

Analysis of behavior of liquid droplets on vertically oscillating substrates

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1. Introduction

In the paint coating process, workability must be controlled by its mechanical properties. For example, wetting phase right after the paint droplet impacts on the substrate in spraying is dominated by paint's surface tension and viscosity. However, the surface tension of the paint cannot be measured by existing methods.

Wilhelmy, du Noüy, and maximum bubble pressure methods are often employed to obtain surface tension. However, the measured values are not satisfactorily accurate for highly viscous samples. The oscillation of flying droplets also gives the surface tension^{1, 2)}. In the method, surface tension is calculated by the resonance frequency of the droplet oscillation, which is not affected by its viscosity. However, this method requires continuous and stable ejection, which is difficult for paint because of its high spinnability.

In our previous study, we reported the oscillation of the droplet excited by the electric tweezer system on the stationary substrate to measure the surface tension³⁾. However, this method cannot be applied for the dynamic surface tension measurement immediately after the droplet is emitted because the metal needle set above the substrate is in the path of the emitted droplet. In this work to solve the problem, we tried to excite the droplet oscillation through the mechanical vertical vibration of the substrate.

2. Experiment

We first summarize the behavior of the droplet oscillated by the electric tweezer system. The behavior is similar to that of the droplet flying in the air. For example, the resonance frequency f_i and the full width at half maximum (FWHM) at the same contact angle are proportional to $R^{-3/2}$ and R^{-2} respectively, where R is the radius of the droplet. In addition, the dependence of the resonance frequency on the contact angle θ_c is qualitatively explained by the theory of surface waves. Concretely, f_i is expressed by

$$f_i = \alpha(\sigma/\rho)^{1/2} L^{-3/2}, \quad (1)$$

where α is a constant, σ is the surface tension, ρ is the density, and L is the meridian length expressed as the $L = 2\theta_c R(\theta_c)$. In addition, the droplet volume V_0 is given as:

$$V_0 = \pi R^3 \left(\frac{1}{3} \cos^3 \theta_c - \cos \theta_c + \frac{2}{3} \right).$$

Secondly, we introduce the newly developed

experimental system. We employed the piezoelectric element as a device to excite the sinusoidal oscillation to the droplet on the glass plate as a substrate. The glass plate is water-repellent except for the areas in contact with the droplet and the contact angle can be controlled by changing the area of non-water-repellent.

Furthermore, we used the stroboscopic technique to observe high-speed oscillation of the droplet. The frequency of the stroboscope used for backlighting is set to be slightly shifted from the frequency of the voltage driving the piezoelectric element. **Figure 1** shows our experimental setup schematically, which is composed them.

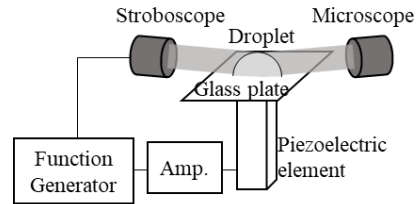


Fig. 1 Schematic diagram of the experimental setup.

We prepared liquid droplets in the volume range of 0.5-4 μ l with micro syringe on the glass plate. We used ethylene glycol and distilled water. We applied the sinusoidal voltage in the range of frequency $f=50-400$ Hz.

3. Results

We measured the position of the top of the droplet $x(t) = A\cos(2\pi ft - \varphi)$, where A and φ are the amplitude and phase of the oscillation.

We start by considering the dependence of the resonance frequency on the radius. **Figure 2** shows the normalized amplitude $A(f)/A_{max}$ obtained for ethylene glycol with contact angle=90-100° and distilled water with contact angle=105-110°, where A_{max} is the maximum amplitude.

The resonance frequency, which was determined from the spectrum peak, increased as the droplet radius decreased. **Figure 3** shows the dependence of the resonance frequency on the droplet radius. The resonance frequency is proportional to $R^{-2/3}$. This result agrees with previous research carried out with the electric tweezer system. The dependences of FWHM on the droplet radius and the resonance frequency on the contact angle are also in good agreement with the previous research.

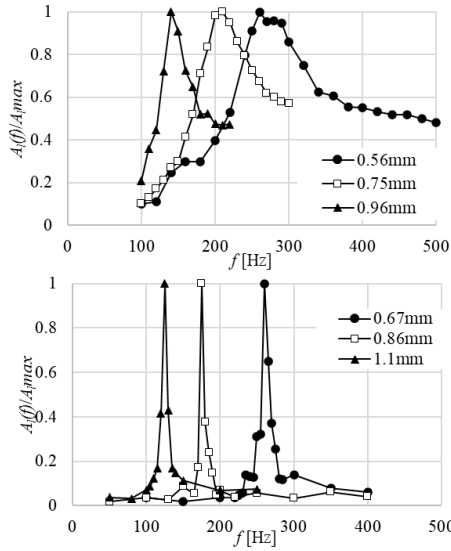


Fig.2 Dependence of the normalized amplitude of ethylene glycol (upper) and distilled water (below) on the frequency. Legends indicate droplet radius.

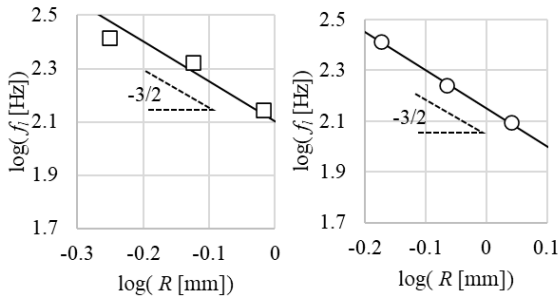


Fig.3 Dependence of the resonance frequency on the droplet radius. The squares (left) and circles (right) indicate the measured values for ethylene glycol and distilled water, respectively. The solid lines indicate the lines with the slope of $-3/2$.

Having discussed the comparison of the inducing method of the droplet oscillation, we turn now to the phase difference, which cannot be measured in the previous research. Let us assume that the droplet behavior follows Voigt model, that is the oscillation is expressed by

$m(d^2x/dt^2) = -k(x - A_s \cos 2\pi ft) - B(dx/dt + 2\pi A_s f \sin 2\pi ft)$, (2) where A_s is the amplitude of the substrate, m is the mass given by the density and volume, k is elastic coefficient given by surface tension, and B is viscous coefficient given by viscosity. The droplet deformation is calculated by Eq. (2) at the steady state. **Figure 4** shows the measured spectra and the theoretical results obtained by Eq. (2) for the droplets of ethylene glycol and distilled water with the contact angle = $94-100^\circ$ and the volume of $1\mu\ell$. The

mass m is calculated by the density and volume of the droplet and k and B are adjusted to reproduce the experimental results.

As shown, the measured and theoretical results show good agreement. Moreover, the ratio between theoretical elastic coefficient of 0.65 is in good agreement with the ratio between their surface tension of 0.66. Similarly, the ratio between theoretical viscous coefficient of 20 agrees with the ratio between their viscosity of 20.

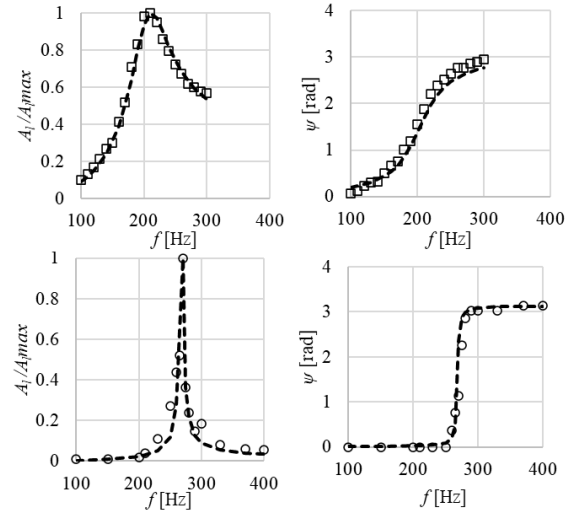


Fig. 4 Comparison of the measured and theoretical results of ethylene glycol (upper) and distilled water (lower). The squares and circles indicate the measured spectra. The dashed lines indicate theoretical results.

4. Conclusion

We observed the behavior of the droplet on the vertically oscillating substrate with nothing in the path of the emitted droplet. We believe these results open up the possibility of the new measurement method of mechanical properties of liquid droplet immediately after being emitted. The final aim of this study is to measure the mechanical properties of the droplet in actual industrial coating, of which size is μm order. The expected oscillation frequency is in the range of 10^5Hz and the expected amplitude is nm order. Therefore, we will attempt to employ the laser as a tool to detect the smaller amplitude at higher frequency. Furthermore, this method will help to clarify the nonlinear behavior of the droplet because it can easily excite the droplet oscillation having large amplitude. The detail would be reported in the presentation.

References

1. B. Stückrad, W. J. Hiller, and T. A. Kowalewski: *Exp. Fluids*. **15** (1993) 332.
2. T. Ishiwata, and K. Sakai: *Jpn. J. Appl. Phys. Express* **7** (2014) 077301-1.
3. S. Ishida, M. Iga, S. Mitani, and K. Sakai: *Jpn. J. Appl. Phys.* **61** (2022) SG1064-1.