# Experimental and Numerical Study of Coalescence Between Two Picoliter-Sized Droplets on a Solid Surface

*Takahiko Matsumoto, Manabu Seo, and Ryohta Suzuki; RICOH Company, Ltd.; Kanagawa, Japan*

## Abstract

*In liquid inkjet printing, a critical quality issue is coalescence between droplets on media surface. To control the coalescence processes, it is useful to study how parameters, including liquid property (density, viscosity and surface tension) and affinity between liquid and a solid surface (advancing and receding contact angle), affect to the processes. However, the effect of changes with the parameters in coalescence processes between picoliter-sized droplets is not studied minutely in previous reports.*

*This paper describes simulation results about deriving equations of the parameters for coalescence processes between two droplets and experimental results for verification of the simulation results under the condition, in which absorption of moisture and evaporation of droplets can be neglected. In our works, experimental and simulation results are almost consistent, and some of the equations can be applied to actual coalescence processes.*

## Introduction

In liquid inkjet printing, a critical quality issue is coalescence between droplets on media surface. To control the coalescence processes, it is useful to study how parameters, including liquid property (density, viscosity and surface tension) and affinity between liquid and a solid surface (advancing and receding contact angle), affect to the processes.

In previous reports, some studies about coalescence on a substrate are reported. For example, an experimental study on the dynamics of coalescence of two water droplets [1], an experimental and numerical study of coalescence on a solid surface at microliter scale [2] and picoliter scale [3], and a numerical study of coalescence of droplets using Lubrication Theory [4] are reported.

However, the effect of changes with the parameters in coalescence processes between picoliter-sized droplets is not studied minutely in previous reports.

This paper describes simulation results of coalescence processes between two droplets on a solid surface when the parameters are changed and experimental results for verification of the simulation results.

## Simulation of coalescence processes and the effect of parameter changes on the processes

Simulation of coalescence processes between two picoliter-sized droplets on a solid surface is carried out with Fluent (Ansys Inc.) when parameters, including liquid property and affinity between liquid and a solid surface, are changed. Table 1 shows the setup contents of simulation. The interface that volume fraction of liquid and air phase equals to 0.5 is defined to the surface of droplets. The parameters are viscosity, surface tension, volume of a droplet, density, advancing contact angle, and receding contact angle. Table 2 shows the ranges of the parameters. Figures 1 (a) and (b) shows that two droplets on a solid surface are touched at a distance of (28/15) $r$ each other (Figure 1 (a)) and the contact angle equals to the advancing contact angle (Figure 1 (b)) under initial condition (t = 0). To set contact angle hysteresis for the simulation, the contact angle of the region on a solid surface, in which two droplets are located at initial condition, is set to the receding contact angle and the other contact angle is set to the advancing contact angle (Figure 1 (c)). Then, contact angle of droplets is adopted advancing or receding contact angle in each region.

**Table 1. Setup contents of simulation with Fluent**

| Setup contents | | | |
|---|---|---|---|
| General | Solver | | Pressure-Based |
| | Time | | Transient |
| | Gravitational Acceleration | | 9.8m/s$^2$ (minus Z Direction) |
| Models | Mutiphase Model | | Volume of Fluid |
| | Viscous | | Laminar |
| Solution methods | Scheme | | SIMPLE |
| | Spacial Discretizarion | Gradient | Least Squares Cell Based |
| | | Pressure | PRESTO! |
| | | Momentum | Second Order Upwind |
| | | Volume Fraction | Geo-Reconstruct |
| | Transient Formulation | | First Order Implicit |

**Table 2. Ranges of simulation parameters**

| Parameters | | | Ranges |
|---|---|---|---|
| $\eta$ | Viscosity | (mPa・s) | 10～1,000 |
| $\gamma$ | Surface tension | (mN/m) | 20～70 |
| $V$ | Volume of a droplet | (pL) | 1～1,000 |
| $\rho$ | Density | (kg/m$^3$) | 500～2,000 |
| $\theta_a$ | Advancing contact angle | (degree) | 30～90 |
| $\theta_r$ | Receding contact angle | (degree) | 0～90 |

### *Conditional equations for prediction of receding or non-receding contact lines*

In coalescence processes of two droplets on a solid surface, there are two cases: one is a case of receding contact lines, and the other is a case of non-receding contact lines (Figure 2). Figure 3 shows the simulation results, in which these cases are discerned. The results depend only on advancing and receding contact angle among the parameters. Thus, the conditional equations for these cases are described by following equation (1) and (2).

(non-receding contact lines)

$$0 \le \theta_r < \frac{2}{3}\theta_a, \tag{1}$$

(receding contact lines)

$$\frac{2}{3}\theta_a \le \theta_r \le \theta_a, \tag{2}$$

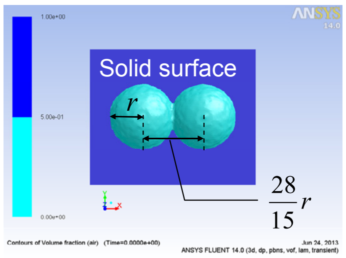



***Figure 1 (a)** Front view under initial condition (t = 0).*

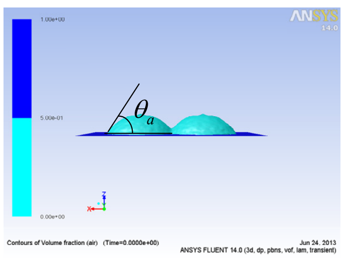



***Figure 1 (b)** Side view under initial condition (t = 0).*

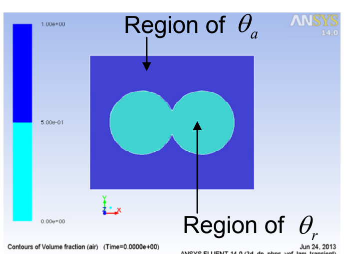



***Figure 1 (c)** Regions of advancing and receding contact angles on a solid surface.*

### *Derivation of equations about $T$ and parameters*

The equations about $T$ and the parameters are derived in each case of receding or non-receding contact lines. $T$ is the time from initial condition (t = 0) to final condition, in which coalescence processes are finished, and is defined to the time until the perimeter of two droplets become constant.

In the case of non-receding contact lines, the equation is described by following equation (3). The equation doesn't depend on density of liquid and receding contact angles.

(non-receding contact lines)

$$T \propto \frac{\rho^0 V^{0.3356}\eta}{\gamma^{0.9814}\left(\theta_r^0 \theta_a^{3.3539}\right)} \cong \frac{V^{1/3}\eta}{\gamma\theta_a^{10/3}}, \tag{3}$$

In the case of receding contact lines, the equation is described by following equation (4). The equation doesn't depend on density of liquid.

(receding contact lines)

$$T \propto \frac{\rho^0 V^{0.3413}\eta}{\gamma^{1.0644}\left(\theta_r^{2.692}\theta_a^{0.6619}\right)} \cong \frac{V^{1/3}\eta}{\gamma\left(\theta_r^{8/3}\theta_a^{2/3}\right)}, \tag{4}$$

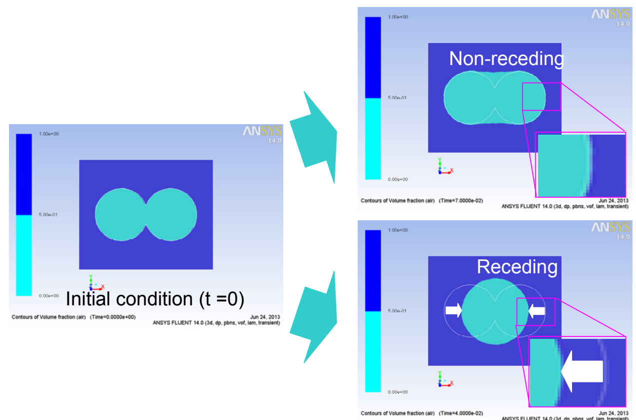



***Figure 2** Each case of receding or non-receding contact lines in coalescence processes.*

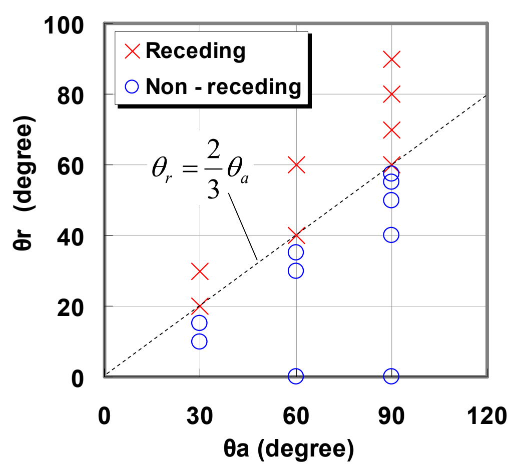



***Figure 3** Simulation results about each case of receding or non-receding contact lines.*

## Experimental setup for verification of simulation results

To verify the simulation results, observation of coalescence processes of two picoliter-sized droplets ejected from a inkjet print head (Cluster Technology, PIJ-25N) on a substrate is carried out with the experimental apparatus shown in Figure 4. Coalescence processes of two droplets are observed and recorded using high speed camera (Keyence, VW-6000). Ethylene glycol (Kanto Chemical, special grade) as the ejected liquid, and a coated slide (Matsunami glass, S081110: a slide coated with aminosilane) as the substrate are used. Trigger signal turns on when the stage passes a particular point, then the inkjet print head eject two set of droplets on the substrate. The liquid property of ethylene glycol can be changed because of absorption of moisture. To estimate the speed of absorption of moisture, the change of viscosity at the nozzle of inkjet print head is estimated by measuring the vibration of meniscus using a laser Doppler vibrometer at 25℃ [5]. As a result, viscosity at the nozzle is constant at relative humidity (RH) not greater than 10 %. Thus, the observation of coalescence is carried out at 25 ℃ and RH 9 % to avoid absorption of moisture. Table 3 shows liquid properties of ethylene glycol at 25 ℃.

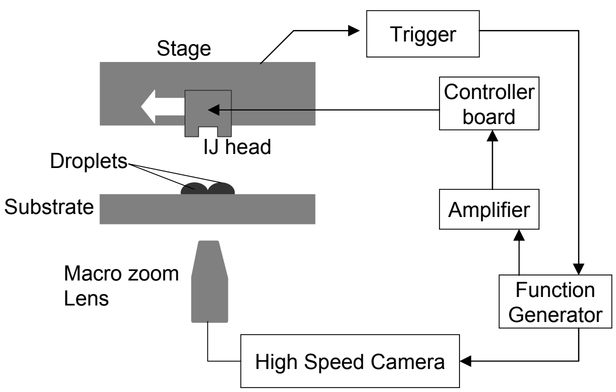



*Figure 4* *Schematic diagram of experimental apparatus for observation of coalescence processes.*

**Table 3. Liquid properties of ethylene glycol at 25 ℃**

| | Viscosity (mPa・s) | Surface tension (mN/m) | Density (kg/m$^3$) |
|---|---|---|---|
| Ethylene Glycol | 16.6 | 50.4 | 1.111 |

## Simulation setup for comparison with experimental results

Simulation of coalescence of two picoliter-sized droplets on a solid surface is carried out for comparison with experimental results. The setup contents of simulation are the same as those mentioned above (Table 1). Liquid properties of ethylene glycol shown in Table 3 are set to simulation parameters. The time, in which two droplets on a solid surface are touched, is set for initial condition (t = 0). To set contact angle hysteresis for the simulation, the contact angle of the region on a solid surface, in which two droplets are located at initial condition, is set to the receding contact angle and the other contact angle is set to the advancing contact angle as mentioned above (Figure 1 (c)). Measurement results of volume of a picoliter-sized droplet of ethylene glycol and advancing and receding contact angles at microliter and picoliter scale are set to simulation parameters.

### *Measurement of advancing and receding contact angles for simulation setup*

Advancing and receding contact angles of ethylene glycol on a coated slide are measured at microliter and picoliter scale for simulation setup. The results are shown in Table 4.

At microliter scale, the advancing and receding angle are measured with expansion-contraction method using a contact angle meter (Kyowa Interface Science, Drop Master 500).

At picoliter scale, the advancing and receding angle are measured by observing the evaporation process [6] of a picoliter-sized droplet of ethylene glycol on a coated slide horizontally with the experimental apparatus shown in Figure 5. The measurements are carried out at 25 ℃ and RH 9 % to avoid absorption of moisture as mentioned above. The contact angle $\theta$ of a droplet is derived from the measurement results of height $h$ and drop base radius $r$ using equation (5). Figure 6 shows $\theta$, $h$ and $r$ of a droplet on a substrate.

$$\theta = 2\tan^{-1}\left(\frac{h}{r}\right), \tag{5}$$

Figure 7 shows changes of $r$, $h$ and $\theta$ after the time when a picoliter-sized droplet ejected from a inkjet print head is landed on a coated slide. The advancing contact angle is decided to the contact angle when wet spread of a droplet is completed, the receding contact angle is decided to the contact angle when it is minimized (Figure 7).

**Table 4. Measurement results of advancing and receding contact angles at microliter and picoliter scale**

| | $\theta_a$ (degree) | $\theta_r$ (degree) |
|---|---|---|
| microliter droplet | 26.2 | 7.5 |
| picoliter droplet | 12.6 | 3.2 |

### *Measurement of volume of a droplet*

Volume of a picoliter-sized droplet of ethylene glycol on a substrate is measured for simulation setup with the experimental apparatus shown in Figure 5. The volume is derived from the measurement results of $h$ and $r$ using equation (6) at the time when a droplet is landed on a substrate.

$$V = \frac{\pi h}{6}\left(3r^2 + h^2\right), \tag{6}$$

As a result, the volume is 3.63 picoliter. The volume reduction rate is almost linear, and all of the volume evaporates in 20 seconds (Figure 7).

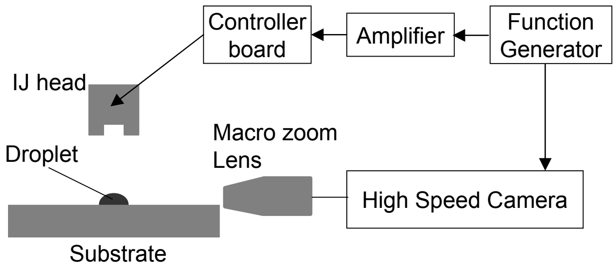



*Figure 5 Schematic diagram of experimental apparatus for observation of evaporation process of a picoliter-sized droplet of ethylene glycol.*

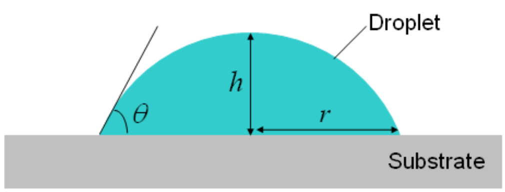



*Figure 6 A picoliter-sized droplet on a substrate.*

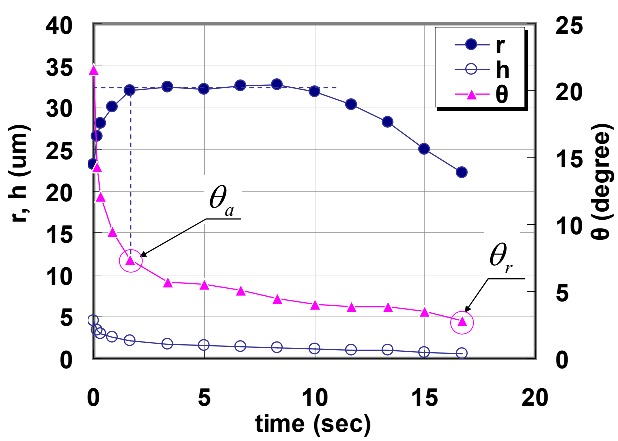


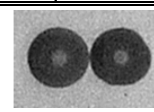
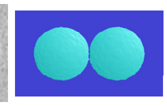
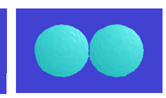
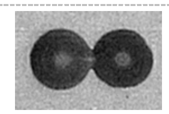
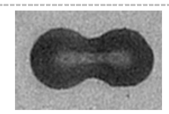
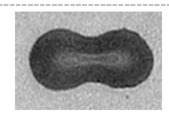
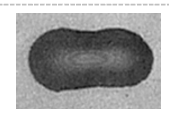
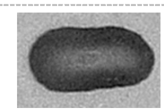

*Figure 7 Changes of h, r, and θ after the time when a picoliter-sized droplet of ethylene glycol is landed on a substrate.*

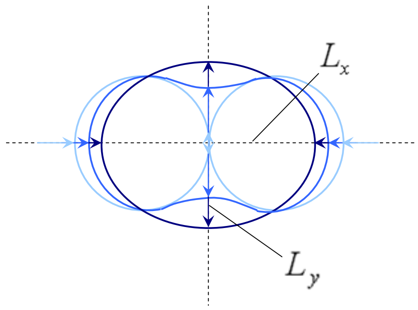
## Verification results and discussion

Table 5 shows experimental and simulation results about coalescence between two picoliter-sized droplets on a coated slide. The advancing and receding contact angle at picoliter scale shown in Table 4 are set to simulation parameters for Simulation 1 shown in Table 5, and those at microliter scale are set for Simulation 2 shown in Table 5. Coalescence processes are almost finished in 100 msec. The volume reduction rate is about 0.5 % after 100 msec, thus evaporation of two droplets can be neglected.

To compare the results quantitatively, changes of $L_x$ and $L_y$ with time are shown in Figure 8, and changes of perimeter of two droplets with time are shown in Figure 9. The experimental results are average values of three separate measurements, error bars represent standard deviation of uncertainty. The results of Simulation 1 show good agreement with experimental results. Setting the advancing and receding contact angle at picoliter scale to simulation parameters is useful to simulate coalescence processes between two picoliter-sized droplets. The experimental values of $L_x$ are constant with time as shown in Figure 8, and the results of contact angles at picoliter scale shown in Table 4 satisfy the equation (1). Thus, the equation (1) can be applied to actual coalescence processes. Experimental and simulation results are almost consistent in the case of non-receding contact lines as shown in Figures 8 and 9, thus the equation (3) can be applied to actual coalescence processes.

**Table 5. Experimental and simulation results of coalescence between two picoliter-sized droplets of ethylene glycol on a coated slide**

| Time (msec) | Experiment | Simulation1 | Simulation2 |
|---|---|---|---|
| 0 |  |  |  |
| 1 |  |  |  |
| 5 |  |  |  |
| 10 |  |  |  |
| 50 |  |  |  |
| 100 |  |  |  |



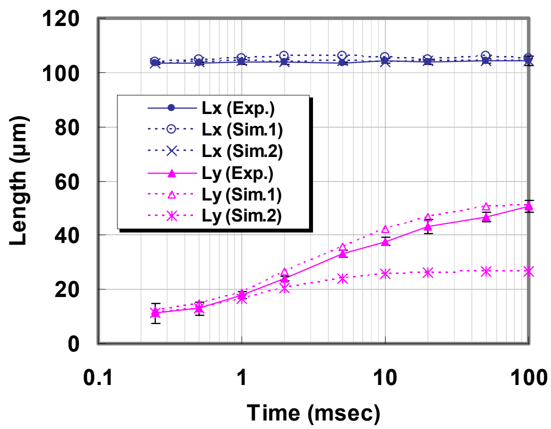




***Figure 8*** *Changes of $L_x$ and $L_y$ of two droplets with time in coalescence processes.*

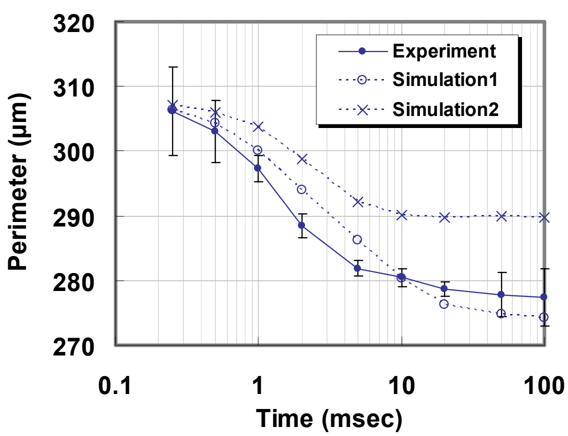



***Figure 9*** *Changes of perimeter of two droplets with time in coalescence processes.*

## Conclusion and Future Work

・The conditional equations described by equation (1) and (2) are derived from simulation of coalescence processes between two droplets on a solid surface for each case of receding or non-receding contact lines. The equations about $T$ and the parameters, which are described by equations (3) and (4), are also derived from the simulation for the cases.

・In the case of non-receding contact lines, experimental and simulation results of coalescence processes between two picoliter-sized droplets are almost consistent by setting the advancing and receding contact angle at picoliter scale to simulation parameters.

・ The equation (1) and (3) can be applied to actual coalescence processes.

We will study coalescence processes between two picoliter-sized droplets in the case of receding contact lines and verify the equation (2) and (4) in future work

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## Author Biography

*Takahiko Matsumoto received his M.S. degree in Department of Advanced Material Science from The University of Tokyo. In 2007, he joined Ricoh Company and has engaged in the research and development of a field of surface chemistry and fluid mechanics. His research interests includes inkjet systems, phenomena of surface chemistry, and CFD simulation.*