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Film Drainage and Coalescence of Droplets Containing Particles in Creeping Flow Through Cylindrical Tube

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Abstract **-** The coalescence of droplets containing particles in a creeping flow through a cylindrical tube is experimentally examined. In particular, the coalescence times of the two droplets containing particles and the diameter of the clearance area between them are measured immediately before coalescence. The effect of those droplets on the coalescence time is discussed. The Reynolds number of the multiphase flow through the test tube is varied to investigate its effect on the film drainage and coalescence of the droplets containing particles. The experimentally measured coalescence times are compared with the coalescence times of droplets without particles and with values predicted using semi-theoretical formulas. The forces exerted between the droplets on a thin film are discussed.

Keywords**:** Coalescence, Droplet, Suspended Particle, Tube, Creeping Flow

1. Introduction

The coalescence of droplets in a viscous flow through a cylindrical tube is 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 flow through the passage results in the same through a cylindrical tube. Muraoka et al. [5] discovered that the coalescence times of droplets containing particles were significantly shorter than those of droplets without particles in a viscous flow through a cylindrical tube. 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 [6], Muraoka et al. [5] 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 [7]. In this study, the coalescence times of two droplets containing particles measuring 30 μm in diameter as well as the diameter of the clearance area between them immediately before coalescence are measured. The Reynolds number (Re) of the multiphase flow through the tube is varied to investigate its effects on the film drainage and coalescence of droplets containing particles. Cases involving droplets containing particles are investigated, and the results are compared with those involving droplets without particles [8]. The experimentally measured coalescence times are compared with values predicted using semi-theoretical formulas. The forces exerted between the droplets on a thin film are discussed.

2. Experiment

Figure 1 shows a schematic illustration of the experimental setup. A glass tube with an inner diameter of 2.0 mm, an outer diameter of 7.0 mm, and a length of 1500 mm was used as the test tube. Silicone oil with a kinematic viscosity of 3000 cSt was employed as the test fluid for the droplets. The droplets used in the experiments contained Au-coated acrylic particles. The employed particles were 30 μm in diameter and 1490 kg/m^3 in density, and the volumetric concentration of particles in the droplets was 0.24%. A mixture of glycerol and pure water was used as the surrounding fluid for creeping flow through the test tube. After the addition of carbon tetrachloride, the density of the droplets was equal to that of the surrounding fluid. In the present experiments, the densities of the droplet and the surrounding fluid were both 1.24×10^3 kg/m^3, the viscosity of the surrounding fluid was 0.24 Pa·s, and the viscosity ratio of the droplet to the surrounding fluid was approximately unity. A large volumetric syringe pump was used to maintain a steady flow through the test tube at a designated average velocity. The test tube was immersed in temperature-controlled water in a tank to maintain a constant system temperature.

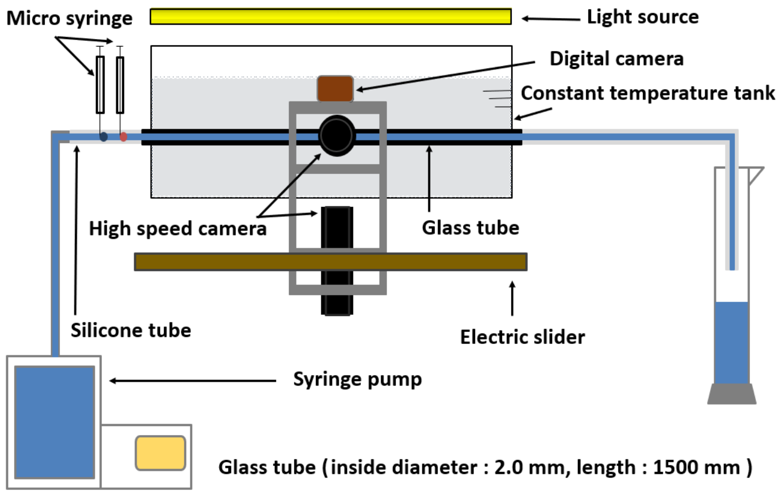


Fig. 1: Experimental setup.

The two droplets containing particles were injected into the test tube using microsyringes placed in front of the tube inlet. The dimensionless undeformed diameter d_1/D of the leading droplet was fixed at 0.9, 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 behavior of the droplets was monitored using a digital camera and two high-speed cameras placed on the sliding stage. The motion of the stage on which the cameras were mounted was electrically controlled to monitor the movement of the droplets through the test tube. The coalescence times of the two droplets containing particles and the diameter of the clearance area between them were measured immediately before coalescence. The Re of the multiphase flow through the test tube was varied to investigate its effect on droplet coalescence.

3. Semi-theoretical formulas of coalescence time of droplets

Based on the semi-theoretical formula of Aul and Olbricht[6], other semi-theoreticla formulas of the coalescence time of droplets have been proposed [5]. As shown in Figure 2, the clearance area between the leading droplet and the subsequent 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.

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

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

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

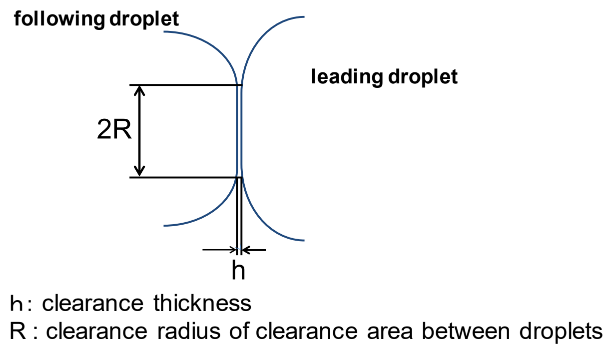


Fig. 2: Radius of clearance area between droplets, and clearance thickness.

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 [7], who expressed the hydrodynamic force exerting on a single droplet in a creeping flow through a tube as

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

where

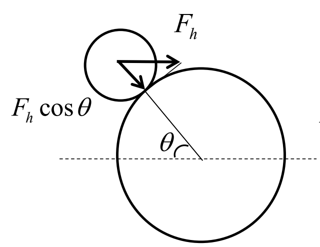
$$\eta = \frac{4\pi(1+\frac{3}{2}\beta)}{(1+\beta)} \quad . \tag{4}$$

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. Substituting Eq. (3) for F_1 and F_2, the hydrodynamic force F_h can be expressed as shown in Eq. (5).

$$F_h = F_1 - F_2 = \left\{ \eta\mu_s(\frac{d_2}{2})K_z U_{z1} + \eta\mu_s(\frac{d_2}{2})K_p U_0 \right\} - \left\{ \eta\mu_s(\frac{d_2}{2})K_z U_{z2} + \eta\mu_s(\frac{d_2}{2})K_p U_0 \right\}$$
$$= \eta\mu_s(\frac{d_2}{2})K_z(U_{z1} - U_{z2}) \tag{5}$$

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. If the following droplet is located at an eccentric position, then F_h can be expressed using Eq. (6).

$$F_h = F_1 - F_2 = \underbrace{\left\{ \eta\mu_s(\frac{d_2}{2})K_z U_{z1} + \eta\mu_s(\frac{d_2}{2})K_p U_0 \right\}}_{\text{without eccentricity}} - \underbrace{\left\{ \eta\mu_s(\frac{d_2}{2})K_z U_{z2} + \eta\mu_s(\frac{d_2}{2})K_p U_0 \right\}}_{\text{with eccentricity}} \tag{6}$$



$$F_h' = F_h \cos\theta$$

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. (7) and (8). For simplicity, C is assumed to be constant in Eqs. (7) and (8), and its value can be determined experimentally.

$$T = C\frac{R^{\frac{8}{3}}}{F_h^{\frac{1}{3}}} \qquad \text{(without eccentricity)} \tag{7}$$

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

The coalescence times of the droplets containing particles were shorter than those of droplets without particles. In terms of the coalescence of the droplets containing particles, the total force F can be expressed as the sum of the hydrodynamic force and van der Waals forces between them, between the particles, and between the droplets and particles (Eq. (9)).

$$F = F_h + \pi R^2 \frac{A_1}{6\pi h^3} + \frac{naA_2}{12h^2} + \frac{naA_3}{3h^2} \quad , \tag{9}$$

where A_2 and A_3 are the Hamaker constants, a is the radius of the particle, and n is the number of particles near the clearance area.

In addition to formulas for describing droplets without particles, semi-theoretical formulas for the coalescence time of droplets containing particles can be obtained by integrating Eq. (1), and the results are as shown in Eq.(10) and (11). Comparing the coalescence times of the droplets without particles (Eqs. (7) and (8)) with those of the droplets containing particles (Eqs. (10) and (11)), only the power of the clearance radius between the droplets containing particles differs from that of the droplets without particles.

$$T = C \frac{R^4}{F_h^{\frac{1}{3}}} \qquad \text{(without eccentricity)} \tag{10}$$

$$T = C \frac{R^4}{F_h'^{\frac{1}{3}}} \qquad \text{(with eccentricity)} \tag{11}$$

4. Results and Discussion

Figures 4–6 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 Re. T is the coalescence time, V is the average velocity of the multiphase flow through the test tube, and R is the radius of clearance area between the droplets. The red lines represent the experimentally measured coalescence times, and the yellow lines represent the experimentally measured coalescence times for the droplets without particles. Each plotted experimental value is the average obtained from 10 to 15 experiments. The blue lines represent the semi-theoretical formulas for the coalescence time, whereas the green lines represent the dimensionless clearance diameters. The coalescence times of the droplets containing particles were significantly shorter than those of the droplets without particles. In the case where the following droplet was small, the coalescence times of the droplets without particles increased with Re. In the case where the following droplet was small, the following droplet deviated from the tube axis and rotated under the effect of the secondary flow caused by the leading droplet. This is because as Re increases, the rotation speed of the following droplet increases and the surrounding fluid enters the clearance area between the droplets. If the following droplet is large, then the droplet will not deviate from the tube axis and thus will not rotate. By contrast, the coalescence times of the droplets containing particles do not increase significantly with Re if the following droplet is similarly small, which is due to the inclusion effect of the particles. The experimentally measured coalescence times of the droplets containing particles agreed roughly with the values predicted using the semi-theoretical formulas. 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 Eqs. (7), (8), (10), and (11)).

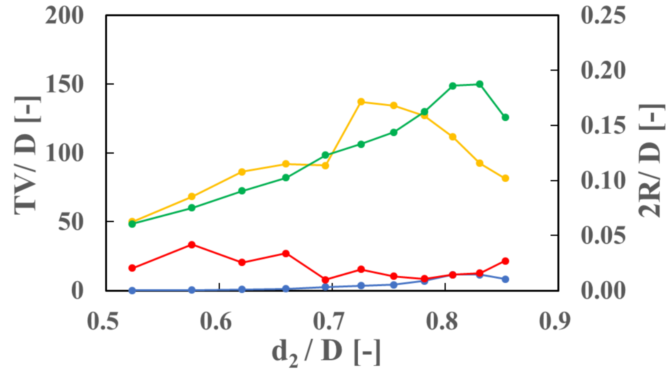


Fig. 4: Dimensionless coalescence time and dimensionless clearance diameter as a function of the dimensionless undeformed diameter of the following droplet for Re = 0.01.

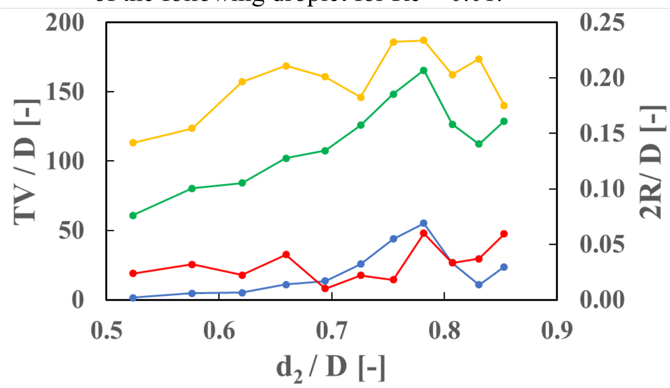


Fig. 5: Dimensionless coalescence time and dimensionless clearance diameter as a function of the dimensionless undeformed diameter of the following droplet for Re = 0.02.

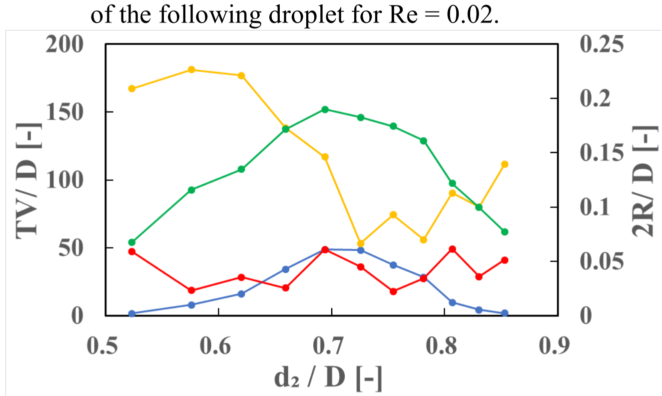


Fig. 6: Dimensionless coalescence time and dimensionless clearance diameter as a function of the dimensionless undeformed diameter of the following droplet for Re = 0.08.

Figures 7 and 8 show the forces exerting on the clearance area as a function of the clearance thickness for different Re. Specifically, the figures show the cases for d_1/D = 0.9 and d_2/D = 0.52, respectively. The red lines represent the total force exerting on the clearance area between the droplets, the yellow lines represent the hydrodynamic force, the blue lines represent the van der Waals force between the droplets, the orange lines represent the van der Waals force between the particles, and the green lines represent the van der Waals forces between the droplets and particles. (See Eqs. (5), (6), and (9)). For Eq. (9), we assume that each 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, the particles and droplets, and the particles increase as the clearance thickness decreases. The van der Waals forces between the particles and droplets, and the particles were extremely small compared with the van der Waals forces between the droplets. When the clearance thickness was larger than 1 nm, the hydrodynamic force was dominant; when it was smaller than 1 nm, the van der Waals force between the droplets was larger the hydrodynamic force. The shorter coalescence time of the droplets containing particles is attributable to the effect of particles near the interface on the shape of the interface. A comparison of the abovementioned figures shows that the Re changed slightly.

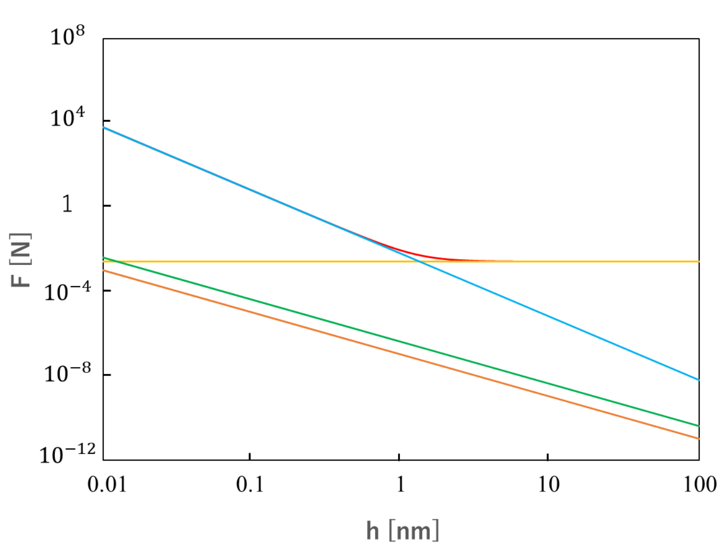


Fig. 7: Forces exerting on clearance area as a function of clearance thickness for Re = 0.01.

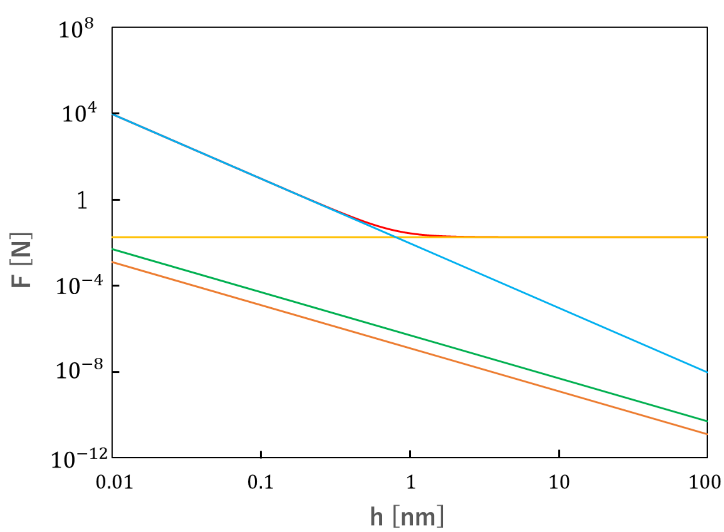


Fig. 8: Forces exerting on clearance area as a function of clearance thickness for Re = 0.08.

5. Conclusion

The coalescence of droplets containing particles in a creeping flow through a cylindrical tube was examined in this study. The coalescence times of droplets containing particles were shorter than those of droplets without particles. The experimentally measured coalescence times of droplets containing particles agreed roughly with the values predicted using the semi-theoretical formulas. The van der Waals forces between the particles and droplets, and the particles were extremely small compared with the van der Waals force between the droplets. When the clearance thickness was larger than 1 nm, the hydrodynamic force was dominant; when the clearance thickness was smaller than 1 nm, the van der Waals force between the droplets was larger than the hydrodynamic force.

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