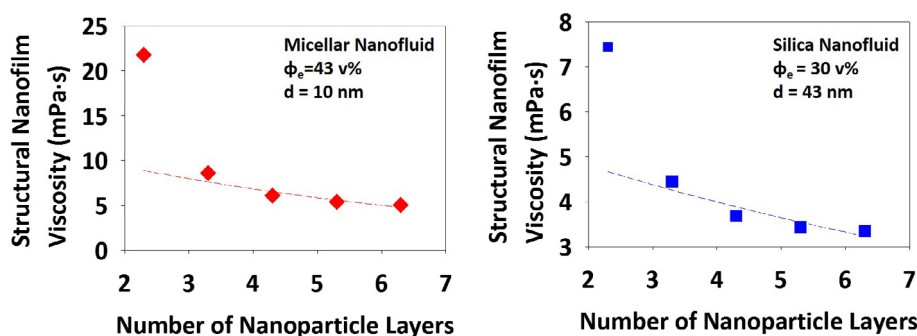Regular Article

Estimation of structural film viscosity based on the bubble rise method in a nanofluid

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GRAPHICAL ABSTRACT

ABSTRACT

When a single bubble moves at a very low capillary number (10^{-7}) through a liquid with dispersed nanoparticles (nanofluid) inside a vertical tube/capillary, a film is formed between the bubble surface and the tube wall and the nanoparticles self-layer inside the confined film. We measured the film thickness using reflected light interferometry. We calculated the film structural energy isotherm vs. the film thickness from the film-meniscus contact angle measurements using the reflected light interferometric method. Based on the experimental measurement of the film thickness and the calculated values of the film structural energy barrier, we estimated the structural film viscosity vs. the film thickness using the Frenkel approach. Because of the nanoparticle film self-layering phenomenon, we observed a gradual increase in the film viscosity with the decreasing film thickness. However, we observed a significant increase in the film viscosity accompanied by a step-wise decrease in the bubble velocity when the film thickness decreased from 3 to 2 particle layers due to the structural transition in the film.

1. Introduction

The self-layering of molecules/nanoparticles in confined geometries is well known [1–15]. It is also well established that the properties (such as viscosity) of thin liquid films are quite different from those of the bulk [16–18]. Many investigators have used the surface force apparatus to determine the film viscosity. They found that the film viscosity depends on the film thickness, and that it is much higher than that of the bulk [19–24]. Several years ago [25], we found that the film viscosity was 100 times greater than that of the bulk by using stratifying foam films formed from latex aqueous suspensions of nanoparticles and by monitoring the rising speed of the dark spots. Langevin and others used a similar method with a surfactant micellar solution and reported that the film viscosity was 30 times greater than the bulk viscosity

[26,27]. Recently, we studied the motion of air bubbles in tubes filled with an aqueous silica dioxide nanoparticle suspension and observed the novel phenomenon of a step-wise change in the bubble velocity, which we attributed to the nanoparticle self-layering phenomenon in the film adjacent to the tube wall [28].

In our research, we measured the rise of a single bubble in a vertical tube filled with a nanofluid; we also used reflected light interferometry to directly monitor the film thickness (the number of nanoparticle layers in the film). We measured the film-meniscus contact angle using reflected light interferometry and calculated the film interaction energy isotherm vs. film thickness.

2. Results

2.1. Rising bubble velocity experiment

We measured the rising speed of bubbles in a vertical tube filled with an aqueous nanofluid containing a surfactant of non-ionic micelles (micelle size = 10 nm) at a constant velocity. The position of the bubble inside the glass tube with time was measured via digital camera (Cannon A720 IS). The video was analyzed to estimate the bubble velocity. Each experiment was conducted at a controlled temperature in the thermochamber (20 °C ± 0.1). Further details regarding the experiment are presented in our previous paper [28]. The bubble rise velocity (U) in the non-ionic micellar solution (Brij 35, φ_e = 43 v%, 0.1 M concentration, which is 2400 times higher than the critical micellar solution) was measured at various tube lengths to diameter (L/D) ratios in the vertical tube (D = 0.4 cm). The diameter of the micelles (10 nm ± 0.4) was measured with the multi-stepwise foam film thinning method [15]. The data for the bubble velocity vs. L/D and Capillary number (Ca) are plotted in Fig. 1.

The Capillary number is a function of the bubble velocity, viscosity, and surface tension, $Ca = \mu U/\sigma$, where μ is the viscosity, U is the bubble velocity, and σ is the surface tension. Since the bubble velocity is slow and the Ca is small (on the order of 10^{-7}), the bubble moving in the tube was considered to be at the equilibrium condition for estimating the role of the structural forces in the bubble's motion.

2.2. Film thickness and contact angle

An air bubble approaching an optical glass plate surface from the micellar nanofluid (0.05 M Brij 35, 1200 times CMC, surface tension, σ = 44.3 mN/m) was monitored using reflected light inter-
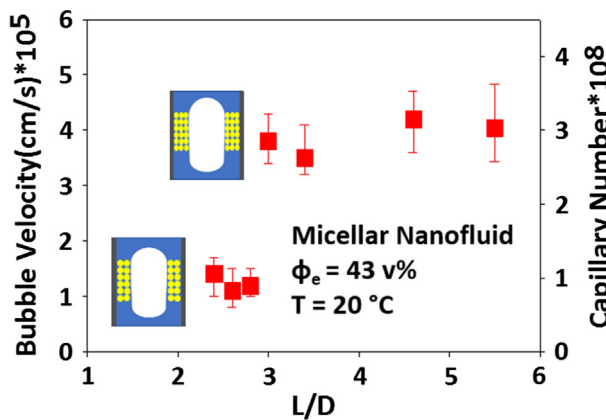

Fig. 1. The step-wise bubble velocity versus L/D ratio was measured in a non-ionic micellar nanofluid (φ_e = 43 v%). The tube diameter was 0.4 cm. The inset sketches indicate the formation of the nanoparticles inside the nanofilm with 2 and 3 micellar layers.

ferometry with a wavelength of 546 nm, and it formed a nanofluid film (868 μm). As the nanofilm between the bubble and glass surface thinned, the film thickness changed, producing interference patterns. The video camera, used in conjunction with the monitor and digital video recorder, recorded the process of the film thinning. Photomicrographs depicting the nanofluid film and the adjoining meniscus are shown in Fig. 2(a). An interferogram depicting the film with 1 micellar layer in contact with the film-meniscus is shown in the inset in Fig. 2(c). Successive maxima and minima in the intensity of the reflected light represent a change in the meniscus region thickness. The details of the experimental measurement are similar to those presented elsewhere [14,29]. The film thickness and the film/meniscus contact angle were estimated from the interference pattern at the equilibrium state. Fig. 2(c) shows the distance from the meniscus vs. the film thickness. We fitted the data with a second order polynomial with a regression coefficient of 0.995. The intercept of the fitted curve with the y-axis provided the film thickness (20.3 nm). The contact angle subtended between the film and meniscus was obtained from the local slope of the meniscus profile. The contact angle with a thickness of 1 micellar layer was obtained (0.6°).

3. Discussion

Fig. 1 shows the air bubble velocity vs. the L/D and Capillary number. Two distinguishing regions of the air bubble velocity were observed. In the region of the L/D from 2.1 to 2.8, there is a constant velocity from 3 to 5.7. However, at an L/D of 2.9, there is a step-wise change in the bubble velocity. A similar phenomenon was reported by us earlier for the bubble rise in another nanofluid (an aqueous silica suspension), and this information is shown in Fig. 3.

It should be noted that this step-wise velocity vs. bubble length was not observed in a common liquid [28]. The step-wise velocity decrease is attributed to the transition of the film from a thickness of 3 micellar layers to 2 micellar layers. We also observed (as previously reported [28]) the same transition (from 3 to 2 layers) for the silica nanofluid (see Fig. 4.).

3.1. Nanofilm structural isotherm vs. film thickness

The nanofilm structural energy ($W_{st}(h)$) is related to the film/meniscus contact angle (θ_{eq}) given by the Frumkin- Derjaguin equation.

$$\sigma(\cos\theta_{eq} - 1) = \Pi'h' + \int_{h'}^{\infty} \Pi(h)dh = \Pi'h' + W_{st} \tag{1}$$

where σ is the surface tension between the bubble and the nanofluid, h′ is the equilibrium film thickness, Π' is represented by the sum of the capillary pressure (P_c) and hydrostatic pressure, and Π is the disjoining pressure. The structural film energy is oscillatory because of nanoparticle layering within the film, and it follows the relation

$$W_{st}(h) = A\cos\left(\frac{2\pi h}{d}\right)\exp\left(-\frac{h}{d}\right) \tag{2}$$

where A is the amplitude of the oscillation, and d is the period of oscillation and decay factor.

Constant A is related to the contact angle and film thickness as shown below:

$$A\cos\left(\frac{2\pi h}{d}\right)\exp\left(-\frac{h}{d}\right) = \sigma(\cos\theta_{eq} - 1) - \Pi'h' \tag{3}$$