Observation of colliding and mixing picoliter droplets inflight and on substrate

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1. Introduction

Using picoliter droplets generated by inkjet technique, we developed measurement system for micro liquid dynamics that could be used for various observations, e.g., capturing time-lapse views of the wetting and penetration process of a liquid on various substrates. ^{1,2)} During such observations, managing the purity of the flying point of the droplets in the air and landing point on the substrate is important. We designed the observation system based on so-called on-demand inkjets, wherein the timing of droplet ejection can be controlled freely by the electrical signal generator. On the other hand, the accuracy of the time from the birth of droplet, that is surface age, is also important for measurements of physical properties, and our system is also able to manage to generate droplets by continuous type of inkjet for that purpose.³⁾ Our current setup has capacity to eject viscous liquid such as aqueous solution of glycerol with high concentration with the viscosity of about 200 mPa s or high-surface-tension liquid, such as water with about 70 mN/m, into droplets.⁴⁾ This means that our device is useful for observations and measurements of micro fluid properties and dynamics of various kinds of liquids.

In this work, we investigated the collision behavior of two droplets with different surface properties but dissolving into each other, with high spatial and temporal resolution, to confirm that one droplet covers the other one owing to the difference in surface tension.

2. Experiments and results

The experimental setup in this work was almost similar with that of our previous work ⁴) except that two generation devices of droplets were installed side by side, as shown in **Fig. 1**. The electrical signal applied to the PZT actuators in each device was identical and the the timing of the collision of two droplets was controlled by spatial distance from each other by manipulators attached to the generation devices. The nozzle ejecting sample



Fig. 1 Photo of our droplet generation and observation system in this work. The nozzle in the right side is for distilled water and the left for dye doped ethanol.

liquid was made of glass and the tip size was 50 μm in diameter, and is disposable.

The materials were distilled water and ethanol with added rhodamine B as a liquid identifier on transmitted light observation not on fluorescence observation. In this work, surface tension of liquid is quite important, and we measured the surface tension values of sample liquids using the Droplet Oscillation Inflight (DOF) method. ³⁾ In this method, oscillation frequency and attenuation of flying droplet provide us the dynamic surface tension and viscosity of liquid, respectively, in a non-contact



Fig. 2 Droplet oscillation analysis of distilled water inflight just after ejected from the nozzle of our device. Data points show the length perpendicular to direction of droplet travel and the curve shows that fitted by Eq. 1.

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manner. In the DOF method, droplet oscillation is, normally, induced by electrical force for the sake of deformation starting at an arbitrary time, but we observed the shape of droplet just after ejection from the nozzle in this work because we need those properties at short time region around 50µs. Figure 2 shows the typical time dependence of the droplet length perpendicular to direction of droplet travel. The curve in this figure was obtained by fitting with the following equation using surface tension σ and viscosity η as fitting parameters,

$$\xi = Ae^{-\gamma t} \sin(\omega t + \delta) + B ,$$

$$\omega = \sqrt{8\sigma/\rho R^3} , \qquad (1)$$

$$\gamma = 5\eta/\rho R^2 .$$

From this figure, we can obtain the surface tension of 70.88 mN/m and the viscosity of 1.05 mPa·s of distilled water. In the same way, the surface tension values of pure ethanol and dye-doped ethanol were 24.07 mN/m and 17.94 mN/m, respectively, and the viscosity values were 1.37 mPa·s and 1.35 mPa·s, respectively.

We managed some experiments in which a water droplet gently collided with an ethanol droplet in air or on glass plate. Owing to the difference in surface tension between water and ethanol, ethanol droplet will completely and immediately cover the water droplet, after that two materials will mix according to the diffusion law. Now we estimate the time required for ethanol to wrap a spherical water droplet by assuming the condition of complete wetting. Driving force for ethanol to cover droplet surface is the difference in surface tension $\Delta\sigma$ between two liquids and this force balances with viscous drag as follows,

$$\pi r d \cdot \eta \left(\frac{v}{d}\right)^2 \sim \Delta \sigma V \quad , \tag{2}$$

where r, d, V and η are radius of water droplet, thickness of ethanol layer, covering velocity of ethanol and the viscosity of ethanol, respectively.



Fig. 3 Colliding droplets of distilled water from the right hand and dye-doped ethanol from the left in air. At the time of $0\mu s$, two droplets came into contact with each other. The blueish region at $15\mu s$ is presumed to be thin ethanol layer covering water droplet.



Fig. 4 Dye-doped ethanol droplet colliding with distilled water droplet on a glass plate. In this figure, we can see the mirror images of actual droplets reflected on the glass surface on the lower side of each photo.

With the values of liquid properties mentioned above and r and d values of 25 μ m and 0.5 μ m, respectively, the estimated time to wrap over the water droplet completely is calculated about 23µs. Figure 3 shows microscopic and stroboscopic time-lapse photos of colliding droplets of water and dye-doped ethanol in air. We can see a boundary surface between ethanol and water and also see that thin layer of ethanol is moving over water surface in the middle picture. From this figure, we estimate that the time for ethanol to wrap completely water droplet is about 30µs, which is nearly equal to the value of our estimation from Eq. (2). We made an experiment to observe droplet collision on glass surface shown in Fig. 4, in which the relative velocity of two droplet was higher than that in Fig. 3. It seems that ethanol covers water surface on substrate more quickly than in the air, and we are trying to find the reason for that by performing additional experiments.

3. Summary

We successfully observed that miscible liquids have boundary surface for a short time region before they are mixed with each other, and we confirmed that the order of that time for ethanol to cover water surface was almost the same as the estimated values of surface tension difference and viscosity.

References

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