Buoyancy Driven Motion of Crude Oil Droplet within Aqueous Solutions

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Abstract

The flow behavior of the crude oil droplet motion within different aqueous solutions was studied in terms of several parameters. These parameters are the effect of surface active material, salt concentration, and polymer concentration. The crude oil droplet motion under low Reynolds number was investigated in terms of rising velocity, steady shapes, and dimensionless geometric parameters. Different Newtonian and Non-Newtonian aqueous solutions were used as continuous phase. The droplets motion due to buoyancy effects was examined in a capillary tube positioned vertically. The mobility of the crude oil droplet and the steady shape deformation are influenced by the presence of the surfactant which, in turn, affects surface tension of the droplets. Presence of salt in the continuous phase also affects the crude oil droplet shape.

Keywords: droplet motion; surfactant; non-Newtonian fluids, crude oil, buoyancy effect.

Introduction

The droplet motion of one immiscible fluid within a continuum phase of another fluid is of great importance in several industrial and technological applications such as extraction of liquid–liquid mixture, breakup of emulsions, pipeline transportation of liquid–liquid mixture, and in the oil industry where oil and water are often produced and transported together. The knowledge of the flow motion behavior of the crude oil droplet within immiscible aqueous solution is essential for the transported industrial processes as in mass, heat, and momentum transfer. The interaction between the dispersed and continuous phases plays an important role in the droplet flow behavior characteristics. The forces created from the continuous phase due to the motion of oil droplet tend to deform the oil droplet, however, the interfacial tension and viscosity of the oil droplet counteracts the external effect and tends to stabilize it.[1]

The investigation of drops and bubbles motion through Newtonian fluids has been found in several experimental and theoretical studies. Taylor[2-3] was one of the first to study the drop deformations through the investigation of a fluid viscosity containing small drops of another fluid. Koh and Leal[4-5] carried out an experimental investigation on the stability of viscous drops through a quiescent fluid. They found that a single drop would not regain a spherical shape if the initial deformation was sufficiently large. Acrivos[6] studied the breakup of small droplets in shear flow environment, and he reported that the droplet breakup will occur at a critical value of capillary number which depends upon the physical properties and the imposed fluid flow. Stone and Leal[7] investigated the relaxation and breakup of an initially extended droplet in quiescent fluid.

The droplet motion within the surrounding fluid creates streamlines which diverge from the axis of symmetry on the upfront side and converge on the trailing side. This behavior will flatten the leading droplet and cause the trailing droplet to stretch.[8] Numerical simulation using a three dimensional boundary integral method of a larger droplet moving past a smaller one which becomes stretched and finally breaks up or becomes entrained and finally coalescent has been investigated.[8-12] Barton et al.[13] studied the liquid droplet migration in a vertical temperature gradient. This droplet migration can be attributed to the effect of temperature to lower the interfacial tension. Therefore, the liquid droplet will move toward warmer regions, which is called thermo-capillary migration. Ha et al.[14] investigated the effect of nonionic surfactant on the deformation and breakup of a liquid droplet in an electric field. It is reported that the nonionic surfactant affects the degree of deformation and the modes of breakup through the Marangoni flow resulting from the non-uniform distribution on the droplet interface. Olbricht and Kung[15] studied the deformation and breakup of liquid droplets in low Reynolds number flow through a straight circular capillary tube for capillary number range of 0.05-1.0 and for three order of magnitude of viscosity ratio. If the viscosity ratio is small, an indentation in the droplet trailing side will grow and entrain the continuous phase inside the droplet boundaries. However, the droplet stretches along the tube axis until it breaks if the viscosity ratio reaches order of 1.0. Ho and Leal[16], Olbricht and Leal[17], and Borhan and Pallinti[18] studied experimentally the creep motion of small immiscible droplets through a circular tube. Their experimental data for droplet speeds are over predicted by the analysis of Martinez and Udell.[19] This discrepancy can be referred to the presence of surfactant in the

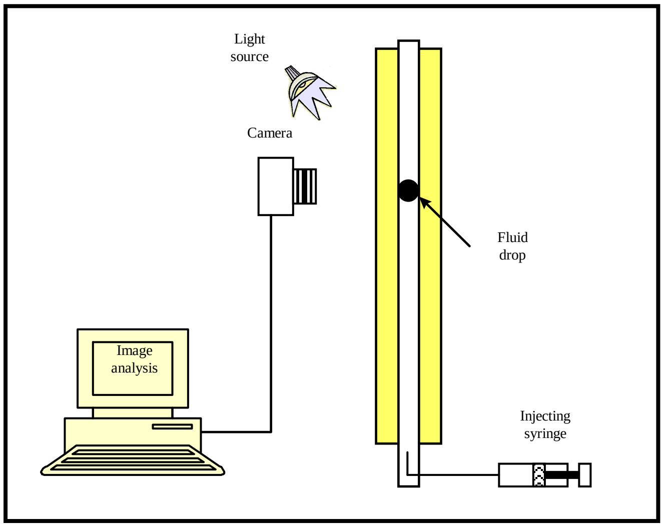

Fig. 1. Schematic illustration of the experimental setup.

experiments (AlMatroushi and Borhan,[20] Borhan and Mao,[21] Johnson and Borhan[22]).

The objective of the current investigation is to study experimentally the crude oil droplets steady motion and their deformation within different aqueous solutions. This investigation employed low viscosity crude oil for liquid droplets with different diameters. Several aqueous solutions will be used as continuous phase such as glycerol, Triton X-100, NaCl, and Alcoflood polymers of AF1235 and AF1285.

Experimental Work

Experimental setup is designed to study the steady motion characteristics of the crude oil droplet flowing through different aqueous solutions of Newtonian and non-Newtonian behavior. A schematic diagram of the experimental setup is shown in Figure 1. The experimental system consists of an outer rectangular tube with base dimensions of 10 x 10 cm with 100 cm height of Plexiglas material. An inner circular tube is made of glass with an outside diameter of 8 cm and 120 cm height. The annular space between the inner glass tube and the outer rectangular Plexiglas tube was filled with a sodium iodide solution. In order to minimize the optical distortions due to the light refractions, the refractive index of the sodium iodide solution was matched with that of the glass tube.

A crude oil droplet of controlled volume is injected through a micrometer syringe fitted at the center line of the bottom section of the inner glass tube. A wide range, 5-70 µl, of crude oil droplet volume was covered in this study. Crude oil from Bu-Hasa oil field-United Arab Emirates is employed throughout this investigation. The crude oil contains 0.42 wt % and 1.33 wt % asphaltene and sulphur, respectively. The dynamic viscosity and density of the crude oil at 25 ^{o}C are 4.79 mPas and 827 kg / m^3, respectively. The apparent viscosity of crude oil was investigated versus shear rate over the range of 0.1-1,000 s^{-1} to study the flow behavior. Crude oil appears to exhibit a Newtonian behavior with constant viscosity of 4.79 mPas at 25 ^{o}C. The inner tube was filled with different aqueous solutions to the height of 80 cm. Table 1 shows the physical properties of all examined aqueous solutions.

Table 1. Physical Properties of Examined Aqueous Solutions

Aqueous Solution	Density kg / m^3	Interfacial Tension mN / m	Viscosity mPa.s	
			m	n
100% glycerin	1260	42.40	977	1.0
90% glycerin	1240	41.94	214	1.0
75% glycerin	1210	74.3	42.2	1.0
50% glycerin	1140	74	7.5	1.0
100% gly.+ 0.05% surf.	1256	31.4	934	1.0
100% gly. + 0.1% surf.	1255	18.5	915	1.0
0% NaCL-Water	996	48	0.9	1.0
1% NaCL-Water	999	45	.97	1.0
2% NaCL-Water	1006	27.2	0.99	1.0
3% NaCL-Water	1016	25.5	1	1.0
100 ppm AF1235	995.2	10.9	11	0.62
1000 ppm AF1235	995.6	19.6	83	0.60
5000 ppm AF1235	996.9	25.7	2000	0.34
100 ppm AF1285	995.3	24.8	11	0.66

The shape of the crude oil droplet was recorded through a high-resolution color video camera from SONY (DCR-PC100E) at various height positions. A powerful light source is employed for the photography recording purposes. All experimental images were recorded at room temperature of 25 ^{o}C. Image-pro image analysis system software was used to characterize and analyze the crude oil droplet shape by measuring the perimeter, maximum axial and radial dimensions. The terminal velocity of the crude oil droplet was estimated by determining the elapsed time required to travel a certain vertical distance. The terminal velocity was determined over a region away from the droplet inlet to avoid the entrance effects.

Several aqueous solutions were selected carefully to cover a wide range of different physical properties as reported in Table 1. Pure glycerol from BDH Middle East L.L.C., United Arab Emirates was employed to represent a Newtonian behavior. Six different concentrations of glycerol solutions were used over the viscosity range of 7.5-977 mPa.s to investigate the role of viscosity on the crude oil droplet deformation. Density of all examined solutions was measured by a pycnometer, however, Ostwald viscometer is employed to measure Newtonian viscosity. Nonionic surfactant of Triton X-100 (iso-Octylphenoxypolyethoxy ethanol from BDH Middle East L.L.C.–United Arab Emirates) in the range of 0-0.1 wt % was used with glycerol solutions to control the interfacial tension quite significantly. The interfacial tensions of crude oil-aqueous solutions were measured by a spinning droplet tensiometer (from Core Laboratories Inc., model 1500, Dallas-USA). Brine solutions of 0-3 wt % NaCl concentration were examined as well.

To investigate the flow behavior of crude oil droplet within non-Newtonian behavior solutions, two Alcoflood polymers were used. These Alcoflood polymers, AF1235 and AF1285 were obtained from Ciba Specialty Chemicals (Bradford, West Yorks, England). Alcoflood polymers are high molecular weight polyacrylamide copolymers supplied in a granular powder with a bulk density of 800 kg / m^3. The intrinsic

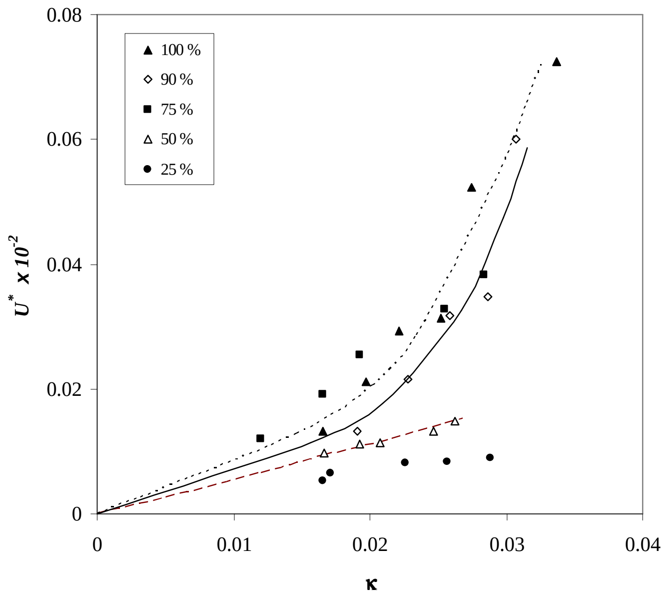

Fig. 2. Effect of glycerol concentration on the rise velocity of crude oil droplet in systems with Bo=0.11-0.13.

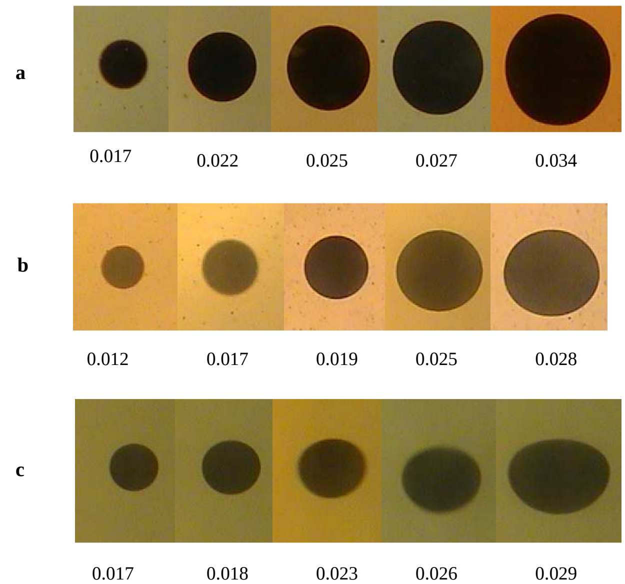

Fig. 3. Steady shapes of oil droplets rising in glycerol-water system; (a) 100% glycerol with *Bo* = 0.13; (b) 75% glycerol-water with *Bo* = 0.09 and (c) 25% glycerol-water with *Bo* = 0.11; where value under each image indicates droplet size ratio κ.

viscosities of the two polymers of AF1235 and AF1285 are 12 and 24 respectively. The Alcoflood polymers are manufactured for the enhanced oil recovery programs floods. AF1235 has good handling characteristics to promote better injection into low to medium permeability reservoirs (50-500 mD), whereas AF1285 is utilized for high permeability reservoirs. Both polymers offer good handling characteristics with excellent viscosity alteration power in water.

The rheological behavior of the Alcoflood aqueous solutions was investigated extensively by Ghannam and Esmail[23] using RheoStress RS100 from Haake. A water bath was connected to the RS100 to control the applied temperature in the rheometer system. The drive shaft of the RS100 was centered by an air bearing to apply the specified stress on the tested sample without any friction. The RS100 offers operating modes of controlled rate (CR) mode, controlled stress (CS) mode, and oscillation (OSC) mode. One of the important specifications of RS100 is its capability to apply shear stress with extremely low inertia. The operating mode of RS100 can be easily switched between CR & CS modes, and it can also apply oscillating stress and frequency sweep. The measurements data was collected using a cone-plate sensor. The sensor system consisted of a stainless steel cone and plate with 35 mm diameter and 4^o of cone angle.

Different polymer concentrations in the range of 100-5000 ppm were used. Power law model sufficiently fits the flow behavior of AF1235 and AF1285 solutions for the reported concentrations. Power law model can be presented as

$$\eta = m\ \dot{\gamma}^{\,n-1} \qquad [1]$$

where η is viscosity in mPa.s, $\dot{\gamma}$ is shear rate in s^{-1}, m is consistency index in mPa.s^n, and n is flow behavior index. The results of modeling analysis in terms of m and n are reported in Table 1.

Results and Discussion

In this section, we present the experimental results in terms of the effect of surfactant concentration, salt concentration and polymer concentration on the steady shape and terminal velocity of crude oil droplets. The Reynolds number based on the terminal velocity of the oil droplet was less than 0.1 in all of the experiments reported here. The Bond number, Bo = $\Delta\rho g R^2/\sigma$, varied in the range $0.11 \le Bo \le 0.13$, where $\Delta\rho$ and σ represent the density difference and interfacial tension between the two phases, respectively. R is the tube radius, and g is the magnitude of the gravitational acceleration. We first consider the effect of the aqueous glycerol concentration on the motion of crude oil droplets. The dependence of the dimensionless terminal velocity on droplet size is shown in Figure 2 where the droplet size κ is made dimensionless with the tube radius and the velocity U is scaled with $\Delta\rho g R^2/\mu$ (μ denotes the viscosity of the suspending fluid). The solid, dotted and dashed curves in this Figure represent the best fits to the experimental data in the glycerol-water system with no addition of surfactant to the continuous phase, taking into account the fact that U must vanish as the bubble size κ tends to zero.

Typical profiles (in the meridional plane) of the steady shapes of crude oil droplet in surfactant free system are shown in Figure 3 for various droplet sizes with different glycerol-water systems. Small droplets have a spherical shape in all three glycerol-water concentrations shown in Figure 3. The steady shape of an oil droplet, in 100% glycerol system, approaches an ellipsoid as the droplet size increases, with a slight loss of fore and aft symmetry. Reducing glycerol concentration to 75% results in radial deformation of larger oil droplet and the droplet develops into an oblate shape. As the droplet size increases in 25% glycerol-water system, the

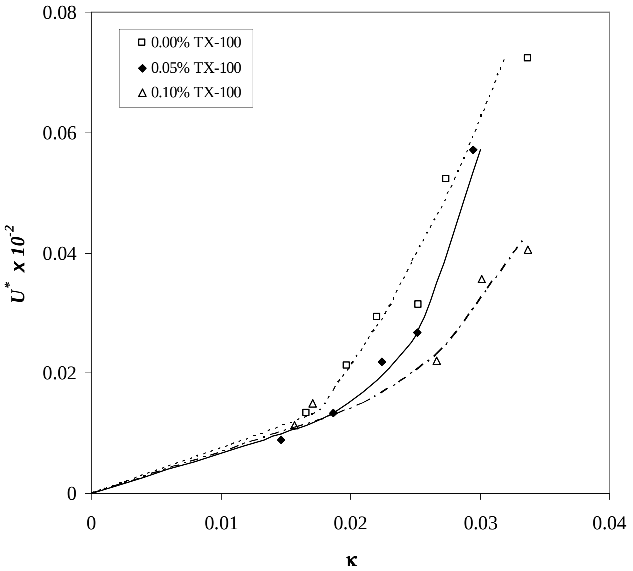

Fig. 4. Effect of surfactant concentration on the translational velocity of crude oil droplet in 100% glycerol-water systems with *Bo* = 0.13-0.12.

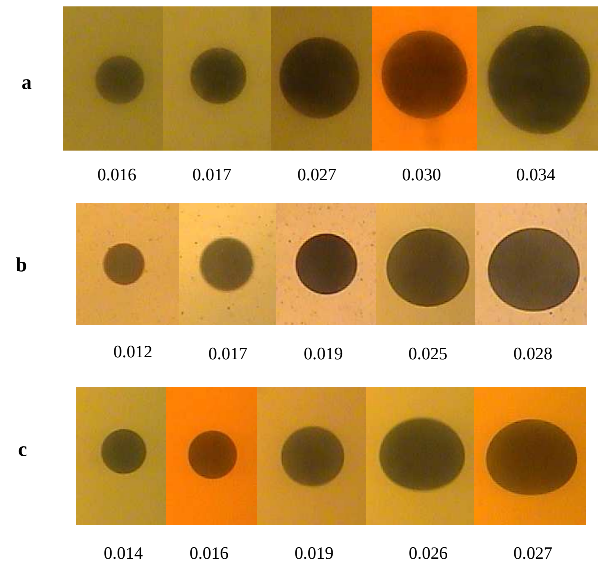

Fig. 5. Surfactant effect on steady shapes of oil drops rising in glycerol-water system; (a) 100% glycerol with 0.1 TX and *Bo* = 0.12; (b) 75% glycerol with 0.00 TX and *Bo* = 0.09; (c) 75% glycerol with 0.05 TX and *Bo* = 0.26; where value under each image indicates drop size ratio κ.

droplet shape becomes oblate with flattened front. This shape changes from ellipsoid to oblate is reflected in oil droplet terminal velocity as shown in Figure 2 where radial reformation of the droplet reduces the terminal velocity especially for larger droplet.

The presences of surfactant in the 100% glycerol-water system results in retarding effect on the crude oil droplet translating velocity as shown in Figure 4. The translating velocity gradually decreases with surfactant concentration where the effect of surfactant is more pronounced at a large value of κ. The addition of surfactant to the system leads to the development of Marangoni stresses on the surface of the rising oil droplet as surfactant monomers diffusing to the oil droplet surface are swept to the trailing end of the oil droplet by surface flow. These Marangoni stresses oppose the surface flow and retard the motion of the oil droplet as a whole in the same manner as that observed for the rise of bubbles in an unbounded fluid. Therefore, the reduction in the terminal velocity in Figure 4 is due to the development of Marangoni stresses which become more pronounced with increasing surfactant concentration.

The addition of surfactant to the 100% glycerol-water system results in an axial droplets deformation as the size of the oil droplet increased above κ = 0.03. This deformation is shown in Figure 5 for various droplet sizes with different surfactant concentrations. Small droplets have a spherical shape in all surfactant concentration systems. As the crude oil droplet size increases, a slight deformation in the axial direction starts to appear and the droplet loses fore and aft symmetry. However, the presence of surfactant in 75% glycerol-water system enhances oil droplet deformation in the radial direction as the size of the oil droplet increased as illustrated in Figure 5-b and Figure 5-c.

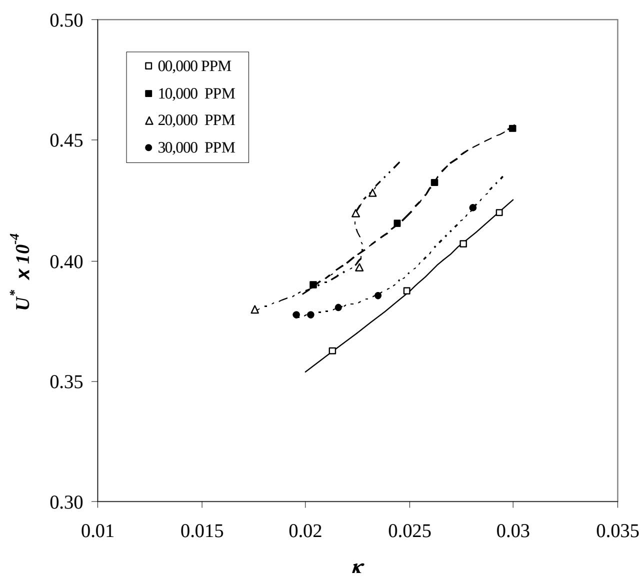

Fig. 6. Effect of NaCl concentration on the rise velocity of crude oil droplet in systems with *Bo*= 0.05-0.11.

To investigate salt effect on crude oil droplet deformation and motion, different concentrations of NaCl-water system are used in this study. Figure 6 reflects the effect of salt concentration on the crude oil droplet translating velocity. The presence of NaCl in the system enhances the crude oil translating velocity for all κ. This increase in translating velocity can be attributed to the effect of salt on the interfacial tension between crude oil-aqueous phases. The presence of salts in the aqueous phase, up to 20,000 ppm, has a strong ability to increase the accumulation of the surface-active species, which are available in crude oil, at the crude oil-

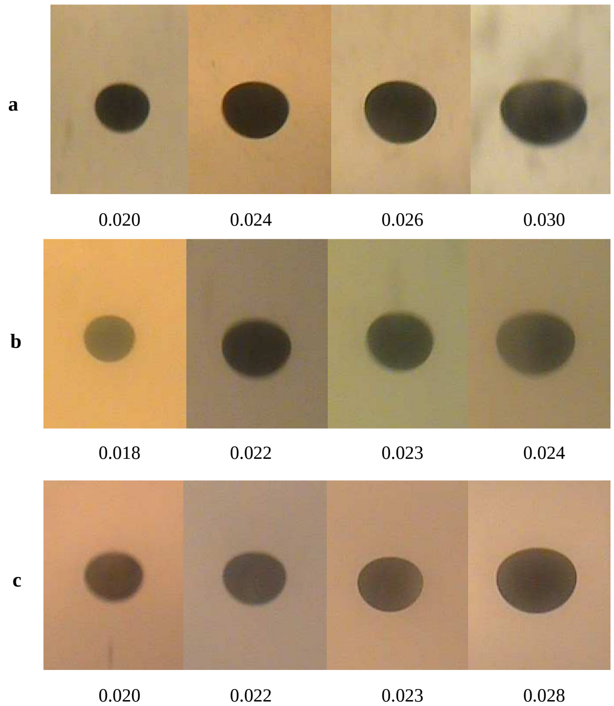

Fig. 7. Steady shapes of oil droplets rising in NaCl-water system; (a) 10,000 PPM with *Bo* = 0.06; (b) 20,000 PPM with *Bo* = 0.10 and (c) 30,000 PPM with *Bo* = 0.11; where value under each image indicates droplet size ratio κ.

aqueous phase interface. These activities reduce the interfacial tension, from 48 to 27.2 mN/m, and thereby enhance the translating velocity. However, further addition of NaCl up to 30,000 ppm will lower the crude oil translating velocity below the upper limit. The further addition of NaCl beyond the 20,000 will increase the repulsive electrostatic double-layer forces which will slightly lower the droplet translating velocity. This reduction in translating velocity leads to the droplet radial deformation even at small droplet size.

The crude oil droplet maintains steady shape as the droplet passed through the tube. Figure 7 shows shape deformation for NaCl-water system, where increasing crude oil droplet size results in radial deformation with reduction in the front curvature for larger droplets. For larger crude oil droplet size, deformation was slightly affected by high concentration of NaCl. Such deformations enhanced at low NaCl concentration and as the NaCl concentration increased, the front curvature of the crude oil droplet increased.

A few runs are made to investigate the polymer concentration effect on crude oil droplet translating velocity as shown in Figure 8. Increasing the polymer concentration reflected in substantial reduction in the terminal velocity for both polymers used in this study. Meanwhile, at a low polymer concentration of 100ppm, the crude oil droplet velocity shows almost similar behavior.

A significant deformation is observed for crude oil droplet motion in the polymer solution where the droplet developed a trailing cusp as illustrated in Figure 9. With small polymer concentration, the crude oil droplet develops into oblate shape

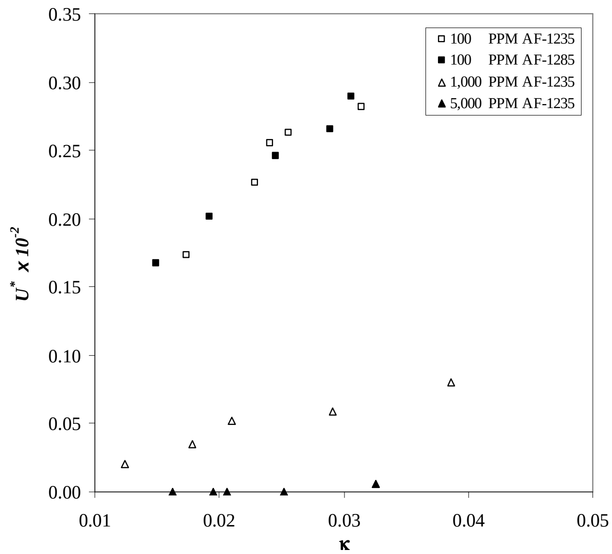

Fig. 8. Effect of polymer concentration on the rise velocity of crude oil droplet in systems with *Bo* = 0.15-0.33.

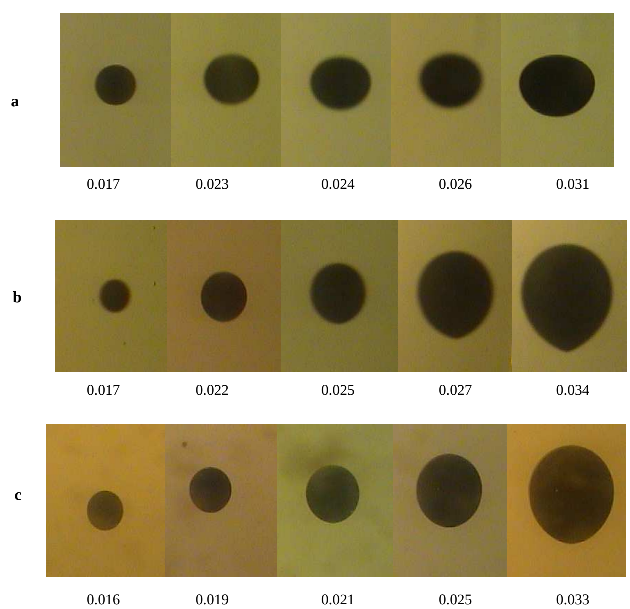

Fig. 9. Steady shapes of oil droplets rising in AF1235 polymer system; (a) 100 PPM with *Bo* = 0.23; (b) 1,000 PPM with *Bo* = 0.13 and (c) 5,000 PPM with *Bo* = 0.10 ; where value under each image indicates droplet size ratio κ.

with flattened front. Increasing the polymer concentration to 1000 ppm, results in cusp appearance at the trailing end of the droplet. This cusp is due to polymer elasticity that reported by several studies. With high polymer concentration of 5000 ppm, this cusp disappears and the crude oil droplet maintains its oblate shape. Ghannam et al.[24] studied the viscoelastic behavior of the Alcoflood polymers. They reported that the concentration of ≤ 1000 ppm, the Alcoflood polymers show viscous behavior for frequency of less than 0.15 s^{-1}, whereas predominantly elastic behavior will be displayed for frequency higher than 0.15 s^{-1}. For higher concentration, the AF1285 display only elastic response, whereas AF1235 shows elastic response if the frequency is higher than 0.3 s^{-1}.

In order to quantitatively characterize the influence of surfactants on the steady droplet shapes, a deformation parameter $D = P^2/4\pi A$ is defined, where P and A represent the perimeter and area of the deformed droplet profile in the meridional plane, respectively. This quantity provides a measure of deviations from a spherical shape (which is characterized by D=1), and it is easily obtained from digitized images of rising crude oil droplets using the Image-Pro image analysis software. Figure 10 shows the dependence of the deformation parameters on the dimensionless droplet size for different surfactant concentrations. Also Figure 10 displays the geometric parameters L_A and L_R, representing the maximum axial and radial dimensions of the steady crude oil droplet profile relative to the tube radius, respectively. The dashed lines in Figure 10 denote the values of the geometric parameters for spherical droplets. Both the radial and axial dimensions of the droplet initially grow linearly as a function of droplet size with the first shape transition occurring at a droplet size of about $\kappa = 0.03$.

Conclusions

The flow behavior of crude oil droplet motion is studied in the presence of different aqueous solutions of glycerol, Triton X-100, NaCl, Alcoflood materials of AF1235 and AF1285. The flow behavior is investigated in terms of rising velocity, steady shapes, and dimensionless geometric parameters. The following conclusions can be made:

1. For all examined aqueous solutions, the dimensionless terminal velocity increases significantly with the dimensionless droplet diameter.
2. For all glycerol solutions, small droplets show spherical shape. For a 100% glycerol, the crude oil droplet approaches ellipsoidal shape as the droplet size increases. Reducing the glycerol concentration causes a radial deformation which leads to oblate shape with flattened front.
3. The terminal velocity of the crude oil droplet decreases gradually with surfactant concentration due to the development of the Marangoni stresses.
4. The presence of NaCl in the aqueous phase increases the droplet translation velocity for all droplet sizes up to a concentration of 20,000 ppm.
5. A substantial reduction in the terminal velocity was reported with polymer concentration for both polymers of AF1235 and AF1285.
6. The presence of polymer causes a significant deformation for the crude oil droplet shape. This deformation depends upon polymer concentration.

For the future of the current study, it is recommended to investigate the crude oil deformation and rising velocity for both medium and high viscosity crude oil. In addition it is important to include the effect of different temperature on the

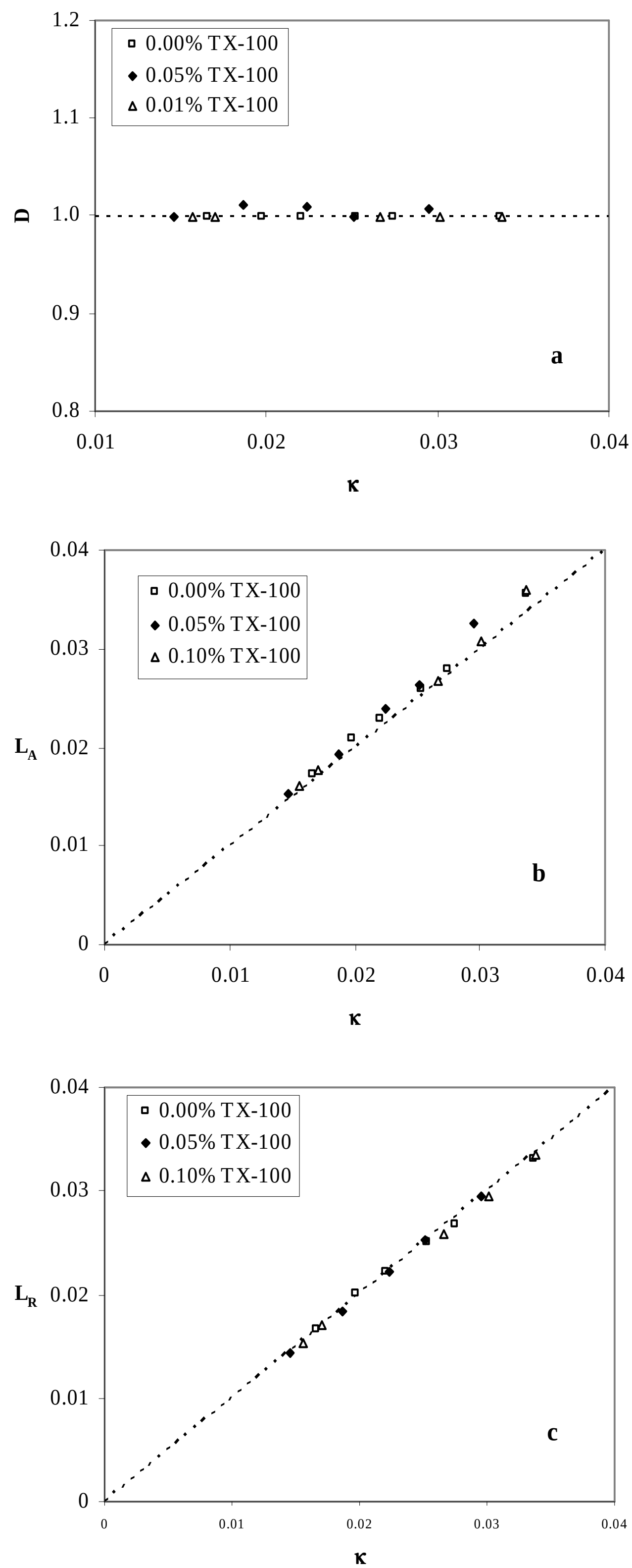

Fig. 10. Variation of the dimensionless geometric parameters with drop size for systems with Bo = 0.12 - 0.13: (a) deformation parameter, D; (b) axial length, L_A; (c) radial length, L_R.

droplet velocity and its deformation. At the end, it is planned to have theoretical and numerical simulation to study the flow behavior of crude oil droplet deformation within aqueous solutions.

Acknowledgments

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