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Scaling laws for jet pulsations associated with high-resolution electrohydrodynamic printing

Hong Kyoong Choi, Jang-Ung Park, O Ok Park, Placid M. Ferreira, John G. Georgiadis, and John A. Rogers

Department of Chemical and Biomedical Engineering, BK21 Graduate Program of KAIST, Korea Advanced Institute of Science and Technology, Daejeon 305-701, Republic of Korea
Department of Material Science and Engineering, Beckman Institute and Frederick Seitz Materials Research Laboratory, University of Illinois at Urbana-Champaign, Urbana, Illinois 61801 USA
Department of Mechanical Science and Engineering, University of Illinois at Urbana-Champaign, Urbana, Illinois 61801 USA

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This paper presents simple scaling laws that describe the intrinsic pulsation of a liquid jet that forms at the tips of fine nozzles under electrohydrodynamically induced flows. The jet diameter is proportional to the square root of the nozzle size and inversely proportional to the electric field strength. The fundamental pulsation frequency is proportional to the electric field strength raised to the power of 1.5. These scaling relationships are confirmed by experiments presented here and by data from the literature. The results are important for recently developed high-resolution ink jet printing techniques and other applications using electrohydrodynamics.

Electric field induced formation of micron and nanometer sized droplets is useful in a number of different fields including electrospray mass spectroscopy and processing of biomaterials, electrohydrodynamic atomization, and other applications. Similar physics, particularly when used to induce pulsating jets as opposed to steady cone jets, can be exploited for printing liquid inks, with the possibility for e-jet printing with ultrafine droplets on the substrate, using a high-speed camera connected to the microscope. In a typical experiment, we observed that with increasing voltage, the meniscus deformed to a classic Taylor cone shape. At sufficiently high voltages, a jet emerged from the tip of the conical meniscus. This jet moved toward the substrate where it impinged on the surface to accumulate a droplet. After breaking, the jet recoiled back to the nozzle, leaving a printed droplet behind.

FIG. 1. (a) Schematic illustration of the nozzle and substrate in e-jet printing, with identification of key parameters used in the scaling analyses. (b) Computed equipotential lines and electric field vectors (arrows) between the nozzle and a flat plate. (c) E-jet printed arrays of \( \sim 1.5 \) \( \mu \)m dots formed using a nozzle with a 2 \( \mu \)m inner diameter. (Ink: 9:1 (v/v) water and glycercine mixture with 0.1M NaCl.)

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The overall flow rate is an important parameter in e-jet printing; it involves the balance of electrical stress, capillary pressure, and applied pressure. Chen et al. suggested a Poiseuille-type flow rate relation for this type of system,\textsuperscript{13,14} according to

\begin{equation}
Q = \frac{\pi d_n^4}{128 \mu L} \left( \Delta P + \frac{1}{2} \varepsilon_0 E^2 - \frac{4 \gamma}{d_N} \right).
\end{equation}

In Eq. (1), $Q$ is the flow rate, $\Delta P$ is the pressure drop, $\mu$ is the viscosity of the liquid, $d_n$ and $L$ are the diameter and length of the nozzle, respectively, $\varepsilon_0$ is permittivity of free space, $\gamma$ is the surface tension of the air-ink interface, and $E$ is the magnitude of the electric field. We use Eq. (1) to establish scaling relationships and the approximate relative magnitudes of key parameters. We approximate $E$ at the tip of the nozzle using a model of a semi-infinite wire perpendicular to an infinite planar counter electrode, according to $E = 4V_0/\left[ d_N \ln(8H/d_N) \right]$,\textsuperscript{15,16} where $H$ and $V_0$ are the standoff height and the imposed potential between nozzle and substrate, respectively.

Upon gradually raising $V_0$ from 0 V, one observes that fluid begins to flow from the nozzle at a certain minimum voltage. This voltage approximately corresponds to stress balance in Eq. (1), when the sum of $\Delta P$ and $\frac{1}{2} \varepsilon_0 E^2$ exceeds the capillary pressure ($4 \gamma/d_N$). We refer to this situation as the “condition for initiation of jetting.” The stress balance in Eq. (1) approximately described this initiation condition. Figure 2 shows that the sum of $\frac{1}{2} \varepsilon_0 E^2$ and $\Delta P$ at the condition for initiation of jetting is nearly constant at a level that corresponds to $\sim 70\%$ of $4 \gamma/d_N$ when the literature value for the surface tension of glycerine is used (63 dyn/cm under no electric field at room temperature). This level of agreement is reasonable, given the approximate nature of the analysis and the uncertainties in the experimental measurements.

The jetting at the apex of the Taylor cone can be explained as a competition between surface tension and electric field forces. When the electric field exceeds a certain critical value, the jetting begins at the tip of the conical meniscus. Due to this physics, it is customary to define an electrical capillary number ($Ca$) as a ratio between the surface tension and the electric field forces according to:\textsuperscript{17}

\begin{equation}
Ca = \frac{\varepsilon_0 (Ed)^2}{\gamma l_1}.
\end{equation}

where $l_1$ and $l_2$ are characteristic length scales associated with the surface tension force and the electric field force, respectively. We choose the diameter of the jet $d$ for $l_2$ and $d_N$ for $l_1$ because the surface tension acts on the entire area defined by the nozzle, while the electric field mainly focuses at the apex of the cone.\textsuperscript{18,19} The quantity $d_N$ can correspond to an anchoring diameter at the nozzle tip. With these assumptions, $d$ can be written as

\begin{equation}
d \propto \sqrt{\frac{\gamma/d_N}{E_0}}.
\end{equation}

To test the dependence of $d$ on $d_N$, we measured $d$ at the condition for initiation of jetting using a high-speed camera. Since $d$ is also time variant, we compared $d$ values at the time when the jet is widest, as determined by high-speed imaging. Here, the $d$ values were averaged at nine different distances from each nozzle tip (indicated as dashed lines), as shown in Fig. 3(b). This dependence of $d$ on $d_N$ predicted by Eq. (3) is consistent with measurements for three different nozzle diameters, as shown in Fig. 3(a). The predicted dependence of $d$ on $E$ is consistent with on literature data\textsuperscript{20} which reports that $d$ linearly decreases with increasing $V_0$ (proportional to $E$).

Margineae et al. reported that the high frequency regime is closely related with capillary waves on the surface of a charged droplet.\textsuperscript{21} From the capillary wave equation, they suggested the following scaling law between the pulsation frequency $f$ and the anchoring radius $r$:

\begin{equation}
f^2 = \frac{2 \gamma}{\pi^2 \rho r^3},
\end{equation}

which corresponds to the lowest excitation mode of a spherical droplet with negligible amount of charge. Here, $\rho$ is the density. From Eq. (4), we can derive another scaling law by using simple dimensional analysis. In most cases, $r$ corresponds best with the jet radius $d/2$ since the pulsating jet occurs at the tip of the cone which has a scale that is comparable to $d/2$. The surface tension force term in Eq. (4) can be replaced with an electrical force term because the jet in this case arises from an instability that roughly occurs when the surface tension force on nozzle $\gamma d_N$ is comparable to the electric force acting on the jet $\varepsilon_0 (Ed)^2$. This assumption, combined with Eq. (3), yields the following scaling law:

\begin{equation}
f \propto \left( \frac{\varepsilon_0}{\rho \gamma} \right)^{1/4} E^{3/2} d_{N}^{-3}.
\end{equation}

Figure 4(a) presents a plot of $f$ versus $E$ in log-log scale. The range of the observed $f$ values corresponds to the maximum that can be easily observed experimentally. We plot $E/d_{N}^{1/2}$ instead of $E$ to compensate for differences associated with nozzle size. Four different sets of data from the litera-
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12This printing behavior is different than that in conventional thermal or piezoelectric drop-on-demand (DOD) systems. In particular, in the method described in this paper, droplets emerge with a characteristic frequency that is not directly controlled, but which instead depends on various parameters of the system. In DOD printing, the time between successive drops can be arbitrarily defined by the user.
14According to Young–Laplace equation, capillary pressure in a tube Δρ is 2γ/R where R is radius of curvature. When the meniscus shape is a hemisphere, Δρ is 4γ/d0 where d0 is capillary inner diameter.