

NANOPARTICLES-SURFACTANT FOAM AND CRUDE OIL INTERACTION IN
POROUS MEDIA

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NANOPARTICLES-SURFACTANT FOAM AND CRUDE OIL INTERACTION
IN POROUS MEDIA

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To the Almighty God be the glory for his love, mercies and favour.

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ABSTRACT

Nanoparticles and surfactant stabilized foams have versatile applications in enhanced oil recovery process. The synergistic advantages of surface tension reduction by surfactant and nanoparticles adsorption at the foam lamellae can be exploited for producing foam with high foamability and longtime stability in the oil producing reservoir. However, the influence of nanoparticles on the static and the dynamic stability of conventional foam is not yet explicit due to limited studies. Moreover, only few studies have considered the pore-scale mechanisms of the nanoparticles-surfactant foams flow process in porous media and the minimization of surfactant adsorption in presence of nanoparticles. Due to limited research in this area, this study was conducted to understand the influence of silicon dioxide (SiO_2) and aluminum oxide (Al_2O_3) nanoparticles on the surfactant foam bulk and dynamic stability and surfactant adsorption on clay mineral. Four main experimental studies comprising the influence of the nanoparticles on surfactant adsorption on kaolinite, bulk and bubble-scale foam stability evaluation in presence of oil and salts, pore-scale visualization studies in etched glass micromodels, and fluid diversion process experiments were conducted. Results of this study showed that the adsorption of surfactant on clay mineral reduced drastically by 40% and 75% in presence of Al_2O_3 and SiO_2 nanoparticles, respectively. The maximum adsorption of surfactant on the nanoparticles occurred at 0.3 wt % sodium dodecyl sulfate (SDS). The foam bulk and bubble scale stability results indicated that 1 wt % of SiO_2 and Al_2O_3 nanoparticles enhanced the stability of the foam in presence of oil and salts. There was a transition salt concentration beyond which the foam stability increased with increasing salt concentrations. The presence of Al_2O_3 and SiO_2 nanoparticles prevented the entering of emulsified oil into the foam lamellae and decreased the transition salt concentrations. From the results of the pore scale studies, the dominant mechanisms of foam propagation in water-wet system were lamellae division and bubble-to-multiple bubble lamellae division. The dominant mechanisms of residual oil mobilization and displacement by the foam in water-wet media were found to be direct displacement and emulsification of oil. The dominant mechanism of foam propagation and residual oil mobilization in oil-wet system was identified as the generation of pore spanning continuous gas foam. Inter-bubble trapping of oil and water, lamellae detaching and collapsing of SDS-foam were observed in presence of oil in both water-wet and oil-wet systems. Generally, the SiO_2 -SDS and Al_2O_3 -SDS foams propagated successfully in oil-filled water-wet and oil-wet systems. Bubble coalescence was prevented during film stretching. The results of the fluid diversion process indicated an effective diversion of fluid in layered macroscopic model with permeability ratio of 8:1 in presence of SiO_2 and Al_2O_3 nanoparticles. The outcomes of this research is a major breakthrough in prospective field applications of nanoparticles-surfactant foams in oil-filled water-wet and oil-wet porous media.

ABSTRAK

Busa zarah nano dan surfaktan mempunyai aplikasi meluas dalam perolehan minyak tertingkat. Kelebihan sinergi melalui penurunan tegangan permukaan oleh surfaktan dan jerapan zarah nano di permukaan gelembung boleh menghasilkan busa dengan kebolehbusaan yang tinggi dan kestabilan busa yang lebih lama dalam menghasilkan takungan minyak. Namun begitu, kesan zarah nano kepada kestabilan statik dan dinamik busa belum dapat dieksplisitkan kerana kajian yang terhad. Selain itu, tidak banyak kajian yang mempertimbangkan mekanisme skala-liang bagi proses aliran busa dalam media liang dan pengurangan penjerapan surfaktan dengan kehadiran zarah nano. Oleh kerana penyelidikan yang terhad, kajian ini dijalankan untuk menentukan kesan zarah nano silika dioksida (SiO_2) dan alumina oksida (Al_2O_3) terhadap kestabilan busa pukal dan penjerapan surfaktan pada mineral lempung. Empat eksperimen utama yang dijalankan adalah kesan zarah nano terhadap jerapan surfaktan pada kaolinit, penilaian kestabilan busa pukal dan skala-gelembung, dengan kehadiran minyak dan garam, kajian pemerhatian skala-liang dalam model mikro gelas terukir, dan eksperimen proses lencongan bendalir dijalankan. Hasil kajian ini menunjukkan jerapan surfaktan pada mineral lempung berkurang secara mendadak sebanyak 40% dan 75% dengan kehadiran zarah nano masing-masing SiO_2 dan Al_2O_3 . Penjerapan maksimum surfaktan pada zarah nano berlaku pada 0.3 % berat sodium dodesil sulfat (SDS). Hasil daripada kestabilan busa pukal dan skala-gelembung menunjukkan peningkatan kestabilan busa pada 1% berat zarah nano SiO_2 dan Al_2O_3 dengan kehadiran minyak dan garam. Terdapat kepekatan garam peralihan yang melampaui kestabilan busa yang meningkat dengan peningkatan kepekatan garam. Kehadiran nano zarah Al_2O_3 dan SiO_2 menghalang kemasukan minyak yang diemulsi ke dalam lamela busa dan menurunkan kepekatan garam peralihan. Daripada hasil kajian skala-liang, mekanisme dominan pergerakan busa dalam sistem basah air adalah pembahagian lamela dan lamela gelembung-ke-multigelembung. Mekanisme dominan untuk pergerakan dan anjakan minyak baki oleh busa dalam sistem basah minyak dikenal pasti sebagai pembentukan liang yang merangkumi busa gas secara berterusan. Inter-gelembung memerangkap minyak dan air, lamela memisah dan meruntuhkan busa SDS yang dicerap dengan kehadiran minyak dalam sistem air-basah dan minyak-basah. Secara umum, pergerakan busa SDS- SiO_2 dan SDS- Al_2O_3 baik dalam sistem berisi minyak basah air dan minyak basah. Tautan gelembung dihalang semasa peregangan filem. Hasil proses lencongan bendalir menunjukkan pelencongan bendalir yang berkesan dalam model makro berlapis dengan nisbah ketertelapan 8:1 dengan kehadiran SiO_2 dan Al_2O_3 . Hasil kajian ini merupakan satu kejayaan utama dalam aplikasi bidang prospektif busa zarah nano-surfaktan dalam media liang berisi minyak basah air dan minyak basah..

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LIST OF SYMBOLS

$\gamma_{\alpha\beta}$	-	Interfacial tension
ΔP	-	The differential pressure
θ	-	The particle contact angle at the interface c
λ	-	Conductivity coefficient
μ	-	Viscosity. coefficient
σ	-	Surface tension coefficient
c_e	-	The equilibrium concentration.
c_o	-	The initial concentration of surfactant
K_f	-	The Freundlich adsorption capacity and intensity
K_L	-	The Langmuir equilibrium constant
K_r	-	Relative permeability
Γ_{max}	-	Maximum amount of surfactant
b	-	The gap thickness of the 2D Hele-shaw cell
W_r	-	The energy required to remove the particle from the interface
L	-	The length of cell
M	-	Mobility ratio
R	-	Radius of the particle
Γ	-	The amount of adsorbate adsorbed
S	-	Surfactant solution
U	-	The velocity of the foam
AI	-	Adsorption index

<i>NP</i>	-	Nanoparticles dispersion
<i>NPS</i>	-	Nanoparticles/surfactant mixtures
μ_{fapp}	-	Foam apparent viscosity

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CHAPTER 1

INTRODUCTION

1.1 Background of Study

Oil recovery from the petroleum reservoirs can be achieved by primary, secondary and tertiary oil recovery methods. Primary and secondary recovery methods depending on the reservoir characteristics, can only recover about 30 to 40 % of the original oil in place (Xing, 2012). Hence, the remaining oil in the petroleum reservoir remains the target of any enhanced oil recovery (EOR) operations such as gas injection, chemical injection, microbial enhanced oil recovery and thermal oil recovery. During enhanced oil recovery process, there is an improvement in the oil displacement and volumetric sweep efficiencies. This can be achieved through reduction of oil viscosity, capillary forces, interfacial tension and the development of a favorable mobility ratio between the displacing and the displaced fluid (Simjoo, 2012). This results in the eventual mobilization and the production of a substantial portion of the trapped residual oil in the reservoir at minimum cost (Payatakes, 1982).

Gas injection with about 39% contributions to world's EOR (Oil & Gas Journal, 2010) remains one of the most commonly used and generally accepted EOR methods. In gas injection, hydrocarbon and non-hydrocarbon gases like methane, air, carbon dioxide, natural gas and nitrogen are injected into the reservoirs for the recovery of residual oil (Liu *et al.*, 2011). Gas injection can either be miscible or an immiscible gas flooding. In miscible gas flooding, the gas is injected either at

minimum miscibility pressure (MMP) or beyond. Oil recovery is enhanced by the reduction of viscosity and interfacial tension as the injected gas mixes completely with the oil. In immiscible flooding, the injected gas does not mix with the reservoir oil. Reservoir pressure is maintained as the gas injection takes place below the minimum miscibility pressure (MMP) (Shokrollahi *et al.*, 2013). However, any gas enhanced oil recovery process suffers from poor macroscopic sweep efficiency because of gas higher mobility and lower density compared to oil or water (Rossen *et al.*, 2010). Gas segregation, gravity override, viscous fingering and channeling through the high permeability streaks are the major challenges of gas injection EOR process (Andrianov *et al.*, 2012).

In order to control the injected gas mobility and improve the poor volumetric efficiency during gas injection EOR, injection of gas slugs and water alternatively known as water-alternating gas (WAG process) has been used for several decades. The synergistic blend of the improved macroscopic sweep of waterflooding and the enhanced microscopic displacement efficiency of gas injection is exploited during WAG process (Sagir *et al.*, 2014). However, as WAG process continues, large volume of oil is considerably trapped by excess production of water that prevents the injected gas from contacting the resident oil in the reservoir. Moreover at some distances away from the wellbore, the process may lead to a poor gravity segregation control due to the large density contrast between the injected gases and the trapped oil (Sohrabi *et al.*, 2001). Consequently, vertical sweep efficiency and total oil recovery are drastically reduced as the process ultimately suffers from viscous instabilities and gravity segregation (Khalil and Asghari, 2006; Farajzadeh *et al.*, 2009).

Due to the inadequacy of WAG, foam, a dispersion of gas in liquid, such that the liquid phase is continuous and some part of the gas phase is made discontinuous by a thin liquid film called lamellae (Falls *et al.*, 1988) emerged in 1958 as a promising solution for controlling gas mobility. Foam controls gas mobility by increasing the apparent viscosity of the displacing fluid and reducing the relative permeability of the gas phase. In heterogeneous porous media, foam helps to divert the injected fluid from the high permeability regions to the low permeability un-swept areas by lowering the

gas mobility in the high permeability zones (Kovscek and Bertin, 2002; Skauge *et al.*, 2002; Blaker *et al.*, 2002). Results of previous studies show that foams apparent viscosities can be up to 1,000 times higher than that of their constituent phases (Zhu *et al.*, 2004; Liu *et al.*, 2005). Foam flooding are also more efficient than WAG process, waterflooding and gas flooding in reducing viscous fingering and improving sweep efficiency (Hirasaki and Lawson, 1985; Liu *et al.*, 2005).

Nevertheless, foams are thermodynamically unstable and require surface active agents for their continuous generation and stability. For effective foam applications in enhanced oil recovery process, the foam have to remain stable and be able to propagate in the reservoir in the presence of resident reservoir brines and oils and at high temperatures (Zhu *et al.*, 2004). Stable foams generation has been achieved using surfactants, polymer and proteins as the conventional foaming and stabilizing agents for several decades (Romero *et al.*, 2002; Murray and Ettelaie, 2004; Romero-Zerón *et al.*, 2010). It has been demonstrated experimentally that gaseous bubbles can be prevented from coalescing by the adsorption of surfactant, polymers and protein molecules at the gas–liquid interface of the foam (Rossen, 1996; Bournival *et al.*, 2014; Zhang *et al.*, 2015).

However, surfactant-stabilized foams, polymer enhanced foams and protein foams are unable to maintain their stability for a long time at reservoir conditions of high salinity, temperatures, and in the presence of oil in porous media. This is due to their high propensity to degrade and their low adhesion energy at the foam interface. Low adhesion of the stabilizing agents at foam lamellae promotes easy desorption and rapid film thinning of foam films (Carrier and Colin, 2003; Adkins *et al.*, 2007; Fameau and Salonen, 2014). The film thinning increases and the foam becomes drier as a result of liquid drainage from the foam films (Fameau and Salonen, 2014). The thinning of the foam films eventually results in foam coalescence, that is, the breaking of smaller unstable bubbles to form bigger bubbles (Carrier and Colin, 2003; Fameau and Salonen, 2014). For surfactant-stabilized foam, the rate of surfactants adsorption on rock surfaces can also be very high thereby reducing the amount of surfactant molecules available for stabilizing the gas-liquid interface of the foam.

Recently, there is an emerging interest in foam stabilized by a mixture of nanoparticles and surfactant. The synergistic advantage of interfacial tension and capillary forces reduction by the surfactant and nanoparticles adsorption at the foam lamellae is exploited for producing foam with high foamability and long time stability (Osei-Bonsu *et al.*, 2015). Results of some previous studies showed that nanoparticles-surfactant foams demonstrated high static and dynamic stability (Hunter, 2008; Cui *et al.*, 2010; Sun *et al.*, 2014; Singh and Mohanty, 2015). This has been attributed to the remarkable stability of the foam films due to the irreversible adsorption and aggregation of nanoparticles at the thin liquid films of the foam. Nanoparticles as the stabilizing components of the foam are solids; therefore, foams stabilized by nanoparticles-surfactant mixtures are more resistant to high salinity, temperatures, and the presence of resident reservoir brines and oils (Adkins *et al.*, 2007). The rate of surfactant adsorption on reservoir rock surfaces and clay minerals is also reduced in presence of nanoparticles (Ahmadi and Shadizadeh, 2013).

1.2 Problem Statement

The performance of foam also depends on the adsorption properties of the foaming agents in presence of resident reservoir brine in porous media. Inorganic salt influences the adsorption of surfactant molecules on clay minerals and at gas-liquid interface of surfactant-stabilized foam. The higher the adsorption of surfactant on clay minerals, the less the available surfactant molecules on the gas-liquid interface of the foam. Effects of different parameters on surfactant adsorption from solution onto reservoir rocks and clay minerals have been investigated in literatures (Zhang and Somasundaran, 2006; Sánchez-Martín *et al.*, 2008; Gogoi, 2009; Muherei *et al.*, 2009; Lv *et al.*, 2011; Amirianshoja *et al.*, 2013; Bera *et al.*, 2013). The results show that surfactants adsorption increases with increasing adsorbent dose, decreasing temperature and NaCl concentration due to their influence on the screening of the electrostatic charge (Behera *et al.*, 2014). However, these previous studies focused only on surfactant adsorption onto reservoir rocks and clay minerals. There is still paucity of information on the influence of electrolyte on the competitive and

co-operative adsorption of surfactant and nanoparticles onto reservoir clay. It is essential to gather information regarding the effect of salts on the adsorption of these foaming/stabilizing agents in order to optimize their performance for foam generation.

Another major concern for ensuring effective foam application in EOR is the stability of foam in the presence of oil. Jensen and Friedmann (1987) discovered from their studies that residual oil saturation of 15% and above in the reservoir will drastically affect foam propagation and performance. Foam stability in the presence of oil depends on aqueous phase composition, type of foaming and/or stabilizing agent, and oil type (Osei-Bonsu *et al.*, 2015). Generally, it has been reported from previous studies that oil has a destabilizing effect on the static and dynamic stability of foam (Vikingstad *et al.*, 2005; Simjoo *et al.*, 2013b; Duan *et al.*, 2014; Osei-Bonsu *et al.*, 2015; Farzaneh and Sohrabi, 2015). Results of these studies further showed that small-chain hydrocarbons with lower density and viscosity are more detrimental to the longevity of foams than long-chain hydrocarbons. Although the influence of oil on the stability of surfactant-stabilized foam has been widely investigated, few studies on the effects of oil on bulk stability of foams stabilized by nanoparticles–surfactant mixtures have been carried out. Thus, the role of nanoparticles on the static stability of surfactant foam in the presence of oil is yet to be well understood.

Porous media wettability is another critical parameter that influence foam stability and performance through their influence on fluid distribution and foam flow characteristics in porous media (Kulkarni and Rao, 2005; Talebian *et al.*, 2013). Results of previous experimental studies suggested divided opinions among researchers on the influence of porous media wettability on foam performance in porous media. Some researchers reported that the ideal reservoir rock wettability for optimum foam performance in porous media is water-wet (Kristiansen and Holt, 1992; Rossen, 1996). Others asserted that foam can be generated and propagated in an oil-wet porous media due to wettability alteration of hydrophobic porous medium to hydrophilic porous medium (Sanchez and Hazlett, 1992; Schramm and Mannhardt, 1996; Mannhardt, 1999). Few other researchers reported optimum foam generation, propagation and stability in oil-wet porous media due to lower surfactants adsorption in the oil-wet porous medium (Lescure and Claridge, 1986; Haugen *et al.*, 2012;

Romero-Zeron and Kantzas, 2007). These results are still contradictory and inconclusive and further studies will be required to obtain consistent results.

Meanwhile, most of the recent studies of nanoparticles-surfactant foams has been focused on either the bulk foam stability static experiments or the macroscopic studies (Yu et al., 2012a; Worthen *et al.*, 2013c; Singh and Mohanty, 2015; Farhadi *et al.*, 2016). The dominant mechanisms controlling the foam generation, propagation and stability in porous media especially in the presence of resident reservoir oils and brines are largely unknown due to limited studies. Knowledge of nanoparticles-surfactant foam propagation and stability in porous media at pore scale is vital for successful field design, application and implementation of nanoparticles-surfactant foam EOR.

1.3 Objectives of Study

The aim of this research is to determine the influence of silicon oxide (SiO_2) and aluminum oxide (Al_2O_3) nanoparticles on the static and dynamic stability of sodium dodecyl sulfate (SDS) foams and to carry out a pore scale mechanistic study of the nanoparticles-surfactant stabilized foam flow process in water-wet and oil-wet porous media. Thus the specific objectives of this study are as follows:

- I. To evaluate the influence of SiO_2 and Al_2O_3 nanoparticles on the adsorption of SDS surfactant by kaolinite at different salinities
- II. To determine the effect of nanoparticles concentration, salinity and oil presence on bulk and bubble scale stability of nanoparticles-surfactant foams
- III. To determine the mechanisms of nanoparticles-surfactant foam flow process at pore scale in water-wet and oil-wet porous media.
- IV. To investigate the role of nanoparticles on the process of fluid diversion by nanoparticles-surfactant foam in heterogeneous porous media.

1.4 Scope and Limitations of Study

This research comprises four main experiments which are surfactant adsorption experiments using two-phase titration method, bulk and bubble scale stability experiments conducted using foam column, dynamic foam analyzer and the 2D Hele-Shaw cell, pore scale visualization studies in the water-wet and oil-wet etched glass micromodels and fluid diversion experiments in unconsolidated visual layered glass bead packed macroscopic models. Some preliminary experiments were conducted in order to support and explain the observations and the results of the main experiments. These includes: surface tension measurements, determination of surfactant adsorption extent on the nanoparticles, determination of particle shape and wettability, determination of foam apparent viscosity in 2D Hele-Shaw cell, determination of foam lamellae thickness and morphology under the Leica EZ4 HD microscope.

The foam was pre-generated before injection into the porous media in all experiments in this study and all experiments were conducted at room temperature and pressure. The foam quality is limited to from 50 % to 90 %. The porosity of the etched glass micromodels ranges from 29 % to 40 % and the permeability ranges from 0.741 to 1.359 Darcy. The flowrate of 0.5 ml/hr (0.00833ml/min) was used in the pore scale visualization experiments. It was difficult to generate foam at lower flowrate than that in this study. The dead end pores investigation experiments were limited to the water-wet system. Influence of pore geometry in terms of aspect ratio and coordination number on the foam performance was not very significant due to the presence of dead end pores. It was difficult to determine any reasonable oil recovery at the production outlet from the diamond shaped micromodels due to its low pore volume (0.47 ml). The permeability contrast of the layered model is 8:1 while the porosity ranges from 30 % to 45 %. The flowrate of fluid diversion experiments could not translate into 2 ft/day at flow rate of 3ml/min-6ml/min. The contact angle of nanoparticles was measured in the absence of oil. Three major salts, NaCl, CaCl₂ and AlCl₃ were used in this research. These salts represent the major monovalent, divalent and trivalent cations, and the major anion found in reservoir brines.

1.5 Significance of Study

A micro-scale understanding of influence of nanoparticles on conventional foam stability and the displacement behaviours of nanoparticles-surfactant stabilized CO₂ foam in oil and water-wet porous media has been provided from the results of these experiments. This will provide the basic guidelines for further research, future field design and implementation of nanoparticles-surfactant CO₂ foam enhanced oil recovery (EOR) process.

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