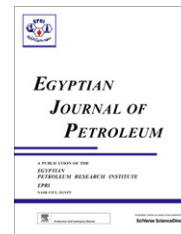




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FULL LENGTH ARTICLE

Application of sustainable foaming agents to control the mobility of carbon dioxide in enhanced oil recovery

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Abstract Carbon dioxide (CO₂) flooding is a conventional process in which the CO₂ is injected into the oil reservoir to increase the quantity of extracting oil. This process also controls the amount of released CO₂ as a greenhouse gas in the atmosphere which is known as CO₂ sequestration process. However, the mobility of the CO₂ inside the hydrocarbon reservoir is higher than the crude oil and always viscous fingering and gravity override problems occur during a CO₂ injection. The most common method to overcome these problems is to trap the gas bubbles in the liquid phase in the form of aqueous foam prior to CO₂ injection. Although, the aqueous foams are not thermodynamically stable, special care should be considered to ensure bulk foam preparation and stability. Selection of a proper foaming agent from a large number of available surfactants is the main step in the bulk foam preparation. To meet this purpose, many chemical and crude oil based surfactants have been reported but most of them are not sustainable and have disposal problems. The objective of this experimental study is to employ Lignosulfonate and Alkyl Polyglucosides (APGs) as two sustainable foaming agents for the bulk foam stability investigations and foam flooding performance in porous media. In the initial part, the bulk foam stability results showed that APGs provided more stable foams in comparison with Lignosulfonate in all surfactant concentrations. In the second part, the results indicated that the bulk foam stability measurements provide a good indication of foam mobility in porous media. The foaming agent's concentration which provided the maximum foam stability also gave the highest value of mobility reduction in porous media.

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1. Introduction

Global warming is the rising average temperature of the Earth's atmosphere and oceans since the late 19th century and its projected continuation. Since the early 20th century, the Earth's average surface temperature has increased by about 0.8 °C (1.4 °F), with about two thirds of the increase occurring since 1980 [1,2]. Warming of the climate system is unequivocal, and scientists are more than 90% certain that

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most of it is caused by increasing concentrations of greenhouse gases produced by human activities such as deforestation and the burning of fossil fuels which makes carbon dioxide as a main product [3,4]. The greenhouse effect is the process by which absorption and emission of infrared radiation by gases in the atmosphere warm a planet's lower atmosphere and surface. It was proposed by Joseph Fourier in 1824 and was first investigated quantitatively by Svante Arrhenius in 1896 [5]. Naturally occurring amounts of greenhouse gases have a mean warming effect of about 33 °C (59 °F) [4]. The major greenhouse gases are water vapor, which causes about 36–70% of the greenhouse effect; carbon dioxide (CO₂), which causes 9–26%; methane (CH₄), which causes 4–9%; and ozone (O₃), which causes 3–7% [6]. Malaysia is one of the countries that produce a large amount of carbon dioxide as a green house gas in the World [7]. The amount of carbon dioxide production in Malaysia has a rough incremental trend which makes Malaysia the first place among the all neighboring countries as shown in the Fig. 1. It was 7.57 metric tons per capita that is much more than the World average about 4.5 metric tons per capita reported by the World Bank in 2012. Therefore, it is necessary to control the amount of CO₂ as a greenhouse gas in Malaysia and also all over the World.

CO₂ injection into hydrocarbon reservoir is reported by many scientists as one of the best methods for CO₂ emission control and also enhanced oil recovery. CO₂ for enhanced oil recovery has favorable characteristics such as dynamic miscibility achievement between CO₂ and oil under most reservoir conditions, intermediate component extraction and heavy oil viscosity reduction which is named CO₂ flooding process. However, CO₂ flooding processes frequently experience viscous fingering and gravity override problems because of the low CO₂ density and viscosity in comparison with the crude oil. As a result, sweep efficiency decreases and significant amounts of oil are left behind [8–11].

The need for mobility control during CO₂ flooding has led to the study of foam processes, which involves the injection of CO₂ together with an aqueous solution of a CO₂-foaming agent [8]. CO₂ has a very low viscosity in comparison to oil and water. However, when CO₂ is a dispersed phase, as in foam, its apparent viscosity is greatly increased; thus, its mobility is improved [12]. From the time when the use of foam in reservoirs was first proposed in a patent by Bond and

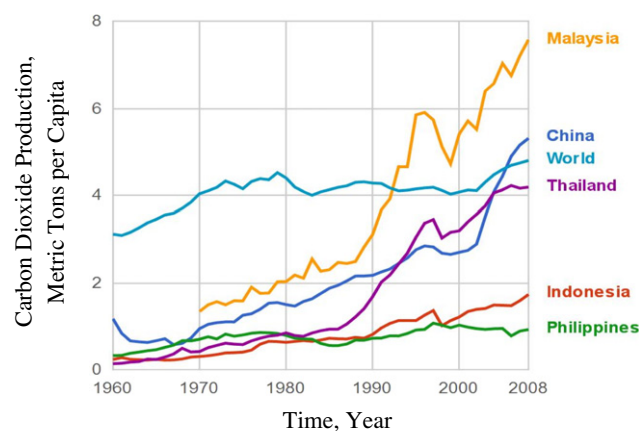


Figure 1 Carbon dioxide (CO₂) emissions per capita (World Bank 2012).

Holbrook (1958) which is reported by Bernard and Holm (1964) [13], it was usually implicitly assumed without specific mention, that foam would preferentially impede flow in the higher permeability layers or fractures in the reservoir that had already been swept of their oil. It was assumed without evidence that the unswept portions of the reservoir would remain at least as accessible and available to have their content displaced and forced into the production wells. Because of this assumption, many, if not most, of the reports of foam investigations included descriptions of core-floods with listings of oil recovered and ranked in the order of those values. This assumption for CO₂ foam floods cannot be examined for validity without more thoroughly considering the processes involved in bulk foam stability. Also, it is necessary to examine the behavior in core samples.

One of the main parameters determines the success of foam flooding is the selection and concentration of proper foaming agents (surfactants). In two-phase colloidal systems such as bulk foam, a thin, intermediate region or boundary, known as the interface, lies between the dispersed and continuous phases. Interfacial properties are very important in foams because the gas bubbles have a large surface area, and even a modest surface energy per unit area can become a considerable total surface energy. If sufficient energy cannot be provided through mechanical energy input, then another alternative is to use surfactant chemistry to lower the interfacial free energy, or interfacial tension. According to this fact, the study of surfactant type and concentration is necessary for strong bulk foam generation. However, the chosen foaming agent should be sustainable, nontoxic and have less environmental disadvantages especially for the offshore field applications. Many screening studies to choose proper foaming agent have been performed by many scientists [14–26] but most of them used various petroleum based surfactants which potentially cause many ecological problems. In the recent years some reports are provided about sustainable surfactants that indicated that they are already used in the petroleum industry. They are applied in environmentally safe drilling fluids [27], in well cleaning agents [28], as foamers in heavy oil recovery [29] and for reducing interfacial tension in surfactant-induced tertiary oil recovery processes [22,30]. However, no attention has been given to the effect of these surfactants as a foaming agent on foam generation and stability to control the mobility of CO₂ injection process. Therefore, the main aim of this paper is to evaluate two types of sustainable surfactants for CO₂ mobility control applications in enhanced oil recovery.

2. Experimental procedure

Two sets of experiments are designed in this paper to achieve the research objectives. In the first set, a series of bulk foam stability tests have been performed using two sustainable foaming agents (surfactants) to understand the effect of surfactant types and concentration in the presence and absence of crude oil on bulk foam stability. In the second part, the core flooding tests are carried out to evaluate the influence of these two surfactants on incremental oil recovery, differential pressure and mobility reduction of gas injection process. The lists of the experimental runs are illustrated in the Tables 1 and 2 for bulk foam stability and core flooding studies, respectively.

Table 1 Bulk foam stability experimental schedule.

Number of runs	Surfactant type	Crude oil concentrations, ppm	Surfactant concentrations, ppm
Run No.1	Alkyl Polyglucoside (APGs)	No oil	500
Run No.2	Alkyl Polyglucoside (APGs)	No oil	1000
Run No.3	Alkyl Polyglucoside (APGs)	No oil	5000
Run No.4	Alkyl Polyglucoside (APGs)	No oil	10,000
Run No.5	Alkyl Polyglucoside (APGs)	10,000	500
Run No.6	Alkyl Polyglucoside (APGs)	10,000	1000
Run No.7	Alkyl Polyglucoside (APGs)	10,000	5000
Run No.8	Alkyl Polyglucoside (APGs)	10,000	10,000
Run No.9	Lignosulfonate	No oil	500
Run No.10	Lignosulfonate	No oil	1000
Run No.11	Lignosulfonate	No oil	5000
Run No.12	Lignosulfonate	No oil	10,000
Run No.13	Lignosulfonate	10,000	500
Run No.14	Lignosulfonate	10,000	1000
Run No.15	Lignosulfonate	10,000	5000
Run No.16	Lignosulfonate	10,000	10,000

Table 2 Core flooding experimental schedule.

Number of runs	Name of the experiments	Gas flow rate, ml/min	Liquid flow rate, ml/min	Foam quality	Capillary number
Run No.1	Gas injection	9	N/A	N/A	10^{-6}
Run No.2	Lignosulfonate foam flooding	9	4	86.2%	10^{-5}
Run No.3	APGs foam flooding	9	4	64.7%	10^{-5}
Run No.4	Water flooding	N/A	4	N/A	10^{-4}
Run No.5	Lignosulfonate flooding	N/A	4	N/A	10^{-4}
Run No.6	APGs flooding	N/A	4	N/A	10^{-4}

2.1. Materials

Lignosulfonate or sulfonated lignin and Alkyl Polyglucosides (APGs) surfactants are applied in the form of 60% active as two sustainable foaming agents for the bulk foam stability investigations. Lignosulfonate is recognized as an anionic surfactant which is a byproduct from the production of wood pulp using sulfite pulping and Alkyl Polyglucosides are known as nonionic surfactant which is regenerated from natural raw

materials such as glucose derived from corn and fatty alcohols from coconut and palm kernel oils (Fig. 2). The detailed properties of both surfactants are provided in a Table 3. Aqueous solutions of surfactants at a concentration from 500 to 10,000 ppm in the range of above, below and equal to critical micelle concentration (CMC) were prepared to generate the foam in the presence and absence of swollen micelles. The synthetic type of mineral oil with the detailed properties in Table 4 is used to simulate the crude oil in both bulk foam stability and core flooding experiments. High purity CO₂ is used in the ambient condition (14.7 psi and 60 °F) for both series of experiments to simulate immiscible injection condition.

2.2. Bulk foam stability experiments

Foam stability test equipment is prepared and modified based on ASTM-D 6082-06, D892-06 and D1881-97 as indicated in Fig. 3. The equipment consisted of 560 cc graduated transparent low pressure cylinder, 6 cm diameter and 60 cm length, spherical gas diffuser stone made of fused crystalline alumina grain, displacement pump and high purity carbon dioxide cylinder. The cell is first filled with the aqueous solution to be tested and the system was brought to the desired pressure by means of a positive displacement pump and temperature equilibrated in a thermostatic water bath. The pressure difference between the CO₂ tank and the solution tank was determined and brought to zero. At this point CO₂ was allowed to flow from the capillary tube into the bottom of the surfactant solution. The CO₂ flowed upward through a gas diffuser at the lower end of the cell. Depending on the effectiveness of surfactants, the bubbles either formed a layer of foam-like dispersion

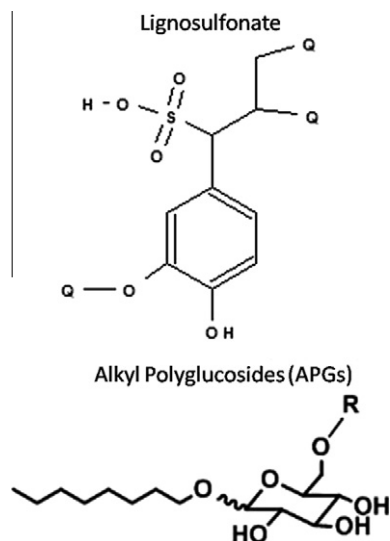
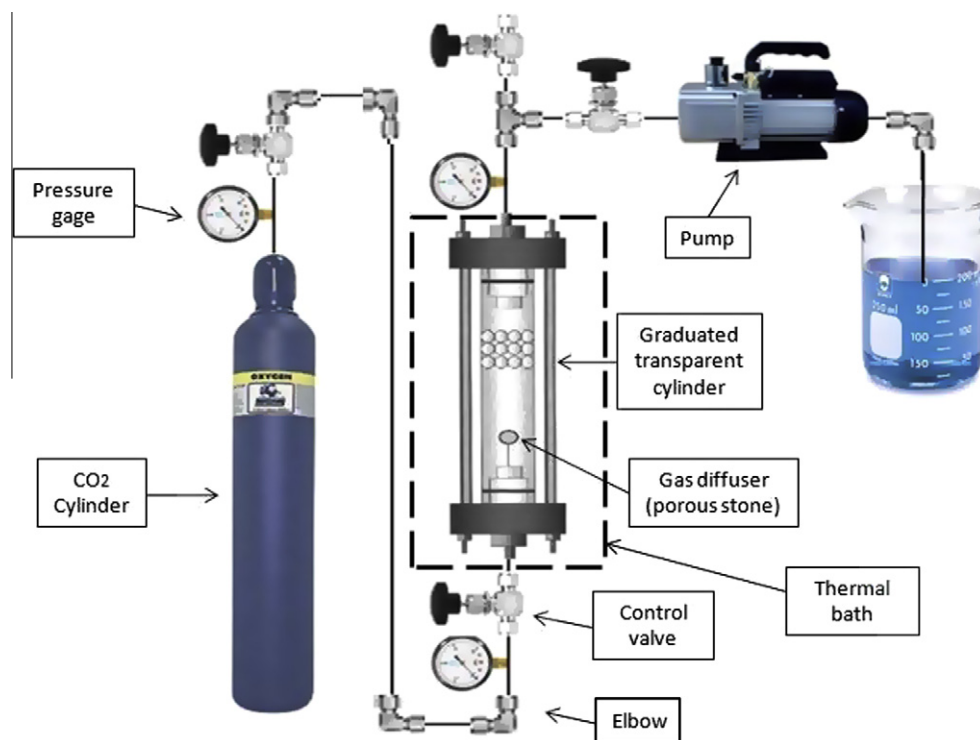
**Figure 2** Chemical structure of Lignosite[®]100 and C₈-APGs.

Table 3 Sustainable surfactants properties.

Trade name	Composition	Supplier	Hydrocarbon chain	Type	Appearance
Lignosite [®] 100	Lignosulfonate	Georgia-Pacific	C ₂₀ H ₂₆ O ₁₀	Anionic	Light brown
APGs	Alkyl Polyglucoside	Mistral Lab Chemicals	C ₁₆ H ₃₂ O ₆	Nonionic	Light yellow

Table 4 Mineral oil properties.

Trade Name	Supplier	Density g/mL at 20 °C	Kinematic Viscosity, cS	CAS No.	Appearance
Paraffin oil	QReC	0.85	30	8012-95-1	Colorless

**Figure 3** Bulk foam stability apparatus.

at the top of the transparent tube or coalesced into a clear layer of CO₂ gas. After a standard volume of CO₂ was introduced, the pump was stopped and the stability of foam determined by measuring the foam layer thickness versus time.

2.3. Core flooding studies

Unconsolidated sand pack with 32 cm length and 3 cm diameter was prepared to simulate the reservoir physical model as shown in Fig. 4. The packing material was the glass bead with the mesh size of 50–100 meshes. The porosity of the sand pack was 34% with the absolute permeability of 2180 md (Table 5). The sand pack is located in the horizontal position to consider that the effect of gravity force is negligible. The rate of the liquid injection in core flooding experiments is maintained 4 ml/min and for gas injection 9 ml/min to consider both gas and liquid injection are valid in the same range of capillary numbers. In the core flooding experiments, the reservoir model initially saturated with deionized water to simulate connate water saturation and to measure absolute permeability. The core

next is flooded with oil to create initial oil saturation. This flood leaves a residual saturation of water comparable to that found as connate water in reservoirs. The sand pack is then flooded with gas, water, surfactant and foam to measure the amount of incremental oil recovery and volumetric sweep efficiency improvement. At the end of the experiment the results of CO₂ injection are compared with water flooding, surfactant flooding and two sustainable CO₂ foams.

3. Experimental results

The results of laboratory experiments for both bulk foam stability and foam flooding in porous media are analyzed and reported in this section.

3.1. Bulk CO₂ foam stability tests

CO₂ foam stability tests were conducted with Alkyl Polyglucosides (APGs) at different surfactant concentrations solubilized

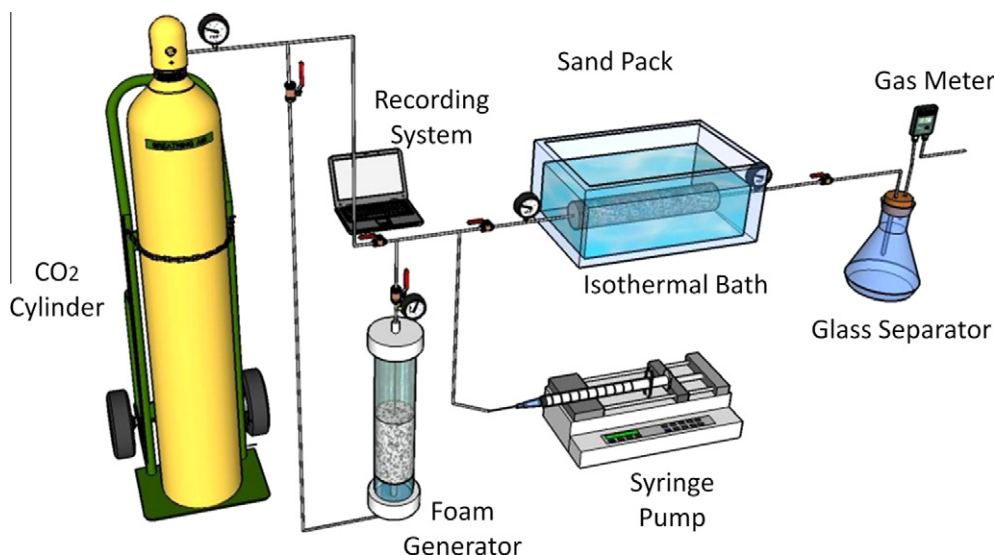


Figure 4 Core flooding apparatus.

Table 5 Sand pack properties.

Packing materials	Mesh size	Length, cm	Diameter, cm	Porosity, %	Absolute permeability, md	Pore volume, ml	OOIP, ml	Wettability
Glass bead	50–70	32	3	0.34	2180	76.9	69	Water wet

in the deionized water, 60 °F, and 14.7 psi. The surfactant concentrations are 500, 1000, 5000 and 10,000 ppm, respectively. In the foam stability experiments, coalescence of bubbles was observed at all concentration versus time. Fig. 5 indicated that the foam collapse of Alkyl Polyglucosides (APGs) is a strong function of surfactant concentration. The results indicated that APGs with concentrations of 1000, 5000 and 10,000 ppm which are equal and more than the CMC are an excellent foaming agent. However, below CMC with 500 ppm concentration, foam stability test indicated very unstable APG foams. The volume reduction occurs because the liquid drains through the lamellae due to the force of gravity after foam generation

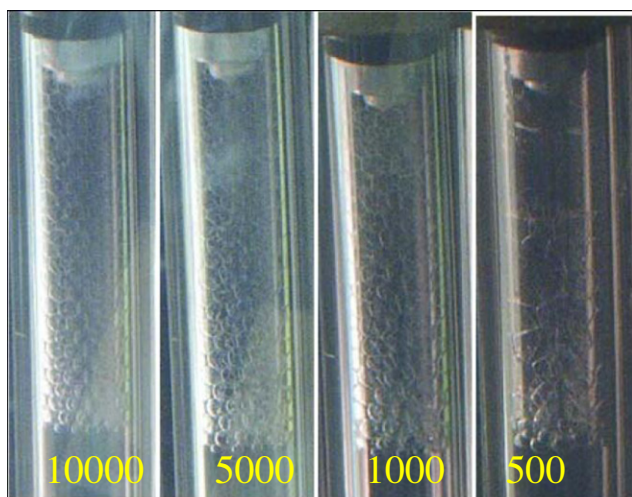


Figure 5 Foam stability of Alkyl Polyglucoside in various concentrations (ppm).

and also surface elasticity reduction due to lack of enough surfactant concentration in the foam lamellas and plateau borders. As the lamellar fluid drains, the amount of surfactant in the lamella and plateau border decreases which causes lower surface viscosity and elasticity in the foam structure. The phenomena change the foam appearance by deforming the shape of the bubbles from spherical to polyhedral. In the tests, it is also being noticed that the lamellae in the upper layer of the foam are thinner than those in the lower layer of the foam due to gravity drainage. Draining continued until capillary forces were going to be equal to gravity forces and the gas–liquid interface curvature increases at the plateau borders. The increased curvature generates a low-pressure region in the plateau border area which caused higher pressure resides at flat thin-film region (lamellas). This pressure difference forces the liquid to flow toward the plateau borders and cause thinning of the films and motion in the foam. Fig. 6 indicated the foam height of different concentrations of Alkyl Polyglucosides (APGs) versus time. The results indicated that using 500 ppm of Alkyl Polyglucosides (APGs) surfactant solution, the height of the foam generated after 25 s was 43.5 cm. The overall time for this foam collapse was 23 min. The foam stands for a much longer time to drain due to increase in surfactant concentration and governing the foam stability below the CMC by increasing interfacial rheological properties of the foam film. The results for 1000 ppm of Alkyl Polyglucosides (APGs) surfactant solution indicated that the initial height of the foam generated in 25 s was 48.0 cm which is higher than the 500 ppm concentration. This concentration of surfactant generates foam to stand as long as 75 min to drain. As shown in Fig. 6, the foam stands for a longer time to drain in comparison with 500 ppm. In the case of 5000 ppm of Alkyl Polyglucosides (APGs) surfactant solution, the height of the foam

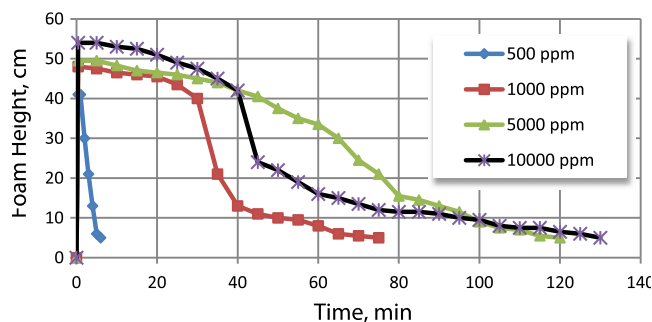


Figure 6 Foam stability of APGs at various surfactant concentrations.

generated after 25 s was recorded 49.5 cm. The overall time for this foam to drain was 120 min. The foam stands for a much longer time to drain due to the stability of the lamellae from this solution above CMC. The final concentration of Alkyl Polyglucosides (APGs) surfactant was 10,000 ppm much more above the critical micelle concentration. In this situation, the foam stability is governed by micelle concentration, structure and layering. The height of the foam generated after 25 s was 54 cm. As shown in Fig. 6, the time recorded for the foam with 10,000 ppm concentration to coalescence was 130 min.

In the second part of the foam stability tests, the foamability and stability of lignosulfonate were examined in the same concentration as Alkyl Polyglucosides (APGs). Fig. 7 represents the result of static decay of CO₂ foam using lignosulfonate solution as a foaming agent with deionized water. The amount of foam in this graph indicates the persistence of foam remaining inside the transparent cell after a standard volume of CO₂ has been injected. The heights of foam generated from these solutions are very small, less than 20 cm. This indicates that this solution is a weak foaming agent. The bubbles formed at 0.5 wt.% concentration of lignosulfonate coalesced in less than a minute. At higher concentrations, the percentage of foam increased and the bubbles lasted more than five minutes, but less than ten. The picture of the foam prepared using minimum to maximum amount of lignosulfonate concentration is provided in Fig. 8.

3.2. Core flooding studies

After the bulk foam stability tests, all concentrations of both surfactants are tested as a foaming agent in the core flooding

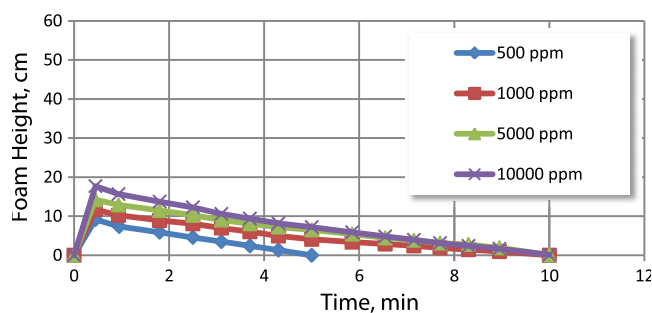


Figure 7 Foam stability of lignosulfonate at various surfactant concentrations.

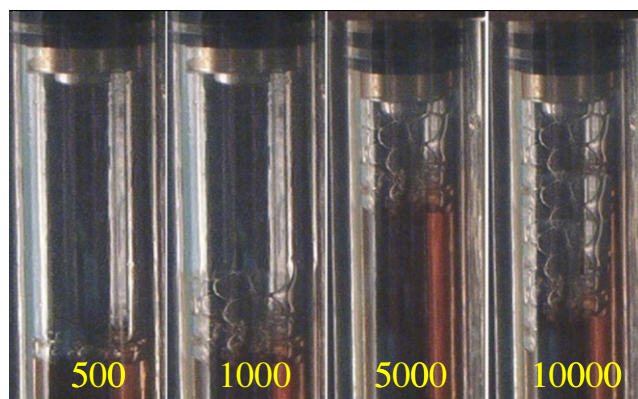


Figure 8 Foam stability of lignosulfonate at various concentrations (ppm).

experiments and the results are compared to the CO₂ injection. The data for extra oil recovery and pressure drop are provided in this section. In the first part of the experiments CO₂ alone was used as a displacing agent in the sand pack, breakthrough occurred after 0.26 PV of fluid was injected. Co-injection of CO₂ with Alkyl Polyglucosides (APGs) solution at 500 ppm simulated short cycles of foam flooding process. Delayed CO₂ breakthrough was observed at 0.42 PV for this run. When Alkyl Polyglucosides (APGs) at 1000 ppm were coinjected with CO₂; the foam appeared to produce more oil and the breakthrough occurred after 0.48 PV of fluid was injected. When Alkyl Polyglucosides (APGs) at 5000 ppm were coinjected with CO₂, carbon dioxide breakthrough occurred at 0.65 PV and the foam flooding performance was improved. In the 1000 ppm of Alkyl Polyglucosides (APGs) as a foaming agent, the gas breakthrough occurred at 0.7 PV which confirms more improvements in the gas oil ratio by surfactant concentration. Fig. 9 indicated the results of plotting the cumulative gas oil ratio (GOR) as a function of total pore volume injected. The highest cumulative GOR occurred when CO₂ only was used as the displacing agent. Coinjected CO₂ with foam reduced the cumulative GOR when surfactant at 500, 1000, 5000 and 10,000 ppm in form of CO₂ foam was injected to the sand pack, the amount of GOR substantially reduced.

The oil recovery from the sand pack is also recorded for various injected fluids as a function of total pore volumes of displacing fluid injected. Each test is repeated for three times to check the repeatability of the experiments. The gas injection is used as a base case of the experiments to determine the amount of oil recovery as a function of pore volume injected. The oil recovery versus pore volume injected for the base case experiment is indicated in Fig. 10. The results indicated that the gas injection can only recover 27.3% of original oil in place. After the gas injection process the foam is introduced into the system to decrease the mobility of gas injection in the sand pack. The results indicated that during each test, 8 PV of fluid was injected, about 56% of the oil was produced by a maximum concentration of Alkyl Polyglucosides (APGs). The results of tests during injection of carbon dioxide as a gas were compared with lignosulfonate and Alkyl Polyglucosides (APGs). The oil production was observed higher for the system of Alkyl Polyglucosides (APGs). Fig. 11 also compared the results of oil production of Alkyl Polyglucosides (APGs) and lignosulfonate both at 10,000 ppm (highest bulk foam

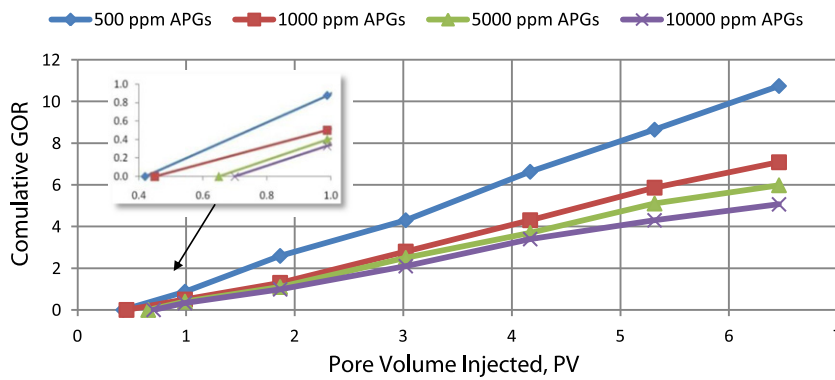


Figure 9 Gas oil ratio (GOR) as a function of pore volume injected.

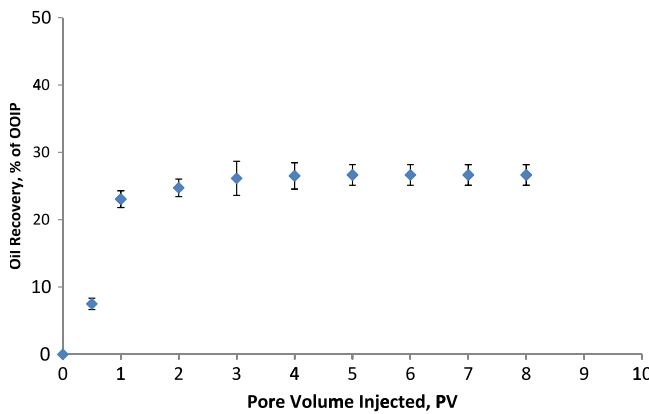


Figure 10 Oil recovery in the single gas injection process.

stability) that indicated the oil production of Alkyl Polyglucosides (APGs) that produced about 56 percent of original oil in place in comparison with 33.3% oil production of lignosulfonate as a foaming agent. The sweep efficiency of lignosulfonate was not very effective, because the foam was not as strong as Alkyl Polyglucosides (APGs). Therefore, the lignosulfonate is not recognized as a strong foaming agent in comparison with Alkyl Polyglucosides (APGs) and oil based surfactants. Pressure drop profiles for Alkyl Polyglucosides (APGs) and lignosulfonate are provided in Fig. 12. The pressure drop of Alkyl Polyglucosides (APGs) at 10,000 ppm was much higher than

lignosulfonate at 10,000 ppm which is because of more stable foams provided by Alkyl Polyglucosides (APGs). At the end of the foam experiments water and surfactant flooding was introduced into the system to compare their results with the foam flooding process. From the results it can be understood that the quantity of the oil recovery by APGs foams is between gas and water flooding processes.

4. Feasibility study

Simplified economics can be estimated with the data gathered for the prices of the conventional oil based surfactants which are compared with the sustainable surfactants proposed in this paper. This is a key point because however technically proven it may be, the process will never be trusted by operators unless incremental barrels of oil can be produced economically. The economical evaluation is performed based on 105 USD/bbl of the crude oil which is the average of OPEC oil price in the years 2011 and 2012. Also, the price of the surfactant systems is evaluated at critical micelle concentration based on the industrial scale preparation. As shown in Fig. 13, the overall cost of oil production using conventional (petroleum based) type of surfactant is between 20 and 30 USD/bbl which is much higher than the sustainable surfactants about 10–15 USD/bbl. The reason of the price difference is that most of the conventional surfactants are petroleum price dependent and their price increases when the crude oil price rises in the World. However, they can be produced easily in the largest

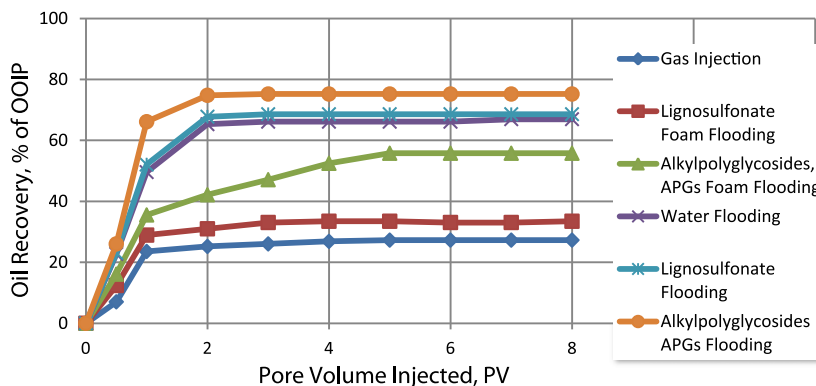


Figure 11 Oil recovery of various injection systems (10,000 ppm of both surfactants).

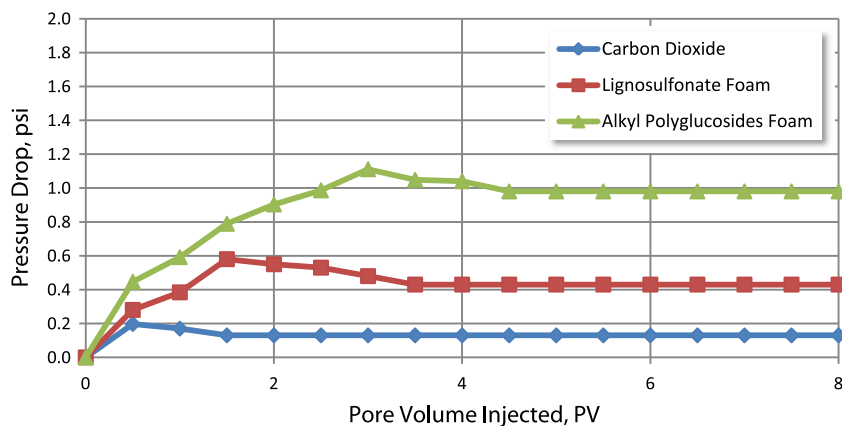


Figure 12 Pressure drop as a function of PV injected (10,000 ppm of both surfactants).

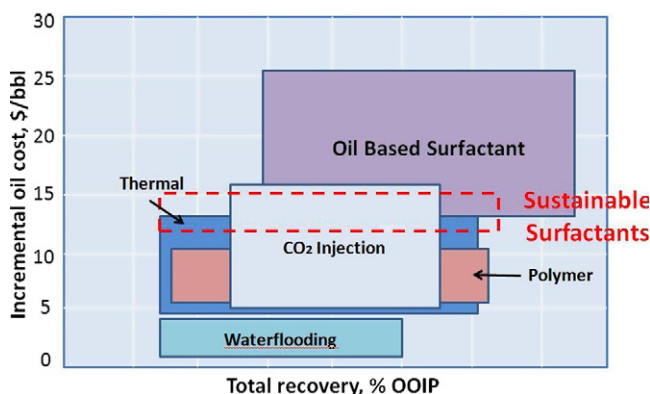


Figure 13 Economical evaluation of using sustainable surfactants.

scale in comparison with the sustainable surfactants. In the case of surfactant application as a foaming agent, the surfactant amount required is only 20–30% of the surfactant which is normally used for the single surfactant flooding. Based on the economical evaluation, it can be concluded that the foam flooding using sustainable surfactants is one of the most economical methods among all other surfactant applications in enhanced oil recovery (see Fig. 13).

5. Conclusions

1. Bulk Foam stability of both lignosulfonate and Alkyl Polyglucosides (APGs) is enhanced by increasing the surfactant concentration.
2. Alkyl Polyglucosides (APGs) are found as good foaming agent that remained about 130 min at maximum (10,000 ppm) surfactant concentration.
3. Alkyl Polyglucosides (APGs) are recognized as a stronger foaming agent that prepared 54 cm initial foam height in comparison with lignosulfonate which only made about 20 cm.
4. Gas Oil ratio (GOR) is decreased during a core flooding studies of Alkyl Polyglucosides (APGs) as the surfactant concentration increased.
5. The quantity of oil recovery increased using both foaming system in comparison with carbon dioxide solely.

6. The results of Alkyl Polyglucosides (APGs) recovery at 10,000 ppm concentration (highest bulk foam stability) indicated 56% of OOIP which is higher in comparison with only 33.3% oil recovery of lignosulfonate as a foaming agent.

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