EFFECT OF FORMATION WATER AND SURFACTANT CHARACTERISTICS ON RESERVOIR ROCK SURFACE CHARGES AND THEIR ZETA POTENTIALS

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ABSTRACT

The loss of injected surfactants in the oil reservoir during a surfactant flooding due to adsorption onto the rock surface weighs heavily on economics. Surfactant adsorption weakens the efficiency for the oil-water interfacial tension reduction. Several factors that influence the surfactant adsorption have been investigated, including the effect of temperature, as well as the concentration and charge of the surfactants. However, the impact of rock surface charge and zeta potential on surfactant adsorption has remained uncertain. This is because the surface charge development of the rock is complex depending on the effect of rock mineralogy, pH, brine salinity, and ionic strength of the ions present. This project aimed to develop an improved understanding of these effects on the rock surface charge and its contribution to the surfactant adsorption. These objectives were achieved through detailed laboratory measurement of surface charge density and zeta potential of silica and kaolinite by electrophoretic light scattering at different pH, brine salinities, and ionic strength. There were three different surfactant types used: anionic, zwitterionic, and non-ionic, which are represented by sodium dodecyl sulphate, coco betaine, and triton X-100, respectively, to investigate their adsorption on the surface of silica and kaolinite. The mineral characterization study shows that SiO_2 and Al_2O_3 were dominant chemical compound in both silica and kaolinite, which reflect their surface charge. The experimental work demonstrates that silica has a negative surface charge for pH exceeding 4.4 at all NaCl concentrations from 0.01M to 1M. However, in $CaCl_2$ solutions, the negative surface charge of silica turns to a positive charge at 0.1M for pH exceeding 8.7 and becomes more positively charged at a concentration of 1M. Different surface charge behavior was observed for kaolinite, which it is negatively charged at all NaCl concentrations from 0.01M to 1M. Moreover, both negative and positive charges were observed in $CaCl_2$ solutions. At pH of less than 10.8, the kaolinite surface indicates a negative charge at both concentrations of 0.01M and 0.1M. The positive charge of kaolinite was observed at a concentration of 1M with pH exceeding 4.4. These results indicate that pH, salt concentration (salinities), and salt types (ionic strength) have a major influence on the surface charge behavior of silica and kaolinite..

ABSTRAK

Kehilangan surfaktan tersuntik di dalam reservoir minyak ketika banjiran surfaktan berikutan penjerapannya pada permukaan batuan memberi impak yang besar terhadap ekonomi. Penjerapan surfaktan mengurangkan kecekapan untuk merendahkan ketegangan permukaan minyak-air. Beberapa faktor yang mempengaruhi penjerapan surfaktan telah dikaji termasuk kesan suhu, kepekatan dan cas surfaktan. Walau bagaimanapun, kesan cas permukaan batuan dan potensi zeta terhadap penjerapan surfaktan masih samar. Puncanya ialah pembentukan cas permukaan pada batuan adalah rumit, yang bergantung pada kesan mineralogi batuan, pH, kemasinan air garam, dan kekuatan ion-ion yang wujud. Projek ini bertujuan untuk menghasilkan pemahaman yang lebih baik tentang pengaruh faktor-faktor terbabit terhadap cas permukaan batuan dan kesannya pada penjerapan surfaktan. Semua objektif ini telah dicapai menerusi kajian makmal terhadap ketumpatan cas permukaan dan potensi zeta silika dan kaolinit, yang telah dilaksanakan secara terperinci dengan mengaplikasi penyerakan cahaya elektroforetik pada pH, kemasinan air garam, dan kekuatan ion yang berbeza. Tiga jenis surfaktan telah diguna: anion, zwiterion, dan bukanion, dengan masing-masing bahan kimia ialah natrium dodesil sulfat, koko betaina, dan triton X-100, bagi mengkaji penjerapan ketiga-tiga surfaktan itu pada permukaan silika dan kaolinit. Kajian pencirian mineral menunjukkan bahawa SiO_2 dan Al_2O_3 ialah sebatian kimia utama yang terdapat pada silika dan kaolinit, yang telah menentukan cas permukaan masing-masing. Data uji kaji menunjukkan bahawa silika mempunyai cas permukaan yang negatif pada pH 4.4 ke atas bagi semua kepekatan NaCl dari 0.01M hingga ke 1M. Walau bagimanapun, dalam larutan $CaCl_2$, cas permukaan negatif silika berubah kepada cas positif pada kepekatan 0.1M dan pH melebihi 8.7, dan seterusnya menjadi cas yang lebih positif pada kepekatan 1M. Tingkah laku cas permukaan yang berbeza belaku pada kaolinit dengan cas negatif terbentuk pada semua kepekatan NaCl dari 0.01M hingga ke 1M. Tambahan lagi, cas negatif dan cas positif telah terbentuk pada larutan $CaCl_2$. Bagi pH kurang daripada 10.8, permukaan kaolinit menunjukkan cas negatif pada kedua-dua kepekatan 0.01M dan 0.1M. Kaolinit bercas positif telah terbentuk pada kepekatan 1M dengan pH yang melebihi 4.4. Hasil kajian ini menunjukkan bahawa pH, kepekatan garam (kemasinan), dan jenis garam (kekuatan ion) mempunyai pengaruh yang besar terhadap tingkah laku cas permukaan silika dan kaolinit.

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LIST OF ABBREVIATIONS

ASP	-	Alkaline surfactant flooding
BET	-	Brunauer-Emmett-Teller
CEOR	-	Chemical enhanced oil recovery
CMC	-	Critical micelle concentration
CPC	-	Critical packing parameter
DLS	-	Dynamic light scattering
EOR	-	Enhanced oil recovery
EDL	-	Electrical double layer
FESEM	-	Field Emission Scanning Electron Microscope
FTIR	-	Fourier Transform Infrared
IFT	-	Interfacial tension
IEP	-	Isoelectric point
IHP	-	Inner Helmholtz Plane
OHP	-	Outer Helmholtz Plane
PZC	-	Point of zero charge
SDS	-	Sodium dodecyl sulphate
SEM	-	Scanning Electron Microscope
TGA	-	Thermal Gravimetric Analysis
XRD	-	X-ray Diffraction
XRF	-	X-ray Fluorescence

LIST OF SYMBOLS

F	-	Force
v	-	Velocity
p	-	Surface charge density
r	-	distance
N_c	-	Capillary number
T	-	Absolute temperature
π	-	pi
V	-	Electric potential
a	-	radius
k_b	-	Boltzmann constant
ε	-	Constant permittivity
H^+	-	Hydrogen ion
OH^-	-	Hydroxyl ion
C_i	-	Ionic strength
Z_i	-	Ion charge
$1/\kappa$	-	Debye length

porous reservoir rock. Strong oil-water IFT keeps oil immobile and traps in porous media as shown in Figure 1.1. This phenomenon is causing about 70% of oil to remain unproducible in the subsurface reservoir rock [5,6]. Thus, IFT plays a vital parameter in controlling oil recovery.

At the microscopic view, the flow of oil in porous rock toward a producing well is subjected to capillary numbers. The capillary number is crucial in determining the remaining oil saturation [7]. Capillary numbers (N_C) is defined as a ratio of viscous forces to the capillary forces [8]. An increase in Nc indicates an increase in oil recovery, which could be achieved either by increasing the viscous power or reducing the capillary forces [9,10]. Lowering oil-water IFT has become central in the reduction of capillary forces; thus, unlocking the trapped oil.
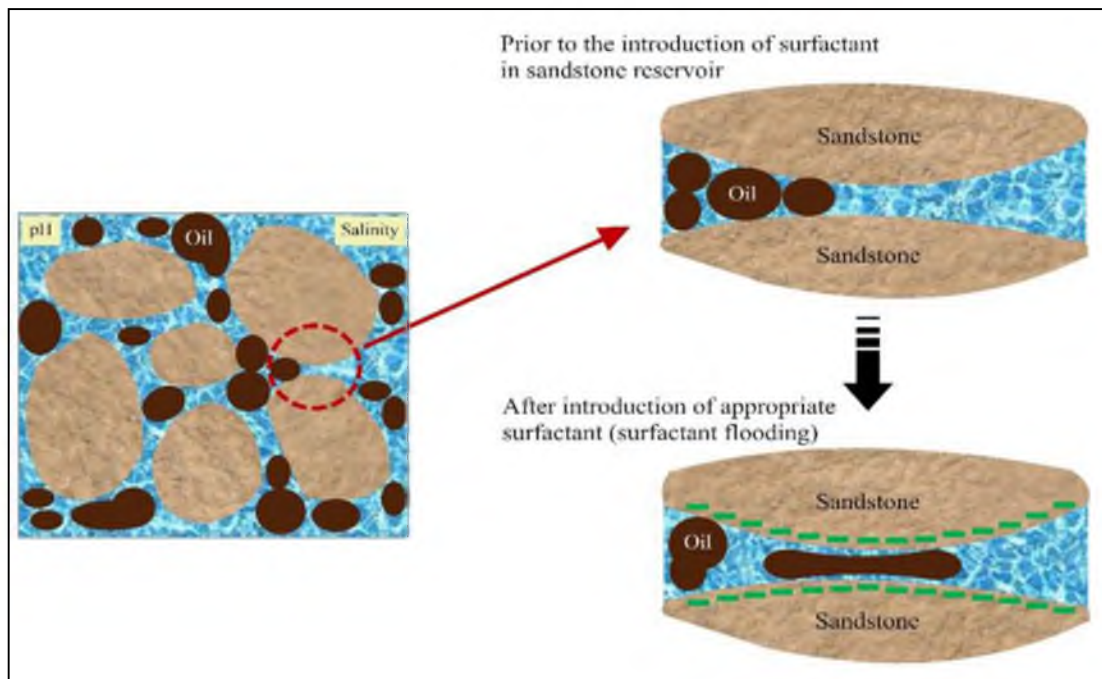
Figure 1.1   The schematic representation of (a) oil trapped within the pore throat (b) oil mobilized by lowering IFT with surfactant solution injection.

To reduce the oil-water IFT, the application of surfactants in the oil industry is introduced. This is known as surfactant flooding [11–13]. Surfactant flooding is a technology that employs surfactant in the injection fluids to lower the oil-water IFT [14–16]. The surfactant flooding process aims to achieve favorable phase behavior of the oil-water interface at ultra-low (0.001mN/m targeted) [17,18] IFT in order to

mobilize the trapped oil [13,19]. Whenever the oil is in contact with water, the surfactants are accumulated at the oil-water interface and formed an adsorbed film, which lowers the IFT between them [20].

Successful implementation of surfactant flooding has been recorded by Gao et al. [21]. They reported that surfactant flooding was implemented in Shengli field since 1922 in over 60 wells. Most surfactant flooding projects showed an increment in oil production and lasted for several years. Likewise, Belhaj et al. [22] documented that successful surfactant flooding has been achieved in other fields in the United States, Germany, Austria, and Canada. However, surfactant flooding shows expensive recovery due to high-cost chemicals.

The high cost of surfactant flooding is associated with surfactant losses during the process. As a result, a substantial amount of surfactant is needed [23]. Surfactant losses occur in the reservoir due to different mechanisms; (1) surfactant adsorption, (2) surfactant precipitation, and (3) surfactant degradation [23,24]. Surfactant losses due to precipitation can be avoided by choosing a surfactant that is tolerant of temperature and salt [22]. However, Kamal et al. [25] and Gogoi et al. [26] reported that surfactant adsorption cannot be avoided but can only be minimized. Surfactant adsorptions [18,27,28] the most dominant factor that controls surfactant losses. Surfactant adsorption on rock surfaces results in a loss, reducing the available surfactant concentration to be used for the oil-water IFT reduction [18,29], thus reduces its feasibility from an economic perspective.

Numerous studies have been done in the past to investigate the mechanism of surfactant adsorption. Various factors were found to affect surfactant adsorption. Azam et al. [30], Paria et al.[31] and Somasundaran [32] reported surfactants concentration, surfactant types, ionic strength, pH, salinity and temperature are the parameters that control the degrees of surfactant adsorption.

To date, the characteristic of surfactant adsorption has been extensively studied for various combination of surfactants such as anionic [33–35], non-ionic surfactant [36–39] zwitterionic surfactant [40–42] and reservoir rock samples [26,29,34,36,43].

According to Dang et al. [44] in 2011, anionic surfactant adsorption increases with increments in sodium chloride (NaCl) concentration. A similar finding reported by Bera et al.[34] in 2013, in which the amount of sodium dodecyl sulfate (SDS) (anionic) adsorption on the sand surface increases with increased salinity. However, the effect of divalent ion such $CaCl_2$ was remain uncertain.

Additionally, surfactant adsorption studies on silica surface [38,45], quartz [46] and carbonate rock [47,48] are described in detail. The studies show that surfactant adsorption is also dependent on the rock mineralogy and its surface charge. This is supported in a recent study by Rahul et al. [49] and Afeez et al. [50] indicating that the mineralogy of the reservoir and their surface charge significantly controls the surfactant adsorption and the selection of surfactants used. Most of the oil reservoir rock is charged either negatively or positively in nature, depending on the mineralogy of the rock [41,51,52]. Practically the adsorption can be minimized by having a similar charge of surfactant with the rock surfaces. A similar charge causes a repulsive force at surfactant-solid interfaces, thus preventing adsorption from taking place.

The role of surface charge in the adsorption mechanism has been documented by Marek et al. [53]. Besides rock mineralogy, the surface charge is also dependant on the pH and salinity of formation water in the reservoir. The isoelectric point (IEP), zeta potential and ionic strength are used to describe the relationship. IEP is a pH where the zeta potential value is zero, which means the negative and positive charge is in balance. The surface charge is highly dependant on the IEP. When the pH of the solution is over the IEP, the surface charge is negative and vice-versa [54]. At the same time, the zeta potential is also influenced by ionic strength. The presence of different types of salt, such as mono and divalent ions will significantly affect the ionic strength, thus effecting the zeta potential and IEP of the solid surface charge [41,54].

The surfactant adsorption system in porous media is genuinely complex. Even though many research have been done, there are still uncertainties associated with the interpretation of measurement, especially concerning the role of surface charge, pH, salinity and ionic strength to the surfactant adsorption mechanism. The surface charge is the key to selecting the appropriate surfactant in the surfactant flooding process.

Therefore, to understand the surface charge development and its association with adsorption is essential, and it has been the motivation of this study.

1.2   Problem Statement

Finding an excellent model system in describing surfactant adsorption phenomena in an oil reservoir can be a problematic process. Many factors have to be dealt with, and Figure 1.2 helps to break the complexity in understanding the surfactant adsorption mechanism in the oil reservoir rock.
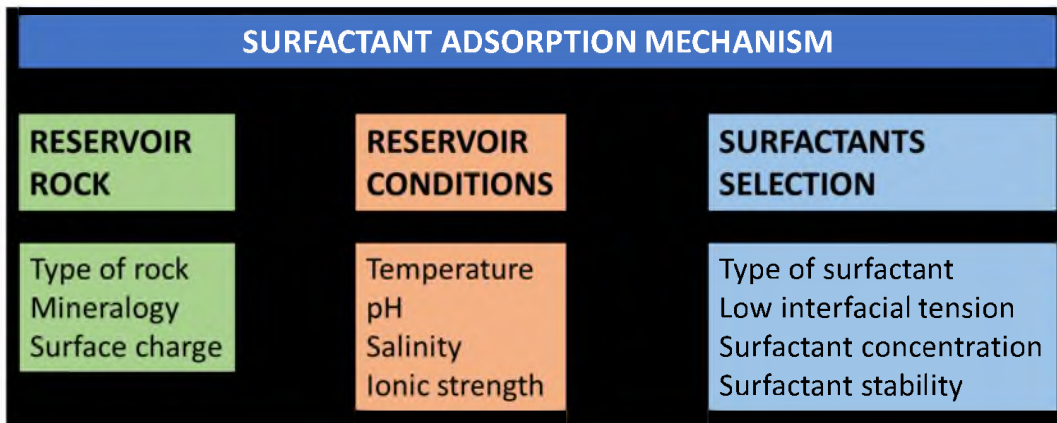
Figure 1.2   The schematic representation of the surfactant adsorption factors.

Surfactant adsorption onto the porous rock is very dependant on the reservoir rock type, reservoir condition, and surfactant types that are used in the surfactant flooding process. The reservoir conditions, especially pH, salinity and ionic strength (due to valent ion) are the central parameters controlling the interaction properties of both reservoir rock and the surfactant.

The interaction between the reservoir fluid and rock surface makes the rock surface positively or negatively charged depending on the mineralogy of the rock. However, the rock mineralogy is not the only factor that controls the rock surface charge. This is because the pH may significantly affect the surface charge. Modification of the surface charge can vary with different pH. Point zero charge (PZC) is an indication where the surface charge can change from positive to negative

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or vice-versa. PZC measures the pH where the negative and positive charge is in balance. When the reservoir pH is above the PZC value, it means the rock surface has a negative charge. With this, the surfactant that has a similar charge should be used for surfactant flooding to minimize the surfactant adsorption.

The role of zeta potential and its relation to surfactant adsorption is still a major debate. Zeta potential also varies with pH. The pH where the zeta potential is zero is called an isoelectric point (IEP). The zeta potential may determine the forces involved in surfactant adsorption process either by a repulsive or attractive force. However, the mechanism of these forces needs further investigation because they are related to the salinity and the presence of valent ions in the reservoir fluid. On the other hand, the correlation between the surface charge and the zeta potential is still uncertain.

1.3   Research Goal

The project aims to develop an improved understanding of the surface charge development on different types of reservoir minerals under different reservoir conditions. The study will further investigate how this surface charge influences the selection of surfactant types and the adsorption into reservoir rock. In detail, the objectives of this research are as follows:

    i. To characterise the mineralogy of reservoir rock minerals and their physicochemical properties.

    ii. To examine the surface zeta potential behavior of the reservoir rock minerals at different pH, salinity, and ionic strength.

    iii. To investigate the stability of different surfactant types on different reservoir environment primarily by temperature and salinity.

    iv. To investigate the zeta potential behavior at the surface of rock minerals with different types of surfactants and salinity.

1.4   Scope of the Study

To achieve the objectives of the research, several parameters within the study are specified. Firstly, reservoir minerals used in this study are silica and kaolinite. Silica is vital because it is widely found in sandstone. Kaolinite is a clay minerals that are also significant because their existence, even in a small amount, is likely to affect the rock surface charge. Various characterization techniques was used to identify their chemical compounds and also physical properties. The main chemical compound present in the silica and kaolinite was measured by X-ray Diffraction (XRD) analysis. An analysis of Fourier Transform Infrared Spectroscopy (FTIR) was carried out to detect the structure and chemical bonding of the chemical compound. The physical properties such as surface area, pore size, particle size, and surface morphology are also measured.

Secondly, there are three different types of surfactants used in this study: anionic, zwitterion, and non-ionic. Sodium Dodecyl Sulphate (SDS), Coco Betaine, and Triton X-100, respectively. The critical micelle concentration (CMC) of the surfactants was determined by measuring their surface tension values at varying surfactant concentrations and different salinity using a tensionmeter. The surfactant stability at various temperature ranges was measured by thermal gravimetric analysis (TGA).

Thirdly, the determination of the surface charge of reservoir minerals is investigated under various ranges of pH, salinity and ionic strength. Hydrochloric acid (HCl) and sodium hydroxide (NaOH) was used to adjust the desired pH of the solutions from pH 2 to 12. Sodium chloride (NaCl) and Calcium Chloride (CaCl$_2$) was used to produce reservoir formation water at different salinity and ionic strength. The surface charge of the reservoir materials was determined by measuring zeta potential using the electrophoretic light scattering approach. The result of the pH/salinity/ionic strength versus the surface zeta potential enables the determination of the isoelectric point (IEP) of the reservoir materials, which will be essential to identify the types of surfactants used for the surfactant flooding process.

The last part is the adsorption study. This experimental study was conducted at static conditions. The surfactant was employed based on the surface charge obtained. Zeta potential approach was used to discuss all the surfactant adsorption phenomena together with explanations from published literature.

1.5   Significance of the Study

This study provides a comprehensive understanding of the underlying principle, surfactant adsorption in the surfactant flooding process   with regards to the surface charge of the rock, the reservoir condition, and surfactant selection.   The findings would benefit the oil industry in the sense of filling the knowledge gap on the role of surface charge for designing an effective surfactant flooding from its losses.

Besides, the adoption of zeta potential measurement not only enables discussion on the adsorption process concerning pH, salinity and ionic strength, but also describes the force involved such as attractive and repulsive forces.

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