FOAMS STABILIZED BY IN-SITU SURFACE ACTIVATION OF SILICA MICRO-PARTICLES WITH SURFACTANT

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To my beloved Mother and Father

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ABSTRACT

Concept of applying gas foam in EOR was first proposed for gas mobility control to alleviate the challenges of gas flooding and WAG process due to its high viscosity and low mobility. Like surfactant, colloidal particles can be employed as surface-active agent to stabilize the foams and can provide better stability due to its irreversible adsorption to the interface. To get ultra-stable particle stabilized foams/emulsions, the surface wettability of commercial raw colloidal particles should be modified (surface activation) from hydrophilic to partially hydrophobic. However, the studies on the in-situ surface activation for the micro-particles are yet to be widely performed. Therefore, the in-situ surface activation of unmodified CAB-O-SIL fumed silica micro-particles by using anionic surfactant such as Sodium dodecyl benzene sulfonate (SDBS) and alpha olefin sulfonate (AOS) has been studied. Its effects on the foamability and foam stability of dispersed solution were analyzed. The research methodology involves the characterization of aqueous foams, measurement of adsorption of particles at the air-water interface, and measurement of surface tensions. The independent variables are concentration of anionic surfactants, concentration of silica micro-particles whereas the dependent variables are foam volume, foam quality, surface tension, and percentage of particles adsorbed at interface. The results showed that hydrophilic silica micro-particles can be surface activated by interacting with anionic surfactants resulting to synergistic effect in foamability and foam stability. The positive charges on the silica micro-particles interact with negative charges of anionic surfactant to generate electrostatic interaction. This generates adsorption of surfactant at the particle-water interface making the particles to be surface active and bubble more stable. SDBS is more efficient anionic surfactant for the surface activation of silica micro-particles compared to that of AOS.

ABSTRAK

Konsep yang menggunakan buih gas dalam EOR diperkenalkan bagi kawalan pergerakan gas supaya dapat mengatasi masalah yang dihadapi dalam pembajiran gas dan proses WAG. Zarah koloid boleh digunakan sebagai ejen pengaktifan permukaan untuk menstabilkan buih dan boleh memberikan kestabilian buih yang lebih baik. Ini adalah disebabkan penjerapan zarah yang sehala di antara permukaan air dan udara. Bagi mendapatkan ultra stabil buih/emulsi yang dihasilkan oleh zarah-zarah, kebolehbasahan permukaan zarah koloid yang didapati dalam pasaran mesti diubah daripada hidrofilik kepada separuh hidrofobik. Namun Demikian, kajian tentang pengaktifan permukaan secara in-situ bagi miko-zarah belum dijalankan secara luas. Oleh itu, kajian tentang permukaan "CAB-O-SIL fumed silica" yang diaktifkan secara in-situ dengan surfactant anionik seperti "Sodium dodecyl benzene sulfonate" (SDBS) and "alpha olefin sulfonate" (AOS) dijalankan. Kesan-kesannya dalam keboleh-buihan dan kestabilan buih juga dikaji. The metodologi kajian sains melibatkan pencirian buih-buih, pengiraan peratusan penjerapan zarah di antara permukaan air-udara, pengiraan ketegangan permukaan. Pembolehubah bebas dalan kajian ini ialah kepekatan surfactant anionik, kepekatan silica mikro-zarah. Pembolehubah tak bebas ialah isipadu buih, kualiti buih, ketegangan permukaan, dan peratusan zarah yang terjerap antara permukaan air-udara. Keputusan menunjukkan bahawa silika mikro-zarah yang hidrofilik boleh diaktifkan permukaannya dengan menginteraksikan dengan surfactant anionik dan menyebabkan kesan sinergistik dalam keboleh-buihan dan kestabilan buih. Caj positif di permukaan silika mikro-zarah berinteraksi dengan caj negatif surfactant anionik dan menjanakan elektostatic interaksi. Interaksi ini menyebabhan penjerapan surfactant di permukaan antara air-udara dan juga mengaktifkan permukaan zarah dan menghasilkan buih yang lebih stabil. SDBS

dibuktikan bahawa lebih effektif sebagai ajen pengakitfan permukaan bagi silika surfactant mikro-zarah daripada AOS.

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LIST OF ABBREVIATIONS

AOS	-	alpha olefin sulfonate
CMC	-	Critical Micelle Concentration
CO_2	-	Carbon dioxide
EOR	-	Enhanced oil recovery
HLB	-	Hydrophile-lipophile
OPEC	-	Organization of petroleum exporting countries
ROS	-	Residual oil saturation
SDBS	-	Sodium dodecyl benzene sulfonate
WAG	-	Water alternate gas flooding

LIST OF SYMBOLS

C_{min}	-	Minimum concentration of surfactant needed to stabilize the foam
C_{min} (Q_b)	-	Minimum surfactant concentration needed to get stable foam based on the foam quality
C_{opt} (S_f)	-	Optimum surfactant concentration that contributes to maximum synergistic effect on foamability
C_{opt} (S_{fs})	-	Optimum surfactant concentration that contributes to maximum synergistic effect on foam stability
Θ	-	contact angle at the fluid-water interface
P_A	-	Liquid pressure at center lamella
P_B	-	Liquid pressure at Plateau border
P_{CA}	-	Capillary pressure at center lamella
P_{CB}	-	Capillary pressure at Plateau border
P_G	-	Pressure in gas- phase
Q_b	-	Foam quality
R	-	Radius of curvature
R_{1A}	-	radius of curvature at the lamella center
R_{1B}	-	radius of curvature at the plateau border
S_f	-	Synergistic effect on foamability (%)
S_{fs}	-	Synergistic effect on foam stability (%)
T	-	Room Temperature
V_0	-	Foam volume generated immediately after shaken
V_0^{P+S}	-	Foam volume generated immediately using surfactant with particles only after shaken

V_0^S	-	Foam volume generated immediately using surfactant only after shaken
V_{30}	-	Foam volume after 30 minutes
V_{30}^{P+S}	-	Foam volume generated using surfactant with particles only after 30 minutes
V_{30}^S	-	Foam volume generated using surfactant only after 30 miniutes
V_g	-	Gas foam volume
V_L	-	Liquid solution volume
w	-	Amount of silica micro-particles drained into the beaker
w_0	-	Amount of silica micro-particles initially added to the dispersion
x (%)	-	Percentage of silica particles adsorbed at the air-water interface
σ	-	Interfacial tension

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CHAPTER 1

INTRODUCTION

1.1 Background of Study

Energy demand increased significantly from 1996 to 2012 reaching 89.6 million barrels per day. Oil and gas has been the global leading energy generating resources. By 2035, it is expected by OPEC that the energy demand figure is increased to 109.7 million barrels per day. It is due to the growing energy demand from developing countries. Nowadays, the global oil supply is gradually outpaced by the global oil demand and it imposes a threat of energy security in the world. On the other hand, it becomes harder to discover new oil well to substitute the produced reserves (Alvarado and Manrique, 2010).

Therefore, enhanced oil recovery (EOR) methods are the key techniques applied to increase the oil recovery from the existing oil reservoirs, and to increase the oil field production life cycle by mobilizing the remaining trapped oil. Enhanced oil recovery (EOR) involves the injection of fluids that are not present in the underground reservoir to mobilize the immobile remaining oil (Lake, 1989). There are several common types of enhanced oil recovery techniques that are chemical flooding, thermal recovery processes, gas flooding, microbial enhanced oil recovery and vibro-seismic technology.

Gas flooding is one of the most widely applied EOR methods in field applications. Gas flooding is normally applied on the reservoir that has been water-flooded. The type of gas injected in this method can be hydrocarbon (light natural gas) and non-hydrocarbon. The non-hydrocarbon gas can be nitrogen, carbon dioxide, flue gas, hydrogen sulfide, and others. The fundamental mechanism of gas flooding method is to increase microscopic sweep efficiency of the oil displacement and reduce the residual oil saturation (Lake, 1989).

The main issues of the gas flooding are the viscous fingering and gravity segregation of displacing gas due to frontal instability. To mitigate these problems, water alternating gas injection (WAG) has been used in several field applications (Dicharry et al, 1973, Warner, 1977). One of the drawbacks is that the contact between the resident oil (displaced fluid) and injected gas (displacing fluid) is blocked by the large quantity of injected water leading to a larger ROS. This water-blocking phenomenon is more detrimental in water-wet reservoir or the reservoir that has been water-flooded previously as secondary recovery technique (Stalkup, 1970). Some of the injected gas (such as CO_2) has high solubility in water and it increases the consumption of that gas making the project less economically feasible.

Concept of applying gas foam for mobility control was first proposed by Bond and Holbrook (1958). To alleviate the challenges of gas flooding and WAG process, foam flooding using the injected gas can be the potential solution to it (Fried, 1961; Kovscek and Radke, 1994; Rossen, 1996; Du *et al.*, 2007; Farajzadeh *et al.*, 2009). It is very useful applying foams in EOR techniques to improve displacement efficiency because of its high viscosity and mobility during the displacement across a porous a medium (Green *et. al.*, 1998). Extensive experimental studies were performed on using surfactant as the foam generation agent to stabilize the foam to reduce the gas mobility in gas flooding (Kim *et al.*, 2005). However, there are several weaknesses on surfactant-stabilized foam that are high surfactant retention in porous media, relatively low foam stability, and degradation of surfactant at reservoir condition (Kim *et al.*, 2004; Ransohoff *et al.*, 1988; Wang, 1984).

Solid particles can be employed as surface-active agent to stabilize the foams besides surfactant. Like the surfactant, the solid particles can be adsorbed at the interface between aqueous and non-aqueous phases, which makes it surface active. The solid-stabilized foams have been applied in a variety of industries due to its higher adhesion energy for the particle adsorption at the foam interface. Surfactants and solid particles have been compared for their ability to stabilize foams/emulsions (Binks *et al.*, 2002, Horozoy 2008). Furthermore, colloidal particles can provide better stability because when the colloidal particles are held at the interface, the adsorption can be irreversible and the particles are not easily desorbed from the interface. Bink and Horozov (2005) reported that a particle with size of larger than 100 nm could be irreversibly adsorbed on the interface. Furthermore, Wilson (1980), and Sun and Gao (2002) reported that solid micro-particles with size of several μm could be employed as the foam stabilizer to generate stable foam or emulsion dispersion.

However, the raw commercial particles are normally not surface active at most of the liquid interfaces. For silica particles, they are normally extremely hydrophilic. To get ultra-stable particle stabilized foams/emulsions, the particle surface must be activated by the wettability modification. The surface activation can be performed by several methods: homogeneous surface coating, Janus particles and in-situ surface activation. Homogeneous surface coating can be done on the silica by having surface silylation to modify the surface hydrophobicity to be more hydrophobic but it costs highly in commercial scales. Janus particles is the particles at which part of surface is coated to be hydrophilic and part of the surface is treated to be hydrophobic but there is a limitation to have large commercial scale of production. In-situ surface activation is the well – known as an easy wettability modification method, which is to just interact the particles with amphiphilic compounds. In-situ surface activation can be relatively less complicated and much cheaper methods to produce the surface-active particles.

Cui *et al.* (2010) highlighted that the unmodified silica particle can be surface activated by the anionic surfactant. The anionic surfactant can provide the negative charges to have electrostatic interaction and form monolayer adsorption of the surfactant at the particle-water interface. The electrostatic interaction modifies the surface properties of the particle from hydrophilic to partially hydrophobic forming an ultra-stable foam or emulsion stabilizer.

1.2 Problem Statement

The implication of foam in mobility control was brought up by Bond and Holbrook (1958). One of the foaming agents commonly used is surfactant. There were many extensive research efforts carried out on surfactant-stabilized foams and they presented several weaknesses that are unstable foam properties, high surfactant retention in porous media and surfactant degradation at reservoir condition (Kim *et al.*, 2004; Ransohoff *et al.*, 1988; Wang, 1984). The surfactants of small molecular weight had been widely researched and applied in the industries; solid particulate stabilizers pique the interest of researchers in their applications.

Bink and Horozov (2005) reported that a particle with size of larger than 100 nm could be irreversibly adsorbed on the interface. Furthermore, Binks (2005) highlighted that there are only two examples in which solid micro-particles are used as foam stabilizers. The functions of micro-particles were reported by Wilson (1980), and Sun and Gao (2002) to be used as the effective foam stabilizer to generate stable foam or emulsion dispersion. However, the raw commercial micro-particles are not surface-active and highly hydrophilic. To obtain ultra-stable particle stabilized foam or emulsion, the surface properties of the particles must be modified to be partially hydrophobic. Cui *et al.* (2010) shows that in-situ surface activation with anionic surfactant can efficiently increase the foamability and foam stability of the particles. However, the studies on the in-situ surface activation for the micro-particles are not widely performed.

1.3 Objectives

In this research, anionic surfactants such as Sodium dodecyl benzene sulfonate (SDBS) and alpha olefin sulfonate (AOS) are used as the in-situ surface activation agent for unmodified CAB-O-SIL fumed silicas with size range of 0.1 – 0.3 μm. The main objectives of this study are:

- To investigate the impact of the treated micro-particles on the foamability and foam stability
- To compare anionic surfactants SDBS and AOS and identify which of it is more efficient.

1.4 Scope of Work

The focus of this study is to examine the behavior of anionic surfactants on the foamability and foam stability without and with the presence of micro-particles. It is also to study the in-situ surface activation of the untreated CAB-O-SIL fumed silicas (micro-particles) by interaction with anionic surfactant, and to assess the impact on the foamability and foam stability of the aqueous dispersions.

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