# Multi-Color Printing

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## Abstract

The impact of liquid drops onto a solid surface, both dry and wet, has been quite well characterized in the literature. However accurate information is still missing about the physics of the relevant phenomena when multiple drops impinge successively onto a solid which is a quite ubiquitous phenomenon in ink-jet printing. In a previous work, the authors have studied the fate of an incoming drop impinging in an axisymmetric manner onto another droplet resting under steady state conditions on the substrate. This situation which is much easier to perform experimentally was also chosen as a test case for studying the coalescence of two drops and some preliminary results have been given.

In this paper, the previous work is extended to rather quite frequent cases in digital printing, such as miss hit and/or the mixture of colors when printing on impervious substrates. For this purpose, a new set-up was built consisting of at least two multi-nozzle printheads mounted on translation and rotation tables which can be used to simulate different printing situations. The experimental methods are based on visualization techniques such as high speed cinematography for large drops and phase controlled ultra short snap shots of the impact process when micro-sized drops are used. The experiments are performed at low enough velocities and the emphasis is now given to asymmetric configurations.

Image processing with specific algorithms helps to delineate the contour of the merging drops and to follow accurately the drainage of the interface film and the subsequent coalescence process. The experimental results are shown to compare favourably with theory for drops colliding at low velocity. Experiments close to actual operating conditions are also performed and the merging behavior is detailed for both axisymmetric and non-axisymmetric configurations. Finally the stability of the printed pattern is analyzed and discussed with relevance to the spreading behavior of drops on differently wetting substrates.

## Introduction

A large number of natural and engineering applications are concerned with drop impact and collisions. Some of these include the study of raindrop formation,[1] the efficient distribution of agricultural spays[2] and the emerging applications of molten metal droplet based manufacturing[3] or OLED fabrication.[4] Detailed knowledge of droplet collision is also critical for the control of advanced processes such as containerless processing in the space environment.[5]

The fluid flow associated with impinging drops is rather complicated because of the extreme deformation of the droplet surface occurring within very short time scales. In the case of low impact velocities such as those occurring in drop on demand printing, the spreading regime is essentially controlled by the interplay between inertial effects which dominate the early spreading of the fluid and viscous and surface tension forces which slow the spreading and eventually bring the fluid to an equilibrium configuration. Two non-dimensional quantities have been demonstrated to characterize such impacts: the initial Reynolds number and the initial Weber number.[6] When impact velocities become important, it has been shown elsewhere that compressibility effects may play a major role.[7]

The work, which is presented here, is connected to ink-jet printing where single or multiple drops may impact onto solid surfaces in a number of different configurations. Much of the work published in the literature concerns the understanding of the fundamental physics of impact phenomena and thus examines only the normal impact of a single droplet onto a flat surface.[8] Few results are available about the axisymmetric impact of a drop onto another resting on a solid surface[9] and even less is known when the drop impact is non axisymmetric and thus must be considered to be fully three dimensional. It is useful to recall here that the latter situation happens quite often in multi-fluid ink-jet printing where a variety of drop onto drop configurations (direct hit, partial hit, and miss-hit) are used to create color patterns. Drop coalescence also arises in many other different contexts such as drop coarsening in dispersions[10] or by condensation.[11]

In the first section of this paper, two different experimental devices each with their own jetting and visualization system are described. Macro-drops (with diameters close to several millimeters) are ejected with a syringe and the drop dynamics are followed with a high speed camera. For the purpose of performing experiments with picoliter sized drops (micro-drops), a particularly elaborate jetting device has been designed and built. This device may carry a large number of multi-nozzle printing heads which can be located in different positions. It has been shown elsewhere[12] that an extremely fast camera taking several millions of frames per second is necessary to fully capture the micro-drop dynamics. Instead of that extremely sophisticated and costly solution, the pseudo-cinematography method combined with a fast shutter high resolution camera has been used. This technique allows following the evolution of the drop dynamics thanks to snap shots of different droplets taken at successive stages of the process. The main assumption which is made is that the process is fully deterministic and this has been checked elsewhere.[12]

In the section on experiments and discussion, the emphasis is on the characterization of phenomena related to low speed impacts of single and multiple drops onto a variety of model and industrial substrates. Through these experiments not only the effects of dynamic impact conditions but also the wetting effects by detailing drop velocity, shape and size at various steps of the spreading phase are investigated. Indeed, a particular effort has been made in capturing the transient interfaces during the merging of several drops. First an axisymmetric landing (direct hit) of the second drop onto the first one[7] is considered. Experiments with micro-drops and macro-drops are shown with evidence that in both cases the incoming drop strikes the sitting one almost as a solid sphere. This facilitates the prediction of the spreading diameter and comparisons with a single drop are given for different wetting conditions. The second configuration which is considered here is the non-axisymmetric coalescence (miss hit). Again both macro and micro-drops are considered with the incoming drop impinging the solid substrate at a given distance of the static drop. The impacting drop spreads under the influence of inertial forces and coalesces with the static one. The experiments show the transient interfaces of the dumbbell shaped composite drop with both side and top views and comparisons with theoretical profiles are given. The purpose of using both macro and micro-drop is to demonstrate the existing similarities when the experiments are performed at similar dimensionless numbers. It is worth emphasizing that both geometric and time scales are larger by an order of three for macro-drops so the experiments are more easily performed and with better accuracy.

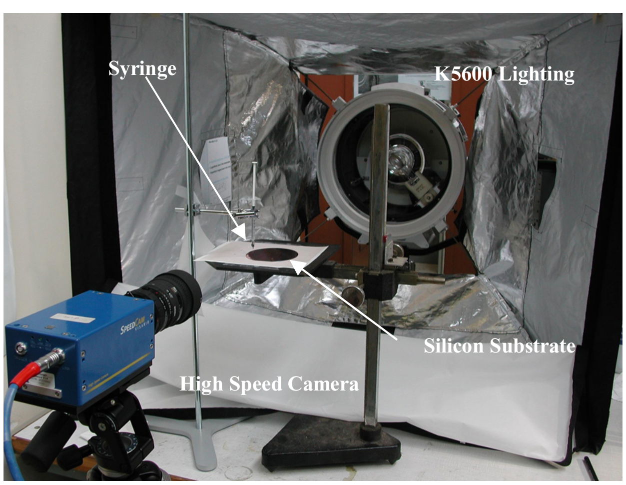
Finally the conclusion section summarizes the main results of this work where situations similar to that encountered in actual matrix ink-jet printing are detailed. It is expected that the results obtained in this work will lead, among other things, to a better understanding of color artifacts due to the hydrodynamics of drop coalescence.

## Experimental

The experimental set-ups used in this work have several droplet generating devices in order to cover a large range of droplet sizes. A syringe pump is used for forming macro-drops of several millimeters (Figure 1a) whilst commercially available printheads allow generating micro-drops in the range of tens of micrometers (Figure 1b). Each set-up also includes an illumination source, a camera (high speed or fast shutter according to the needs) and an optical system. The high speed camera is a stand-alone system whilst special triggering electronics combined with appropriate software is necessary for capturing the dynamics of micro-drops using the pseudo-cinema-tography technique.

### Fluids and Substrates

The fluids that were used in this study were distilled water, a mixture of distilled water, glycerol and surfactant and model UV inks available from Agfa-Gevaert® and SunJet® especially tuned for piezoelectric print-heads. The range of viscosities covered goes from 1 mPa.s to 30 mPa.s at a temperature of 20°C. Static surface tension for these different fluids varied between 30 and 73 mN/m. These characteristics were respectively measured using a Brookfield DVII viscometer and a Krüss K9 digital tensiometer.



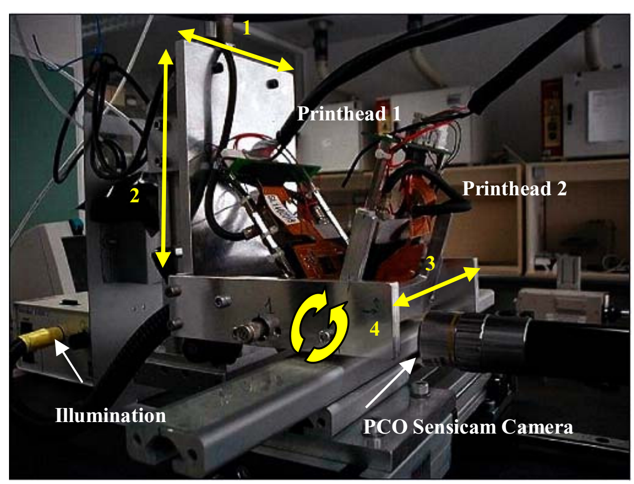




*Figure 1. (a) Experimental set-up for macro-drops , (b) Micro-drop jetting and measurement device*

Substrate materials with different wetting properties were used throughout this work in order to pinpoint the main differences which appear when the contact angles are different. The model substrates, onto which drops im-pinged, were wafers of oxidized silicon and hydrophobic silanized silicon that showed equilibrium contact angles of respectively 30° and 110° with water. These model substrates were all cleaned in a bath with oxygenated water and sulphuric acid and dried thoroughly before use in order to get rid of any organic contamination. The industrial substrate used was an hydrophobic polyester with a true polyesterterephtalate surface (PET) with no pores and of course no absorption. Distilled water deposited on this substrate showed a contact angle larger than 80° whilst the equilibrium angles found on this substrate using the above described model UV inks was close to 35°. In contrast to the microporous and polymeric blend materials that are used elsewhere (typical coating thickness of 20 to 40 µm) for single drop impact and spreading with imbibition the above described hydrophobic and hydrophilic substrates can be considered to be totally non-absorbing and thus only superficial spreading effects are supposed to be active.

### Jetting Devices and Visualization Systems

Water based droplets with radii in the millimeter range were created by pumping the mixture (water/glycerol/ surfactant) through a needle using a syringe pump. The size of the droplets could be tuned by changing the diameter of the needle and/or the surface tension of the jetted fluid. The velocity of the drops could be varied by almost one order of magnitude (0.3 to 3 m/s) by simply changing the height of fall (distance between needle and the substrate). The velocity was calculated knowing the distance of travel of the drop and the time needed for that travel. The camera used to capture the impact and collision processes of macro-drops was a high speed camera (Visario 1500 from Weinberger AG®) with a CMOS sensor and a full resolution of 1536 x 1024 pixels at 1000 frames/s (Figure 1a). A binning procedure was used to reduce the image format to 762 x 512 pixels and increase the framing rate to 4000 images/s. This was largely sufficient for most of the experiments performed in this work. Above that rate some illumination problems occurred although a very powerful lamp (4 kW) with controllable beam pattern (K5600 Lighting®) was used as shown in figure 1a. The images acquired were stored directly on the camera during the experiment and then transferred to the computer for further analysis.

Micro-drops with an average size of 41 µm in diameter were formed using Spectra® SL printheads. Manipulation of the drive waveform and variation of the ink composition allowed forming drops with velocities varying from 4 to 8 m/s. As already stated in the introduction an innovative jetting system has been designed and built for studying the coalescence processes with micro-drops and it is detailed here. The printheads were mounted on a large mechanical structure as shown in figure 1b. Four different mobile parts were available on this system and rendered it quite versatile for a number of uses. Part #1 allowed for a vertical translation of 30 mm and could be moved transversally over a distance of 100 mm as shown by the double headed arrows on figure 1b. The printhead #1 could be shifted transversally on a length of about 8 mm whilst printhead #2 could be slanted with an angle of plus or minus 35° with reference to the vertical position. The mechanical structure also comprised a translation table on which different substrates could be positioned and printed. The impact and collision processes with micro-drops were captured using a PCO Sensicam® fast shutter video capturing system from PCO Computer Optics GMBH. This is a high resolution 1280 x 1024 pixels, 12 bit thermo-electrically cooled imaging camera which allows taking sharp snap shots of different experiments with a shutter time comprised between 500 ns and several µs. Two to three microseconds of exposure time revealed to be well adapted for this purpose. Multiple shots taken at different times of multiple droplets were combined into one single "video" according to the pseudo-cinematography technique that has been described elsewhere.[8] The reproducibility from one experiment to another was good provided that all experimental inaccuracies were made as small as possible. Dual-exposure shot measurements - possible with this camera -were used to determine the velocity of the impacting drops. The illumination used for this camera was far less sophisticated compared to the previous one and was a cooled high intensity fiber light source available from ILC Technology model R400-1 with front thermal glass. The cover glass prevented unwanted heating of the sample and also protected this very light sensitive camera.

Finally once the grabbing process is terminated, image processing software (Matlab® Image processing toolbox) was used to measure the transient diameter and height and eventually contact angle of the spreading drop from a gray scale image. The filters available within the toolbox are sufficient for processing macro-drop images but do not succeed with micro-drops. Specific algorithms combining segmentation, edge detection, search of drop profile and statistical processes have been written and allow for much better analysis of micro-drop impact and spreading processes.[8] Moreover the user environment is friendly and can be rendered fully automatic so as to process a full video film image after image.

## Results and Discussion

To fulfill the objectives listed in the introduction, experiments were performed with both large and small drops impacting either onto a solid substrate or colliding one onto another. In both cases, the final and transient spreading diameter and height were measured for a large range of impact Reynolds and Weber numbers given by:

$$Re = \frac{\rho U_0 D_0}{\mu} \quad ; \quad We = \frac{\rho U_0^2 D_0}{\sigma} \qquad (1)$$

where $D_0$ and $U_0$ are the initial drop diameter and the impact velocity of the drop. The other quantities are the characteristics of the fluid.

## Impact of a Single Drop

Before details and results on the coalescence of drops are considered, it is instructive to examine the temporal spreading of a single drop. When the drop touches down the substrate, there is only a contact point so the diameter can be considered to be very small whilst the initial height is equal to the diameter. The drop then begins to spread first as a truncated sphere before a lamella is ejected from the base of the drop and eventually forms a thin film bounded by a rim in the case of drops having high enough inertia. The rate of spreading is driven by the inertia of the drop and slowed by viscous effects. When the drop reaches its maximum radius, all the initial energy has been dissipated and the profile of the drop is close to that of a pancake. The characteristic time for this process can be taken as $D_0/U_0$[12]. This first inertial stage is followed by a second intermediate stage during which, depending on the wetting behavior of the substrate, two different things may happen:

- If the drop does not wet the surface, the surface tension forces will cause the drop to retract to sessile drop shape with eventually several oscillations.
- If the drop does wet the surface, the drop goes from a pancake shape to a truncated sphere without hardly any movement of the contact line.

The third stage is that of capillary spreading during which the drop wetting the substrate slowly moves to an equilibrium configuration with the diameter proportional to the 1/7 or 1/10 power of time depending on the history of the drop dynamics.

It is useful to predict the maximum radius achieved during the inertial stage since most of the size of a printed drop, which greatly affects the print quality, is obtained during this first stage. Prediction of the maximum spreading diameter has already been attempted by several authors. Most of them are empirical or semi-empirical. In this paper focus was given on the correlation described by Asai *et al.*[15] although other equations could also be used. The equation below, gives the maximum spreading ratio $D_{max}/ D_0$ as a function of the impact Reynolds and Weber numbers:

$$\frac{D_{\max}}{D_0} = 1 + 0.48 We^{0.5} \exp\left[-1.48 We^{0.22} \mathrm{Re}^{-0.21}\right] \quad (2)$$

where $D_{max}$ is the maximum diameter attained at the end of the inertial stage. Figure 2 gives the comparison between the experimental results and the above equation. The agreement is quite good since all the experimental points lay more or less close to the solid straight line which indicates perfect matching between data and correlation. It is also useful to note here that the experimental data included spreading results obtained with both macro and micro-drops. This equation can thus be used as an initial condition for different impacting conditions and for any kind of substrate either impervious or porous.

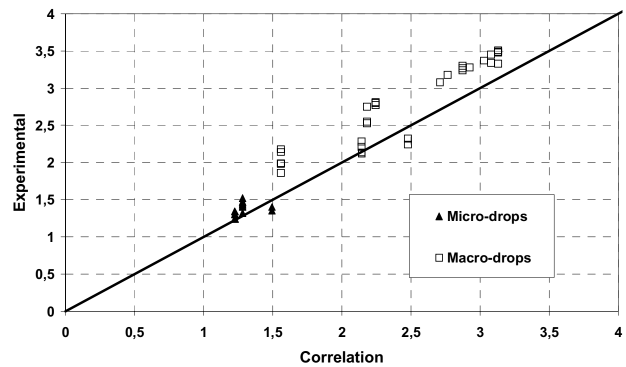



*Figure 2. Comparison between experimental data and correlation for the maximum spreading ratio*

## Collision and Axisymmetric Coalescence

Although there is a lot of literature[16,17] on the impact of a single drop onto a liquid film of uniform thickness, very few papers are really concerned with the collision process and coalescence of two drops.[14,9,18] The first paper summarizes the main observations without too many explanations on the hydrodynamic behavior during collision and coalescence. The other two papers give quantitative observations of coalescence of macro-drops (several millimeters) and of intermediate sized drops (0.6 mm) but no comparison is provided.

The experiments shown in this paper were performed with the following procedure. The first drop impinges onto the solid and reaches steady state, after some time, the second incoming drop impacts the first in a manner as close as possible to an axisymmetric landing. This procedure is followed both for macro-drops and for micro-drops with the devices shown in Figure 1a and 1b. It would also have been possible, using device 1b to impact on drops which are not yet in their equilibrium position but this has not been attempted in this study.

Figures 3a and 3b show the axisymmetric coalescence of macro and micro-drops. The inserts on the snap-shots give the time in ms for macro-drops and in μs for micro-drops and the horizontal lines give the position of the substrate. For the microdrops the dual exposure technique has been used to correct for the very short delay times: this leads to the variation in density of the observed second droplet compared to the first one.

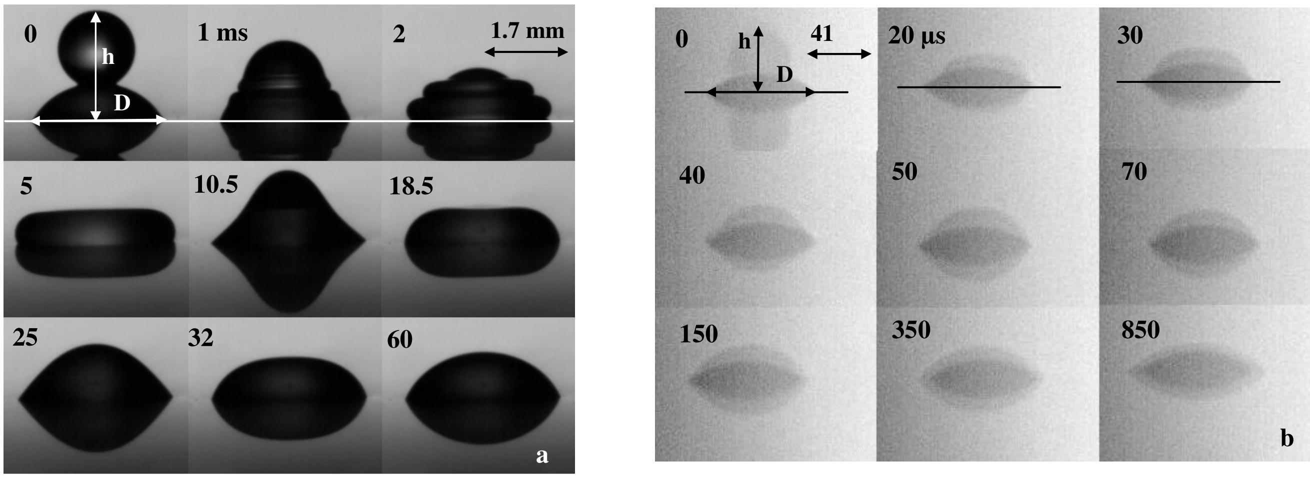



*Figure 3a and 3b. Axisymmetric coalescence of macro-drops and micro-drops*

The pictures on macro-drops of figure 3a were obtained with a glycerol-water-surfactant mixture on a hydrophobic silanized silicon wafer. The impacting drop diameter and velocity were 1.7 mm and 0.7 m/s. The dimensionless numbers for this experiment were Re = 48 and We close to 33. The height of the static drop was almost equal to 0.7 mm so at time 1 ms the snap shot shows the impacting drop touching the substrate. There was no movement of the contact line until 2 ms where one can note a change of the wetting angle to an advancing position. From then on, there was a merging of the two drops into one single mass which eventually could oscillate with a maximum being obtained at about 10.5 ms as shown on the picture. This however does not tell us more about the proper mixture of two colored inks.

The dynamics for the micro-drop axisymmetric coalescence are represented in figure 3b. The times at which the snap shots have been taken for this particular experiment were such that the dynamics of the coalescence at the initial stages cannot be captured but nevertheless the profiles are close to that observed with macro-drops. Because the Re number is even smaller than for the macro-drop experiment (16 instead of 48), and by reference to figure 2, it is expected that in this particular case the oscillations of the micro-drop obtained after coalescence will be smaller if any. The pictures at long times (350 to 850 µs) show the capillary spreading of a single mass of fluid. It is useful to note that when the drop is exposed more than once (multi-exposure shots) it is darker. The static drop is thus always much darker than the impinging one and could reveal to be useful for observation purpose.

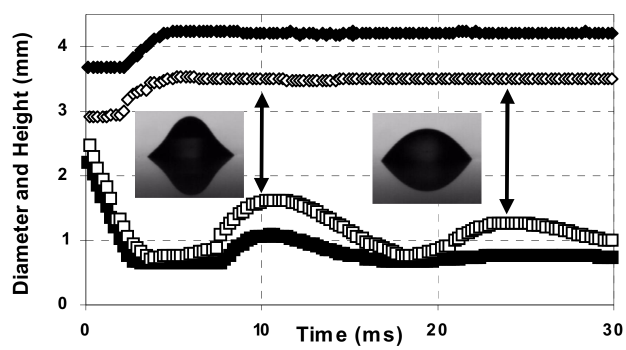



*Figure 4. Evolution of the transient diameter and heights*

Figure 4 above shows the evolution of the transient diameters and heights during macro-drop coalescence on oxidized silicon (solid symbols) and on silanized silicon (blank symbols) wafers. The diameters and heights were measured as shown on figures 3a and 3b. As expected, since the wetting is more pronounced, the initial diameter was larger for the static drop sitting on the oxidized substrate. The transient evolutions of the diameter were similar for the drops on both substrates but the height evolved differently due to receding behavior which was more important on the silanized surface. The profiles of the drop on the silanized substrate at two particular times (indicated by double-headed arrows) when the drop reaches a maximum in height during its oscillation are given as insert in the figure. This frequency of oscillation is that of a drop having a volume double to the impacting one which again is consistent with the fact that the two drops have merged into one single mass of fluid.

An essential requirement for the ink-jet process is the maximum size of the drop which is obtained when the collision process is terminated. This diameter can be predicted using the assumption of volume conservation. Based on the experiments, it is shown that when the impacting drop touches the sitting drop there is an evolution towards a profile close to a truncated sphere sitting on top of a cylinder (# 1ms on figure 3) and finally a perfect cylinder which is obtained around 3 ms. From then on there is no change of the diameter and the size is similar to that obtained on the 4th snap-shot of figure 3a (time #5 ms). The calculation using the above assumptions gives:

$$D_{\max}^{2} = \frac{2}{3}\left(\frac{D_0 - h}{h}\right)\left[\frac{3}{4}D_i^2 + (D_0 - h)^2\right] + D_{contact}^{2} \qquad (3)$$

where $D_{max}$ is the maximum diameter of the merged drop, $D_0$ the diameter of the impacting drop, $D_i$ is the diameter of the drop atop a cylinder (i.e. when the impinging drop intersects the solid substrate), $D_{contact}$ is the diameter of the static drop before impact and h the height of the final cylinder with diameter $D_{max}$. The approximate time at which the drop attains the maximum diameter is found by using the following assumptions:

- The impinging drop goes through the sitting drop without any loss in velocity.
- The initial velocity of the contact line is similar to that of the impinging drop.

The predicted time was very close to the experimental one which points that the drag experienced by the impinging drop was not too big (the height of the sitting drop is small) and the average calculated velocity is close to the actual one.

**Non-Axisymmetric Coalescence of Two Drops**

Although the coarsening mechanism of two or more drops is of critical importance in two-phase systems[10] the literature on the non-axisymmetric coalescence of two moving drops is quite limited.[9,19] Figures 5a and 5b given below show the essential features of the experiments that were done in this work for non-axisymmetric coalescence of macro and micro-drops.

Figures 5a represents the dynamics of coalescence which happens when a macro-drop is gently deposited close to a sitting drop. The coalescence proceeded by forming a connecting neck just at the time of deposition of the second drop and pursued up to almost 12 ms (snap shot #6) at which the height was maximum. As for figure 3a and 3b the time, shown in the inserts, is in ms for the macro-drop experiment and in µs for micro-drop one.

Figure 5b shows the non-axisymmetric coalescence for micro-drops with the second drop jetted at a velocity of 4 m/s and landing on a PET surface. The pitch (distance from drop center to drop center) was around 50 µm. Thanks to the gray level difference between static and incoming drops (multi-exposure shots) it is easily seen that the spreading occured in a one-sided way since the left end was almost immobile. Once again the late times are useful for obtaining the behavior of the surface tension dominated regime.

Figure 6 shows the profiles at different dimensionless times when the two drops given in figure 5a begin to merge. All the images of coalescence recorded at a rate of 4000 frames/s have been processed using the specific software detailed above. The processed profiles proved to be quite valuable in getting accurately the evolution of the height of the connecting neck as a function of time. The dimensionless time scales as:

$$t^* = t \times \sqrt{\frac{\sigma}{\rho D_0^3}} \qquad (4)$$

with the characteristic capillary time equal to 12.9 ms for this experiment.

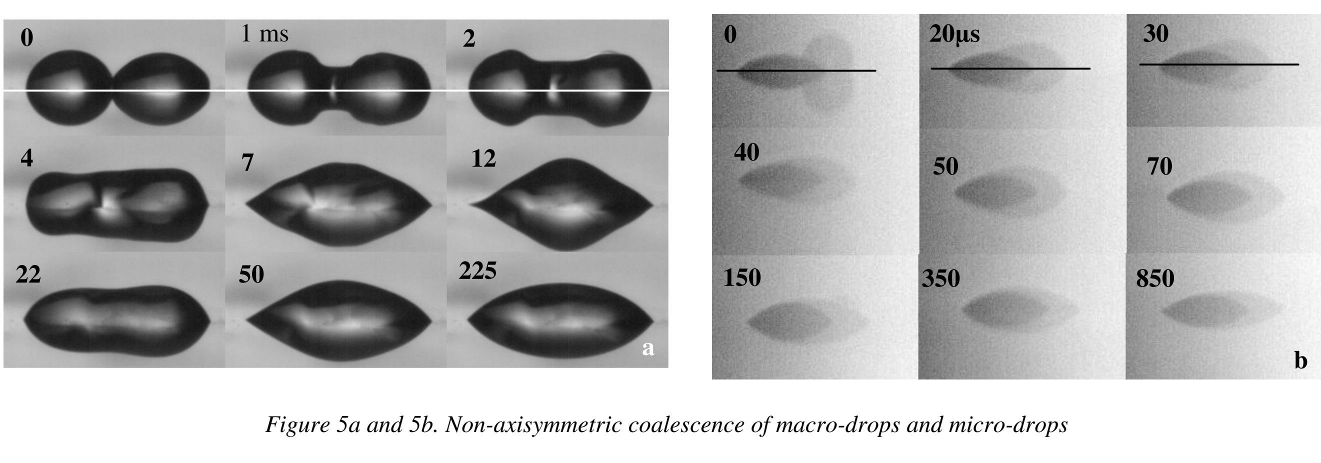



*Figure 5a and 5b. Non-axisymmetric coalescence of macro-drops and micro-drops*

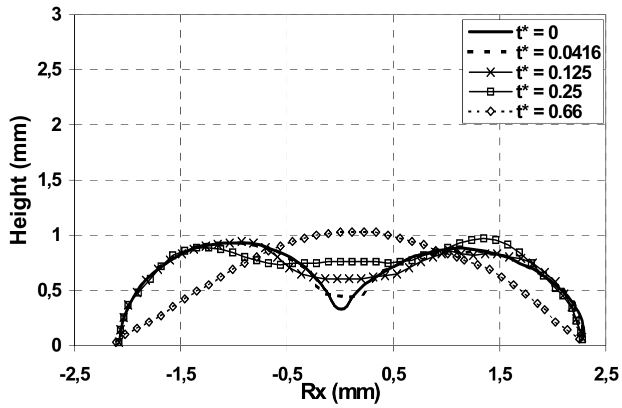



*Figure 6. Evolution of the height of the connecting neck during merging*

The evolution of the height was compared with predictions in the case of head-on collision of two drops with relative velocity U. A purely geometrical consideration[20] leads to:

$$h = \sqrt{\frac{UD_0 t}{2}} \qquad (5)$$

The experimental results compared quite favorably with the predictions if the collision velocity was taken to be 0.2 m/s. This value is consistent with the fact that the drop is gently deposited on the substrate.

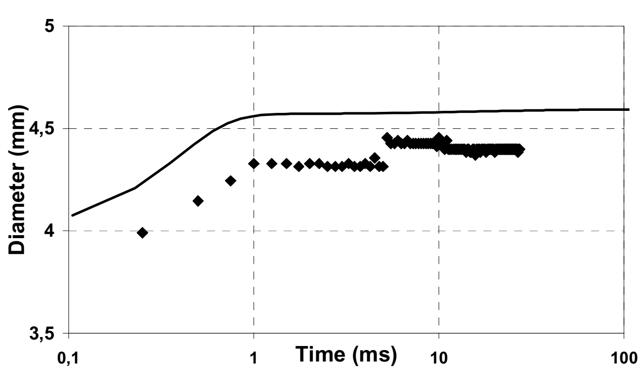



*Figure 7. Evolution of drop diameter during non-axisymmetric coalescence*

Finally figure 7, gives the comparison between experimental results (symbols) and predictions (solid line) for non-axisymmetric coalescence of macro-drops on silanized silicon wafer. The predictions were based on the fact that the spreading of the drop essentially happened on one side with the transient diameter given by the variational equation.[21] Indeed on the other side when the spreading drop encountered the static one, the net result was only an evolution of the height of the connecting neck (figure 6) without any movement of the formerly sitting drop. This model represents fairly well the expanding diameter and is probably sufficient for representing coalescence at low dimensionless numbers. It can be rendered more elaborate for predicting collision processes at high velocity by taking into account further developments reported elsewhere.[22] Other experiments are necessary in order to better assess the limits of the proposed model.

## Conclusions

Previous work on the coalescence of drops that occur in ink-jet printing has been fairly phenomenological in nature. In this study it was attempted to quantify the influence of various parameters and to improve the prediction capabilities of the coalescence process. In this context, two specific devices essential for collecting the data on macro-drops and on micro-drops were designed and built. Experiments with different sizes of drops for a variety of situations corresponding to actual matrix printing were performed. Predictions of drop behavior at various stages of the process were given while pointing out the simplifications in the models that have been considered. It is unambiguously shown in this work that drops put side by side coalesce and may or may not move over the substrate. This is a basic process taking place in every ink jet color printing technology. Color artifacts such as color shifts and mottle are very strongly related to the deviations in ideal dot positioning. Part of the cause of misplacement of dots can come from the jetting performance of the head. However, as shown in this paper also wel-positioned droplets may move on the substrate due to the presence of neighbouring droplets of other color inks. The basic processes causing these phenomena have been pinpointed, and this preliminary work should help in better understanding and defining the constraints for high quality multi-color ink jet printing.

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## Biography

**Guido Desie** got a Ph.D. at the K.U.Leuven, in the field of physicochemical analysis of enzymatic systems. In 1987, he joined Agfa Gevaert, Belgium, where he was involved in R&D of physical properties of film materials. From 1991, he was involved in R&D of Ink Jet and Toner based digital printing techniques. He is the co-author of about forty granted patent families mainly in the fields of Ink Jet and Toner Jet printing.