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## A Systematic Literature Review on Interfacial Tension and Zeta Potential Analyses Behind Smart Waterflooding

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### Abstract

Smart waterflooding is a widely discussed topic for oil recovery improvement, with the main mechanism believed to be wettability alteration. However, the exact phenomena behind this effect are still being debated. This systematic literature review (SLR) utilized the Scopus database to analyze and assess studies on the interfacial tension (IFT) and zeta potential (ZP) concepts and their impact on smart waterflooding between 2017 and 2021. The review was conducted using the Preferred Reporting Items for Systematic Reviews and Meta-Analyses (PRISMA) methodology. The review found inconsistencies in the literature, with some studies suggesting that the reduction of IFT from smart waterflooding was not the reason for oil recovery improvement. Further evidence on the alteration of surface charge through ZP analysis is needed, and gaps are present in the area of simulation approach. The study suggests that improvements in model development and experimental setup design are necessary for future studies to obtain comprehensive conclusions. In summary, this SLR provides a critical overview of the recent understanding of the impact of IFT and ZP on smart waterflooding and highlights areas for further research, including a potential model that incorporates each mechanism with the ultimate oil recovery.

## INTRODUCTION

Smart waterflooding has attracted much attention of academia and industry to be beneficial in producing more oil. The performance of smart waterflooding to enhance oil recovery was potentially superior to the conventional seawater injection (Al-Shalabi & Sepehrnoori, 2016). Moreover, a relatively low cost and environmental benefits are added value of smart waterflooding making it favorable compared to other enhanced oil recovery (EOR) methods such as polymer or surfactant flooding (Ding et al., 2019). The term of smart water refers to the modification of different types of salt concentration that disturbs the original equilibrium in a reservoir, and smart water can be injected either in the secondary or tertiary recovery process (Maghsoudian et al., 2020). Smart waterflooding is often known as low salinity waterflooding (LSWF) for sandstone rocks (Afekare & Radonjic, 2017; Bartels et al., 2019; Sohal et al., 2016). However, smart waterflooding in carbonates means injecting predesigned brine with altered ion content from the former injected fluid which will result in the production of additional oil (Hao et al., 2019). Even though, injecting low salinity water into carbonate reservoir is possible (Derkani et al., 2018), adjusting particular salt content, i.e., magnesium, calcium, sulfate, and other ions are also plausible (Hao et al., 2019).

The common understanding of LSWF positive effect is because of the interaction between injected brine and clay minerals (Bernard, 1967). In contrast, the clay content in carbonate rocks is very low or even zero (Lager et al., 2008), thus LSWF was not considered feasible EOR method for carbonate. However, the modification of injected water composition in a process called controlled salinity water-flooding (CSW) can increase the oil recovery from carbonate reservoir by changing the mineral-brine ZP that is of identical polarity as the crude-oil brine interface (Collini et al., 2020; Jackson et al., 2016). The wettability of

sandstone reservoir are mostly strong water-wet or mixed-wet, while carbonate reservoirs are either completely oil-wet or mixed-wet (Chilingar & Yen, 1983; Treiber & Owens, 1972). Smart waterflooding affects the interaction between rock fluid and fluids in oil reservoirs to alter the wettability towards more water-wet and optimize the oil gain (Maghsoudian et al., 2020). Proposed mechanisms have to describe the reason behind the positive outcome of the smart waterflooding on the oil production include the multicomponent ionic exchange, double layer expansion, reduce the IFT, increase in pH to in-situ saponification, alter the surface charge, wettability alteration and fines migration (Etemadi et al., 2017; Lashkarbolooki et al., 2014, 2017; Maghsoudian et al., 2020; Rezaei Gomari & Hamouda, 2006; Zhang et al., 2007).

The present study highlights the mechanism related to IFT and ZP in evaluating the effect of smart waterflooding to increase oil recovery. To the best of our knowledge, there still no published paper of a systematic literature review (SLR) that is written by applying the Preferred Reporting Items for Systematic Reviews and Meta-Analysis (PRISMA) method regarding smart waterflooding, particularly the interfacial interaction between oil/rock/brine. SLR is a review of a clearly formulated question that uses systematic and explicit methods to identify, select, and critically appraise relevant research, and to collect and analyze data from the studies that are included in the review (Moher et al., 2009). SLR is selected because it summarizes and synthesizes the finding of existing literature on a research topic or field. According to a report by Donthu and colleagues in 2021, SLR is suitable to be used when the scope of the review is specific, dataset is small and manageable enough that its content can be manually reviewed (Donthu et al., 2021). Meanwhile, PRISMA is a tool to help authors improving the reporting of systematic reviews and meta-analyses (Moher et al., 2009). PRISMA is applied because it serves as the basis for reporting systematic reviews of other types of research, particularly evaluations of interventions (Moher et al., 2009). The existing publications comprises of numbers of experimental studies and review papers consisting all the possible mechanisms behind the increase of oil recovery (Afezare & Radonjic, 2017; Al-Shalabi & Sepehrnoori, 2016; Bartels et al., 2019; Derkani et al., 2018; Hao et al., 2019; Purswani et al., 2017; Sohal et al., 2016). Although these reviews have provided comprehensive information on smart waterflooding, focusing on the oil/rock/brine interfaces through IFT, ZP, and interfacial rheology will provide clearer understanding on interfacial interaction between rock, oil, and water.

A well-known characteristic of water-wet reservoir pores is the presence of a thin layer of water that leads to the indirect adhesion of oil to the rock surface (Tetteh et al., 2020). In some pore spaces, oil molecules can directly adhere to the pore surface over time, leading to a mixed-wet condition (Brady & Thyne, 2016). This means that some pores are strongly oil-wet, while others are either intermediate or water-wet, based largely on the heterogeneous nature of the rock and the differing surface properties. The water film has an impact on the wetting condition of the rock, either by making it thicker or thinner, depending on the balance of forces at the crude oil-brine-rock (COBR) interface.

The DLVO theory is employed to relate the electrokinetics of the rock surface to the thermodynamic interactions involved in altering wettability and the function of the water film (Brady & Thyne, 2016; Ding & Rahman, 2017; Hirasakl, 1991; Mahani et al., 2017). This theory takes into account the total disjoining pressure, which is the combination of van der Waals forces, electrostatic or electrical double layer (EDL) forces, and structural forces. A negative total disjoining pressure indicates an attractive force between the rock and oil surfaces, which leads to the adhesion of oil molecules to the rock, resulting in a thin water film. This enhances the direct interaction between the polar and asphaltene components of crude oil and the rock surface, making it more oil-wet. Conversely, a positive total disjoining pressure represents a repulsive force between the oil and rock surfaces and results in a thick and continuous water film, making the rock water-wet.

Carbonate surfaces have a positive charge in brine with pH values lower than 8-9 (Ding & Rahman, 2017; Hirasakl, 1991). Crude oil particles, which are negatively charged due to the presence of carboxylic, stearate, and fatty acids (Mahani et al., 2015, 2016), interact with carbonates and generate an attractive electrostatic force at the COBR interface, leading to oil-wetness. Changing the wettability of carbonates from oil-wetness to intermediate or water-wetness is advantageous for improving oil recovery. The mechanisms of altering wettability at the COBR interface of carbonates, which are influenced by the DLVO theory, can be divided into three categories: 1) Expansion of the EDL and variation in surface charge, 2) Multivalent ionic exchange (MIE), and 3) Electrostatic bond linkages.

Typically, when the ionic strength decreases, the EDL expands, leading to increased water wetting due to the greater distance between oil and calcite (Alotaibi et al., 2011, 2018; Alotaibi & Nasr-El-Din, 2011; Alotaibi & Yousef, 2016; Mahani et al., 2015, 2016). It's important to note that a charged colloidal particle dispersed in water creates two charged layers on its surface, known as the EDL. The inner layer, which consists of counterions balancing the colloid charge, is referred to as the Stern layer and is strongly attached

(Chilingar & Haroun, 2014). The outer layer, composed of loosely bound ions, is called the diffuse layer (Chilingar & Haroun, 2014). Zeta potential is used to measure the thickness of the EDL. When both the crude oil-brine and rock-brine interface have the same polarity, a force of repulsion forms between the oil and rock surface. This leads to the enlargement of the EDL at both interfaces, causing the water film to thicken and the wettability to change from oil-wet to water-wet (Afekare & Radonjic, 2017; Ding & Rahman, 2017; Hirasaki, 1991; Myint & Firoozabadi, 2015). A decrease in the salinity of the brine can result in a decrease in the attractive force at the crude oil-brine interface, resulting in an increased Debye length and a change in rock wettability.

McGuire and colleagues in 2005 suggested that the increment in oil production caused by the injection of low-salinity brine leads to a rise in brine pH, causing a change in wettability (McGuire et al., 2005). This rise in pH would result in a decrease in the IFT between the brine and oil, thus increasing the capillary number and enhancing oil recovery. Tetteh and colleagues in 2018 stated that the lowest IFT between crude oil and brine was found within a salinity range that corresponds to seawater (30,000 ppm) (Tetteh & Barati, 2018). This was explained by the existence of natural surface-active agents in the crude oil that concentrate at the brine/oil interface and interact with divalent ions, such as  $\text{SO}_4^{2-}$ , which forms a stable interface, leading to a decrease in IFT. The creation of small oil clusters due to oil snap-off and phase entrapment in large pores would decrease sweep efficiency and thus hinder improved oil recovery (IOR). A decrease in IFT can improve oil recovery by increasing surface elasticity and preventing snap-off at the crude oil/brine interface (Chávez-Miyauchi et al., 2016; Garcia-Olvera & Alvarado, 2017; Tetteh & Barati, 2018). Mokhtari and colleagues concluded, based on their observation and other research, that the dissociation of polar components in crude oil into the brine solution caused a decrease in solution pH and IFT, making it the primary fluid-fluid interaction mechanism responsible for the IOR observed with LSWF (Mokhtari et al., 2019; Mokhtari & Ayatollahi, 2019).

In this review, we conduct systematical review on exploring the contribution of IFT and ZP to bring effect during smart waterflooding, observing the methodology of examining the relation between oil/brine/rock interfaces to the oil recovery improvement, summarizing different point of view of discussing IFT and ZP in smart waterflooding from the collected previous publications for the past five years, i.e., 2017 to 2021, studying the limitations, and offering suggestions for further development. This review focuses on two main research questions as follows: (1) How does the IFT is measured and being discussed to have an impact on the fluid/fluid interface under smart waterflooding during these five years and (2) How does the ZP is measured and being discussed to have an impact on the of fluid/rock or fluid/fluid interface under smart waterflooding during these five years. The aim of this review is to obtain the status of understanding the IFT and ZP as the potential contributing factor to enhance oil recovery in a SLR.

## **MATERIALS AND METHOD**

This study constructed a SLR on the contribution of IFT and ZP in smart waterflooding effect. This study was designed by following guidelines of the PRISMA checklist (Moher et al., 2009). It is essential to define research questions to be responded in this study, thus assuring the clarity of the research objective. The data mining source was explained including the selection criteria for further analysis.

### **A. Search Strategy**

The search strategy was conducted by visiting Scopus database. Applying smart waterflooding as the keyword focused the search onto the study area. Both review and research articles were taken to support the aim of this study. The articles must be indexed by Scopus because it has been proven to be a reliable source. The data were being gathered from October to November 2021.

The method to define whether a research article or review article suited the inclusion criteria was begin by collecting the list of the article titles from Scopus database with applied keyword and transferring the list to Mendeley. The inclusion criteria started from narrowing the publication year into 2017 to 2021 to obtain the current status of understanding in regards to the topic. The process was continued by screening the title based on the source title. Only articles form journal source were considered.

### **B. Relevance and quality appraisal**

The title search from Scopus database referring the keyword and data collection were completed by using Mendeley. The publication years and the titles then were being scanned by one reviewer to define the suitability of the title to the journal source criteria which are published from 2017 to 2021. One reviewer appraised the articles thoroughly. The quality assessment instrument examined the fundamental background and objective, sample selection, methodology, interpretation of results and major findings. The valuation of publication bias was not performed because the absence of combined effect size in this

systematic review. Publication bias is defined as the failure to publish the results of a study on the basis of the direction or strength of the study findings. This may mean that only studies which have statistically significant positive results get published and the statistically insignificant or negative studies does not get published. Of the several reasons of this bias the important ones are rejection (by editors, reviewers), lack of interest to revise, competing interests, lack of motivation to write in spite of conducting the study (Nair, 2019). Meanwhile, in statistics, an effect size is a value measuring the strength of the relationship between two variables in a population, or a sample-based estimate of that quantity. The criteria to decide the paper to be selected for synthesis consist of availability of IFT and/or ZP measuring technology and/or clear definition of IFT and/or ZP.

### C. Data extraction and analysis

The inclusion and quality criteria extracted the data from the collected articles by using a data collection table. The data presented in the table include references, smart water variation, oil sample, methodology, and main finding (Table I and II). In particular, the table for ZP analysis have additional parameter of system and rock types. A descriptive synthesis of the results and tabulated data system were used to summarize the strength of evidence around the different effect of IFT and ZP in smart waterflooding context.

## RESULTS AND DISCUSSION

The results from the article search and selection are introduced in Figure 1. The article search collected 52 distinctive titles to screen for relevance and each abstracts were subsequently selected for inclusion. 34 studies match the inclusion criteria and complete manuscript were gathered for examination. 23 studies were then omitted, leaving 11 studies for further appraisal. The primary causes for exclusion for each study were the absence of IFT and/or ZP measurement and discussions.

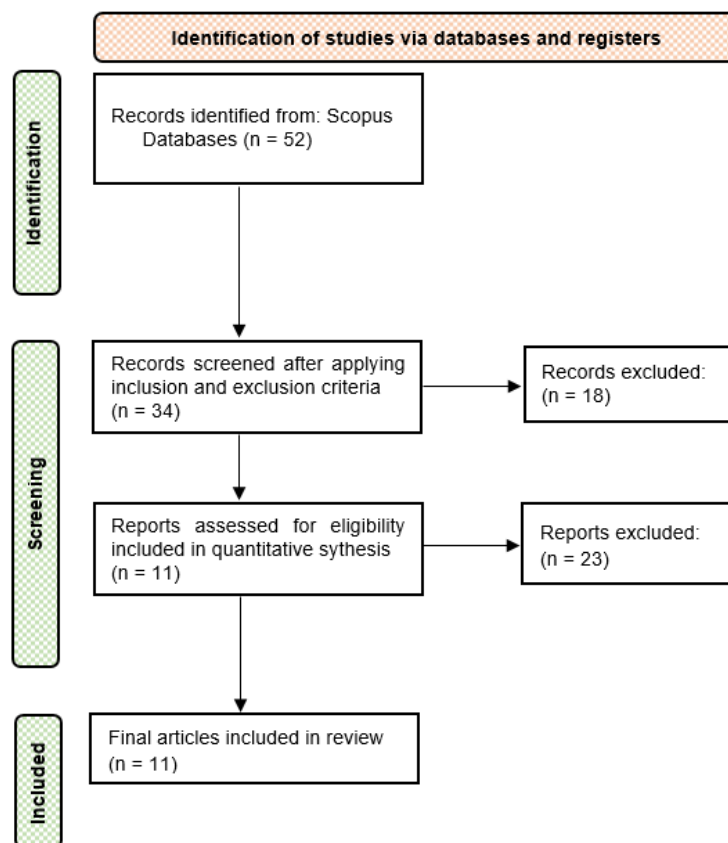


Figure 1. The article selection strategy

### A. Study characteristics

All 11 articles selected were descriptive and covered the IFT and ZP related topic in smart waterflooding. The investigated topics of IFT and ZP were presented in 40% and 60% of the total included articles, respectively. 4 studies for IFT and 3 studies for ZP were conducted in the laboratory with measuring apparatus by applying different ion type and concentration within the water, rock samples, and crude oil. A

study was a review of the mechanism of smart waterflooding in carbonate which has IFT and ZP as discussion topic. One study used numerical approach to analyze ZP. The remaining of the studies did not mention the methodology of measuring IFT and/or ZP, but the analysis related to IFT and/or ZP were available.

B. How does the IFT is measured and being discussed to have an impact on the fluid/fluid interface under smart waterflooding during these five years?

6 out of the 11 included studies shared the analysis of IFT in smart waterflooding (Ayirala, Al-Yousef, et al., 2018; Garcia-Olvera & Alvarado, 2017; García & Saraji, 2021; Hao et al., 2019; Maghsoudian et al., 2020; Wang et al., 2020). 4 studies mentioned pendant drop analysis to explain the IFT between oil and brine in smart waterflooding (Ayirala, Al-Yousef, et al., 2018; García & Saraji, 2021; Maghsoudian et al., 2020; Wang et al., 2020). Most of the studies used crude oil sample from the field to analyze the IFT (Ayirala, Al-Yousef, et al., 2018; Garcia-Olvera & Alvarado, 2017; García & Saraji, 2021; Hao et al., 2019; Maghsoudian et al., 2020), however one study employed toluene with different amounts of stearic acid in brine-oil systems (Wang et al., 2020). In 3 studies the analysis of IFT was supported by the interfacial rheology analysis (Ayirala, Al-Yousef, et al., 2018; García & Saraji, 2021; Wang et al., 2020). 1 study observed the effect of IFT on the water salinity (García & Saraji, 2021), whereas the other studies focused on specific anion or cation content in the water (Maghsoudian et al., 2020; Wang et al., 2020). There were also studies investigating the effect of both salinity and individual ion (Ayirala, Al-Yousef, et al., 2018; Garcia-Olvera & Alvarado, 2017; Hao et al., 2019). Two studies proved the validity of the Young-Laplace equation and oscillatory pendant-drop measurements on crude oil-brine systems (Ayirala, Al-Yousef, et al., 2018; García & Saraji, 2021). Hao and colleagues (Hao et al., 2019) reviewed the mechanism of smart waterflooding on carbonate. One of the topic was regarding IFT measurement which taken from the collected previous works from 2010 to 2017.

Ayirala and colleagues in 2018 and Wang and colleagues in 2020 examined the brine ion interactions at the interface of oil and water through crude-oil-droplets coalescence time that influenced by the rigidity of interfacial film (Ayirala, Al-Yousef, et al., 2018; Wang et al., 2020). The low IFTs observed with modified brine in comparison with de-ionized (DI) water indicate the promotion of surface-active component in the crude oil (asphaltenes and resins) transfer to the crude oil/water interface by the addition of salts (Ayirala, Al-Yousef, et al., 2018; Maghsoudian et al., 2020). The amount of surface-active component in the oil defines the coalescence time of oil droplets, where the more the amount, the longer the time, due to the decrease of IFT (Wang et al., 2020). The electrostatic force at the interface explained the IFT value variation as a result of ion concentration through Debye length (García & Saraji, 2021; Maghsoudian et al., 2020). High ion concentration thinned the Debye length, thus resulting in higher gradient of electrostatic potential that led to faster surface-active component transfer to the interface and caused IFT to decrease (García & Saraji, 2021). The interfacial rheology was viewed as an assessment tool for ion and crude oil interaction through snap-off and coalescence (Garcia-Olvera & Alvarado, 2017; García & Saraji, 2021). Favorable interface elasticity suppressed the occurrence of snap-off and increased oil recovery (Garcia-Olvera & Alvarado, 2017). Sulfate increased the rigidity of the interfacial film thus hindered the coalescence between oil droplets (Ayirala, Al-Yousef, et al., 2018) and might favored the elasticity of the interface thus depressed snap-off (Garcia-Olvera & Alvarado, 2017). The summary of the IFT studies was tabulated in Table I.

**Table I Summary of IFT study in smart waterflooding**

References	Smart water	Oil	Methodology	Discussions
(Ayirala, Al-Yousef, et al., 2018)	Salinity and ion variations	Field crude oil	Pendant drop analysis, dynamic and static measurement	Salts promote the transfer of surface-active components in the crude oil to the interface that indicated by the low measured IFT.
(Maghsoudian et al., 2020)	Ion variations	Field crude oil	Pendant drop analysis	Excess amount of divalent cations shows better impact on IFT reduction.
(García & Saraji, 2021)	Salinity variations	Field crude oil	Pendant drop and oscillating pendant drop	IFT reduces rapidly as the transfer of polar component to the interface becomes faster at higher ion concentration.
(Garcia-Olvera & Alvarado, 2017)	Salinity and ion variations	Field crude oil	N/A	Sulfate favor the elasticity of the interface, therefore the occurrence of

					snap-off can be depressed and oil recovery increased.
(Wang et al., 2020)	Ion variations	Toluene with stearic acid	Dynamic drop	pendant	Surface-active component stearic acid content in oil results in a longer coalescence time.
(Hao et al., 2019)	Salinity and ion variations	Reservoir live oil	Review study		The IFT reduction was too small to impact the oil recovery.

N/A: Not available

C. How does the ZP is measured and being discussed to have an impact on the of fluid/rock or fluid/fluid interface under smart waterflooding during these five years?

7 studies was conducted to analyze zeta potential during smart waterflooding. Electrophoretic mobility was mainly used as the principal in measuring zeta potential (Abbasi et al., 2020; Ayirala, Yousef, et al., 2018; Ding et al., 2019). The preparation of the sample conducted by several methods such as sonicating (Ding et al., 2019), magnetic oscillator stirring (Ding et al., 2019), and multi-wrist shaking (Maghsoudian et al., 2020). According to the reviewed studies, there are studies which measured zeta potential in brine/rock system (Ding et al., 2019; Hao et al., 2019; Maghsoudian et al., 2020; Namaee-Ghasemi et al., 2021; Shaik et al., 2020) and also in oil/brine system (Ayirala, Yousef, et al., 2018; Ding et al., 2019). Calcite powder (Maghsoudian et al., 2020), Austin chalk powder (Ding et al., 2019), Iranian carbonate (Abbasi et al., 2020), Iceland Spar calcite (Shaik et al., 2020), carbonate (Hao et al., 2019; Namaee-Ghasemi et al., 2021) were combined with smart water solutions to measure brine/rock system zeta potential. Adding mineral oil onto smart water will provide oil-brine emulsion for oil/brine system zeta potential (Ding et al., 2019). Zeta potential measurements are conducted on rock/brine/oil systems to analyze the chemical composition and surface charge of rock powders dispersed in various brines. The rock powders were kept in crude oil for a duration of one month in a laboratory cell, at a temperature of 80 °C and pressure of 1000 psi (Abbasi et al., 2020). The surface charges of the aged powders were evaluated by dispersing them in various brines to examine the impact of brine chemistry and ion composition (Abbasi et al., 2020). The results of the zeta potential measurements on the brine/powder samples showed negative values. The tested brines were a mixture of seawater and river water, and they underwent ion modification. This study measured the electrophoretic mobility of carbonate particles dispersed in the specified brine using the Zetasizer Nano ZS apparatus. The zeta potential, which reflects the value of electrophoretic mobility, was recorded for each sample. The combinations of carbonate particles and brine were subjected to 20 minutes of sonication and then given a two-day period to settle in order to achieve equilibrium (Abbasi et al., 2020). One study implemented simulation approach to examine the contributing parameter on wettability alteration by observing surface charge change (Namaee-Ghasemi et al., 2021). The extraction of oil using four brines of varying levels of salinities was successful, and the chemical composition of the resulting liquids was analyzed separately. The concentrations of the extracted substances in the simulation matched the actual results, indicating a satisfactory outcome. The concentrations of  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$  indicated the dissolution of Calcite and Dolomite in all cases. When seawater was used, Calcite dissolution was dominant in higher salinities, causing an underestimation of  $\text{Ca}^{2+}$  and an overestimation of  $\text{Mg}^{2+}$ . Lowering the salinity to 5000 ppm resulted in similar  $\text{Mg}^{2+}$  concentrations in both the experiment and the simulation, indicating a higher contribution of Dolomite dissolution. Reducing the salinity to 2000 ppm led to a decrease in  $\text{SO}_4^{2-}$  concentration and an overestimation of  $\text{Mg}^{2+}$ , which indicates the effect of the surface charge change mechanism in lower salinities. The review study by Hao and colleagues in 2019 highlighted the topic of the change of surface charge through interaction between the double layer and the surface complexation, zeta potential, effect of pH, and effect of non-potential determining ions (Hao et al., 2019). In particular, the discussion of zeta potential were drawn from previous publications from the year of 2006 to 2016.

Discussion of ZP is closely related with the surface charge (Abbasi et al., 2020; Maghsoudian et al., 2020; Namaee-Ghasemi et al., 2021; Shaik et al., 2020), where the change of surface charge that is obtained from ZP measurement can be used as a guide to assess wettability alteration toward water-wet state. ZP value is affected by the presence of ion in the water and salinity. ZP chalk was made more negative by reducing  $\text{Ca}^{2+}$  or  $\text{Mg}^{2+}$  concentration or raising  $\text{SO}_4^{2-}$  concentration in the water (Ding et al., 2019; Maghsoudian et al., 2020). The zeta potential tests in the research carried out by Maghsoudian and colleagues in 2020 were conducted under room temperature conditions, and the preparation of the solid/liquid solution required 72 hours of mixing (Maghsoudian et al., 2020). The surface charge density of calcite increases (positive)

with NaCl concentration which then allow adsorption of negatively charged naphthenic acid content in the oil, thus create oil-wet condition (Shaik et al., 2020). When the salinity of the brine is reduced, the zeta potential becomes negative. The expansion of the EDL in low total dissolved solids (TDS) results in a sparser distribution of ions on the surface and facilitates the diffusion of active ions (Abbasi et al., 2020). The ZP becomes negative at lower salinity brine due to the behavior of the EDL around the particles in the brine. When there are fewer dissolved salts (i.e., lower salinity), the EDL surrounding the particles in the brine is less dense and more spread out, which allows the particles to repel each other, and the ZP becomes negative. In contrast, at higher salinity, the EDL is more compact and crowded with ions, leading to an attraction between particles and a positive ZP. This phenomenon is due to the changes in the electrostatic interactions between the particles and the ions in the solution, which affect the ZP. The interfacial rheology was also discussed with ZP of oil droplets, where larger magnitude of ZP negatively charged oil droplets in brine indicated stronger electrostatic repulsion between droplets (Ayirala, Yousef, et al., 2018). The studies regarding ZP from the reviewed paper were summarized in Table II.

**Table II Summary of ZP study in smart waterflooding**

<b>References</b>	<b>System: (rock, smart water, oil)</b>	<b>Methodology</b>	<b>Discussions</b>
(Ding et al., 2019)	Brine/rock, oil/brine: (Austin chalk, Ion variations, Light mineral oil)	Electrophoretic mobility	Reducing $Ca^{2+}/Mg^{2+}$ concentration or raising $SO_4^{2-}$ concentration in seawater makes ZP chalk becomes more negative, whereas the change of ZP oil due to variation of $Ca^{2+}$ , $Mg^{2+}$ , and $SO_4^{2-}$ concentration in seawater is minor.
(Abbasi et al., 2020)	Oil/brine/rock: (Iranian carbonate, Salinity and ion variations, Field crude oil)	Electrophoretic mobility	A decrease in the salinity of the brine results in a negative zeta potential. This can be explained by the fact that at lower total dissolved solids (TDS), the EDL expands and becomes less densely populated with ions, resulting in a facilitation of active ions diffusion.
(Ayirala, Yousef, et al., 2018)	Oil/brine: (N/A, Ion variations, Stock tank crude oil)	Electrophoretic mobility	The ZP oil in different brines is the indication of strength of electrostatic repulsion between oil droplets that influence the thickness of water film.
(Maghsoudian et al., 2020)	Brine/rock: (Calcite, Ion variations, Field crude oil)	N/A	The negatively charge increases with excess amount of $SO_4^{2-}$ due to the disruption of the balance between divalent anions and cations, which led to a significant adsorption of $SO_4^{2-}$ on the surface of calcite. The strong interaction between the negatively charged $SO_4^{2-}$ and the positively charged surface of calcite ( $Ca^{2+}$ ) in the Stern layer, along with better chemical bonding, caused this adsorption. As a result, these cations strongly adhered to the calcite surface rather than to the oil components.
(Namaee-Ghasemi et al., 2021)	Brine/rock: (Carbonate, Salinity and ion variations, Field crude oil)	Numerical simulation	The surface charge changes actively when the brine salinity is lowered. The mechanism of surface charge change is contributing more significantly when the value of $C/C_0$ for $Mg^{2+}$ quickly becomes one, suggesting that $Mg^{2+}$ is adsorbed on the rock surface and causing a change in surface charge. Additionally, there is a decreasing trend in the concentration of $SO_4^{2-}$ , indicating that it is being increasingly absorbed by the surface. The study utilized a geochemical engine, which is a tool that performs batch reaction equilibrium

			calculations. This tool is integrated with the simulator UTCHEM and is used for a variety of purposes, including evaluating compatibility, determining equilibrium composition, and calculating pH values for the mixing of different aqueous solutions. However, certain parameters such as surface charges, EDL expansion, near surface diffusion rates, and dispersion in porous media were not considered in UTCHEM. The focus of the study was instead to observe the general behaviors of individual ions and gain a better understanding of the underlying mechanisms of LSE.
(Shaik et al., 2020)	Brine/rock: (Iceland Spar calcite, Salinity and ion variations, Model oil (n-dodecane and toluene))	N/A	Surface charge density that is influenced by NaCl dictates the adsorption of negatively charged naphthenic and desorption of naphthenic acid, consequently the wettability alteration.
(Hao et al., 2019)	Brine/rock: (Carbonate, Salinity and ion variations, N/A)	Review study	Reduction of oil adsorption on mineral surface having negative value of zeta potential by repelling the negatively charged carboxylic groups.

#### D. Discussion

IFT between oil and water is important parameter in producing oil through enhanced oil recovery. Several studies have discovered that smart waterflooding affects the IFT between water/oil and oil droplets interfaces (Al-Attar et al., 2013; Alameri et al., 2014; Ayirala, Al-Yousef, et al., 2018; Garcia-Olvera & Alvarado, 2017; García & Saraji, 2021; Hao et al., 2019; Khaksar Manshad et al., 2016; Maghsoudian et al., 2020; Mahani et al., 2015; Wang et al., 2020; Yousef et al., 2012; Zhang & Austad, 2005). IFT decreases with the increase of ion concentrations in the brine (Garcia-Olvera & Alvarado, 2017; Maghsoudian et al., 2020).

Oil has natural surfactant component namely asphaltene and resin (Ayirala, Al-Yousef, et al., 2018; Behrang et al., 2021; García & Saraji, 2021; Hasiba & Jessen, 1968; Maghsoudian et al., 2020; Shepherd et al., 2005; Sheu & Shields, 1995; Varadaraj & Brons, 2012; Wang et al., 2020). Ions in the water triggered the transfer of asphaltene and resin within the oil to the oil and water interface. The concept of IFT reduction by modifying the ion in the smart waterflooding is slightly different in the surfactant flooding. Surfactant is commonly utilized and proven effectively to reduce the IFT between oil and formation water (Riswati et al., 2020). However, surfactant is expensive and results in complicated chemical reaction when interacting with formation water, oil, and rock surface. Oil droplets coalescence time is longer due to rigidity of the interfacial film that hinder coalescence between oil droplets (Ayirala, Al-Saleh, et al., 2018; Ayirala, Al-Yousef, et al., 2018; Wang et al., 2020). Beside, increase of the electrostatic repulsion of the head group of fatty acid in the oil also delay the coalescence of oil droplets (Ayirala, Yousef, et al., 2018; Wang et al., 2020).

The interface between two contacting fluids may form elastic (solid-like) or viscous (liquid-like) interface or intermediate states which is attributed to the surface active component in the oil (Bidhendi et al., 2018; Criddle & Meader, 1955; García & Saraji, 2021; Hao et al., 2019). Elastic interface eases the desorption of oil from the rock surface, thus elastic interface is favorable near the surface while the ion causing viscous interface should stay dissolve in the solution. An increase of interfacial viscoelasticity between oil and water due to injection of modified brine minimizes the oil trapping by snap-off, thus create continuous oil phase and greater oil recovery (Garcia-Olvera & Alvarado, 2017).

Reducing the number of divalent ions, particularly  $Mg^{2+}$ , increased IFT (Behrang et al., 2021; Maghsoudian et al., 2020). The atomic radius of  $Mg^{2+}$  is smaller than  $Ca^{2+}$ , thus higher charge density. Therefore,  $Mg^{2+}$  interacts effectively with surface active agent in the crude oil, i.e., asphaltene and resin. Moreover,  $SO_4^{2-}$  resulted in less significant effect to the IFT compared to divalent ions (Maghsoudian et al., 2020). It tends to chemically interact with the rock surface. Meanwhile, the IFT is increased for lower salinity brines.

The effect of ion on the IFT trend is different according to the range of pH level (Behrang et al., 2021; Shepherd et al., 2005). At pH 3.5 and 7, divalent cation,  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$ , played important role in reducing IFT, more than monovalent cation ( $\text{K}^+$  and  $\text{Na}^+$ ). Conversely, monovalent cation showed better efficiency in reducing IFT at pH 11 compare to divalent cations. The molecular size of divalent cation is bigger than monovalent cation, so that monovalent cation is easily positioned between the branches of resin molecules and decrease the surface charge of resin. Resulting in more stable state thus lowering repulsive force while entering interface.

An experimental study on naphthenate deposits in the mixture of aqueous solution with crude oil containing naphthenic acid carried on by Shepherd and colleagues in 2005 explained that the reason for the reduction in IFT with pH for the  $\text{Na}^+$  solution can be attributed to the abundance of cations in the aqueous phase. This abundance of cations leads to a decrease in the number of  $\text{H}^+$  ions (counterions) at the interface, which in turn increases the tendency of acidic groups to ionize (Shepherd et al., 2005). The cause of higher IFT for calcium solution is due to surface inactivity as a result of contribution of calcium in the precipitation or binding with the naphthenic acid at higher pH, which makes them surface inactive (Cooke Jr. et al., 1974; Goldszal et al., 2002; Healy & White, 1978; Mohammed & Sorbie, 2010; Spildo & Høiland, 1999)). A study by Shirazi and colleagues (Shirazi et al., 2019) proved that the use of nanoparticle to the smart water shows diverse IFT reduction behavior. Diluting smart water to improve nanoparticle stability gave negligible change in wettability alteration, but drastic change in IFT reduction, that contribute to enhancing the oil recovery.

The concept of IFT is proportionally related to the coalescence of oil droplets where the coalescence time is shorten by designing magnesium and calcium ion amount in the water, lowering brine salinity and decreasing pH (Ayirala, Al-Yousef, et al., 2018; Wang et al., 2020). In the study by Wang and colleagues (Wang et al., 2020), stearic acid as the surface active component is deprotonated into stearate at high pH, thereby increasing the electrostatic repulsion of the head group of fatty acid and the IFT. This stabilize the interface between oil droplets and water and as a result, coalescence time increases. Anion  $\text{SO}_4^{2-}$  affects in hindering oil droplet coalescence by hardening the interface (Ayirala, Al-Saleh, et al., 2018; Ayirala, Al-Yousef, et al., 2018; Ayirala, Yousef, et al., 2018). In other study, sulfate favor elasticity of the interface hence avoid snap off (Garcia-Olvera & Alvarado, 2017).

Crude oil in contact with low salinity water creates physical phenomena of spontaneous formation of micelles in the form of microdispersions (Emadi & Sohrabi, 2013; Sohrabi et al., 2017). Smart waterflooding affects contact angle rather than IFT or in other word smart water (in the case of diluted seawater) affects the interaction between rock/oil/brine instead of oil/brine (Yousef et al., 2010, 2011). A study by Alameri and colleagues (Alameri et al., 2014) proved that IFT increases by decreasing salinity in contrast with improving oil production effect by the low salinity waterflooding. Studies postulated that the IFT reduction by smart water was not sufficient in increase the oil recovery (Ayirala, Al-Yousef, et al., 2018; Garcia-Olvera & Alvarado, 2017; Hao et al., 2019; Maghsoudian et al., 2020).

Zeta potential analysis is another technique to study the interaction occurs at the interface between rock/oil/brine. One of the method to assess the optimum adjusted chemical of injected brine to yield the best performance of waterflood was by integrating surface analytical technique such as acid number, zeta potential, x-ray diffraction and etc. (Purswani et al., 2017). Suppose a surface having positive charge submerge in a solution containing ions. Negative charge layer will be accumulated at some position away from the surface and additional of positive and negative layers may also exist (Hao et al., 2019). When a solid is in proximity to another surface, it has the potential to push it away, and the intensity of this repulsive force is measured as disjoining pressure (Hao et al., 2019; Hiorth et al., 2010). The arrangement of the ions can be effectively explained by a double layer that has two distinct properties: thickness and potential difference. In addition, when brine comes into contact with charged surfaces, an additional layer of ions will accumulate near the surface, known as the double layer (Abbasi et al., 2020). ZP can be used to analyze injection water from the aspect of EOR performance (Abbasi et al., 2020). Double layer expansion is closely related to the stability improvement and increase the thickness of thin brine films at the interface (Hirasaki & Zhang, 2004; Myint & Firoozabadi, 2015). The relation between measured zeta potential and the surface retentions was understood by developing a surface-complexation model (SCM) in PHREEQC software (Parkhurst & Appelo, 2013). Another approach by SCM was carried out by Song and colleagues (Song et al., 2017) to predict ZP of calcite in brines through generalized double layer model.

Repulsive and attractive forces exerted between charged surfaces described the distribution of charges near a surface under the DLVO theory (Derjaguin & Landau, 1993; Elimelech et al., 1998; Verwey, 1947). When carbonate rock is dispersed in deionized water, its surface charge is positive. In 1967, Somasundaran and Agar conducted research on the zero point of charge (ZPC) in relation to pH. They discovered that the ZPC

of carbonate was between pH 8 and 9.5. When the pH of the solution was more alkaline, the surface of the carbonate rock was negatively charged due to the abundance of negative ions at the interface. On the other hand, when the pH was more acidic, the surface was positively charged (Hao et al., 2019). According to sources (Austad et al., 2009; nouar sofiane & Djebaili, 2008), the point of zero charge (PZC) for the calcite powder was approximately pH 8.9. Additionally, all of the water solutions that varied in pH were found to be within the range of 7 to 8.1 (Maghsoudian et al., 2020). In contrast, crude oil carries a negative charge. As a result, it is reasonable to assume that the surface charge of carbonate rock will turn negative after being mixed with crude oil and aged for a period of time (Abbasi et al., 2020). Typically, the acidic parts of the unrefined oil that carried a negative charge were absorbed by the carbonate surface, which had a positive charge. This process led to the formation of a complex called  $\text{Ca}^{2+}$ -carboxylate on the surface of the rock. This complex was responsible for altering the wettability of the rock towards the areas that were more likely to be covered in oil (Maghsoudian et al., 2020). Surface charge influenced the wettability of calcite by hydrocarbon adhesion of oil basic counterparts (Erzuah et al., 2017). High amount of excess ions intensify the alteration of surface charge (Maghsoudian et al., 2020; Mahani et al., 2016). Meanwhile, the surface charge of carbonate became positive when aged in deionized water (Abbasi et al., 2020). The electric surface charge turned to be more negative by  $\text{SO}_4^{2-}$  adsorption onto the rock surface. It was also represented by the value of ZP chalk that was more negative in smart water due to the increase in  $\text{SO}_4^{2-}$  or decrease of  $\text{Mg}^{2+}$  or  $\text{Ca}^{2+}$  concentration (Ding et al., 2019).  $\text{SO}_4^{2-}$  has strong acidic property while  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$  are alkaline earth metal, so that  $\text{SO}_4^{2-}$  tends to establish basic environment while interacting with ion  $\text{H}^+$  and the hydration bond breakdown.  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$  interact with ion  $\text{OH}^-$  and generate acid environment.  $\text{Mg}^{2+}$  is able to change the surface charge into positive and alter the wettability of rocks towards water-wet condition. In this state, the strongly adsorbed carboxylate groups of oil can be desorbed (Maghsoudian et al., 2020). Zhang and Austad found that lower ZP was obtained from higher  $\text{SO}_4^{2-}/\text{Ca}^{2+}$  ratio (Zhang & Austad, 2005). Moreover, amount of inactive salt, NaCl, is also essential in oil recovery from carbonate beside active ions,  $\text{SO}_4^{2-}$ ,  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$  (Fathi et al., 2010). pH affected the electrostatic interactions between oil, brine, and carbonate hence the surface charge, in which calcite surface became negatively charged at higher value (Hirasaki & Zhang, 2004; Sohal et al., 2016).

The increase of  $\text{SO}_4^{2-}$  makes ZP oil more positive while the decrease of  $\text{Mg}^{2+}$  results in more negative ZP oil. These happen due to ionic screening effect which will be reduced by decreasing the concentration of  $\text{Mg}^{2+}$  or promoted by increasing  $\text{SO}_4^{2-}$  (Ding et al., 2019). The ZP reduced to negative value by lowering the brine salinity (Mahani et al., 2016). ZP data can be utilized to estimate contact angles and disjoining pressure by the DLVO theory (Alshakhs & Kovscek, 2016). Above all, the brine has to be designed to make surface charge of carbonate rock surface having the same sign with the surface charge of oil, thus create water-wet condition. In other words, the negative magnitude of carbonate rock ZP should be large enough to maintain stable water film and prevent adsorption of the oil onto the rock surface.

The oil droplets has negative charge in the brine, thus higher magnitude of negative ZP hinder the oil droplets coalescence. The ZP value of crude oil droplets in brine varies due to the influence of ionic strength, specific adsorption, and pH (Ayirala, Yousef, et al., 2018). The dissociation of surface active organic acids in oil is triggered by increasing pH above the isoelectric point of oil and water interface. Thus, negatively charged crude oil droplets is observed at pH 6 which is higher than the dissociation pH of surface active compound in oil. Coalescence of oil droplet is easily achieved when the brine contain  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$  because of the low magnitude of ZP and high ionic strength of  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$ . Brine containing  $\text{SO}_4^{2-}$  or  $\text{Na}^+$  makes negative magnitude of ZP high which indicate rigid interface, hence the coalescence of oil droplet is difficult (Ayirala, Yousef, et al., 2018).

The effect of nano particles addition in smart water was able to change wettability from oil wet to intermediate/water-wet state (Shirazi et al., 2019).  $\text{TiO}_2$  NPs was capable of desorbing the oil components by being adsorbed onto the rock surface because the positive surface charge  $\text{TiO}_2$  NPs acted identically to divalent cations with stronger bond to the carboxylate component (Dai et al., 2017). Some inconsistency between the improvement of oil recovery and zeta potential by smart waterflooding can still be observed in which the changing of surface potential solely cannot dictate the oil recovery (Hiorth et al., 2010; Zhang et al., 2006). In addition, Yutkin and colleagues concluded that EDL expansion was only possible when the ionic strength of the brine was lower than 0.1 M (Yutkin et al., 2017).

## CONCLUSION

This systematic review has identified the research trend pertaining analysis of IFT and ZP in smart waterflooding within five years, from 2017 to 2021. Based on the answers to the questions posed, the pendant drop and electrophoretic mobility were used to measure IFT and assess ZP, respectively. The

analyzed water consisted of ion variations and salinity-modified water with crude oil sample from the field were used in some studies. ZP measurement was carried out upon brine/rock, oil/brine, and oil/brine/rock systems. IFT reduction was a result of the transfer of surface active component in the oil to the interface which was affected by the ion composition in the water. The relationship between IFT and the coalescence of oil droplets is directly proportional. The coalescence time can be reduced by manipulating the levels of magnesium and calcium ions in the water, decreasing the salinity of the brine, and lowering the pH. Anion  $\text{SO}_4^{2-}$  affects in hindering oil droplet coalescence by hardening the interface. ZP measurement was a guidance for assessing surface charge alteration, thus allowed the analysis of wettability alteration toward water-wet state due to smart waterflooding. ZP chalk was made more negative in smart water by increasing  $\text{SO}_4^{2-}$  or decreasing  $\text{Ca}^{2+}$  or  $\text{Mg}^{2+}$  concentrations. The negative value of rock ZP should be large enough to facilitate stable water film, thus create water-wet condition. Meanwhile, high magnitude of ZP oil indicated rigid interface, hence complicated the coalescence of oil droplet.

This SLR showed gaps and provided research opportunities in the area of interfacial mechanism under smart waterflooding, particularly related to IFT and ZP. The findings of this review shared the current development of evidence-based understanding for wettability alteration in smart waterflooding. As previously discussed, several authors had highlighted the important role of IFT and ZP to support the reasoning of smart waterflooding positive effect to the oil recovery. IFT and ZP provided a lens of interfacial phenomena through which wettability was altered. However according to the present review, doubt and inconsistency were still observed. Several authors concluded that IFT reduction from smart waterflooding was not enough to be the reason of oil recovery improvement. Moreover, surface charge alteration from ZP measurement also needed more evidence to prove the effectiveness towards improving oil recovery. The simulation approach was not able to directly cover the surface charge change mechanism.

An experimental setup should be designed to facilitate the observation of each mechanism separately, so that the impact to the smart waterflooding performance can be clarified. Advance visualization analysis to really witness the wettability alteration by IFT reduction or surface charge alteration through ZP measurement is worth considering for future development. It is necessary to develop a model for IFT measurement in smart waterflooding considering the chemical reaction involved. In addition, the improvement of simulation approach on ZP is also important. At last, a model which incorporate each mechanism with the ultimate oil recovery will be a great advancement, thus a comprehensive conclusion can be drawn.

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