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Coalescence of Droplets Rising in a Quiescent Fluid Confined in a Vertical Cylindrical Tube in Stokes Regime

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Abstract – The coalescence of droplets rising in a quiescent fluid confined in a vertical cylindrical tube is experimentally examined. The viscosity ratio of the droplet to the surrounding fluid is unity, keeping the undeformed diameter of the leading droplet constant while varying the kinematic viscosities of the droplet and surrounding fluid, as well as the diameter of the following droplet. The coalescence times of the two droplets and the diameter of the clearance area between them are measured immediately before coalescence. The experimentally measured coalescence times are compared with the coalescence times of droplets predicted using semi-theoretical formula. The forces acting on the thin film between the droplets are discussed.

***Keywords*:** Coalescence, Droplet, Creeping motion, Vertical tube, Quiescent fluid

1. Introduction

The coalescence of droplets rising in a quiescent fluid confined in a vertical cylindrical tube is potentially useful for different purposes including the handling of fluids, control of chemical reaction and so on. The phenomenon is also the basis for analyzing the flow of multiphase fluids through porous media such as in enhanced oil recovery (e.g., [1]–[4]) and the breaking of emulsions in porous coalescers. We focus on a narrow passage in porous media, where if we assume the passage as a cylindrical tube, then the coalescence of droplets in viscous fluid through the passage results in the same through a cylindrical tube. There are few cases that investigated the coalescence of droplets in a tube, such as by Olbricht W.L.'s group [5], [6]. Aul and Olbricht proposed semi-theoretical formula of the coalescence time of droplets in creeping flow through a cylindrical tube [6]. The coalescence time is defined as the period between the instant when the relative velocity of the two droplets becomes zero after their apparent contact, and when coalescence occurs. Based on Aul and Olbricht's semi-theoretical formula, Muraoka et al. [7] proposed other semi-theoretical formulas for the coalescence time in terms of the resistance experienced by a liquid droplet in a viscous flow through a cylindrical tube in the Stokes regime [8].

In this study, the coalescence times of two droplets as well as the diameter of the clearance area between them immediately before coalescence are measured. The experimentally measured coalescence times are compared with values predicted using semi-theoretical formula. The forces acting on the thin film between the droplets are discussed.

2. Experiment

Figure 1 shows a schematic illustration of the experimental setup. A glass tube with an inner diameter of 3.5 mm, an outer diameter of 8.0 mm, and a length of 1500 mm was used as the test tube. The test tube was filled with a quiescent fluid which is a mixture of glycerol and pure water. The test tube was immersed in temperature-controlled water in a tank to maintain a constant system temperature. Silicone oil with a kinematic viscosities of 30cSt and 50cSt were employed as the test fluid for the droplets. The viscosity of the droplets was equal to that of the surrounding fluid. Two droplets were injected into the test tube using micro-syringe set on a syringe pump placed in front of the tube inlet. The behavior of the droplets was monitored using three digital cameras placed on the sliding stage. The motion of the stage was electrically controlled to monitor the movement of the droplets through the test tube. The dimensionless undeformed diameter d_1/D of the leading droplet was fixed at 0.76, and the dimensionless undeformed diameter d_2/D of the following droplet was varied, where d_1 is the undeformed diameter of the leading droplet, d_2 is the undeformed diameter of the following droplet, and D is the inner diameter of the test tube. The velocity of each leading and following droplet, the deviation from the tube central axis of each droplet and coalescence time were measured. The coalescence times of the two droplets and the diameter of the clearance area between them were measured immediately before coalescence.

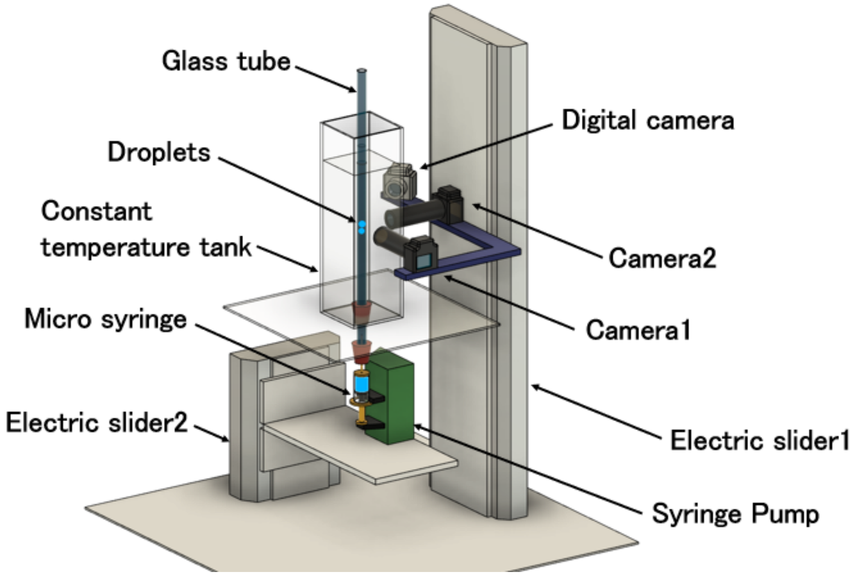

Fig. 1: Experimental setup.

3. Semi-theoretical formula of coalescence time of droplets rising in a quiescent fluid confined in a vertical tube in Stokes regime

Based on the semi-theoretical formula of Aul and Olbricht [6], other semi-theoretical formulas of the coalescence time of droplets have been proposed [7]. As shown in Figure 2, the clearance area between the leading droplet and the following droplet is assumed to be flat and discoid. Eq. (1) was derived by Reynolds [9] under the assumption of two parallel-plane surfaces approaching each other.

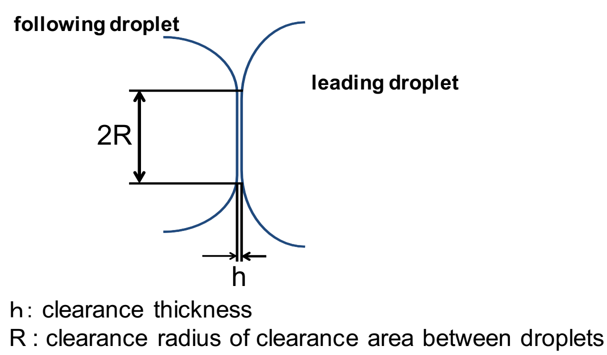

Fig. 2: Radius of clearance area between droplets, and clearance thickness.

$$\frac{dh}{dt} = -\frac{2Fh^3}{3\pi\mu_s R^4} \tag{1}$$

$$F = F_h + \pi R^2 \frac{A_1}{6\pi h^3}, \tag{2}$$

where F is the total force compressing the clearance area between the droplets, and A_1 is the Hamaker constant. The total force F is expressed as the sum of the hydrodynamic force F_h and van der Waals force between the droplets (Eq. (2)). In this case, the hydrodynamic force decelerates the following droplet until the relative velocity of the two droplets becomes zero after their apparent contact. The hydrodynamic force can be expressed as F_1 - F_2, where F_1 is the hydrodynamic force exerting on the following droplet when the velocity of the following droplet equals that of a single droplet, and F_2 is the hydrodynamic force exerting on the following droplet when the relative velocity of the two droplets becomes zero after their apparent contact. Because the acceleration of the droplet was low, the virtual mass [10] was extremely small compared with the hydrodynamic force and thus not considered. The present experiments confirmed that the leading droplets stabilized before and after their apparent contact. Aul and Olbricht investigated the coalescence of droplets using a glass tube (inner diameter, 54 μm; length, 25 mm) and proposed a semi-theoretical formula for the coalescence time of droplets in a creeping flow through a tube. They defined the hydrodynamic force as the force exerting on a single rigid sphere after experimentally confirming that the velocity of the droplet was similar to that of the rigid sphere. In this study, F_1 and F_2 were determined using the numerical procedure developed by Higdon and Muldowney [8], who expressed the hydrodynamic force exerting on a single droplet in a creeping flow through a tube as

$$F_0 = \eta\mu_s(\frac{d}{2})K_z U_z + \eta\mu_s(\frac{d}{2})dK_p U_0, \tag{3}$$

where

$$\eta = \frac{4\pi(1+\frac{3}{2}\beta)}{(1+\beta)} \tag{4}$$

Here, F_0 is the hydrodynamic force exerting on a single droplet in a creeping flow through a tube, μ_s the viscosity of the surrounding fluid, d the undeformed diameter of a single droplet, K_z and K_p the resistance coefficients, U_z the velocity of a single droplet, U_0 the maximum velocity of the parabolic pressure-driven flow, β the viscosity ratio of the droplet to that of the surrounding fluid, and K_z and K_p include the center-to-center distances between the droplets and tube axis. The form of Eq.(3) is based on the similar idea as that of Haberman and Sayre [11]. Haberman W.L. and Sayre R.M. replaced the resistance acting on a sphere moving with velocity U in a creeping flow with maximum velocity V through a cylindrical tube, with the resistance acting on a sphere fixed in a flow with maximum velocity of V-U at the tube axis, where the tube wall moves at velocity U in the opposite direction of the flow. They expressed the resistance acting on the sphere in this case with the following equation.

$$Drag = 6\pi\mu a\left(UK_1 - VK_2\right) = 6\pi\mu aUK_1 - 6\pi\mu aVK_2 \tag{5}$$

Here, K_1 and K_2 are wall correction coefficients. Therefore, the first term represents the resistance acting on a sphere moving at a velocity U in quiescent fluid within a cylindrical tube, while the second term represents the resistance acting on a sphere fixed in creeping flow with maximum velocity V inside the cylindrical tube.

Substituting Eq. (3) for F_1 and F_2, the hydrodynamic force F_h can be expressed as shown in Eq. (6).

$$F_h = F_1 - F_2 = \left\{\eta\mu_s(\frac{d_2}{2})K_z U_{z1} + \eta\mu_s(\frac{d_2}{2})K_p U_0\right\} - \left\{\eta\mu_s(\frac{d_2}{2})K_z U_{z2} + \eta\mu_s(\frac{d_2}{2})K_p U_0\right\}$$
$$= \eta\mu_s(\frac{d_2}{2})K_z(U_{z1} - U_{z2}) \tag{6}$$

where U_{z1} is the velocity of a single droplet with the diameter of the following droplet, and U_{z2} is the velocity of the following droplet when the relative velocity of the two droplets becomes zero after their apparent contact. Eq.(6) is an equation for the case where both the leading droplet and the following droplet move on the tube axis. That is, the density ratio between the droplets and the surrounding fluid is unity. With regard to the motions of the two droplets during the creeping flow through the cylindrical tube, as the following droplet became smaller in size, the effect of the secondary flow produced by the presence of the leading droplet caused it to move to an eccentric position. As the following droplet continued to decrease in size, it was more easily affected by the secondary flow. If the following droplet is located at an eccentric position, then F_h can be expressed using Eq. (7).

$$F_h = F_1 - F_2 = \underbrace{\left\{\eta\mu_s(\frac{d_2}{2})K_z U_{z1} + \eta\mu_s(\frac{d_2}{2})K_p U_0\right\}}_{\text{without eccentricity}} - \underbrace{\left\{\eta\mu_s(\frac{d_2}{2})K_z U_{z2} + \eta\mu_s(\frac{d_2}{2})K_p U_0\right\}}_{\text{with eccentricity}} \tag{7}$$

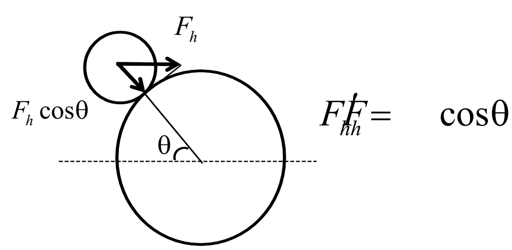

Fig. 3: Illustration of F_h'.

As shown in Figure 3, the hydrodynamic force in this case can be denoted by F_h', which is equal to $F_h\cos\theta$. Here, θ is the angle between the tube axis and the line joining the centers of the leading and following droplets. The coalescence time, T, can be calculated by integrating the numerator and denominator on the left-hand side of Eq. (1) using the method employed by Aul and Olbricht. Without considering the details of the integration process, the coalescence time T can be expressed using Eqs. (8) and (9). For simplicity, C is assumed to be constant in Eqs. (8) and (9), and its value can be determined experimentally.

$$T = C\frac{R^{\frac{8}{3}}}{F_h^{\frac{1}{3}}} \qquad \text{(without eccentricity)} \tag{8}$$

$$T = C\frac{R^{\frac{8}{3}}}{F_h'^{\frac{1}{3}}} \quad \text{(with eccentricity)} \tag{9}$$

When two droplets rise in a quiescent fluid confined in a vertical cylindrical tube, in Eqs. (3) and (5), the second term disappears, leaving only the first term. Eq. (3) is replaced by Eq.(10).

$$F_0 = \eta\mu_s(\frac{d}{2})K_z U_z \tag{10}$$

Substituting Eq. (10) for F_1 and F_2, the hydrodynamic force F_h can be expressed as shown in Eq. (11). The Eq.(11) takes into account the deviation of the leading and following droplets from the tube central axis.

$$F_h = F_1 - F_2 = \eta\mu_s(\frac{d_2}{2})K_z(U_{z1} - U_{z2}) \tag{11}$$

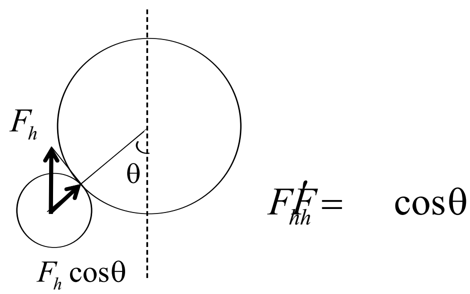

Fig. 4: Illustration of F_h'.

As shown in Figure 4, the hydrodynamic force in this case can be denoted by F_h', which is equal to $F_h\cos\theta$. Here, θ is the angle between the vertical straight line and the line joining the centers of the leading and following droplets. The coalescence time T can be expressed by Eq. (12) in a similar manner to Eqs. (8) and (9).

$$T = C\frac{R^{\frac{8}{3}}}{F_h'^{\frac{1}{3}}} \tag{12}$$

4. Results and Discussion

The motion of droplets rising in a quiescent fluid confined in a vertical cylindrical tube in Stokes regime could be classified into three types: type 1 where both leading and following droplets rise almost straight, type 2 where the leading droplet rises almost straight while the following droplet undergoes spiral motion, and type 3 where both leading and following droplets undergo spiral motion. Approximately 65% of the motion of droplets were the type 1. Approximately 25% of that were the type 2. Approximately 10% of that were type 3. Figures 5–6 show the dimensionless coalescence times and dimensionless clearance diameter between the droplets as functions of the dimensionless undeformed diameter of the following droplet for different values of kinematic viscosity of the droplets. T is the coalescence time, and R is the radius of clearance area between the droplets. The red lines represent the experimentally measured coalescence times. The blue lines represent the semi-theoretical formula for the coalescence time, whereas the yellow lines represent the dimensionless clearance diameters. The dimensionless coalescence time indicates how many tube diameters the two droplets travel during the coalescence time. The experimentally measured coalescence times of the droplets agreed well with the values predicted using the semi-theoretical formula. The trend for the clearance diameter was the same. Meanwhile, the effect of the clearance radius was greater than that of the hydrodynamic force, since the power of the clearance radius was much higher than that of the hydrodynamic force. (See Eq. (12)).

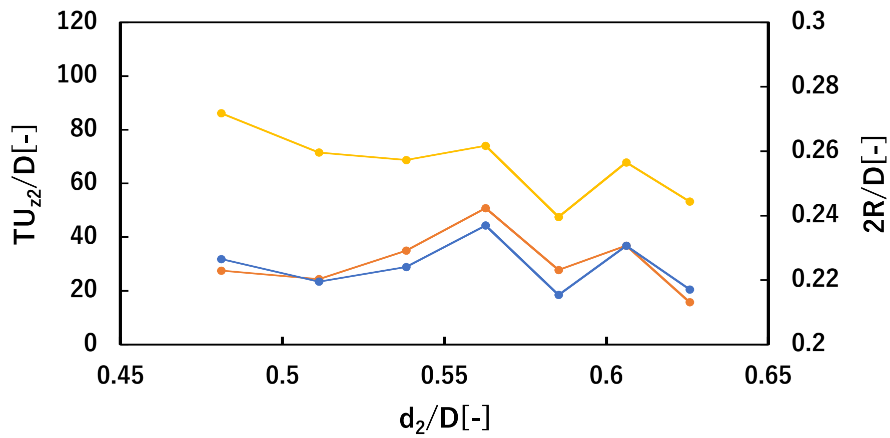

Fig. 5: Dimensionless coalescence time and dimensionless clearance diameter as a function of the dimensionless undeformed diameter of the following droplet for the droplets with a kinematic viscosity of 30cSt.

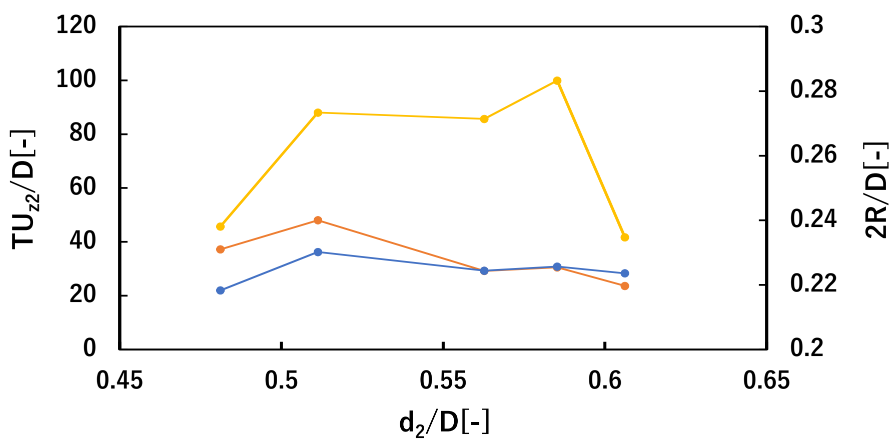

Fig.6: Dimensionless coalescence time and dimensionless clearance diameter as a function of the dimensionless undeformed diameter of the following droplet for the droplets with a kinematic viscosity of 50cSt.

Figures 7 and 8 show the forces acting on the clearance area as a function of the clearance thickness for different kinematic viscosity of the droplets. The figures are shown in log-log format. Specifically, the figures show the cases for $d_1/D = 0.76$ and $d_2/D = 0.\ 48$, respectively. The red lines represent the total force acting on the clearance area between the droplets, the blue lines represent the hydrodynamic force, the yellow lines represent the van der Waals force between the droplets (See Eqs. (2), (11)). For Eq. (2), we assume that Hamaker constant is 10^{-20} J. The hydrodynamic force F_h is constant regardless of the clearance thickness h. However, the van der Waals forces between the droplets increases as the clearance thickness decreases. When the clearance thickness was larger than about 30 nm, the hydrodynamic force was dominant; when it was smaller than 30 nm, the van der Waals force between the droplets was larger than the hydrodynamic force. A comparison of the abovementioned figures shows that the kinematic viscosity of the droplets changed slightly.

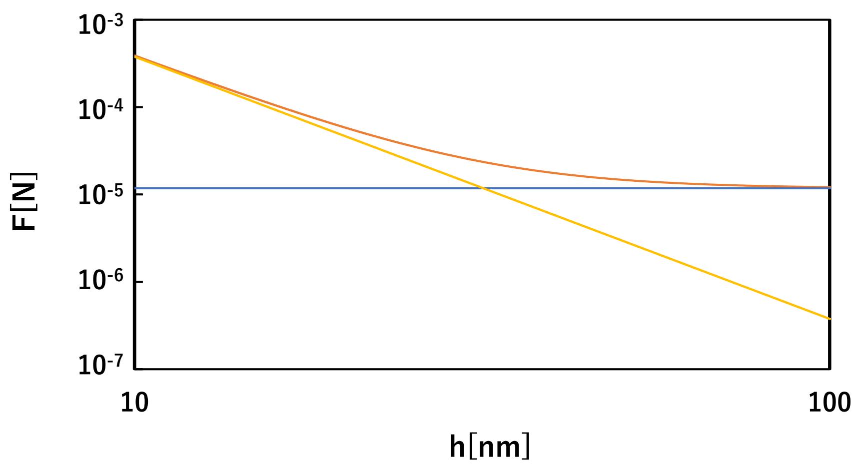

Fig. 7: Forces acting on clearance area as a function of clearance thickness for the droplets with a kinematic viscosity of 30cSt.

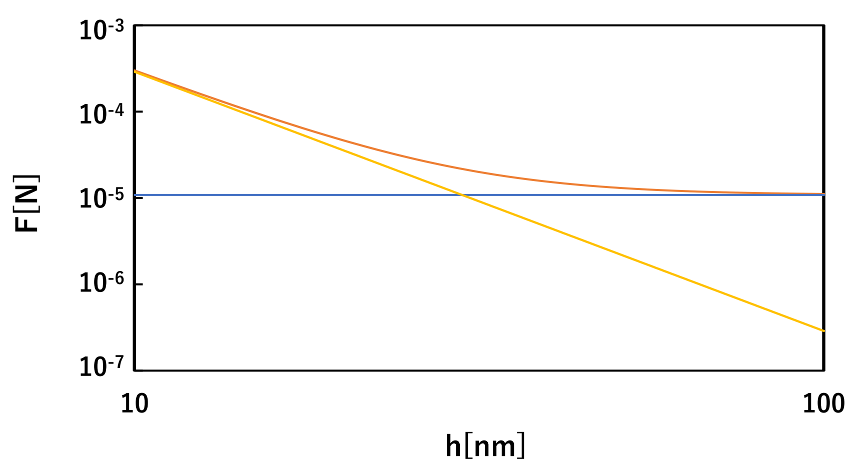

Fig. 8: Forces acting on clearance area as a function of clearance thickness for the droplets with a kinematic viscosity of 50 cSt.

5. Conclusion

The coalescence of droplets rising in a quiescent fluid confined in a vertical cylindrical tube in Stokes regime was examined in this study. The motion of droplets could be classified into three types: type 1 where both leading and following droplets rise almost straight, type 2 where the leading droplet rises almost straight while the following droplet undergoes spiral motion, and type 3 where both leading and following droplets undergo spiral motion. Approximately 65% of the motion of droplets were the type 1. In case of type 1 where both leading and following droplets rise almost straight, semi-theoretical formula of coalescence time of droplets was obtained. The experimentally measured coalescence times of droplets agreed well with the values predicted using the semi-theoretical formula. When the clearance thickness was larger than about 30 nm, the hydrodynamic force was dominant; when the clearance thickness was smaller than about 30 nm, the van der Waals force between the droplets was larger than the hydrodynamic force.

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