to 6410 ppm	+7% OOIP
			Mg ²⁺	1530 to 4589 ppm	+5% OOIP
			Ca ²⁺	460 to 920 ppm	+6% OOIP
(Mushabe et al., 2022)	Modified SW	96°C	SO ₄ ²⁻	0.056 to 0.112 mol/l	+3% OOIP
			Mg ²⁺	0.210 to 0.421 mol/l	+10% OOIP
(Yasuda et al., 2022)	Modified SW	63°C	NaCl	23.47 to 0 g/L	+19.4% OOIP
(Montazeri et al., 2024)	Modified SW	55°C	SO ₄ ²⁻	0.06 to 0.12 mol/l	+1-2% OOIP

2.3 Hybrid EOR Approaches

Hybrid EOR techniques involve the combination of two or more chemical methods to improve oil recovery through synergistic mechanisms, such as IFT reduction, wettability alteration, ion exchange, and EDL expansion. In many of these approaches, LSW plays a key role by shifting the rock surface toward more water-wet conditions, thereby enhancing the effectiveness of subsequently injected agents (Johannessen & Spildo, 2013; Shakeel et al., 2022).

For example, when combined with surfactants, wettability alteration enhances the efficiency of IFT reduction, allowing effective oil mobilization (Sekerbayeva et al., 2020). As a result, the combined process enhances both oil detachment and displacement, leading to improved recovery compared to standalone methods (Esfandiarian et al., 2020; Hirasaki et al., 1983; Shaddel & Tabatabae-Nejad, 2015).

One critical aspect of this strategy is its sensitivity to the composition of brine, especially when considering its effects on surfactant activity. Low-salinity conditions are often favorable for surfactants, especially when they reduce adverse interactions with ions such as Ca^{2+} and Mg^{2+} . This helps prevent precipitation and activity loss (Pal et al., 2018; Shaddel & Tabatabae-Nejad, 2015). At the same time, adjusting the ion composition, especially for SO_4^{2-} , Ca^{2+} , and Mg^{2+} , also helps improve wettability alteration without impairing surfactant stability, especially under conditions (Esfandiarian et al., 2020). This balance between salinity reduction and ion tuning is important for maximizing the effectiveness of this hybrid system.

Apart from surfactant-based methods, hybrid approaches can also include combinations with alkali, in which conditions of low salinity affect the chemical environment in which alkali-based reactions take place (Khlaifat et al., 2022). Unlike conventional alkali flooding, in which processes are often limited by fast consumption and precipitation reactions, the influence of LSW changes ionic competition at the interface between rock and fluid (Gong et al., 2016; Honarvar et al., 2020). Lower concentrations of non-active ions, such as Na^+ and Cl^- , improve accessibility to reactive species and can improve the effectiveness of interactions between alkali and crude oil (Shaddel et al., 2013). Therefore, in-situ surfactant generation can take place under improved conditions, possibly requiring lower concentrations of alkali to attain similar effects on interfaces.

Another important aspect of this hybrid approach is its influence on reaction pathways. Under modified ionic conditions, the interaction between alkali and crude oil components may become more selective, reducing losses due to side reactions with formation water or rock minerals (Al-Saedi et al., 2020; Honarvar et al., 2020; Shaddel et al., 2013). This can contribute to improved chemical efficiency and more stable system behavior compared to standalone alkali flooding.

Similarly, nanoparticle-based hybrid systems offer an innovative form of hybrid EOR, where reduced ionic strength enhances the interactions between nanoparticles, brine, and rock surface, which improves nanoparticle stability. The stability enables better dispersion of nanoparticles, thereby reducing aggregation and increasing penetration into the porous medium, facilitating better interaction with the rock surface (Devakumar et al., 2023; Jafarbeigi et al., 2022b; Mumbere et al., 2025; Shakeel et al., 2024). Moreover, this hybrid system can enhance disjoining pressures in thin liquid films, thus aiding oil detachment from the rock surface.

Despite these potential benefits, the performance of nanoparticle-based hybrid systems is highly sensitive to nanoparticle type, concentration, and brine composition. Improper design may lead to particle aggregation, pore blockage, or excessive retention within the porous medium, which can negatively impact injectivity and recovery efficiency (Devakumar et al., 2023; Shakeel et al., 2024; Sze Lim et al., 2023). Furthermore, the interaction between nanoparticles and reservoir minerals under varying salinity conditions remains insufficiently understood, particularly in imbibition processes (Mumbere et al., 2025).

However, hybrid EOR approaches are not limited to LSW-based systems and may involve various combinations of chemical agents depending on reservoir conditions.

A comparison of the dominant mechanisms of standalone and hybrid EOR approaches is summarized in Table 6.

Table 6. Comparison of mechanisms in standalone and hybrid EOR methods

Category	Method	Wettability Alteration	IFT Reduction
Standalone	Surfactants	Moderate	Strong
	Alkali	Moderate	Moderate-strong
	Nanoparticles	Strong	Weak
	LSW	Strong	Weak
	Ion-modified LSW	Very strong	Weak-moderate
Hybrid	LSW-Surfactant	Strong	Strong
	LSW-Alkali	Strong	Moderate-strong
	LSW-Nanoparticles	Very strong	Weak-moderate

2.4 Effect of Injection Sequencing in Hybrid EOR Processes

In hybrid chemical EOR processes, the selection of the chemicals, as well as the sequence of their use, is of critical importance in the evaluation of the efficiency of the EOR process. Injection sequencing has a direct effect on the evolution of the interactions, surface chemistry, and interfacial conditions, which directly affect the SI process in fractured carbonate reservoirs. Even though the EOR processes are well studied, the dynamic interplay among the EOR mechanisms, which are activated during the injection process, has not been clearly understood.

One of the important functions of sequencing is to provide beneficial initial conditions for subsequent chemical interactions. For instance, LSW treatment is used as preconditioning to alter the rock wettability and modify the surface charge using mechanisms such as MIE, EDL, and calcite dissolution. This treatment is used to reduce the adhesion forces between crude oil and the carbonate surface. Consequently, this enables the injected subsequent chemicals to be more effective. When surfactants are injected after LSW treatment, they are more effective in reducing the IFT and mobilizing the oil due to reduced adsorption loss to the rock surface and better contact with the oil-water interface (Johannessen & Spildo, 2013; Sekerbayeva et al., 2020; Shakeel et al., 2022). Figure 11 is an illustration of the oil recovery profile for LSW-surfactant injection after LSW preconditioning done by Shakeel et al. (2022). Here, the RF achieved an OOIP of 82.5%. Moreover, in LSW-nanoparticle systems, preconditioning with LSW was used to improve the stability of nanoparticle dispersion and promote more uniform adsorption on the rock surface. This allowed stronger wettability alteration and disjoining pressure effects (Jafarbeigi et al., 2022b).

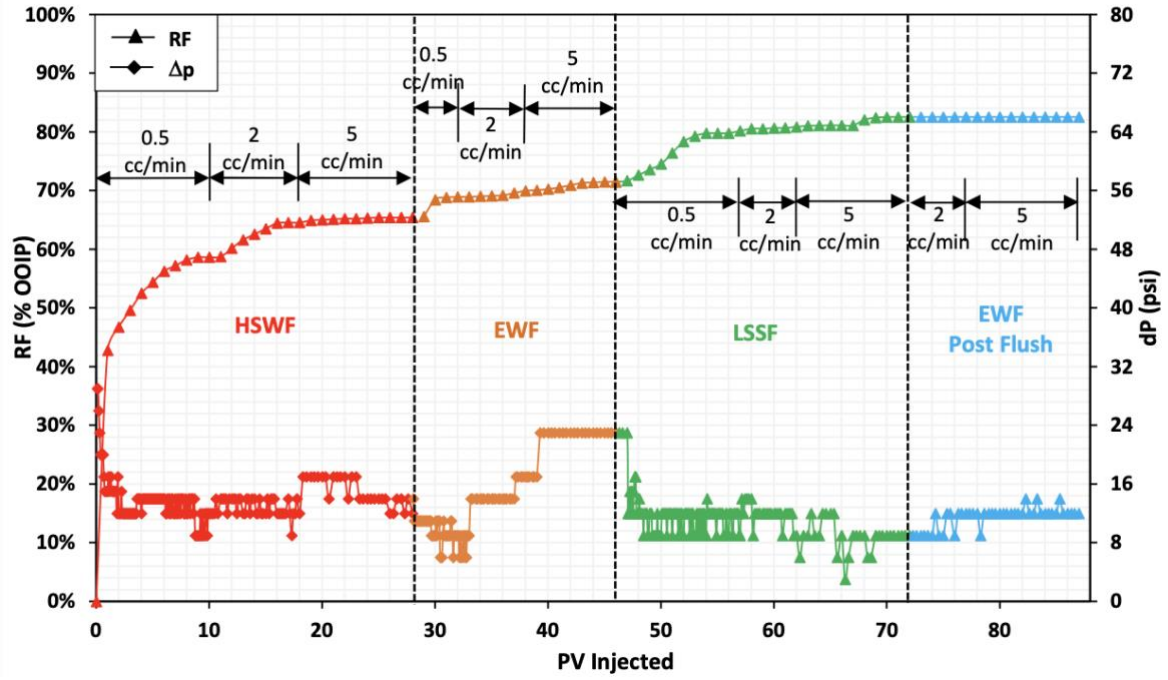


Figure 11. Oil recovery profile for LSW-surfactant injection after LSW preconditioning (Shakeel et al., 2022)

On the other hand, the direct injection of hybrid chemical systems, which are not preconditioned, may lead to suboptimal performance due to unfavorable initial reservoir conditions. In high salinity reservoirs, the performance of the hybrid systems can be compromised, as high salinity can inhibit the performance of the surfactant, enhance the aggregation of nanoparticles, and reduce the exchange of ions (Devakumar et al., 2023; Shakeel et al., 2024). In addition, the competitive adsorption of different chemicals can occur when injected in a simultaneous manner, which can reduce the availability of active agents at the rock-fluid interface. These effects highlight the importance of controlling not only the composition but also the stage-by-stage sequence of injection.

Sequencing is also important in controlling the balance between the capillary and gravity-driven mechanisms during the process of SI. Alteration of wettability in the early stages of the process, normally implemented using LSW or ion-modified brines, increases the strength of the capillary mechanism by moving the system towards a more water-wet system, thus promoting the process of imbibition (Alvarez & Schechter, 2017; Austad & Standnes, 2003; Ghandi et al., 2019). Subsequent reduction of the IFT, normally implemented using surfactant or alkali-based soaps, increases the oil recovery and reduces the residual oil saturation (Hincapie et al., 2022; Jia et al., 2019; Kamal, Hussein, et al., 2017; Shakeel et al., 2024). However, the reduction of the IFT at the early stages of the process may result in a weakening

of the capillary mechanism before the process of wettability alteration is complete, thus slowing down the process of imbibition (Hou et al., 2015; Santanna et al., 2014). Therefore, an optimal sequence often involves first wettability modification and then controlled IFT reduction to maximize both imbibition rate and ultimate recovery.

Several experimental studies have demonstrated the significance of injection sequencing in hybrid EOR systems. Coreflood and imbibition experiments have shown that LSW pretreatment followed by chemical injection can yield higher recovery compared to simultaneous injection or reversed sequences (Alagic et al., 2011; Sekerbayeva et al., 2020; Shakeel et al., 2022; Tavassoli et al., 2016). These findings suggest that the effectiveness of hybrid EOR approaches is not solely dependent on the individual mechanisms involved, but also on how and when these mechanisms are activated during the injection process.

2.5 Research Gap

Although there has been extensive research conducted to explore the application of chemical EOR in carbonate reservoirs, there are some gaps in understanding and optimizing the SI process under natural reservoir conditions. While individual EOR techniques, like LSW, surfactant, alkali, and nanoparticles, have been explored in detail, most of these studies have been conducted to assess individual EOR mechanisms, like wettability alteration, IFT reduction and ion-exchange, rather than understanding their combined and synergistic effects during the imbibition-based EOR process.

Hybrid EOR techniques have attracted attention in recent times, and most of these studies have been conducted under coreflooding conditions. Although coreflood experiments provide valuable insight into displacement efficiency under forced flow, they do not adequately represent SI, which is the dominant recovery mechanism in fractured carbonate reservoirs.

Furthermore, while various hybrid combinations of chemical agents have shown significant performance potential, there is a lack of systematic experimental testing of different hybrid EOR techniques under identical conditions. Specifically, there is no sufficient evaluation of the combined effects of wettability alteration, IFT reduction, and their effects on SI kinetics in carbonate formations.

A lack of understanding of injection sequencing effects in hybrid EOR techniques is another significant limitation in the available knowledge in this area. Specifically, while

preconditioning is assumed to enhance chemical injection performance, little is known about optimal sequencing effects and direct effects on SI kinetics.

Finally, while most available research has focused on RF as a primary performance metric in EOR techniques, there is limited use of mechanistic metrics such as contact angles and IFT in these techniques. As a result, this limits the ability to establish direct relationships between oil RF improvements and physicochemical mechanisms associated with SI kinetics.

Therefore, this study provides a systematic experimental evaluation of both standalone and hybrid EOR strategies under SI conditions, with particular emphasis on injection sequencing effects and mechanistic interpretation. By directly linking oil recovery performance with wettability alteration and IFT measurements, this work aims to establish a mechanistic framework for optimizing hybrid EOR design in fractured carbonate reservoirs.

3. METHODOLOGY

To attain the objectives of the current study, a systematic approach has been proposed to evaluate the performance of the SI in oil-wet fractured carbonates using standalone and hybrid EOR processes. To begin, the research has been initiated by carrying out fluid, chemical, and rock characterization, followed by screening tests and SI experiments under controlled conditions.

To evaluate the performance of the standalone EOR processes, including surfactant, alkali, nanoparticles, and LSW, a set of experiments has been conducted. In addition, a set of experiments has been designed to evaluate the performance of the hybrid EOR process, which combines the effects of the LSW and chemical agents. Moreover, additional experiments have been designed to evaluate the effect of the sequencing, in which the hybrid formulations have been introduced after the implementation of chemical flooding or LSW pretreatment.

In addition, the research was conducted to assess the effect of the brine composition, in which the modification ions, including Ca^{2+} , Mg^{2+} , and SO_4^{2-} , has been incorporated in the experimental design to evaluate the effect of the combined ions on the imbibition process. Furthermore, the CA and IFT experiments were done to evaluate the wettability alteration and the IFT reduction.

The detailed materials, experimental procedures, and analysis are explained in the following sections.

3.1 Materials

3.1.1 Crude Oil Properties

In this study, a heavy crude oil sample from the Uzen field in Kazakhstan was diluted with decane at a 1:1 volume ratio. The crude oil is characterized by an acid number of 0.08 mg KOH/g and a base number of 0.71 mg KOH/g. The viscosity and density of the resulting diluted oil were determined using an Anton Paar viscometer, and the measured properties are summarized in Table 7. Prior to usage, the filtration of crude oil was conducted to remove minerals and rock particles.

Table 7. Properties of diluted oil

Temperature	25°C	63°C
Density (g/cm ³)	0.789	0.758
Viscosity (cP)	4.402	1.846

3.1.2 Brine Composition

Porous media were saturated with formation water (FW) obtained from the Uzen field, the composition of which is provided in Table 8. To evaluate the impact of conventional water flooding and low-salinity water flooding as potential EOR techniques, Caspian seawater (SW) and its two-fold diluted form (LSW) were prepared and filtered prior to use. The effectiveness of the designed LSW to alter wettability of this specific field was already evaluated and confirmed by Kashiri et al. (2024). The ionic composition of FW, SW and LSW, obtained through ion chromatography, is also reported in Table 8.

Table 8. Chemical composition of brines

Brine	Ion Concentration (ppm)								TDS (ppm)
	Na ⁺	Ca ²⁺	Mg ²⁺	Cl ⁻	SO ₄ ²⁻	K ⁺	HCO ₃ ⁻	CO ₃ ²⁻	
FW	18902.0	3307.0	972.8	36159.0	75.8	390.4	276.0	390.4	60083.0
SW	3513.1	400.8	790.4	6026	3138	87.6	256.2	36	14248.1
LSW	1756.5	200.4	395.2	3013	1569	43.8	128.1	18	7124

To further enhance the performance of the selected hybrid EOR configuration, additional ion-modified LSW formulations were designed. These modified brines were developed by selectively adjusting the concentration of PDIs, including SO₄²⁻, Ca²⁺, and Mg²⁺, while maintaining the TDS close to the base LSW composition. Several ion-modified brines were prepared to systematically investigate the effect of ion concentration, including sulfate-enriched and divalent-cation-enriched systems. The calculated ionic compositions of these modified LSW formulations are presented in Table 9.

Table 9. Chemical composition of ion-modified LSW

Brine	Ion Concentration (ppm)								TDS (ppm)
	Na ⁺	Ca ²⁺	Mg ²⁺	Cl ⁻	SO ₄ ²⁻	K ⁺	HCO ₃ ⁻	CO ₃ ²⁻	
LSWCaMg-3SO4	1560.8	200.4	395.2	394.2	4707	43.8	128.1	18	7447.5
LSW3Ca3Mg-SO4	55.0	601.2	1185.6	3401.6	1569	43.8	128.1	18	7002.3
LSW3Ca3Mg-3SO4	99.1	601.2	1185.6	1198.6	4707	0	0	0	7791.5

The salts given in Table 10 were used to prepare these aqueous solutions.

Table 10. List of salts used to prepare brines

Required chemicals	Chemical formula	Purity	Producer
Sodium bicarbonate	NaHCO ₃	≥99.0%	Sigma-Aldrich
Sodium sulfate	Na ₂ SO ₄	≥99.0%	Sigma-Aldrich
Sodium chloride	NaCl	≥99.0%	Sigma-Aldrich
Calcium chloride	CaCl ₂	≥99.0%	Sigma-Aldrich
Magnesium chloride hexahydrate	MgCl ₂ .6H ₂ O	≥99.0%	Sigma-Aldrich
Potassium chloride	KCl	≥99.0%	Sigma-Aldrich
Magnesium sulfate	MgSO ₄	≥99.0%	Sigma-Aldrich

3.2 Chemicals Used in EOR Experiments

This study considered surfactant, alkali, and nanoparticles (NPs) as candidate chemicals for EOR methods. The screening phase involved testing multiple possible formulations using a range of static evaluation methods and selection criteria, followed by detailed analysis. A concise overview of each chemical and the corresponding screening procedure is provided below.

3.2.1 Surfactant

An anionic internal olefin sulfonate, Soloterra O332 (C15-C18 IOS), supplied by Sasol, was used. Additionally, the selected co-surfactant is an anionic alcohol propoxy sulfate supplied by the Alfoterra series, AlfoTerra S23-7S-90. Previous studies by Dauyltayeva et al. (2023) investigated the adsorption behavior of the selected surfactant. It was established that the selected surfactant exhibited standalone dynamic adsorption of 9.2 mg/g rock. It is important to note that the selection of the surfactant had already been carried out. Detailed results were established by Kashiri et al. (2024). From the results, it is evident that the optimal composition is 1 wt.% total surfactant with a 10:90 surfactant-to-co-surfactant ratio. This composition exhibited the minimum IFT. For the preparation of the solutions, the required amounts of the selected surfactant and the selected co-surfactant were accurately measured. Subsequently, the measured amounts were gradually introduced into the synthetic brine while stirring at 200 rpm to prevent the formation of foam. Stirring is done for 1 to 2 hours to ensure the complete dissolution of the solutions. Subsequently, the solutions were sealed with parafilm and stored in a dry environment.

3.2.2 Alkali

Sodium carbonate (Na_2CO_3) was selected as the alkali candidate for the SI experiments, following the screening methodology outlined in earlier work by Kashiri et al. (2023). To prepare the alkaline solutions, the required amount of Na_2CO_3 (0.008-0.015 g/L) was accurately weighed and gradually dissolved in the synthetic brines under continuous stirring at 600 rpm. Mixing was maintained for 20-30 minutes to ensure complete dissolution.

3.2.3 Nanoparticles

The spherical porous silica NPs were 99.5% pure, had a density of 2.4 g/cm^3 , and had a particle size of 10-20 nm. The NPs were sourced from SkySpring Nanomaterials. To find the optimum concentration of NPs for the SI experiments, aqueous stability and wettability alteration tests were conducted. After finding the optimum concentration, nanofluid solutions were made by adding the nanoparticles to brine and stirring them with a magnetic stirrer for one hour. The mixture was then subjected to ultrasonic homogenization for another hour. This method employs high-frequency ultrasound waves to create cavitation and acoustic streaming, which increases the dispersion, decreases the particle size, and increases the stability of the nanofluid.

3.3 Rock Sample Preparation

The limestone outcrop samples were used as test materials in the wettability alteration experiments for NPs screening stage, contact angle measurements and SI tests. The results of the X-ray diffraction (XRD) test showed that the mineral composition of the rock was primarily composed of calcite, which formed about 96.97% of the total composition, while only quartz was detected in small quantities.

For the wettability alteration experiments, the limestone outcrop samples were crushed to powder form using a Retsch Disc Mill DM 200. The crushed sample was then sieved to have sizes of 40 and 125 μm . The sample that passed through the 40 μm sieve but was retained by the 125 μm sieve, which had an average size of 82.5 μm , was used for the floatation tests.

For the contact angle test, the core plug samples were cut to 0.5-1.0 cm thick sections and then polished to have minimal roughness. The samples were then dried in an oven for at least 3 days. After drying, the samples were then subjected to formation brine for 24 hours to restore the

initial reservoir conditions. The samples were then subjected to crude oil at 80 °C for 30 days to restore the initial wettability.

For the SI experiments, core plugs were cut to the required length and oven-dried at 80 °C for at least three days. Their dry weights and dimensions were recorded before porosity measurements were performed using a Helium Porosimeter. To eliminate residual or trapped gases that could affect pore accessibility or capillary behavior, the cores were vacuum-deaerated prior to saturation. Formation brine was then introduced under a pressure of 1100 psi for 24 hours using a Manual Saturator to ensure complete saturation. Absolute brine permeability was subsequently determined by Aging Cell flooding apparatus which allows measuring the pressure drop across the core at different injection flow rates, with a confining pressure of 1000 psi and a back pressure of 500 psi. Darcy's law, shown In Equation 3.1, was applied to calculate permeability by incorporating pressure drop, flow rate, fluid viscosity and core dimensions. Following this step, oil flooding was carried out at varying flow rates until irreducible water saturation (S_{wi}) was achieved. The oil-saturated cores were then aged in crude oil at 80 °C for 60 days to restore reservoir-like wettability conditions. Despite the uniform mineralogical composition (96.97% calcite), the K_{abs} values varied considerably across cores, which is attributed to natural heterogeneity in grain size and pore structure inherent to the limestone outcrop samples. A summary of the core properties and measured parameters is provided in Table 11.

$$k = \frac{q\mu L}{A\Delta p} \quad (3.1)$$

Table 11. Properties of core samples for SI tests

Core for Test #	D, mm	L, mm	Porosity, %	PV, cc	K_{abs} , mD	S_{wc} , %
1	38.10	59.90	14.53	9.920	42.4	27.79
2	38.10	61.70	14.25	10.030	51.4	37.12
3	37.20	59.20	12.64	8.129	20.2	41.57
4	37.20	59.10	12.79	8.211	25.4	45.20
5	38.10	61.70	14.25	10.030	51.4	37.12
6	38.00	61.40	19.00	13.190	9.9	30.86
7	37.90	61.20	17.99	12.460	10.7	42.98
8	38.05	59.49	20.27	13.710	36.0	42.38
9	38.05	60.37	21.10	14.490	92.1	34.30
10	38.05	60.85	20.62	14.270	58.8	44.95
11	38.07	60.43	19.12	13.150	20.3	18.63
12	38.04	59.98	16.54	11.280	28.4	37.5

3.4 Nanoparticles Screening Experiments

3.4.1 Aqueous Stability Test

For the aqueous stability assessment, silica nanoparticle solutions with concentrations between 0.05 and 0.2 wt.% were prepared in SW and LSW. These solutions were stored in an oven at 63 °C for one week. Solutions that remained clear and free of precipitation were considered stable. The stable dispersions were then used in wettability alteration tests.

3.4.2 Wettability Alteration Test

The alteration in the wettability caused by nanoparticles was assessed using the floatation method with crushed limestone samples. During this process, 2 g of crushed limestone powder is first mixed with 10 mL of crude oil in a test tube. The mixture is left to stand at 63 °C for 48 hours to ensure oil-wet conditions. After the oil-wet conditions have been established, 20 mL of the nanoparticles dispersed in either SW or LSW is introduced into the mixture. Subsequently, the test tube is shaken well to ensure the right interaction between the phases. It is then left to stand in the oven at 63 °C for 24 hours to ensure the right phase separation. At this point, the oil phase together with the particles suspended is removed. Particles with water-wet properties, which settle at the bottom of the test tube, are collected. They are subsequently washed with toluene to remove any oil left. Finally, the weight of the water-wet particles is recorded to determine the level of alteration in the wettability. The degree of alteration in the wettability is evaluated using Equation 3.2.

$$\text{Wettability Alteration (\%)} = \frac{W_{\text{water-wet}}}{W_{\text{total}}} * 100 \quad (3.2)$$

3.5 Contact Angle and IFT Measurements

Once the aging process is over, the oil-saturated core slices were wiped lightly to remove the oil in excess. These oil-wet slices were then subjected to standalone or hybrid EOR formulations at 80 °C. Contact angle tests were carried out to observe the changes in wettability by using an IFT700 device from Vinci Technologies, as shown in Figure 12. The contact angle measurements were recorded at 0, 1, 3, 6, 9, and 12 days to observe the changes in wettability. The angle between the oil droplet and the rock surface was used as a quantitative measure of the wettability of the carbonate rock. The angle decreases toward more water-wet conditions.

The experiments were repeated at least twice for reproducibility of the results. The results were recorded in the form of average values.



Figure 12. IFT700 interfacial tension and contact angle measurement system (Vinci Technologies, n.d.)

IFT measurements were conducted using the same IFT700 system (Vinci Technologies) to evaluate the effectiveness of different brine and chemical formulations in reducing oil-water interfacial forces. The measurements were performed using the pendant drop method, where an oil droplet was formed in the aqueous phase and its shape was analyzed to determine IFT values. All measurements were repeated to ensure consistency, and average values were reported.

The Table 12 below summarizes the standalone and hybrid EOR methods for which CA and IFT measurements were performed, based on their primary mechanisms identified from the literature.

Table 12. Selection of standalone and hybrid EOR methods for CA and IFT measurements based on dominant recovery mechanisms

Category	Method	CA	IFT
Standalone	Surfactants		+
	Alkali		+
	Nanoparticles	+	+
	LSW	+	+
	Ion-modified LSW	+	+
Hybrid	LSW-Surfactant	+	+
	LSW-Alkali	+	+
	LSW-Nanoparticles	+	+

3.6 Spontaneous Imbibition Experiments

SI experiments were performed at 63°C using Amott cells supplied by Pan Terra Geoconsultants. After the aging process, the cores, with all faces exposed, were placed

vertically inside the cells, which were then filled with SW through the top inlet. This phase represents imbibition by a fluid that does not noticeably affect the capillary force. Once oil production reached a plateau, the imbibing fluid was sequentially switched to the EOR agents under study, including LSW, surfactant solution, alkali solution, NPs solution and hybrid formulations. Oil recovery was tracked over time by recording the graduated scale readings on the Amott cell, and the RF was expressed as the percentage of the original oil in place (%OOIP). Figure 13 shows selected SI tests performed in Amott cells as representative examples.



Figure 13. Representative SI experiments conducted in Amott cells

Each test number in Table 13 corresponds to the core sample of the same number listed in Table 11. Table 13 represents the list of SI experiments that have been completed to assess the recovery performance of various brine and chemical combinations, as well as different sequences and LSW-preconditioning. In this design, Phase 2 in Tests 1-4 evaluated standalone chemical floods, while Phase 2 in Tests 5-7 assessed hybrids. Phase 3 in Tests 1-3 represents hybrids injected after a standalone chemical stage, whereas Phase 3 in Tests 8-10 represents hybrids injected after LSW pretreatment.

Following the evaluation of standalone, hybrid, and sequencing strategies, an optimal injection pathway was identified based on preliminary analysis of SI experiments. To further investigate and enhance this configuration, additional experiments were designed. Specifically, two

additional SI tests (Tests 11-12) were conducted using ion-modified LSW formulations to evaluate the effect of ionic composition on wettability alteration and imbibition performance. In these tests, the injection sequence was modified by introducing ion-modified LSW after the conventional LSW stage. This design enabled a systematic assessment of whether ion optimization can further improve the effectiveness of LSW preconditioning and its subsequent hybrid application.

Table 13. List of SI experiments

Test Number	Phase 1	Phase 2	Phase 3
1	SW	S	LSW+S
2	SW	NP	LSW+NP
3	SW	A	LSW+A
4	SW	LSW	
5	SW	LSW+S	
6	SW	LSW+NP	
7	SW	LSW+A	
8	SW	LSW	LSW+S
9	SW	LSW	LSW+A
10	SW	LSW	LSW+NP
11	SW	LSW	LSW-M
12	SW	LSW	LSW-M+S

Note:

SW = Seawater; S = Surfactant in SW; NP = Nanoparticles in SW; A = Alkali in SW; LSW = Low salinity water; LSW-M = Ion-modified low salinity water; LSW+S = Surfactant in LSW; LSW+NP = Nanoparticles in LSW; LSW+A = Alkali in LSW; LSW-M+S = Surfactant in LSW-M.

3.7 Experimental Design

The experimental design, as presented in Figure 14, has been developed to systematically investigate EOR approaches under SI conditions in fractured carbonate reservoirs.

First, EOR fluids, such as surfactants (S), alkali (A), nanoparticles (NP), and LSW, were initially screened to determine the scope of EOR fluids investigated in this study. To understand the underlying recovery mechanisms, CA and IFT measurements were carried out for selected standalone and hybrid formulations. This helps to understand oil recovery mechanisms, such as wettability alteration and IFT reduction, which are the main recovery mechanisms for oil displacement in fractured carbonate reservoirs.

Next, SI experiments were carried out to assess oil recovery performance. This was also designed to compare oil recovery performance for standalone, hybrid methods (LSW+S,

LSW+A, and LSW+NP), and different sequences, such as hybrid chemical injection after standalone methods and after LSW preconditioning.

To further improve oil recovery performance, additional experiments were carried out, incorporating ion-modified LSW, whereby the concentrations of key PDIs, such as Ca^{2+} , Mg^{2+} , and SO_4^{2-}), were optimized to improve wettability alteration effects. The optimized sequences, such as $\text{SW} \rightarrow \text{LSW} \rightarrow \text{LSW-M}$ and $\text{SW} \rightarrow \text{LSW} \rightarrow \text{LSW-M+S}$, were carried out using SI experiments.

Lastly, oil recovery and rates by SI tests, along with CA and IFT measurements, were analyzed to understand relationships between fluid-rock interactions and observed production performance.

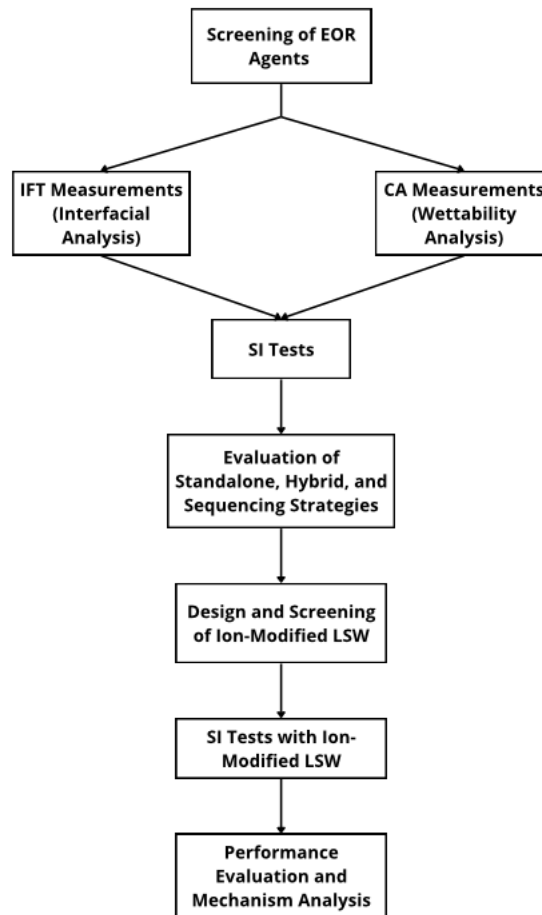


Figure 14. Experimental workflow for evaluation and optimization of EOR strategies

4. RESULTS

4.1 Nanoparticle Screening and Selection

4.1.1 *Aqueous Stability Test*

From Figures 15 and 16, the aqueous stability tests of nanoparticles in SW showed that solutions with concentrations of 0.05 wt.% and 0.1 wt.% remained stable, while higher concentrations (0.15 wt.% and 0.2 wt.%) exhibited precipitation after one week. A similar trend was observed in LSW, where only the 0.05 wt.% and 0.1 wt.% concentrations remained stable over the same period. Based on these observations, nanoparticle concentrations of 0.05 wt.% and 0.1 wt.% in both SW and LSW were selected for further experiments.



Figure 15. Aqueous stability test for NPs in SW



Figure 16. Aqueous stability test for NPs in LSW

4.1.2 *Wettability Alteration Test*

The flotation test technique was used to evaluate the wettability alteration of crushed limestone samples. From Table 14, in seawater, increasing nanoparticle concentration from 0.05 wt.% to

0.1 wt.% improved wettability alteration, with the 0.1 wt.% concentration achieving the highest alteration of 89.8%.

In contrast, in LSW, the 0.05 wt.% concentration resulted in higher wettability alteration (88.7%) compared to 0.1 wt.% (85.1%). These results indicate that nanoparticle performance in wettability alteration depends on both concentration and brine salinity.

Table 14. Wettability alteration test results for NPs screening

Brine	NP Concentration	Initial Powder Weight (gr)	Water-Wet Particles (gr)	Remaining Oil-Wet Particles (gr)	Water-Wet Weight (gr) / Initial powder weight (gr)	Wettability Alteration (%)
SW	0.05 wt.%	2	1.687	0.313	0.844	84.4
	0.1 wt.%	2	1.795	0.205	0.898	89.8
LSW	0.05 wt.%	2	1.774	0.226	0.887	88.7
	0.1 wt.%	2	1.722	0.278	0.861	86.1

Based on these results, 0.1 wt.% nanoparticle concentration was selected for SW, whereas 0.05 wt.% was determined to be optimal for LSW and these concentrations were used in all subsequent experiments.

4.2 Mechanistic Measurements (IFT and Contact Angle)

Based on the experimental design and selection criteria defined in the methodology (Table 12), selected standalone and hybrid EOR systems were evaluated through IFT and CA measurements.

4.2.1 IFT measurements

The results of the IFT measurements for the selected systems are shown in Table 15. SW, as the baseline, had the highest IFT value of 45.5 mN/m, indicating high interfacial force and hence less potential for mobilization of the oil by the capillary force. The IFT was reduced in the case of LSW, which had a value of 33.2 mN/m.

Table 15. IFT values for selected standalone and hybrid EOR systems

Methods	IFT values (mN/m)
SW	45.5
LSW	33.2
S	0.2
LSW + S	0.3

A	0.4
LSW + A	0.4
NP	25.2
LSW + NP	21.5

The addition of the surfactant system caused a significant reduction in the IFT. The lowest value of IFT was obtained by S, which was 0.2 mN/m. The hybrid system, namely, LSW+S, also produced an extremely low value of 0.3 mN/m, indicating the significant influence of the surfactant system on the IFT, irrespective of the type of brine solution.

In contrast, the alkali-based systems also produced significant reduction in the IFT, and the values obtained were 0.4 mN/m for both A and LSW+A systems. Although the values are slightly high compared to the surfactant-based system, they are still within the low IFT region, indicating effective reduction in IFT. The nanoparticle-based system, however, showed limited influence on IFT, and the values remained high, namely 25.2 mN/m and 21.5 mN/m, for NP and LSW+NP, respectively.

From the above results, it is evident that the system containing the surfactant produced the most pronounced reduction in the IFT, and the ultra-low value was obtained. The alkali-based system also resulted in significant IFT reduction, although slightly less compared to the surfactants. The nanoparticle-based system failed to show strong influence on the IFT, and the LSW system produced only moderate reduction in the IFT compared to the SW system.

4.2.2 Contact Angle Measurements

To further investigate the alteration in wettability, CA measurements were performed on the EOR systems, and the results are presented in Table 16. The initial CA for all EOR systems was 178.5°, indicating strongly oil-wet conditions. The CA images for the evaluated EOR systems are shown in Appendix A.

Table 16. Contact angle values for selected standalone and hybrid EOR systems

Methods	Days					
	0	1	3	6	9	12
LSW	178.5°	126.2°	120.8°	118.0°	118.0°	118.0°
NP	178.5°	173.4°	168.5°	163.8°	161.9°	154.3°
LSW + S	178.5°	175.1°	173.8°	173.7°	173.4°	173.4°
LSW + A	178.5°	172.3°	170.2°	168.0°	167.1°	167.1°
LSW + NP	178.5°	169.3°	166.7°	161.6°	157.3°	150.0°

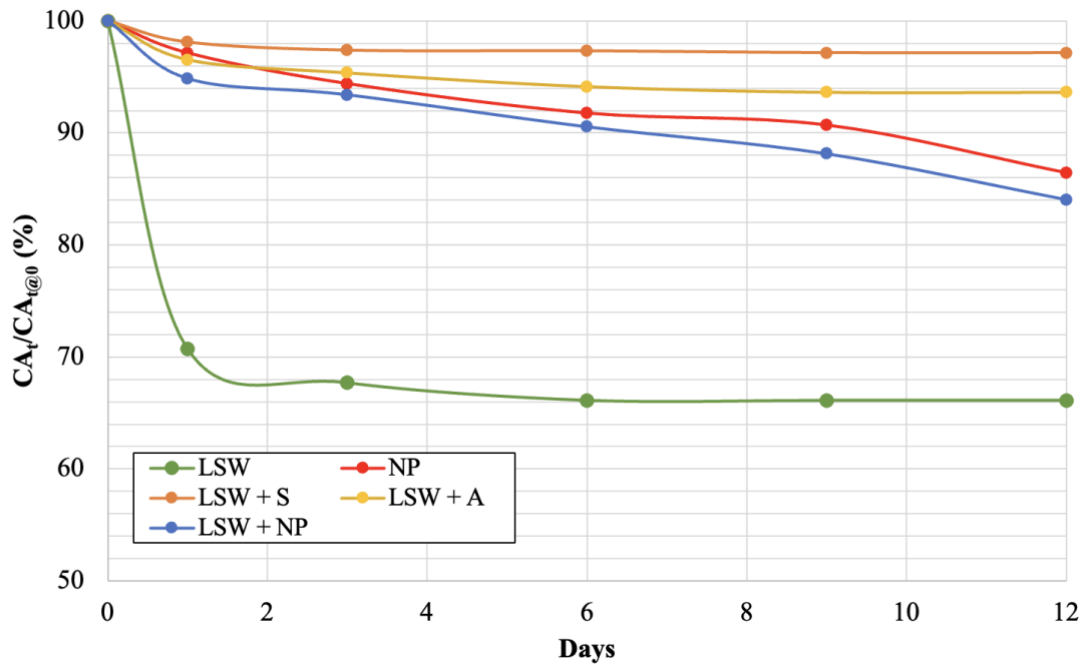


Figure 17. Normalized CA ratio over time for selected standalone and hybrid EOR systems

According to the results presented in Table 16 and further illustrated in Figure 17, the CA was significantly reduced from 178.5° to 118° over time for the LSW system, indicating the wettability alteration conditions from strongly oil-wet to weakly oil-wet conditions. The CA measured for the NP-based EOR system, indicated slower wettability alteration, as the contact angles reduced to 154.3° over 12 days.

The CA measured for the LSW+NP system indicated slightly improved wettability alteration, as the CA reached 150° . The CA measured using the LSW+A and LSW+S EOR systems indicated moderate and minimum wettability alteration, as the contact angles reduced to 167.1° and remained high, reaching 173.4° , respectively. The results indicate that wettability alteration is most significant for the LSW system, while surfactant-based systems show minimal CA reduction despite their high IFT reduction potential. This distinction highlights the different dominant mechanisms governing each EOR method. As shown in Figure 17, LSW demonstrates the most rapid and substantial wettability alteration, stabilizing at approximately 66% of its initial value by day 12, while LSW+S shows the least alteration, remaining above 97% throughout the experiment.

Based on the combined IFT and contact angle results, distinct imbibition behaviors are expected for the evaluated EOR systems. Surfactant-based systems, characterized by ultra-low IFT, are anticipated to exhibit rapid early-time imbibition due to reduced capillary resistance

and enhanced oil mobilization. In contrast, LSW and nanoparticle-based systems, which show more pronounced wettability alteration, are expected to promote slower but sustained imbibition over time as the rock surface gradually shifts toward water-wet conditions. Alkali-based systems, demonstrating both low IFT and moderate wettability alteration, are expected to provide intermediate performance. Overall, these mechanistic observations suggest that different EOR methods will exhibit distinct imbibition dynamics, which are evaluated in the subsequent sections.

4.3 Imbibition Performance of Standalone EOR Methods

As the first approach, we compared the effect of different standalone EOR approaches on SI and oil recovery. Table 17 represents four tests designed for this comparison.

Table 17. Interpretation goals for standalone methods

	Test Number	Comparison		Interpretation Goal
		Phase 1	Phase 2	
<i>Standalone methods</i>	1	SW	S	IFT reduction
	2	SW	A	IFT reduction
	3	SW	NP	Wettability shift and interface alteration
	4	SW	LSW	Wettability shift due to low ionic strength

To address the primary objective of this study, the standalone EOR designs were first evaluated in terms of their influence on the SI process and the corresponding imbibition rates. The results, showing oil recovery over time and imbibition rates for the different standalone EOR methods, are presented in Figure 18 and Table 18.

For all standalone EOR techniques, the initial SI stage was conducted with SW to provide a common baseline for comparison. Figure 17 presents the RF values normalized to the %OOIP after SW imbibition. Imbibition with SW was weak and nearly identical across all cores, with imbibition rates ranging from 0.0156 to 0.0567 %/h and an average of 0.0304 %/h. The average value of RF after SW imbibition equals 9.17 %OOIP. The weak response is attributed to the predominantly oil-wet to mixed-wet nature of the carbonate rock, where capillary forces act against water invasion and limit SI. This average value is therefore adopted as the baseline reference for evaluating the subsequent EOR stages.

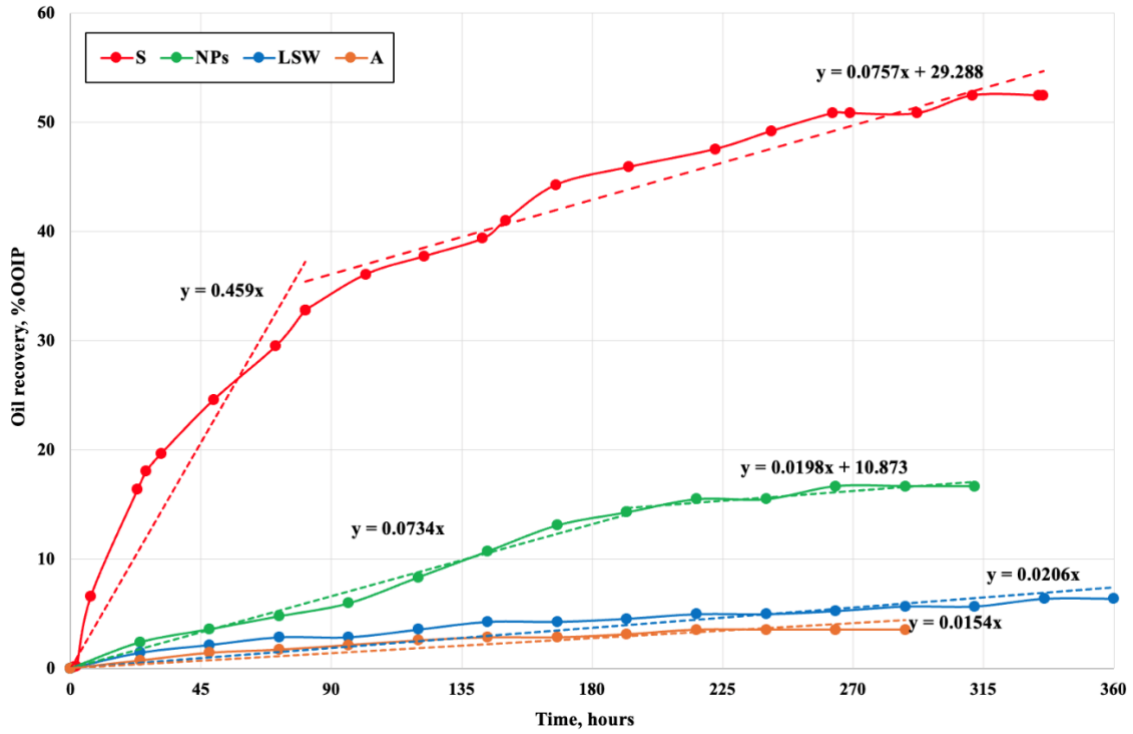


Figure 18. Comparison of standalone EOR methods

Table 18. Comparison of imbibition rates for different standalone EOR methods

Test Number	Phase	Early Time Imbibition Rate (%/h)	Late Time Imbibition Rate (%/h)	RF (%OOIP)
1	S	0.4590	0.0757	52.46
2	A	0.0154		3.54
3	NP	0.0734	0.0198	16.67
4	LSW	0.0206		6.38

As shown in Figure 18, the addition of surfactant to SW markedly enhanced SI compared to the weak baseline provided by SW alone (average 0.0304 %/h). The early-time imbibition rate increased sharply to 0.4590 %/h. Importantly, imbibition remained enhanced for more than two weeks, well beyond the short-lived activity of SW. During the late-time period, imbibition with S slowed to 0.0757 %/h, yet still exceeded SW performance. In terms of overall performance, surfactant flooding phase resulted in RF of 52.46 %OOIP, a dramatic improvement compared to the minimal recovery achieved by SW alone (9.17 %OOIP).

Switching from SW to alkali flooding resulted in only a marginal improvement in SI performance. The imbibition rate remained essentially unchanged at 0.0154 %/h, and the oil recovery achieved was less than 4 %OOIP.

The addition of nanoparticles brought a moderate improvement, as the early time imbibition rate improved to 0.0734 %/h, while the RF improved to 16.67 %OOIP. The effect of

nanoparticles, however, remained much lower compared to the effect of the surfactants. Unlike the surfactants, which start working immediately, nanoparticles start working gradually as they are adsorbed on the surface of the rocks. As a result, the oil production via imbibition slowed down over time, reaching a plateau at a rate of 0.0198 %/h.

The change from SW to LSW gave a relatively low kinetic response among all the fluids, but the recovery was still achieved. After the switch, the rate of imbibition remained very low and almost constant, 0.0338 %/h, indicating that the rate of oil displacement was very slow. However, the process was effective, as shown by the fact that RF was 6.38 %OOIP. Thus, although the change from SW to LSW did not accelerate the rate of imbibition significantly, the recovery was still achieved.

Overall, the effectiveness of the standalone EOR methods was found to follow the ranking: Surfactant > Nanoparticles > LSW > Alkali, in terms of imbibition kinetics and oil recovery. On the basis of these findings, hybrid EOR systems are further examined to check the synergistic effects of the various mechanisms.

4.4 Imbibition Performance of Hybrid EOR Methods

After establishing the performance of the standalone methods, the next step was to evaluate the hybrid formulations. Table 19 represents three tests designed for this analysis.

Table 19. Interpretation goals for hybrid methods

	Test Number	Comparison		Interpretation Goal
		Phase 1	Phase 2	
<i>Hybrid methods</i>	5	SW	LSW+S	Any synergy from IFT + ionic interactions
	6	SW	LSW+A	IFT reduction + wettability shift
	7	SW	LSW+NP	Enhanced interface/wettability alteration

The results, showing oil recovery over time and imbibition rates for different hybrid EOR methods, are presented in Figure 19 and Table 20.

As shown in Figure 19, switching to LSW+S increased the early-time imbibition rate to 0.145 %/h and achieved an RF of 9.98 %OOIP. Compared to LSW alone, which produced only a slow and limited recovery (0.0206 %/h; 6.38 %OOIP), the hybrid clearly accelerated oil displacement by introducing surfactant. Imbibition was sustained for nearly two weeks.

However, both the rate and recovery were lower than those of the standalone surfactant (0.4590 %/h; 52.46 %OOIP).

Switching to LSW+A increased the early-time imbibition rate to 0.0817 %/h, but the RF was only 2.68 %OOIP. Compared to LSW alone (0.0206 %/h; 6.38 %OOIP), the hybrid not only failed to provide a meaningful improvement but actually resulted in lower recovery. Performance also remained weaker than that of alkali alone (0.0154 %/h; 3.54 %OOIP), confirming the absence of any synergy.

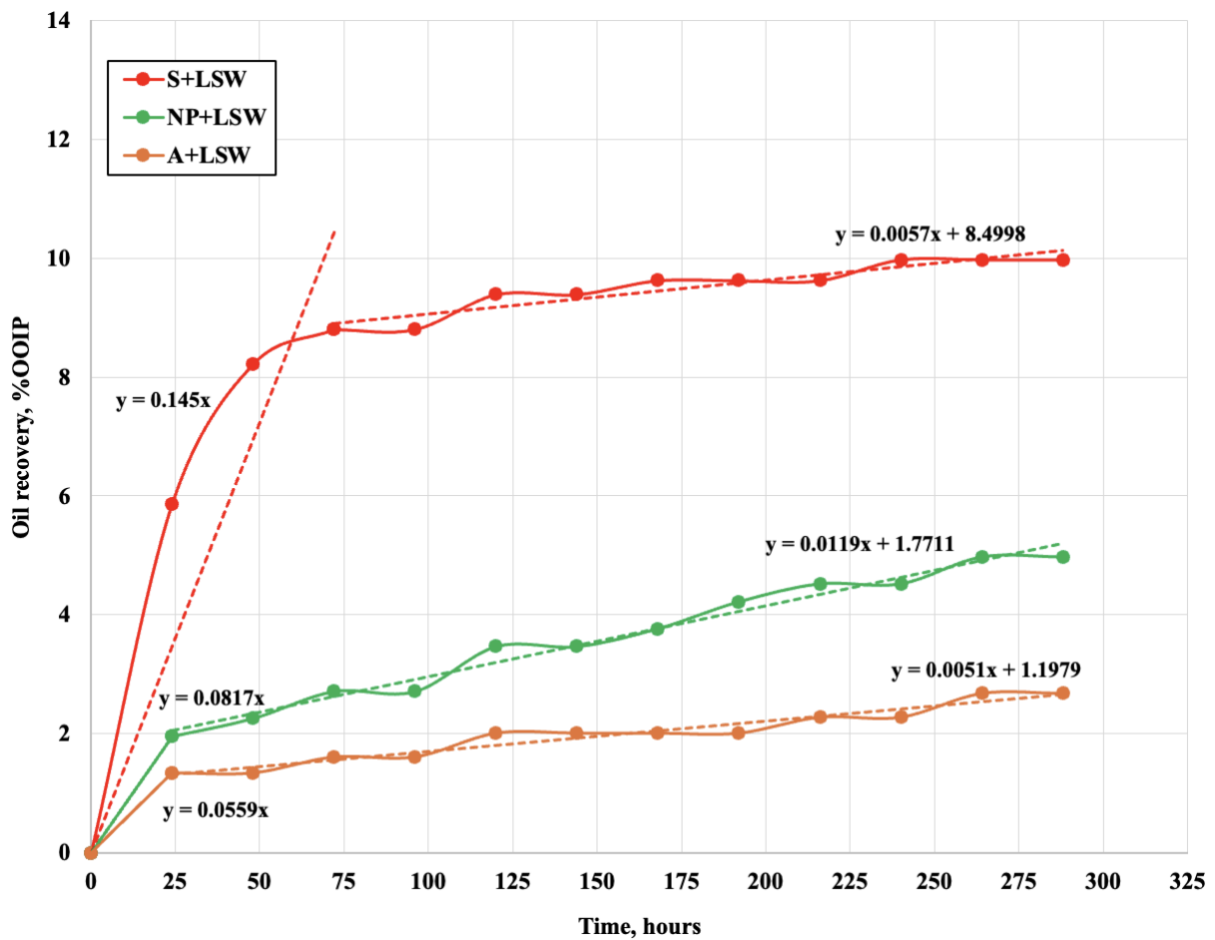


Figure 19. Comparison of hybrid EOR methods

Table 20. Comparison of imbibition rates for different hybrid EOR methods

Test Number	Phase	Early Time Imbibition Rate (%/h)	Late Time Imbibition Rate (%/h)	RF (%OOIP)
1	LSW+S	0.1450	0.0057	9.98
2	LSW+A	0.0817	0.0119	2.68
3	LSW+NP	0.0559	0.0051	4.98

When nanoparticles were combined with LSW, the early-time rate decreased to 0.0559 %/h and the late-time rate further declined to 0.0051%/h. The RF also dropped to 4.98 %OOIP. This indicates that instead of synergy, LSW reduced nanoparticle effectiveness. As a result, although NPs worked as a standalone, the hybrid design did not provide synergy under the tested conditions.

Overall, the results demonstrate that, while surfactant flooding in SW remains the best standalone option, hybridization makes weaker strategies, such as LSW, more competitive by accelerating the process of early-time imbibition and modestly increasing recovery. As a result, in fractured formations where SI dominates, hybrids should be preferred over weaker strategies, while ensuring chemical compatibility.

4.5 Sequencing Effects

The next stage of investigation focuses on the optimal sequencing of EOR methods. Specifically, the key question is whether hybrid solutions should be applied after standalone chemical flooding or after LSW pretreatment. The objective of the further study is therefore to identify the sequencing pathway that maximizes recovery and accelerates imbibition, providing clear guidelines for the practical design of hybrid EOR strategies. The SI tests analyzed and their corresponding interpretation objectives are summarized in Table 21. For all tests listed, phase 1 is SI with SW.

Table 21. Interpretation goals for optimal sequence analysis

Test Number	Comparison		Interpretation Goal
	Phase 2	Phase 3	
Hybrid after Standalone Methods			
1	S	LSW+S	Enhanced IFT + ionic interactions
2	A	LSW+A	Enhanced IFT change
3	NP	LSW+NP	Particle stability and enhanced wettability alteration
LSW preconditioning			
1	LSW	LSW+S	IFT reduction after wettability alteration
2	LSW	LSW+A	Enhanced IFT change
3	LSW	LSW+NP	Particle stability and interaction in low salinity

4.5.1 Hybrid Injection After Standalone Flooding

To evaluate whether hybrid formulations provide additional benefits once a chemical-enhanced flooding stage has already been applied, SI tests were performed in which surfactant, alkali, or nanoparticles in SW were followed by their corresponding hybrid with LSW. The results are summarized in Tables 22-23 and Figures 20-22. For all tests listed, phase 1 is SI with SW. RF values for Phases 2-3 are normalized to the %OOIP measured after the preceding phase has reached plateau.

Table 22. Comparison of imbibition rates for different standalone and subsequent hybrid methods

Test Number	Phase Number	Phase	Early Time Imbibition Rate (%/h)	Late Time Imbibition Rate (%/h)
1	2	S	0.4590	0.0757
	3	LSW+S		0.0019
2	2	A		0.0154
	3	LSW+A		0.0101
3	2	NP	0.0734	0.0198
	3	LSW+NP		0.0700

Table 23. Comparison of recovery factors for standalone and subsequent hybrid flooding

Test Number	Phase 2	Phase 3	RF of Phase 2 (%OOIP)	RF of Phase 3 (%OOIP)	Interpretation
1	S	LSW+S	52.46	0.69	Collapse after strong surfactant effect; LSW diluted surfactant activity
2	A	LSW+A	3.54	2.94	No synergy; alkali weakened in LSW
3	NP	LSW+NP	11.58	21.43	Sustained recovery; RF nearly doubled

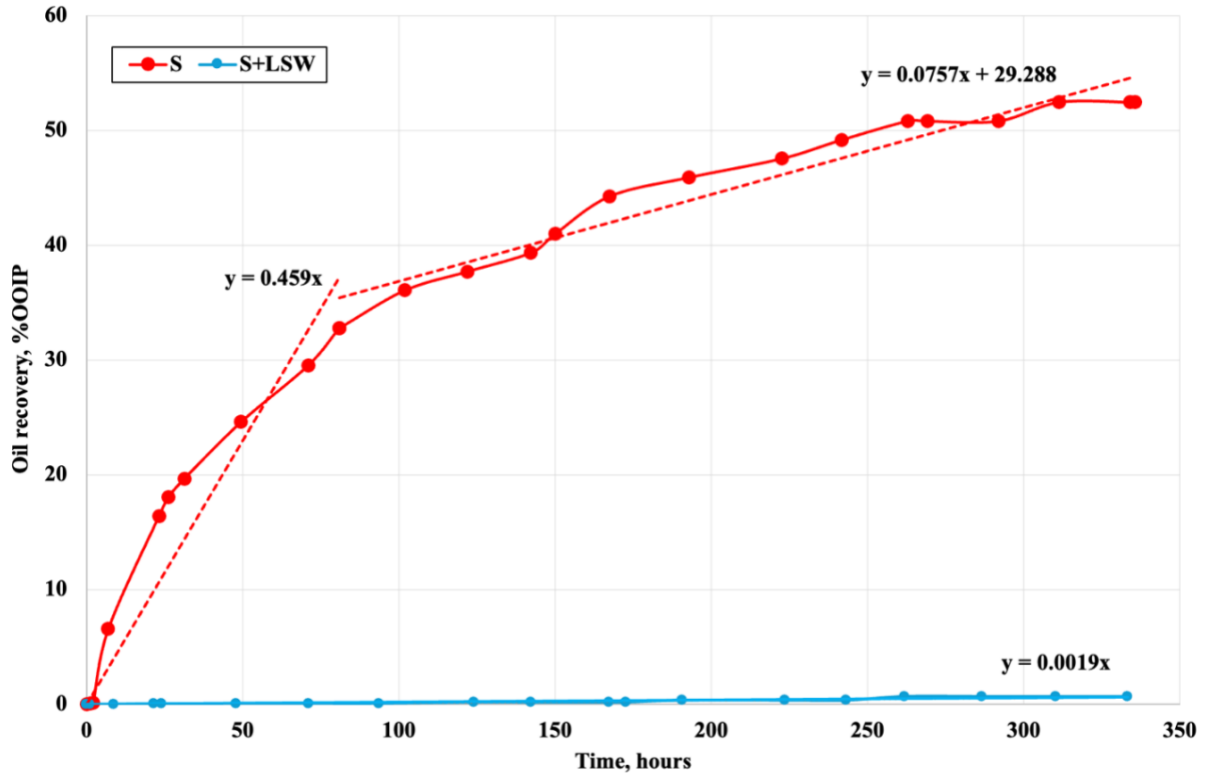


Figure 20. Comparison of standalone and hybrid surfactant flooding methods

As shown in Figure 20, the surfactant stage (S) exhibited the strongest response, with an early-time imbibition rate of 0.459 %/h and an RF of 52.46 %OOIP. However, upon transitioning to S+LSW, the imbibition rate collapsed to 0.0019 %/h, and recovery plateaued with only a minimal gain (+0.69 %OOIP). This outcome indicates that the strong IFT reduction achieved during surfactant flooding was already sufficient to mobilize the oil located near the fracture surfaces. Once this oil was displaced, switching to additional wettability-driven mechanism (S+LSW) could not provide additional benefit, as most of the readily accessible oil had already been recovered.

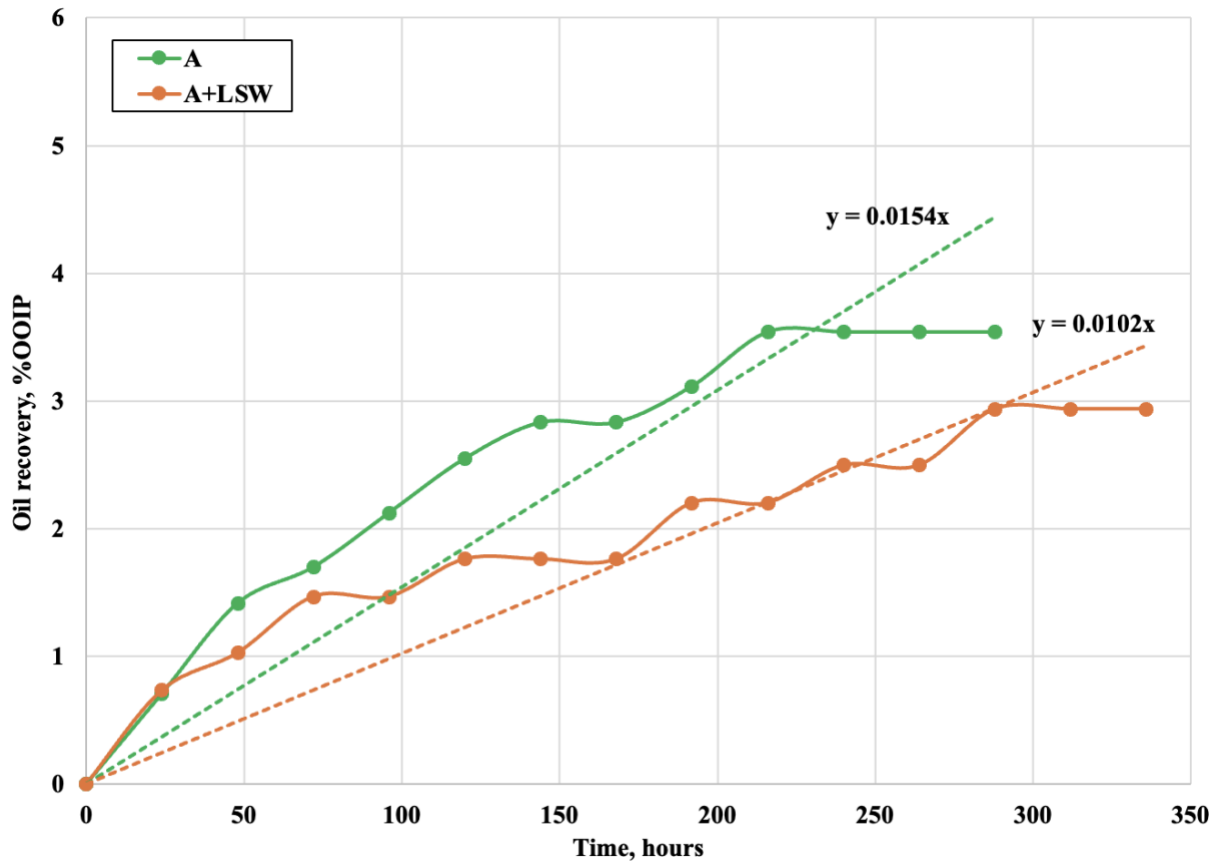


Figure 21. Comparison of standalone and hybrid alkali flooding methods

From Figure 21, the standalone alkali flooding in SW (A) was found to achieve only minimum recovery, as indicated by the rate of 0.0154 %/h and RF of 3.54 %OOIP. However, when the sequence was continued with the application of the hybrid LSW+A, some additional oil was recovered (0.0102 %/h; 2.94 %OOIP). The above results, therefore, indicate that although the alkali-based method is weak as standalone, the application of the hybrid is found to have mobilized an additional, although small, amount of oil.

From Figure 22, the application of nanoparticles in SW (NP) was found to perform moderately, as indicated by the recovery rate equal to 0.0734 %/h and RF of 11.58 %OOIP. However, when the sequence was continued with the application of the hybrid NP+LSW, additional recovery was achieved, nearly doubling the RF to 21.43%OOIP. This indicates that although the hybrids LSW+A and LSW+NP are not strong methods, their application immediately after standalone stage can still extract additional oil, extending the recovery process.

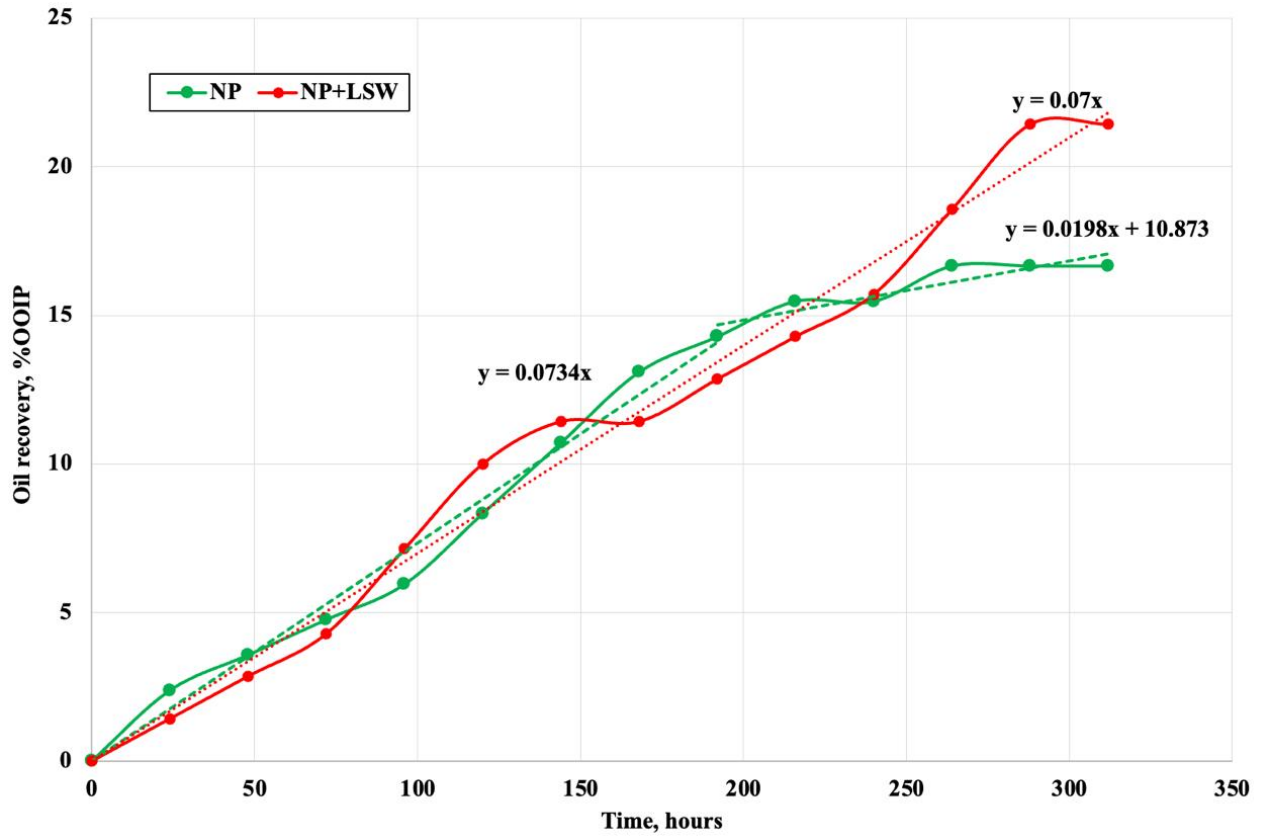


Figure 22. Comparison of standalone and hybrid nanoparticles flooding methods

4.5.2 Hybrid Injection After LSW Pretreatment

To evaluate whether hybrid formulations perform better when applied after an initial wettability alteration stage, SI tests were conducted where LSW flooding was followed by surfactant, alkali, or nanoparticle hybrids. The results are summarized in Tables 24-25 and Figure 23. For all tests listed, phase 1 is SI with SW and phase 2 is SI with LSW. RF values for phase 3 are normalized to the %OOIP measured after the preceding LSW phase has reached plateau.

For all hybrid EOR techniques, the second SI stage with LSW was used to provide a common baseline for comparison. Figure 23 presents the RF values normalized to the %OOIP after LSW imbibition. Imbibition with LSW was moderate and nearly identical across all cores, with imbibition rates ranging from 0.0157 to 0.0524 %/h and an average of 0.0339 %/h. The average value of RF after LSW imbibition equals 10.25 %OOIP. The response is slightly higher than average imbibition rate and RF reached by SI with SW (0.0304 %/h and RF 9.17 %OOIP). This can be attributed to the gradual wettability alteration induced by ionic dilution during LSW flooding.

Table 24. Comparison of imbibition rates for hybrid methods after LSW pretreatment

Test Number	Phase	Early Time Imbibition Rate (%/h)	Late Time Imbibition Rate (%/h)
1	LSW+S	0.1666	0.0433
2	LSW+A		0.0099
3	LSW+NP		0.0314

Table 25. Comparison of recovery factors for hybrid methods after LSW pretreatment

Test Number	Phase 2	Phase 3	RF of Phase 3 (%OOIP)	Interpretation
1	LSW	LSW+S	25.23	Synergistic improvement, strong recovery gain
2		LSW+A	3.04	Negative effect, recovery declined
3		LSW+NP	9.72	Similar to LSW phase

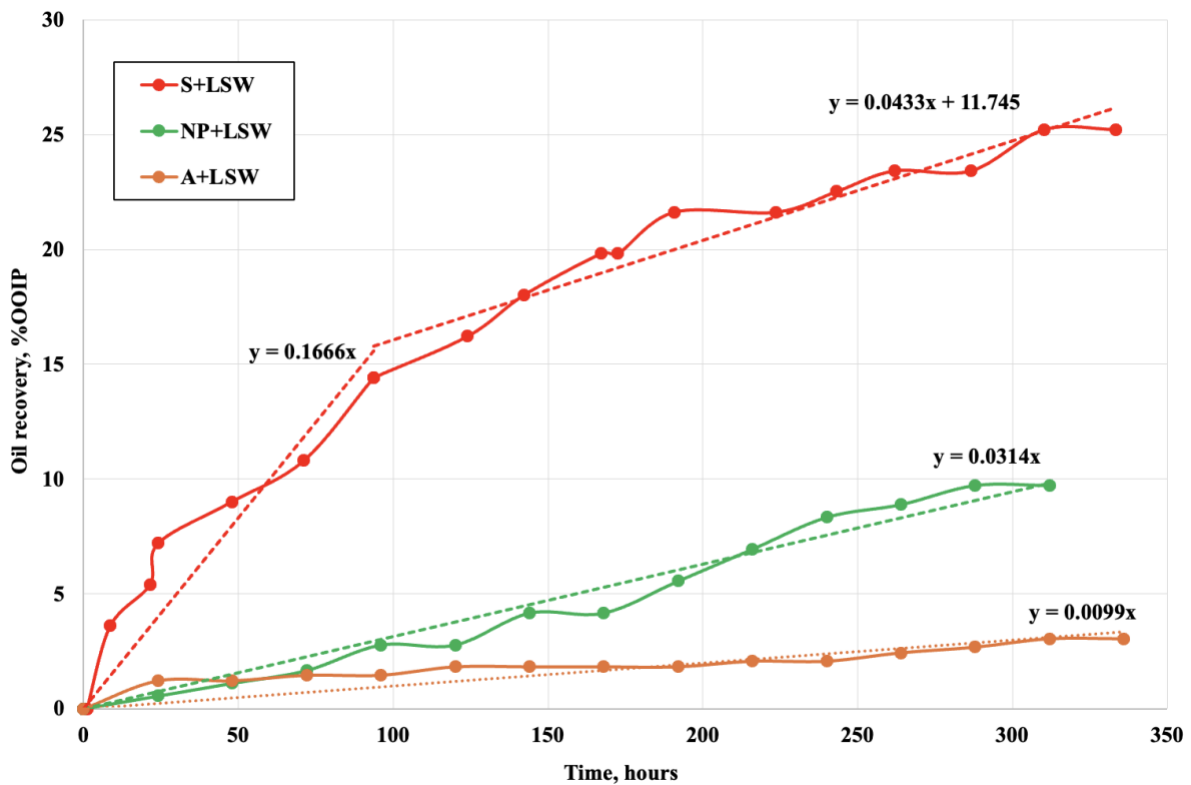


Figure 23. Comparison of hybrid EOR methods after LSW pretreatment

From Figure 23, after transitioning from LSW to LSW+S, the early-time imbibition rate increased sharply to 0.1666 %/h - almost five times higher than LSW alone - and the RF rose by a further 25.23 %OOIP. This shows that the hybrid stage was able to mobilize oil beyond what LSW had already displaced, combining the wettability alteration effect of LSW with the strong IFT reduction of the surfactant. Importantly, imbibition remained active for nearly two weeks, demonstrating that the hybrid not only accelerated recovery but also sustained it over a

longer period. Overall, this sequence confirms that applying LSW+S after LSW pretreatment is highly effective, as it recovers additional oil and maintains imbibition far longer than LSW flooding alone.

Once LSW flooding had already established RF of 10.25% OOIP, no additional benefit was gained with LSW+A. Instead, the imbibition rate decreased to 0.0099%/h, and the additional RF gained was only 3.04% OOIP. This suggests that the use of alkali in the LSW flooding process decreased the effectiveness of the LSW flooding process. The use of LSW+NP resulted in an additional gain. Although the imbibition rate remained relatively unchanged at 0.0314%/h, an additional gain of 9.72% OOIP was achieved. This suggests that the use of the hybrid process resulted in an additional gain, but it did not accelerate the imbibition process.

4.6 Ion-Modified Brine Effects

4.6.1 Screening of Ion-Modified LSW

In order to further improve the effectiveness of injection strategy, ion-modified brines were also investigated to assess whether a tailored adjustment in ionic composition could improve the effectiveness of wettability alteration. The goal here was to find an optimized formula for LSW, one that could induce a stronger and quicker transition to water-wet conditions.

CA measurements were carried out for a variety of ion-modified LSW formulations, and the results are presented in Table 26 and Figure 24. Images of contact angles for the investigated systems are presented in Appendix B. The initial CA for all analyzed systems were 178.5°, confirming strongly oil-wet conditions before treatment. Over time, all tested brine formulations displayed a decrease in CA, confirming their ability to alter wettability to a certain extent.

Table 26. Contact angle measurements for ion-modified LSW compositions

	Days			
	0	1	3	6
LSWCaMg-3SO ₄	178.5°	133.5°	133.3°	142.6°
LSW3Ca3Mg-SO ₄	178.5°	133.3°	137.0°	47.3°
LSW3Ca3Mg-3SO ₄	178.5°	168.6°	156.6°	45.1°

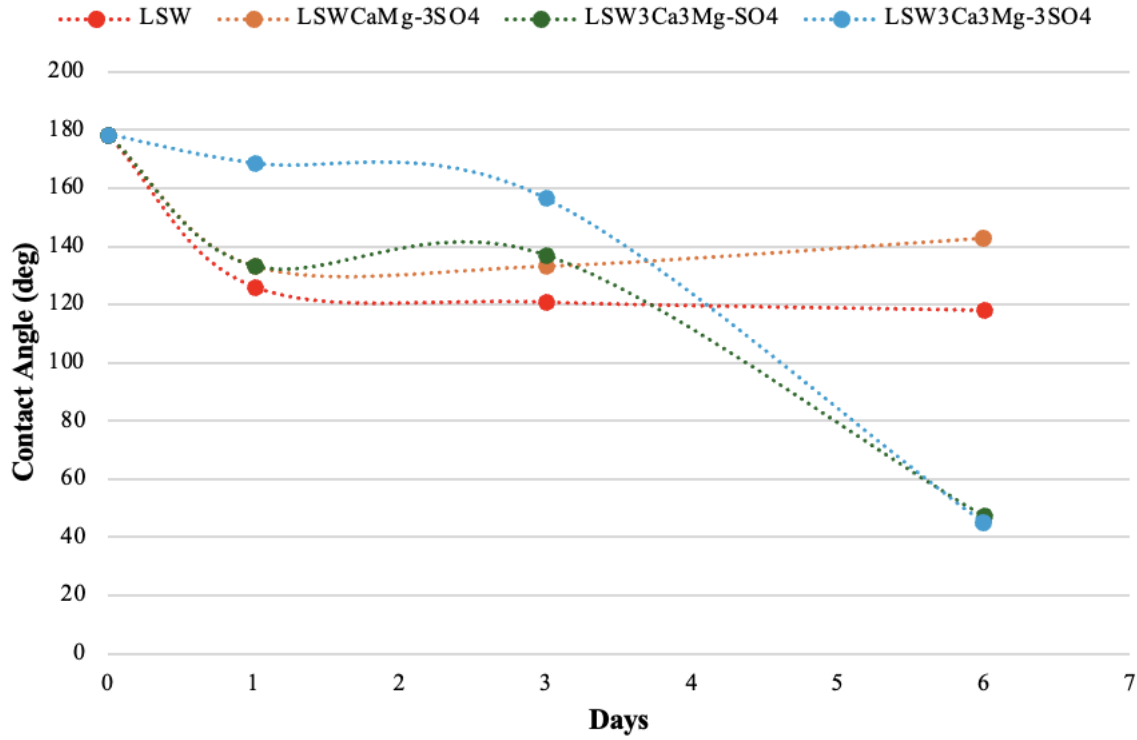


Figure 24. Contact angle measurements for ion-modified LSW compositions

Among the evaluated systems, the LSWCaMg-3SO₄ system showed limited improvement, with the CA remaining relatively high at 142.6°, suggesting weaker wettability modification. In contrast, the LSW3Ca3Mg-SO₄ system demonstrated the most pronounced wettability alteration, with the CA decreasing sharply to 47.3° after 6 days. This indicates a strong transition toward water-wet conditions and suggests enhanced interaction between the modified brine and the rock surface. Similarly, the LSW3Ca3Mg-3SO₄ system also resulted in a substantial reduction in CA to 45.1°, although the reduction occurred more gradually compared to the LSW3Ca3Mg-SO₄ system.

Based on these results, the LSW3Ca3Mg-SO₄ formulation was selected as the optimized LSW design for subsequent experiments, as it provided the most effective and rapid wettability alteration among the tested compositions. In addition, IFT measurements for this brine showed a value of 17.5 mN/m, indicating a noticeable reduction compared to conventional LSW (33.2 mN/m). This suggests that, in addition to wettability alteration, ion modification also contributes to improved interfacial behavior, further supporting the selection of this formulation for enhanced oil recovery applications.

Furthermore, since the LSW→LSW+S sequence was previously identified as the most effective hybrid design, additional mechanistic measurements were conducted for the ion-

modified system (LSW3Ca3Mg-SO₄+S). The results showed that the contact angle slightly decreased from 178.5° to 173.1°, indicating limited wettability alteration in the presence of surfactant. However, the IFT was significantly reduced to 0.15 mN/m, confirming that the surfactant dominates interfacial behavior in the hybrid system.

4.6.2 Imbibition Performance of Ion-Modified LSW

To evaluate the impact of ion modification on oil recovery, SI tests were conducted using the optimized LSW3Ca3Mg-SO₄ brine and compared with baseline and previously identified EOR systems. The results are summarized in Table 27 and Figure 25. For all tests listed, phase 1 is SI with SW. RF values are normalized to the %OOIP measured after the preceding phase has reached plateau.

Table 27. Comparison of imbibition rates for LSW-based methods

Phase	Early Time Imbibition Rate (%/h)	Late Time Imbibition Rate (%/h)	RF (%OOIP)
LSW		0.0206	6.38
LSW3Ca3Mg-SO ₄	0.0395	0.0024	6.81
LSW+S	0.1666	0.0433	25.23
LSW3Ca3Mg-SO ₄ +S	0.6944	0.0372	40.15

As shown in Figure 25 and summarized in Table 27, the ion-modified LSW3Ca3Mg-SO₄ system exhibited a higher early-time imbibition rate of 0.0395 %/h compared to the base LSW (0.0206 %/h), indicating faster initial oil displacement. This is reflected in the steeper initial slope of the recovery curve. However, the late-time imbibition rate decreased significantly to 0.0024 %/h, suggesting that imbibition slowed considerably after the initial stage. Despite the enhanced early-time kinetics, the overall recovery factor showed only a slight improvement, reaching 6.81 %OOIP compared to 6.38 %OOIP for LSW. This indicates that while ion modification accelerates early-stage imbibition, it does not significantly increase ultimate recovery when applied as a standalone brine.

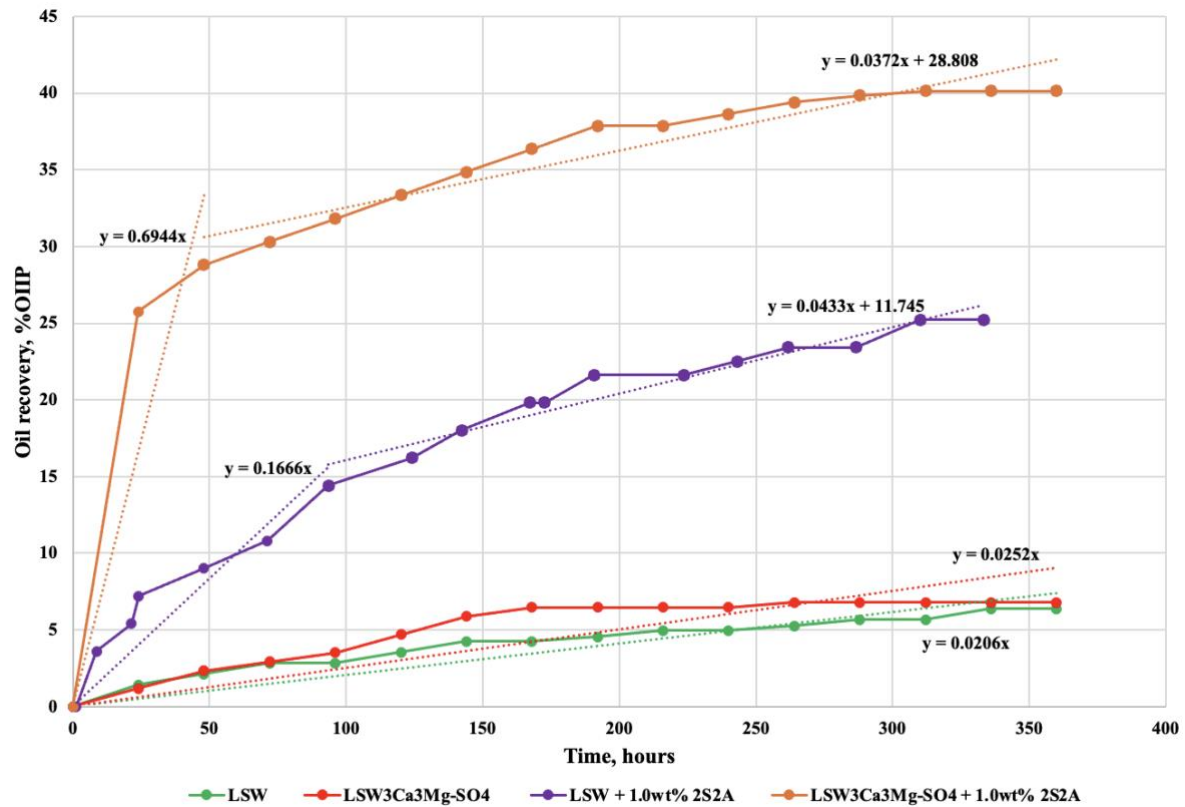


Figure 25. Comparison of hybrid EOR methods after LSW pretreatment

By comparing the conventional sequence (LSW→LSW+S) and the ion-modified sequence (LSW→LSW3Ca3Mg-SO₄+S), it is evident that the ion-modified method further improves the overall performance of the EOR process. Although the previous tests found that the LSW→LSW + S sequence is the best injection strategy, the inclusion of ion-modified LSW injection prior to surfactant injection significantly improved the imbibition kinetics and ultimate recovery. As shown in Figure 25 and Table 27, the ultimate recovery was significantly improved for the ion-modified sequence (LSW3Ca3Mg-SO₄+S), reaching 40.15 %OOIP compared to 25.23 %OOIP for the LSW+S method. Moreover, the early-time imbibition rate was significantly improved for the ion-modified sequence, reaching 0.6944 %/h compared to 0.1666 %/h for the LSW+S sequence, indicating a much faster oil displacement rate. It is evident that the improved wettability alteration, obtained through the ion-modified brine injection, enhances the effectiveness of the subsequent surfactant injection.

In summary, even though LSW→LSW+S was found to be the optimal conventional sequence, the optimized sequence using ion-modified LSW is an even better and more efficient method for maximizing oil recovery.

5.1 Mechanisms of Imbibition Enhancement

Figure 10 is a log-log plot showing the relationship between the change in contact angle (ΔCA , %) on the y-axis and the change in interfacial free tension (ΔIFT , mN/m) on the x-axis. The y-axis ranges from 0 to 80, and the x-axis ranges from 0.1 to 100. The plot is divided into three main regions: 'Wettability-dominated system' (top right), 'Intermediate (synergistic) regime' (middle), and 'IFT-dominated systems' (bottom right). Data points are labeled with material combinations: LSW3Ca3Mg-SO4, LSW, NP, LSW + NP, LSW + A, LSW3Ca3Mg-SO4 + S, LSW + S, A, and S. Green text labels indicate 'Stronger wettability alteration' (top left), 'Limited IFT reduction' (middle left), 'Strong IFT reduction' (middle right), and 'Limited wettability alteration' (bottom left).

As illustrated in Figure 26, the evaluated systems can be grouped into three mechanistic regimes. Wettability-dominated systems, such as LSW and ion-modified brine (LSW3Ca3Mg-SO₄), exhibit high ΔCA values with relatively moderate ΔIFT , indicating that their performance is primarily controlled by wettability alteration. These systems promote a gradual shift toward more water-wet conditions, which enhances capillary forces and supports sustained imbibition. However, this process is inherently slow, as it relies on surface interactions such as ion exchange and EDL expansion. This is reflected in the relatively low

imbibition rates observed for LSW (~ 0.02 %/h), despite its ability to contribute to incremental recovery over time.

In contrast, surfactant-based systems fall within the IFT-dominated regime, characterized by minimal ΔCA and significant ΔIFT reduction. In these systems, the dominant mechanism is the reduction of capillary resistance, which facilitates rapid oil detachment and mobilization. As a result, surfactant flooding exhibits significantly higher early-time imbibition rates (~ 0.46 %/h), demonstrating that IFT reduction is the primary factor controlling imbibition kinetics. However, because these systems do not significantly alter wettability, their performance is mainly associated with rapid initial recovery rather than gradual capillary-driven displacement.

Nanoparticle and alkali-based systems are positioned closer to the IFT-dominated region in Figure 26 due to their relatively higher ΔIFT compared to ΔCA . However, their overall performance indicates that neither IFT reduction nor wettability alteration is sufficiently strong to dominate the imbibition process. Despite their apparent proximity to the IFT region, these systems exhibit low imbibition rates and limited recovery, confirming that they do not behave as true IFT-dominated systems. Instead, their positioning reflects weak or ineffective contributions from both mechanisms, rather than the presence of a strong governing process. This highlights that the location of a system in the ΔCA - ΔIFT space represents the relative magnitude of measured effects and must be interpreted in conjunction with imbibition performance.

The direct comparison between the selected systems clearly demonstrates that imbibition rate and ultimate recovery are controlled by different mechanisms. IFT reduction is the dominant factor governing imbibition rate, as it reduces resistance to flow and enables rapid oil mobilization, while wettability alteration controls the extent of recovery by strengthening capillary forces and sustaining imbibition over time. Therefore, when comparing individual mechanisms, IFT-driven systems are more effective for accelerating oil production, whereas wettability-driven systems are essential for achieving sustained recovery.

This comparison also explains the superior performance of combined approaches. The most effective EOR strategy is achieved by integrating both mechanisms, where wettability alteration first prepares the rock surface and activates capillary forces, followed by IFT reduction that accelerates oil mobilization. This is consistent with the observed performance of LSW+S sequences, where the combination of mechanisms results in both improved imbibition

rate and increased recovery. Furthermore, ion-modified brine enhances this effect by strengthening wettability alteration, thereby improving the efficiency of subsequent IFT-driven processes.

Overall, Figure 26 serves as a mechanistic selection and comparison framework, demonstrating that effective SI enhancement requires not only identifying dominant mechanisms but also understanding how they influence imbibition rate and recovery. The results confirm that neither wettability alteration nor IFT reduction alone is sufficient, and that optimal performance is achieved through their combined and properly sequenced application.

5.2 Comparison of Standalone and Hybrid EOR Approaches

The comparison of the standalone approaches with the hybrid EOR designs, shown in Figure 27, evaluates how the dominant mechanisms translate into differences in imbibition rate and ultimate recovery.

Among the standalone EOR approaches, the improvement in the imbibition rate is most pronounced in the surfactant flooding method, which clearly indicates the dominance of the IFT reduction mechanism in governing the oil imbibition kinetics. The high early-time imbibition rates clearly indicate the ability of the IFT reduction approach to mobilize the oil quickly, allowing immediate oil production from the pore network. This confirms that IFT-driven systems are the most effective for accelerating SI and achieving fast oil recovery.

In contrast, wettability-driven systems such as LSW and nanoparticles exhibit substantially slower imbibition kinetics. The relatively low imbibition rates observed for these systems reflect the time-dependent nature of surface-controlled processes, including ion exchange, EDL expansion, and nanoparticle adsorption. As a result, oil displacement occurs more gradually, with recovery increasing steadily over time rather than through rapid initial production. This confirms that wettability alteration primarily contributes to sustained recovery, rather than acceleration of imbibition.

A key observation from this comparison is that the presence of a mechanism alone does not guarantee effective performance; rather, its strength and activation under reservoir conditions are critical. Alkali flooding exhibited the weakest performance among the standalone methods, which can be attributed to the absence of an effective governing mechanism. The low TAN of the crude oil limits the potential for in-situ surfactant generation, preventing meaningful IFT

reduction. As a result, both imbibition rate and recovery remain low, confirming that the mechanism is not activated. Similarly, nanoparticle systems, although capable of altering wettability, exhibit limited effectiveness under the tested conditions, indicating that weak or partially activated mechanisms do not translate into significant improvements in recovery.

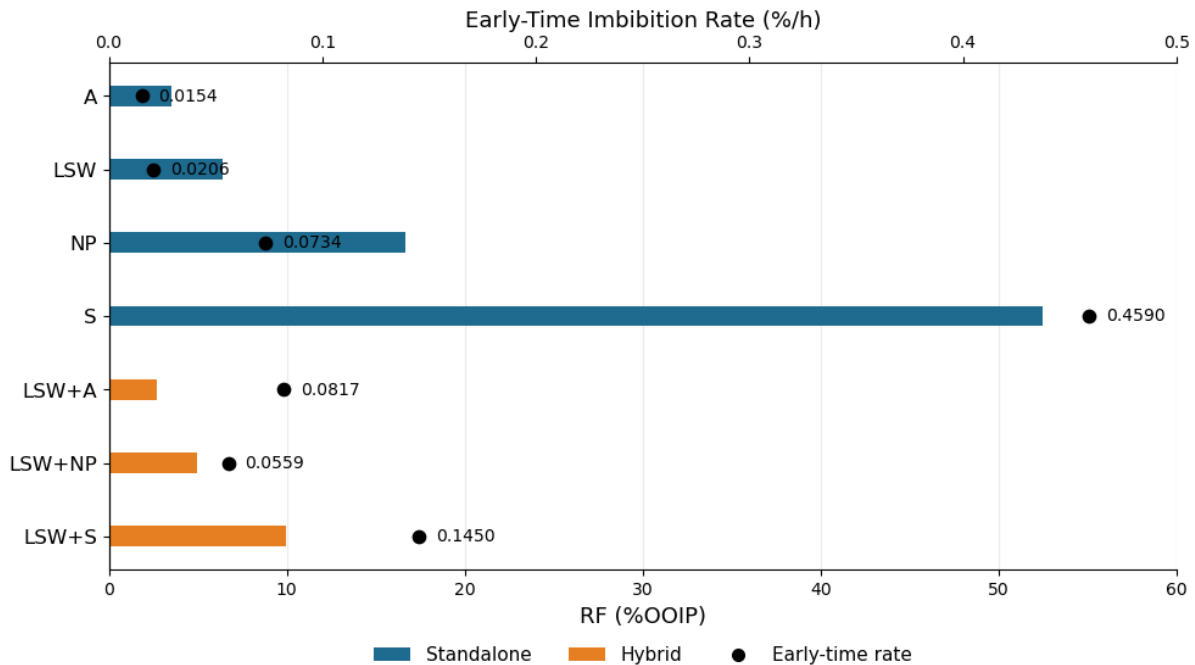


Figure 27. Comparison of recovery factors and imbibition rates for standalone and hybrid EOR systems

Hybrid EOR systems were expected to combine the advantages of multiple mechanisms; however, the results demonstrate that hybridization does not inherently lead to improved performance. Among the evaluated systems, only the LSW+S hybrid showed a meaningful improvement compared to LSW alone, indicating that hybrid effectiveness depends on the preservation of a strong governing mechanism. In this case, the surfactant retained its ability to reduce IFT, resulting in an increase in imbibition rate and improved recovery.

However, the performance of the LSW+S system remained lower than that of the standalone surfactant, indicating that the effectiveness of IFT reduction was partially reduced under low-salinity conditions. This decline can be attributed to several coupled mechanisms: (i) dilution of surfactant micelles in the low-salinity medium, which raises the CMC and reduces the extent of IFT reduction; (ii) increased surfactant adsorption onto the carbonate surface at lower ionic strength due to weaker electrostatic shielding, leading to rapid loss of active surfactant from solution. These findings demonstrate that even strong mechanisms can be weakened when transferred into hybrid systems if reservoir conditions are not favorable.

In contrast, hybrid systems including LSW+A and LSW+NP did not show improvement and, in some cases, resulted in reduced performance. The main reason for underperformance of A + LSW is the low TAN (0.08 mg KOH/g) of the crude oil, which limits the possibility of generating sufficient in-situ surfactants. Second, under low-salinity conditions the aqueous buffering capacity, ability of a fluid to resist changes in pH, is weaker. In addition, reduced ionic strength in LSW increases electrostatic repulsion between negatively charged carbonate surfaces and alkali anions, further limiting beneficial rock-fluid interactions. Similarly, nanoparticle performance is reduced in LSW due to decreased stability and weaker adsorption caused by changes in EDL interactions and reduced concentrations of divalent ions (Ca^{2+} , Mg^{2+}). As a result, neither system is able to effectively activate its governing mechanism in hybrid form.

Overall, the comparison demonstrates that EOR performance is controlled not by the number of mechanisms present, but by the strength and preservation of the dominant mechanism. IFT-driven systems are the most effective for accelerating oil production due to their high imbibition rates, while wettability-driven systems contribute to slower but sustained recovery. Hybrid systems can outperform standalone methods only when they successfully combine these mechanisms without diminishing their effectiveness. Therefore, optimal EOR performance is achieved through mechanism-based design, where strong wettability alteration is complemented by efficient IFT reduction under conditions that keep both mechanisms.

5.3 Influence of Sequencing on Oil Recovery

The results of this study clearly demonstrate that the order in which EOR methods are applied has a significant impact on oil recovery performance. While standalone and hybrid systems showed varying effectiveness, the sequencing of mechanisms was found to be a critical factor governing both imbibition kinetics and ultimate recovery.

As shown in Figure 28, the impact of injection order varies significantly depending on the strength and nature of the governing mechanism. For IFT-dominated systems such as surfactant flooding, the majority of recovery is achieved during the standalone stage. The high imbibition rate associated with strong IFT reduction enables rapid oil mobilization at early times, leaving limited remaining oil for subsequent recovery. As a result, the additional contribution from the hybrid stage (LSW+S) is minimal, indicating that applying wettability alteration after IFT-

driven mobilization does not significantly enhance performance once the primary mechanism has been exhausted.

In contrast, systems governed by wettability alteration exhibit a different response to sequencing. Nanoparticle-based systems, which rely on gradual surface modification, do not fully mobilize oil during the initial stage. Consequently, the subsequent injection of LSW+NP contributes to additional recovery, as further wettability alteration and fluid-rock interaction continue to enhance capillary-driven displacement. However, despite this improvement, the overall performance remains moderate due to the relatively weak governing mechanism and the absence of significant IFT reduction.

For alkali systems, both standalone and hybrid contributions remain low regardless of injection order. This confirms that sequencing cannot compensate for the absence of an effective governing mechanism. As a result, neither imbibition rate nor recovery is significantly improved, and no synergistic effect is observed.

A key outcome of this analysis is that applying IFT-driven mechanisms first leads to rapid recovery, whereas applying wettability-driven mechanisms first enables subsequent processes to act more effectively. This is clearly demonstrated in Figure 29, where LSW is used as a preconditioning stage prior to hybrid injection. In this sequence, LSW gradually shifts the rock toward more water-wet conditions, enhancing capillary forces and preparing the system for oil displacement. When followed by surfactant injection, the reduction in IFT results in a sharp increase in imbibition rate, leading to both faster oil production and higher incremental recovery.

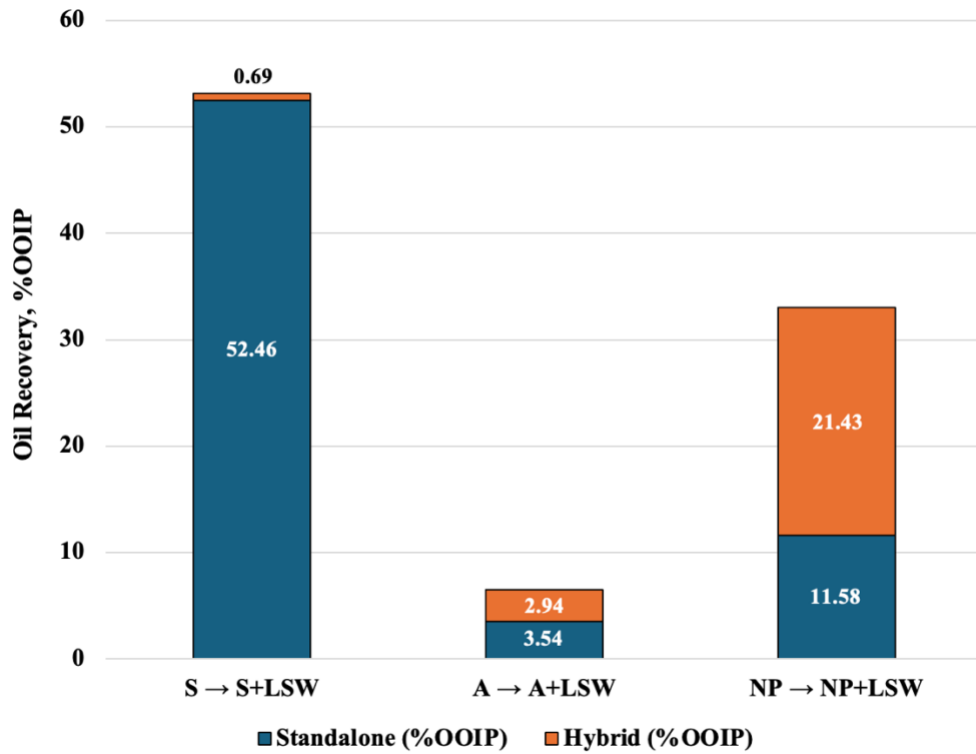


Figure 28. Comparison of oil recovery factors for standalone flooding and the additional recovery obtained after subsequent hybrid flooding

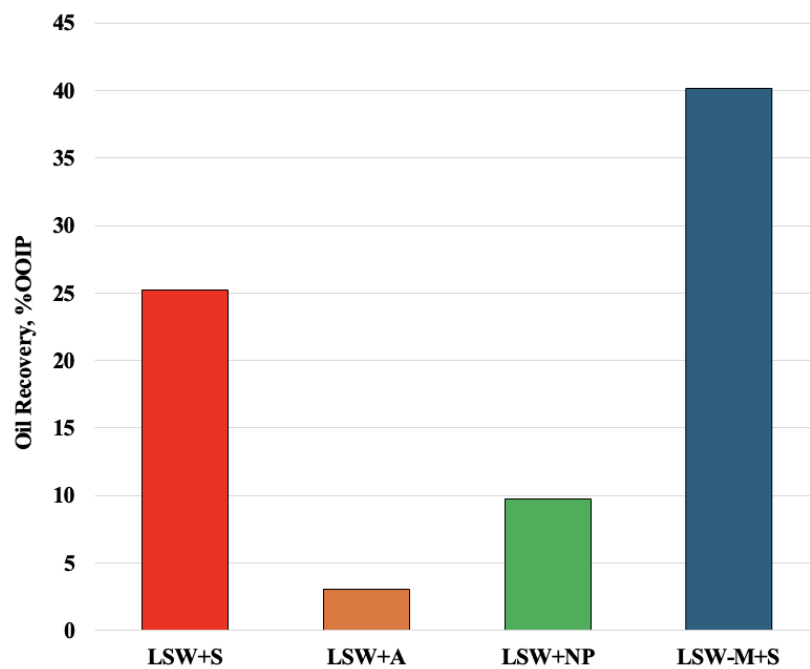


Figure 29. Comparison of recovery factors of hybrid methods after LSW pretreatment

Among the evaluated configurations, the LSW→LSW-M+S sequence provides the most effective combination of mechanisms. The initial wettability alteration stage activates capillary-driven imbibition, while the subsequent IFT reduction stage accelerates oil

mobilization. This sequence successfully integrates both mechanisms without compromising their effectiveness, resulting in superior performance compared to other tested strategies. In contrast, LSW+NP provides only limited additional recovery without significantly improving imbibition rate, while LSW+A remains ineffective due to the absence of a strong governing mechanism.

Furthermore, the effectiveness of the LSW→LSW-M+S sequence is strongly influenced by the ionic composition of the chemical flood applied in the final stage. In this study, ion-modified brine was introduced in the LSW-M+S stage to enhance surfactant effectiveness by optimizing ionic interactions at the rock-fluid interface, thereby improving IFT reduction, oil mobilization, and overall recovery performance.

Overall, the results demonstrate that optimal EOR performance in fractured carbonate systems is achieved through mechanism-based sequencing rather than standalone application or simple hybridization. Wettability alteration should be applied first to prepare the system and activate capillary forces, followed by IFT reduction to accelerate oil mobilization. This approach maximizes both imbibition rate and ultimate recovery, providing a clear strategy for designing effective EOR processes in fractured reservoirs.

5.4 Methodology for Designing EOR Strategies in Fractured Reservoirs

The findings from this study prove that successful EOR strategy design in fractured carbonate reservoirs involves a combination of wettability and IFT reduction through a proper sequence of stages during injection. The findings also show that successful evaluation of the performance of EOR strategies does not only consider ultimate recovery, but also considers imbibition rate, which controls the speed of oil production. Therefore, EOR strategies must be designed to balance rapid imbibition kinetics with sustained recovery efficiency.

The proposed methodology for developing EOR strategy in fractured carbonate reservoirs has been presented in Figure 30, and it includes a combination of reservoir characterization, mechanism selection, and injection strategy to optimize recovery and imbibition rate in reservoir conditions.

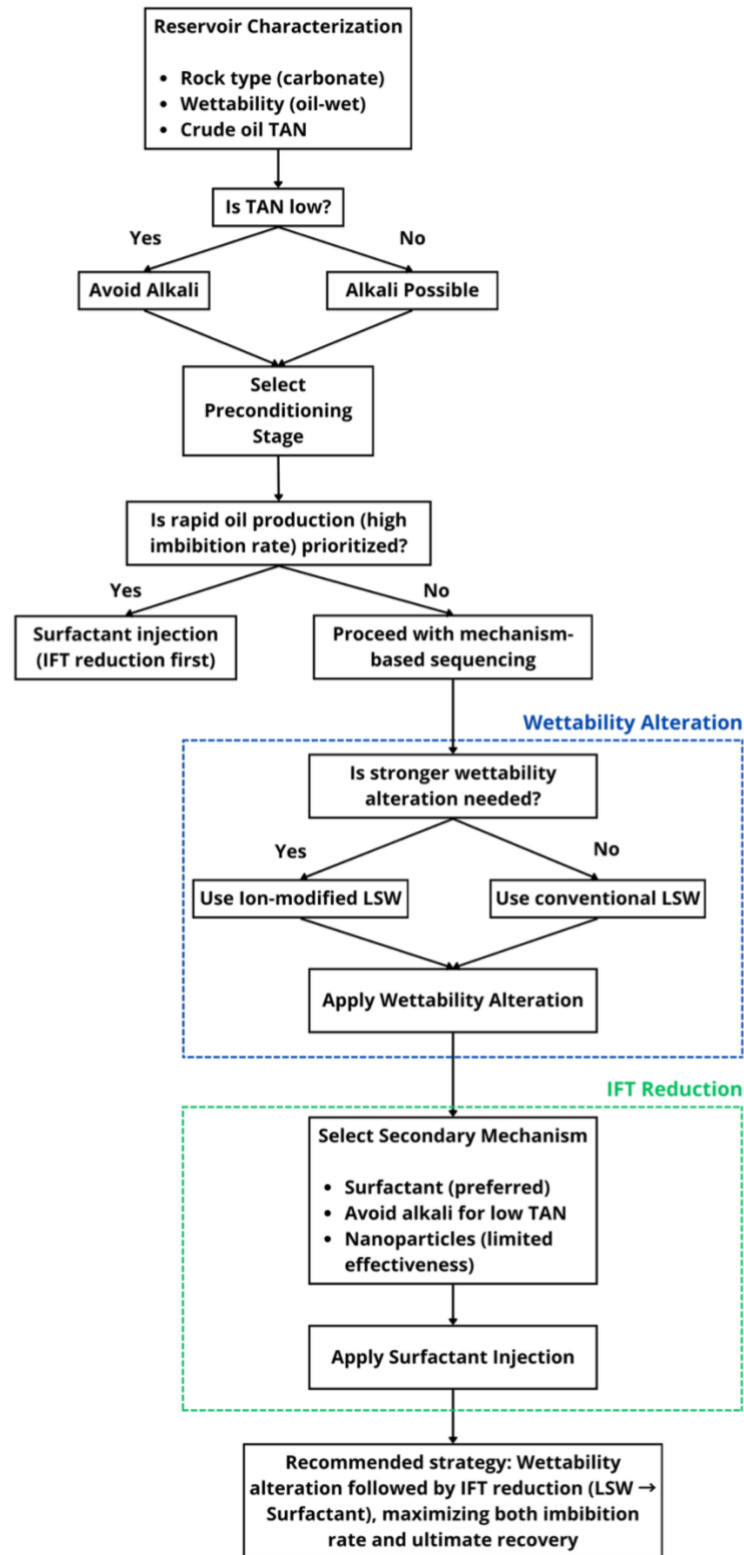


Figure 30. Workflow for designing EOR strategies in fractured carbonate reservoirs, highlighting the sequential application of wettability alteration and interfacial tension reduction mechanisms

The design process starts with the characterization of the reservoir, where important parameters such as the type of rock, the nature of the wettability condition, and the properties of the crude

oil, such as TAN, are considered. The parameters are important because they affect the feasibility and efficiency of different EOR methods. For instance, low-TAN oil does not allow for the application of alkali methods, and these methods are to be avoided.

Once the reservoir screening has been carried out, a preconditioning stage is chosen, and a critical choice has to be made depending on the main production goal. This includes determining whether rapid oil production (high imbibition rate) has to be emphasized. If rapid oil production is a necessity, then IFT-driven methods such as direct surfactant injection can be employed first to accelerate oil production. However, if the goal includes optimizing both recovery and production rate, a mechanism-based sequencing approach is employed.

In the mechanism-based approach, wettability alteration is incorporated as the first step to modify the interactions between rock and fluid, and to improve the capillary forces. Depending on the required extent of wettability alteration, either conventional LSW or ion-modified LSW can be employed. The findings from this study prove that ion-modified LSW has a stronger and quicker effect on wettability alteration, thus making it a more efficient approach for preparing the rock surface for subsequent processes.

Subsequently, a second mechanism is chosen for its potential to further improve oil mobilization. Of all the mechanisms considered, surfactant flooding is determined to be the best approach based on its high capacity for IFT reduction and imbibition rate increase. In contrast, alkali and nanoparticle-based systems show limited potential for EOR, either because of low activation of the respective mechanisms or due to sensitivity reservoir properties.

The above process demonstrates how the proposed approach can be utilized to identify an optimal EOR design, where it is evident that wettability alteration and IFT reduction must be applied together. By considering wettability alteration followed by surfactant flooding, it is possible to achieve high rates of capillary-based mobilization and oil production.

6. CONCLUSIONS AND RECOMMENDATIONS

The study aimed to assess the performance of standalone and hybrid EOR methods and their effect on SI in a fractured carbonate reservoir. The results indicate that the recovery efficiency is significantly affected by the major recovery mechanism and the sequencing strategy for the chemical methods.

The results indicate that, for a fractured carbonate system, the major recovery mechanism for accelerating imbibition is IFT reduction. Surfactant flooding in SW produced the best results, especially for the early-time rate and ultimate RF, proving that the rate of oil production is controlled by the IFT reduction. The results also show that LSW and nanoparticles, on the other hand, affected the system through a gradual change in wettability, resulting in moderate recovery, although with a slower rate. This confirms that imbibition rate and ultimate recovery are governed by different mechanisms: IFT reduction controls production rate, while wettability alteration controls recovery extent.

From the analysis of standalone and hybrid systems, it is apparent that the hybrid approach does not automatically ensure improved performance. In this regard, while LSW+NP systems only showed modest improvement, alkali systems were found to be ineffective when subjected to low-TAN conditions. The greatest improvement was achieved through mechanistic sequencing rather than simple combination.

The results clearly demonstrate that the effect will be the best when the wettability shift takes place before the IFT modification. The experiment conducted using LSW→LSW+S shows that there is an apparent synergy effect as a result of which the rock surface was prepared through LSW and after that IFT reduction takes place by surfactant addition. This design resulted in both increased imbibition rate and higher ultimate recovery.

These findings offer clear guidelines for EOR design in oil-wet fractured carbonate formations. LSW is most effective as a preconditioning process rather than as a standalone EOR method. The optimal approach for EOR strategy involves application of LSW pretreatment followed by ion-modified LSW and surfactant flooding, ensuring both kinetic enhancement and maximized recovery.

Additionally, ion-modified LSW was found to be more effective in altering wettability than LSW, making the preconditioning process more effective.

Based on the findings of this study, further research is recommended to expand and validate the findings of this study:

1. Dynamic flooding experiments: Extend the testing of selected standalone and hybrid EOR approaches with coreflooding experiments to consider dynamic flow behavior, including the effects of capillary-viscous interactions, fracture-matrix transfer, with emphasis on optimized injection strategy (wettability alteration followed by IFT reduction).
2. Numerical modeling: Develop and apply numerical models to simulate SI under varying wettability and IFT conditions, enabling quantitative assessment of the combined effects of wettability alteration and IFT reduction at the core scale.
3. Upscaling to reservoir-scale applications: Integrate the experimental and modeling results into reservoir simulation studies to evaluate the performance of optimized injection strategies in fractured carbonate systems.

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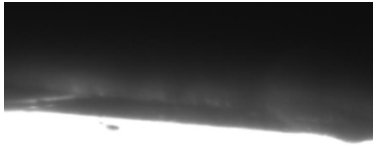
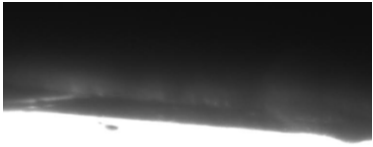
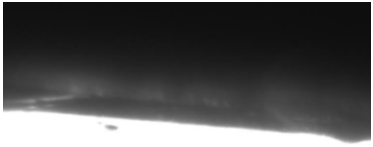
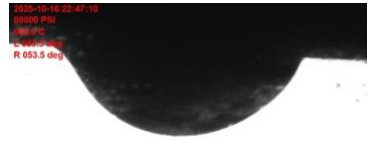
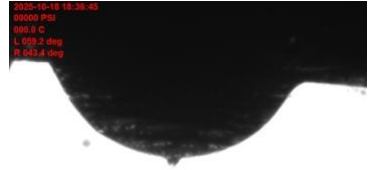
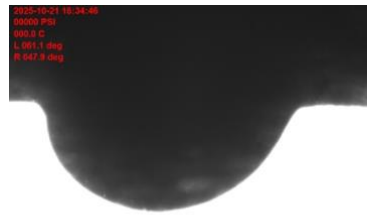
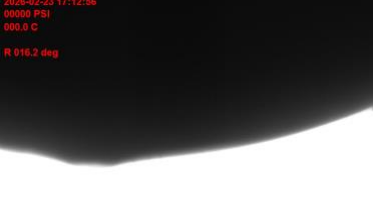
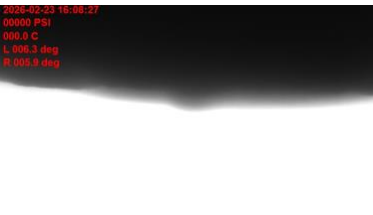
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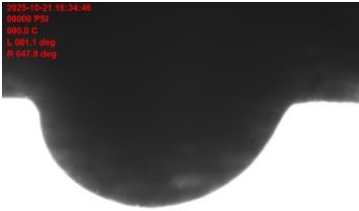
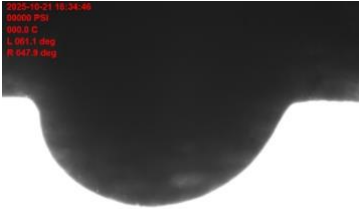
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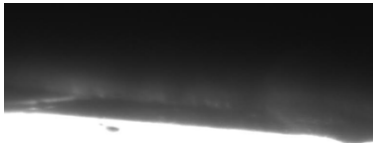
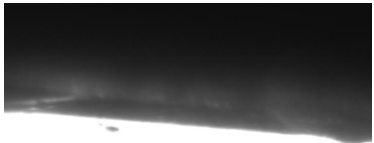
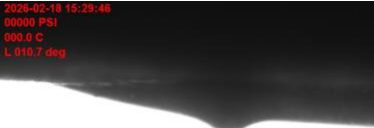
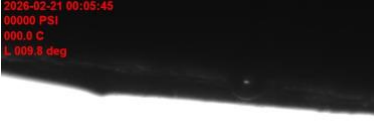
8. APPENDICES

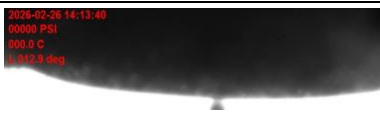
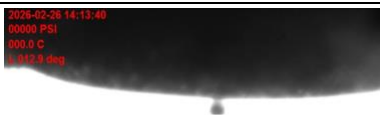
Appendix A1. Contact angle measurement images for selected EOR systems

Time (days)	LSW	NP	LSW + S
0	 178.5°	 178.5°	 178.5°
1	 126.2°	 173.4°	 175.1°
3	 120.8°	 168.5°	 173.8°
6	 118.0°	 103.0	 173.7°

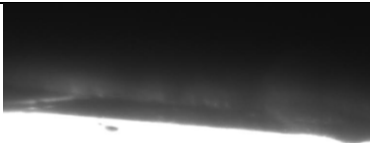
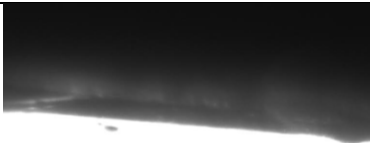
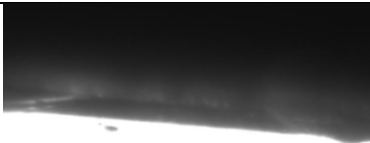
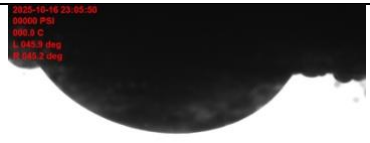
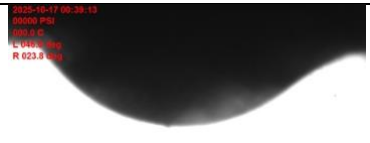
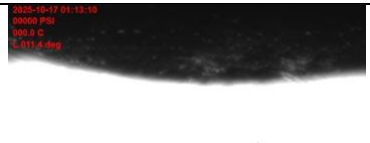
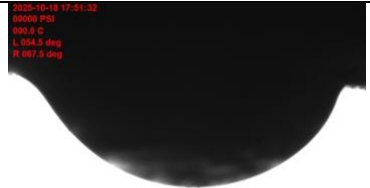
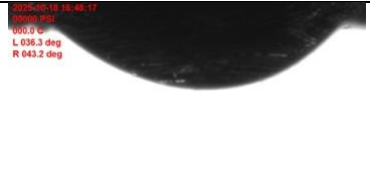
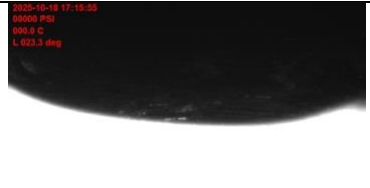
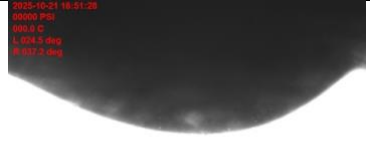
9	 118.0°	 161.9°	 173.4°
12	 118.0°	 154.5°	 173.4°

Appendix A2. Contact angle measurement images for selected EOR systems

Time (days)	LSW+A	LSW+NP
0	 178.5°	 178.5°
1	 172.3°	 169.3°
3	 170.2°	 166.7°

6	2026-02-23 17:24:38 00000 PSI 000.0 C L 012.6 deg  100.0	2026-02-23 16:59:52 00000 PSI 000.0 C L 018.4 deg  161.6°
9	2026-02-26 14:13:40 00000 PSI 000.0 C L 012.8 deg  167.1°	2026-02-26 12:51:22 00000 PSI 000.0 C L 022.7 deg  157.5
12	2026-02-26 14:13:40 00000 PSI 000.0 C L 012.8 deg  167.1°	2026-02-26 13:02:37 00000 PSI 000.0 C L 031.3 deg R 022.7 deg  150.0°

Appendix B. Contact angle measurement images for ion-modified LSW systems

Time (days)	LSWCaMg-3SO ₄	LSW3Ca3Mg-SO ₄	LSW3Ca3Mg-3SO ₄
0	 178.5°	 178.5°	 178.5°
1	 133.5°	 133.3°	 168.6°
3	 133.3°	 137.0°	 156.5°
6	 142.6°	 47.3°	 45.1°