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The Investigation of Silica Nanoparticles-CO_2 Foam Stability for Enhancing Oil Recovery Purpose

David Maurich[1*]
[1]Department of Petroleum Engineering, Faculty of Engineering and Design, Institut Teknologi Sains Bandung, Kota Deltamas Lot-A1 CBD, Jl. Ganesha Boulevard, Pasirranji, Kec. Cikarang Pusat, Bekasi, Jawa Barat 17530
*Corresponding Author: davidmaurich@gmail.com

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Abstract

Carbon dioxide (CO_2) gas injection is one of the most successful Enhanced Oil Recovery (EOR) methods. But the main problem that occurs in immiscible CO_2 injection is the poor volumetric sweep efficiency which causes large quantities of the oil to be retained in pore spaces of reservoir. Although this problem can be improved through the injection of surfactant with CO_2 gas where the surfactant will stabilize CO_2 foam, this method still has some weaknesses due to foam size issue, surfactants compatibility problems with rocks and reservoir fluids and are less effective at high brine salinity and reservoir temperature such as typical oil reservoirs in Indonesia. This research aims to examine the stability of the foams/emulsions, compatibility and phase behavior of suspensions generated by hydrophobic silica nanoparticles on various salinity of formation water as well as to determine its effect on the mobility ratio parameter, which correlate indirectly with macroscopic sweep efficiency and oil recovery factor. This research utilizes density, static foam, and viscosity test which was carried out on various concentrations of silica nanoparticles, brine salinity and phase volume ratio to obtain a stable foam/emulsion design. The results showed that silica nanoparticles can increase the viscosity of displacing fluid by generating emulsions or foams so that it can reduce the mobility ratio toward favorable mobility, while the level of stability of the emulsion or foam of the silica nanoparticles suspension is strongly influenced by concentration, salinity and phase volume ratio. The high resistance factor of the emulsions/foams generated by silica nanoparticles will promote better potential of these particles in producing more oil.

INTRODUCTION

Petroleum is still one of the main energy sources in terms of its wide availability and integration with current technology. Demand for energy, especially oil which continues to increase is not followed by availability (energy supply) which continues to decline each year. The method for increasing the oil recovery factor after the primary and secondary recovery stages is called Enhanced Oil Recovery (EOR) methods. One of the many EOR methods applied in the oil field to increase the oil recovery factor is the injection of carbon dioxide (CO_2) gas. Injection of CO_2 gas (CO_2 flooding) into the reservoir causes viscosity of oil and reservoir fluid interface tension decreases so that oil easily flows (Abdurrahman, Permadi, Hidayat, & Pangaribuan, 2018; Samba, Aldokali, & Elsharaf, 2019). Another advantage is that CO_2 has the ability to invade reservoir zones that were not previously invaded by water, so that trapped oil can be displaced. However, due to the viscosity of CO_2 is lower than the oil viscosity at the reservoir conditions, the viscosity instability will occur so that it will reduce the sweep efficiency of the EOR process and will significantly reduce the oil recovery factor (Abdurrahman, Ferizal, Saputra, & Sari, 2019; Samba & Elsharafi, 2018). In addition, reservoir rock conditions that are very heterogeneous in terms of permeability distribution can cause channeling or viscous fingering as a result of very low CO_2 viscosity. It can also cause gravity segregation as a result of low CO_2 density which will further reduce volumetric

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sweep efficiency (Li, Wang, & Li, 2020). In order to overcome these issues, various studies to improve the stability and viscosity of CO_2 have been conducted in laboratories, especially the use of surfactant-stabilized CO_2 foam. In the study of injection of surfactant to form CO_2-in water foam, it is generally known that the surfactant could reduce the mobility of CO_2 and increased sweep efficiency by forming CO_2-in water foam that has a greater viscosity than brine. However, the viscosity of CO_2-in water foam is less stable in reservoir conditions because some surfactants are adsorbed to the rock surfaces and are degraded due to high reservoir temperature and brine salinity (Maurich, 2019). To overcome these problems, a substance that can maintain or increase foam stability effectively during the process of injection of CO_2 in an oil reservoir is highly required (Jikich, 2012).

Several nanotechnology studies in the field of petroleum exploitation processes have produced the concept of using silica nanoparticles (Ahmadi & Shadizadeh, 2013; Bera & Babadagli, 2017; Cheraghian, Hemmati, Masihi, & Bazgir, 2013; Emadi, Shadizadeh, Manshad, Rahimi, & Mohammadi, 2017; Hendraningrat, Li, & Torsater, 2013; Kang, She, Zhang, You, & Song, 2016; Novrianti, 2016; N Rita, Mursyidah, Erfando, Herfansyah, & Ramadhan, 2019; Novia Rita, Erfando, & Munandar, 2019; Skauge, Spildo, & Skauge, 2010). Basically, silica nanoparticles have easily adsorbed properties at the brine-oil interface or brine-CO_2 interface so that they can generate foams or emulsions that can increase the viscosity and stability of CO_2 during the injection process in the reservoir (Jikich, 2012). Meanwhile other researchers such as Espinoza, Caldelas, Johnston, Bryant, & Huh (2010) concluded that under conditions of high salinity, it was required high concentrations of silica nanoparticles to maintain the stable foams at a temperature of 95 ^oC. High salinity and temperatures will usually prevent the formation of foam, while increasing pressure results in more foam (Yu, Mo, Liu, & Lee, 2013). In a subsequent study, Yu, Wang, Liu, & Lee (2014) concluded that the structure of silica nanoparticles had no effect on the shape and size of the CO_2 bubble, but in the process of displacement of oil in the core samples, it was concluded that hydrophilic silica nanoparticles could displace residual oil left in rock pore spaces after water injection (waterflooding) thus increasing the total oil recovery factor at room temperature conditions. To find out the exact character of silica nanoparticles in the EOR process it still requires in-depth research before it can finally be applied in the oil field. This research was set to investigate the stability of silica nanoparticles-CO_2 foam where silica nanoparticles used are hydrophobic. Since the test is carried out in the room condition where CO_2 is very difficult to put into a test tube, iso-octane is used instead of CO_2 gas which according to Al Otaibi, Kokal, Chang, AlQahtani, & AlAbdulwahab (2013) that the character of iso-octane is almost similar to CO_2 gas in supercritical conditions. Furthermore, the stability and performance of the emulsion formed between silica nanoparticles and iso-octane were evaluated in the laboratory at different brine salinities to determine its potential as an EOR agent.

The fluid parameters associated with the EOR process which was evaluated in this study include; density, viscosity, mobility ratio (M) and resistance factor (R). Where mobility ratio (M) is defined as the ratio between the mobility of displacing fluid to the mobility of displaced fluid. An EOR process that has good macroscopic sweep efficiency will have a small mobility ratio value. Mobility ratio can be written in the following equation:

$$M = \frac{\lambda_D}{\lambda_d} = \frac{\lambda_{np}}{\lambda_o} = \frac{(k_{rnp}/\mu_{np})}{(k_{ro}/\mu_o)} = \frac{(k_{rnp}\times\mu_o)}{(k_{ro}\times\mu_{np})} \quad (1)$$

where λ_D is the displacing phase mobility, λ_d is the displaced phase mobility, λ_{np} is the nanoparticles suspension mobility, λ_o is the oil mobility, μ_{np} is the nano particles suspension viscosity, μ_o is the oil viscosity, k_{rnp} is the relative permeability to nanoparticles suspension at the average water saturation behind the waterfront, k_{ro} is the relative permeability to oil ahead of the flood front at S_{wirr} behind the waterfront.

Unfortunately, since the process of displacement of oil in porous media by silica nanoparticles suspension was not carried out in this study (optional), then the relative permeability (kr) value could not be obtained, consequently the mobility ratio parameter could not be calculated. Meanwhile the resistance factor (R) parameter is defined as the ratio between the mobility of water to the mobility of the displacing phase (nanoparticles). Resistance factor is also often associated with the reduction of the relative permeability to water because amount of the displacing phase is adsorbed onto the rock surfaces. Generally, an EOR process which has good volumetric sweep efficiency will have a high resistance factor value (Green & Willhite, 1998). Resistance factor can be written in the following equation:

$$R = \frac{\lambda_w}{\lambda_{np}} = \frac{(k_{rw}/\mu_w)}{(k_{rnp}/\mu_{np})} = \frac{(k_{rw}\times\mu_{np})}{(k_{rnp}\times\mu_w)} \quad (2)$$

where λ_w is the water mobility, k_{rw} is the relative permeability to water at the average water saturation behind the waterfront, μ_w is the water viscosity, k_{rw} is the relative permeability to water at the average water saturation behind the waterfront.

Because this research was conducted in static conditions, the calculation of resistance factor parameter only relied on the magnitude of the viscosity ratio between nanoparticles and water.

MATERIAL AND METHOD

Equipment

Ostwald viscometer, picnometer, microscope, digital balance, hot plate magnetic stirrer, beaker glass, filter paper, thermometer, test tubes and racks.

Materials

Hydrophobic silica nanoparticles, iso-octane, aquadest, NaCl, solvents.

Measurement of Fluid Properties

The properties of the fluids that were measured include: density with picnometer and fluid viscosity with Ostwald viscometer at room temperature, concentration and salinity that have been determined in the study. All measurements and testing were conducted in room temperature condition.

Static Foam/Emulsion Test

The indicator in stability testing for a static foam/emulsion is when foam/emulsion is produced, there is no change in the bubble/blob column volume and there is no change in their size. The stages of testing are as follows:

a) Static foam/emulsion stability testing is conducted by mixing silica nanoparticles dispersion in brine with CO_2 or iso-octane in test tubes at room temperature with varying concentrations of silica nanoparticles between 0 to 2 w/v% and brine salinity (10000, 32500, and 65000 ppm), then the suspension of the silica nanoparticles with CO_2 or iso-octane was shaken vigorously until the foams/emulsions were completely formed. The suspensions, contain foams/emulsions, were kept for several weeks to observe their stability. The selection of brine concentration was based on the range of typical Indonesian oil field reservoir salinity. Meanwhile the concentration of nanoparticles was varied to adopt several studies conducted by some researchers.
b) Foams/emulsions column height in the test tubes were measured as a function of time. The macroscopic bubble/blob structure of stable emulsion was also characterized by using an optical microscope equipped with a digital camera.
c) Image recording was conducted during the stability test to observe the changes in bubbles/blobs size in the test tubes.

Tests were carried out on various concentrations of silica nanoparticles, brine salinity and phase volume ratio to obtain a stable foam/emulsion design.

Displacement Test in Porous Media (Optional)

To find out the performance of the foams/emulsions on increasing the oil recovery, it is necessary to conduct oil displacement test in 2D porous media. However, this work was not performed because of inadequate laboratory equipment. The flowchart of research methodology can be seen in Figure 1.

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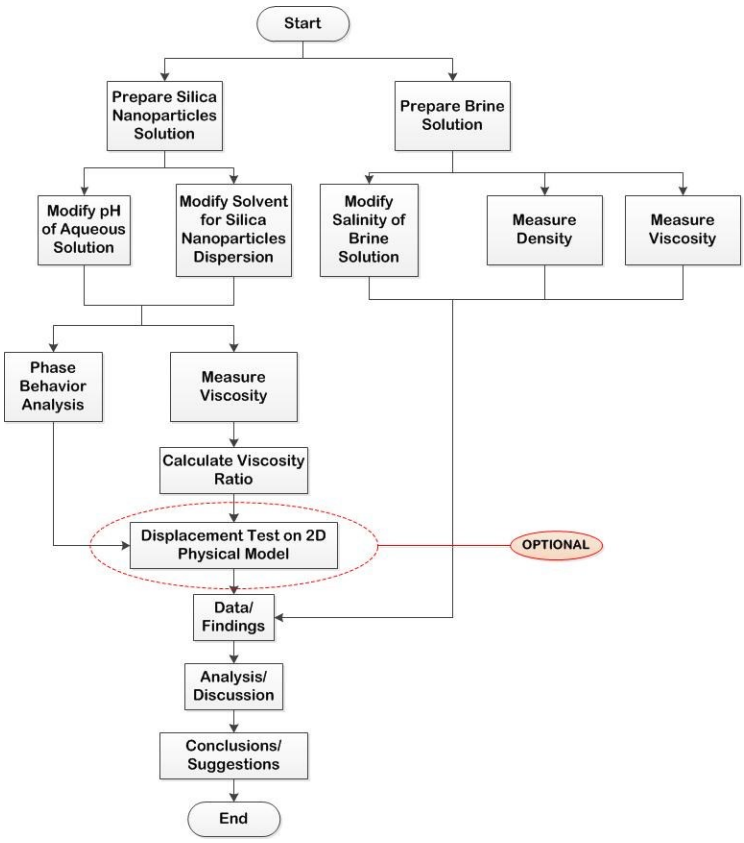

Figure 1. Research methodology.

RESULTS AND DISCUSSION

Viscosity

Based on results of measurements of density and viscosity of pure NaCl solution and NaCl solution added with silica nanoparticles, it was concluded that high brine salinity (high NaCl concentration) will result high density of solution but did not have a major effect on brine viscosity. The addition of silica nanoparticles caused the viscosity of water to increase, the higher the concentration of silica nanoparticles added, the greater the viscosity of the suspension. This is also indicated by the increased viscosity ratio between silica nanoparticles to NaCl solution as shown in Table 1. Theoretically, a higher viscosity ratio will cause a lower mobility ratio and as a result the sweep efficiency during enhanced oil recovery processes will increase.

Table 1. Fluids density and viscosity data.

Fluid	Density (gr/cc)	Viscosity (cP)	μ_{np}/μ_w
Brine 10.000 ppm NaCl	1.11	0.955	1
Brine 10.000 ppm NaCl + Silica Nanoparticles 0.5 w/v%	1.12	1.041	1.09
Brine 10.000 ppm NaCl + Silica Nanoparticles 2 w/v%	1.125	1.097	1.15
Brine 32.500 ppm NaCl	1.12	1.008	1
Brine 32.500 ppm NaCl + Silica Nanoparticles 1.25 w/v%	1.125	1.069	1.06
Brine 65.000 ppm NaCl	1.15	1.075	1
Brine 65.000 ppm NaCl + Silica Nanoparticles 0.5 w/v%	1.143	1.095	1.02
Brine 65.000 ppm NaCl + Silica Nanoparticles 2 w/v%	1.15	1.159	1.08

Phase Behavior Analysis

Based on the results of the phase behavior test (as shown in Figure 2), it was shown that above 0.05 w/v% the suspension of hydrophobic silica nanoparticles at room temperature will form more foam volume in line with the concentration and time. Silica nanoparticles depositions were observed to be relatively stable along with time as shown in Figure 3. Basically, a good nanofluid injection as an EOR agent must has a large, viscous and stable volume of foam/emulsion and very little or no deposition (Mo, Yu, Liu, & Lee, 2012). The temperature fluctuations during measurements in room conditions were considered to affect the results slightly. Nevertheless, the results of the study indicate that stabilized foams generated by silica nanoparticles and injected gas can be recommended for use as mobility control agent in the EOR processes.

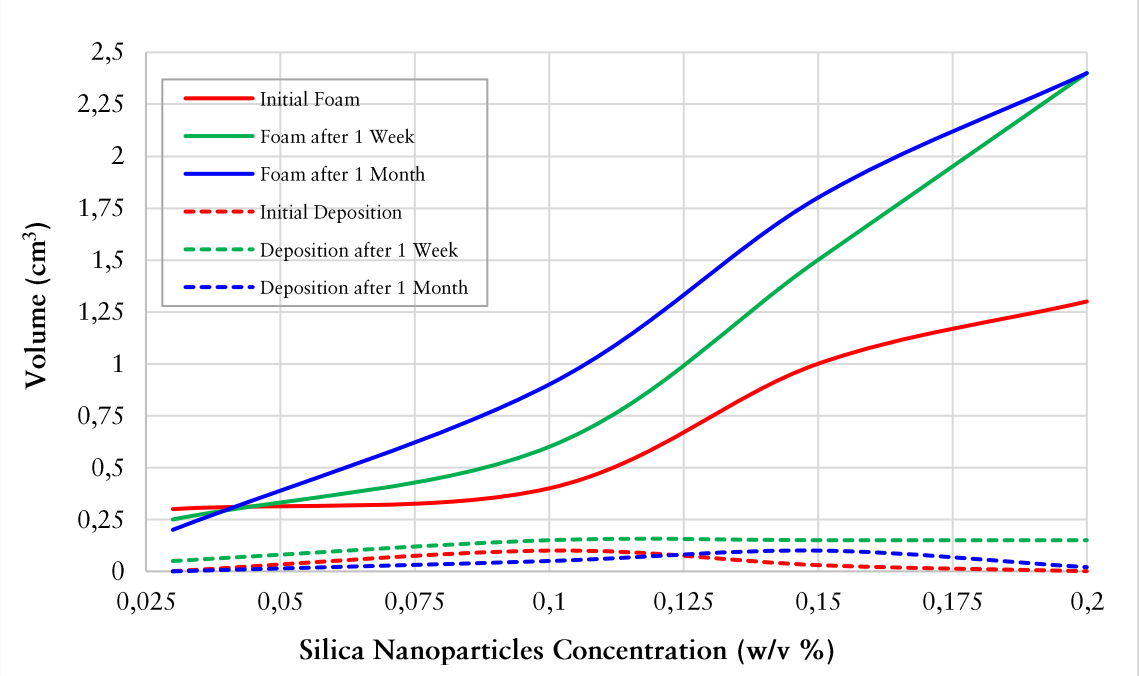

Figure 2. Foam stability curve and silica nanoparticles suspension deposition.

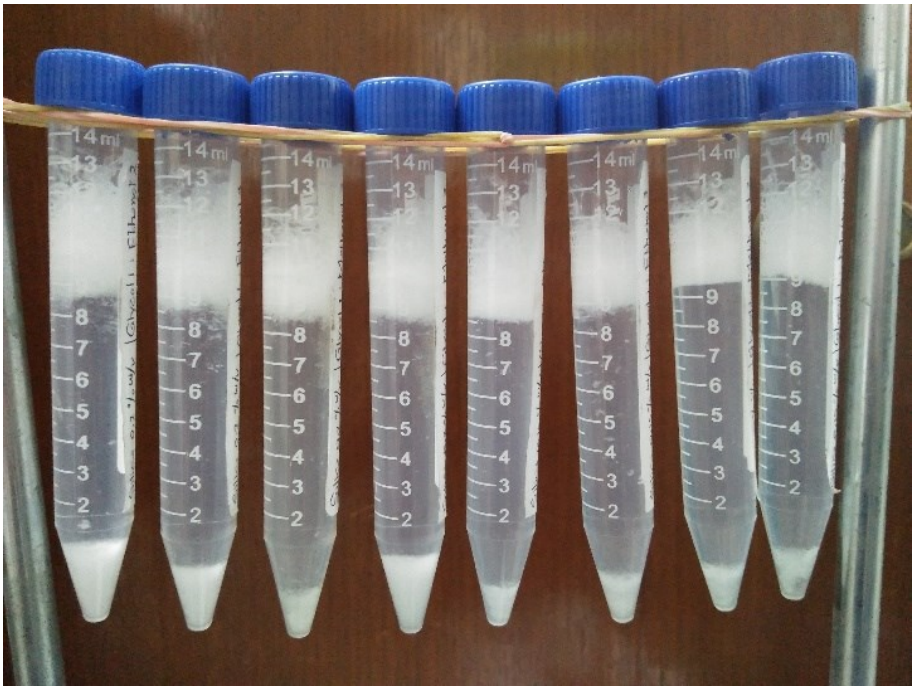

Figure 3. Phase behavior analysis (foam & deposition observation).

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Emulsion Stability

By varying the phase volume ratio between silica nanoparticles and iso-octane (as CO_2 surrogate substance) in laboratory, the best injection volume composition between silica nanoparticles and CO_2 will be obtained eventually. The thick red line as shown in Figure 4 represents the suspension of silica nanoparticles (NP) with iso-octane (IO) with a volume ratio of 5:5 (ml/ml), while the dashed red line shows the performance of the mixture after one month. Based on this curve it was interpreted that the best injection composition with the highest emulsion volume and good stability (highest stable emulsion volume percentage as shown in Table 2) was obtained by combining silica nanoparticles with CO_2 with a composition of 1:1 at concentrations around 1.3 w /v% (highest foam volume but less stable) or 0.8 w/v% (second best option with lowest concentration and more stable foams). The resulting suspension has a clear color (translucent) and the emulsion formed was water in iso-octane (w/o) emulsion type (Figure 5).

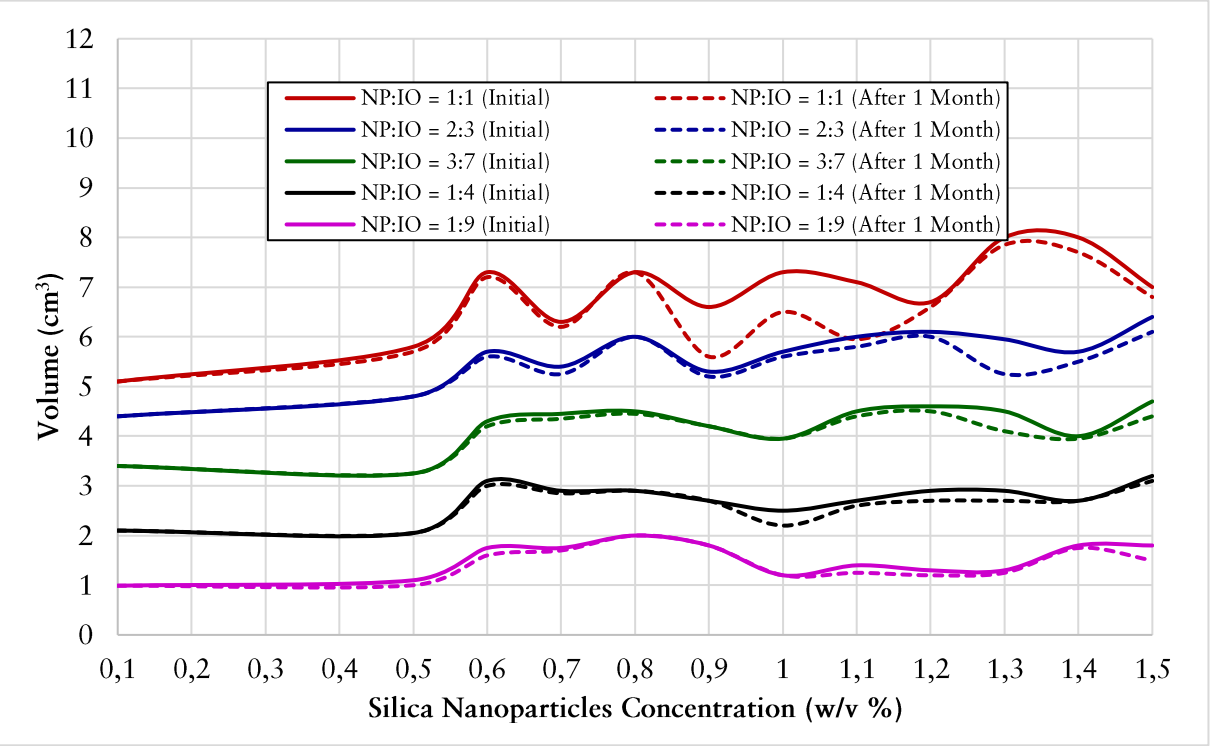

Figure 4. The relationship between nanoparticles/iso-octane phase volume ratio and emulsion volume.

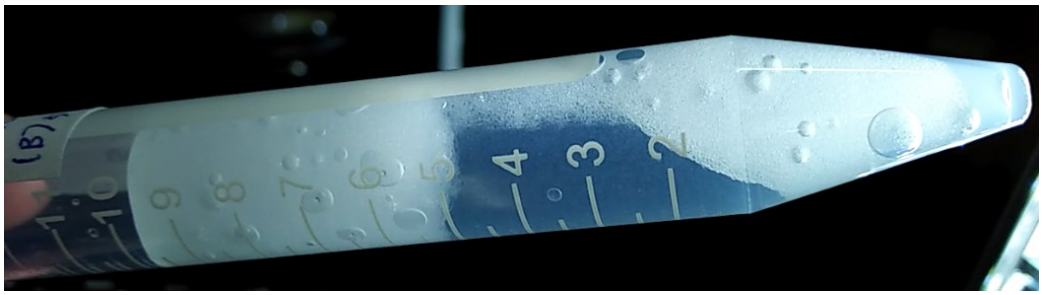

Figure 5. Photo of emulsion formed by silica nanoparticles-iso octane suspension.

Table 2. Emulsion volume and stable emulsion volume percentage data.

NP: IO Ratio	NP Concentration (w/v %)	Volume (cm^3)		Stable Emulsion (%)
		Initial	After 1 Month	
1:1	0.1	5.1	5.1	100.0
1:1	0.5	5.8	5.7	98.3
1:1	0.6	7.3	7.2	98.6

1:1	0.7	6.3	6.2	98.4
1:1	0.8	7.3	7.3	100.0
1:1	0.9	6.6	5.6	84.8
1:1	1	7.3	6.5	89.0
1:1	1.1	7.1	5.95	83.8
1:1	1.2	6.7	6.6	98.5
1:1	1.3	8	7.85	98.1
1:1	1.4	8	7.7	96.3
1:1	1.5	7	6.8	97.1
2:3	0.1	4.4	4.4	100.0
2:3	0.5	4.8	4.8	100.0
2:3	0.6	5.7	5.6	98.2
2:3	0.7	5.4	5.25	97.2
2:3	0.8	6	6	100.0
2:3	0.9	5.3	5.2	98.1
2:3	1	5.7	5.6	98.2
2:3	1.1	6	5.8	96.7
2:3	1.2	6.1	6	98.4
2:3	1.3	5.95	5.25	88.2
2:3	1.4	5.7	5.5	96.5
2:3	1.5	6.4	6.1	95.3
3:7	0.1	3.4	3.4	100.0
3:7	0.5	3.25	3.25	100.0
3:7	0.6	4.3	4.2	97.7
3:7	0.7	4.45	4.35	97.8
3:7	0.8	4.5	4.45	98.9
3:7	0.9	4.2	4.2	100.0
3:7	1	3.95	3.95	100.0
3:7	1.1	4.5	4.4	97.8
3:7	1.2	4.6	4.5	97.8
3:7	1.3	4.5	4.1	91.1
3:7	1.4	4	3.95	98.8
3:7	1.5	4.7	4.4	93.6
1:4	0.1	2.1	2.1	100.0
1:4	0.5	2.05	2.05	100.0
1:4	0.6	3.1	3	96.8
1:4	0.7	2.9	2.85	98.3
1:4	0.8	2.9	2.9	100.0
1:4	0.9	2.7	2.7	100.0
1:4	1	2.5	2.2	88.0
1:4	1.1	2.7	2.6	96.3
1:4	1.2	2.9	2.7	93.1
1:4	1.3	2.9	2.7	93.1
1:4	1.4	2.7	2.7	100.0
1:4	1.5	3.2	3.1	96.9
1:9	0.1	0.99	0.99	100.0

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1:9	0.5	1.1	1	90.9
1:9	0.6	1.75	1.6	91.4
1:9	0.7	1.75	1.7	97.1
1:9	0.8	2	2	100.0
1:9	0.9	1.8	1.8	100.0
1:9	1	1.2	1.2	100.0
1:9	1.1	1.4	1.25	89.3
1:9	1.2	1.3	1.2	92.3
1:9	1.3	1.3	1.25	96.2
1:9	1.4	1.8	1.75	97.2
1:9	1.5	1.8	1.5	83.3

Emulsion Size

Based on observations under a microscope with a magnification of about 500 times as shown in Figure 6, it was shown that the size of the nanoemulsions formed were relatively uniform (2.4 µm-8.8 µm) and spread evenly on suspensions made without aggregates. Therefore, silica nanoparticles are recommended to be used as injection fluid in the oil field for an EOR process due to their stability, performance and nano size which assumed to not cause plugging in microscopic pore spaces of oil reservoir rocks.

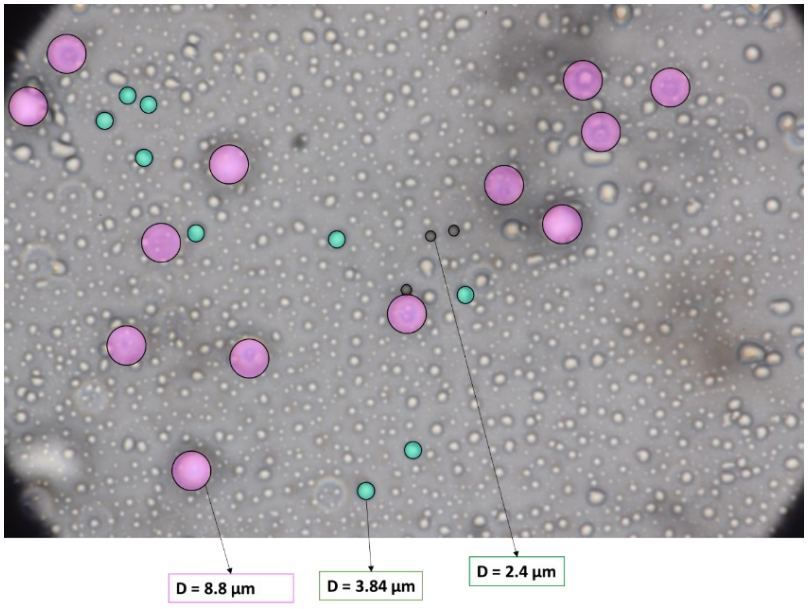

Figure 6. Structure of silica nanoparticles-iso octane emulsion with magnification under microscope.

CONCLUSION

This research aims to examine the stability of the foams/emulsions, compatibility and phase behavior of suspensions generated by hydrophobic silica nanoparticles on various salinity of formation water as well as to determine its effect on the mobility ratio parameter. It was found that increasing the concentration of silica nanoparticles will significantly impact the volume of emulsions or foams formed. This will also cause the viscosity to increase but contrary will decrease the mobility ratio towards favorable mobility. Higher brine salinity will reduce the viscosity ratio between the suspension of silica nanoparticles and brine, consequently the resistance factor will also indirectly decrease and finally will reduce the sweep efficiency of oil in the reservoir. The stability of emulsions or foams generated by silica nanoparticles were particularly affected by concentration, salinity and phase volume ratio between suspension of silica nanoparticles with iso-octane or CO_2 gas.

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