R&D NOTES

FLUID MECHANICS AND TRANSPORT PHENOMENA

Feasibility of N_2/Sunflower Oil Compound Drop Formation in Methanol Induced by Bubble Train

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Introduction

Alcohol and vegetable oil are immiscible and can be mixed to produce either emulsion or biodiesel, which can be used as alternative fuel for a diesel engine. Apparently, mechanical agitator has been using in either the batch (Noureddini and Zhu,[1] Darnoko and Cheryan,[2] Van Gerpen et al.[3]) or the continuous (Darnoko and Cheryan[4]) biodiesel processes. Its capital and maintenance costs can be lowered significantly if a new energy efficient mixing method is offered instead. In this case, mechanical moving parts in the tanks or vessels may have to be removed and replaced by a nonmechanical mixer. Some researchers used ultrasonic to produce biodiesel (Stavarache et al.[5,6]), with the increase of the production yields couple with reductions of processing time, and of the amount of catalyst used. Alternatively, besides the famous applications in flotation processes, a rising gas bubble is considered possibly to be one effective way to disperse alcohol into vegetable oil or vegetable oil into alcohol (Duangsuwan et al.[7]).

A preliminary attempt has been made by applying a gas bubble in the form of air/alcohol gas/liquid compound drop to rise in vegetable oil (Duangsuwan et al.[7]), with the increase of the contacting area between the alcohol and the vegetable oil through a thin film of alcohol on the bubble, and a toroidal film of alcohol flowing out of the bubble surface. This work aims to observe the feasibility of the formation of another type of liquid films in such a system, the thin film of vegetable oil engulfing the gas bubble and rising in alcohol. The simple way is to let the gas bubble to rise from the lower layer of the denser vegetable oil to the upper layer of the lighter alcohol. Single bubble streams or bubble trains were used in this work for continuous bubble generation instead of using the large single bubbles.

A large bubble easily penetrates through a liquid/liquid interface due to its buoyancy force being large enough to overcome the liquid/liquid interfacial tension force. A bubble train can also perform the penetration if the total buoyancy force acting on it is sufficient. A successful penetration can cause a transport of a lower liquid into an upper liquid in the form of entrained droplets following behind the bubble (Greene et al.[8], Reiter and Schwerdtfeger,[9] Kemiha et al.[10] and Dietrich et al.[11]) or a lower liquid film engulfing around the bubble (Hashimoto and Kawano,[12] Kawano et al.[13,14]). Based on the criteria of Torza and Mason[15] and Johnson and Sadhal[16] for static compound drop configurations, the fully and partially engulfing configurations of the air/sunflower oil compound drop in methanol are impossible (Duangsuwan et al.[7]). The results for the dynamic cases obtained from this work appear also to support the earlier predictions based on the static compound drop configurations.

Experimental Setup

Figure 1 shows the experimental setup schematically. The cylindrical glass column was 0.04 m inner dia. and 0.3 m height. It was filled with edible sunflower oil as a lower liquid phase, and methanol as an upper liquid phase. The height of the lower phase from capillary tube tip was 0.05 m, whereas the height of the upper phase from the liquid/liquid interface was 0.10 m. The 0.07 × 0.07 m square cross-sectional Plexiglas jacket filling with water was used to eliminate the light distortion caused by the cylindrical

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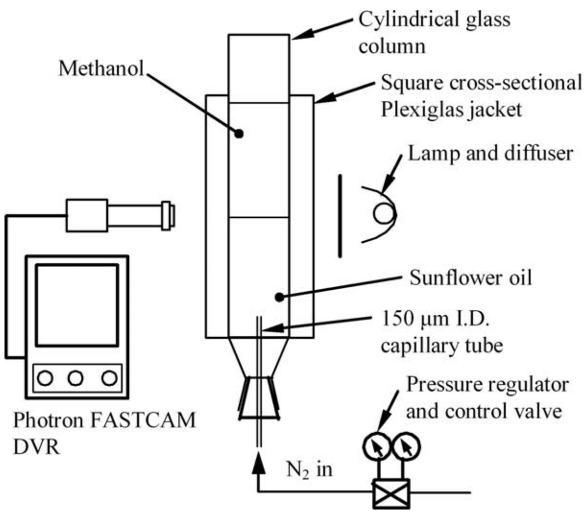

Figure 1. Experimental setup.

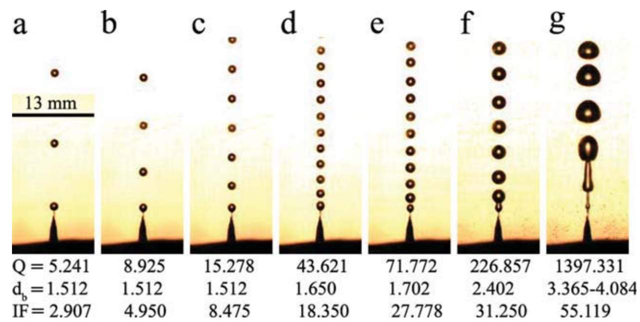

Figure 2. Example images of bubble trains in sunflower oil.

Liquid depth is 50 mm. Q = gas-flow rate (mm^3/s); d_b = bubble diameter (mm); IF = injection frequency (bubble/s). [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

column. The single bubble trains were formed by blowing N_2 gas at constant flow rates through a 150 μm inner dia. capillary tubes vertically submerged in sunflower oil phase. Photron FASTCAM Digital Video Recorder (DVR) was used to capture the experimental images. The bubble size, the rise velocity, the gas-flow rate and the bubble-injection frequency were measured by calculations from the image frames obtained. The set frame frequency for the camera was 500 frames per second. The properties of sunflower oil (see, method of determination in Duangsuwan et al.[7]), methanol (Dean[17,18]), and N_2 (Lide[19]) are shown in Table 1.

Results and Discussion

At first, the formation in sunflower oil of the bubble trains via a 150-μm I.D. capillary tube was observed, with two liquid depths from capillary tip, 50 mm and 150 mm. Figure 2 shows the example images of bubble trains formed in the 50 mm depth sunflower oil. The bubble trains in Figures 2a–f are stable because they almost have constant bubble sizes and constant distances between the adjacent bubbles. However, the unstable bubble train is observed at a very high gas-flow rate as shown in Figure 2g.

The relationship between the gas flow rate, the bubble size and the bubble injection frequency obtained are shown in Figure 3a. It shows that the bubble size is independent of the liquid depth and seems to be constant when the value of the gas-flow rate is lower than about 43 mm^3/s. In this case, surface tension force only plays a major role during detaching of the gas bubble without coalescence from the capillary tip, and the size of the bubble is independent of the gas-flow rate and could be approximated by using equation (Van Krevelen and Hoftijzer[22]; Bondarev and Romanov[23]; Snabre and Magnifotcham[24]) $d_b = [6 d_a \sigma/((\rho - \rho_g) g)]^{1/3}$, where d_b is the bubble diameter, d_a is the capillary tube inside diameter, σ is the surface tension of the continuous liquid, ρ is the density of the continuous liquid, ρ_g is the density of the gas bubble, and g is the gravitational acceleration. Higher than this flow rate, the coalescence of the bubbles during formation happens and increases the bubble size, see also, for example, Figure 2d–g. The bubble injection frequency increases as the gas-flow rate increases. However, the increase rate of the injection frequency slows down when the coalescence occurs.

Based on the results in Figures 2 and 3a, and neglecting the turbulent mixing at liquid/liquid interface caused by the large bubble and the high-injection frequency, the bubble trains with small bubble sizes at low gas-flow rate were selected to observe the feasibility of the compound drop formation. The experimental terminal velocity of the bubble train as a function of the low gas-flow rate is shown in Figure 3b. The plot shows 10 bubble trains with liquid depth 50 mm (closed circles), and 10 bubble trains with liquid depth 150 mm (opened circles) having a linear relationship with the gas-flow rate. Compared with single bubble of the same bubble size, terminal velocity of the bubble train is higher than that of the single bubble depending on the gas-flow rate. A detailed study of formation and rise of a bubble train in a viscous liquid can be found in Snabre and Magnifotcham.[24]

Table 1. Properties of the System Used

Fluid at 298 K	Density (kg m^{-3})	Dynamic viscosity (mPa s)	Surface tension (mN m^{-1})	Interfacial tension in contact with sunflower oil (mN m^{-1})
Sunflower oil	921.8	56.863	32.3	0
Methanol	787.5	0.551	22.068	4.15[a]
Nitrogen	1.131	0.01791	–	–

[a]Estimation was made from equation $\sigma_{AB} = \sigma_A + \sigma_B - 2K(\sigma_A \sigma_B)^{1/2}$ (Girifalco and Good[20]), where σ_A is the surface tension of sunflower oil, σ_B is the surface tension of methanol or ethanol; $K = K_m x\ \varphi$, where $K_m = 4(V_A)^{1/3}(V_B)^{1/3}/[(V_A)^{1/3}+(V_B)^{1/3}]^2$, V_A = molar volume of sunflower oil, V_B = molar volume of methanol or ethanol, $\varphi = 1.05$ (obtained by using the soybean oil/methanol interfacial tension from Wu et al.[21])

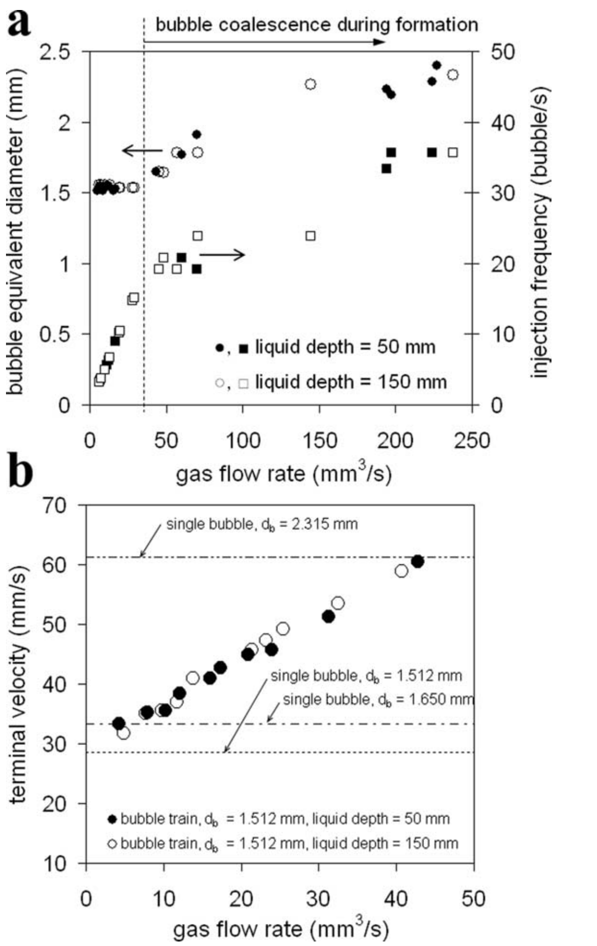

Figure 3. Formation of N_2 bubble train through a 150 μm I.D. capillary tube in sunflower oil at 298 K.

(a) Equivalent diameter and injection frequency as functions of gas flow rate, and (b) terminal velocity as a function of gas-flow rate.

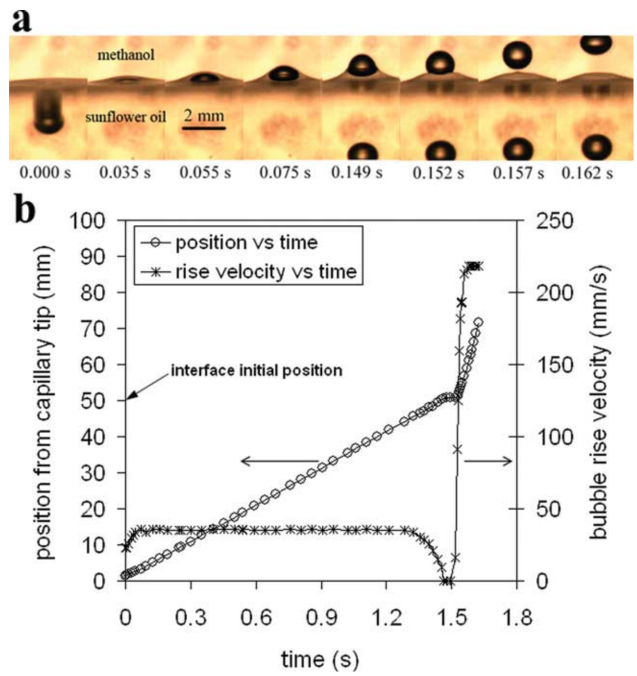

Figure 4. Penetration of bubble train A through the sunflower oil/methanol interface.

(a) Sequential images, and (b) Positions and rise velocities of a bubble in bubble train A from detachment at capillary tip until penetration at liquid/liquid interface. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

Two bubble trains, namely A and B (see formation details in Table 2), were selected for presentation. Sequential images of bubble train A during penetration through sunflower oil/methanol interface is shown in Figure 4a. The position and velocity as functions of time of a penetrating bubble in bubble train A are shown in Figure 4b. During approach to the interface, the bubble slows down until it is captured at the interface. However, after sticking at the interface for a little while, the bubble finally breaks the interface and rises into the methanol upper phase.

Transport of sunflower oil into methanol is successful when using bubble train B. During penetration, it drags a sunflower oil column into methanol phase as shown in Figure 5. The top bubble detaches from the oil column without any amount of the oil carried with it. However, it leaves a space at the tip of the oil column for the second bubble to continue rising and extending the column. Figure 6a shows that the 1st –4th bubble detach from the oil column at the different heights, whereas Figure 6b shows that these detachments accelerate the velocities of the detaching bubbles and, at the same time, decelerates those of the following adjacent bubbles (see, i.e. at: t = 0.024–0.028 s for 1st and 2nd bubbles; t = 0.086–0.090 for 2nd and 3rd bubbles; t = 0.158–0.168 s for 3rd and 4th bubbles; and t = 0.248–0.254 s for 4th and 5th bubbles). Figure 6b also indicates that velocity of the oil column increases along the height.

Figure 7 shows the continuing extension of the oil column until its part detaches from the sunflower oil/methanol interface. The stream of the bubble during penetration and detachment is not stable due to the detachment velocity is faster than the velocity of the bubble below the detaching part of the oil column. The detaching point is not certain, depending on the position, the volume and the velocity of the detaching upper part. For bubble train B, the detaching part normally has two bubbles inside, but finally it remains only one bubble due to another one splits quickly. The

Table 2. Formation Details of the Selected Bubble Trains

Bubble train	Capillary tube I.D. (μm)	Gas flow rate (mm^3 s^{-1})	Bubble diameter (mm)	Injection frequency (bubble s^{-1})	Bubble train terminal velocity (mm s^{-1})	Distance between centers of adjacent bubbles (mm)
A	150	7.665	1.512	4.237	35.02	6.50
B	150	43.621	1.650	18.350	64.38	2.85

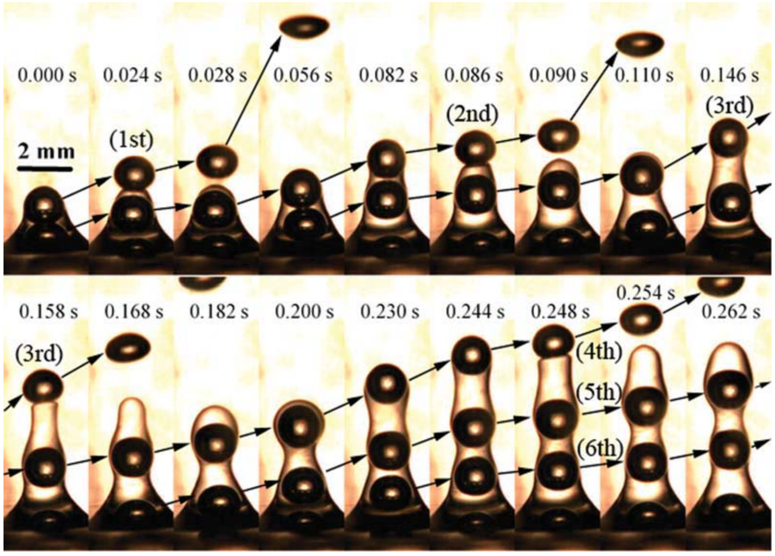

Figure 5. Sequential images showing sunflower oil column extended into the methanol upper phase by bubble train B.

[Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

entrained oil droplet and the bubble in the successful detaching part form a fully engulfed compound drop and remain that configuration for a very short time, see Figure 7a at $t = 0.190–0.234$ s, and $t = 0.326–0.346$ s, or in Figure 7b at $t = 0.052–0.064$ s. Finally, the bubble splits from the oil and rise upward, leaving the oil droplet to fall downward. Besides the results from the static equilibrium prediction (Duangsuwan et al.[7]), the phenomenon shown in Figure 7 confirms the impossibility of the dynamic formation and stabilization under gravitational field of the fully engulfing configuration gas-bubble/pure sunflower oil compound drop rising in pure methanol.

Conclusions

We have used a high-speed photography technique to observe the passage of single N_2-bubble trains through the sunflower oil/methanol interface in order to produce the fully engulfed N_2 bubble/sunflower oil compound drop rising in methanol. Two main results are obtained depending on bubble size and bubble injection frequency: (1) bubble train penetration with no entrainment, and (2) bubble train penetration with the entrained sunflower oil droplets. The phenomenon of the bubble and the entrained sunflower oil droplet separating from each other in methanol clearly confirms that the stabilization of the thin sunflower oil film around a bubble rising under a gravitational field in methanol is difficult. Further numerical simulations of the flow fields of the continuous fluid around rising compound drops could illuminate the details of the transients involving the interactions between the gas/engulfing film and engulfing film/continuous liquid interfaces. These may in turn allow possible optimization of the system and material properties to provide the maximum stability for prolonged periods. However, these early results already provide a reproducible mechanism of transport and dispersion of vegetable oil droplets in a short chain alcohol.

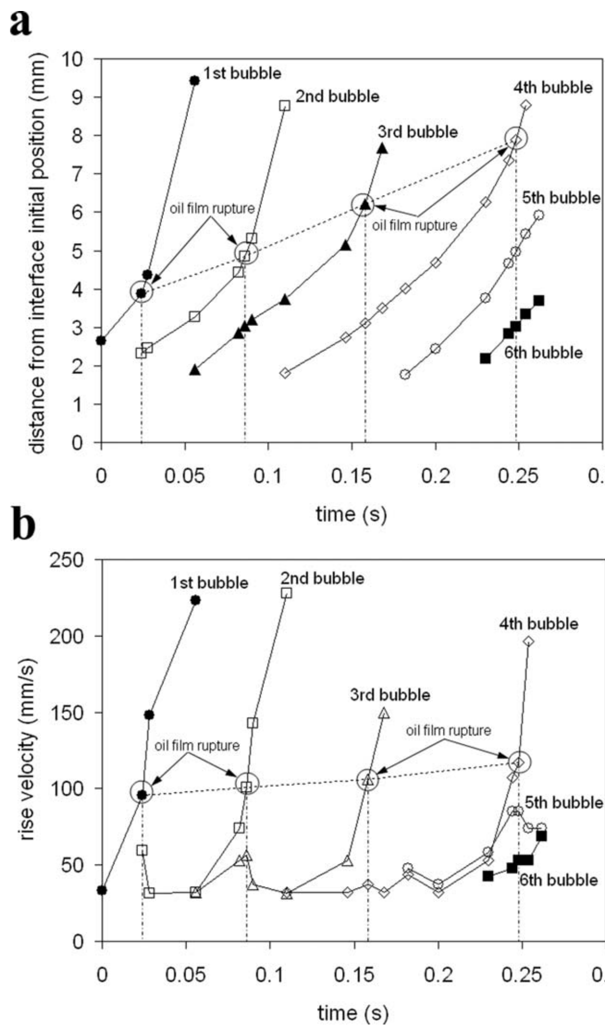

Figure 6. Motions of bubble train B based on Figure 5.

(a) Distances from interface initial position, and (b) rise velocities of gas bubbles.

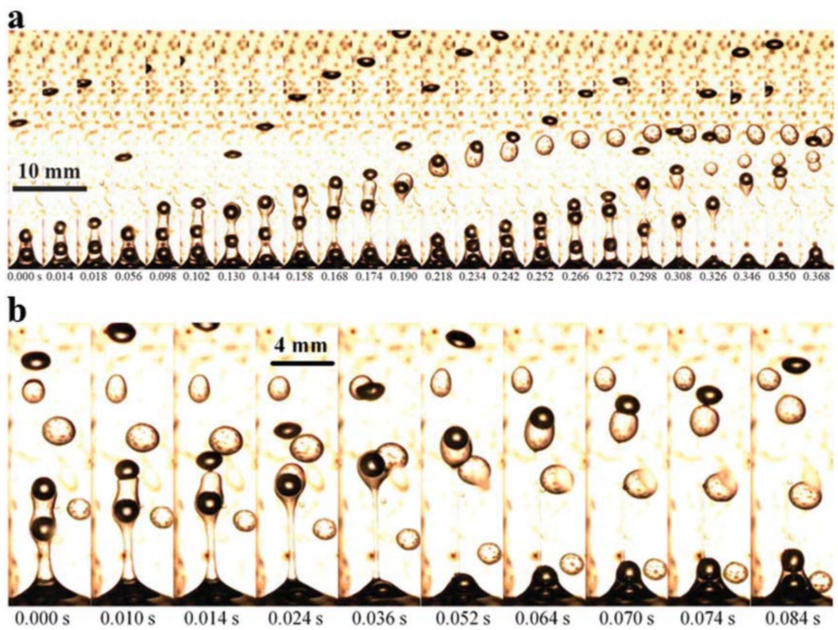

Figure 7. Sequential images showing entrainment of sunflower oil droplets into the methanol upper phase by bubble train B.

[Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

Acknowledgments

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