

Satellite drop formation during piezo-based inkjet printing

F. Marangon¹, W. K. Hsiao¹, G. Brenn², and C. Planchette^{2*}

¹Research Center Pharmaceutical Engineering, A-8010 Graz, Austria

²Institute of Fluid Mechanics and Heat Transfer, Graz University of Technology, A-8010 Graz, Austria

*Corresponding author: carole.planchette@tugraz.at

Abstract

The increasing use of ink-jet printing in manufacturing processes and especially its emergence in pharmaceutical industry has considerably risen the required control level. The formation of satellite drops causing off-target deposition and drop volume deviation remains poorly understood. Based on experimental data obtained over two print-heads and 16 inks made of water, ethanol and sugar in various proportions, we proceed to a careful analysis of the drop formation process and deduce a practical criterion to predict the formation of satellite drops. Our criterion results from the comparison of the kinetics of two simultaneous processes occurring right after the detachment of the liquid portion from the nozzle. One of them is the recoil of the liquid ligament into the main drop. Its kinetics is modeled using a modified Taylor velocity accounting for finite ligament length and viscous effects. For the other process, namely the ligament pinch-off close to the drop, two regimes must be considered, found for short and long ligaments. We explain this phenomenon by different distributions of the recoiling liquid and propose for each regime a scaling law providing good predictions of the experimental pinch-off times. Finally, we show that the comparison of the newly established theoretical recoil and pinch-off times successfully predict the formation of satellite drops.

Keywords

inkjet printing, drop, satellite, ligament

Introduction

In the last decades, driven by the apparent versatility of the technology, inkjet printing has emerged in application fields as diverse as printed electronics, biochips arrays and printed medicines [1, 2, 3, 4]. The latter application consists in manufacturing dosage forms by printing ink(s) which contains the active ingredient(s) onto appropriate substrates (paper, placebo pills. . .) which are then administrated to the patient [5, 6]. While such an approach opens routes to personalized medicines where the doses and drug combination are adjusted to each patient, it requires a very strict control of the printing process. In contrast to standard graphics arts, not only the position of the drops must be controlled but also their individual volume which is strongly modified by the presence of satellite drops.

The wide-spread use of piezo based drop-on-demand technology has resulted in the accumulation of empirical and theoretical knowledge about ink properties and jettability in relation to the driving signal (pulse) for various commercially devices. Since the 80's already, several experimental and numerical studies [13, 14, 15] showed that to keep a constant momentum of the printed liquid, an increase of ink viscosity or surface tension must be compensated by an increase of the driving pulse voltage or length. This understanding of Newtonian liquids printing has been reported in the form of operability diagrams based on Ohnesorge and Weber numbers [8, 9] which provides a strong basis to standard practical implementations. The functioning modes of drop-on-demand print-heads, pushing and squeezing, have been widely studied and numerically simulated, essential in industries research centers and have provided a sound understanding of the physical principles at stake [10, 11, 12].

The challenge caused by potential satellite drops remains however poorly documented. In a work by Dong et al. [16] a criterion to predict the recombination of primary and satellite drops is proposed but their formation is not predicted. Understanding this process asks about the stability of a finite and asymmetric liquid ligament. The stability of liquid jet has been deeply investigated [17] and several experimental numerical and theoretical studies have focused on the case of finite ligament covering a wide range of Ohnesorge number ($0.005 < Oh < 5$) and ligament aspect ratio ($1 < \lambda < 70$) [18, 19, 20]. Yet the ligaments were symmetric i.e. without the bulgy extremity that forms during inkjet and no consensus has emerged for the parameter range of interest ($0.01 < Oh < 0.5$; $5 < \lambda < 50$). Finally, Hoath et al. [7] proposed to apply the former results treating the ligaments produced by inkjet printing as symmetric ones of doubled aspect ratio - the bulge playing the role of a symmetric plane. The aim of our study is to analyze and model the fragmentation of liquid ligaments produced by inkjet printing accounting for both their finite and asymmetric characters. By filling this gap, we provide a new practical criterion enabling to predict satellites formation during ink-jet printing. Based on the experimental observation of the drop formation, we identify two competing processes: the recoil of the ligament in the main drop and the ligament pinch-off close to the main drop. Our approach then consists in comparing the kinetics of both processes for the geometry of the ligament at stake and to determine if satellite drops will appear or not.

The details of the experimental set-up and image analysis method are given in the section **Materials and methods**.

The measured time scales of each process are presented in the section **Results and discussion** where they are compared with the models we derive. This section finishes with the establishment of a criterion for satellite drops formation. Finally the paper ends with conclusions.

Materials and methods

Different liquids are mixed using ultra pure water (from TKA purification unit), ethanol (analytical grade, Merck GmbH) and sugar (Wiener Zucker, Agrana AG) in various proportions. After filtration with acetate syringe filters (pore size $1 \mu\text{m}$), the liquids are characterized. The density ρ is measured using a density meter (DSA5000M, Anton Paar GmbH), the surface tension σ is deduced from pendant drop experiments (Easy Drop, Krüss GmbH) and the viscosity μ is obtained in a double gap cylinder applying a shear rate of 200 s^{-1} (MCR300, Anton Paar GmbH, Austria). All measurements are performed at $22^\circ\text{C} \pm 3^\circ\text{C}$. The liquid composition, properties and jettabilities are presented in Table 1 together with $Oh_n = \mu/\sqrt{\rho\sigma d_n}$, the Ohnesorge number based on the inner nozzle diameter $d_n = 100 \mu\text{m}$.

Table 1. Liquid composition and properties measured at 25°C

Liquid number	Ethanol-Sugar (w:w) - (w:w)	ρ kg.m^{-3}	μ mPa.s	σ mN.m^{-1}	Jettable? yes/no	Oh_n
1	20 - 47.3	1170	41.00	29.4	no	0.699
2	0 - 45.0	1201	6.89	71.8	no	0.074
3	0 - 52.0	1237	13.9	71.5	yes	0.148
4	0 - 55.0	1255	20.7	72.8	yes	0.216
5	0 - 45.0	1266	27.2	72.9	no	0.282
6	1.0 - 45.0	1198	7.35	64.1	yes	0.084
7	6.0 - 21.0	1073	2.90	49.7	no	0.040
8	4.2 - 42.0	1175	6.84	52.0	no	0.087
9	3.5 - 48.5	1211	12.4	53.0	yes	0.155
10	3.5 - 53.0	1235	21.0	52.4	yes	0.261
11	3.5 - 55.0	1250	28.0	52.5	no	0.345
12	10.0 - 37.5	1141	6.68	41.4	yes	0.097
13	47.0 - 0	925	2.90	28.5	no	0.074
14	29.0 - 30.0	1066	7.01	29.9	yes	0.124
15	20.0 - 41.0	1135	13.9	31.5	yes	0.261
16	20.0 - 44.5	1151	19.8	29.4	no	0.699

The drops are produced using two piezo-based print-heads (Microdrop Technologies GmbH) of the same type and geometry (dispenser head MD-K-140; inner nozzle diameter $100 \mu\text{m}$). The printing temperature is set constant to 25°C and the shape of the electric signal driving the piezo is unchanged. The voltage, duration and frequency f of this pulse are varied covering $80 - 110 \text{ V}$, $20 - 40 \mu\text{s}$, and $100 - 1000 \text{ Hz}$, respectively. The drop formation is imaged using a standard camera (Guppy PRO, Allied Vision GmbH) and a stroboscopic illumination at f (LED). Different instants of the process are obtained by tuning the delay between the piezo- and LED- pulses. The typical spacial resolution is $1.05 \mu\text{m}/\text{px}$.

Image sequences similar to those of Fig. 1 (a-b) are analyzed with Matlab to deduce the following parameters (see Fig. 1 for corresponding sketches):

- t_{detach} the instant (counted from the start of the piezo pulse) when the ligament detaches from the nozzle ,
- if applicable, t_{recoil} the instant when the ligament has just recoiled in the main drop and $\tau_{recoil}^{exp} = t_{recoil} - t_{detach}$,
- if applicable, t_{po} the instant when the ligament pinches off the main drop and $\tau_{po}^{exp} = t_{po} - t_{detach}$,
- d the ligament diameter at t_{detach} ,
- l_0 and l the ligament lengths at t_{detach} and at $t > t_{detach}$, respectively.

Similarly to the work by Hoath et al. [7], we further define $\lambda = 2 l_0/d$ the initial aspect ratio of the ligament. Finally, using the temporal evolution of l , we observe a linear decrease and obtain the experimental recoiling velocity $V_{recoil}^{exp} = -dl/dt$, see Fig. 1 (c-d). Note that this velocity is found constant during the entire recoil and as long as the ligament does not pinch off.

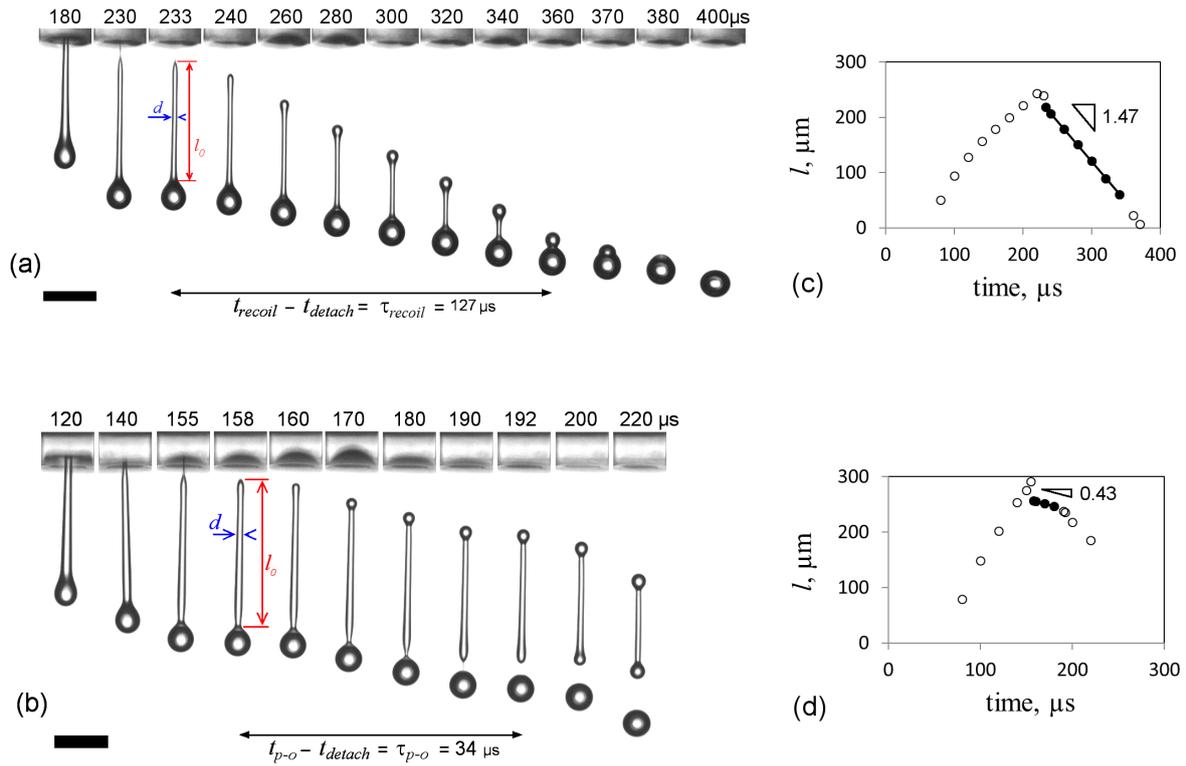


Figure 1. (a) Recoil observed with liquid No. 15 for 100 Hz, 99 V, 33 μs . (b) Pinch-off observed with liquid No. 6 for 100 Hz, 105 V, 36 μs . Ligament length as a function of time elapsed since the start of the piezo pulse for (c) liquid No. 15 for 100 Hz, 99 V, 33 μs (images of Fig. 1a) and (d) liquid No. 6 for 100 Hz, 105 V, 36 μs (images of Fig. 1b). The full symbols are used from the ligament detachment until its recoil or break-up. The recoiling velocity estimated from these points is found to be 1.47 and 0.43 m/s, respectively. The scale bars indicate 100 μm , the time elapsed since the start of the piezo pulse is indicated in μs .

Results and discussion

Ligament recoil

The recoiling velocities obtained for all experiments without satellites are plotted in Fig. 2 and compared with the classical and proposed models. The classical Taylor-Culick velocity is obtained by balancing the unsteady term of the Navier-Stokes equation $\rho \frac{du}{dt}$ with the pressure gradient, neglecting viscous effects. The surface tension being the driving force, the typical time scale is the capillary one $\sqrt{\rho D^3/\sigma}$, and the pressure to consider is the Laplace pressure σ/d . The jet being infinite, d is the only length scale at stake. This provides:

$$V_{Taylor}^{theo} = \sqrt{\sigma/\rho d}$$

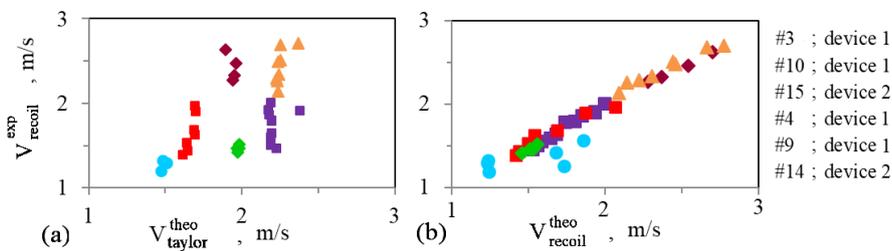


Figure 2. Experimental recoil velocity as a function of (a) V_{Taylor}^{theo} and (b) V_{recoil}^{theo} from Eq. 1 with $\alpha = 3.32$, $\beta = 3.25$ and $\gamma = 0.68$.

Despite its common use [7, 16] and as shown in Fig. 2a, V_{Taylor}^{theo} does not reproduce the experimental data V_{recoil}^{exp} . A similar discrepancy has already been documented for the retraction velocity of infinite liquid curtains [21]. In that case the deviation comes from the liquid viscosity. The latter governs the shape and dimension of the rim, and therefore its mass, which itself affects the inertia associated to its acceleration. The relevant dimensionless parameter is found to be the Ohnesorge number Oh , and the rim retraction velocity always tends toward the Taylor-Culick velocity for long times.

For the retraction of a liquid ligament similar to that observed in printing two modifications can therefore be proposed. First, the viscosity must be kept. Second, the ligament being finite in size, the pressure gradient must be estimated on a typical length scale l_0 . Three terms must then be accounted for: the unsteady term $\rho \frac{du}{dt}$ scaling as $\rho V_{recoil} \sqrt{\sigma/\rho D^3}$, the pressure gradient estimated by σ / dl_0 , and the viscous term taken as $\mu V_{recoil}/d^2$. Doing so provides the modified Taylor-Culick velocity:

$$V_{recoil}^{theo} = \beta \sqrt{(1 + \alpha Oh) \sigma d / \rho l_0^2} + \gamma \quad (1)$$

where α , β and γ are constants which can be adjusted to obtain a quantitative agreement. The results obtained using $\alpha = 3.32$, $\beta = 3.25$ and $\gamma = 0.68$ are plotted in Fig. 2b. The agreement is very good for most of the cases. Indeed, three points are deviating (liquid No. 14) which correspond to short ligaments ($\lambda \leq 22$).

The recoiling velocity being constant during the recoil phase (Fig. 1c-d) and its modeling being very satisfying using V_{recoil}^{theo} (Eq. 1, Fig. 2b), the recoiling time can be estimated by $\tau_{recoil}^{theo} = (l_0 - 2d) / V_{recoil}^{theo}$. Redefining the constants, this expression becomes at first order:

$$\tau_{recoil}^{theo} = \tilde{\beta} l_0^2 \sqrt{\frac{\rho}{\sigma d}} (1 - \tilde{\alpha} Oh) + \tilde{\gamma} \quad (2)$$

The theoretical predictions of the recoil time obtained using $\tilde{\alpha} = 0.6$, $\tilde{\beta} = 0.058$ and $\tilde{\gamma} = 25 \cdot 10^{-6}$ are plotted in Fig. 3, showing excellent agreement. The deviations observed for short ligaments (see Fig. 2b, liquid No. 14) are not visible any more indicating some compensation process.

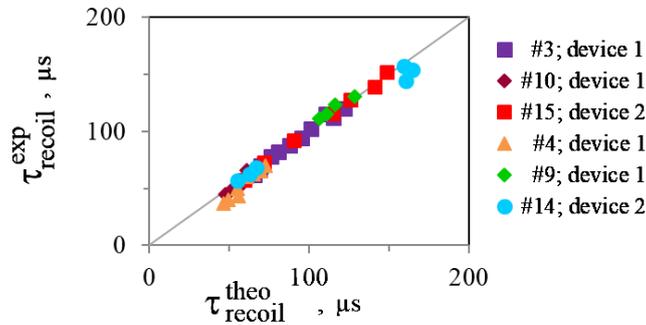


Figure 3. Experimental recoil time τ_{recoil}^{exp} as a function of τ_{recoil}^{theo} the theoretical predictions obtained using Eq. 2 with $\tilde{\alpha} = 0.6$, $\tilde{\beta} = 0.058$ and $\tilde{\gamma} = 25 \cdot 10^{-6}$.

Ligament pinch-off

Simultaneously to the ligament recoil, the development of its pinch-off takes place. This process is caused by a local Laplace pressure gradient which drains the liquid away thus enhancing the draining flux and potentially leading after some time to the ligament pinch-off. This process is also at the origin of the well-known Plateau-Rayleigh instability of an infinite inviscid liquid jet. For our system, the pinch-off is expected to happen close to the main drop where the changes of interface curvature are important. Classically for the range of Oh at stake, the viscosity cannot be neglected and the typical time scale of this process is obtained by balancing the capillary pressure with the viscous stress. This provides the well-known visco-capillary time scale : $\tau_{visc} = \mu d / \sigma$ used by Hoath et al. to predict the formation of satellite drops during inkjet printing [7]. Note that the finite length of the ligament l_0 is not accounted for in this time scale. The comparison of τ_{visc} with the experimental measurements is presented in Fig. 4.

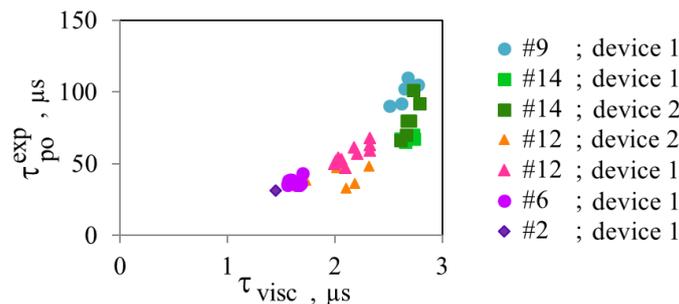


Figure 4. Experimental pinch-off time τ_{po}^{exp} as a function of τ_{visc} the typical capillary viscous time scale used by Hoath et al. [7]. Beside a matching trend, the agreement remains very poor.

The agreement is quite poor and to better understand the limit of this approach, it is interesting to look at the variations of τ_{po}^{exp} with the ligament length or directly with λ . As it can be seen in Fig. 5a for a given liquid, τ_{po}^{exp} is varying with λ and these variations are not monotonic. Instead, the pinch-off time of short ligaments increases with λ while an opposite trend is found for long ligaments. This indicates that the finite size of the ligament cannot be neglected. We explain this observation considering the recoil process which occurs simultaneously to the pinch-off. This recoil causes a liquid transfer toward the main drop with a typical velocity V_{recoil} . This recoiling liquid potentially delays the pinch-off by accumulating at the neck. Using the expression given by Eq. 1 provides at first order a kinetic pressure $P_{kin} \approx (1 + \alpha Oh)^2 \sigma d / l^2$. This liquid transfer is opposed by the capillary pressure that squeezes the neck causing the pinch-off. It can be evaluated by $P_{cap} = \sigma / d$. Thus, if $P_{kin} > P_{cap}$ which is equivalent to $\lambda < K(1 + \alpha Oh)$, the liquid flux is drained into the main drop, no liquid accumulates and the pinch-off is not significantly delayed. As observed in Fig. 5c, the ligament remains cylindrical (constant diameter except at the pinching-off) during the whole process. In contrast, for if $P_{kin} < P_{cap}$ and thus $\lambda > K(1 + \alpha Oh)$, the inertia of the recoiling liquid cannot overcome the capillary squeezing; the liquid accumulates at the neck forming a *bulp* (see Fig. 5b) which is expected to slow down the pinch-off. Here K is a constant which needs to be determined. Using $K = 19$ we obtained the limits plotted as vertical bars in the Fig. 5a) which seem - despite the restricted number of points - compatible with the experiments. Note that the asymmetric character of the ligament is essential to observe the liquid transfer from one side (the ligament tail) to the other one (the bulge, precursor of the main drop). Having this finding in mind, two models must be proposed for large and small λ , respectively.

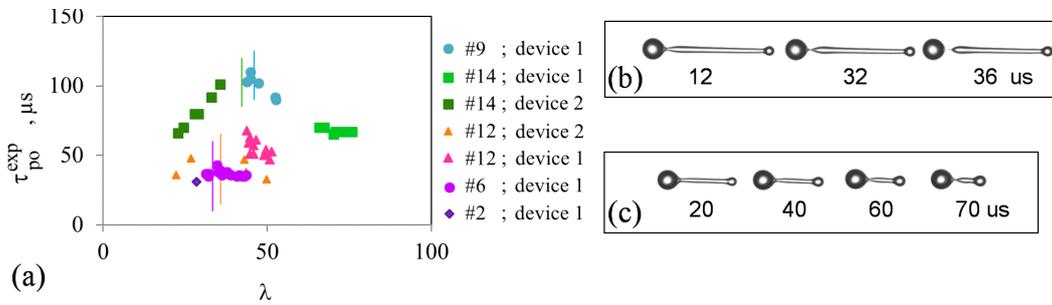


Figure 5. (a) τ_{po}^{exp} as a function of λ for all experiments where pinch-off is observed. For a given liquid, a maximum is observed for intermediate values of λ . The vertical lines represent $K(1 + \alpha Oh)^2$ with $K = 19$. (b and c) Pictures of the pinch-off process with the time elapsed from the ligament detachment in μs ; (b) liquid No. 6, printed with 105 V, 37 μs pulse duration and $f=100$ Hz, $\lambda = 40.5$ and (c) liquid No. 14, printed with 84 V, 33 μs pulse duration and $f=100$ Hz, $\lambda = 29.0$. For $\lambda = 40.5$ a *bulp* appears close to the neck which is not the case for $\lambda = 29.0$

Pinch-off, large λ

An effective ligament diameter d^* must be computed that accounts for the recoil flux feeding the neck. The volume of recoiled liquid varies with the time elapsed from detachment t as $\Omega_{recoil}(t) = \pi d^2 V_{recoil} t / 4$. This volume accumulates at the neck forming as a first approximation a cylinder of volume $\Omega_{visc}(t) = \pi d^{*2} l_{visc}(t) / 4$ where $l_{visc}(t) = \sqrt{\mu t / \rho}$ is the thickness of the viscous boundary layer. Applying volume conservation between $\Omega_{recoil}(t)$ and $\Omega_{visc}(t)$ leads to $(d^*/d)^2 = V_{recoil} \sqrt{\rho t / \mu}$. The theoretical pinch-off time for large λ , τ_{po}^{large} is thus obtained using τ_{visc} where d is replaced by $d^* \left(\tau_{po}^{large} \right)$. Further replacing V_{recoil} by Eq. 1 and simplifying the expression to the leading order in Oh provides:

$$\tau_{po}^{large} \propto \tau_{visc} ((1 + \alpha Oh) / \lambda)^{2/3} \quad (3)$$

The predictions are compared to the experimental data in Fig. 6a. Full and empty symbols correspond to λ larger, respectively smaller, than 35. As expected, the agreement is very good for the large ligaments but not for the small ones for which a different model is developed below.

Pinch-off, small λ

For short ligaments, the recoiling flux is not accumulating in the neck. The pinch-off time can be estimated by balancing the capillary pressure gradient σ / dl_o with the viscous losses $\mu u / d^2$ where u is the axial velocity component. This provides a modified viscous-capillary time scale $\mu d \lambda / \sigma$. For these short ligaments, the pinch-off process is so fast that the ligament diameter is already locally reduced when it detaches from the nozzle. Thus the ligament diameter is better evaluated by a constant D close to the nozzle inner diameter than by d obtained from the image analysis at t_{detach} . Thus we propose :

$$\tau_{po}^{small} = \mu D \lambda / \sigma \quad (4)$$

The theoretical results τ_{po}^{small} estimated with $D = 120 \mu m$ are compared to experimental data τ_{po}^{exp} in Fig. 6b where the empty symbols correspond to points for which $\lambda < 35$ and full symbols for the others. As expected the agreement is very good but only for the small ligaments.

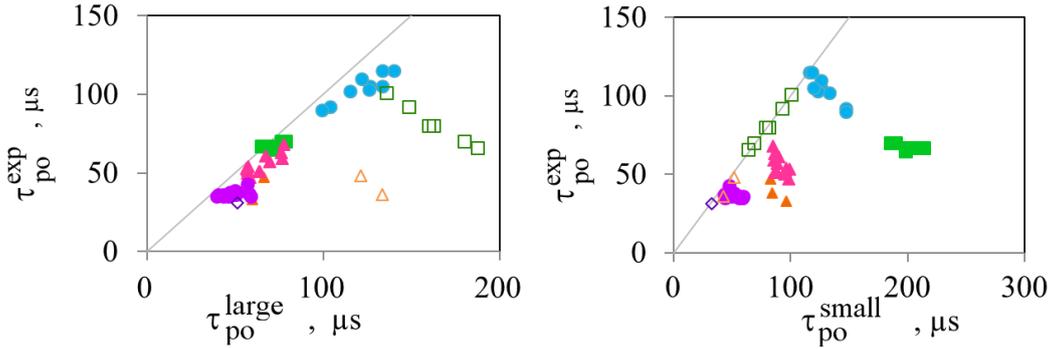


Figure 6. Experimental pinch-off time as a function of (a) τ_{po}^{large} given by Eq. 3 and (b) τ_{po}^{small} given by Eq. 4. Empty symbols are points obtained for $\lambda < 35$, full symbols correspond to $\lambda > 35$. The proposed models work very well for their respective application domains.

Criterion for satellite drop formation

Finally, we propose to compare the kinetics of the recoil and the pinch-off to deduce the outcome of a printing event. To do so, we calculate $T = \tau_{po}^{theo} / \tau_{recoil}^{theo}$. Here τ_{po}^{theo} is taken as τ_{po}^{small} (Eq. 4) and as τ_{po}^{large} (Eq. 3) for λ smaller and larger than 35, respectively. τ_{recoil}^{theo} is given by Eq. 2. The results are plotted in Fig. 7 and show that T is well-suited to predict the outcome of a printing process. Furthermore, to estimate T , only the liquid properties and the initial shape of the ligament l_0 and d are needed, which makes it very versatile to use.

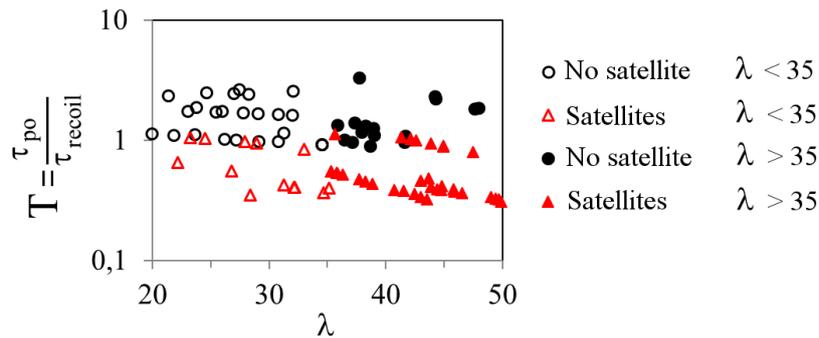


Figure 7. T as a function of λ . Open symbols: small λ , full symbols: large λ . The circles correspond to observed recoil (no satellite) while the triangles are used when pinch-off was recorded (satellite). The proposed parameter T is well separating these two kinds of events.

Conclusions

This experimental study has shed light on the evolution of the drop and the ligament produced by an ink-jet print-head. As expected, two competing processes have been identified corresponding to the recoil of the ligament into the main drop and to its pinch-off of from the main drop. The kinetics of these two processes have been investigated in detail using 16 ink formulations and 2 printing devices. We demonstrated that, despite its common use, the Taylor-Culick velocity is not appropriate to describe the velocity at which the ligament recoils in the main drop. Instead, one must account for the liquid viscosity and, more importantly, for the finite size of the ligament via its initial length. The proposed model of the recoil velocity was successfully used to obtain a prediction of the recoil time (from the detachment of the ligament from the nozzle) based only on the liquid properties and the initial dimensions of the ligament. Further, we proved that the pinch-off time of such asymmetric and finite ligaments cannot be described without account for the liquid transfer from the tail to the main drop. It is noteworthy that this transfer is caused by the geometry of the expelled liquid entity and can therefore not be modeled accurately without considering it carefully. More precisely, two regimes have been evidenced. For small initial aspect ratio of the ligament, the liquid transfer does not significantly affect the pinch-off process and the pinch-off time increases with the ligament length. In contrast, for large initial aspect ratio of the ligament, the kinetic pressure of the recoiling flux cannot overcome the capillary pressure squeezing the neck: the liquid accumulates at the neck and delays the pinch-off. Two corresponding scaling laws are proposed that agree very well with the data on their respective domain of validity. Finally, we compared the predicted recoil and pinch-off time scales and showed that their ratio efficiently predicts the formation of satellite drops during ink-jet printing. This ratio can be computed from the liquid properties and the initial dimensions of the liquid ligament only. Thus, it is of high practical importance and can be used to tune the printing parameters accordingly for both small and long ligaments.

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