Breakup Length of Electrified Liquid Jets: Scaling Laws and Applications

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The growth rate of infinitesimal perturbations in electrified jets exhibits different behavior at the viscid and inviscid limits. Using Saville’s approach to estimate the growth rate for perturbations in the long-wavelength limit and by ignoring the effects of gravity, we derive two scaling laws for the jet breakup length in two regimes of the Taylor-cone mode. Our experimental measurements show clear dependency of the jet length on the flow rate; however, change of the applied voltage appears to affect only the cone angle but not the jet itself. The experimental data agree excellently with our theoretical model in both cases. On the basis of the jet diameter, the transition between viscid and inviscid limits appears to occur at an electric Reynolds number of approximately 5. Finally, we show how to enhance the quality and the resolution in electrostatic-inkjet-printing applications by setting the printing distance less than the jet length and predict the line width as a function of the operational parameters.

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I. INTRODUCTION

The breakup of liquid jets in the capillary regime, which is caused by the growth of perturbations, has been a focus for many scientists over several decades. The first attempt to explain the capillary instability was undertaken more than 100 years ago by Rayleigh [1]. He showed that axisymmetric disturbances grow downstream until a breakup happens for uncharged inviscid liquid jets if the wavelength exceeds the circumference of the jet. For gravitational jets, Javadi et al. [2] introduced a theory supported by experiments that explains how viscosity tends to increase the jet intact length. If the jet is produced out of a liquid meniscus as a result of application of an electric field, the so-called cone-jet mode or Taylor-cone mode, the instability mechanism will be different. Local stability analysis has been used extensively to understand the breakup mechanism of electrified jets since the analysis of Taylor [3]. Taylor showed that the electric charge stabilizes the jet for long-wavelength perturbations, correcting errors in Basset’s analysis [4]. Saville [5] adopted a linear theory to estimate the growth rate of both axisymmetric and nonaxisymmetric perturbations in electrified jets taking into account the effect of viscosity and inertia. There have also been studies related to the nonlinear breakup of an electrified jet, but in which details of the breakup process cannot be captured with simplified one-dimensional models. As an example, Conroy et al. [6] used a nonlinear slender jet model to investigate ionic surfactants and electrokinetic influences on the size of satellite droplets produced at jet breakup; however, their numerical simulations suggest that the effect on the breakup time is weak. In contrast, Collins et al. [7] solved the full Navier-Stokes equations assuming a perfectly conducting cylindrical jet, which is bounded by a concentric ground electrode, and they showed that nonlinear effects tend to delay the breakup process, and hence increase jet length. The configuration investigated by Collins et al. is, however, different from that of investigated by Saville [5], who assumed a ground electrode at infinity. Knowing the length of electrified jets is crucial in many fields, such as analytical chemistry, fabrication of nanofibers and biological materials, the textile industry, and continuous inkjet printing. Although much research has been conducted into the breakup of electrified jets for inviscid liquids [8–10] and highly viscous liquids [7,11–13], none of these have introduced a quantitative estimation for the breakup length. In this work, we propose two scaling laws to predict the breakup length of an electrified jet in the viscid and inviscid limits on the basis of Saville’s approach to estimate the growth rate of perturbations in the long-wavelength limit. These scaling laws are set in the context of a systematic experimental investigation. We then use these scaling laws to print grids from nanoparticle inks with high quality and high resolution, giving an accurate prediction of the width of the printed lines.
II. SCALING OF THE JET LENGTH IN THE VISCID LIMIT

If we consider a liquid meniscus emanating from a metal nozzle at a voltage difference $V$ that is applied between the nozzle and a counter-electrode plate, electric charges will accumulate on the liquid surface, producing stresses that deform the meniscus into a cone shape. Because of the singularity created at the apex, a fine jet, much smaller than the nozzle, can be produced from the cone. At relatively low voltage, a pulsating mode occurs in which the ejection is periodic. If the voltage is increased above a critical value, a steady jet will be emitted, which then breaks up at some point downstream (Taylor-cone mode). The jet produced in this mode typically breaks up into either an axisymmetric fashion (varicose) or a nonaxisymmetric fashion (whipping). Our focus in this study is on the intact length of the jet in the Taylor-cone mode when the breakup is axisymmetric. The parameters that mainly characterize the problem are the density $\rho$, dynamic viscosity $\mu$, surface tension $\gamma$, electrical conductivity $K$, electrical permittivity of the environment $\varepsilon_0$, relative electrical permittivity $\beta$, flow rate $Q$, and jet radius $R$. Here the jet diameter is assumed to be constant as the thinning of the jet is negligible. If we consider an approximation of a linear theory and use the viscous time $t_v = \mu R / \gamma$ and the viscous velocity $v_v = \gamma / \mu$ as characteristic parameters and if we ignore the effects of gravity, the nondimensional equations that govern the jet can be written as follows:

$$\nabla \cdot \mathbf{v} = 0,$$

$$\frac{1}{\text{Oh}^2} \frac{\partial}{\partial t} \mathbf{v} = -\nabla p + \nabla^2 \mathbf{v},$$

where the Ohnesorge number $\text{Oh} = \mu / \sqrt{\rho \gamma R}$. The infinitesimal perturbations $\{\tilde{u}, \tilde{v}, \tilde{w}, \tilde{p}\}$ for the velocity and the pressure that propagate through the jet, written in cylindrical coordinates $(r, z)$, read

$$\{\tilde{u}, \tilde{v}, \tilde{w}, \tilde{p}\} = \{u, v, w, p\}(r, z) \times \exp(i\omega t),$$

where $u, v, w, p$ are the velocities and the pressure amplitude, and $\omega$ is the growth rate. Saville solved Eqs. (1)–(3) together with Maxwell’s equations, assuming that the ground electrode is at infinity, to obtain a dispersion relation. Considering that the most-unstable axisymmetric perturbation corresponds to an infinitely long wavelength, he then estimated the growth rate for a viscous jet breaking up through a varicose mode [5] to be

$$\omega \sim \frac{\gamma}{6\mu R} \left(1 - \frac{R E_0^2}{4\pi^2}\right),$$

where $E_0$ is the electric field strength at the surface of the jet. In Eq. (4), the term in parentheses has to be less than 1 for the jet to have axisymmetric breakup, which is the case for all the experimental data in this study.

We now extend this approach to identify the breakup length of the jet. To do this, Eq. (4) can be used to estimate the breakup time $t_b$ for the jet, where $t_b \sim 1/\omega$, from which, if $v_j$ is the jet velocity, the breakup length of the jet is

$$L_b \sim v_j t_b.$$  \hspace{1cm} (5)

Knowing the jet velocity $v_j \sim Q/R^2$ from the continuity equation and the electric field strength $E_0$ estimated from $E_0 = (\rho \gamma^2 K^2/\varepsilon_0)^{1/6}$ [14], we see the only unknown in Eq. (5) is the jet radius. Gañán-Calvo [15] suggested a scaling law for the jet radius. This was subsequently validated experimentally in Ref. [16]:

$$R = \left(\frac{\rho Q^2 \varepsilon_0}{\gamma K}\right)^{1/6}.$$  \hspace{1cm} (6)

This relation is limited to the following conditions:

$$\alpha_\rho \gg \alpha_\mu^{1/4}, \quad \alpha_\rho (\beta - 1) \gg 1,$$

where $\alpha_\rho = \rho KQ/\gamma \varepsilon_0$ and $\alpha_\mu = K^2 \mu^3 \varepsilon_0^{3/2} \gamma$. These conditions hold for the experimental scope considered in this study.

From Eqs. (4)–(6), the scaling for the jet breakup length can be written as

$$L_b = k_v \frac{6\mu}{Z_v},$$  \hspace{1cm} (8)

where $k_v$ is a constant and

$$Z_v = \left[\frac{(\rho \gamma^2 \varepsilon_0)}{Q^2 K}\right]^{1/6} - \frac{1}{4\pi} \left(\frac{\rho^2 \gamma K}{\varepsilon_0}\right)^{1/3}.$$  \hspace{1cm} (9)

III. EXPERIMENT

To validate this scaling law, we measure the jet breakup length for different liquids with a wide range of properties. The jet length $L_b$ is measured from the point where the change of the cone jet diameter starts to be very small ($d_d/dz < 0.4$, where $z$ represents the coordinate in the axial direction). The liquids used are ethanol, ethylene glycol, 2-propanol, 1-methoxy-2-propanol, propylene carbonate, triethylene glycol, dipropylene glycol, glycerol, and a mixture of 10% water and 90% glycerol by weight (see Table I). For each test, ten measurements of the jet length are taken, and the standard deviation from the mean value in all cases is less than 10%. Sodium iodide is added to some of the liquids to provide a broad range of conductivity from 0.1 to 10 $\mu$S/cm. Conductivity higher than 10 $\mu$S/cm is not considered in this study as at higher conductivity the jet tends to have nonaxisymmetric breakup.

The experimental setup consists of a tapered metal nozzle of outer diameter at the tip $d_n$ ranging between 30
and 250 μm. In all cases the fluid meniscus wetted the outer edge of this nozzle. This nozzle is connected to high-voltage power supply (Fug HCP 14-6500), which is able to supply a voltage difference of up to 10 kV between the nozzle and a ground plane. A thin metal plate is used as the ground electrode. This plate has a 2-mm hole at the center to avoid flow accumulation on the electrode surface. The separation between the nozzle and the plate is set to be 3 mm, this separation thus being much larger than the jet diameter. In view of this, the assumption of having the ground electrode at infinity in our analysis is justified. Images of the jet are captured with a CCD camera (uEye UI-2230-C, IDS) with a microscopic lens (Optem Zoom 125) and a flash lamp (High-Speed Photo-Systeme) with an illumination time of 20 ns. The magnification of the microscopic lens can be adjusted to give a resolution ranging from one to four pixels per micrometer. A waveform generator (HP 33120A, Hewlett-Packard) and a delay-pulse generator (DG535, Stanford Research Systems) are used to synchronize the triggering pulses of the camera with the flash lamp. A diffuser is placed between the light source and the nozzle to ensure uniform lighting. The images are processed with image-analysis software (ImageJ) to measure the jet intact length.

The working fluid is fed into the nozzle by use of either a syringe pump (Harvard Apparatus PHD4400) or a height difference of the liquid level between the nozzle tip and a reservoir, with a flow rate \( Q \) between 0.0006 and 0.1 ml/min. The flow rate is measured with a nanoflow sensor (model N-565, Upchurch Scientific). Fig. 1 shows the breakup length of the jet as a function of the flow rate for individual liquids. As one may expect, increase of the flow rate and/or the viscosity leads to a longer jet. Also, for the same solvent at different conductivity in Fig. 1, it may be seen that increased conductivity delays the breakup. This effect can be elucidated in terms of an increase of the electric field strength on the jet, which is known to stabilize conducting jets [17]. Also, the increase of conductivity yields a reduction of the jet diameter and, therefore, an increase of the Ohnesorge number. This increase would stabilize the jet too. On the other hand, we find that the voltage difference has no direct influence on the jet length itself. This can be explained simply in terms of insensitivity of both the field near the apex of the cone and that on the jet to the geometric conditions of the electrodes that create the field [18]. However, from our observations it is evident that increasing the voltage does lead to an increased cone angle, and hence it is the cone length in the axial direction that declines (see Fig 2). In addition, from our data it appears that the diameter of the nozzle does not have any influence on the jet breakup length. This is because the nozzle diameter is much greater than the jet diameter, and hence the jet breakup itself is a local mechanism. These observations are consistent with the scaling proposed in Eq. 8, where the breakup length is a function only of the flow rate and the liquid properties.

![Graph showing breakup length vs flow rate for different liquids.](image)

**FIG. 1.** Jet length as a function of the flow rate for liquids with different viscosities and conductivities. The nozzle diameter is 250 μm. EG0.78, ethylene glycol 0.78 μS/cm; EG4, ethylene glycol 4 μS/cm; Prop0.1, propanol 0.1 μS/cm; Prop2, propanol 2 μS/cm; W+90%G, 10-wt% water and 90-wt% glycerol.
Figure 2 shows the measured breakup length normalized by $d_n$ for a total of 190 experiments against the theoretical model in Eq. (8). Excellent collapse for most of the data (open symbols) occurs when the constant $k_v$ has a value of 7, with a maximum standard deviation of 16.8%, for a data range that spans almost 3 orders of magnitude; this provides strong support for the proposed scaling law. The solid symbols, which diverge from the theoretical prediction, have electric Reynolds number $Re > 5$. The electric Reynolds number is defined here as $Re = [(\rho^3Q^2\gamma)/(\mu\varepsilon_0)]^{1/6}$. This suggests that the scaling is limited to a low Reynolds number, which is consistent with our approach because the growth rate used in this analysis is valid only when viscosity is dominant.

### IV. SCALING OF THE JET LENGTH IN THE INVISCID LIMIT

To predict the breakup length for inviscid jets, one needs to estimate the growth rate when viscous effects are negligible in comparison with the inertia of the jet. If we replace the scale factors for time and velocity with $t_c = (\rho R^3/\gamma)^{1/2}$ and $v_c = (\rho R/\gamma)^{1/2}$, respectively, in the inviscid limit, then Eq. (2) can be rewritten as

$$\frac{\partial}{\partial t} v = -\nabla p + \text{Oh}\nabla^2 v. \quad (10)$$

Following Saville’s approach, the general equation for the growth rate [5] is

$$\frac{\hat{\omega} I_m(\xi)}{\xi I'_m(\xi)} = 1 - m^2 - \xi^2 - \frac{R\varepsilon_0 E_0^2}{4\pi\gamma} \left[ 1 + \frac{K'_m(\xi)}{K_m(\xi)} \right]. \quad (11)$$

where $\hat{\omega}$ is the growth rate in nondimensional form, $I_m$ and $K_m$ are the modified Bessel functions, $m$ is the azimuthal wavenumber, and $\xi$ is the axial wavenumber. If viscosity effect is negligible in comparison with the effect of inertia, $\hat{\omega}$ can be defined as $\omega(\rho R^3/\gamma)^{1/2}$. For axisymmetric motion, the general equation reduces to

$$\frac{\omega R^3}{\gamma} = \frac{1 - \xi^2 - \frac{(R\varepsilon_0 E_0^2/4\pi\gamma)}{2 \left[ \xi I_0(\xi)/I_1(\xi) \right]^2 - \xi^2 - 1}}{1}. \quad (12)$$

For long wavelength ($\xi \rightarrow 0$), where the most-unstable axisymmetric breakup is observed, the growth rate in the inviscid limit can be written as

$$\omega^2 \sim \frac{\gamma}{6\rho R^3} \left( 1 - \frac{R\varepsilon_0 E_0^2}{4\pi\gamma} \right). \quad (13)$$

This is similar to Rayleigh’s expression for an inviscid jet produced from a charged liquid sphere [19]. On the basis of Eq. (13), it is easy to estimate the breakup length in the inviscid limit as

$$L_b = \frac{k_i 6}{Z_i}. \quad (14)$$
where

$$Z_i = \left[ \left( \frac{\gamma^5 \varepsilon_0}{\rho^3 Q^2 K} \right)^{1/6} - \frac{1}{4\pi} \left( \frac{\gamma K}{\rho^3 Q^5 \varepsilon_0} \right)^{1/3} \right]^{1/2}. \quad (15)$$

Comparison of our experimental data in the inviscid limit with the new scaling in Eq. (14) this shows excellent agreement with a fitting factor $k_i$ of 7 (see Fig. 4). The maximum standard deviation, with data over 2 orders of magnitude, is 15%. It should be emphasized that the collapse in Fig. 3 happens only for the data for which $Re > 5$. This observation provides cogent evidence of the credibility of the two models, wherein each model is valid only within the limits proposed in the analysis.

In the following section, we illustrate the importance of our models for improving the electrostatic-inkjet-printing technology.

### V. PRINTING METAL GRIDS WITH HIGH QUALITY AND HIGH RESOLUTION

Direct printing of metal grids with good optical and electrical performance shows great potential to fabricate transparent conductive electrodes, especially in the case when high resolution of metal grids is preferred to provide invisibility and when flexibility of electrodes is needed for flexible devices.

In this section, the objective is to print uniform grid lines from nanoparticle inks with the minimum possible width to enhance the resolution. We use silver and copper nanoparticle inks with the physical properties given in Table II. The printing system consists of a glass nozzle with a tip diameter of 50 $\mu$m. A metal needle is inserted inside the glass-tube nozzle to work as an electrode. This electrode is connected to a high-voltage power supply. The glass nozzle is fixed to the print-head body, which has two inlets: one inlet is for the electrode; the other inlet is connected to a tube with a reservoir at its end to introduce the ink. A high-speed $X$-$Y$ stage (AeroTech PRO 165 LM) is used to move a glass substrate placed on a metal plate horizontally.

<table>
<thead>
<tr>
<th>Ink</th>
<th>$\rho$ (kg/m$^3$)</th>
<th>$\mu$ (kg/m s)</th>
<th>$\gamma$ (mN/m)</th>
<th>$K$ (µS/cm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>55-wt% silver nanoparticles</td>
<td>5700</td>
<td>1</td>
<td>40</td>
<td>0.01</td>
</tr>
<tr>
<td>50-wt% copper nanoparticles</td>
<td>4980</td>
<td>0.8</td>
<td>35</td>
<td>0.05</td>
</tr>
</tbody>
</table>

In Fig. 5, copper lines printed at a printing distance of (a) 1 mm (b) 0.8 mm, and (c) 0.4 mm. The jet breakup during the printing is shown in (d).
The width $W$ is a function of the cross-section area of the printed line $A_l$ and the contact angle $\theta$. The area can be calculated as a segment of circle as follows:

$$A_l = \frac{r^2}{2} (2\theta - \sin 2\theta),$$

where $r = W/2 \sin \theta$.

If we assume that the stage velocity is set to be equal to the jet velocity as discussed earlier, then the line cross-section area should be equal to the cross-section area of the jet $A_j = \pi d_j^2/4$. Finally, the line width can be expressed in terms of the jet diameter and the contact angle as follows:

$$W = d_j \sin \theta \sqrt{\frac{2\pi}{2\theta - \sin 2\theta}}.$$

To print a uniform silver grid with lines 10 $\mu$m in width, we need to calculate the corresponding jet diameter from Eq. (17). The measured contact angle between the silver ink and the glass substrate is 14°; as a result, the jet diameter should be 2.2 $\mu$m. The flow rate that gives
If the viscous force is dominant compared with inertia, the regime in which the jet breaks up is identified. The universal scaling laws are of great importance to enhance the quality and resolution of the printed lines in electrostatic-inkjet-printing applications as an example of the practical impact of these models. On the basis of our analysis, we finally present an accurate prediction of the line width as a function of the jet diameter and the contact angle.

VI. CONCLUSION

In the present work, we derive scaling laws of the intact length of an electrified jet in two different regimes. The scaling laws are validated with experimental data obtained with a wide range of governing parameters. The jet intact length is independent of the voltage applied to the electro-spray. The voltage has an effect only on the cone angle and hence the cone axial length. Further, the jet length is not affected by the nozzle size because $d_n \gg R$. On the basis of competition between inertia and the viscous force, the regime in which the jet breaks up is identified. If the viscous force is dominant compared with inertia, being $Re \lesssim O(1)$, the breakup length $L_b \sim 6\mu Z_v$, while for $Re \gg O(1)$, the scaling is $L_b \sim 6/Z_v$. The experimental results reveal a threshold validity limit between the two scaling laws at $Re \simeq 5$.

These results have an impact on a wide range of application areas, including chemical engineering, the textile industry, and inkjet printing. We show how these scaling laws are of great importance to enhance the quality and resolution of the printed lines in electrostatic-inkjet-printing applications as an example of the practical impact of these models. On the basis of our analysis, we finally present an accurate prediction of the line width as a function of the jet diameter and the contact angle.

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