Terminal Velocity of Single Drops in Stagnant Liquids*

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Abstract
Terminal velocities of single drops rising through infinite stagnant liquids under wide ranges of fluid properties were measured to examine the effects of initial shape deformation, surfactants and the viscosity ratio. As a result, the following conclusions were obtained: (1) the terminal velocity $V_T$ of a drop is not affected by initial shape deformation due to strong viscous damping of shape oscillation, (2) the drop drag coefficient $C_D$ is a function of the drop Reynolds number $Re$, the viscosity ratio $\kappa$ and surfactant concentration, (3) surfactants increase $C_D$ and the influence of surfactants on $C_D$ becomes weaker as $\kappa$ increases, which is consistent with the Levich’s drag model, (4) the effect of $\kappa$ on $C_D$ can be evaluated by the factor in the Levich’s model, $[(2 + 3\kappa)/(1 + \kappa)]$, even at intermediate drop $Re$ numbers, and (5) the combination of the Levich’s model and Schiller & Nauman’s correlation gives good estimation of $C_D$ of single drops in clean and fully contaminated systems under the conditions of $-11.6 < \log_{10} M < -0.9$, $1.7 \times 10^{-1} < Re < 2 \times 10^{2}$, $1.7 \times 10^{-2} < Eo < 12.1$ and $0.1 < \kappa < 100$.

Key words: Drop, Terminal Velocity, Viscosity Ratio, Drag Coefficient, Surfactant

1. Introduction
Sufficient knowledge on terminal velocities $V_T$ of single fluid particles rising or falling through infinite stagnant liquids is required for modeling and simulation of dispersed two-phase flows in various industrial fields. Many researches(1)−(11) on $V_T$ of single fluid particles or solid particles in stagnant liquids have been conducted and reliable models and correlations for $V_T$ or the drag coefficient $C_D$ are available. Grace et al.(1) proposed a well-known graphical correlation among the Eötvös number $Eo$, Reynolds number $Re$ and Morton number $M$ for single fluid particles in infinite stagnant liquids. Here, $Eo$ is the ratio of buoyant force $F_b$ to interfacial tension force $F_s$, $Re$ is the ratio of inertial force $F_i$ to viscous force $F_\mu$, and $M$ represents the fluid properties of two fluids. They are defined by

$$Eo = \frac{F_b}{F_s} = \frac{(\rho_c - \rho_d)gd^2}{\sigma}$$

$$Re = \frac{F_i}{F_\mu} = \frac{\rho_c V_T d}{\mu_c}$$

$$M = \frac{\rho_c^2 F_b}{\rho_d^2 F_s} = \frac{g \mu_c^2 (\rho_c - \rho_d)}{\rho_d^2 \sigma^3}$$

where $d$ is the sphere-volume equivalent diameter of the fluid particle, $g$ the acceleration due to gravity, $\rho$ the density, $\mu$ the viscosity and $\sigma$ the interfacial tension. The subscripts $c$ and $d$ denote the continuous and dispersed phases, respectively. Bond and Newton(2) pointed out that small fluid particles with high viscosity behave as solid particles and the drag forces acting...
on them follow the Stokes’s law\(^{(3)}\). Hadamard\(^{(4)}\) and Rybczynski\(^{(5)}\) deduced a theoretical \(C_D\) model for single spherical fluid particles at very low Reynolds numbers as a function of the viscosity ratio \(\kappa (= \mu_d/\mu_c)\):

\[
C_D = \frac{8}{Re} \left(\frac{2 + 3\kappa}{1 + \kappa}\right)
\]

(4)

For bubbles, the viscosity ratio is close to zero (\(\kappa \approx 0\)). For rigid particles, it becomes infinity (\(\kappa \approx \infty\)) and Eq. (4) reduces to the Stokes’s drag model\(^{(3)}\). Levič\(^{(6)}\) proposed a \(C_D\) model which takes into account the effects of not only \(\kappa\) but also surfactants:

\[
C_D = \frac{8}{Re} \left[\frac{2 + 3\kappa + 3C}{1 + \kappa + C/\mu}\right]
\]

(5)

where \(C\) is a coefficient expressing the retardation of interface motion by surfactants. This equation is applicable only to spherical fluid particles at extremely low Reynolds numbers (\(Re < 1\)).

As for bubbles at medium or high Reynolds numbers, Tomiyama et al.\(^{(10)}\) proposed \(C_D\) correlations for single bubbles rising through clean, slightly contaminated and fully contaminated liquids:

(a) For a clean system

\[
C_D = \max \left\{ \min \left\{ \frac{16}{Re} \left(1 + 0.15Re^{0.687}\right), \frac{48}{Re} \left[\frac{8}{3} \frac{Eo}{Eo + 4}\right]\right\} \right\}
\]

(6)

(b) For a slightly contaminated system

\[
C_D = \max \left\{ \min \left\{ \frac{24}{Re} \left(1 + 0.15Re^{0.687}\right), \frac{72}{Re} \left[\frac{8}{3} \frac{Eo}{Eo + 4}\right]\right\} \right\}
\]

(7)

(c) For a fully contaminated system

\[
C_D = \max \left\{ \min \left\{ \frac{24}{Re} \left(1 + 0.15Re^{0.687}\right), \frac{8}{3} \frac{Eo}{Eo + 4}\right\} \right\}
\]

(8)

They also pointed out that \(V_T\) of a bubble depends on the bubble shape, which is strongly affected by initial shape deformation\(^{(11)}\). However, there are few studies on \(V_T\) of a drop at intermediate and high Reynolds numbers.

In this study, terminal velocities of single drops rising through infinite stagnant liquids under a wide range of \(Re\) (0.17 < \(Re\) < 200) were measured to investigate the influences of surfactants and viscosity ratio \(\kappa\) on \(C_D\) and \(V_T\) of a drop. The effect of initial shape deformation on \(V_T\) was also discussed.

2. **Experimental**

Experiments were conducted at room temperature and atmospheric pressure. Figure 1 shows the schematic of experimental apparatus. The acrylic tank and the measurement system were the same as those used in our previous experiments on bubble terminal velocity\(^{(11)}\). The width, depth and height of the tank were 0.20, 0.20 and 0.69 m, which were large enough to avoid the wall effects\(^{(9)}\). Silicon oils (Sin-etsu Silicone, KF96-30 and KF96-100) and glycerol-water solutions were used for the dispersed and continuous phases, respectively. Experimental conditions are summarized in Table 1. The concentration of glycerol-water solution was changed to examine the effects of the viscosity ratio \(\kappa\) on \(C_D\) and \(V_T\) of a drop. The effect of initial shape deformation on \(V_T\) was also discussed.
Table 1  Experimental conditions

<table>
<thead>
<tr>
<th>Fluid (continuous phase)</th>
<th>Fluid (dispersed phase)</th>
<th>log_{10} M</th>
<th>κ</th>
<th>\rho_c (kg/m³)</th>
<th>\rho_d (kg/m³)</th>
<th>\mu_c (m.Pa.s)</th>
<th>\mu_d (m.Pa.s)</th>
<th>\sigma (N/m)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Water (clean)</td>
<td>KF-30</td>
<td>−11.6</td>
<td>31.5</td>
<td>998</td>
<td>960</td>
<td>1</td>
<td>31.5</td>
<td>0.0546</td>
</tr>
<tr>
<td>Water (clean)</td>
<td>KF-100</td>
<td>−11.6</td>
<td>100</td>
<td>998</td>
<td>967</td>
<td>1</td>
<td>100</td>
<td>0.0508</td>
</tr>
<tr>
<td>Water (contaminated)</td>
<td>KF-30</td>
<td>−10.6</td>
<td>31.5</td>
<td>998</td>
<td>960</td>
<td>1</td>
<td>31.5</td>
<td>0.0246</td>
</tr>
<tr>
<td>Glycerol-water solution (clean)</td>
<td>KF-30</td>
<td>−6.0</td>
<td>1.9</td>
<td>1178</td>
<td>960</td>
<td>17</td>
<td>31.5</td>
<td>0.05</td>
</tr>
<tr>
<td>Glycerol-water solution (clean)</td>
<td>KF-30</td>
<td>−4.8</td>
<td>0.9</td>
<td>1198</td>
<td>960</td>
<td>34</td>
<td>31.5</td>
<td>0.0496</td>
</tr>
<tr>
<td>Glycerol-water solution (contaminated)</td>
<td>KF-30</td>
<td>−4.1</td>
<td>0.9</td>
<td>1198</td>
<td>960</td>
<td>34</td>
<td>31.5</td>
<td>0.0288</td>
</tr>
<tr>
<td>Glycerol-water solution (clean)</td>
<td>KF-30</td>
<td>−3.0</td>
<td>0.4</td>
<td>1221</td>
<td>960</td>
<td>87</td>
<td>31.5</td>
<td>0.0467</td>
</tr>
<tr>
<td>Glycerol-water solution (clean)</td>
<td>KF-100</td>
<td>−3.0</td>
<td>1.1</td>
<td>1223</td>
<td>969</td>
<td>89</td>
<td>100</td>
<td>0.0466</td>
</tr>
<tr>
<td>Glycerol-water solution (contaminated)</td>
<td>KF-30</td>
<td>−3.0</td>
<td>0.4</td>
<td>1221</td>
<td>960</td>
<td>87</td>
<td>31.5</td>
<td>0.0456</td>
</tr>
<tr>
<td>Glycerol-water solution (clean)</td>
<td>KF-30</td>
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<td>0.1</td>
<td>1246</td>
<td>960</td>
<td>280</td>
<td>31.5</td>
<td>0.0455</td>
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</tbody>
</table>

(C₈H₁₇C₆H₄(OCH₂CH₂)₁₀OH) was used for surfactants to examine its effects on \( V_T \). The concentration of Triton X-100 was \( 2.68 \times 10^{-1} \) mol/m³, which was four times higher than the maximum concentration in the research by Zhang and Finch(12) \( (6.7 \times 10^{-2} \) mol/m³) and about a half of the critical micelle concentration \( (4.2 \times 10^{-1} \) mol/m³)(13). Therefore, the contaminated systems in the present experiments could be regarded as fully contaminated systems and the influence of micelle was negligible.

Since all single drops in the present experiments rose rectilinearly and their shapes were axisymmetric, measurements of \( V_T, d \) and the aspect ratio \( E \) (the ratio of minor to major axes) were carried out using a single high-speed video camera (Kodak Motion Corder Analyzer SR-1000, shutter speed = 1/5000 s, frame rate = 250 frame/s, 512 × 480 pixels). Images for measuring \( V_T \) were taken at 400 mm above the nozzle tip, at which all drops reached terminal conditions. The spatial resolution of images ranged from 0.02 to 0.18 mm/pixel. The maximum error in the \( V_T \) measurement was 0.5% and the uncertainty in measured \( d \) estimated at 95% confidence was 0.81%. To examine the influence of initial shape deformation, rise velocity \( V_Z \) and aspect ratio \( E \) of a drop were measured near the nozzle tip using the high-speed video camera (Fig. 2). The liquid densities, viscosities and interfacial tension \( \sigma \) were
measured by a densimeter (JIS B7525, accuracy: ±0.1%), a viscometer (Rion, Viscotester VT-03E, accuracy: ±3%) and capillary tubes (glass, inner diameter: 0.63, 1.02 and 1.40 mm), respectively. The measurements of $\rho_c$, $\rho_d$, $\mu_c$, $\mu_d$ and $\sigma$ were carried out at least three times before and after each experiment. Uncertainties estimated at 95% confidence were 2.0, 3.0 and 2.0 % for $\rho$, $\mu$ and $\sigma$, respectively. The temperature of the continuous phase was monitored by a digital thermometer (Sato, SK-1250MC, accuracy: ±0.53 K) and it was within the range of 293 – 295 K through all the experiments.

3. Results and discussions

The effect of initial shape deformation on $V_T$ of single drops was examined and compared with that of single bubbles\(^{(14)}\). Figure 2 shows recorded images, measured aspect ratio $E$ and rise velocities $V_Z$ of single drops and bubbles of $E_o \equiv 1.0$ released from nozzles with different diameters $D_N$. Bubbles or drops released from smaller diameter nozzles exhibit deformed initial shapes as shown in Fig. 2 (a) and (c), whereas the initial deformation of bubbles or drops released from larger diameter nozzles is small as shown in Fig. 2 (b) and (d). As for single bubbles, a slight difference in initial shape deformation causes a large difference in $E$ and $V_Z^{(11)}$. To the contrary, $V_Z$ and $E$ of single drops are not affected by initial shape deformation. This might be due to the higher viscous damping of shape oscillation of single drops than that of single bubbles. In fact, the strong fluctuation of $E$ just after the release is observed only for bubbles as shown in Fig. 2 (c). Measured drop Reynolds numbers in clean and contaminated systems at $\log_{10} M = -3.0$ are plotted against $E_o$ in Fig. 3 (a). For comparison, measured bubble Reynolds numbers at the same Morton number\(^{(14)}\) are shown in

![Fig. 2 Effects of initial shape deformation (log10 $M \equiv -4.8$ and $E_o \equiv 1.0$)](image)
There is a large scatter of bubble Reynolds number in the clean system due to the difference in initial shape deformation, and the scatter in the contaminated system is very small because of the high damping of shape oscillation caused by surfactants. On the other hand, no scatter of drop Reynolds numbers is observed not only in the contaminated systems but also in the clean systems. These experimental data imply that there is little influence of initial shape deformation on $V_T$ of single drops for clean and contaminated systems, and therefore, $V_T$ and $C_D$ are uniquely determined by fluid properties, $d$ and surfactant concentration.

Comparisons of measured $V_T$ of single drops between clean and contaminated systems are shown in Fig. 4. The $V_T$ of single contaminated drops is lower than that of single clean drops. The difference in $V_T$ between clean and contaminated systems becomes small in a high $\kappa$ system as shown in Fig. 4 (c). Though the addition of surfactants changes $\sigma$, this change does not cause any change in $V_T$ because the terminal condition in the present experimental conditions is determined by the viscous and inertial forces (viscous force dominating regime), in other words, by the drop Reynolds number. Measured $C_D$ is plotted against $Re$ in Fig. 5, which clearly indicates that $C_D$ does not depend on $M$ but on $Re$. As the viscosity ratio $\kappa$ becomes smaller, $C_D$ of contaminated drops becomes higher than that of clean drops. This increase of $C_D$ must be due to the well-known Marangoni effect caused by the concentration gradient of adsorbed surfactants on the interface. To the contrary, the difference in $C_D$ between
the clean and contaminated systems becomes smaller as $\kappa$ increases. According to Eq. (5), $C_D$ of a spherical drop at a low $Re$ number depends on $(\kappa + C/\mu)$, and therefore, the influence of surfactants on $C_D$ and $V_T$ becomes weaker as $\kappa$ becomes larger and larger than $C/\mu$. Hence, the experimental results indicate that the Levich’s term $[(2 + 3\kappa + 3C/\mu)/(1 + \kappa + C/\mu)]$ in Eq. (5) might be applicable to $C_D$ and $V_T$ models not only for low $Re$ number conditions but also for intermediate $Re$ number conditions.

Figure 6 shows measured $Re$ numbers of single clean drops with Eqs. (6) and (8) for bubbles in pure and fully contaminated systems under various conditions; $-11.6 < \log_{10} M < -0.9$, $0.1 < \kappa < 31.5$, $1.7 \times 10^{-1} < Re < 2 \times 10^2$ and $1.7 \times 10^{-2} < E_o < 12.1$. Drop shapes are also shown (the discussion on drop shapes will be reported in elsewhere). The measured $Re$ under the largest $\kappa$ condition ($\kappa = 31.5$) is close to Eq. (8) for contaminated bubbles. As $\kappa$ decreases, the measured $Re$ approaches to Eq. (6) for clean bubbles. This indicates that $\kappa$ strongly affects the drop $Re$ number, and that drop $Re$ numbers would lie in the region between the curves of Eqs (6) and (8). The first term in the right-hand side of Eq. (6) or Eq. (8) can be obtained by setting $(\kappa, C/\mu) = (0,0)$ or $(\kappa, C/\mu) = (0,\infty)$ in the following equation:

$$C_D = \frac{8(2 + 3\kappa + 3C/\mu)}{Re(1 + \kappa + C/\mu)}(1 + 0.15Re^{0.687}) \quad (9)$$

The above equation is the combination of Eq. (5) and the following equation proposed by Schiller and Nauman$^{15}$ for single spherical solid particles.

$$C_D = \frac{24}{Re}(1 + 0.15Re^{0.687}) \quad (10)$$

For $(\kappa, C/\mu) = (\infty, 0)$, the right side of Eq. (9) becomes equal to the first term of Eq. (8), i.e., Eq. (10). Hence, the experimental results shown in Fig. 6 indicate that (i) the values of $C_D$ for single drops are in the range between the values of $C_D$ for clean bubbles and for solid particles, and (ii) Eq. (9) will be applicable to the evaluation of $C_D$ for single drops under various $\kappa$ conditions.

To examine the applicability of Eq. (9) to clean drops ($C = 0$) under various $\kappa$ conditions, $C_D[(1 + \kappa)/8(2 + 3\kappa)]$ for clean drops for various systems is plotted against $Re$ and compared with Eq. (9) in Fig. 7. There is a strong correlation between $C_D[(1 + \kappa)/8(2 + 3\kappa)]$ and $Re$. 

![Fig. 5 Measured $C_D$ and $Re$ in clean and contaminated systems](image)

Fig. 5 Measured $C_D$ and $Re$ in clean and contaminated systems
The calculated $C_D$ using Eq. (9) with $C = 0$ agrees well with the measured $C_D$ of single clean drops. This result supports the speculation that the influence of $\kappa$ on $C_D$ for single drops can be evaluated by the term $[8(2 + 3\kappa)/(1 + \kappa)]$ not only in low Re but also in intermediate Re conditions ($Re < 200$).
Then, measured drag coefficients $C_D$ of single drops in fully contaminated systems ($C \to \infty$) are plotted against $Re$ and compared with Eq. (9) in Fig. 8. The measured $C_D$ agrees well with $C_D$ calculated by Eq. (9) with $C \to \infty$. This indicates that the influence of $\kappa$ on $V_T$ is negligible in fully contaminated systems as Eq. (9) implies. Comparison between all the measured and predicted terminal velocities are shown in Fig. 9. The calculated $V_T$ agrees well with the measured $V_T$ and the maximum error is about 10% under the conditions of $-11.6 < \log_{10} M < -0.9$, $1.7 \times 10^{-1} < Re < 2 \times 10^2$, $1.7 \times 10^{-2} < Eo < 12.1$ and $0.1 < \kappa < 100$. 
4. Conclusion

Terminal velocities of single drops rising through infinite stagnant liquids under wide ranges of fluid properties were measured to examine the effects of initial shape deformation, surfactants and the viscosity ratio. As a result, the following conclusions were obtained.

(1) The terminal velocity $V_T$ of a drop is not affected by initial shape deformation due to strong viscous damping of shape oscillation.

(2) The drop drag coefficient $C_D$ is a function of the drop Reynolds number $Re$, the viscosity ratio $\kappa$ and surfactant concentration.

(3) Surfactants increase $C_D$ and the influence of surfactants on $C_D$ becomes weaker as $\kappa$ increases. This trend is consistent with the Levich’s drag model.

(4) The effect of $\kappa$ on $C_D$ can be evaluated by the factor in the Levich’s model, $[(2 + 3\kappa)/(1 + \kappa)]$, even at intermediate drop $Re$ numbers.

(5) The combination of the Levich’s model and Schiller & Nauman’s correlation gives good estimation of $C_D$ of single drops in clean and fully contaminated systems under the conditions of $-11.6 < \log_{10} M < -0.9$, $1.7 \times 10^{-1} < Re < 2 \times 10^2$, $1.7 \times 10^{-2} < Eo < 12.1$ and $0.1 < \kappa < 100$.

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