DATA-DRIVEN ANALYSES OF LOW SALINITY WATERFLOODING IN CARBONATES

by

RASHIDA SALIMOVA

2021

Thesis submitted to the School of Mining and Geosciences of Nazarbayev University in Partial Fulfillment of the Requirements for the Degree of
Master of Science in Petroleum Engineering

Nazarbayev University
April, 2021

Acknowledgements

I would like to express my deep gratitude to my supervisor Professor Peyman Pourafshary for giving me the opportunity to do this research, for his patience and motivation, for providing invaluable guidance throughout the research and thesis writing. I am also grateful to my co-supervisor Professor Lei Wang for his supervision and valuable recommendations. The completion of this work could not have been accomplished without their support.

I would like to thank my family and friends for their support and encouragement throughout my life.

Originality Statement

I, Rashida Salimova, hereby declare that this submission is my own work and to the best of my knowledge it contains no materials previously published or written by another person, or substantial proportions of material which have been accepted for the award of any other degree or diploma at Nazarbayev University or any other educational institution, except where due acknowledgement is made in the thesis.

Any contribution made to the research by others, with whom I have worked at NU or elsewhere is explicitly acknowledged in the thesis.

I also declare that the intellectual content of this thesis is the product of my own work, except to the extent that assistance from others in the project's design and conception or in style, presentation and linguistic expression is acknowledged.

Signed on DATE

ABSTRACT

Maximizing crude oil recovery is a main objective of the oil and gas industry. Oil recovery by natural production in carbonates is usually lower than 30%. Thus, Enhanced Oil Recovery (EOR) methods are used to increase the oil production in carbonate reservoirs. Low salinity water (LSW) injection is a promising EOR technique, which have been studied by many researchers for potential improvement of oil recovery.

LSW flooding in carbonates has been widely evaluated by coreflooding tests in prior studies. A closer look in the literature on LSW in carbonates indicates a number of gaps and shortcomings. It is difficult to understand the exact relationship between different controlling parameters and the LSW effect in carbonates. The active mechanisms involved in oil recovery improvement are still uncertain, and more analyses are required. To predict the LSW performance and study the mechanisms of oil displacement, data collected from available experimental studies on LSW injection in carbonates were analyzed using data analysis approaches.

In this thesis, I collected data from 26 secondary and 117 tertiary coreflooding tests. Machine learning (ML) and statistical approaches were utilized to analyze the extracted main parameters. We used a linear regression model to study the linear relationship between single parameters and incremental recovery factor (RF). Correlations between rock, oil, brine properties and tertiary RF were negligible and weak. Subsequently, we analyzed the effect of brine and oil/brine parameters (oil acidity, alteration in salinity and active ions concentration) on LSW performance using multivariable linear regression. Relatively stronger linear correlation was found for a combination of oil/brine parameters and RF. We also studied the nonlinear relationship between parameters by applying ML nonlinear models, such as Artificial Neural Networks (ANN), Support Vector Machine (SVM) and Decision Tree (DT). These models showed better data fitting results compared to linear regression. Strong and very strong relationships between properties and RF were achieved by ML models. Among the used ML models, DT provided the best correlation for oil/brine parameters, as ANN and SVM overfitted the testing data. Finally, different mechanisms involved in the LSW effect were analyzed based on the changes in the effluent PDIs concentration, interfacial tension, pH, zeta potential, pressure drop. Wettability alteration by LSW was commonly observed in coreflooding tests.

TABLE OF CONTENT

List of Figures

List of Tables

Nomenclature

Abbreviations

AN	Acid Number
ANN	Artificial Neural Networks
BN	Base Number
CBR	Crude oil-Brine-Rock
DT	Decision Tree
EDL	Electric Double Layer
E&P	Exploration & Production
EOR	Enhanced Oil Recovery
HS	High Salinity
IFT	Interfacial Tension
LS	Low Salinity
LSW	Low Salinity Water
MIE	Multicomponent Ionic Exchange
ML	Machine Learning
MRE	Mean Relative Error
OOIP	Original Oil in Place
PDIs	Potential Determining Ions
RF	Recovery Factor
SVM	Support Vector Machine
TDS	Total Dissolved Ions
PVT	Pressure-volume-temperature

Symbols

S_{or}	Residual oil saturation
S_{wi}	Irreducible water saturation
k	Permeability
β_o	Intercept
β_1	Slope of the line
ε_i	Error
x_i	The i^{th} observation of independent variable
y_i	The i^{th} observation of dependent variable
z_j	Sum of the weighted inputs
w_i	Weight factor
b_j	Bias
${w_j}^n$	The output layer weight
ω	Vector variable
C	Constant
ξ_i	Slack variable
ε	Margin

CHAPTER 1: INTRODUCTION

1.1 Background

Maximizing crude oil recovery is a main objective of the oil and gas industry. Thus, hydrocarbon primary, secondary, and tertiary recovery operations are applied. The tertiary recovery method, also known as Enhanced Oil Recovery (EOR), mobilizes residual oil left behind by secondary methods. Estimates show that approximately 60% of world's oil reserves are held in carbonate reservoirs (Akbar et al., 2000). The amount of oil that can be produced from these reservoirs by natural production is below 30%. This small value of oil recovery can be the result of the heterogeneous parameters, low matrix permeability, fractures, and oil-wet conditions in carbonates. Hence, EOR methods are required to reduce the residual oil and increase the oil production.

Low salinity water (LSW) flooding is one of the promising techniques for EOR in carbonate formations. It is a process of injecting low saline water with an optimized ion composition into the reservoir in order to recover incremental oil (Derkani et al., 2018). Recent research has shown that brine with the controlled salinity and ionic composition can achieve higher oil recovery (up to 10% and more) than the regular waterflooding method (Strand et al., 2008). LSW destabilizes the equilibrium of the initial crude oil-brine-rock (CBR) system and alters the original wettability conditions (Derkani et al., 2018). The advantages of the injection of low saline brine are minimal cost and no increased injection problems. Although most EOR techniques are not applicable during the late life cycle of the reservoir, LSW can be used during the late stages of the oil recovery process. It is also an environmentally friendly method of EOR (Austad et al., 2012).

Machine learning is a part of computer science in which data analysis is used to make predictions and decisions with minimal human intervention (Mitchell, 1997). Machine learning models have been successfully applied in different disciplines of petroleum industry (Wang & Fu, 2018; Mohamed & Kederitz, 2000; Bakshi et al., 2017). A closer look in the literature on LSW in carbonates indicates a number of gaps and shortcomings. It is challenging to understand the relationship between various parameters and the low salinity effect in carbonates. The mechanisms involved in increasing oil recovery are still not clear and more analyses are required. Available data from the literature can be analyzed using data analysis methods to predict the performance of LSW in carbonates and study the active mechanisms of oil displacement.

1.2 Literature Review

Different EOR techniques, such as LSW, are used to extract more oil from carbonate rock. It is believed that main parameters which control the oil recovery improvement by LSW are the composition of injected water, formation water, and oil, temperature, pressure, and pH. The amount of active and inactive ions in the injected brine and the porous media is also considered to be essential for the LSW performance.

Different coreflooding and imbibition experiments have been conducted in order to observe and study governing parameters and active mechanisms during LSW. The main target of coreflooding tests is to study the effect of LSW design and injection mode on incremental oil recovery. Some fluid/rock properties are reported to affect the performance of LSW more than others. Based on these parameters, a few mechanisms have been proposed to explain the incremental oil recovery by LSW injection.

1.2.1 LSW governing parameters

a. Carbonate rock properties

Carbonates are sedimentary rocks formed of minerals, such as calcite and dolomite. The petrophysical properties of carbonate reservoir, such as porosity, permeability, wettability, are complicated, so there are challenges in crude oil extraction from carbonate formations. The typical characteristics of carbonate rock are fractures with high permeability and low permeable matrix. The reservoir behavior prediction is very challenging due to these rock properties.

The wetting conditions of reservoirs play a main role in oil recovery processes. Most carbonates are observed to be neutral or oil wet (Legens et al., 1998). This wettability can be related to the retention of carboxylic group with negative charge of oil heavy compounds on the positively charged rock surface. Injection of ions and interaction between the injected active ions, which are called potential determining ions (PDIs), and rock surfaces may alter the initial wettability, resulting in the detachment of the oil and incremental oil recovery. PDIs are primarily sulfate, calcium and magnesium (SO_4^{2-}, Ca^{2+}, Mg^{2+}) ions that interact with the carbonate surface. Their presence in LSW is critical in a wettability alteration and a positive LSW effect in carbonates.

Interaction between the rock and ions in the contacted water is the main factor that affects the wettability of the rock. Arif and co-workers investigated the effect of mineral composition and surface roughness on the contact angle and wettability (Arif et al., 2019). The interaction between surface and ions as well as the activity of PDIs can be different for various types of carbonates (Awolayo et al., 2018b). For example, chalk is highly reactive compared to limestone rock, which is more heterogeneous (Strand et al., 2008). Hence, the rock composition and initial wettability are important parameters that affect the active mechanisms for improving oil recovery by LSW injection.

b. Oil acidity

It is proved that crude oil composition is essential for positive LSW effect (Austad et al., 2012; Ravari, 2011; Puntervold, 2008). The polarity of crude oil corresponds to the presence of S, N, O atoms in functional groups of acidic and basic organic molecules, e.g., asphaltenes and resins (Awolayo et al., 2018b). The acidic number (AN) is defined as the amount of KOH in mg required to neutralize 1 g of oil (Dubey et al., 1993). AN is a controlling parameter during LSW injection, as it determines the carboxylic group amount in the crude oil, which has a major influence on the carbonate wettability. It is found that with lower AN, the oil recovery is higher (Austad, et al., 2012). At lower AN, the attachment of carboxylic groups to the rock surface is weaker, which makes wettability alteration towards more water-wet easier during secondary watreflooding. Figure 1 compares oil recoveries for two cores saturated with high and low AN. In another study, the core was less water-wet for the oil with the higher AN (Ravari, 2011). Generally, in carbonates, at higher AN, the rock is more oil wet, which results in higher residual oil after waterflooding or after natural depletion. Hence, the performance of LSW is affected by the oil AN and should be considered as an influencing parameter.

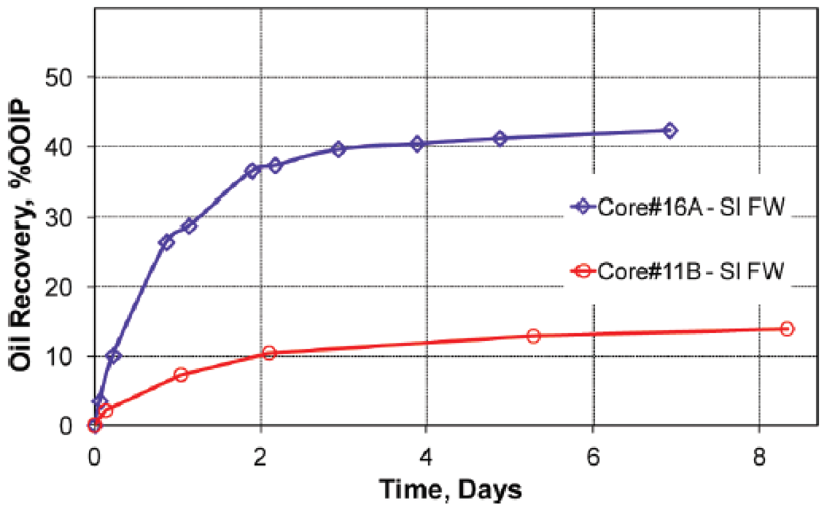

Figure 1.Effect of different AN (for Core #16A AN=0.08 and for Core #11B AN=0.7 mgKOH/g) on oil recovery from limestone cores at 110°C (Austad et al., 2012)

The effect of base number (BN), which is the quantity of the basic components in oil, is less than the effect of AN (Puntervold, 2008). Puntervold studied the effect of BN on wettability alteration and oil recovery in carbonate rocks. The results showed that higher amount of BN increases the water wetness, and thus, the oil recovery.

c. Injected water composition

Seawater contains active and inactive ions. Ions, such as SO_4^{2-}, Mg^{2+}, and Ca^{2+}, which can interact and affect the rock surface are active ones toward the carbonate surface (Zhang & Austad, 2006). Inactive ions, e.g., Na^+ and Cl^-, are the ions that are indifferent toward the rock surface (Awolayo et al., 2018b).

Concentration of active and inactive ions in injected water is essential to contribute to the LSW effect in carbonates (Yi & Sarma, 2012; Strand et al., 2008; Austad et al.,2012). Ca^{2+}, Mg^{2+}, and SO_4^{2-} are active toward the limestone surface, and their concentrations are important to activate LSW mechanisms and the LSW efficiency. The concentrations of inactive ions, such as Na^+ and Cl^-, are also critical in influencing different mechanisms which will be discussed later.

Yi and Sarma studied the effect of the amount of SO_4^{2-} on wettability change in limestone cores. They proved that sulfate in brine contributes to the wettability change (Yi & Sarma, 2012). Injection of the brine with high concentration of PDIs recovered more oil and changed wetting condition towards more water-wet (Figure 2).

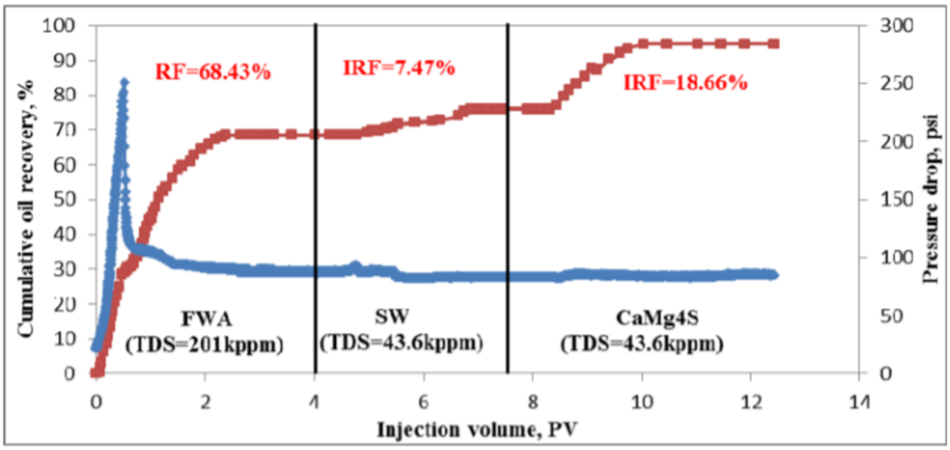

Figure 2. Effect of formation water (FW), seawater(SW) and brine with Ca^{2+}, Mg^{2+}, SO_4^{2-} on cumulative oil recovery and pressure difference (Yi & Sarma, 2012)

The affinities of divalent cations Ca^{2+} and Mg^{2+} were investigated by different researchers (Sekerbayeva et al., 2020; Bazhanova & Pourafshary, 2020; Awolayo et al., 2018a; Strand et al., 2008). At lower temperature, calcium showed more reactivity than $Mg^{2+,}$ but with higher temperature, Mg^{2+} got adsorbed more to the limestone surface and detached additional oil droplets from the surface (Awolayo et al., 2018a).

Despite successful cases where the presence of PDIs in seawater significantly improved the oil recovery, some researchers reported that the surface activity of limestone towards PDIs in seawater was low, and no incremental oil recovery was observed (Austad et al., 2012; Fathi et al., 2010).

The dilution of the injected brine reduces the amount of inactive ions close to the rock surface and enhances the interaction between active ions and rock. Na^+ and Cl^- do not interact with carbonate surface, but they prevent access of active ions to the surface to alter the wettability (Awolayo et al., 2018b). The reduction in salinity of injected brines plays a significant role in the oil recovery. Different studies proved this effect (Yi & Sarma,2012; Austad et al., 2012). As an example, in an experiment, the cores were flooded with 40 times diluted seawater after injection of seawater (Yi & Sarma, 2012). As can be seen from Figure 3, the diluted seawater achieved additionally almost 18% OOIP. Fani et al (2018) also investigated the importance of dilution and reported a similar behavior (Fani et al., 2018).

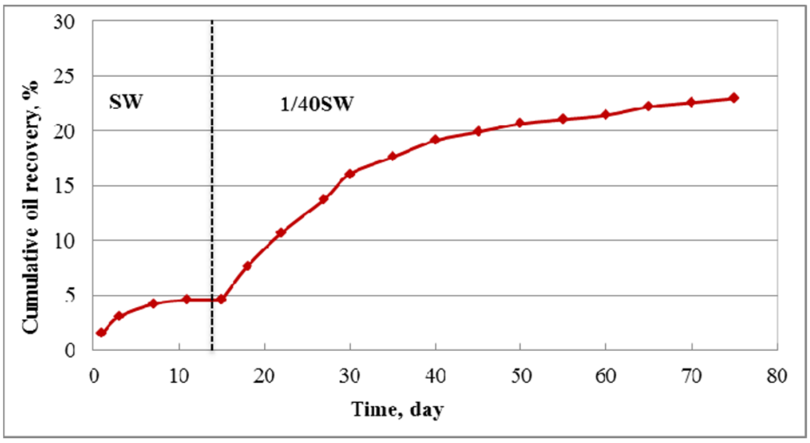

Figure 3. Effect of seawater (SW) and diluted seawater (1/40 SW) on cumulative oil recovery in limestone cores (Yi & Sarma, 2012)

Carbonate surface charge changes with the PDIs concentration in brine. Rock surfaces becomes positively charged when injected brine contains Ca^{2+} and Mg^{2+} and less anions, such as SO_4^{2-} (Dubey et al., 1993). Zeta potential determines the sign of the charge at the oil-brine or rock-brine interfaces (Dubey et al., 1993). The sign and the magnitude of zeta potential is controlled by the concentration of PDIs, temperature, pH and type of the rock (Mahani et al., 2017; Mahani et al., 2015; Jackson et al.,2016; Gomari et al., 2006).

Adsorption of SO_4^{2-} to rock surface changes it toward less positive. Divalent cations (Mg^{2+} and Ca^{2+}) interact with the carboxylic acid group of oil. This results in the release of negatively charged oil compounds from carbonate surface and wettability alteration towards a more water-wet state.

d. Temperature

The importance of temperature in LSW in limestone were investigated in different studies (Collini et al., 2020; Sekerbayeva et al., 2020; Tetteh et al., 2018; Austad et al., 2012). Alotaibi et al. (2010) observed that when formation brine was injected, contact angles changed from neutrally wet towards more a water-wet state as temperature increased from 122°F to 266°F. It was further reported that divalent cations affinity toward limestone cores was influenced by temperature change. Awolayo et al. (2018a) noted that temperature increase from 20°C to 130°C resulted in a significant increase in the adsorption of Mg^{2+} and the desorption of Ca^{2+}. The same tendency of substitution of calcium cation was demonstrated in the study of Puntervold (2008). According to Strand et al. (2008), the water wetness of a reservoir limestone flooded with seawater was improved at high temperature of 130°C.

However, a number of authors have recognized that injection of seawater at high temperature did not improve the oil recovery and ions affinity towards carbonate surfaces (Mahani et al., 2017; Fathi et al., 2010; Ravari, 2011). It was observed that injection of seawater at elevated temperature did not improve the water-wet fraction on the rock surface (Fathi et al., 2010). No incremental oil was recovered from outcrop limestone in imbibition experiments conducted by Ravari (2011) at 130°C. Overall, a few studies have examined a positive effect of temperature on wettability alteration and incremental oil recovery during LSW, whereas other studies reported an unclear dependency of LSW efficiency on temperature.

e. pH

Measurements from several studies have identified that the wettability alteration in carbonates involves the change in the effluent pH during LSW injection (Mohammadkhani et al., 2018; Gandomkar et al., 2015).

Mohammadkhani and co-workers reported the effluent pH rise in comparison to the injected water pH (Mohammadkhani et al., 2018). They also noticed that along with the pH change, Ca^{2+} concentration in the effluent brine increases. In another experiment, the effluent pH values were measured after high and low-salinity water injection stages (Gandomkar et al., 2015), indicating that the pH value is controlled by rock/fluid interactions.

The importance of the parameters described above has been observed in numerous studies. Some researchers observed the contribution of the parameters discussed above to the incremental oil recovery by LSW, others did not discover any effect on the oil recovery improvement. Hence, the exact relationships between the incremental oil recovery and the governing parameters are still not clear. In this work, we aimed to analyze the effect of controlling parameters on the LSW efficiency in carbonates.

1.2.2 EOR Mechanisms of LSW in Carbonates

A survey of the literature shows that based on the type of crude oil and the properties of reservoir and injection/formation brines, there are several EOR mechanisms of LSW interpretation proposed. Multicomponent ionic exchange (MIE), reduction in interfacial tension, expansion of electric double layer, and rock dissolution are the main mechanisms suggested by researchers to explain the incremental oil recovery by LSW in carbonates. Most of these mechanisms result in the wettability alteration of the carbonate rock which is the most desirable and widely accepted reason of improving oil recovery by LSW.

a. MIE

The mechanism of MIE occurs between the injected brine and the rock surface. The weakening of ionic bonds between oil compounds and carbonate surface is caused by exchange of ions (Austad et al., 2012; Al Kharousi et al., 2018). Sulfate ions adsorb onto the carbonate surface, lowering the charge of the carbonate surface. Then, cation Ca^{2+} is co-adsorbed by rock surface, and its excess reacts with carboxylic acid groups of polar oil components, which are originally bonded to the rock surface. So, the bonds between the polar oil components and rock surface are broken. It releases the oil in the form of Ca^{2+}-carboxylic compounds, and wettability is altered toward more water-wet state (Austad et al., 2012; Strand et al., 2008). To activate this mechanism, presence of PDIs is essential and this is a reason that PDIs concentration is considered as an influencing parameter during LSW flooding.

b. Reduction in IFT

Several researchers correlated the IFT reduction with the incremental oil recovery during LSW flooding (Meng et al., 2015; Alotaibi et al., 2010; Okasha et al., 2009). The interfacial tension (IFT) characterizes the capillary forces occurred between two liquid interfaces (Okasha et al., 2009). The wettability alteration was observed for these tests, where the interfacial tension reduced, from 17.4 mN/m to 14.2 mN/m (Meng et al., 2015). Increased temperature and dilution of high salinity brine can help reduce interfacial tension. Alotaibi et al (2010) observed an IFT reduction from 23.01 to 16.3 mN/m, when a temperature increases from 77 to 194°F.

However, there are contradictions reported in literature. Al-Attar and co-workers (2013) assessed the relationship between IFT and different concentrations of salt, Ca^{2+} and SO_4^{2-} at ambient conditions, and discovered no correlation between IFT and oil recovery.

c. Expansion of Electric Double Layer

It has been suggested that expansion of electric double layer is one of the mechanisms of LSW flooding in carbonates (Lingthelm et al., 2009; Awolayo et al., 2018a; Mahani et al., 2015; Al Mahrouqi et al., 2016). The layer of ions, which is formed on the rock-brine interface, together with the layer of opposite ions on the rock or fluid surface, is called the electric double layer (EDL) (Lingthelm et al., 2009). An electrical potential, also known as zeta-potential, is usually developed at the interfaces of rock/fluid and fluid/fluid pairs. The sign and the magnitude of the surface charges at rock-brine and oil-brine interfaces control the wetting condition of carbonate rock (Dubey et al., 1993). Lingthelm et al. investigated that the reduced concentration of divalent cations, Mg^{2+} and Ca^{2+}, in low saline water causes the predominance of repulsive forces and, hence, the expansion of the EDL (Lingthelm et al., 2009). Thus, it results in the desorption of oil components and the oil recovery improvement.

d. Rock dissolution

The release of adsorbed oil components occurs with the dissolution of minerals as a result of the physicochemical instability of the rock surface (Hiorth et al., 2008; Hiorth et al., 2010; Yousef et al., 2011). The dissolution affects wettability change towards water-wet and improves the oil recovery. Yousef and co-workers have noticed microscopic anhydrite dissolution triggered by injecting slugs of seawater with lower salinity (Yousef et al., 2011).

A discrepancy in the rock dissolution mechanism was found by Austad and others (Austad et al., 2009). They conducted experiments using LSW and concluded that injecting fluid with Ca^{2+} ion brought in higher oil recovery, which contradicts to the findings of previous researchers (Hiorth et al., 2008). Thus, no clear correlation between rock dissolution and oil recovery was found.

The following conclusions are drawn from the detailed examination of literature:

a) Different parameters, such as rock properties, oil acidity, injected water composition, temperature, and pH, are effective in LSW performance, but the relationship between them and oil recovery remains unclear;
b) The influence of parameters on the active mechanisms has not been clarified.

Hence, a comprehensive study is required to examine all oil displacement studies in the literature to answer to these questions. in this work the available data of oil displacement at the core scale are collected and the effect of different parameters on active mechanisms and the performance of LSW are analyzed.

1.2.3 Machine Learning

As we discussed, different parameters are effective to enhance the oil recovery during LSW injection. The behavior of LSW can be modeled as a function of these active parameters. By data analysis, it is possible to develop linear and nonlinear relationships between variables and recovery factor. Machine learning can be applied as a powerful tool to develop these models.

Linear regression analysis is the most common method to find the relationships between a variable or group of variables and output. Simple linear regression can be applied for prediction of response variable using a single variable. Multiple regression is used to explore how multiple variables explain the behavior of an output variable.

Simple and multiple linear regression approaches have been implemented for analyzing LSW flooding in sandstones (Wang & Fu, 2018). More than 200 experimental results of tertiary recovery processes in sandstones were collected from the literature and analyzed using linear regression methods. They concluded that multivariable linear regression demonstrated a stronger correlation between a set of active parameters and the incremental oil recovery in comparison to a single variable linear regression. Incremental oil recovery was observed to be positively related to base number, salinity, and clay content in sandstones (Wang & Fu, 2018). However, they observed that the strengths of linear relationships are classified as ‘moderate’, which are insufficient for prediction of LSW performance in sandstones.

Mohamed Ibrahim & Koederitz worked on over 400 groups of relative permeability data and aimed to develop relative permeability equations for oil-water systems (Mohamed & Kederitz, 2000). They used a forward stepwise multivariable method of linear regression, which is based on automatic searching for the best set of variables. Novel equations for relative permeability forecasting were found using this method. A good fit of their model to the collected data with was demonstrated (Mohamed & Kederitz, 2000).

Various nonlinear Machine learning regression methods are more accurate in data analysis, such as decision trees (DTs), artificial neural networks (ANNs), random forest (RF), support vector machines (SVMs)nonlinear. Models based on nonlinear regression try to find a connection between input and output variables assuming that the relationship between coefficients is not linear. Non-linear regression models uses Gaussian, power, exponential, logarithmic functions to fit the data.

Nonlinear models are used in different aspects of Exploration & Production (E&P) operations (Wang et al., 2020; Mahmoud et al., 2020; Abdulmalek et al., 2019; Schuetter et al., 2015; Alkinani et al., 2020; Venna et al., 2018). Wang and co-workers (2020) investigated the capability of three ML techniques, namely ANN, SVM and RF, as prediction models in estimating LSW effect in sandstones. These ML models developed were trained and tested using 178 tertiary LSW flooding data points, including total salinity, AN, BN, and clay content of sandstone (Wang et al., 2020). In their study, 1000 realizations were run, and the average correlation coefficient R was reported for all models. Some ML models achieved 'very strong' nonlinear relationships for the data set (Wang et al., 2020).

ANN is based on correlations between components which are also known as neurons in input and output layers. Models based on a net of neurons were used in fracture pressure predictions (Abdulmalek et al., 2019), relative permeability estimation (Kalam et al., 2020), prediction of the rate of penetration (Mahmoud et al., 2020), and evaluation of reservoir porosity (Al-AbdulJabbar et al., 2020). Abdulmalek (2019) collected about 4000 data points, and developed an ANN model to predict the pressure in fracture with a very good correlation and high accuracy. In another study, ANN was applied to predict the relative permeability to oil and water based on various parameters, such as porosity, absolute permeability, saturation, and wettability (Kalam et al., 2020). Using feed-forward neural networks, the porosity was predicted from the drilling parameters (Al-AbdulJabbar et al., 2020), which aimed to investigate the potential of ANN approach in real-time prediction of porosity while drilling. ANN-based model was also used to predict the rate of penetration by analyzing more than 3000 data points (AbdulJabbar et al., 2020).

DT technique of regression is based on a group of nodes called trees and leaves, which are predictor variables (Schuetter et al., 2015). DT is categorized as a non-parametric ML model. In a study of Schuetter (2015), this method is used to find the best combination of parameters for drilling process based on available surface data. DT model was also implemented in estimation of liquid holdup in two-phase flow (Almashan et al., 2020), and evaluation of pressure-volume-temperature (PVT) properties of oil systems (Almashan et al., 2019).

SVM is able to fit variables using nonlinear transformation equation to create a linear response of data (Schuetter et al., 2015). SVM-based model is also non-parametric. It can be used for regression and classification. In a regression, SVM finds the multidimensional hyperplane that fits the maximum number of data points, and the distance from this hyperplane to the nearest data point is maximized (Alkinani et al., 2020). Alhashem (2020) used SVM to predict multiphase flow regimes from gas and liquid properties and diameter of horizontal pipe. Also, SVM was applied in the lost circulation zones prediction (Alkinani et al., 2020) and phase classification problem (Venna et al., 2018).

To sum up, the linear and nonlinear methods of regression analysis are successfully used in different E&P operations. They predicted the performances of results based on the dataset of available parameters. Some of the researchers compared different modeling methods and discovered the best regression model for fitting the data. Such approaches can also be used to predict the performance of LSW in carbonates.

1.3 Problem Statement

Oil recovery by natural production in carbonates is usually lower than 30%. Hence, EOR methods are necessary to increase the amount of oil produced from carbonates. LSW injection is a promising EOR technique in improving oil recovery which has been studied by many researchers. LSW performance in carbonates has been investigated at the laboratory scale. The effect of important parameters, such as total salinity and composition of diluted water and oil acidity were studied. However, relatively small number of experimental studies on LSW injection in carbonates were conducted in contrast to the experiments in sandstones.

Detailed examination of previous studies on LSW injection reveals a number of shortcomings. The oil displacement response observed by different researchers were not in an agreement together, and there is no certainty in the relationship between controlling parameters and LSW performance in carbonates. Different mechanisms, namely MIE, rock dissolution, IFT reduction, and expansion of EDL, were suggested in prior studies. Also, there is no exact explanation of how the injection of diluted water can enhance oil recovery, and the governing mechanisms of LSW flooding are not clear. Thus, in this thesis, I collected available secondary and tertiary flooding data and analyzed the main parameters using data analysis approaches. The mechanisms were investigated by analyzing changes in different properties, such as zeta potential, IFT, effluent PDIs concentration, and others.

Nowadays, one of the powerful data analysis tools is Machine Learning (ML). ML methods have been implemented to predict the performance in different petroleum disciplines. Linear and nonlinear regression models have been successfully used in petroleum data analysis. The research to date on LSW flooding in carbonates has tended to focus on experiments, and no accurate data analysis of LSW performance in limestone exists. To understand the conditions for LSW to work and exact active mechanisms, machine learning models and statistical approaches were applied in this study.

1.4 Objectives of the thesis

The following objectives are deduced from the problem definition:

- to have a better understanding of oil displacement during LSW flooding in carbonates by analyzing coreflooding tests
- to study the dependence of incremental oil recovery on oil-rock-fluid parameters using data analysis approaches
- to determine the effect of different parameters on the active mechanisms and to find out the dominant active mechanisms during LSW injection in carbonates.

1.5 Thesis structure

The thesis is organized as follows: Introduction, Methodology, Results and Discussion, Conclusion and Recommendations. Chapter 1 first reviews the literatures that include the experimental studies on LSW injection in carbonates and lists governing parameters in LSW performance. Then, the EOR mechanisms proposed by researchers are discusses. At the end, linear and nonlinear models are briefly introduced, and different examples of their application in petroleum disciplines are provided. The methods used to analyze the data are described in Chapter 2. This chapter explains data collection process, and Machine learning and statistical methods. The obtained results are analyzed and discussed in Chapter 3. Finally, main conclusions are drawn and recommendations are suggested for future research in Chapter 4.

CHAPTER 2: METHODOLOGY

Different parameters are effective in improving oil recovery by LSW flooding. However, the exact effect of controlling parameters on LSW performance in carbonates is still not clear. The active mechanisms which explain the positive effect of LSW injection on oil recovery enhancement are difficult to determine. Thus, to investigate the mechanisms and conditions for LSW to work, the data from available coreflooding experiments are collected and analyzed using data analysis approaches.

2.1 Data collection and cleaning

Experimental studies of LSW flooding in carbonates were carefully studied and relevant oil displacement tests were extracted. Fluid/rock properties and experimental results were collected from the most recent papers shown in Table 1. Each data entry corresponds to a coreflooding test. Both secondary and tertiary modes of LSW flooding in carbonates were considered in the data extraction process. The collection of data was conducted in an unbiased manner from tables and graphs in available studies. The data from 145 core flooding tests were extracted and compiled.

The laboratory experiments of oil displacement tests by LSW injection in limestone cores were categorized to extract information about the injection mode, injection sequences, and the main parameters, which are considered to affect the oil recovery.

Rock/fluid properties controlling the performance of LSW flooding are shown in Table 2, which shows the number of available data points for each controlling parameter. As not all parameters were reported in every LSW flooding experiment, there is a significant number of missing data for some parameters, such as crude oil base number (BN), residual oil saturation (S_{or}), pH, interfacial tension (IFT), and effluent concentrations of ions. This affects the accuracy of our models in some cases due to the lack of data. Table 3 shows the minimum, maximum and mean values of main parameters.

Table 1. Number of coreflooding data collected from papers

#	Paper	Number of data points
1	Samanova, 2021	2
2	Collini et al., 2020	7
3	Li et al., 2020	2
4	Sekerbayeva et al., 2020	1
5	Feldmann et al., 2020	3

6	Tetteh et al., 2019	6
7	Tetteh et al., 2018	4
8	Mohammadkhani et al., 2018	5
9	Nasralla et al., 2018	20
10	Chandrasekhar et al., 2018	2
11	Amiri et al., 2018	9
12	Mirchi et al., 2018	2
13	Tetteh et al., 2017	2
14	Jalilian et al., 2017	2
15	Mohsenzadeh et al., 2016	4
16	Chandrasekhar et al., 2016	6
17	Awolayo et al., 2016	3
18	Jackson et al., 2016	2
19	Gandomkar et al., 2015	3
20	Shehata et al., 2014	4
21	Nasralla et al., 2014	4
22	Awolayo et al., 2014	3
23	Al-Harrasi et al., 2012	3
24	Austad et al., 2012	5
25	Romanuka et al., 2012	16
26	Winoto et al., 2012	3
27	Vo et al., 2012	4
28	Yi & Sarma, 2012	6
29	Yousef et al., 2012	2
30	Gupta et al., 2011	5
31	Yousef et al., 2011	2
32	Alotaibi et al., 2010	2
33	Austad et al., 2008	1

Table 2. Number of data points extracted from literature for parameters

Parameter	**Number of data points**	
	Secondary mode	**Tertiary mode**
Porosity, %	20	112
Permeability , mD	28	117
Initial water saturation Swi, %	23	116
Formation water composition, ppm	26	117
Formation water salinity, ppm	24	114
Secondary injected brine composition, ppm	25	114
Secondary injected brine salinity, ppm	25	116

Tertiary injected brine composition, ppm	-	112
Tertiary injected brine salinity, ppm	-	114
Crude oil acid number, mgKOH/g	7	87
Crude oil base number, mgKOH/g	1	64
Viscosity of oil, cp	16	106
Density of oil, cp	25	98
Residual oil saturation S_{or}, %	7	37
pH of effluent brine	0	27
Test temperature, °C	8	100
Secondary recovery factor, %OOIP	28	117
Tertiary recovery factor, %OOIP	-	117
IFT, mN/m	5	25
Contact angle	2	27
Effluent cations concentration, ppm	3	20
Effluent SO_4^{2-} concentration, ppm	1	9
Pressure drop, psi	2	60
Zeta potential	9	18

Table 3. Statistical measures of the parameters

Parameter	**Min**	**Max**	**Mean**
Permeability, mD	0.4	200.6	32,7
Low salinity, ppm	0	193230	22315
SO_4^{2-} concentration (LS), ppm	0	9222	930
Cations concentration (LS), ppm	0	13454.5	1416
SO_4^{2-} concentration (HS), ppm	0	4290	534
Cations concentration (HS), ppm	14.34	61480	15879
AN, mgKOH/g	0.08	4.6	0.57
BN, mgKOH/g	0.01	2.49	0.5
Temperature, °C	20	250	88

RF is collected for both secondary and tertiary injection modes to analyze the effect of controlling parameters on LSW injection. There are 28 data points reporting secondary recovery factors, ranging within [7%-85%] of OOIP (original oil in place), with the mean value of 45%, and standard deviation of 23.1%. The probability of the secondary recovery factors are shown in Figure 4.

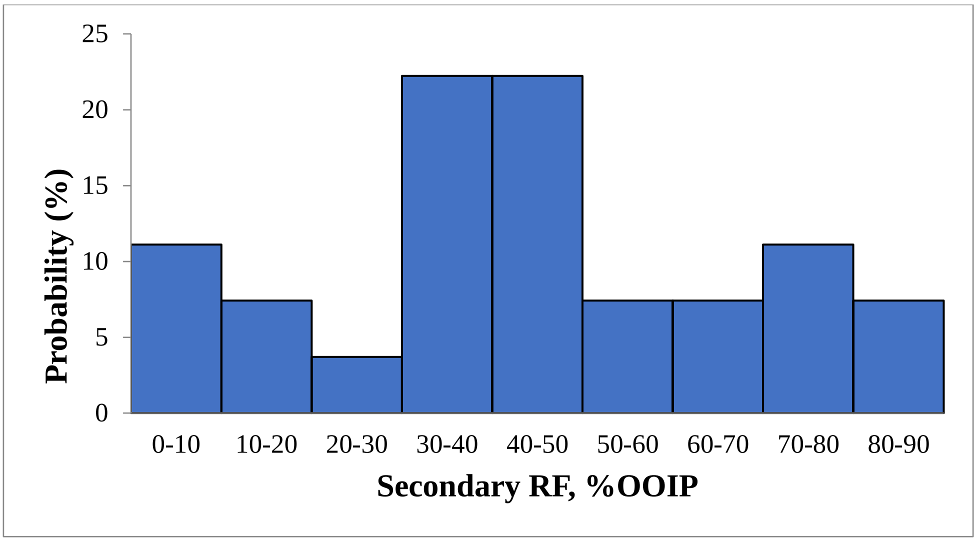

Figure 4. Probability of incremental recovery factor after the secondary stage injection, %OOIP

There are 117 data which shows the incremental oil recovery by injection of LSW in the tertiary mode. These data points range within [0-42%] of OOIP (original oil in place), mean 6.17%, and standard deviation of 7.6%. Figure 5 shows the distribution of incremental oil recovery by tertiary recovery.

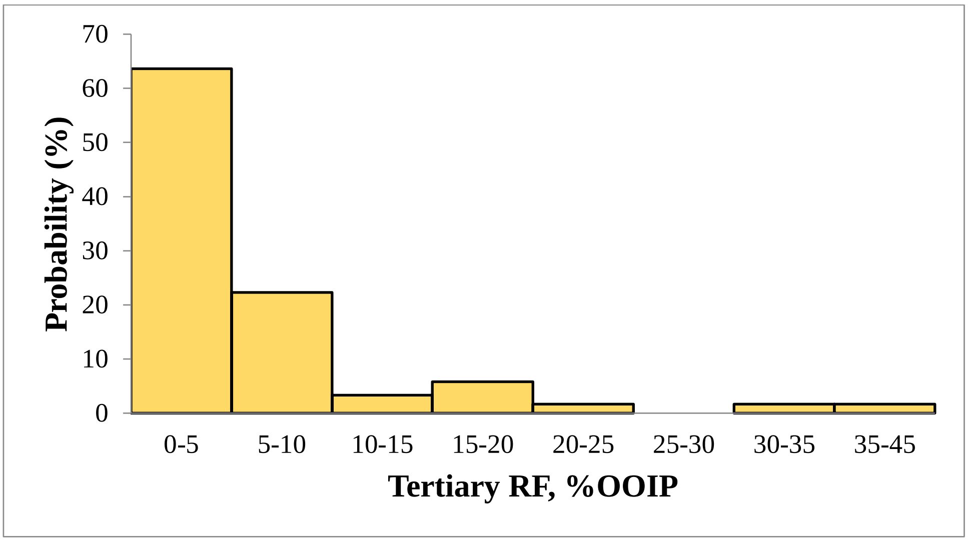

Figure 5. Probability of incremental recovery factor achieved by the tertiary stage injection, %OOIP

LSW flooding, as an EOR approach, is applied at the tertiary stage. The low number of secondary core flooding tests in the literature proves the more importance of the application of LSW in tertiary. Hence, this study focused more on analyzing data collected from tertiary core flooding experiments.

Collected data points were organized and prepared for regression analysis. Different units for parameters, such as compositions of brines, total salinities, temperature, pressure drop, were reported in the literature. At this stage, all data were converted to a unified unit system.

Data points were measured at different experimental conditions. Fluid and rock properties were also different. To make comparative analyses, different dimensionless numbers were developed to scale controlling parameters while preserving their physical significance. For example, for ions composition, the relative change of the concentration was calculated and used in the modeling process. Equations 1-3 show dimensionless numbers describing the alteration in the sulfate and cations concentration, and total dissolved solids. Equation 4 shows the dimensionless acid number (AN), and Equation 5 defines the dimensionless oil recovery. The conversion into dimensionless parameters reduced the number of data points.

$$SO_4{}^{2-}(d) = \frac{SO_4{}^{2-}(HS) - SO_4{}^{2-}(LS)}{TDS(HS)} \quad , \tag{1}$$

$$Cations(d) = \frac{Cations(HS) - Cations(LS)}{TDS(HS)} \quad , \tag{2}$$

$$TDS(d) = \frac{TDS(HS) - TDS(LS)}{TDS(HS)} \quad , \tag{3}$$

$$AN(d) = \frac{AN}{BN} , \tag{4}$$

$$\text{and } \Delta RF_3(d) = \frac{RF_3 - RF_2}{100 - S_{wi} - RF_2} , \tag{5}$$

where RF_3 is the recovery factor after tertiary flooding (% OOIP), RF_2 is the recovery factor after secondary flooding (%OOIP), and S_{wi} is the initial water saturation (%).

2.2 Data analysis methods

As mentioned earlier, different parameters are effective in potentially enhancing the oil recovery during LSW injection in carbonates. LSW performance in carbonates can be predicted based on these controlling parameters. Using Machine Learning techniques, linear and nonlinear relationships between parameters and recovery factor can be developed. In this section, predefined modules in MATLAB were applied to analyze LSW controlling parameters and study the effect of them on the oil displacement. Different mechanisms are proposed by researchers to support the hypotheses behind the incremental oil recovery by LSW in carbonates, but the effect of main fluid/rock parameters on the mechanisms has not been clarified.

2.2.1 *Machine Learning methods*

Machine learning methods were used to analyze the effect of single and multiple controlling parameters on the incremental oil recovery by LSW. Linear and nonlinear correlations were developed nonlinear between different independent variables such as dimensionless rock/fluid properties and oil recovery factor as the dependent parameter. The correlation coefficients were estimated to determine the strength of dependence between variables and recovery factor.

Linear regression method

Simple and multivariable linear regression models were applied to analyze the data. Simple linear regression is used to estimate the strength of influence of individual independent parameters on the dependent parameter. It assumes that the relationship between variables is linear. After preliminary analysis, acid number, base number, total salinity of low and high saline brines, potential determining ions concentration, permeability, and temperature were selected as the governing independent parameters for regression. Linear correlations were developed by the least squares method, which minimizes the summed squares of the vertical separation between the actual values and the predicted values from regression of independent variable.

As shown in Figure 6, simple linear regression determines the coefficients β_0 and β_1 for the estimation of the linear relationship between parameters as

$$y_i = \beta_0 + \beta_1 x_i + \varepsilon_i \tag{6}$$

Where y_i – the ith observation of the dependent variable, β_0- the intercept, β_1 – the slope of the line, x_i – the ith observation of the independent variable, ε_i – the error.

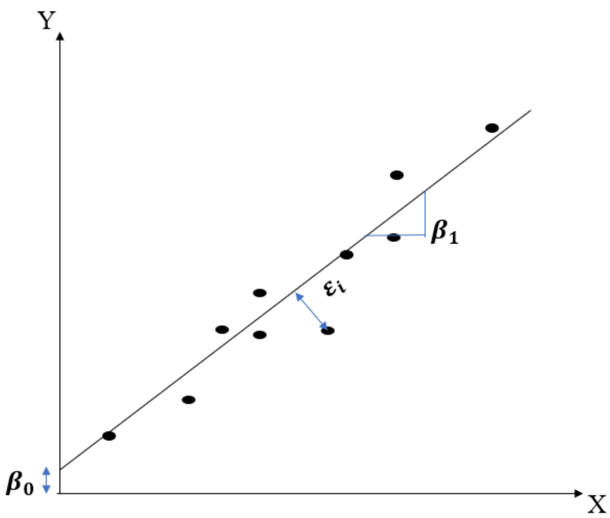

Figure 6. Parameters of a simple linear regression

Multivariable linear regression models are used to study the correlation among a group of independent variables and a dependent variable. Two groups of parameters were investigated. Group 1 is the brine parameters consisting of SO_4^{2-}, Mg^{2+} and Ca^{2+} concentration and injection brine salinity. Group 2 is the oil/brine parameters consisting of the group 1 parameters and the dimensionless AN. The predicted RF values are calculated based on the estimated regression coefficients using independent variables.

Nonlinear regression methods are more accurate in data analysis, because they assume that the relationship between coefficients is not linear which is more realistic in many cases. Machine learning algorithms, such as decision trees (DTs), artificial neural networks (ANNs), and support vector machines (SVMs) were applied to assess the contribution of different parameters to the LSW effect. For ML models, data points were randomly divided into 70% for training and 30% for testing. In our work, 5000 realizations were run for nonlinear regression for each type of ML models.

Artificial Neural Networks

ANN is based on connections between components, which are also known as neurons in input, hidden, and output layers. Variables called weight are assigned to the connections to represent the contribution of the input variables to output. After determination of the parameters between input and hidden layers, ANN finds the nonlinear relationship between input and output variables (Mitchell, 1997). In this work, feedforward neural network were used to analyze the relationship between the input to output layers (Babuska, 2010).

Each input data point x_i is multiplied by a weight factor, w_i (Figure 7). Then the sum of the weighted inputs is passed through a nonlinear function to generate an output (Babuska, 2010).

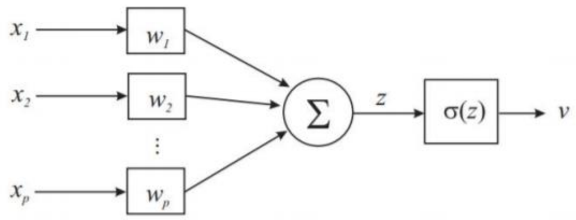

Figure 7. Artificial Neural Networks (Babuska, 2010)

Mathematically, the transformation of inputs is described by (Babuska, 2010):

$$z_j = \sum_{i=1}^{n} w_{ij}{}^t x_i + b_j{}^t, j = 1,2,..t \quad (7)$$

where b is bias, w_i is weight factor.

After computation of z_j, the outputs of the hidden layer, v_j, are calculated.

$$v_j = \sigma(z_j), j = 1,2,..t \tag{8}$$

Finally, the neurons of output layer are computed.

$$y_l = \sum_{j=1}^{t} w_{jl}{}^{n} x_i + b_l{}^{n}, l = 1,2,..n \tag{9}$$

where $b_l{}^n$ is the output layer bias, and $w_{jl}{}^n$ is the output layer weight.

The structure of ANN model was chosen based on the sensitivity analysis (Table 4). Oil/brine parameters based on 500 data entries were used in the analysis. In the ANN used here, 1 hidden layer with 4 neurons was chosen, as shown in Figure 8.

Table 4. Sensitivity analysis for ANN

#	Number of layers	Number of neurons	Results	
			Training R	Testing R
1	1	2	0,162	0,034
2	1	4	0,204	0,044
3	2	2	0,150	0,017

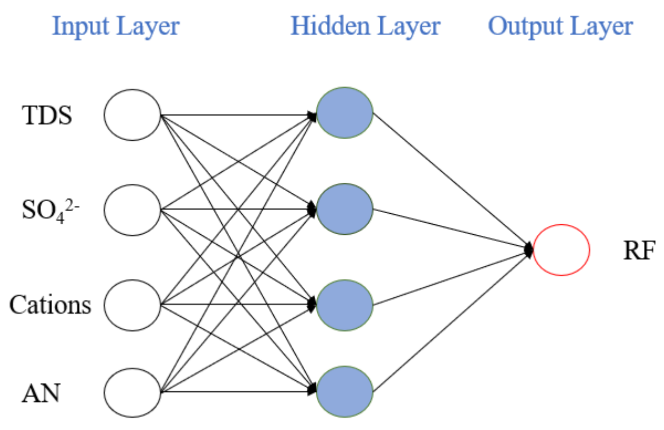

Figure 8. ANN-based model topology for regressing RF to oil/brine parameters

Decision Tree

DT method is a supervised Machine Learning algorithm. As a non-parametric ML model, it is widely used in solving classification and regression problems. DT algorithm is based on a group of nodes called root, decision, and leaf nodes (Kitts, 1999). The objective of this ML method is to build a model that predict the class or the value of variable by making simple decisions. This study used the Regression Decision Tree, each leaf of which represents a numeric value for important independent variables (Kitts, 1999).

Figure 9 shows the organization of a typical DT and connections between nodes. Root node represents a variable which splits into two or more sub-nodes. Sub-nodes that do not split are leaf nodes. Each split in DT is associated with a discriminant made on a particular dimension. The goodness of splitting is determined by variance. If the variance is lower, classification is good, and vice versa (Kitts, 1999).

One problem of DT is overfitting which occurs due to noise in the training data. To eliminate the overfitting, pruning algorithm can be implemented in DT to remove the least reliable nodes or branches (Kitts, 1999).

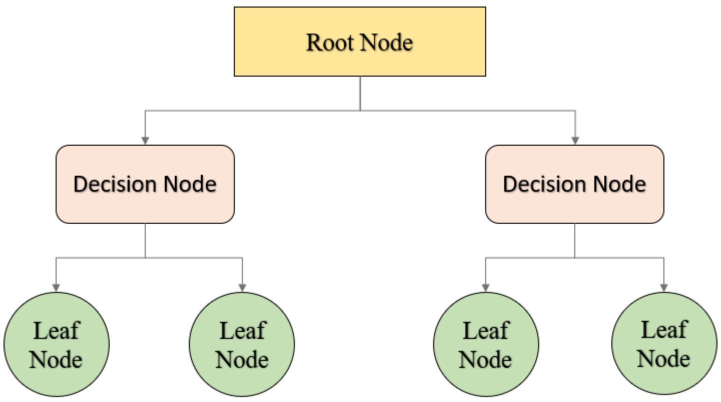

Figure 9. Decision Tree structure

Support Vector Machine

SVM model is also a kind of supervised learning methods that are used in handling regression and classification problems (Schuetter et al., 2015). This model can fit variables using a nonlinear transformation equation to predict response of predictor data.

We have a training data set $\{(x_1, y_1),..(x_n, y_n)\}$, and the input data $X \in R$. In our case, these input data are LSW governing parameters. A decision boundary is created to separate the LSW parameters by maximizing the distance or margin ε from supporting data points (Vapnik, 1995). If the data set is not linearly separable, SVM creates higher-dimensional space, where data can be separated by hyperplanes (Figure 10), which would be used for the prediction of output. The data points closest to the hyperplane are called support vectors (Vapnik, 1995).

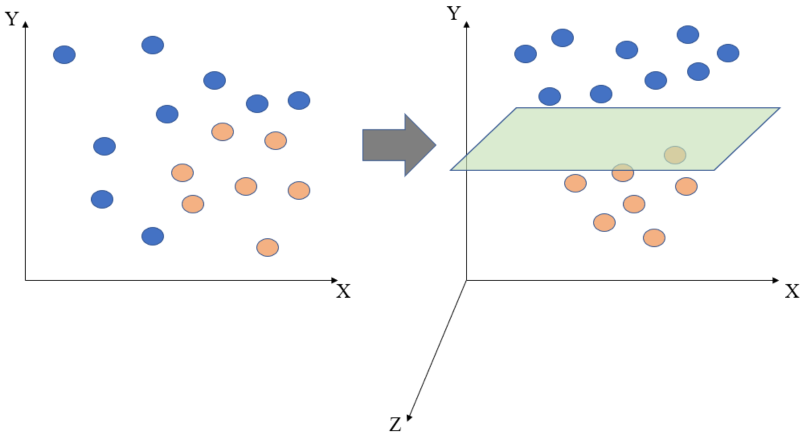

Figure 10. SVM higher-dimensional hyperplane for nonlinear problems

All deviations from the margin ε are represented by the distance ξ_i, a slack variable. The objective of SVM can be described by the following equations (Vapnik, 1995):

$$MIN\,\frac{1}{2}\|\omega\|^2 + C\sum_{i=1}^{n}|\xi_i|\,, \tag{10}$$

$$\text{and } |y_i - \omega_i x_i| \leq \varepsilon + |\xi_i|\,, \tag{11}$$

where MIN is a minimization, C is a constant (C>0) and ω is a vector variable.

The purpose of Equation 10 is to minimize the distance from deviated data points to decision boundaries. Equation 11 describes the constraints from both sides of a hyperplane. Hyperplanes are illustrated in Figure 10.

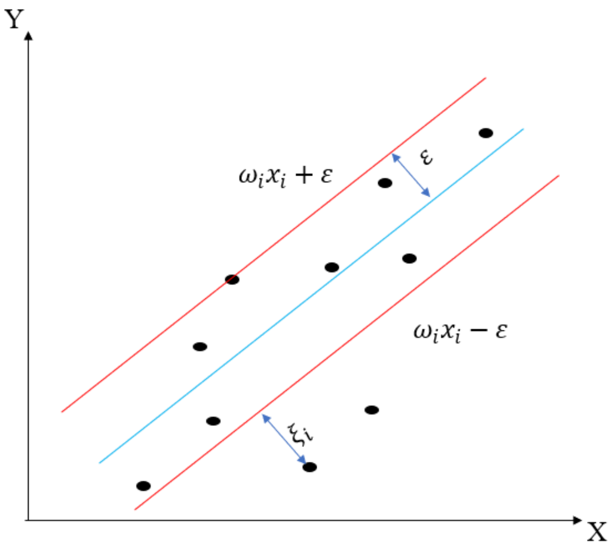

Figure 11. Support Vector Machine

The transformation of inputs to another space is enabled using a Kernel function, K, which helps to create a hyperplane without increasing the computational cost (Alkinani et al., 2020). There are different types of Kernel functions. We used a Gaussian Kernel function:

$$K\ (x_i, x_j) = e^{-\frac{1}{2\sigma^2}\|x_i - x_j\|^2}\ , \tag{8}$$

where x_i, x_j are input data points, and σ is a kernel width parameter. As the value of σ decreases, the SVM model overfits.

P-value, mean relative error (MRE) and coefficients of correlation and determination, R and R^2, respectively, were calculated for each regression model. P-value represents the probability that the null hypothesis is true, and it shows if the change in the model or random circumstances are the causes of the desired result. A low p-value is preferable for regression models.

MRE is the ratio of the absolute error of a measurement to the measurement taken. It is described by:

$$MRE = \frac{Predicted\ RF - Actual\ RF}{Actual\ RF} \frac{1}{N} * 100\%, \tag{9}$$

Coefficient of determination, R^2, is the squared correlation coefficient, R. The values of the coefficients vary from 0 to 1, representing no linear relationship and good linear relationship, respectively. R^2 is the percentage of variation in the dependent variable explained by the input independent variables. It is calculated by:

$$R^2 = 1 - \frac{\sum_{i=1}^{N}(RF_i - \widehat{RF_i})^2}{\sum_{i=1}^{N}(RF_i - \overline{RF_i})^2}\ , \tag{10}$$

where y_i is the i^th^ observation of the dependent variable y, RF_i is the i^th^ observation of RF, $\widehat{RF_i}$- the predicted RF, $\overline{RF_i}$ is the mean of RF.

When the number of variables increases, the coefficient of determination usually increases even with the same data set. The adjusted R^2 is used to minimize the impact of the number of variables. It is calculated by:

$$Adjusted\ R^2 = 1 - \frac{N-1}{N-k}(1 - R^2)\ , \tag{11}$$

where N is the number of data points, k is the number of independent variables, and R^2 is coefficient of determination.

Each of the coefficients shows the strength of the relationship between input parameters and the output parameter (RF). The qualitative interpretation of the relationship strength based on the correlation coefficient is represented in Table 5 (Wang & Fu, 2018).

Table 5. The strength of relationship between parameters and coefficient of correlation (Wang & Fu, 2018)

Absolute value of correlation coefficient R	**Strength of relationship**
[0.7, 1.0]	Very strong
[0.4,0.7)	Strong
[0.3,0.4)	Moderate
[0.2,0.3)	Weak
[0.01, 0.2)	Negligible
0	No correlation

2.2.2 Statistical methods

The active mechanisms which explain the positive effect of LSW injection on oil recovery enhancement are difficult to establish. To study the relationships between proposed mechanisms and conditions for LSW to work, data collected from available coreflooding tests were statistically analyzed.

Mechanisms such as MIE, rock dissolution, IFT reduction, EDL expansion, and micro-dispersions were evaluated using these methods. These mechanisms are suggested as governing mechanisms of LSW in the literature. In this work, by statistical analysis of controlling parameters, such as PDIs effluent concentration, wettability pressure drop, IFT, pH of effluent brine, and zeta potential, we proved the occurrence of these mechanisms (Figure 12). For example, analysis of the concentrations of PDIs (Ca^{2+}, Mg^{2+} and SO_4^{2-} ions) in the effluent can explain possible ion-exchange and rock dissolution mechanisms. In our study this parameter was extracted and analyzed using statistical methods. In another example, alteration in zeta potential values is an indicator to show the electric double layer expansion mechanism.

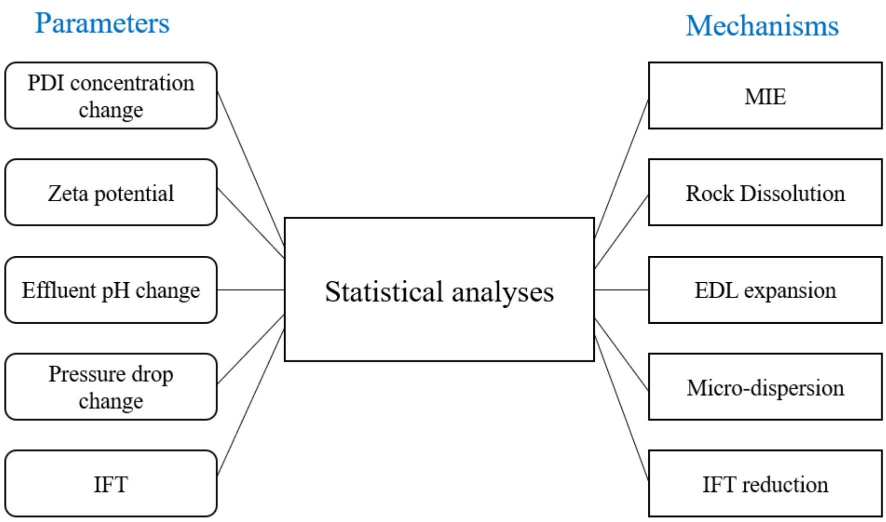

Figure 12. Schematic illustration of the statistical analyses of controlling parameters and mechanisms

CHAPTER 3: RESULTS AND DISCUSSION

Understanding the active mechanisms involved in improving the performance of LSW in carbonate formations is still challenging. The effect of controlling parameters on the incremental oil recovery by LSW injection has been examined experimentally in prior studies (Sekerbayeva et al., 2020; Bazhanova & Pourafshary, 2020; Collini et al., 2020; Tetteh et al., 2018; Awolayo et al., 2018a;). However, some cases in the literature showed no clear evidence of the contribution of main parameters to the LSW effect. The exact relationships between main parameters and additional recovery factors are important questions to be addressed. Using data analysis approaches, we studied different rock/fluid properties to seek more effective parameters. We also analyzed the effect of the rock, fluid, and crude oil properties individually and together on the incremental oil recovery achieved by LSW flooding. In this chapter the findings by data analysis methods from clarifying effective parameters and active mechanisms during LSW flooding are presented.

3.1 Effect of LSW governing parameters on oil recovery

LSW is recognized as effective to increase the oil recovery from carbonate formations, whereby the equilibrium between fluids and rock is disturbed. Injection of LSW affects the interaction between rock and fluids and alters parameters such as wettability and surface charges. Different rock/oil/brine properties are involved in this procedure. Although the dependence of LSW performance on single controlling parameters in carbonates has been proved in different ex

perimental studies, there are some contradictions reported in the literature. For example, the importance of PDIs in LSW performance were investigated by a number of researchers

(Sekerbayeva et al., 2020; Bazhanova & Pourafshary, 2020; Awolayo et al., 2018a). However, some studies reported that the presence of PDIs in injected brine did not guarantee incremental oil recovery by LSW (Austad et al, 2012; Fathi et al., 2010).

To study the effect of single rock/fluid/oil properties on the incremental oil recovery by LSW, data analysis approaches were applied to analyze the collected data. Based on the literature and different experiments observed, a group of parameters have been selected as candidates of controlling parameters. The parameter values from 149 core flooding tests were extracted and compiled. The main oil/brine/rock parameters that were analyzed are listed in Table 6. Among the carbonate rock parameters that were analyzed individually, it is found that permeability is the most influential one. Hence, in this study, permeability of the porous media is considered. For brine properties, studies showed that the concentration of PDI and salinities of injected brines are correlated with the incremental recovery factors (RF). Among crude oil parameters that were considered, acid number (AN) and base number (BN) are the most effective ones which were selected for data analysis. Another main operational parameter used in data analysis is the temperature of LSW flooding tests.

Table 6. List of rock/brine/oil parameters

Parameters		**Number of data points**
Rock	Permeability	122
Brine	Low salinity brine	117
	Salinity change	117
Oil	Cations	110
	SO_4^{2-}	107
	AN	80
	BN	60
	Temperature	98

Using the single-variable linear regression model, we first investigated the effect of individual parameters on the incremental oil recovery of LSW in core flooding.

3.1.1 Rock properties

The importance of carbonate rock parameters has been investigated in different studies (Arif et al., 2019; Strand et al., 2008). We decided to analyze the effect of rock permeability, as the most influential one, on the incremental oil recovery by LSW injection. Permeability values of carbonate rock samples used in coreflooding tests were collected. The incremental oil recovery factors during the tertiary recovery period were linearly regressed against the permeability based on 118 data points. The obtained correlation coefficient is 0.1721, which indicates negligible strength of the relationship between permeability and recovery factor. Figure 13 scatters the incremental recovery factors after tertiary flooding mode against permeability. For different ranges of permeability, average RF was calculated (Figure 14). RF achieved at given permeability varies, but the incremental RF is slightly higher for core samples with low permeability. After the secondary waterflooding, residual oil saturation S_{or} is relatively high in low permeable cores. So, LSW flooding could possibly recover more oil.

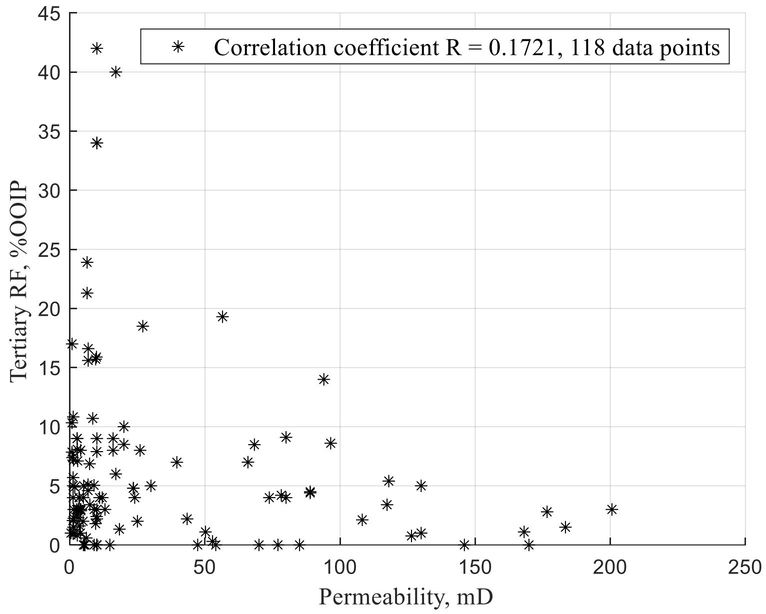

Figure 13. Incremental RF after tertiary flooding vs permeability

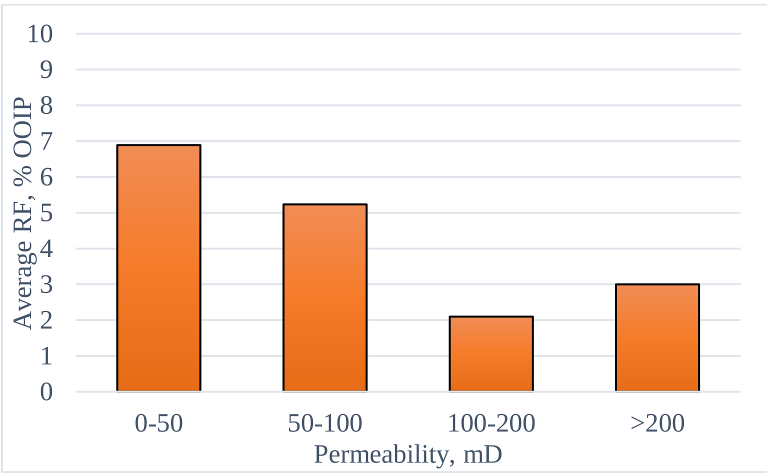

Figure 14. Average RF vs permeability

3.1.2 Brine properties

LSW is generally used as an EOR method after high saline water flooding. The change in salinity and ion composition of the injected brine affects the tertiary oil recovery. There are many experimental studies performed to investigate the contribution of low salinity brine composition to the enhanced oil recovery (Yi & Sarma, 2012). Ion composition and specially concentration of active ions, such as Mg^{2+}, Ca^{2+}, and SO_4^{2-} , and total salinity of injected LSW and its contrast with the high saline water applied during secondary waterflooding, are reported to affect the performance of LSW in carbonates. Using linear regression, we analyzed the relationship between these parameters and the incremental oil recovery.

Different dimensionless numbers for brine parameters were developed to preserve the physical significance of these parameters. Equations 1-3 that are described in the previous chapter were used for calculating of the relative change of PDIs and salinity.

Salinity of injected low saline and high saline brines

Total salinities of injected low saline brines were linearly regressed against the incremental RF achieved after tertiary flooding. The total number of data entries that reported the total salinity of low saline brines is 121. Negligible correlation between absolute values of low salinity and RF is established with a correlation coefficient of 0.1059. Figure 15 shows that the linear regression of the recovery factor against total salinity of low saline brine.

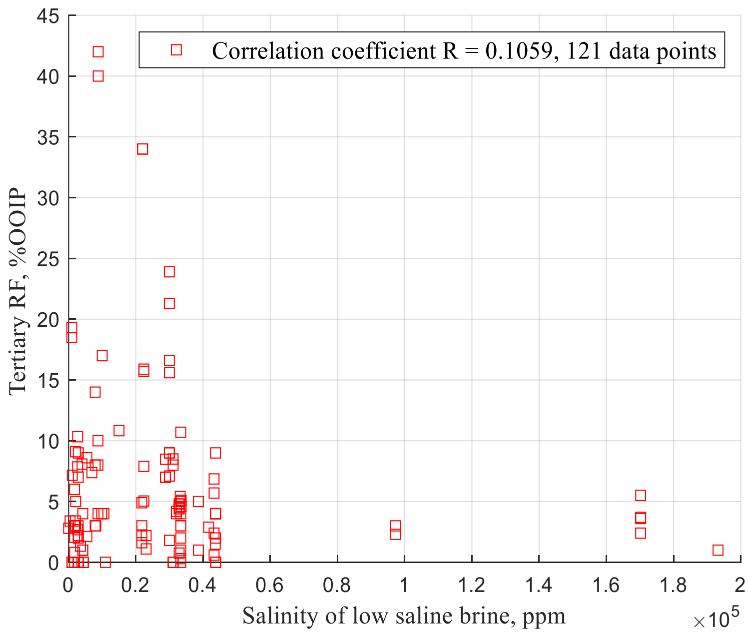

Figure 15. Incremental RF vs total salinity of low saline brine

To investigate whether the change in total salinity of injected brines can affect the LSW performance, we plotted the RF as a function of the dimensionless salinity values and obtained the correlation coefficient. The change in salinity does not contribute to oil recovery enhancement individually as the correlation coefficient (0.046) is even lower than the coefficients for absolute values of low salinity. Figure 16 illustrates the linear regression of salinity change and tertiary RF.

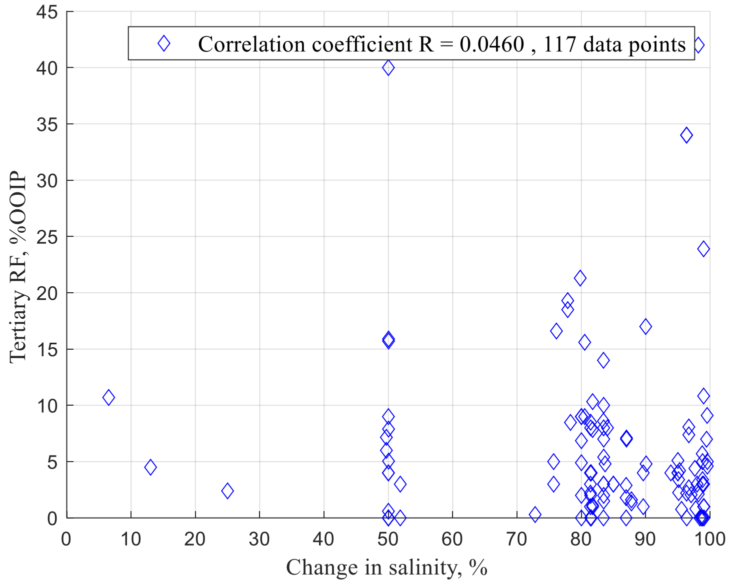

Figure 16. Incremental RF vs change in salinity

PDIs concentration

The concentration of potential determining ions, such as Mg^{2+}, Ca^{2+} and SO_4^{2-}, is one of the necessary conditions for LSW flooding to take effect (Sekerbayeva et al., 2020; Bazhanova & Pourafshary, 2020; Awolayo et al., 2018a). However, there is no clear relationship between active ions concentration and oil recovery improvement by LSW reported in the literature. To verify the effect of these parameters on RF, we analyzed the relative change in ions concentration in injected high and low saline water using a linear regression model.

Regression incremental recovery factor after tertiary flooding against change in concentration of cations, Mg^{2+} and Ca^{2+} is based on 110 data entries (Figure 17). According to Table 2, the obtained correlation coefficient, 0.0299, corresponds to negligible strength of the relationship between this parameter and RF.

The effect of the difference in SO_4^{2-} concentration in low and high saline brines on LSW performance was also analyzed by linear regression. Figure 18 shows negligible strength of correlation between these two parameters. The correlation coefficient is 0.1593 for 107 data points.

Thus, individual cations and SO_4^{2-} concentration does not contribute a positive LSW effect in carbonates.

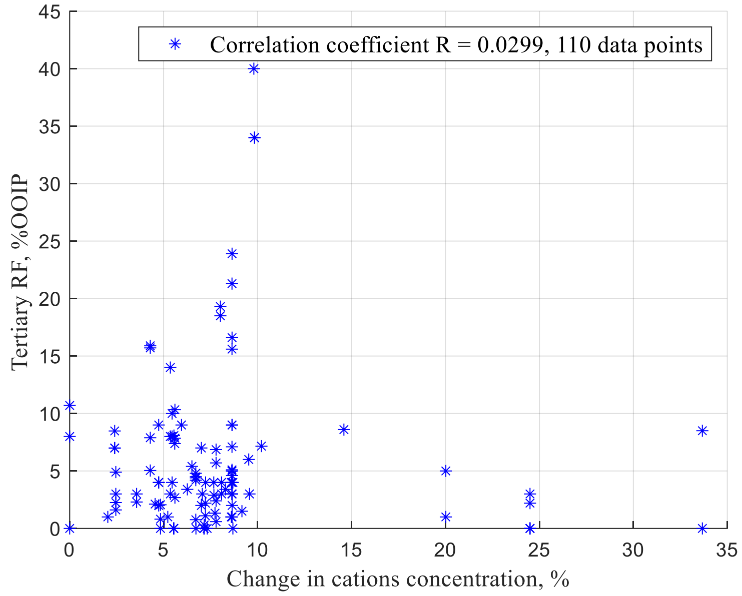

Figure 17. Incremental RF vs change in cations concentration

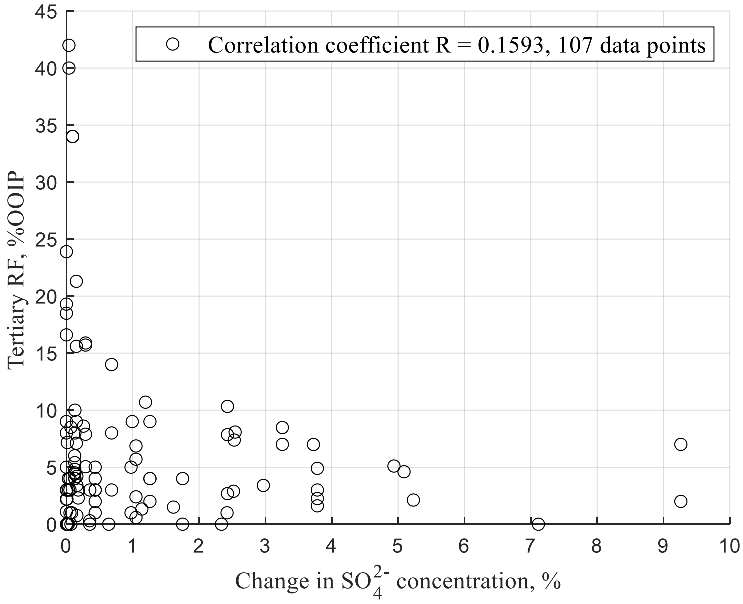

Figure 18. Incremental RF vs change in SO_4^{2-} concentration

3.1.3 Oil properties

The contribution of oil parameters, such as the acid number (AN) and base number (BN), to the EOR potential of LSW flooding in carbonates, has been identified in prior studies (Austad et al., 2012; Ravari, 2011). It is believed that at higher AN, there is a higher chance that LSW can significantly alter the wettability to a more water-wet state, thus more oil could be recovered by LSW in carbonates (Yi & Sarma, 2012). The effect of base number (BN), which is the quantity of the basic components in oil, is less significant than the effect of AN (Puntervold, 2008).

To observe the effect of crude oil components, we analyzed the effect of AN and BN on oil recovery based on 80 and 60 data points, respectively. The relationship between the acid number and incremental oil recovery was found to be negligible, as the correlation coefficient is 0.1848 (Figure 19). For various ranges of AN, we calculated the average RF (Figure 20). It is clear, that at higher AN (more than 1.5 mgKOH/g) LSW can better improve the oil recovery. Higher AN means more oil adheres to the rock surface at initial condition. And after waterflooding, more oil remains in the core. So, there is a higher chance for LSW flooding to alter the wettability and recover more oil.

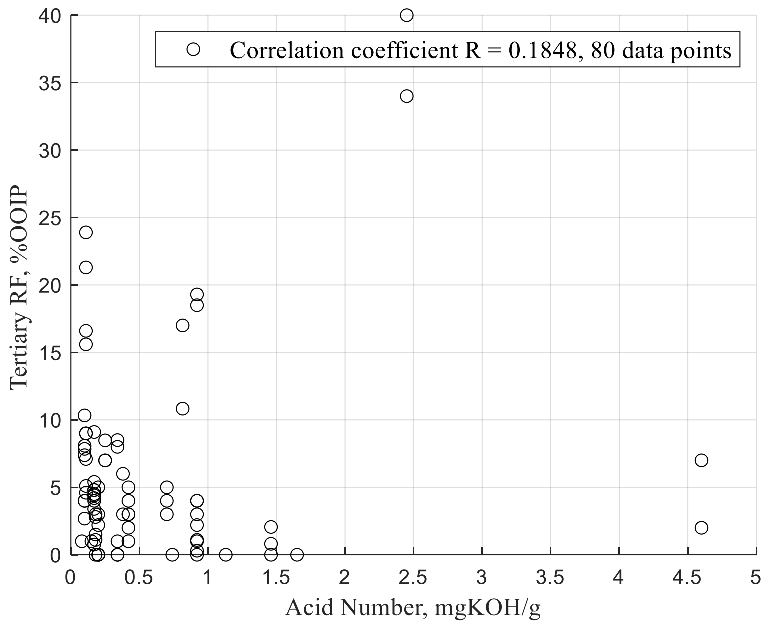

Figure 19. Incremental RF and AN of crude oil

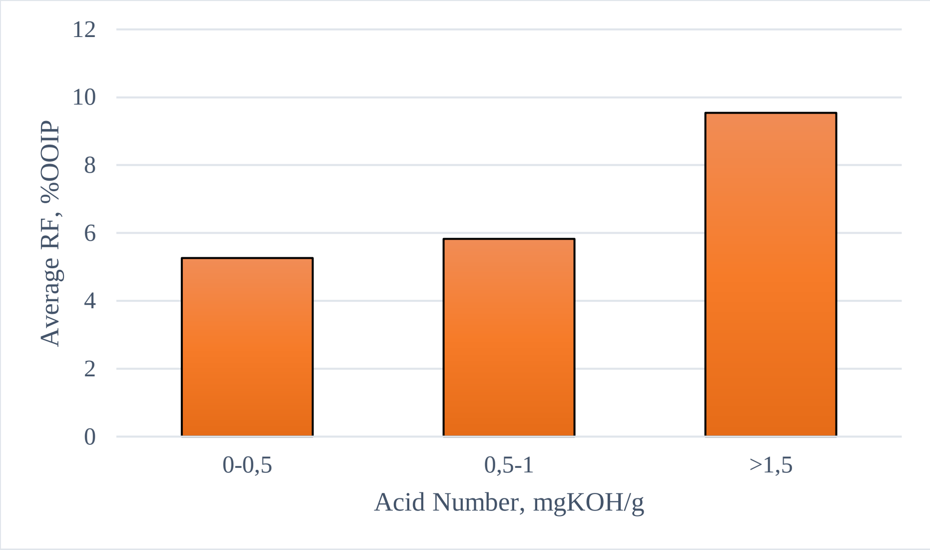

Figure 20. Average RF vs Acid Number

Figure 21 scatters the additional recovery factor after tertiary LSW flooding against oil base number. A weak correlation is established between base number and tertiary recovery factor (R=0.2334). At low values of BN, higher RF were achieved by LSW.

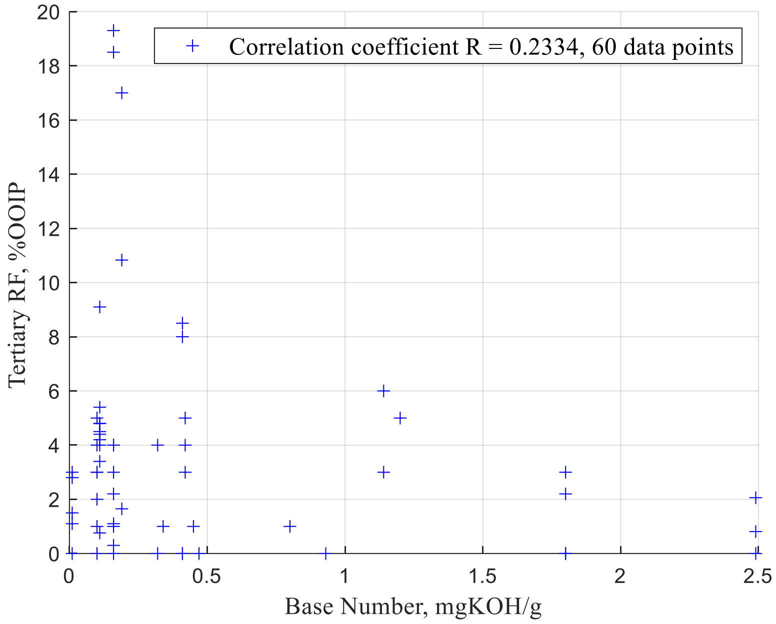

Figure 21. Incremental RF and the base number of crude oil

3.1.4 *Temperature*

A few studies have identified a positive effect of temperature on wettability alteration and incremental oil recovery during LSW, whereas some studies also reported an unclear dependency of LSW efficiency on temperature (Mahani et al., 2017; Fathi et al., 2010; Ravari, 2011). Temperature was linearly regressed with RF based on 98 data entries (Figure 22). The strength of correlation observed is weak, as the correlation coefficient (R=0.2647) is not sufficient for explaining the variance of LSW performance. Figure 23 shows different ranges of experimental temperature and average RF obtained in corresponding ranges. It can be seen that at higher temperature (more than 100°C) the incremental oil recovery is greater. So, elevated temperature can be one of the possible reasons for enhancing oil recovery during LSW injection.

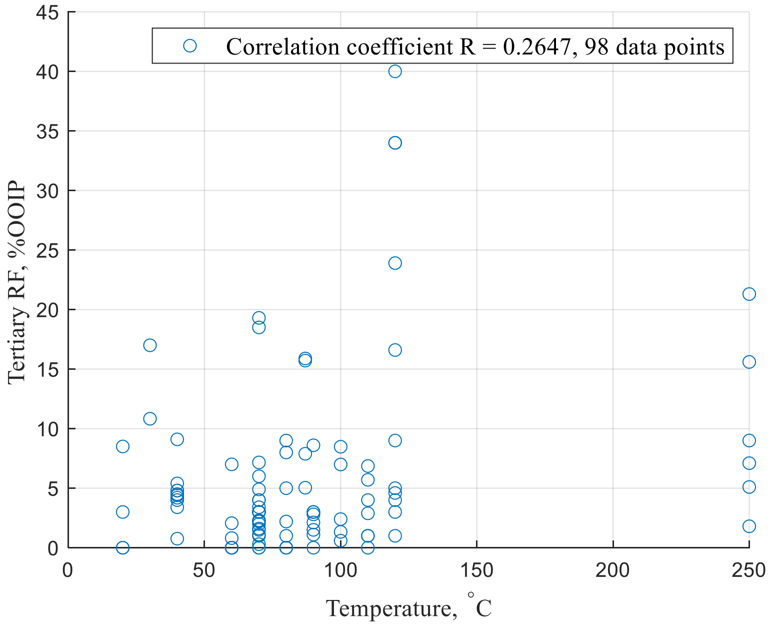

Figure 22. Incremental RF and temperature

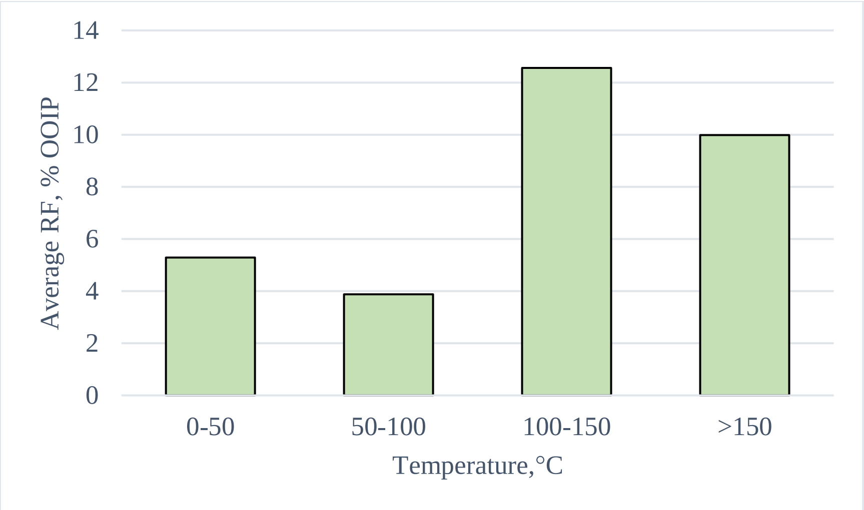

Figure 23. Average RF in different temperature ranges

Table 7 summarizes all results obtained from single-variable linear regression for main parameters. Only coefficients of correlation obtained for temperature and BN showed weak linear relationships between parameters and RF. Among brine parameters, linear regression of SO_4^{2-}

concentration against RF showed better data fitting than others. However, it is not enough for explaining of the variance of LSW effect. The linear relationships between single parameters and the improved recovery factor are mostly negligible, so single parameter cannot explain the LSW performance in carbonates. It is thus inferred that LSW effect is probably the synergistic result of several properties.

Table 7. Single-variable linear regression results

Parameters		**Number of data points**	**Correlation Coefficient R**	**Strength of relationship**
Rock	Permeability	118	0.1721	Negligible
	Low salinity	117	0.1059	Negligible
Brine	Change in salinity	117	0.046	Negligible
	Cations	110	0.029	Negligible
	SO_4^{2-}	107	0.1593	Negligible
Oil	AN	80	0.1848	Negligible
	BN	60	0.2334	Weak
Temperature	T	98	0.2647	Weak

3.2 Effect of a group of parameters on oil recovery

As we discussed, linear regression between single variables and incremental RF failed in the interpretation of LSW performance. Hence, combinatorial effect of controlling parameters on the LSW effect were investigated. For this purpose, we analyzed two of properties, brine and oil/brine, using multivariable regression and nonlinear regression techniques. Dimensionless numbers for all parameters were developed and used to preserve the physical significance of controlling parameters.

3.2.1 Linear multivariable regression

Alteration in salinity and in composition of PDIs are reported to be both responsible in increasing oil recovery. Hence, we developed new parameters to consider effects of salinity, cations, and anions (Equations 1-5). The parameters are represented in Table 7 and showed better results compared to other studied parameters. Comparison of models is based on coefficients of determination R^2, adjusted R^2, and p-value. These coefficients show the strength of the linear relationship for two or more independent variables.

The group of brine parameters include concentration of PDIs and salinity of injected fluids. Totally 96 data points reported simultaneously PDIs and salinity in available experimental studies. We compared different combinations of these parameters based on the main coefficients (Table 8).

The group of oil/brine properties was analyzed using the multivariable linear regression. To the previous set of parameters, we added another variable, the ratio of acid to the base number, to consider all effective properties of fluids. It reduces the number of data points to 42. As it can be seen from Table 8, inclusion of salinity improves the regression model, as the adjusted R^2 becomes higher and the p-value for ions concentration variables decreases. Adjusted R^2 increases when TDS was added to the model, suggesting a better data fitting. Figures 24-29 show the predicted RF from linear regression models for brine and oil/brine parameters against actual RF values .

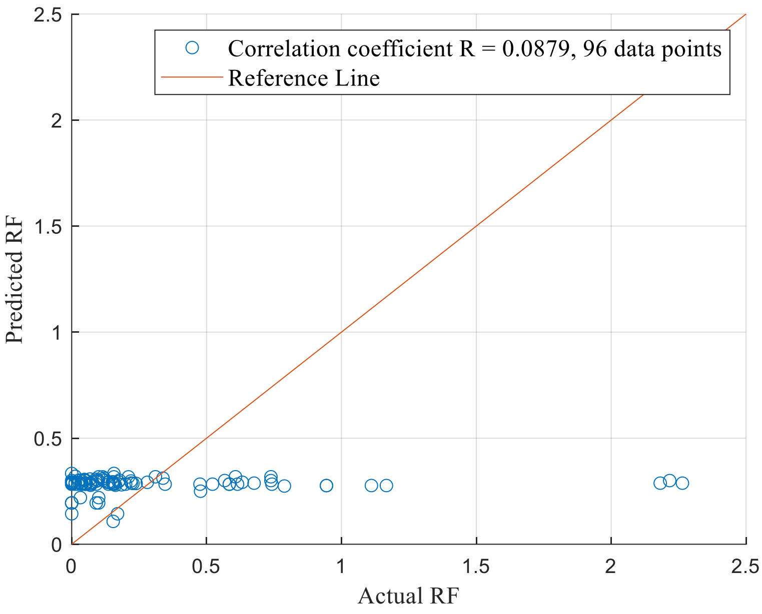

Figure 24. Predicted RF from linear regression and actual RF from experiments for brine parameters (Predicted RF=0.33-0.557*Cations)

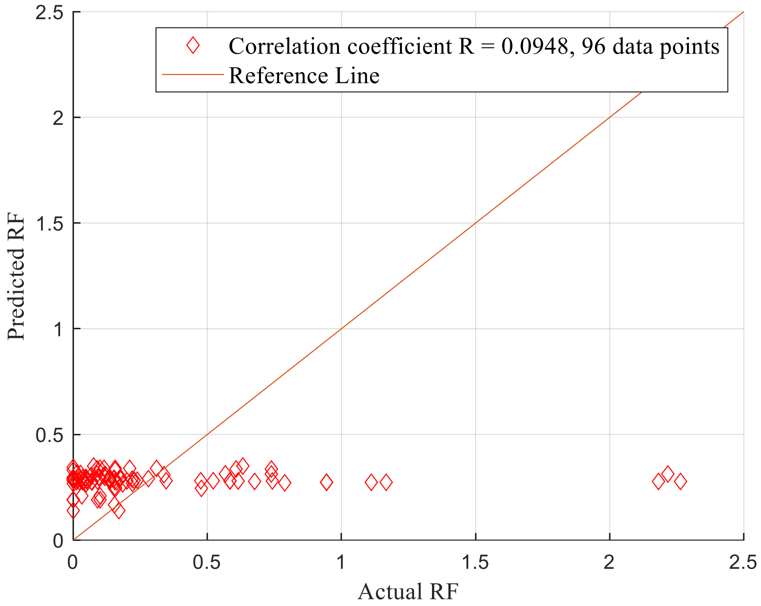

Figure 25. Predicted RF from linear regression and actual RF from experiments for brine parameters (Predicted RF=0.328-0.56*Cations+0.663*SO_4^{2-})

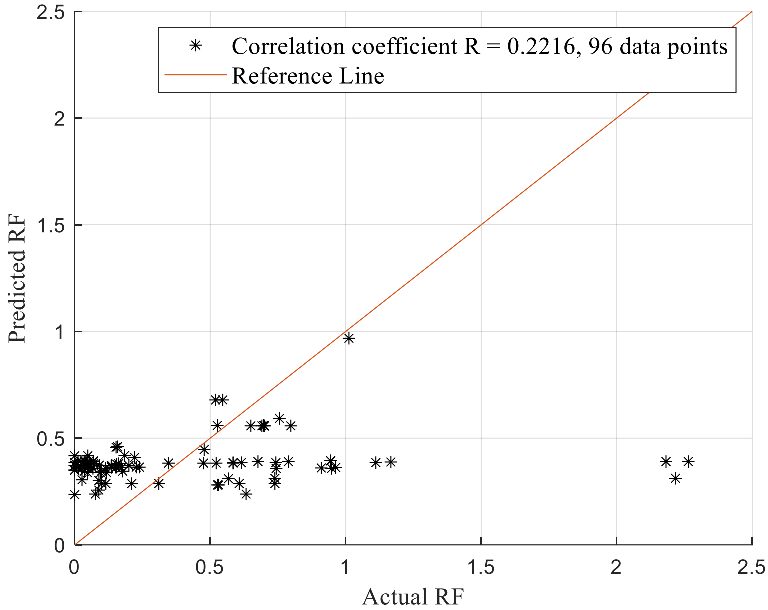

Figure 26. Predicted RF from linear regression and actual RF from experiments for brine parameters (Predicted RF=0.35+1.17*Cations-1.433*SO_4^{2-}- 0.085*TDS)

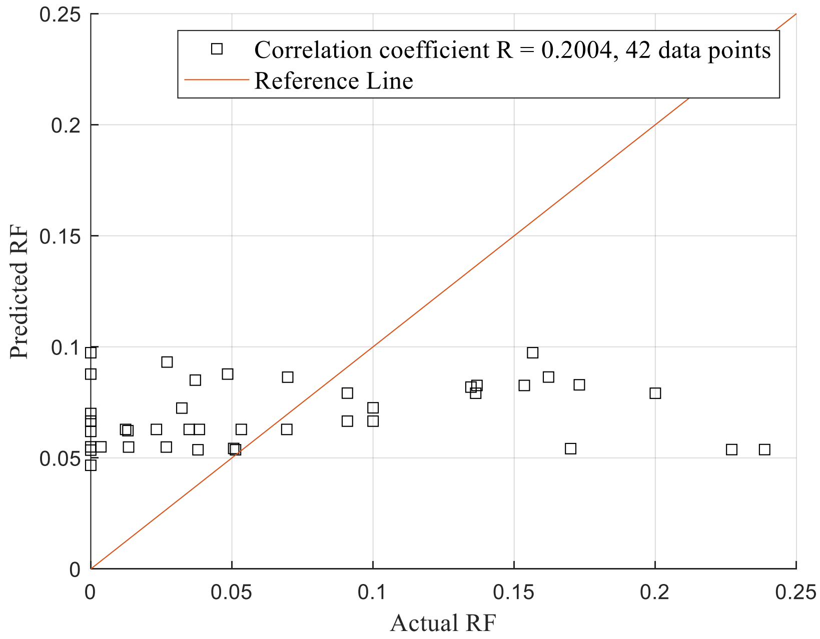

Figure 27. Predicted RF from linear regression and actual RF from experiments for oil/brine parameters (Predicted RF=0.102-0.14*Cations-0.006*AN)

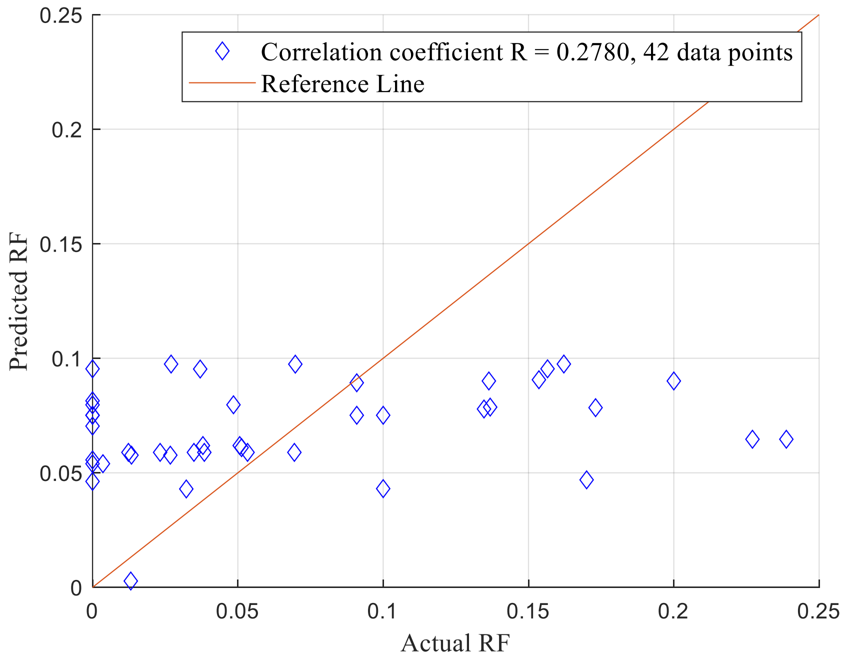

Figure 28. Predicted RF from linear regression and actual RF from experiments for oil/brine parameters (Predicted RF=0.033-0.158*Cations-0.0065*AN+0.082*TDS)

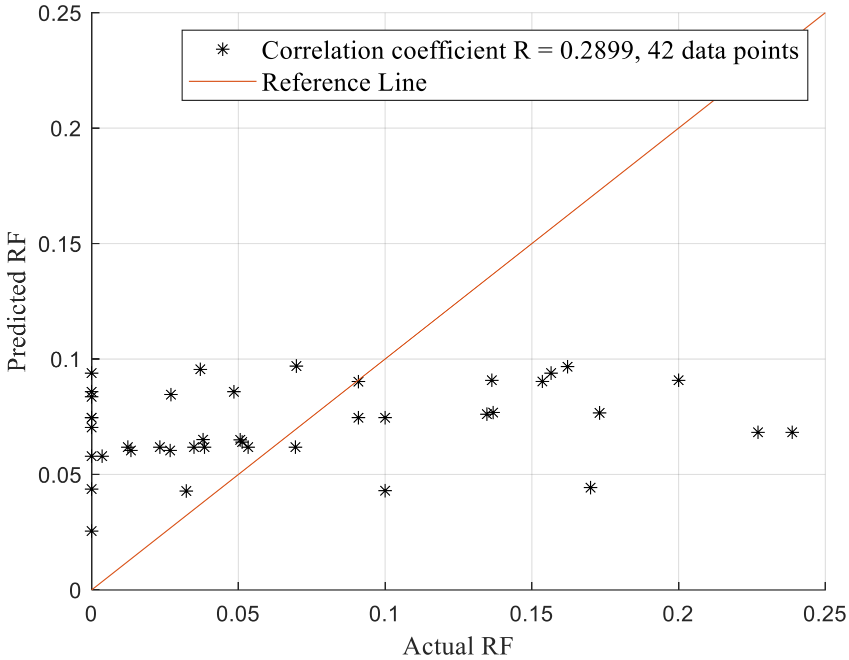

Figure 29. Predicted RF from linear regression and actual RF from experiments for oil/brine parameters (Predicted RF=0.023-0.158*Cations-0.0058*AN+0.091*TDS-0.48*SO_4^{2-})

Table 8. Multivariable linear regression results for brine parameters

Properties	**Variable**	**R**	**Adjusted R^2**	**p-value**	**MRE**	**№ of data points**
Brine	C+ Cations	0.0879	-0.0028	1.89e-05; 0.39	421	96
	C+ Cations+ SO_4^{2-}	0.0948	-0.0012	2.7e-05; 0.39; 0.73	415	96
	C+ Cations+ SO_4^{2-} +TDS	0.2216	0.0181	0.14; 0.09; 0.05; 0.77	470	96
Oil/Brine	C+Cations+AN	0.2	-0.009	0.001; 0.35; 0.23	156	42
	C+Cations+AN+TDS	0.278	0.0044	0.6; 0.3; 0.21; 0.22	154	42
	C+Cations+AN+TDS +SO_4^{2-}	0.289	-0.015	0.72; 0.31; 0.29; 0.19; 0.6	153	42

Correlation coefficients show that the combination of oil/brine parameters can better explain the LSW effect than a group of brine parameters, but the results obtained from linear regression model are still not sufficient to explain the effect of controlling parameters. Table 9 shows the strengths of linear relationships between these properties according to the magnitude of correlation coefficient R. Weak relationships between AN and brine properties and RF were obtained, indicating that AN in combination with other brine parameters is playing a nonnegligible role in LSW performance in carbonates.

Table 9. Strength of linear relationship for brine and oil/brine parameters

Properties	Variable	Strength of relationship
Brine	C+ Cations	Negligible
	C+ Cations+ SO_4^{2-}	Negligible
	C+ Cations+ SO_4^{2-}+TDS	Weak
Oil/Brine	C+Cations+AN	Weak
	C+Cations+AN+TDS	Weak
	C+Cations+AN+TDS+SO_4^{2-}	Weak

3.2.2 Nonlinear multivariable regression

Linear regression analysis did not show acceptable results to explain the relationship between governing parameters and the LSW effect. The strengths of the relationships from the multivariable linear regression model for different sets of variables were found to be from negligible to weak. As expected, no linear relationship between parameters and RF was established. Hence, we applied Machine Learning approaches, nonlinear regression models, for further analyses of these parameters. Data analyses were conducted using three different ML models: Support Vector Machine (SVM), Artificial Neural Networks (ANN), and Decision Tree (DT). They have been successfully used in different petroleum areas to predict performances and analyze complex data. The random division of data points was conducted by separating them into training and testing groups in the proportion 0.7 to 0.3. Average correlation coefficients were obtained from 5000 simulations. The best fitted model was found to interpret the LSW performance.

Oil/brine parameters were analyzed in this section. There are 96 data points containing brine parameters analyzed by ML models. The best interpretation of LSW flooding based on brine parameters was achieved by Decision Tree with the Minimum Leaf Size of 10, as the correlation coefficients for training and testing data are the highest among all ML models (Table 10). A set of oil/brine properties, including dimensionless brine parameters and AN, were analyzed based on 42 data entries. All three ML models showed strong and very strong relationships between oil/brine parameters and RF, and data was fitted better than the case with only brine parameters. The best results were obtained from the Decision Tree method. Both coefficients for training and testing data are high without obvious overfitting. Figures 30-35 show predicted RF values from ANN, SVM, DT models and actual RF values for brine and oil/brine parameters. Correlation coefficient R was averaged upon 5000 model simulations.

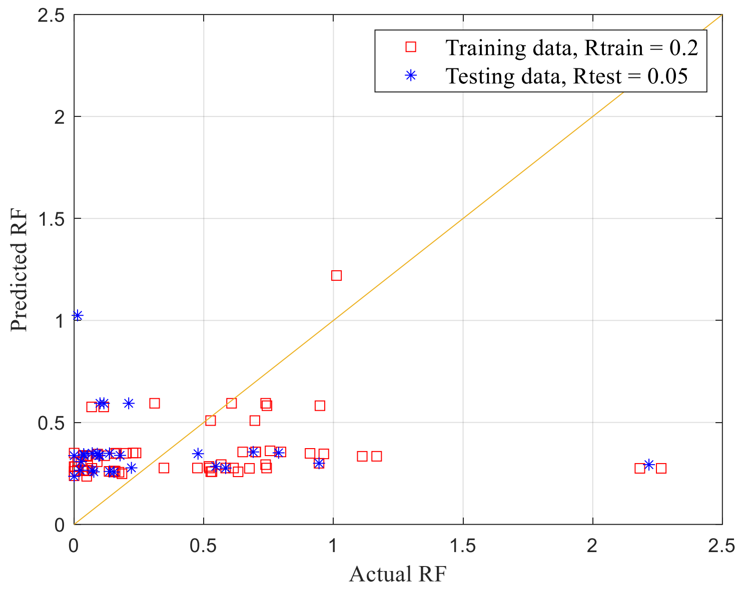

Figure 30. Predicted RF from ANN and actual RF from experiments for brine parameters

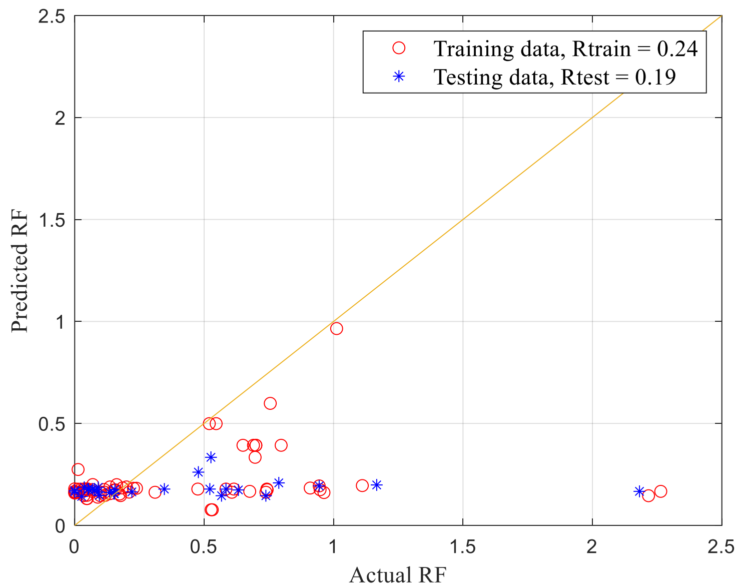

Figure 31. Predicted RF from SVM and actual RF from experiments for brine parameters

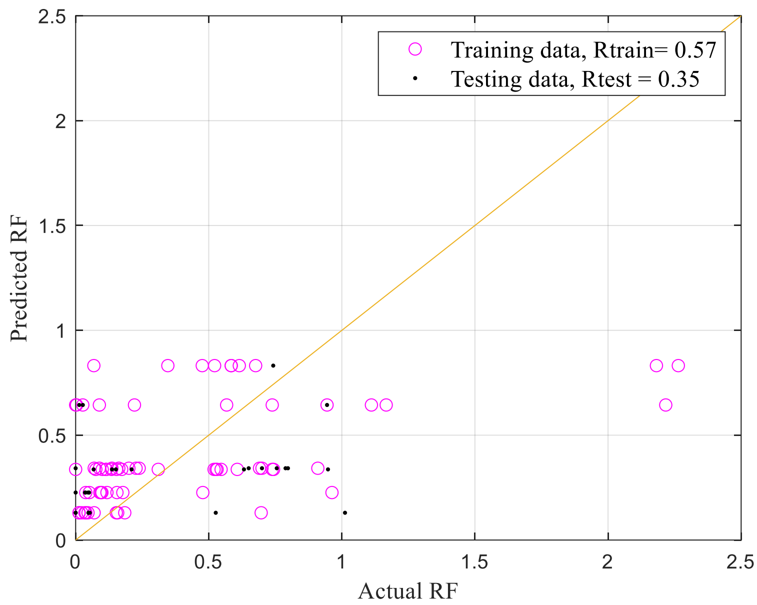

Figure 32. Predicted RF from DT and actual RF from experiments for brine parameters

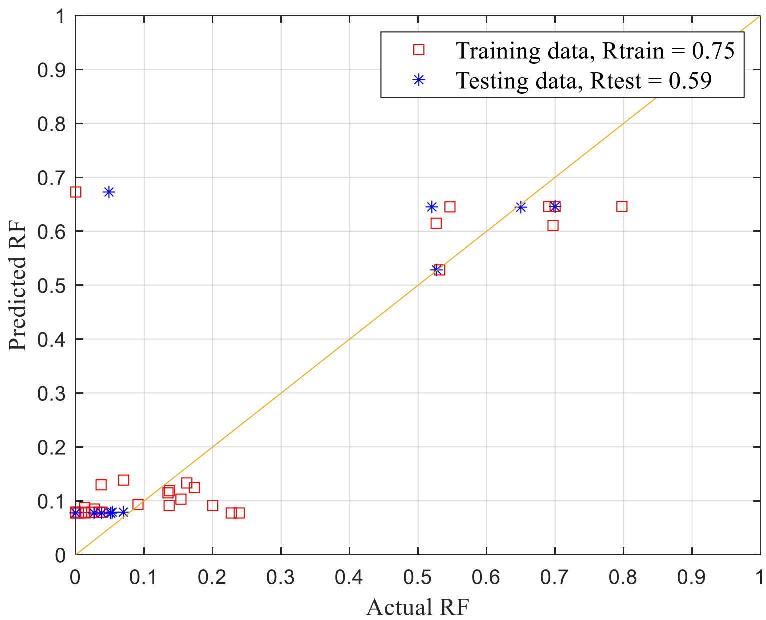

Figure 33. Predicted RF from ANN and actual RF from experiments for oil/brine parameters

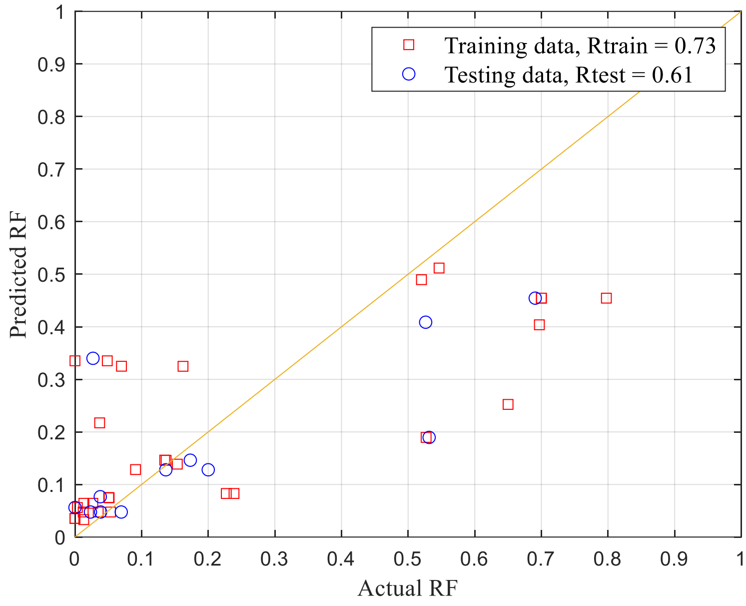

Figure 34. Predicted RF from SVM and actual RF from experiments for oil/brine parameters

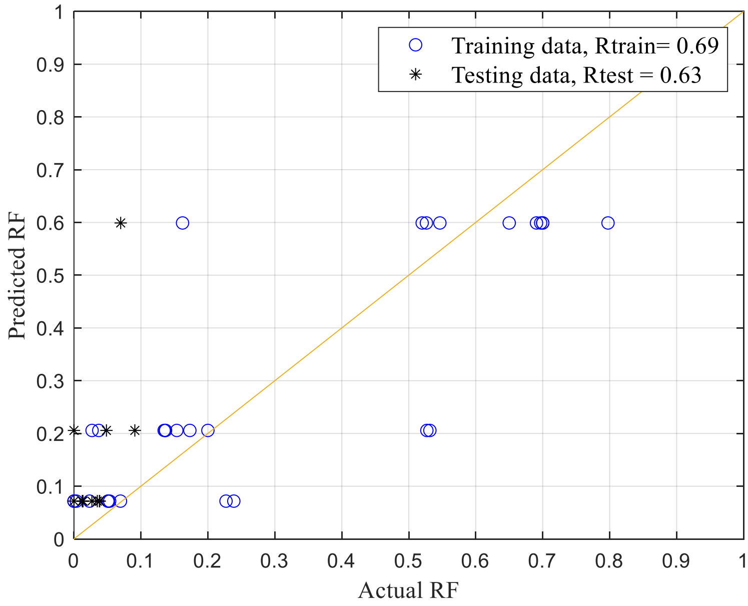

Figure 35. Predicted RF from DT and actual RF from experiments for oil/brine parameters

Table 10 summarizes the average coefficients of correlation obtained from three ML models. Table 11 shows the strengths of nonlinear relationships for these ML models. The average values and ranges of R obtained from 5000 simulations are illustrated in Figures 36-37.

Table 10. The results obtained from three ML models for different sets of parameters

Parameters	**Number of data points**	**Model**	**Average R for training data**	**Average R for testing data**	**MRE**

C+Cations+ SO_4^{2-}+TDS	96	ANN	0.2	0.04	484.7
		SVM	0.24	0.18	220.8
		DT	0.57	0.35	176.7
C+Cations+ SO_4^{2-} +TDS+AN	42	ANN	0.75	0.59	184.7
		SVM	0.73	0.61	243.6
		DT	0.68	0.63	240.5

Table 11. Strength of nonlinear relationship for brine and oil/brine parameters

Parameters	**Model**	**Strength of relationship**	
		Training data	Testing data
C+Cations+ SO_4^{2-}+TDS	ANN	Weak	Negligible
	SVM	Weak	Negligible
	DT	Strong	Moderate
C+Cations+ SO_4^{2-} +TDS+AN	ANN	Very strong	Strong
	SVM	Very strong	Strong
	DT	Strong	Strong

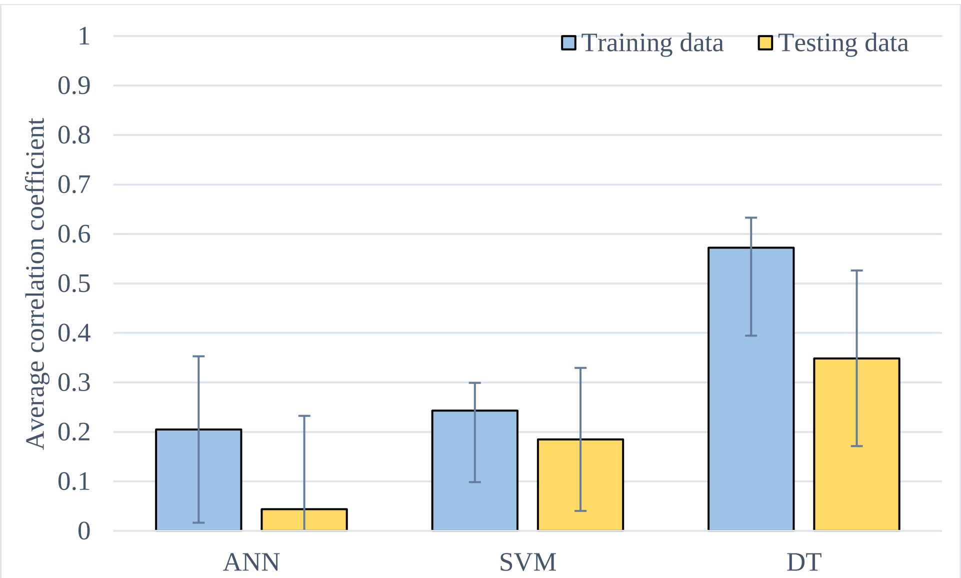

Figure 36. Average correlation coefficients of ML models for brine parameters

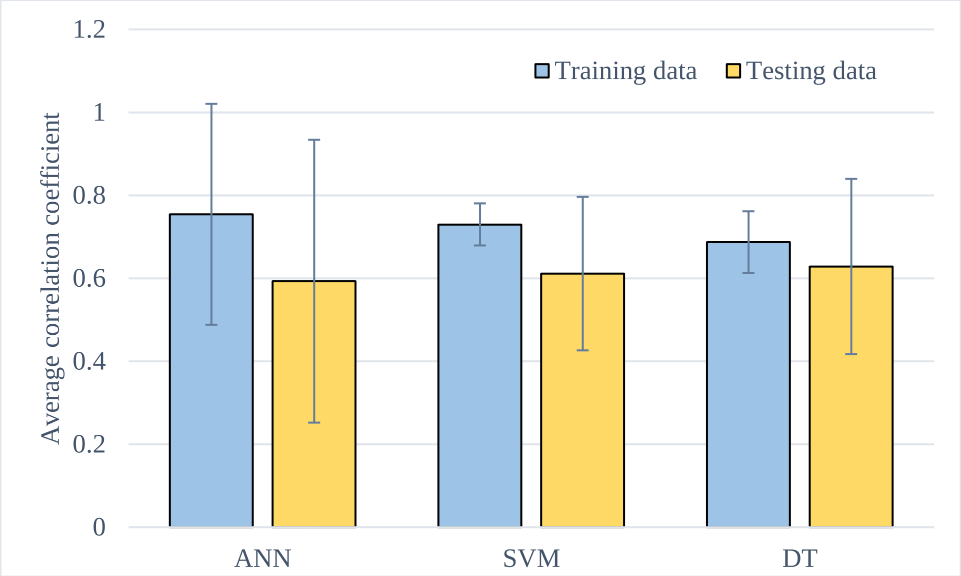

Figure 37. Average correlation coefficients of ML models for oil/brine parameters

Nonlinear relationship between controlling parameters and incremental recovery factor better explains the LSW performance than linear regression. For brine properties, Decision Tree provided the best fit, as the average values of R are considered to be strong and moderate for training and testing data, respectively, and MRE is the lowest (176.7%). For oil/brine parameters, ANN showed the highest results for average R (R_training=0.75), but overfits the data. Decision Tree yielded high correlation coefficients (0.68 and 0.63 for training and testing) and MRE=240.5% with negligible overfitting, exhibiting good performance.

Using linear regression, we showed that LSW effect could not be modeled based on a single parameter, so it is a result of a combined contribution of several parameters. So, we make predictions of LSW based on a set of main parameters and discovered that the best prediction was made using oil/brine properties. Machine Learning models help to achieve better results in explaining the connection between a set of controlling parameters and the positive LSW effect.

3.3 Linking mechanisms to parameters

Different mechanisms governing LSW performance in carbonates were proposed in prior studies (Al Kharousi et al., 2018; Meng et al., 2015; Ligthelm et al., 2009; Hiorth et al., 2010). Multicomponent ionic exchange (MIE), interfacial tension (IFT) reduction, expansion of electric double layer (EDL), and rock dissolution were the main mechanisms suggested by researchers to explain the incremental oil recovery by LSW. Wettability alteration is widely accepted as a reason for a LSW on oil recovery improvement.

In this section, using statistical analysis of some parameters, such as PDIs effluent concentration, pressure drop, IFT, pH of effluent brine, and zeta potential, we studied the occurrence of the main mechanisms. The change in PDI concentration (Ca^{2+}, Mg^{2+} and SO_4^{2-} ions) in the injected and effluent brine can be used to study the dominance of the MIE and rock dissolution mechanisms. Measured IFT values can explain the IFT reduction mechanism. Change in zeta potential can be a tool to show the mechanism of EDL expansion. From tertiary coreflooding results, we extracted the values of these parameters.

3.3.1 PDIs concentration

Active ions concentration in the effluent brine can be an indicator of two mechanisms: MIE and rock dissolution. When MIE is dominant, Ca^{2+}, Mg^{2+} and SO_4^{2-} decrease due to the adsorption of ions onto rock surface. In contrast, the rock dissolution mechanism involves a rise in the effluent Ca^{2+} and SO_4^{2-} concentrations.

Totally 24 data recordings of Ca^{2+} concentration change in the effluent were found in the literature. Increase, decrease and no change in Ca^{2+} concentration are compared in Figure 38. Relatively equal number of rise and reduction of Ca^{2+} concentration was found in the experimental studies. Average RF was calculated for these cases (Figure 39). It is clear that approximately similar RF is achieved when either of these two different mechanisms is dominantly active.

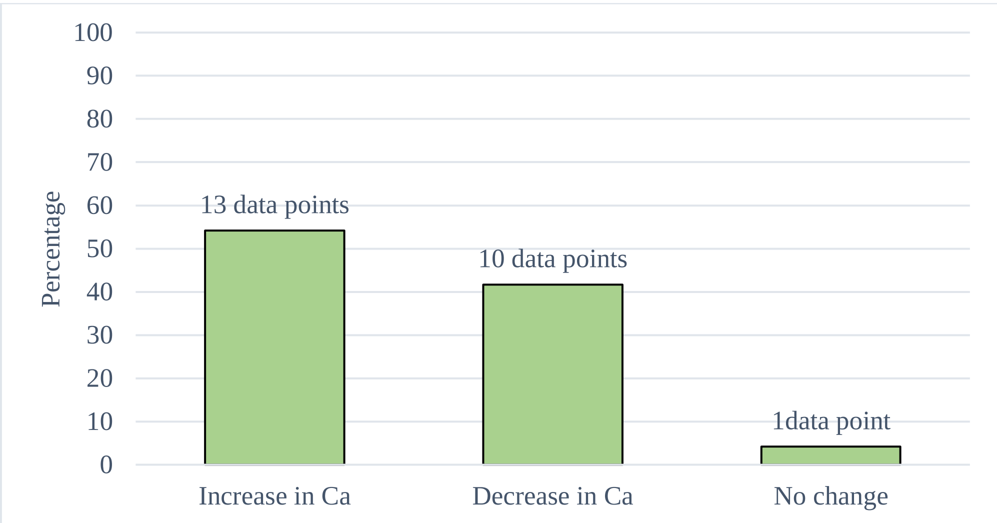

Figure 38. Number of data points for Ca^{2+} concentration change

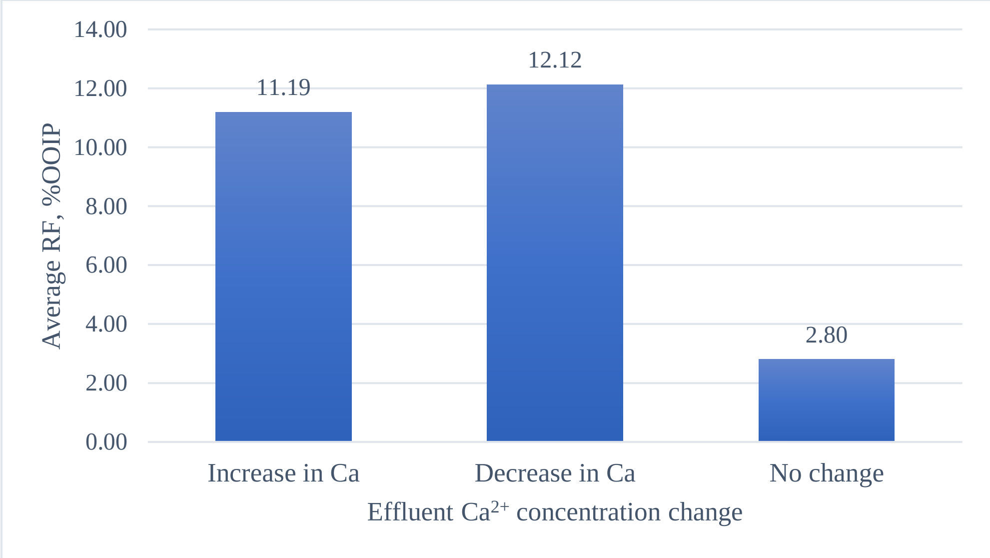

Figure 39. Average RF vs effluent Ca^{2+} concentration change

There are 13 data points containing the effluent Mg^{2+} concentration collected from experimental studies (Figure 39). Approximately the same number of data points reported an increase and decrease in Mg^{2+} concentration. Average RF and temperature are higher when Mg^{2+} ions concentration decreases in the effluent (Figure 40 and Figure 41). It can be explained by the effect of temperature on increased Mg^{2+} activity toward the carbonate surface, which results in adsorption of cation (Awolayo et al., 2018a).

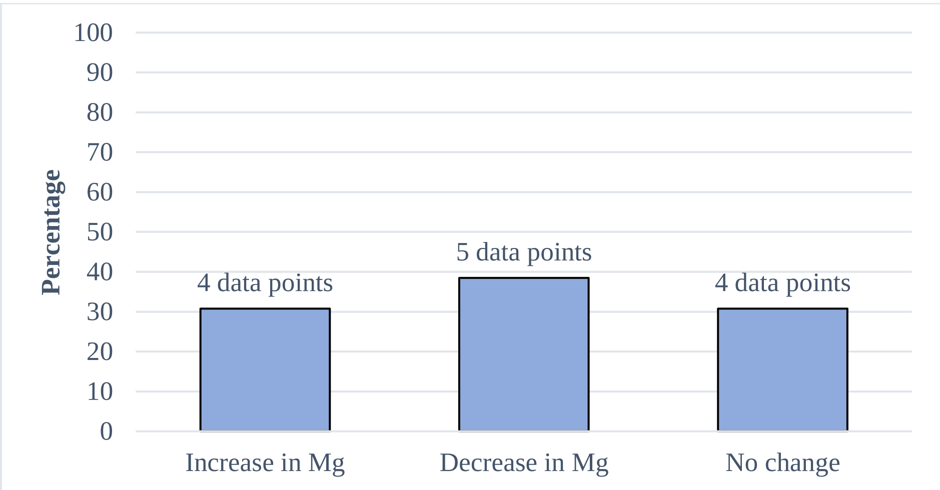

Figure 40. Number of data points for Mg^{2+} concentration change

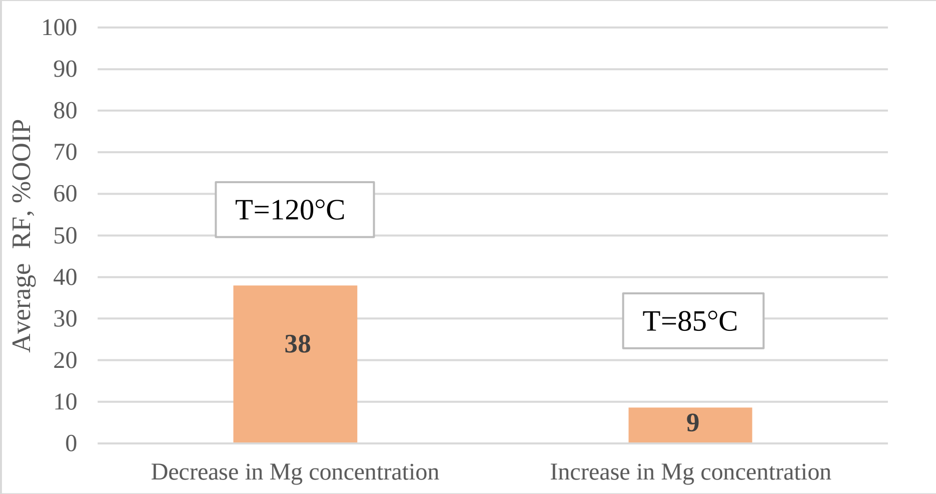

Figure 41. Average RF vs Mg^{2+} concentration change

SO_4^{2-} concentration change in the effluent brine was reported 14 times (Figure 42). Reduction of SO_4^{2-} concentration in effluent was recorded in 7 coreflooding tests, and an increase in anion concentration was indicated in 6 experiments. Higher average RF corresponds to decrease of SO_4^{2-} concentration, which in combination with Ca^{2+} reduction supports the rock dissolution mechanism (Figure 43).

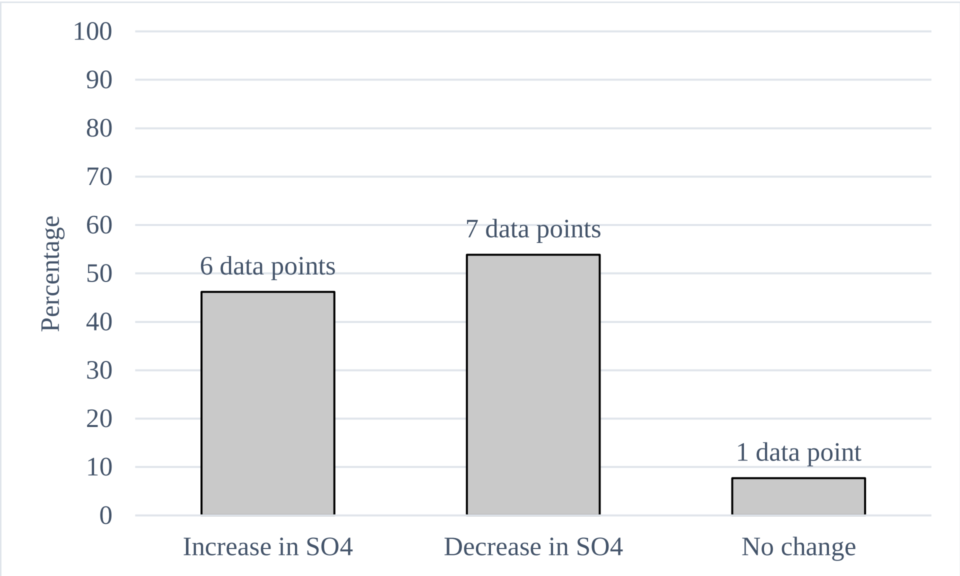

Figure 42. Number of data points for SO_4^{2-} concentration change

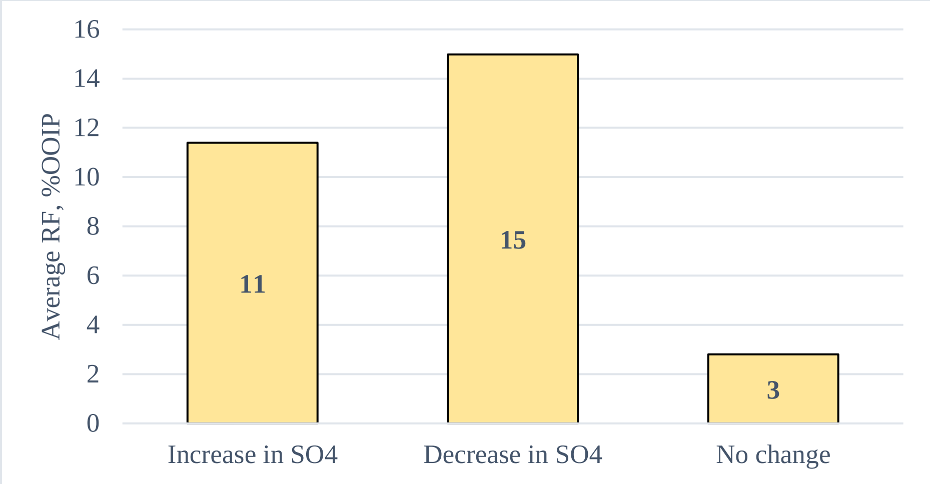

Figure 43. Average RF vs SO_4^{2-} concentration change

As the change in concentrations of separate PDIs are not sufficient for an indication of mechanism, we analyzed combinatorial effect of all three active ions in LSW. Table 12 shows the recordings of ions change in the effluent brine and proposed mechanisms for these cases. Most experiments indicated the rock dissolution effect by measuring the PDI concentration. However, there are also some cases where MIE was active mechanism for LSW effect in carbonates.

Table 12. Proposed mechanisms based on PDIs concentration change

Paper	Mg^{2+}	Ca^{2+}	SO_4^{2-}	Proposed Mechanism
Austad et al., 2012		Increase	Increase	Rock dissolution
Chandrasekhar et al., 2016	No change	Increase	Increase	Rock dissolution
Awolayo et al., 2014		Decrease	Decrease	MIE
Awolayo et al., 2016		Increase	Decrease	Rock dissolution
Shehata et al., 2014		No change	No change	-
Gupta et al., 2011	No change	Increase		Rock dissolution
Mohammadkhani et al., 2018	Increase	Decrease		MIE
Vo et al., 2012	No change	Increase		Rock dissolution
	No change	Increase		-
Chandreskaer et al., 2018	Decrease	Decrease		MIE

3.3.2 IFT

Brine/oil interfacial tension reduction was suggested as one mechanism of LSW flooding. There are 17 measurements of IFT after secondary and tertiary stages of flooding found in literature. In coreflooding tests, measurements mostly indicated decrease in IFT (Figure 44). We also analyzed RF for different ranges of IFT decrease and noticed that average RF increases with bigger difference in IFT (Figure 45). Hence, big change in IFT can improve oil recovery more in contrast to small difference in IFT values during tertiary injection of LSW.

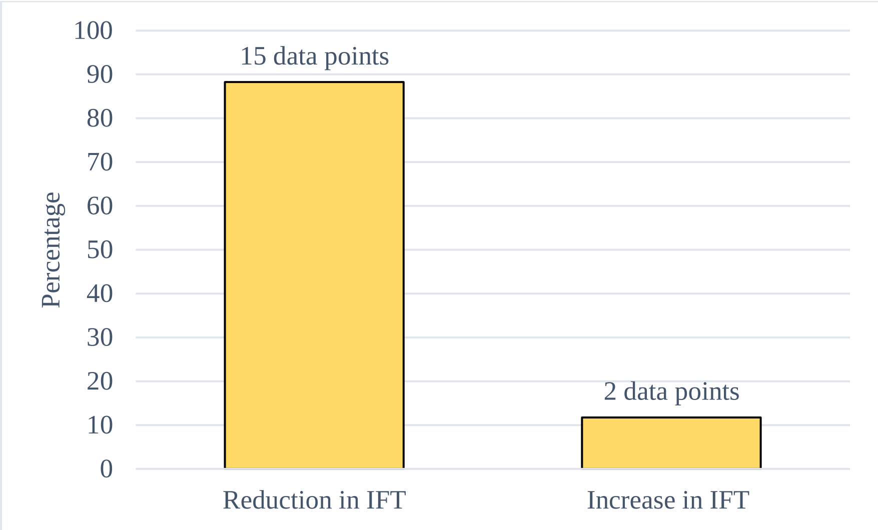

Figure 44. Number of data points for IFT change

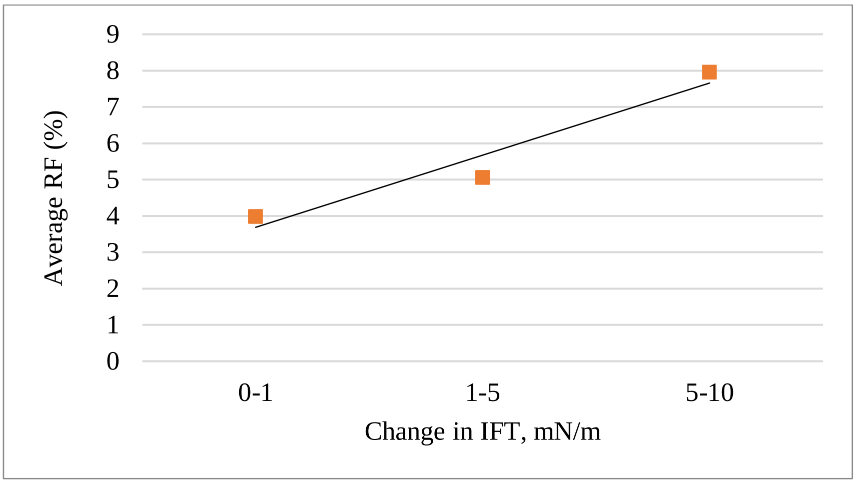

Figure 45. Average RF vs Change in IFT

3.3.3 pH

The effect of pH was studied by a number of researchers (Ligthelm et al., 2009; Gandomkar et al., 2015; Mohammadkhani et al., 2018). It is believed that the pH of the effluent brine increases when injected brine is switched from high salinity to low salinity ones. pH increase can be explained by a rock dissolution mechanism (Mohammadkhani et al., 2018). pH increase also can alter surface charge toward more negative and increase repulsive electrostatic forces at the rock/fluid interface (Lingthlem et al., 2009). It results in EDL expansion and a thicker water-wet film near the rock surface.

There are 24 recordings of pH increase, decrease or no change (Figure 46). The most common case is pH increase. Among these 24 data points, about 14 experimental tests reported the numeric value of effluent brine pH. We divided these data points into two categories: small change (less than 10%) and big change (more than 10%). Average RF for both cases were calculated and compared (Figure 47). It is clear that pH increase is more common than decrease in pH, and close RF values were achieved with small and big changes of pH in the effluent.

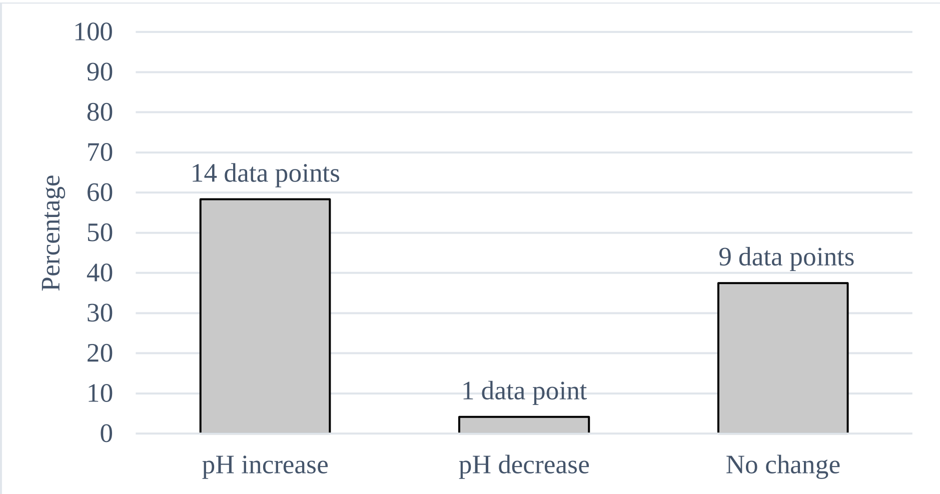

Figure 46. Number of data points for pH change

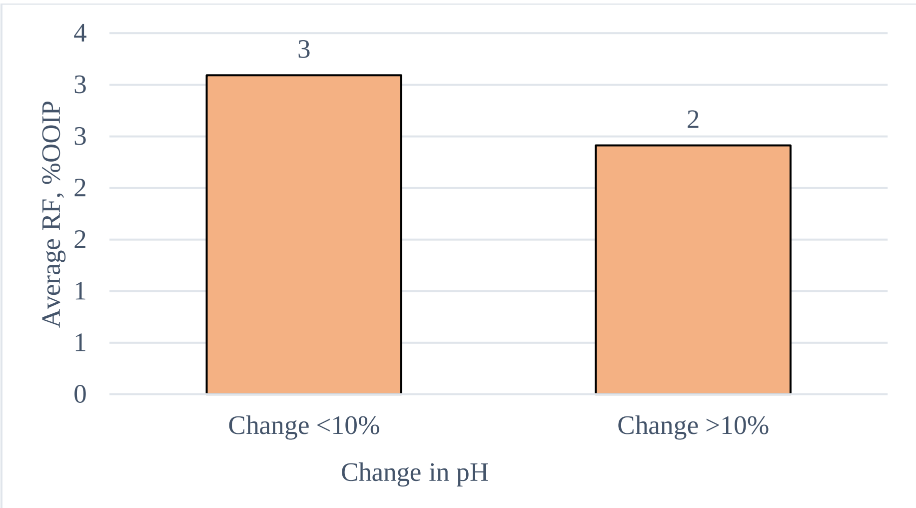

Figure 47. Average RF vs pH change

3.3.4 Zeta potential

Zeta potential changes due to the reduced concentration of cations, e.g., Mg^{2+} and Ca^{2+}, in LSW, which results in the predominance of repulsive forces (Ligthelm et al., 2009). Thus, the EDL expands, and water-wet films become thicker and more stable. As a result, oil components are desorbed and the oil recovery is improved by LSW (Lingthelm et al., 2009).

There are 14 measurements of zeta potential collected from existing experimental studies, 12 of them reported zeta potential change of more than 6 mV. Mostly, zeta potential after LSW injection became more negative. As can be seen from Figure 48, different RF is achieved at different values of zeta potential change. However, even small change in zeta potential can yield a noticeable oil recovery improvement.

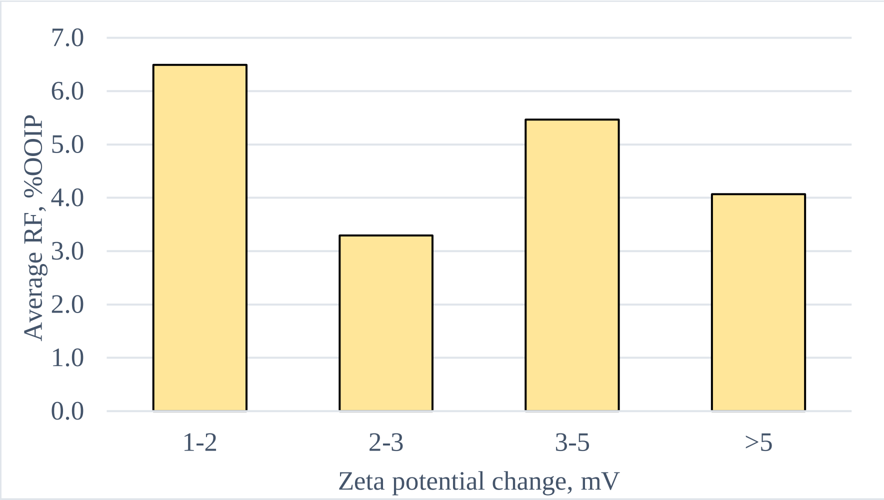

Figure 48. Average RF vs zeta potential change

3.3.5 Pressure change

During LSW injection, pressure drop is expected to decrease. We collected the recordings of pressure change and found 56 data points reporting decrease of pressure drop. Figure 49 compared the number of cases with differential pressure decrease and no change. Approximately same average RF was indicated for both cases (Figure 50). This effect can be explained by change in relative permeability as a result of switching from high to low salinity brines (Nasralla et al., 2018).

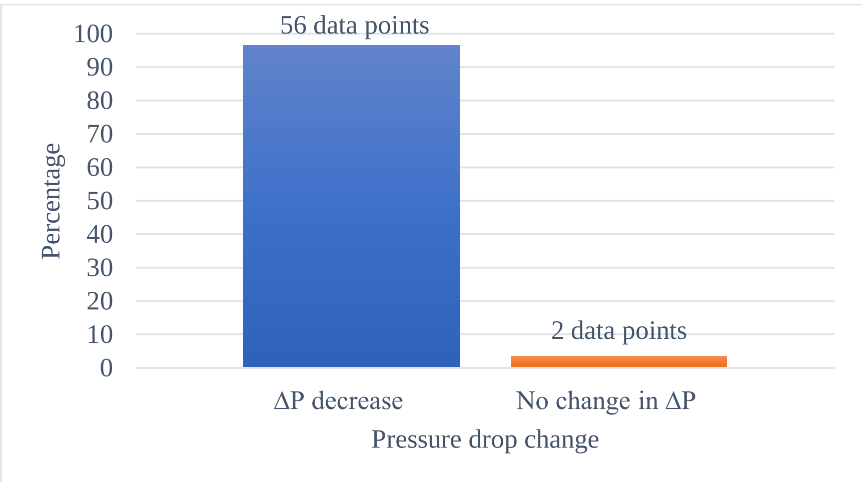

Figure 49. Number of data points for change in pressure drop

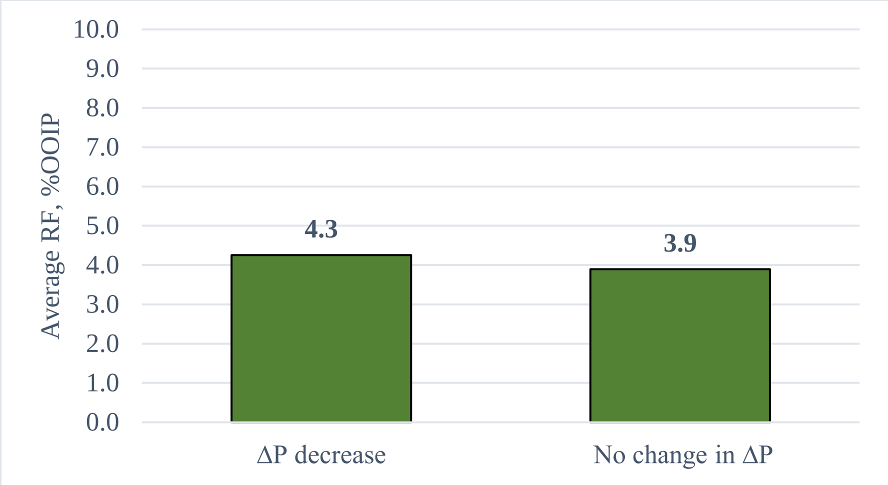

Figure 50. Average RF vs pressure drop change

3.3.6 Wettability alteration

Wettability alteration was indicated in different experimental studies by measuring contact angle and wettability index. Totally 57 coreflooding tests reported a change of wettability of the system toward a more water-wet state (Figure 51). Alteration toward more oil-wet conditions was found only 4 times. By activating different mechanisms discussed earlier, LSW probably induces wettability alteration.

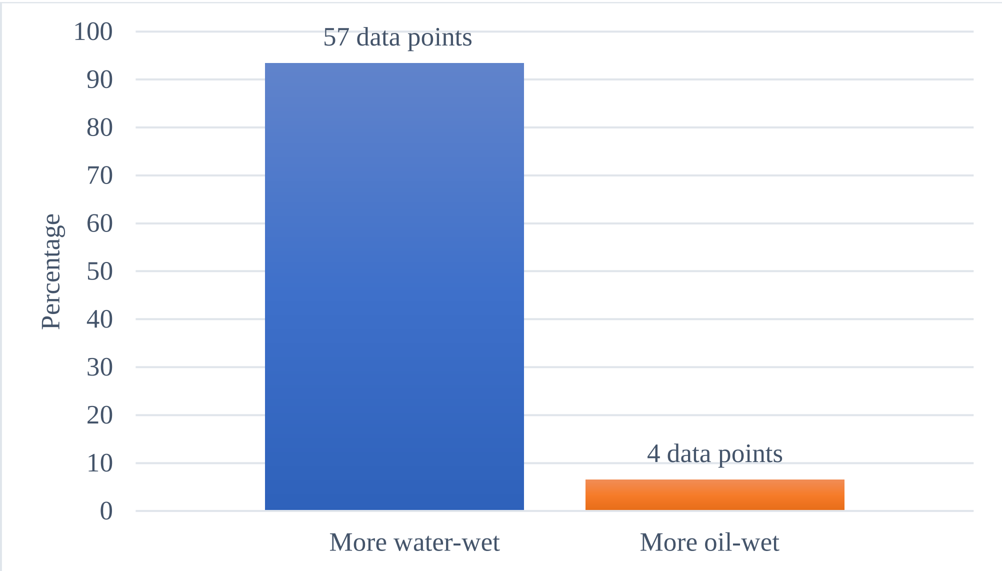

Figure 51. Number of data points for wettability alteration

Different mechanisms were suggested by researchers. There are 50 recordings of mechanisms proposed for LSW flooding tests in literature. Figure 52 compares the number of tests that mentioned different mechanisms. The most popular mechanisms were rock dissolution and EDL expansion. MIE was suggested as an active mechanism for LSW injection 11 times based on PDIs concentration measurements. IFT reduction is the least popular mechanism in experimental studies.

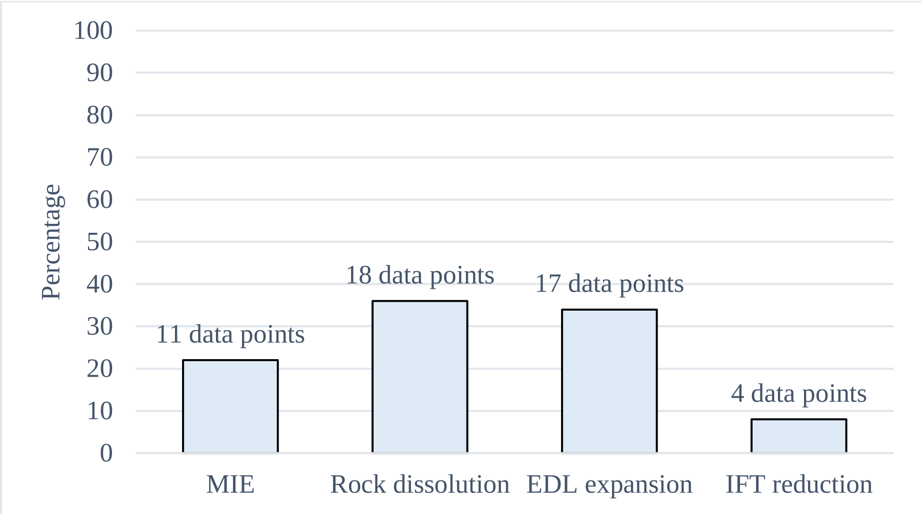

Figure 52. Number of data points for proposed mechanisms

Different mechanisms were linked to parameter changes reported in literature. We also analyzed the ranges of different parameters to investigate if there a linkage between the parameter changes and RF. By analyzing the concentrations of PDIs in effluent, MIE and rock dissolution are explained. For some parameters, like zeta potential and IFT, the contrast was found to be important in LSW effect. LSW performance cannot be explained by one mechanism, as different parameters, such as PDIs concentration change, IFT reduction, and zeta potential are found to have a correlation with oil recovery. However, all of these mechanisms contribute into the wettability alteration, and after the change of wettability toward a water-wet state, oil recovery is improved by LSW.

CHAPTER 4: CONCLUSIONS AND RECOMMENDATIONS

The main objective of this thesis was to understand the conditions for LSW to work and the active mechanisms. Machine learning models and statistical approaches were applied in this study. We developed ML models to predict LSW effect based on controlling parameters and used statistical approaches to study the mechanisms involved in the process. The following conclusions can be drawn based on the obtained results:

- Different single parameters, such as salinity, contrast in salinity change, PDIs concentration, oil acidity, base number of crude oil, permeability, and temperature, were individually analyzed using linear regression. Negligible and weak relationships between single parameters and incremental RF were established. So, a single parameter is not sufficient to explain the performance of LSW injection.
- Among groups of parameters, a set of oil/brine parameters that include AN, alteration in salinity, SO_4^{2-} and cations concentration, showed better correlation than only brine parameters. However, a combination of properties does not linearly correlate with RF. So, linear correlations are insufficient to forecast LSW potential.
- A nonlinear relationship between parameters and RF was observed using Machine learning models. Among ML models, DT produced the best correlation for brine only parameters, the correlation coefficients for training and testing data were 0.57 and 0.35, respectively, and the lowest MRE =176.7%. For oil/brine parameters, all models showed strong and very strong relationships between parameters. However, ANN and SVM showed unsatisfactory results for testing data due to overfitting. In contrast, less overfitting was achieved by DT (R_training= 0.68 and R_testing=0.63).
- Several mechanisms involved in the LSW process, and LSW effect cannot be explained by a single mechanism. MIE and rock dissolution are the most widely accepted mechanisms found in literatures. These mechanisms result in wettability alteration in coreflooding tests in carbonates.
  - Our studies showed that by analyzing oil/brine parameters, a better understanding of the active mechanisms during LSW can be achieved, and it is possible to predict the mechanism by analyzing parameters such as salinity, ions concentration, pH, and IFT.

REFERENCES

1. Ahmed S, A., Elkatatny, S., Ali, A. Z., Abdulraheem, A., & Mahmoud, M. (2019, March). Artificial neural network ANN approach to predict fracture pressure. In *SPE middle east oil and gas show and conference*. Society of Petroleum Engineers.
2. Akbar, M., Vissapragada, B., Alghamdi, A. H., Allen, D., Herron, M., Carnegie, A., ... & Saxena, K. (2000). A snapshot of carbonate reservoir evaluation. *Oilfield Review*, *12*(4), 20-21.
3. Al-Kharusi, B., Pourafshary, P., Mosavat, N., & Al-Wahaibi, Y. (2018, October). Design and performance of smart water shock injection SWSI in carbonate reservoirs. In *SPE Annual Caspian Technical Conference and Exhibition*. Society of Petroleum Engineers.
4. Al-AbdulJabbar, A., Al-Azani, K., & Elkatatny, S. (2020). Estimation of reservoir porosity from drilling parameters using Artificial Neural Networks. *Petrophysics*, *61*(03), 318-330.
5. Al-Attar, H. H., Mahmoud, M. Y., Zekri, A. Y., Almehaideb, R., & Ghannam, M. (2013). Low-salinity flooding in a selected carbonate reservoir: experimental approach. *Journal of Petroleum Exploration and Production Technology*, *3*(2), 139-149.
6. Al Harrasi, A., Al-maamari, R. S., & Masalmeh, S. K. (2012, January). Laboratory investigation of low salinity waterflooding for carbonate reservoirs. In *Abu Dhabi international petroleum conference and exhibition*. Society of Petroleum Engineers.
7. Alhashem, M. (2020, January). Machine learning classification model for multiphase flow regimes in horizontal pipes. In *International Petroleum Technology Conference*. International Petroleum Technology Conference.
8. Alkinani, H. H., Al-Hameedi, A. T., & Dunn-Norman, S. (2020, September). Predicting the risk of lost circulation using Support Vector Machine model. In *54th US Rock Mechanics/Geomechanics Symposium*. American Rock Mechanics Association.
9. Al Mahrouqi, D., Vinogradov, J., & Jackson, M. D. (2017). Zeta potential of artificial and natural calcite in aqueous solution. *Advances in colloid and interface science*, *240*, 60-76.
10. Almashan, M., Narusue, Y., & Morikawa, H. (2019, October). Estimating PVT properties of crude oil systems based on a Boosted Decision Tree Regression modelling scheme with K-Means Clustering. In *SPE/IATMI Asia Pacific Oil & Gas Conference and Exhibition*. Society of Petroleum Engineers.
11. Almashan, M., Narusue, Y., & Morikawa, H. (2020, November). Decision Tree Regressions for estimating liquid holdup in two-phase gas-liquid flows. In *Abu Dhabi International Petroleum Exhibition & Conference*. Society of Petroleum Engineers.

12. Amiri, S., & Gandomkar, A. (2019). Influence of electrical surface charges on thermodynamics of wettability during low salinity water flooding on limestone reservoirs. *Journal of Molecular Liquids*, *277*, 132-141.
13. Arif, M., Abu-Khamsin, S. A., Zhang, Y., & Iglauer, S. (2020). Experimental investigation of carbonate wettability as a function of mineralogical and thermo-physical conditions. *Fuel*, *264*, 116846.
14. Austad, T., Strand, S., Puntervold, T., & Ravari, R. R. (2008, October). New method to clean carbonate reservoir cores by seawater. In *SCA2008-15 presented at the International Symposium of the Society of Core Analysts* (Vol. 29).
15. Austad, T., Strand, S., & Puntervold, T. (2009, September). Is wettability alteration of carbonates by seawater caused by rock dissolution. In *Paper SCA2009-43 presented at the International Symposium of the Society of Core Analysts held in Noordwijk, The Netherlands, September* (pp. 27-30).
16. Awolayo, A., Sarma, H., & AlSumaiti, A. M. (2014, March). A laboratory study of ionic effect of smart water for enhancing oil recovery in carbonate reservoirs. In *SPE EOR Conference at Oil and Gas West Asia*. Society of Petroleum Engineers.
17. Awolayo, A. N., & Sharma, H. K. (2016). Impact of multi-ion interactions on oil mobilization by smart waterflooding in carbonate reservoirs. *J. Pet. Environ. Biotechnol*, *7*(278), 2.
18. Awolayo, A. N., Sarma, H. K., & Nghiem, L. X. (2018a). Modeling the characteristic thermodynamic interplay between potential determining ions during brine-dependent recovery process in carbonate rocks. *Fuel*, *224*, 701-717.
19. Awolayo, A. N., Sarma, H. K., & Nghiem, L. X. (2018b). Brine-dependent recovery processes in carbonate and sandstone petroleum reservoirs: review of laboratory-field studies, interfacial mechanisms and modeling attempts. *Energies*, *11*(11), 3020.
20. Babuška, R., & Kober, J. (2010). Knowledge-based control systems. Delft University of Technology.
21. Bakshi, A., Uniacke, E., Korjani, M., & Ershaghi, I. (2017, April). A novel adaptive non-linear regression method to predict shale oil well performance based on well completions and fracturing data. In *SPE Western Regional Meeting*. Society of Petroleum Engineers.
22. Bazhanova, M., & Pourafshary, P. (2020). Impact of SO_4^{2-}, Ca^{2+} , and Mg^{2+} ions in Caspian Sea ion-engineered water on the rate of wettability alteration in carbonates. *Journal of Petroleum Exploration and Production Technology*, *10*(8), 3281-3293.

23. Chandrasekhar, Sriram, Himanshu Sharma, and Kishore K. Mohanty. "Wettability alteration with brine composition in high temperature carbonate rocks." In *SPE Annual Technical Conference and Exhibition*. Society of Petroleum Engineers, 2016.
24. Chandrasekhar, S., Sharma, H., & Mohanty, K. K. (2018). Dependence of wettability on brine composition in high temperature carbonate rocks. *Fuel*, *225*, 573-587.
25. Collini, H., Li, S., Jackson, M. D., Agenet, N., Rashid, B., & Couves, J. (2020). Zeta potential in intact carbonates at reservoir conditions and its impact on oil recovery during controlled salinity waterflooding. *Fuel*, *266*, 116927.
26. Dubey, S. T., & Doe, P. H. (1993). Base number and wetting properties of crude oils. *SPE Reservoir Engineering*, 8(03), 195-200.
27. Fani, M., Al-Hadrami, H., Pourafshary, P., Vakili-Nezhaad, G., & Mosavat, N. (2018, November). Optimization of smart water flooding in carbonate reservoir. In *Abu Dhabi International Petroleum Exhibition & Conference*. Society of Petroleum Engineers.
28. Fathi, S. J., Austad, T., Strand, S., Frank, S., & Mogensen, K. (2010, September). Evaluation of EOR potentials in an offshore limestone reservoir: A case study. In *Proceedings of the Eleventh International Symposium on Reservoir Wettability, Calgary, AB, Canada* (pp. 6-8).
29. Feldmann, F., Strobel, G. J., Masalmeh, S. K., & AlSumaiti, A. M. (2020). An experimental and numerical study of low salinity effects on the oil recovery of carbonate rocks combining spontaneous imbibition, centrifuge method and coreflooding experiments. *Journal of Petroleum Science and Engineering*, *190*, 107045.
30. Gandomkar, A., & Rahimpour, M. R. (2015). Investigation of low-salinity waterflooding in secondary and tertiary enhanced oil recovery in limestone reservoirs. *Energy & Fuels*, *29*(12), 7781-7792.
31. Gomari, K. R., Hamouda, A. A., & Denoyel, R. (2006). Influence of sulfate ions on the interaction between fatty acids and calcite surface. *Colloids and Surfaces A: Physicochemical and Engineering Aspects*, *287*(1-3), 29-35.
32. Gupta, R., Smith, P. G. J., Hu, L., Willingham, T. W., Cascio, M. L., Shyeh, J. J., & Harris, C. R. (2011). Enhanced waterflood for Middle East carbonate cores–impact of injection water composition. Presented at SPE Middle East Oil and Gas Show and Conference, Manama, Bahrain.
33. Hiorth, A., Cathles, L. M., & Madland, M. V. (2010). The impact of pore water chemistry on carbonate surface charge and oil wettability. *Transport in porous media*, *85*(1), 1-21.

34. Hiorth, A., Cathles, L. M., Kolnes, J., Vikane, O., Lohne, A., & Madland, M. V. (2008, October). Chemical modelling of wettability change in carbonate rocks. In *10th Wettability Conference, Abu Dhabi, UAE* (pp. 1-9).
35. Ibrahim, M. N., & Koederitz, L. F. (2000, January). Two-phase relative permeability prediction using a linear regression model. In *SPE eastern regional meeting*. Society of Petroleum Engineers.
36. Jackson, M. D., Al-Mahrouqi, D., & Vinogradov, J. (2016). Zeta potential in oil-water-carbonate systems and its impact on oil recovery during controlled salinity water-flooding. *Scientific reports*, *6*(1), 1-13.
37. Jalilian, M., Pourafshary, P., Sola, B. S., & Kamari, M. (2017). Optimization of smart water chemical composition for carbonate rocks through comparison of active cations performance. *Journal of Energy Resources Technology*, *139*(6).
38. Kalam, S., Khan, M., Mahmoud, M., Khan, R. A., & Abu-Khamsin, S. A. (2020, November). New vision into relative permeability estimation using Artificial Neural Networks. In *SPE Asia Pacific Oil & Gas Conference and Exhibition*. Society of Petroleum Engineers.
39. Kitts, B. (1999). Representation operators and computation. *Minds and Machines*, 9(2), 223-240.
40. Legens, C., Toulhoat, H., Cuiec, L., Villieras, F., & Palermo, T. (1999). Wettability change related to adsorption of organic acids on calcite: Experimental and ab initio computational studies. *SPE Journal*, *4*(04), 328-333.
41. Li, S., Jackson, M. D., & Agenet, N. (2020). Role of the calcite-water interface in wettability alteration during low salinity waterflooding. *Fuel*, *276*, 118097.
42. Ligthelm, D. J., Gronsveld, J., Hofman, J., Brussee, N., Marcelis, F., & van der Linde, H. (2009, January). Novel waterflooding strategy by manipulation of injection brine composition. In *EUROPEC/EAGE conference and exhibition*. Society of Petroleum Engineers.
43. Mahani, H., Keya, A. L., Berg, S., Bartels, W. B., Nasralla, R., & Rossen, W. R. (2015). Insights into the mechanism of wettability alteration by low-salinity flooding (LSF) in carbonates. *Energy & Fuels*, *29*(3), 1352-1367.
44. Mahani, H., Menezes, R., Berg, S., Fadili, A., Nasralla, R., Voskov, D., & Joekar-Niasar, V. (2017). Insights into the impact of temperature on the wettability alteration by low salinity in carbonate rocks. *Energy & Fuels*, *31*(8), 7839-7853.
45. Mahmoud, A. A., Elkatatny, S., Al-AbdulJabbar, A., Moussa, T., Gamal, H., & Shehri, D. A. (2020, September). Artificial Neural Networks model for prediction of the rate of

penetration while horizontally drilling carbonate formations. In *54th US Rock Mechanics/Geomechanics Symposium*. American Rock Mechanics Association.

46. Meng, W., Haroun, M. R., Sarma, H. K., Adeoye, J. T., Aras, P., Punjabi, S., ... & Al Kobaisi, M. (2015, November). A novel approach of using phosphate-spiked smart brines to alter wettability in mixed oil-wet carbonate reservoirs. In *Abu Dhabi International Petroleum Exhibition and Conference*. Society of Petroleum Engineers.
47. Mirchi, V. (2018, September). Pore-Scale investigation of the effect of surfactant on fluid occupancies during low-salinity waterflooding in oil-wet carbonates. In *SPE Annual Technical Conference and Exhibition*. Society of Petroleum Engineers.
48. Mitchell, T. M. (1997). Machine learning.
49. Mohsenzadeh, A., Pourafshary, P., & Al-Wahaibi, Y. (2016, March). Oil recovery enhancement in carbonate reservoirs via low saline water flooding in presence of low concentration active ions; A case study. In *SPE EOR Conference at Oil and Gas West Asia*. Society of Petroleum Engineers.
50. Mohammadkhani, S., Shahverdi, H., & Esfahany, M. N. (2018). Impact of salinity and connate water on low salinity water injection in secondary and tertiary stages for enhanced oil recovery in carbonate oil reservoirs. *Journal of Geophysics and Engineering*, *15*(4), 1242-1254.
51. Nasralla, R. A., Sergienko, E., Masalmeh, S. K., van der Linde, H. A., Brussee, N. J., Mahani, H., ... & Alqarshubi, I. (2014, November). Demonstrating the potential of low-salinity waterflood to improve oil recovery in carbonate reservoirs by qualitative coreflood. In *Abu Dhabi International Petroleum Exhibition and Conference*. Society of Petroleum Engineers.
52. Nasralla, R. A., Mahani, H., van der Linde, H. A., Marcelis, F. H., Masalmeh, S. K., Sergienko, E., ... & Basu, S. (2018). Low salinity waterflooding for a carbonate reservoir: Experimental evaluation and numerical interpretation. *Journal of Petroleum Science and Engineering*, *164*, 640-654.
53. Okasha, T. M., & Alshiwaish, A. (2009, January). Effect of brine salinity on interfacial tension in Arab-D carbonate reservoir, Saudi Arabia. In *SPE Middle East oil and gas show and conference*. Society of Petroleum Engineers.
54. Puntervold, T. (2008). Waterflooding of carbonate reservoirs: EOR by wettability alteration.
55. Ravari, R. R. (2011). Water-Based EOR in Limestone by Smart Water: A study of surface chemistry.

56. Romanuka, J., Hofman, J., Ligthelm, D. J., Suijkerbuijk, B., Marcelis, F., Oedai, S., ... & Austad, T. (2012, January). Low salinity EOR in carbonates. In *SPE Improved Oil Recovery Symposium*. Society of Petroleum Engineers.
57. Samanova, A. (2021). Surfactant/LSW flooding in carbonates: an investigation of hybrid EOR method design to improve oil displacement.
58. Schuetter, J., Mishra*, S., Zhong, M., & LaFollette, R. (2015, July). Data analytics for production optimization in unconventional reservoirs. In *Unconventional Resources Technology Conference, San Antonio, Texas, 20-22 July 2015* (pp. 249-269). Society of Exploration Geophysicists, American Association of Petroleum Geologists, Society of Petroleum Engineers.
59. Sekerbayeva, A., Pourafshary, P., & Hashmet, M. R. (2020). Application of anionic Surfactant\Engineered water hybrid EOR in carbonate formations: An experimental analysis. *Petroleum*.
60. Shehata, A. M., Alotaibi, M. B., & Nasr-El-Din, H. A. (2014). Waterflooding in carbonate reservoirs: does the salinity matter?. *SPE Reservoir Evaluation & Engineering*, *17*(03), 304-313.
61. Tetteh, J. T., Rankey, E., & Barati, R. (2017, October). Low salinity waterflooding effect: Crude oil/brine interactions as a recovery mechanism in carbonate rocks. In *OTC Brasil*. Offshore Technology Conference.
62. Tetteh, J., Janjang, N. M., & Barati, R. (2018, August). Wettability alteration and enhanced oil recovery using low salinity waterflooding in limestone rocks: a mechanistic study. In *SPE Kingdom of Saudi Arabia Annual Technical Symposium and Exhibition*. Society of Petroleum Engineers.
63. Tetteh, J. T., & Barati, R. (2019). Crude-oil/brine interaction as a recovery mechanism for low-salinity waterflooding of carbonate reservoirs. *SPE Reservoir Evaluation &* Engineering, 22(03), 877-896.
64. Vapnik, V., Guyon, I., & Hastie, T. (1995). Support vector machines. *Mach. Learn*, 20(3), 273-297.
65. Venna, A. R., Ang, Y. Y., Nguyen, N., Lu, Y., & Walters, D. (2018). Support-vector-machine phase classification of downhole leak flows based on acoustic signals. *Petrophysics*, *59*(06), 841-848.
66. Vo, L. T., Gupta, R., & Hehmeyer, O. J. (2012, January). Ion chromatography analysis of advanced ion management carbonate coreflood experiments. In *Abu Dhabi International Petroleum Conference and Exhibition*. Society of Petroleum Engineers.

67. Wang, L., & Fu, X. (2018). Data-driven analyses of low salinity water flooding in sandstones. Fuel, 234, 674-686.
68. Wang, L., Tian, Y., Yao, B., & Yu, X. (2020). Machine learning analyses of low salinity effect in sandstone porous media. Journal of Porous Media, 23(7).
69. Winoto, W., Loahardjo, N., Xie, S. X., Yin, P., & Morrow, N. R. (2012, January). Secondary and tertiary recovery of crude oil from outcrop and reservoir rocks by low salinity waterflooding. In *SPE Improved Oil Recovery Symposium*. Society of Petroleum Engineers.
70. Yi, Z., & Sarma, H. K. (2012, January). Improving waterflood recovery efficiency in carbonate reservoirs through salinity variations and ionic exchanges: A promising low-cost" Smart-Waterflood" approach. In *Abu Dhabi International Petroleum Conference and Exhibition*. Society of Petroleum Engineers.
71. Yousef, A. A., Al-Saleh, S. H., Al-Kaabi, A., & Al-Jawfi, M. S. (2011). Laboratory investigation of the impact of injection-water salinity and ionic content on oil recovery from carbonate reservoirs. *SPE Reservoir Evaluation & Engineering*, *14*(05), 578-593.
72. Yousef, A. A., Liu, J., Blanchard, G., Al-Saleh, S., Al-Zahrani, T., Al-Tammar, H., & Al-Mulhim, N. (2012). SmartWater flooding: industry’s first field test in carbonate reservoirs, SPE-159526 presented at the SPE Annual Technical Conference and Exhibition held in San Antonio, Texas, USA, 8–10 October.
73. Yousef, A. A., Al-Saleh, S., & Al-Jawfi, M. S. (2012, January). Improved/enhanced oil recovery from carbonate reservoirs by tuning injection water salinity and ionic content. In *SPE improved oil recovery symposium*. Society of Petroleum Engineers.
74. Zhang, P., & Austad, T. (2006). Wettability and oil recovery from carbonates: Effects of temperature and potential determining ions. *Colloids and Surfaces A: Physicochemical and Engineering Aspects*, *279*(1-3), 179-187.