



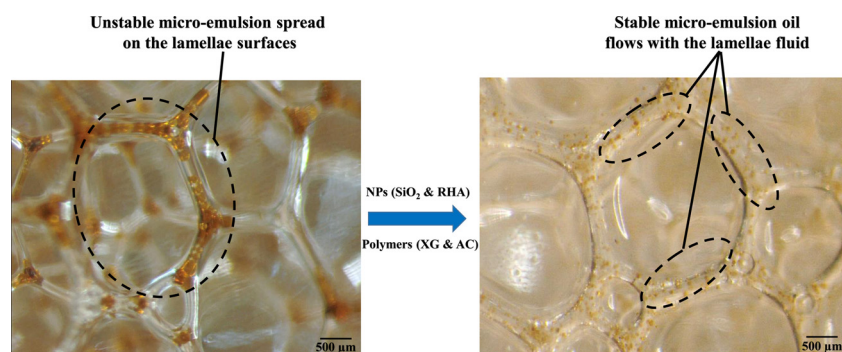
Nanoparticle/polymer-enhanced alpha olefin sulfonate solution for foam generation in the presence of oil phase at high temperature conditions

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GRAPHICAL ABSTRACT



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ABSTRACT

Foam flooding can improve oil recovery performance through reducing gas mobility in subsurface porous media. However, foam in porous media is thermodynamically unstable, especially in reservoir conditions where it is in contact with hydrocarbons (oil phase). In this study, the use of silica (SiO_2) and rice husk ash (RHA) nanoparticles with xanthan gum (XG) and acacia gum (AG) polymers, is proposed to improve the foam stability at high temperature and high salinity conditions in the presence of oil phase. Moreover, it was highlighted that the potential use of RHA, extracted from waste materials, and natural acacia gum can substitute previously proposed synthesized nanoparticles and polymers as foam additives for immiscible displacement in subsurface porous media.

The results show that the addition of polymers do not change the surface tension at high temperatures, and its combination with nanoparticles can increase the viscosity of surfactant remarkably. It was found that an increase in the stability of CO_2 -foam can be achieved with increasing nanoparticles and polymer concentrations to optimum concentrations. Furthermore, foam stability increases with decreasing the nanoparticles sizes and increasing the molecular weight of the polymer.

In order to understand the performance of the optimum foam composition at high temperature and salinity conditions, the foam was brought into contact with a reservoir oil. It was shown that the CO_2 -foam stability in the presence of oil, could be enhanced by using nanoparticles and polymers, compared to the CO_2 -foam system with no additives. The reason for such improvement is due to the presence of nanoparticles and polymers in lamellae that break the oil into the emulsion droplets system that can flow easily through the lamellae without draining the entire surfactant solution from them.

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1. Introduction

Gas injection has been used as one of the methods to increase oil production in many reservoirs [1–3], in which gases such as carbon dioxide, nitrogen, methane, and hydrogen sulfide are injected into the subsurface reservoirs to overcome capillary and viscous forces in porous media and push the oil out of pores [4–6]. Carbon dioxide (CO₂) has been used for oil recovery from conventional reservoirs, and it is proposed for heavy oil recovery processes, this is due to its non-flammable and non-toxic nature, and economic viability [7–9]. Besides that, one of the solutions to mitigate greenhouse gas emissions is to use it in enhanced oil recovery (EOR) processes and store it in depleted oil reservoirs.

The drawbacks of CO₂ injection into subsurface reservoirs are related to the low density and viscosity of CO₂ which lead to gas channelling and its early breakthrough during EOR processes that means a lower sweep efficiency may be achieved [8,10]. To overcome these drawbacks, foams have been introduced where the co-injection of a gas and surfactant solution could reduce gas mobility (i.e., increasing apparent gas viscosity) and in turn improves the oil sweep efficiency [11].

Foam is developed by diffusing a large amount of gas into a continuous liquid phase and generating gas bubbles that are surrounded by liquid films called lamellae, which directs gas to flow through the low-permeability regions and leads to a reduced gas fingering and improved sweep efficiency in EOR processes [6,12].

There are many factors influencing the success of a foam flooding process, such as the type of surfactant, reservoir temperature, salinity, pressure, wettability, surface adsorption, and the presence of oil. Therefore, foam is classified as a metastable structure by its nature at reservoir conditions and this is due to the drainage of liquid surfactant from the lamellae that causes an increase in capillary pressure and decreases the lamellae thickness, that eventually initiates the foam coalescence and lamellae rupture [13].

The reservoir hydrocarbons introduce an additional detrimental effect on foam stability. Many investigators studied the effect of reservoir oil on foam properties [14–16]. Generally, the presence of oil decreases the foam stability by rupturing bubbles through the spreading of oil droplet on the gas-liquid interface in the foam lamellae. However, foam remains stable if the presented oil creates a stable pseudoemulsion film when the oil and gas phases are separated by the surfactant solution. Therefore, the foam stability in the presence of reservoir hydrocarbons is governed by the stability of pseudoemulsion phase [16,17].

One of the proposed solutions to enhance the foam stability is the use of nanoparticles in the foam structure. Recently, nanotechnology has been introduced in the oil and gas industry as promising solutions in well and reservoir engineering concepts, especially for enhanced oil recovery methods [18–20]. Since surfactants are not able to maintain the foam stability for a long period of time at reservoir conditions in the presence of oil, nanoparticles have been used with surfactants to increase the foam stability [18,21]. Nanoparticles have very small sizes, which can easily move without plugging the rock pores, and they are sustainable materials at reservoir conditions such as heightened temperatures and salinities [22–24].

Manan et al. [25] conducted several experiments to measure the foam stability in the presence of different types and concentrations of nanoparticles such as silicon dioxide, aluminium oxide, copper oxide and titanium dioxide in alpha olefin sulfonate (AOS) surfactant. The stability was tested using Ross-Miles method using a half-life test at ambient temperature and atmospheric pressure. The results revealed that all types of nanoparticles improved the stability of CO₂-foam at low concentrations of nanoparticles, and aluminium oxide nanoparticles with a concentration of 0.1 wt% showed higher stability compared to others. Moreover, the results from displacement tests that were conducted on sand packs confirmed the enhancement achieved by using nanoparticles.

The main parameters that control the stability of foam with

nanoparticles are: (1) the large adhesion energy between nanoparticles and thin liquid films of the foam that allows the particles to cling with each other in the foam interface which protects the foam bubble from the rupture or coalescence [24,26], (2) the presence of nanoparticles can increase the lamellae thickness, which increase the foam stability [27], (3) nanoparticles helps to decrease the liquid drainage from foam lamellae, through creating monolayer or a gel structure inside the liquid lamellae [28].

Alzobaidi et al. [29] demonstrated nanoparticles have the ability to lower the interfacial tension at the gas-liquid interface, thus reduce the capillary pressure across the gas-liquid interface, which in turn increase the foam stability. The improved foam stability was attributed by the increase of the foam lamellae thickness, due to the adsorption and aggregation of nanoparticles between the bubbles in thin liquid films, therefore reducing the capillary pressure between the gas-liquid interface [27]. Thus, the lower the capillary pressure the higher the foam stability and the lower the bubbles coalescence rate.

According to the findings by Espinoza et al. [24], 5.0 nm silica nanoparticles created a supercritical stabilised CO₂-foam with an improved foam viscosity. Different ranges of nanoparticle concentrations, salinities, flow rates and temperatures were tested with supercritical CO₂. The results revealed that silica nanoparticles with the concentration of 0.05 wt%, creates a stable CO₂-foam at high-temperature conditions with a mobility ratio eighteen times lower than the foam generated by the surfactant solution and supercritical CO₂ without any nanoparticles. Also, similar results were reported in a study by Zhang et al. [30], in which they confirmed that CO₂ in a surfactant solution with surface treated silica, can generate stable foams that will sustain at harsh conditions such as high temperatures and high salinities. Furthermore, other studies demonstrated that as the concentration of the silica nanoparticles increases, the stability of supercritical CO₂-foam improves further.

Guo and Aryana [31] investigated the possibility of using nanoclay and silica nanoparticles with pure and mixed surfactants of AOS and Lauramidopropyl Betaine (LAPB), which is a Zwitterionic type, to improve the foam stability. Foam behaviour was evaluated by using modified bulk foam tests for foamability and the foam stability, and by using a microfluidic 2-D model for foam flow in porous media. The results of the foamability test showed that silica nanoparticles or nanoclay with AOS have the best foamability response. Therefore, nanoclay can be the best alternative to silica nanoparticles in improving foam properties for EOR processes. In addition, the mixed solution of AOS and LAPB was not effective compared to individual surfactants, as the mixed solution of AOS and LAPB showed a reduction in foamability in the presence of silica nanoparticles and nanoclay.

Nanoparticles from waste products were also used to increase the foam stability, which are considered as environmentally-friendly solutions to dispose and sequester waste products in oil reservoirs. Some of these waste products are rice husk ash (RHA) and fly ash (FA) nanoparticles that can be used to stabilise the CO₂-foam and improve foam mobility. Several studies have addressed the effect of using FA NPs to improve the foam stability, however the use of RHA NPs to improve the foam stability has not been investigated to a high extend, and only a few studies have reported their use in the literature. RHA is a natural material that is generated from rice milling and is converted into ash during the firing process, it contains up to 90% amorphous silica [32]. In a recent study by Jie et al. [33], they demonstrated that using RHA NPs can generate stable foams in the presence and absence of oil. Their bulk foam stability tests revealed that RHA NPs improved the foam stability and increased the half-life time from 123 min to 145 min, when the RHA concentration increased from 0.1 wt% to 0.9 wt%. Additionally, the RHA-enhanced foam demonstrated slightly better stability compared to the commercial silica NPs. However, for both types of nanoparticles, the presence of oil had detrimental effects on the half-life time, with less detrimental effect in the presence of silica NPs.

Singh et al. [34] reported a more stabilised foam generated by

carbon dioxide, and thermally treated FA nanoparticles that were produced from coal power plants. The foam texture study demonstrated that using non-ionic or anionic surfactants and carbon dioxide with thermally treated FA nanoparticles, can generate small bubbles and strong foam in porous media. Also, in Berea core, the study showed that a great improvement in stability of CO₂-foam can be achieved with an even lower concentrations of thermally treated FA nanoparticles in the absence of oil [30]. Moreover, Eftekhari et al [35] tested the possibility of using FA nanoparticles to stabilise foam for EOR applications using bulk and porous media tests. The results showed that with a small concentration of FA nanoparticles, foam stability was significantly increased in the presence of oil. In addition, since the oil with surfactant solution generated stable emulsions when FA nanoparticles were added, a strong foam structure was formed and a high oil recovery from core samples was achieved.

Many researchers investigated and introduced different types of polymers to improve the viscosity of foam and its stability, and in turn enhance the mobility ratio and increase the oil recovery factor [13,36,37]. Ahmed et al. [13] reported improvements in the CO₂-foam stability by using conventional and associative polymers. CO₂-foam was generated by using AOS surfactant with 3.0 wt% of NaCl, 0.5 wt% betaine as foam stabilizer, and 0.2 wt% of hydrolysed polyacrylamide and associative polymers (superpusher) at 80 °C and atmospheric pressure. The results proved that the generated CO₂-foam without polymer was unstable and it was decayed faster than other foams in which polymers were used. This is due to low viscosity of liquid solution of CO₂-foam without polymer, which makes the liquid drainage fast and lamellae become thin. By using polymer, the viscosity was increased, and the foam stability was improved with a better stability through using associative polymer compared to conventional polymer (reported half-lives were 1876 and 1500 s for associative and conventional polymers, respectively). Acacia gum (gum Arabic) has also been used as a type of polymer in enhanced oil recovery processes [38–40], and it has a similar molecular structure as xanthan gum. Solomon et al. [41] conducted core flood experiments using different polymers (xanthan gum and acacia gum) with sodium hydroxide and a surfactant (Shell Enordet 0242), in an alkali/surfactant/polymer (ASP) flood process. The study demonstrated that xanthan gum in the ASP process provided a stable front displacement than acacia gum, thus high acacia gum concentration was recommended to achieve better displacement process.

The synergistic effects of nanoparticles and polymers with a surfactant have been studied by many researchers in recent years. Emrani and Nasr-El-din in 2017 studied the effect of nanoparticles and polymers on the CO₂-foam stability to be used as a hydraulic fracturing fluid [42]. The results showed that the foam stability and its mobility reduction were improved when guar gum and silica nanoparticles were added to alpha olefin sulfonate solution. Silica nanoparticles covered the surface of bubbles and made them more stable at high temperature and salinity conditions while polymer decreased the liquid flow rate

from lamellae by increasing its viscosity. In addition, Kalyanaraman et al. [43] studied the effect of using nanoparticles with a polymer on the improvement of foam rheology. A surfactant solution containing polyelectrolytes (PEI) and polyelectrolyte complex nanoparticles was used to generate CO₂-foam. The results revealed that using polyelectrolyte complex nanoparticles (PECNP) produced a stable foam in the presence of oil while the foam viscosity was improved using both the PEI and PECNP surfactants.

Most of the studies to date, discussed the CO₂-foam stability at harsh conditions in the presence of nanoparticles or polymers, and only few studies have addressed the synergetic effect and properties of nanoparticles and polymers on the foam stability for EOR processes at high temperature and salinity conditions. However, the effect of these additives on the foam stability in the presence of oil phase was not extensively investigated.

Therefore, in this study we investigated the properties of nanoparticles/polymers enhanced foams at high temperature and salinity conditions in the presence of oil. We used different nanoparticles (silica and rice husk ash), and polymers (xanthan gum and acacia gum) to determine the CO₂-foam stability. Then, the effects of oil phase on the foam stability at high temperature and salinity conditions were presented.

2. Experimental setup and procedure

2.1. Materials

An anionic surfactant (alpha olefin sulfonate, AOS) with a viscosity of 1.0 cp at 22 °C, and pH of 7.0–8.0, was used in this study. AOS is widely used in EOR processes to provide a solution with low interfacial tension and low adsorption rate on rock surfaces. The salinity of 2.0 wt % of NaCl was adjusted by adding sodium chloride with a purity greater than 99%. Carbon dioxide (CO₂) with a purity greater than 99% was used as our gas phase, and distilled water was mixed with surfactants in all the experiments.

Fumed silica (SiO₂), and rice husk ash (RHA) are two types of nanoparticles were used in this study. Silica nanoparticles have a molecular weight of 60.08 g/mol, and an average particle diameter of 7.0 nm. However, when silica NPs dispersed in the water solution, the Z-average particles diameter was increased to 133 nm as shown in Fig. 1 (particles size distributions for silica and RHA NPs were measured using zetasizer nano-zs). This is due to the large surface area of silica NPs and the process used to produce them, which favour the nanoparticles to form stable aggregates in aqueous suspension even after the ultrasonic treatment [44]. The rice husk ash nanoparticles are a by-product of burning rice husk, with a bulk density of 550–650 kg/m³, pH of 7.10. The RHA sample had large particles sizes and they were decreased by using the ball milling process, the Z-average particle size was 420 nm. As shown in Fig. 1 the size distribution by intensity of RHA

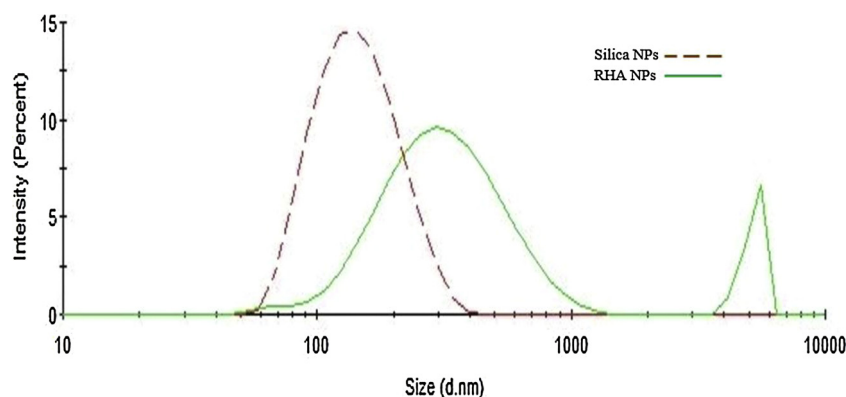


Fig. 1. Size distribution by intensity of Silica and RHA NPs.

(green curve) has two peaks recorded as 350 nm for peak 1, and 500 nm for peak 2.

Two types of polymers with different molecular weights (xanthan gum and acacia gum) were used to find their impacts on foam stability at high-temperature conditions.

In our experiments, xanthan gum polymer (XG) has a molecular weight of 3.2 million g/mol and acacia gum (AG) has a molecular weight of 1.0 million g/mol (molecular weights were measured using zetasizer nano-zs).

To investigate the impact of oil phase on the foam stability, a North Sea oil sample with an API degree of 24.75 and a viscosity of 54.71 cP at 23 °C (7.99 cP at 70 °C) was used.

2.2. Sample preparation

To prepare a solution that includes AOS surfactant, NaCl, nanoparticles and polymers, different steps were taken in a specific order. Firstly, 2.0 wt% NaCl was added to 200 ml of distilled water, then required mass of nanoparticles (0.05, 0.2, 0.5 and 1.0 wt%) were mixed with the solution for one hour using a magnetic stirrer (all weight percentages are referenced to the weight of 200 ml of distilled water). Then the sample was placed in an ultrasonic water bath at the temperature of 35 °C for a period of two hours, this process helps in further agitation and dispersion of the nanoparticles into distilled water. The ultrasonic bath is normally used to generate high shear forces to disperse the nanoparticles when small particles are mixed in water, this avoids any precipitation and agglomeration of them in the solution [44]. Then required masses of AOS surfactant (0.5 wt%) and polymers (0.1, 0.3 and 0.5 wt %) were added to the solution of brine and nanoparticles, and the sample was stirred for a period of 24 h with the magnetic stirrer.

2.3. Surface tension and viscosity

Surface tension of the surfactant solutions was measured using Sigma tensiometer 703D based on DuNouy ring method with different concentrations of nanoparticles at different temperatures. Furthermore, viscosity of the surfactant solutions and their shear rate profiles with different concentrations of nanoparticles and polymers at different temperatures were measured using Rheometer DHR-3.

2.4. Bulk foam stability test

There are different standard methods to evaluate the foamability of surfactants and foam stability in bulk and porous media. In bulk foam tests the main criteria are the foam height and its half-life time [25], and in foam tests in porous media the main criteria are the pressure gradient, viscosity and mobility reduction factor [45].

Although the column test has been used in many studies to evaluate foam stability and foamability, it is difficult to use the column test to analyse the foam stability at high-temperature conditions. Therefore, bottle test is selected in this research to test the foam stability and foamability.

Bottle test, ASTM D3601, is used to measure the foamability and foam stability with different concentrations of additives at high temperature conditions. The test requires a 16 oz. Boston round bottle with the height of 169 mm and diameter of 75 mm. A transparent waterproof tape label (marked from 0 to 169 mm) was fixed to the outside of the bottle to monitor the foam height. The foam solution is poured into the bottle and placed in a water bath, consisting of a beaker that is filled with water and placed on a hot plate for one hour at the desired temperature (85 °C), as shown in Fig. 2. Then the solution was removed from the water bath and the CO₂ was injected into the surfactant solution for two minutes. At this time the height of the liquid–CO₂ interface was recorded, and it is the initial liquid height. The bottle was returned to the water bath for 30 min. The bottle was then shaken at a

speed of 40 shakes per 10 s with a minimum 8-inch stroke. After the shaking process, the maximum total height is recorded (total foam height at time zero) and the timer was started. The bottle was placed back into the water bath and remained undisturbed for the rest of test period. The time required for the foam to decay to the initial height was recorded and analysed as foam stability criteria.

The bubble size distribution and foam texture were investigated by using an Olympus microscope SZX100 and ImageJ software.

The foam stability can be analysed by using Eqs. (1) and (2) which show the maximum foam height and normalized foam height, respectively:

$$F_t = M_t - I \quad (1)$$

$$F_{\text{hnt}} = \frac{F_t}{F_{t0}} \quad (2)$$

where, F_t , is the foam height at time t ,

M_t , is the total height of liquid and foam at time t ,

I , is the height of liquid at time t ,

F_{hnt} , is the normalized foam height at time t ,

and F_{t0} , is the total foam height at the beginning of the test.

3. Results and discussion

3.1. Surfactant concentrations and the CMC value

The Critical Micelle Concentration, CMC, value for AOS was identified using a DuNouy ring tensiometer at 25 °C and 85 °C. As it can be seen from Fig. 3, by increasing the concentration of AOS surfactant, surface tension decreases continuously until it reaches the CMC point (0.1 wt%); in which the surface tension is 29.3 mN/m, and 25.6 mN/m at 25 °C and 85 °C, respectively. As the temperature increases, the kinetic energy of the molecules increases, which leads to a decrease in the attraction forces between the molecules at the interface. Furthermore, this allows a higher number of surfactant molecules to be present at the liquid-gas interface, thus a lower surface tension can be achieved [47–49].

The impacts of different additives on the surface tension of the AOS solution at different temperatures were measured. To investigate the effect of NaCl, nanoparticles, and polymers on the surfactant surface tension, the AOS solution was mixed with NaCl at optimum concentrations of nanoparticles (SiO₂ and RHA), and polymers (XG and AG). The surface tension values of the mixtures of 0.5 wt% AOS, 2.0 wt % NaCl, 0.2 wt% SiO₂ NPs, and 0.2 wt% RHA NPs are shown in Fig. 4. Adding NaCl decreased the surface tension at 25 °C, as the salt reduces the electrostatic repulsion between the surfactant ionised head-groups, in which surfactant head groups will be expelled to the surface, therefore, it reduces the surface tension [50,51].

At higher temperatures, besides the effect of salinity, the temperature can further decrease the surface tension, due to the increase in kinetic energy of the AOS molecules. Therefore, in the optimum mixture solution, the surface tension was decreased due to the dominant effects of salinity and temperature.

Fig. 5 shows the influence of adding XG on increasing the AOS surface tension at 25 °C, where the surface tension was increased from 32.4 mN/m to 35.2 mN/m by adding 0.3 wt% XG to the AOS solution, and in the presence of NaCl the surface tension increased from 29.4 mN/m to 34.5 mN/m when adding XG and NPs. This is because of the reduced mobility of surfactant molecules in the presence of XG due to the association of surfactant and polymers, and a high viscosity of the solution, which led to an increase in the surface tension of the AOS solution [52]. The effect of XG was minimised at 85 °C as the surface tension of the AOS solution and other additives with XG was dramatically decreased from 35.2 mN/m to 29.0 mN/m when the temperature raised from 25 °C to 85 °C.

The impact of AG on the surface tension is shown in Fig. 6. It has no

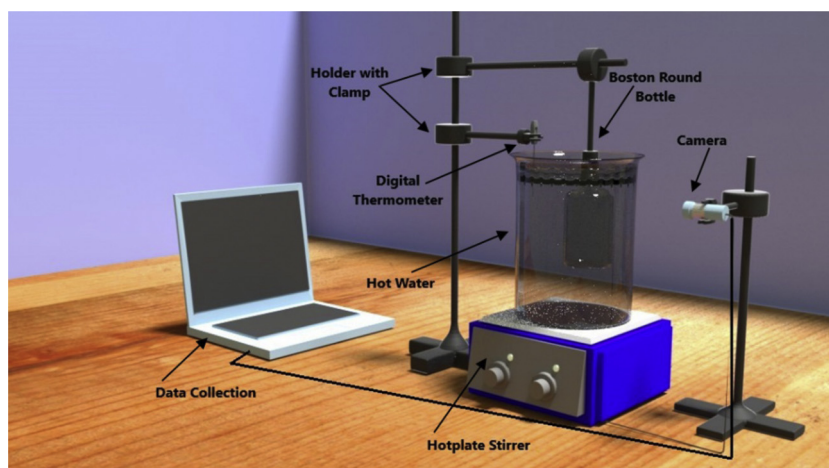


Fig. 2. Schematic of the bottle test experiment [46].

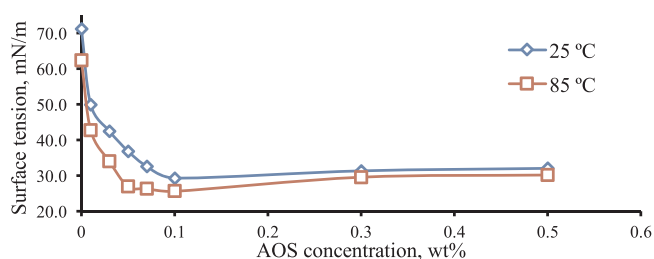


Fig. 3. Surface tension of AOS surfactant at different temperatures.

remarkable effect on the surface tension of the saline solution at 25 °C and 85 °C. The AOS surfactant molecules occupied the gas-liquid interface and decreased the surface tension, without any influence from the AG molecules, thus adding such polymers will not affect the surface tension.

3.2. Surfactant viscosity measurement

Viscosity versus shear rate was measured for the solutions with the optimum concentration of XG and AG (0.3 wt%), the surfactant concentration of 0.5 wt%, the salinity of 2.0 wt% and the silica/RHA NPs concentration of 0.2 wt%, at the temperature of 85 °C. As shown in Figs. 7 and 8, viscosities of the solutions were gradually decreased with increasing shear rates, demonstrating shear-thinning behaviours. Overall, the solutions of XG and AG were showed higher viscosities than the AOS solution, with a higher value in the XG solution compared to the AG solution.

According to the results shown in Figs. 7 and 8, adding AOS, NaCl, and SiO₂/RHA NPs to the XG or AG solutions slightly decreased the viscosity of them at low shear rates, and the changes in viscosity of the solutions were negligible at higher shear rates. The reduction in viscosity is attributed to the addition of NaCl as a result of weakening intermolecular forces between the polymer molecules, and limiting the

molecules extension therefore, it decreases the solution viscosity.

The XG solutions with and without additives showed higher viscosities than the AG solutions in all shear rates, this can be due to the molecular weight of XG which is much higher than the molecular weight of AG. Therefore, using XG as an additive with the AOS solution to generate CO₂-foam will lead to an increase in the CO₂-foam apparent viscosity.

3.3. Bulk foam stability

To verify the capability of the bottle test in measuring the foam stability, two foam stability experiments were conducted using the column and bottle tests with 0.5 wt% AOS surfactant at ambient temperature (25.6 °C). In the column test, 200 ml of the AOS surfactant was placed in a 1000-mL graduated glass cylinder, using the ASTM D 1173 method foam was generated by CO₂ injection through the porous stone (pore diameter 0.2 μm) at the pressure of 1.5 bar. Fig. 9 shows the normalised foam height versus time for the bottle and column tests using the AOS solution. The normalised foam heights are reasonably comparable for both test methods. The two tests were repeated to confirm that they can predict the behaviour of foam in a consistent way.

Furthermore, the bubble-size distributions after five minutes for both methods are shown in

Fig. 10. Based on the results from ImageJ analysis, foam bubble-size distributions from both methods showed reasonably similar trends. Through these analyses we could conclude that the predictions and results of foam stability and texture analysis by these methods are comparable.

The AOS solution with a concentration of 0.5 wt% (five times the critical micelle concentration, CMC) was used at 26.1 and 85 °C with high-salinity condition, to determine the effect of temperature and salinity on the AOS foam stability. In these experiments, the foam stability was determined by monitoring the normalized foam height as a function of time.

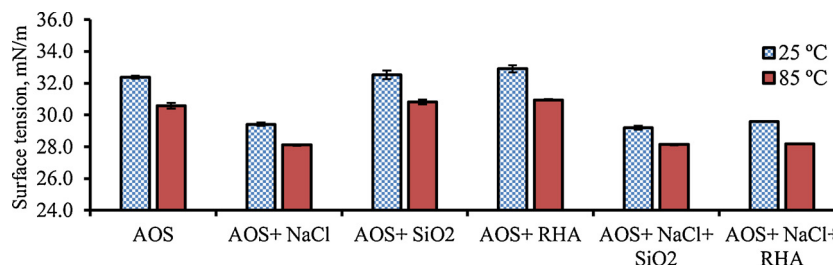


Fig. 4. Surface tension of the AOS solution with 2.0 wt% of NaCl and 0.2 wt% of SiO₂ NPs and RHA NPs.

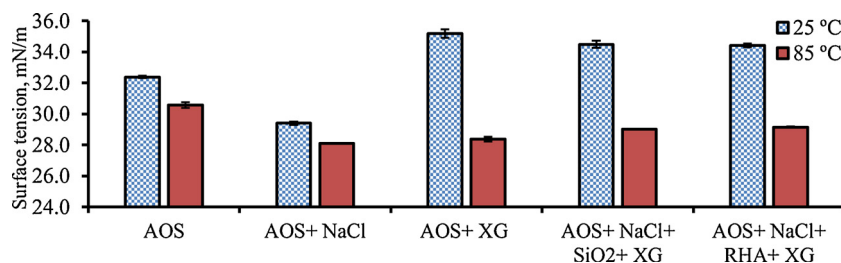


Fig. 5. Surface tension of the AOS solution with and without 2.0 wt% of NaCl, 0.2 wt% of SiO₂ NPs / RHA NPs, and 0.3 wt% XG.

The results revealed that foam stability for the foam produced by the AOS were sharply decreased at a higher temperature as shown in Fig. 11. This is due to the increase in the drainage rate from thin lamellae, which caused a higher bubble coalescence rate. When salinity of the AOS solution was increased at different temperatures, the foam stability was further decreased. The results showed that adding 2.0 wt% of NaCl affected the foam half-life time drastically (half-life is the time taken to reach to the half of the initial foam height after its generation), from 5420 s to 360 s at 26.1 °C. The change in the half-life time was from 261 s to 100 s when the temperature was 85 °C. This behaviour could be justified as salt ions can attract polar molecules of water and repel surfactant molecules out of the solution, which in turn reduces the concentration of surfactant molecules in the solution and destabilises foams.

3.4. Nanoparticle-stabilized CO₂-foam

To investigate the performance of nanoparticle-stabilized CO₂-foam, static foam experiments were conducted using the bottle test method with different concentrations of SiO₂ and RHA nanoparticles at the salinity of 2 wt% (NaCl) and the temperature of 85 °C.

As shown in Fig. 12, the CO₂-foam stability increases with increasing the concentration of SiO₂ and RHA NPs up to a certain concentration. Nanoparticles at low concentrations (0.05 and 0.1 wt %) generated foams with lower stability and half-life time compared to the higher concentrations. At lower concentrations of nanoparticles, their presence at the interface was not enough to prevent the liquid drainage and improve the stability.

At the concentration of 0.2 wt% of the nanoparticles, stable foams were generated with half-life and total decay times of 420 and 1800 s for SiO₂ NPs, and 320 and 1200 s for RHA NPs, respectively. Therefore, the concentration of 0.2 wt% was considered as an optimum concentration for both types of nanoparticles. At this concentration, the drainage rate from the thin lamellae was reduced by enough nanoparticles accumulated at the gas-liquid interface and plateau boarders. However, beyond this concentration, the effect of increasing nanoparticles concentration was deteriorating to the foam stability. As, the concentration of nanoparticles increases, more flocs are generated inside the lamella, thus these large and dense aggregates accelerate the foam coalescence and decrease the foam stability. This is because of extra gravity forces which increase the rate of the fluid drainage and expedite the rupturing of the bubbles. According to AlYousef et al. [53]

the addition of NPs at low to intermediate concentrations produced a more stable foam by generating small flocs that provide barriers between the gas bubbles and delaying the coalescence of bubbles. However, an excessive amount of NPs, reduced the maximum capillary pressure of coalescence and the value of zeta potential was decreased, as a result of the formation of extensive agglomerates.

Furthermore, as shown in Fig. 13, the half-life time of the nanoparticle-stabilized foam increases until it reaches to the nanoparticles concentration of 0.2 wt%, and then it starts to decrease due to the gravity effect that accelerates the nanoparticles aggregation at higher concentrations.

Additionally, due to the difference in the average size of nanoparticles in the solution (133 nm for SiO₂ NPs, and 420 nm for RHA NPs), at the optimum concentrations of nanoparticles, SiO₂ NPs generated higher stability foam with a longer half-life time than RHA NPs. This is because the nanoparticles with large sizes require more energy to be attached on the gas-liquid interface compared to the particles with smaller sizes, which means larger particles decrease the foam stability [54].

Although the RHA NPs have lower stability than SiO₂ NPs, it could be considered as the optimum nanoparticles for foam stability, as they are a bi-product of waste materials, therefore using them can provide a sustainable solution to manage waste materials, besides they are low-cost materials compared to other synthetic nanoparticles. Moreover, the RHA NPs have most of the properties of the SiO₂ NPs, naturally more silica content in the RHA (90% amorphous silica) [55], which makes the RHA the best alternative nanoparticles to be used effectively in EOR process. This will reduce the cost of producing synthetic SiO₂ NPs. However, to improve the efficiency of the RHA nanoparticles in the foam stability, further reduction in the range of RHA particles sizes is needed.

Fig. 13 shows that at lower concentrations of nanoparticles (0.05 wt % and 0.1 wt %), larger sizes of nanoparticles have slightly longer half-life time compare to smaller sizes. This might be due to the mechanism of nanoparticles accumulation at the foam lamellae, bigger particles at low concentrations will cover most of the lamellae surfaces and demonstrate a better stability than smaller particles at low concentrations. However, further studies are required to investigate the effect of different particles sizes of NPs at low and high concentrations on the foam stability.

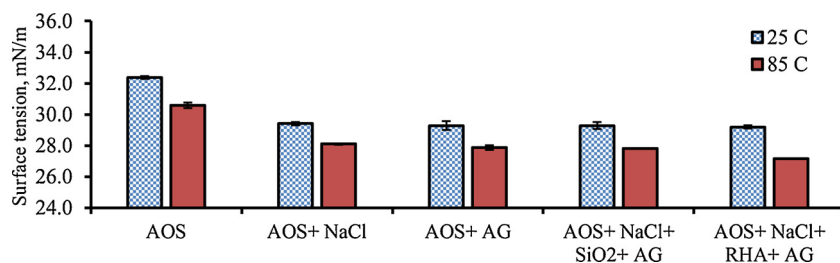


Fig. 6. Surface tension of the AOS solution with and without 2.0 wt% of NaCl, 0.2 wt% of SiO₂ NPs / RHA NPs, and 0.3 wt% AG.

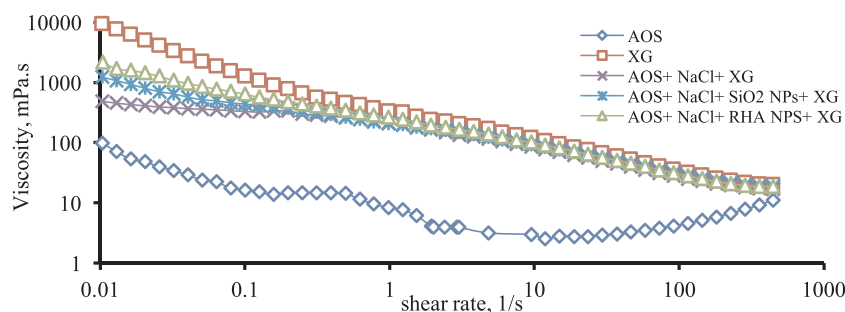


Fig. 7. Viscosity of xanthan gum solution with different additives as a function of shear rate at 85 °C.

3.5. Nanoparticle/polymer-stabilized CO₂-foam

The effect of polymer concentration on nanoparticle-stabilized CO₂-foam stability was examined by performing a series of experiments to study the synergetic effect of nanoparticles and polymers in enhancing the CO₂-foam properties at 85 °C and 2.0 wt% salinity.

The foam stability was tested for solutions with the optimum concentration of nanoparticles (SiO₂ and RHA) and different concentrations of polymers (XG and AG). Three concentrations of polymers (0.1, 0.3, and 0.5 wt%) were used while the concentrations of AOS surfactant, nanoparticles and NaCl were remained constant at 0.5 wt%, 0.2 wt%, and 2.0 wt%, respectively. Increasing the polymers concentration enhancing the foam stability for both types of nanoparticles as shown in Fig. 14. The foam that was enhanced by XG with SiO₂ NPs, demonstrated a high stability with the XG concentration of 0.3 wt% (3900 s) which was higher than the measured values for the XG concentrations of 0.1 wt% and 0.5 wt%, 2400 and 3300 s, respectively. For the foam that was enhanced by XG and RHA NPs, also the XG concentration of 0.3 wt% showed the highest stability with a decay time of 3000 s that was higher than the measured values for the XG concentrations of 0.1 and 0.5 wt%, 1200 and 2700 s, respectively. Adding XG to the surfactant solution increases its viscosity and lamellae thickness, which in turn decreases the rate of the liquid drainage from the foam structure. Therefore, using an optimum XG concentration, 0.3 wt%, improves the foam stability and reduces the rate of foam coalescence. For the XG concentration of 0.1 wt%, the viscosity of surfactant solution is too low to slow down the rate of the liquid drainage from the lamellae. On the other hand, at the XG concentration of 0.5 wt%, liquid solution becomes too dense in which a large gravity force is exerted on the liquid film that accelerates the rate of the liquid drainage i.e. the gravity force becomes dominant over the viscous force, and hence it expedites the rate of foam coalescence.

In the presence of nanoparticles and polymer, a larger area of the gas-liquid interface is covered by these additives in addition to the surfactant molecules, therefore, it has stronger molecular interaction/adhesion forces, which help to increase the foam life and thus the foam stability. The foam stability will be affected by bubbles disappearance (foam coarsening) as shown in Fig. 15, due to the pressure difference between bubbles [56]. The small bubbles start to disappear, as the gas

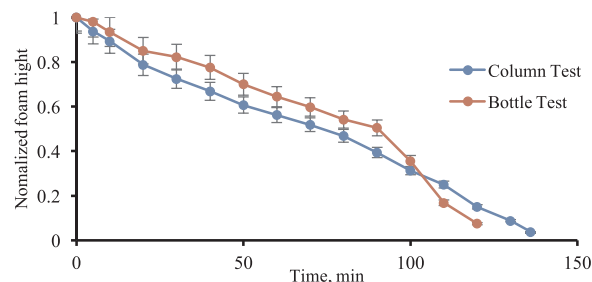


Fig. 9. Normalized foam height from column test and bottle test with AOS concentration of 0.5 wt%.

starts to diffuse into the bigger bubbles through the liquid film, and bigger bubbles will be grown, in which they eventually coalesce and rupture the foam structure. Von Neumann's study on 2D foam bubbles [57] demonstrated that the time evaluation of a 2D bubble is a function of the number of the sides in each bubble. The bubbles with 6 sides and more start to expand, whereas for the bubbles with less than 6 sides, they rapidly shrink and completely vanish as shown in Fig. 15. That is because these bubbles start to diffuse to other bubbles through the thin film to grow the neighbouring bubbles.

Addition of AG to the surfactant solution with nanoparticles demonstrated relatively low foam stability compare to XG as shown in Fig. 16, this is due to the difference in the molecular weight of the polymers. AG has an average molecular weight of 1.0 million g/mol which is three times lower than XG's molecular weight (3.2 million g/mol). The higher molecular weight increases the viscosity of the solution, which increases the disjoining pressure between the gas and liquid phases in the lamellae, that controls the foam stability [58]. In the solution of XG polymer, viscosity is high enough to generate thick lamellae, which makes the disjoining pressure positive and hence increases the foam stability. The generated foam with nanoparticles and AG polymer has thinner lamellae compare to the foam generated with nanoparticles and XG, as shown in Fig. 17. Using XG polymer increased the lamella thickness from 59 μm (in the absence of nanoparticles and polymer) to 188 μm and 134 μm in the presence of SiO₂ and RHA NPs, respectively, and when AG was used lamella thickness reached to

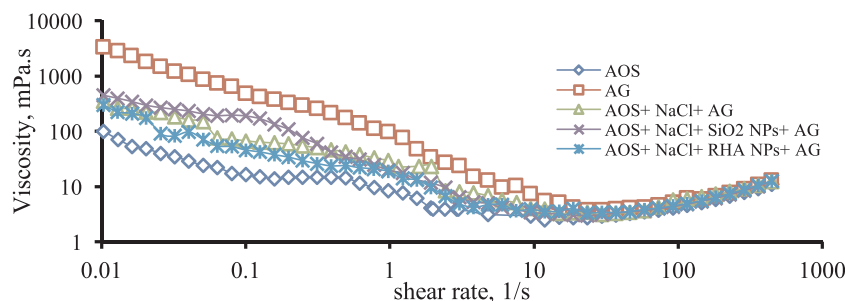


Fig. 8. Viscosity of acacia gum solution with different additives as a function of shear rate at 85 °C.

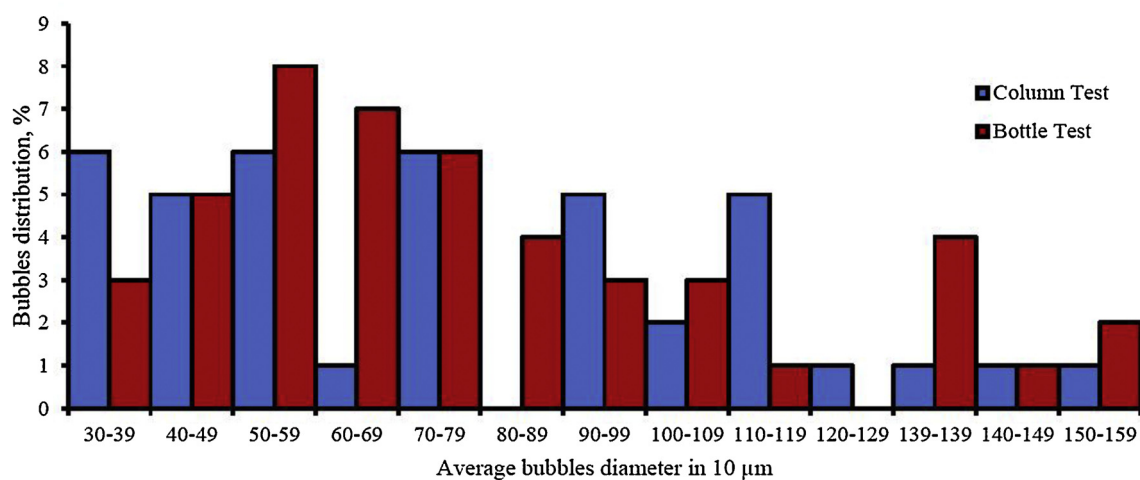
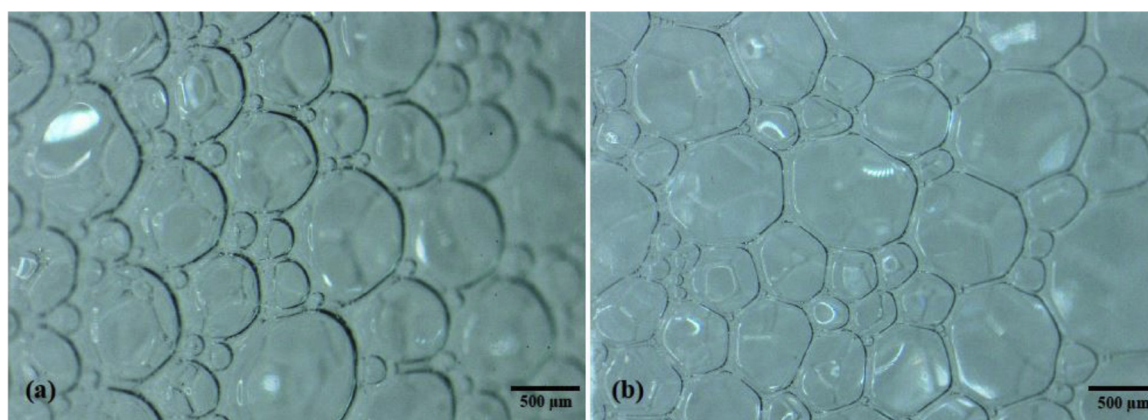


Fig. 10. Bubble-size distribution after 5 min for 0.5 wt% of AOS from (a) column test, (b) bottle test.

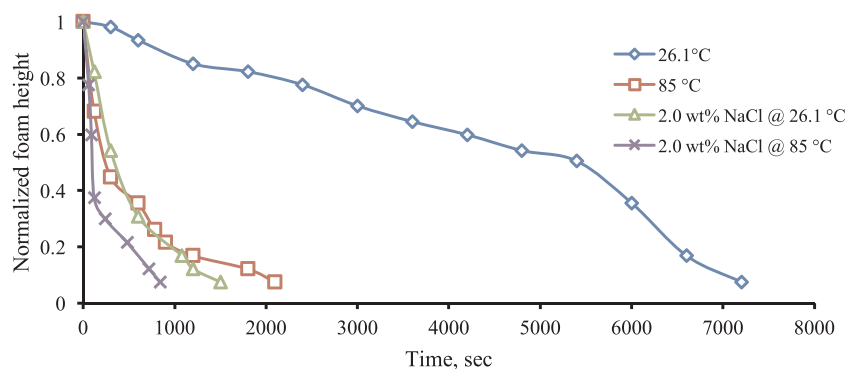


Fig. 11. AOS bulk foam stability with and without NaCl at different temperature conditions.

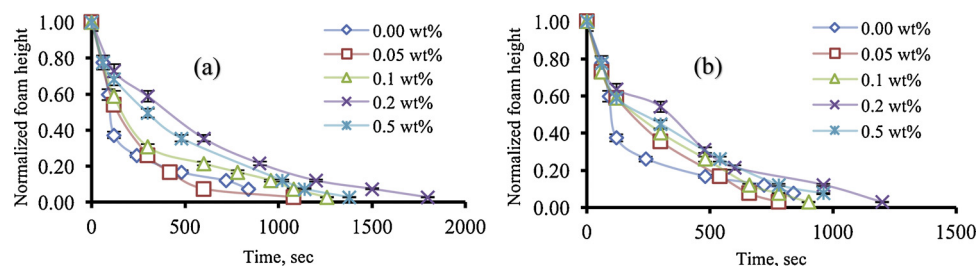


Fig. 12. Foam stability with 0.5 wt% AOS and 2.0 wt% NaCl at 85 °C and different concentrations of silica nanoparticles (a) SiO₂ (Bashir et al. [46]) NPs, (b) RHA NPs.

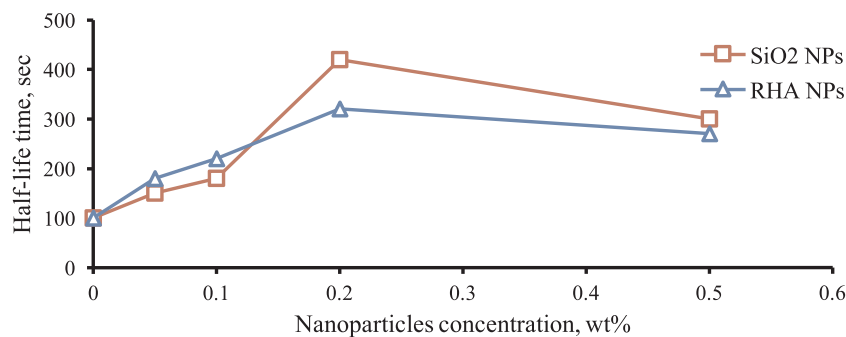


Fig. 13. Foam half-life time for different types and concentrations of nanoparticles with 0.5 wt% AOS and 2.0 wt% NaCl at 85 °C.

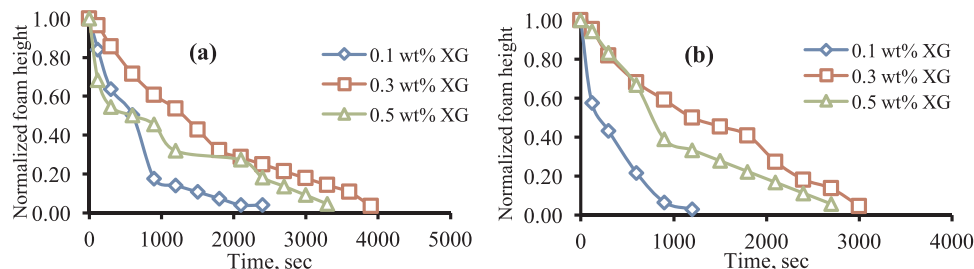


Fig. 14. Foam stability at 0.5 wt% of AOS, 2.0 wt% NaCl in the presence of different concentrations of xanthan gum at 85 °C and different nanoparticles, (a) SiO₂ (Bashir et al. [46]), (b) RHA.

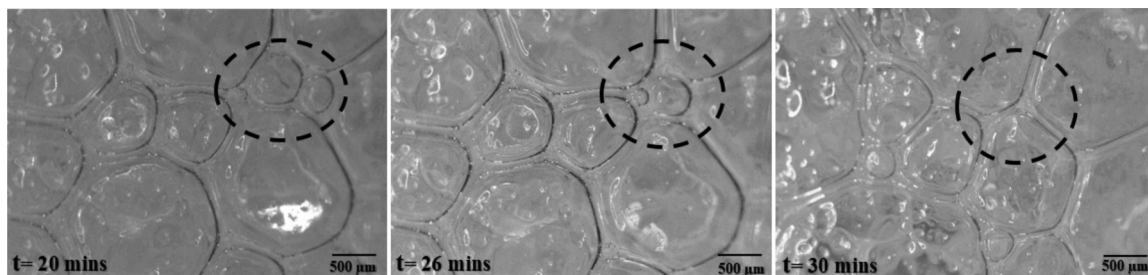


Fig. 15. A 2D images of foam bubbles coarsening by time. The foam was generated by 0.5 wt% of AOS, 2.0 wt% NaCl, 0.2 wt% SiO₂ NPs and 0.3 wt% XG at 85 °C, and the black circles indicate the merging of small bubbles into larger ones, i.e., coarsening bubbles.

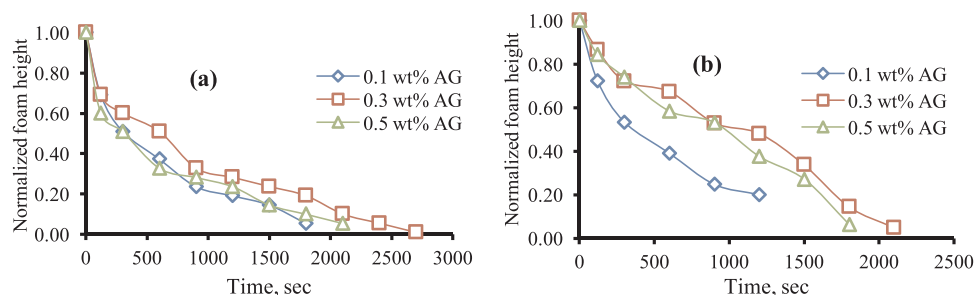


Fig. 16. Foam stability at 0.5 wt% of AOS, 2.0 wt% NaCl in the presence of different concentrations of acacia gum at 85 °C and different nanoparticles, (a) SiO₂, (b) RHA.

117 μm and 100 μm in the presence of SiO₂ and RHA NPs, respectively. The difference between the lamella thickness enhancements for two polymers is due to the viscosity of solutions of XG and AG polymers. The viscosity of the AG solution was not high enough to sustain thick lamellae for a long period compared to the viscosity of the XG solution, therefore, the solution with AG showed a lower foam stability.

3.6. Nanoparticles/xanthan gum polymer-stabilized foam in the presence of oil

The application of foam for enhanced oil recovery processes is strongly affected by the foam stability in the presence of reservoir hydrocarbons (oil phase). Reservoir hydrocarbons can flow in the liquid films of the injected foam in the form of emulsions or continuous phase [59,60]. Therefore, the interaction of foam with reservoirs hydrocarbons should be determined before introducing foams as an enhanced oil recovery process. To study these effects, three oil fractions (5, 10 &

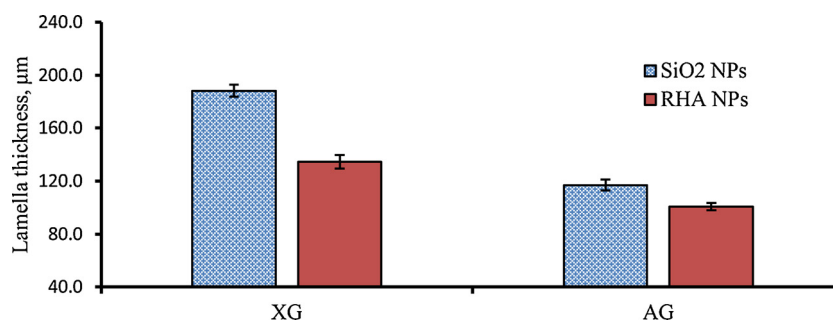


Fig. 17. Average foam lamella thickness for the optimum concentrations of nanoparticles and polymers at 85 °C (after 5 min of foam generation).

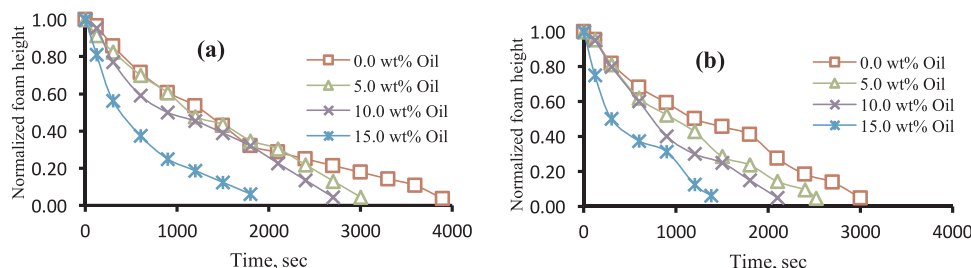


Fig. 18. Foam stability at 0.5 wt% AOS, 2.0 wt% NaCl, and 0.3 wt% XG in the presence of different concentrations of oil at 85 °C and different nanoparticles, (a) SiO₂ NPs (Bashir et al [46]), (b) RHA NPs.

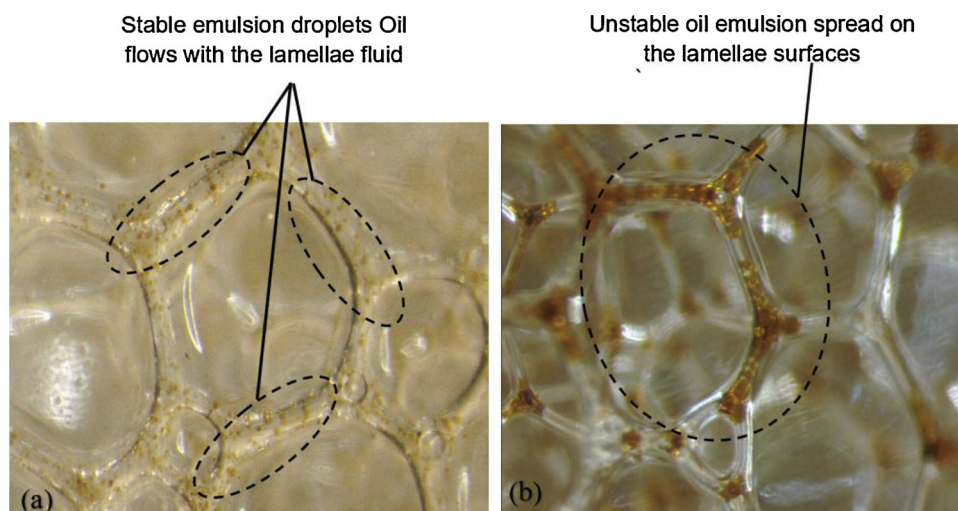


Fig. 19. Entering and spreading of oil at the foam lamellae in the, (a) presence of SiO₂ NPs and XG polymer, (b) absence of SiO₂ NPs and XG polymer. A video in .avi format is provided comparing the drainage of foam lamellae in the presence of oil phase.

15 vol%, volume fraction relative to the total solution volume) were used with the optimum concentrations of nanoparticles (0.2 wt%) and XG polymer (0.3 wt%). The required volume of oil was added to the test chamber after it was filled with the prepared AOS solution and additives at 85 °C. Fig. 18 shows the foam stability in the presence of oil for SiO₂/RHA NPs with XG polymer. The foam stability was slightly affected by addition of oil up to 10.0 vol% in both nanoparticle-based foams, and it was sharply decreased when 15.0 vol% oil was added. Furthermore, both nanoparticles with XG polymer improved the CO₂-foam stability, with a better performance in the presence of SiO₂ NPs.

The NPs accumulated at the gas-liquid interface reduced the interfacial area between the liquid and gas (increased the lamella thickness), and polymers improved the viscosity of the liquid in the lamellae which led to a decrease in the drainage rate from them. Therefore, when the oil was added to the AOS solution, it could not drainage the viscous liquid from the lamellae. Consequently, the oil was mixed with the viscous liquid, and flowed within the viscous lamellae as stable

emulsified droplets, without entering or penetrating the gas-liquid interface as shown in Fig. 19a. However, in the absence of additives, the oil was mixed with the AOS solution and created large oil emulsions inside the foam lamellae, which could merge into each other and become larger droplets (no viscous liquid in lamella to prevent the coalescence of the oil droplets leading to large droplets). Therefore, due to the imbalance of gravity (density difference between two phases) and capillary forces, the rate of liquid drainage was faster than the oil phase. Furthermore, the coalesced oil droplets started to spread over the lamella surface leading to the drainage of the surfactant from the lamella, thus the stable gas-liquid interface was replaced by the unstable oil-gas interface as shown in Fig. 19b. Hence through this process oil can rapidly reduce the lamella thickness and rupture the foam lamella. The process of foam lamella stability in the presence of oil is animated in Fig. 20. Also, this can be visualized in the provided video file that shows the oil phase can flow as stable emulsions or they can rupture the foam structure as coalesced droplets, depending on the type of the solution.

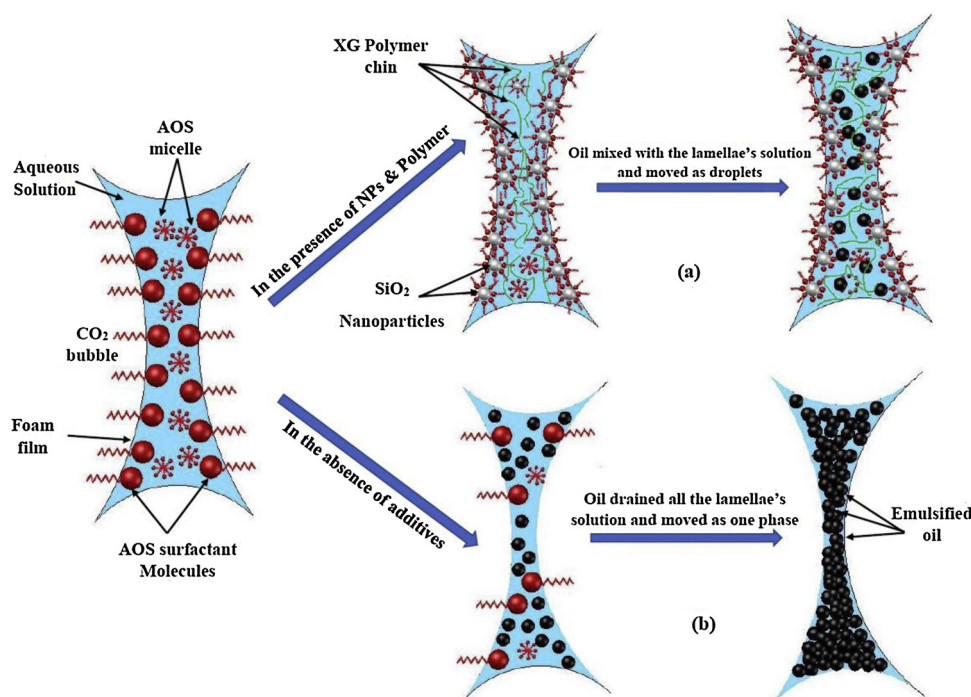


Fig. 20. A schematic diagram of the foam lamella stability in the presence of oil and (a) in the presence of SiO₂ Nanoparticles and XG polymer, (b) in the absence of additives.

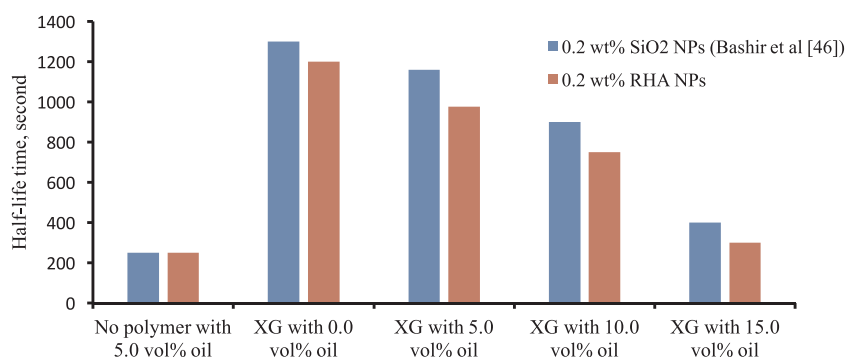


Fig. 21. Foam half-life time of 0.5 wt% AOS, 2.0 wt% NaCl, 0.3 wt% Xanthan gum, for different oil fractions at 85 °C.

From Fig. 21, the foam half-life time in the presence of SiO₂ NPs was slightly decreased from 1300 s (half-life time of the foam with XG) to 1160 s (11.0% decrease) and 900 s (30.0% decrease) in the presence of 5.0 vol% and 10.0 vol% oil, respectively. In the presence of RHA NPs, the half-life time decreased from 1200 s (half-life time of foam with XG) to 975 and 750 s in the presence of 5.0 vol% and 10.0 vol% oil, respectively. Furthermore, for 15.0 vol% of oil, the half-life time and stability were sharply decreased to 400 s (69.0% decrease) for SiO₂ NPs based foam, and 300 s (75.0% decrease) for RHA NPs based foam. As the oil fraction increases, the oil spreads and enters the lamellae that causes an instability in the bubble structures, faster liquid drainage from the lamellae, and decreases the foam life.

Similarly, using proper additives in the foam structure can improve the foam texture in the presence of oil as shown in Fig. 22. The average bubbles diameter was reduced from 2200 μm (in the absence of additives) to 320 μm (in the presence of additives). In the presence of SiO₂ NPs and XG polymer (Fig. 22a), the rate of liquid drainage was decreased, and the oil was able to flow in the form of emulsified droplets through the lamellae, without interrupting the gas-liquid interface. Therefore, small bubbles with spherical shapes and considerable lamellae thickness were created, resulting in an increase in the foam stability. However, in the absence of the nanoparticles and polymer

(Fig. 22b), the oil could rapidly drain all the liquid surfactant from the lamellae. This led to creation of big bubbles with polyhedral shapes, thus bubbles rapidly ruptured and coalesced.

4. Conclusions

In this study, the improvement of the CO₂-foam stability at high temperature and salinity conditions, in the presence of oil phase, was investigated. The focus of this investigation was on the ability of nanoparticles (SiO₂ and RHA NPs) and polymers (XG and AG) to increase the foam life by using a standard bottle test. The results provide us with a better understating of their potential applications in improving rheological properties of surfactant solutions and foams.

Addition of SiO₂ or RHA NPs can increase the CO₂-foam stability and half-life time at 85 °C and 2.0 wt% NaCl, with an optimum concentration of 0.2 wt% for both SiO₂ and RHA NPs.

Furthermore, using xanthan gum and acacia gum with nanoparticles improved the foam stability to a higher level, and increased the lamella thickness by enhancing the liquid viscosity.

Even though, RHA NPs showed inferior performance compared to SiO₂ NPs, they can be used as an additive to improve the foam stability in high temperature and salinity conditions, as they are economically

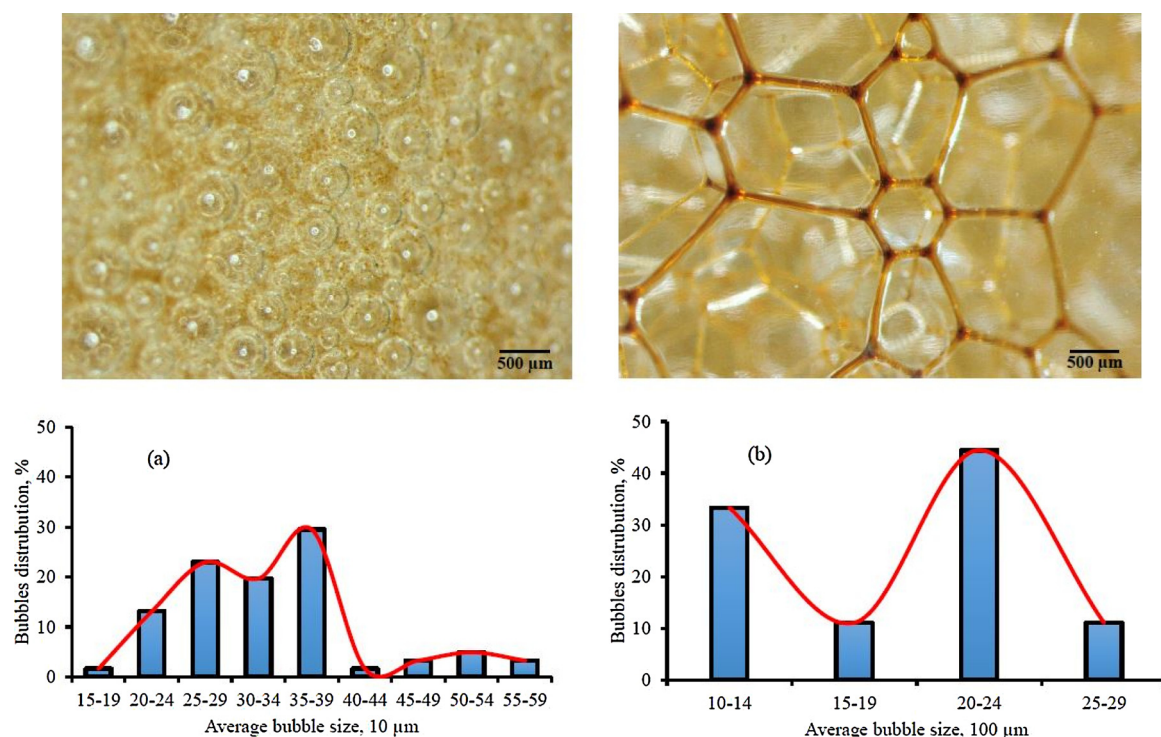


Fig. 22. Effect of oil on the bubbles size distribution after 5 min in the (a) presence of SiO_2 NPS and XG polymer, (b) absence of nanoparticles and polymer.

cost-effective materials. However, further studies are recommended to decrease the particles sizes to a lower value to improve their performance in foam stability applications. One of the main challenges of foams is their stability in the presence of oil phase. In our study we demonstrated that CO_2 -foam can show a better stability in the presence of oil when the optimum concentrations of SiO_2 /RHA NPs (0.2 wt %) and xanthan gum (0.3 wt %) were added. The stability of the CO_2 -foam with these additives was decreased insignificantly, when it was exposed to 5 vol% and 10 vol% of the oil, and the foam was sharply coalesced when the oil volume increased to 15 vol%. This sharp coalescence happened at the oil concentration of 5 vol% when the surfactant was used with no additives. This shows that using optimum concentrations of additives can enhance the stability of the foam structure at high temperature conditions.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary material related to this article can be found, in the online version, at doi:<https://doi.org/10.1016/j.colsurfa.2019.123875>.

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