

Adsorption of anionic surfactant on surface of reservoir minerals in alkaline-surfactant-polymer system

^{a,b} Tengku Amran Tengku Mohd *, ^a Muhammad Ikhran Abdul Wahib, ^a Nik Khairul Irfan Nik Ab Lah,

^a Muhammad Shafiq Mat Shayuti, ^a Munawar Zaman Shahrudin, ^a Rohani Mohd Zin, ^b Mohd Zaidi Jaafar

^aSchool of Chemical Engineering, College of Engineering, Universiti Teknologi MARA, Selangor, Malaysia

^bSchool of Chemical and Energy Engineering, Faculty of Engineering, Universiti Teknologi Malaysia, Johor, Malaysia

*Corresponding email: amran865@uitm.edu.my

Abstract

Alkaline-surfactant-polymer (ASP) flooding is significant to the oil and gas industry due to synergistic interaction between alkaline, surfactant and polymer. However, chemical losses due to adsorptions of surfactant and polymer on the rock surface could lead to inefficiency of the process. There are also significant uncertainties on adsorption mechanism when surfactant is flooded with presence of alkaline and polymer. This study highlights the static adsorption tests using anionic sodium dodecyl sulphate (SDS), hydrolysed polyacrylamide (HPAM) and sodium carbonate (Na_2CO_3) as the surfactant, polymer and alkaline, respectively. Sand particles and kaolinite clay were used as the reservoir minerals. The adsorption tests were conducted at various surfactant concentrations ranging from 50 to 2000 ppm. Sodium chloride (NaCl) concentration was investigated from 0 to 2 wt.%, while the local sand and kaolinite was mixed in surfactant solution at a fixed mass to volume ratio of 1:5. The static adsorption test was conducted by shaking the mixture samples and centrifugation before analysing the supernatant liquid using UV-Visible spectrophotometer. The results showed that the surfactant adsorption was higher on kaolinite compared to sand particle. The higher the salinity, the higher the adsorption of surfactant due to higher ionic strength. The adsorption of SDS surfactant on sand particles and kaolinite was lesser in ASP system compared to the presence of surfactant solution alone. Thus, it can be concluded that the presence of polymer and alkaline in ASP solution have great potential to reduce the surfactant adsorption on both sand particle and kaolinite.

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1.0 Introduction

Conventional oil, which includes crude oil, condensate, and natural gas, is anticipated to account for about 90% of global production by 2030. Conventional oil production is expected to fulfil global demand in the 21st century, as higher prices drive new discoveries and improved recovery techniques (Monadjemi, 2016; Isma'il, 2012). Enhanced oil recovery (EOR) is widely recognised for its efficiency in increasing oil production while recovering remaining oil from the conventional stage by various methods such as immiscible, miscible, microbial enhanced oil recovery (MEOR), thermal and chemical. Chemical enhanced oil recovery (CEOR) denotes as

one of the effective technologies for recovering bypassed oils and residual oils stranded in reservoirs, particularly in mature oil fields. Chemical injection is used in this EOR method to improve oil recovery (Gbadamosi et al., 2019). CEOR slowed down in the 1980s and was significantly behind immiscible, miscible, and thermal EOR techniques in terms of worldwide project implementation.

Alkaline-surfactant-polymer (ASP) flooding, one of the rising CEOR techniques, is extensively utilised in the China, United States, India, and Venezuela fields, due to its potential to lower interfacial tension (IFT) and controlling mobility (Zhang et al., 2019). Thus, monitoring the efficiency of ASP flooding is important as reviewed and suggested by

Mohd et al. (2017a; 2017b), who employed a real-time approach of streaming potential measurement using electrodes permanently installed downhole. Recent practice in China demonstrates that ASP can be an invaluable tool for surviving the drop in oil prices. By simultaneously injecting polymer, surfactant, and alkali, ASP flooding was proved to enhance and maximise displacement and sweep efficiency as well as recovery of hydrocarbon up to 20% (Yang et al., 2018). Both alkali and surfactant help in reducing the IFT, thus improving sweep efficiency and enhancing oil recovery. Alkali may also be used to inhibit the higher rate of adsorption of high-priced surfactants. Reduction of mobility ratio is achieved through the use of polymer, which adds substantially to the improvement in sweep efficiency (Lüftenegger & Clemens, 2017; Guo et al., 2017; Mohd et al., 2020). Another CEOR process providing mobility control is foam flooding, involving the investigation of mobility (Mohd et al., 2015), the improvement of foam stability with the addition of nanoparticles (Azizi et al., 2015; Mohd et al., 2018), as well as the simulation studies using molecular dynamic simulation (Azmi et al., 2019; 2020).

Despite many advancements in CEOR techniques that incorporate the surfactant, some concerns must be addressed to ensure that the surfactant is effective at improving recovery factors in oil reservoirs (Gbadamosi et al., 2019). Typical problems include surfactant loss via surfactant retention mechanisms such as adsorption, precipitation, or trapping during flooding (Kamal et al., 2017). Surfactant adsorption occurs during chemical flooding when the molecules of surfactant are adsorbed in porous medium from the bulk to the surfactant-rock interface. The rock surfaces may have a variety of charges, including negative, positive, negative-positive, and no charge, all of which influence the surfactant's adsorption differently. Positively charged surfaces attract anionic surfactants while cationic surfactants are attracted towards negatively charged surfaces. No net electrical charge happens when the rock surface is at its isoelectric point (IEP). Therefore, the surface carries a positive charge and negative charge at pH below and above the IEP, respectively. Surfactant loss was found because of surfactant adsorption onto reservoir rocks, resulting in decreased efficiency and economic losses (Kamal et al., 2017). It is worth noting that although adsorption surfactant cannot be completely prevented, it may however, be reduced. At the interface, the behaviour of

surfactants is controlled through mechanisms such as electrostatic attraction, hydrophobic bonding, hydrogen bonding, species solvation and covalent bonding. When negatively charged surfactant interact with opposite charge of solid interface, higher adsorption is observed due to electrostatic interactions. In contrast, opposite interaction occurs when the surfactant has similar charge with the solid interface, which slows down the adsorption process (Ercan et al., 2021). For example, anionic surfactant which has negative charge will be attracted to positive surface charge (carbonate) while cationic surfactant that has positively charged will be attracted to the negative charged rock surface such as sandstone (Wang et al., 2015; Saxena et al., 2019).

Solid surfaces could have both positive and negative charges in ionised solution, it can be either one of it or it also appears by ions adsorption from aqueous solution onto non-charged surfaces (Amirianshoja et al., 2013). At low solution concentration, surfactant molecules are adsorbed onto rock sediment or surface as single monomer depending on the electrical charges of the solid surface (Richard & Rendtor, 2019). As the concentration of the solution increases, monomers of the surfactant are aggregately adsorbed and tend to form micelles concentration. This aggregation can form one or two layer(s), and the hydrophobic interaction of the surfactant molecules leads to level of critical micelle increase in the adsorption (Azam et al., 2013; Wu et al., 2017). Physical-chemical properties such as pH, ionic strength, temperature, and adsorbent concentration can affect the adsorption of surfactant and have the influence in the behaviours of the dissolution of the mineral and surfactant precipitation (Tagavifar et al., 2018; Hoff et al., 2001). These properties can result in significant changes of the adsorption system.

Electrostatic attraction between the charge of surfactant head group and the charge of rock surface is crucial mechanisms in evaluating surfactant adsorption process. According to Bera et al. (2013), anionic surfactants adsorb less than cationic surfactants onto sandstone surfaces, while non-ionic surfactants perform intermediately. Previously conducted research on an anionic surfactant showed that it adsorbed ineffectively onto sandstone, where the surfactant's electrostatic interactions with the substrate resulted in weak interactions between the rock surfaces and surfactant (Curbelo et al., 2020). When the surfactant

head group has the same charge as the reservoir mineral surfaces, strong repulsion forces take place, forming a weak attraction. This phenomenon occurs when an anionic surfactant, such as sodium dodecyl sulphate (SDS) attracts to the quartz sand containing silica, where the surfactant's adsorption rate on the rock surfaces is quite low and decreasing (Mohd & Jaafar, 2019).

In ASP, adsorption occurs due to losses of surfactant and polymer into the formation. Theoretically, the addition of polymer in ASP could reduce surfactant adsorption. However, there have been limited number of studies on the adsorption behaviour for this case and thus, there are many uncertainties that motivate further investigation. Therefore, this research is carried out to evaluate the adsorption mechanism of anionic SDS surfactant on the local sand and kaolinite surfaces using static adsorption test at varying salinity with and without presences of alkaline and polymer. The outcomes of this research could provide the prospect of salinity and kaolinite in altering the adsorption behaviour of SDS surfactant in the ASP system. Higher adsorption is expected at higher brine concentration due to increase of the ionic strength, while alkaline and polymer are potential in reducing the surfactant adsorption on both local sand and kaolinite clay. Thus, the findings from this research could be significant for reservoirs comprised of shaly formation with the presence of kaolinite clay mineral for future EOR planning.

2.0 Methodology

2.1 Materials

SDS was purchased from Merck, while Hydrolyzed Polyacrylamide (HPAM, MW: 16–19 million g/mol) used as polymer was acquired from Vchem. Sodium carbonate (NaCO_3) used for preparation of alkaline solution was obtained from QReC. Sodium chloride for brine preparation was purchased from sigma Aldrich. Potassium bromide (KBr) used as standard reference for FTIR analysis was purchased from Sigma-Aldrich, Malaysia. Local sand and kaolinite clays were the reservoir minerals used for adsorption tests. The sand particles were obtained from local sand in Pantai Bagan Lalang, Malaysia, sieved at desired range of size (60 to 200 μm) and purified, while kaolinite clay mineral was purchased from Sigma-Aldrich, Malaysia.

2.2 Methods

2.2.1 Critical micelle concentration (CMC)

Measurement of surface tension was carried out using tensiometer (KRUS K20 EasyDyne) for CMC determination of SDS surfactant by plotting surface tension values against surfactant concentrations. In this study, surface tension was measured using Du Nouy ring method at atmospheric pressure at different concentrations of SDS surfactant from 0 to 0.6 wt.%. The CMC is the concentration at the inflexion point of the curve. Acetone was used during experiment to clean the ring before flame-dried for each measurement.

2.2.2 Static adsorption tests

Local sand and kaolinite clay were used for static adsorption test as the solid samples. SDS solution was prepared by dissolving the SDS powder in distilled water at varying concentration from 50 to 2000 ppm. Clean local sand was added and mixed with the SDS solution in the Schott bottle at a fixed mass to volume ratio of 1:5 for adsorption tests. Then, the mixture sample was placed in the incubator shaker for 24 hours at constant room temperature, centrifuged at 2000 rpm for 20 minutes to segregate liquid and solid part effectively. The upper liquid supernatant was then collected and ready for adsorption analysis by UV-Vis spectrophotometer. Calibration curve was required to determine the surfactant concentration after adsorption. Those procedures were repeated for kaolinite clay mineral as the solid sample and varying the salinity from 0 to 2 wt.% of brine concentration. Then, the SDS solution was formulated with alkaline and polymer as ASP system, and the same procedures were repeated for adsorption tests.

2.2.3 Adsorption measurement

Calibration curve is a standard curve plotted linearly between absorbance and concentration to determine the adsorption of surfactant onto sand particles or kaolinite clay mineral. UV-Vis spectrophotometer was used to obtain the absorbance at each concentration for constructing the calibration curve. The unknown concentration of the supernatant liquid after adsorption test was estimated by using calibration curve from the surfactant solutions of known concentrations. The amount of adsorbed surfactant, $A(t)$ in mg/g was determined using Eq. (1) (Saxena et al., 2019):

$$A(t) = \frac{\Delta C(t) \cdot V}{m} = \frac{(C_i - C_f) \cdot V}{m} \quad (1)$$

where C_i and C_f are the initial and final equilibrium concentrations of surfactant, V is volume of the solution used and m is the mass of adsorbent represented by the mineral samples.

3.0 Results and discussion

3.1 Critical micelle concentration

CMC determination of surfactant was done using surface tension measurement. The CMC was obtained by plotting surface tension versus surfactant concentration as shown in Fig. 1. The surface tension was observed to decrease as the surfactant concentration increased until it reached at the inflexion point, from which the curve started to deflect. Thus, the CMC was found at 0.22 wt.%. At the CMC, large amounts of micelles started to form in the surfactant solution. Further increase in surfactant concentration beyond the CMC did not affect the surfactant adsorption on the solid surface (Hoff et al., 2001; Azam et al., 2013).

3.2 Static adsorption

3.2.1 Adsorption of SDS surfactant

The effect of salinity on the static adsorption of SDS surfactant onto the surfaces of sand particle and kaolinite clay was investigated and the results are depicted in Fig. 2. From Fig. 2, a sudden increase in surfactant adsorption with increase in concentration was observed, which was followed by a gradual increase until reaching plateau, where adsorption was constant. At this region, additional concentration added to the system had no effect to the surfactant adsorption as most of the active sites were adsorbed with surfactant, and micelles were repelled by the adsorbed surfactant molecule (Elias et al., 2016). This pattern shown at all three salinity cases complies with the typical adsorption curve found in previous research (Paria et al., 2004; Bera et al., 2013). For salinity effect, brine concentration of 2 wt.% demonstrated the highest surfactant adsorption, followed by 1 wt.% and 0 wt.%. At the same concentration of anionic surfactant, the increase in salinity can increase the adsorption of surfactant onto sand particle. At surfactant concentration of 2000 ppm, it was found that surfactant adsorption onto the sand particles was 0.93 mg/g at 2 wt.% brine, while 0.83 mg/g and 0.64 mg/g surfactant adsorption onto the sand particles was obtained for

1 wt.% and 0 wt.%, respectively. Increasing the sodium chloride concentration from 0 to 2 wt.% in the surfactant solution had caused the charge sites to increase, resulting in an abrupt increase of the anionic SDS surfactant adsorption on the negatively charged surfaces (Azam et al., 2013). Thus, introducing negative salinity gradient was reported to be able to overcome moderate surfactant retention to a certain degree. Negative salinity gradient is favourable when

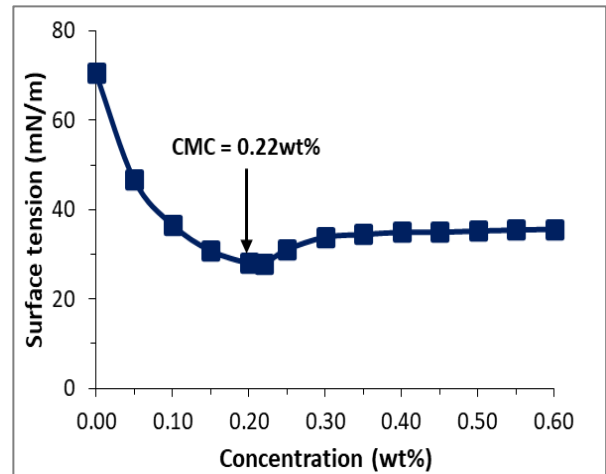


Fig. 1: CMC of anionic SDS surfactant

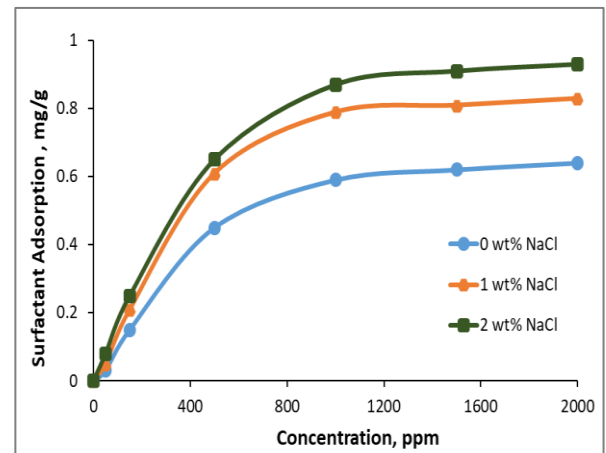


Fig. 2: Static adsorption of SDS onto sand particle

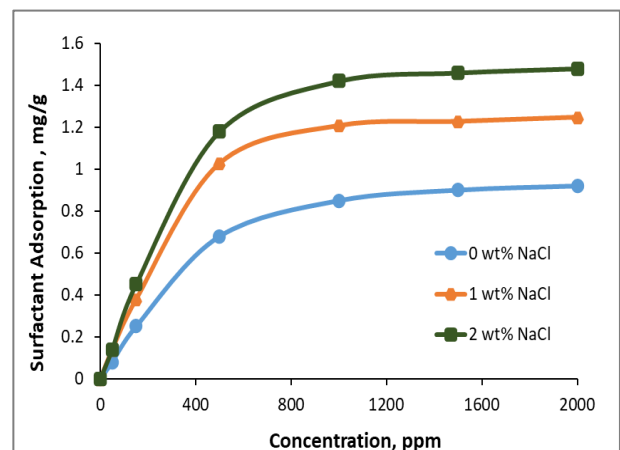


Fig. 3: Adsorption of SDS onto kaolinite mineral

decreasing salinity is required as the surfactant is diluted (Grigg and Bai, 2005; Sheng, 2010). Increasing the salt concentration also increases the ionic strength, which in turn boosts up the surfactant adsorption. This is related to the salting-out effect of the surfactant. The thickness of the electrical double layer (EDL) at the solid surface is diminished due to increase in ionic strength, leading to reduction in electrostatic repulsion of anions.

Kaolinite displayed higher surfactant adsorption compared to the sand particles as illustrated in Fig. 3. At surfactant concentration of 2000 ppm, the surfactant adsorption was 1.48 mg/g, 1.25 mg/g, and 0.92 mg/g for 2 wt.%, 1 wt.%, and 0 wt.% brine, respectively. From Fig. 3, similar adsorption behaviour was observed on kaolinite as on the sand particles. At the interface between surfactant and kaolinite, there was an unequal distribution of electrical charges, which led to a rise in the potential across the interface, resulting in the formation of EDL (Pethkar & Paknikar, 1998). As sodium chloride concentration increased, surfactant adsorption on kaolinite surface was high due to compression of EDL, which was similar to the adsorption on sand surface. Therefore, high ionic strength at high brine concentration could favour the adsorption of SDS on the kaolinite surface.

3.2.2 Adsorption of SDS surfactant in ASP system

Static adsorption tests of SDS surfactant in ASP system were conducted on sand particles and kaolinite clay minerals. Fig. 4 illustrates surfactant adsorption behaviour on sand particles, while Fig. 5 depicts the surfactant adsorption on kaolinite clay, both in ASP system at brine concentration ranging from 0 to 2 wt.%. From the figures, it was observed that higher surfactant concentration in ASP system had led to the increase of adsorption until reaching a stationary point, after which a slight increase was observed, followed by no further increase of adsorption as the surfactant concentration increased (Paria and Khilar, 2004). This is a typical adsorption pattern, which is similarly observed in the adsorption curves shown in Fig. 2 and Fig. 3. As a typical surfactant adsorption behaviour, adsorption is directly proportional to the surfactant concentration due to the formation of hemi-micelles. SDS was adsorbed by quartz due to the capability of the formation mineral to generate a variable charge (Azam et al., 2013).

Fig. 4 and Fig. 5 show that the increase of brine concentration increased the surfactant adsorption in ASP system. Sand particles as adsorbent exhibited a

maximum surfactant adsorption of 0.71 mg/g at brine concentration of 2 wt.%, followed by 0.61 mg/g and 0.42 mg/g at brine concentration of 1 wt.% and 0 wt.%, respectively. Kaolinite clay acted as a good adsorbent, whereby the amount of adsorbed surfactant was 0.99 mg/g, 0.8 mg/g, and 0.54 mg/g at 2, 1, and 0 wt.%, respectively. At 2 wt.% brine, surfactant adsorption in

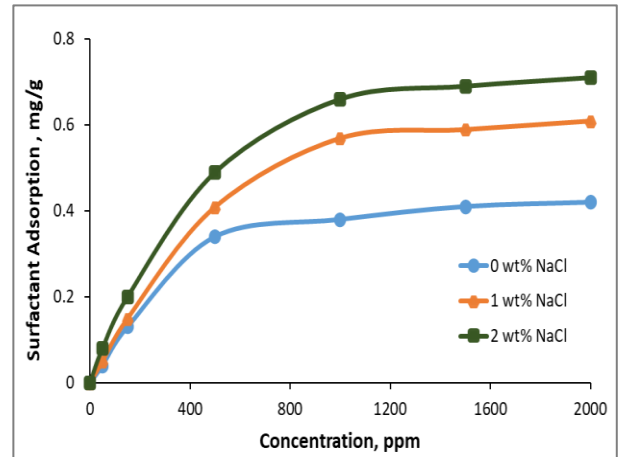


Fig. 4: Adsorption of SDS surfactant on sand particles in ASP system

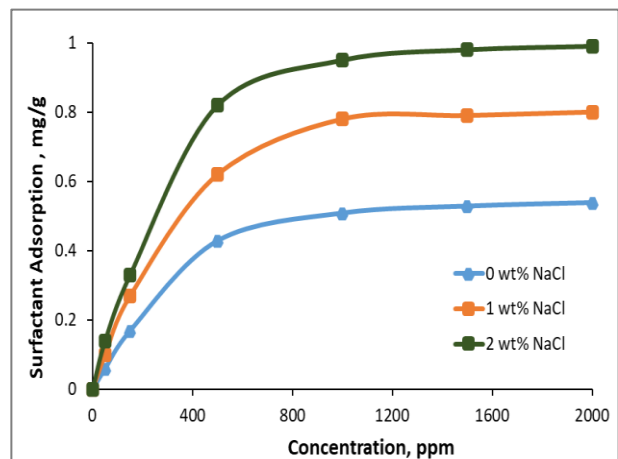


Fig. 5: Adsorption of SDS surfactant on kaolinite clay in ASP system

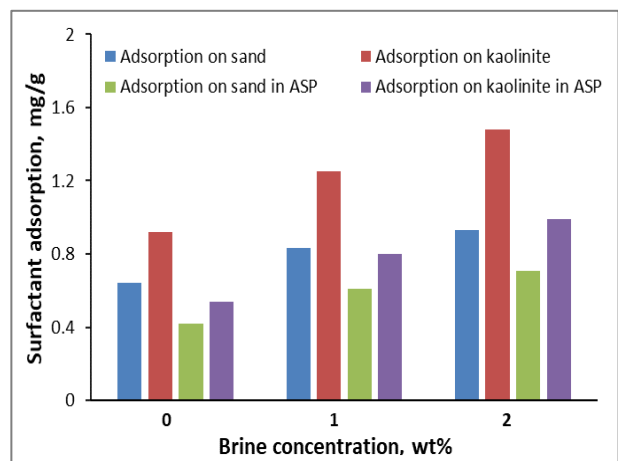


Fig. 6: Summary of surfactant adsorption on sand and kaolinite in surfactant formulation and ASP system at different brine concentration

ASP system increased by approximately 40% when changing the sand particles to kaolinite clay as the adsorbent. For salinity effect from 0 to 2 wt.%, the surfactant adsorption in ASP system exhibited an increase of approximately 70% and 83% on sand particles and kaolinite clay, respectively. These findings proved that salinity plays an important role on surfactant adsorption behaviour, which could not be simply ignored. High brine concentration provides high ionic strength leading to reduction in repulsion of anions due to compression of EDL, which in turn increases the adsorption capacity (Pethkar & Paknikar, 1998).

Addition of polymer and alkaline with surfactant formulation resulted in a significant decrease of surfactant adsorption. Without addition of alkaline and polymer surfactant formulation (Fig. 2 and Fig. 3) a maximum adsorption of 0.93 mg/g and 1.48 mg/g was exhibited on sand particles and kaolinite respectively at 2 wt.% brine. However, addition of sodium carbonate alkaline and HPAM polymer as ASP system has reduced the surfactant adsorption to 0.71 mg/g and 0.99 mg/g, respectively. The presence of sodium carbonate as sacrificial agent reduces the surfactant adsorption for most of reservoir rocks. Alkali plays a crucial role in this process by enhancing the formation of the anionic hydroxyl group (OH^-) in aqueous solution (Azam et al., 2013). The reduction of surfactant adsorption is due to the dissociation of alkaline into weak carbonic acid and generation of OH^- ions upon interaction with water molecules (Saxena et al., 2019). The synergistic effect between alkaline and surfactant in reducing the adsorption is more effective than surfactant alone (Zhang et al., 2012). HPAM is an anionic polymer, thus the negative charged of polymer gives advantage to the reduction of the surfactant adsorption. Therefore, the synergistic effect between surfactant, alkaline and polymer resulted in a gradual decrease of surfactant adsorption.

Fig. 6 presents the summary of surfactant adsorption on surfaces of sand and kaolinite in surfactant formulation and ASP system at various brine concentrations. From Fig. 6, it can be observed that brine concentration, types of reservoir mineral as adsorbent, and system formulation for adsorption analysis play important roles in surfactant adsorption behaviour. As the brine concentration increased, the surfactant adsorption increased, with kaolinite as the adsorbent exhibited higher surfactant adsorption

compared to sand particles. In contrast, formulation of ASP system had significantly minimised the surfactant adsorption capacity for all the investigated cases. The highest surfactant adsorption was obtained by surfactant formulation with kaolinite as adsorbent at 2 wt.% brine (1.48 mg/g), while the lowest surfactant adsorption was exhibited by ASP system with sand as adsorbent at 0 wt.% brine (0.42 mg/g). Thus, synergistic effect of alkaline, surfactant and polymer were proven to have given significant impact in minimising surfactant adsorption. Alkaline promotes the formation of OH^- in aqueous solution (Azam et al., 2013), while anionic HPAM polymer favours the electrostatic repulsion, which provides advantage to the reduction of the surfactant adsorption. Besides that, polymer introduced into ASP flooding can reduce the mobility ratio, thus enhancing the oil recovery (Negin et al., 2017; Mao et al., 2018). According to Thomas & Farouq (2001), the key to successful ASP flooding is the good performance and the displacement ability. The effectiveness of ASP system proved that synergistic of chemicals used can greatly minimise surfactant adsorption. It was found that as the surfactant concentration increases, SDS adsorbs strongly on the surface in the order of alumina > kaolinite > ilmenite > silica (Elias et al., 2016).

4.0 Conclusions

In this research, adsorption of anionic SDS surfactant on surfaces of reservoir minerals namely sand and kaolinite clay, was investigated in surfactant formulation and ASP system. CMC of SDS surfactant was found at 0.22 wt.%, below which the surfactant adsorption was observed to occur. From the adsorption tests, it was found that ASP system exhibited the lowest surfactant adsorption on sand particles at 0 wt.% brine, while the highest surfactant adsorption was obtained on kaolinite clay by surfactant formulation at 2 wt.% brine. Therefore, it can be concluded that ASP system has a huge impact in minimising the surfactant adsorption. However, presence of clay in the formation and salinity should not be simply neglected. Indeed, they should be thoroughly considered in the selection of EOR methods as they could increase the surfactant adsorption, thus, affecting the efficiency of the process.

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