

Study on droplet formation with surface tension for electrohydrodynamic inkjet nozzle[†]

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Abstract

Droplet ejection from an electrohydrodynamic (EHD) inkjet nozzle depends on many factors such as an onset voltage, liquid conductivity, surface tension, etc. Since the surface tension has an influence on the contact angle between the nozzle surface and the liquid droplet, the surface tension change should be investigated for the control of the droplet ejection. In this study, surfactant, which can weaken the surface tension force, was used to analyze the effect of the surface tension. Furthermore, hydrophobic coating of the nozzle surface was considered as another factor for the droplet ejection. Also, a flow visualization technique was developed to observe the droplet formation and ejection from the EHD inkjet nozzle by various surface tension values.

Keywords: Droplet; Electrohydrodynamic; Inkjet; Nozzle; Surface tension

1. Introduction

Formation of micro and nanometer size droplets from an electrostatic nozzle is an important problem. Since the conventional jetting devices based on thermal bubble or piezoelectric pumping have some fundamental limitations on the droplet size which depends on the nozzle size as well as the ejection frequency, electrostatic jetting based on the direct manipulation of liquid by an electric field appears to be more promising.

The electrostatic jetting is useful in a number of different fields including ink-jet printing, electrospray mass spectroscopy and processing of biomaterials, electrohydrodynamic atomization and other applications. This process can be considered as a branch of fluid mechanics concerned with electrical force effects. The variety of variables affecting electrohydrodynamic ejection often makes it difficult to investigate and predict its operation, for instance, the difficulty in getting an exact formulation of the dependence of droplet size with other parameters such as applied voltage, flow rate of solution, size of capillary nozzle, electrode configuration and properties of liquid like viscosity, electric conductivity, relative permittivity, and surface tension. Among these effects, the electrospray can be classified in many modes such as dripping, micro-dripping,

pulsating, cone-jet and instability of cone-jet including varicose, kink and multi-jet. This classification is important to consider and define main factors (main variables) in establishment of controlling equation of droplet size. Usually, if a smaller droplet size or a higher frequency is required, an applied voltage should be increased, allowing for the observation of unstable ejection phenomena under a special condition. When the voltage increases, the droplet size decreases and the reduction rate increases because the electrostatic force increases and the surface tension decreases to reduce the droplet size for the balance of both forces. However, detailed analysis of the transition dynamics of this process is still lacking.

According to above classification, the first onset voltage which can be defined as a threshold voltage for transferring from the dripping mode to the pulsating mode is an obviously important parameter for the stable jetting because it can be used to analyze the control of the droplet size and the emission regimes corresponding to the experimental configuration. Also, the second onset voltage can be considered as a boundary between the pulsating and the cone jet mode. Hence, the electrospray can be applied in various fields such as the stable cone-jet regime which is relevant to the production of macromolecular ions and is used in conjunction with mass spectrometry in fundamental research involving biological molecules, and other regimes can be used in electrostatic painting, drug delivery, drug micro-encapsulation, etc. by the control of the onset voltage.

In this study, a formulation was determined to calculate the

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onset voltage in the range of the end of dripping mode to the start of the oscillation mode. Thus, an equation was developed to estimate the droplet size depending on the applied voltage in a given condition. The equation was also confirmed by many experimental data which were found in this research and other references. Also, surfactants were used to investigate the effect of the surface tension. Although an anion surfactant such as a SDS (sodium dodecyl sulfate) is mostly used, there are other kinds of surfactants such as a cation, an amphoteric ion, and a nonionic. Thus, the ejection characteristics with the onset voltage were studied by varying surfactants at the same surface tension condition.

2. Numerical analysis method

The onset voltage is defined as the applied voltage where the dripping mode for ejection of droplet from electrostatic nozzle transfers to pulsating cone-jet mode or other modes. The equation for calculation of onset voltage can be built based on the competition between electric force and surface tension force. The magnitude of the normal electric field in configuration between the capillary and the earthed electrode can be calculated as follows [1]:

$$E_n = \frac{\sqrt{2}U}{r \ln\left(\frac{4D}{r}\right)} \quad (1)$$

where U is the applied voltage, r is the outer radius of the capillary, and D is the distance between the nozzle tip and the earthed electrode plate. Hence, the normal electric force around a pendant drop on the electrostatic nozzle tip in the non-uniform electric field is formulated as

$$F_e = \frac{1}{2} S \varepsilon_n^2 = 4\pi\varepsilon_0 \frac{U^2}{\ln^2\left(\frac{4D}{r}\right)} \quad (2)$$

The surface area of the pendant droplet can be approximated by the outer radius of the nozzle tip r as $S = 2\pi r^2$. Thus, the electric force acting on the pendant droplet is a function of the applied voltage, the distance between the nozzle tip and the earthed electrode plate D and the outer radius of the capillary.

The surface force due to the surface tension γ of the liquid in ambient air is also approximated with the assumption that the radius of curvature of the droplet equals the outer radius of the capillary nozzle. It can be calculated as follows:

$$F_{st} = 2\pi r \gamma \varphi \quad (3)$$

where φ is Harkins correction factor [2] that depends on the ratio of the radius of the nozzle and the capillary length. The oscillation of the meniscus starts since the electric force approaches the surface force, then, the onset voltage can be established. Combining Eqs. (2) and (3), the onset voltage can

be expressed as

$$U_{ons1} = \sqrt{\frac{r\gamma\varphi}{2\varepsilon_0}} \ln\left(\frac{4D}{r}\right). \quad (4)$$

Eq. (4) is used to calculate the onset voltage at which the dripping mode stops. Furthermore, if the voltage increases, the pulsating regimes will stop and the stable cone-jet mode will occur. Thus, another threshold voltage appears at which the transition from the pulsating regimes to the stable cone-jet mode is observed. In this model the onset field at the tip of the hyperboloid surface is also defined as the equilibrium between the electrical stress pulling the liquid toward the electrode and the surface tension pulling the liquid back to the needle [3, 4]. Assuming that once the onset voltage in the needle-type emitter is reached, the liquid snaps-over to a Taylor cone and jet emission starts. This voltage can be calculated by the following equation:

$$U_{ons2} = \sqrt{\frac{r\gamma}{\varepsilon_0}} \ln\left(\frac{\beta D}{r}\right) \quad (5)$$

where the shape factor β is from 2 to 4.

As mentioned above, the electrospray behaves in dripping mode below the onset voltage. In this mode the electrical Bond number ($r\varepsilon_0 E^2 / \gamma$) that measures the strength of the electric force versus the capillary force is less than or comparable to the gravitational Bond number ($\rho g r^2 / \gamma$) that measures the relative strength of the gravitational force versus the capillary force [5]. In case of small Weber number ($\rho u^2 r / \gamma$) that measures the inertial force versus the capillary force, the diameter of the droplet ejecting from the nozzle can be approximated by balancing the gravitational force F_g and the electric force with the surface area.

$$4\pi\varepsilon_0 \frac{U^2}{\ln^2\left(\frac{4D}{r}\right)} + \frac{\pi d^3}{6} \rho g = 2\pi r \gamma \varphi \quad (6)$$

Hence, the diameter of the droplet in dripping mode can be calculated approximately by

$$d = \sqrt[3]{\frac{12}{\rho g} \left[r\gamma\varphi - 2\varepsilon_0 \left(\frac{U}{\ln\left(\frac{4D}{r}\right)} \right)^2 \right]} \quad (7)$$

On the other hand, with the numerical method, the droplet diameter with various voltages can be calculated as follows [6]:

$$d(U) = d_0 \left[1 - \left(\frac{U}{U_{ons1}} \right)^2 \right]^{1/3} \quad (8)$$

where d_0 is the droplet diameter at $U = 0$, and U_{ons1} is the onset voltage. Since the calculated results from Eq. (7) did not show good agreement with the experimental data in this study, a new equation has been developed to improve the accuracy for the relationship between the droplet diameter and the applied voltage in the range of dripping mode. The equation developed to calculate more accurate droplet diameters is as follows:

$$d(U) = d_0 = [1 - aU - bU^2]^{1/3} \quad (9)$$

where a and b are factors whose values are determined by various parameters such a surface tension of liquid, distance between the nozzle and the electrode, inside diameter of the nozzle, etc.

$$a \propto \sqrt{\frac{\epsilon_0}{\gamma D}} \quad (10)$$

$$b \propto \frac{\epsilon_0}{\sqrt{2\gamma d} \ln\left(\frac{4D}{r}\right)} \quad (11)$$

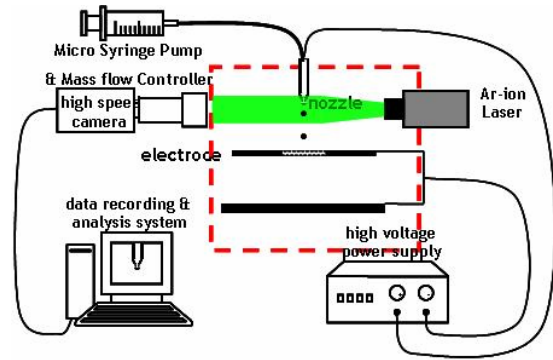
After the units of the parameters have been adjusted, the equation for the droplet diameter can be developed as follows:

$$d_n = d_0 \left[1 - K_1 \sqrt{\frac{\epsilon_0}{\gamma D}} U - K_2 \frac{\epsilon_0}{2\sqrt{2\gamma r} \ln\left(\frac{4D}{r}\right)} U^2 \right]^{1/3} \quad (12)$$

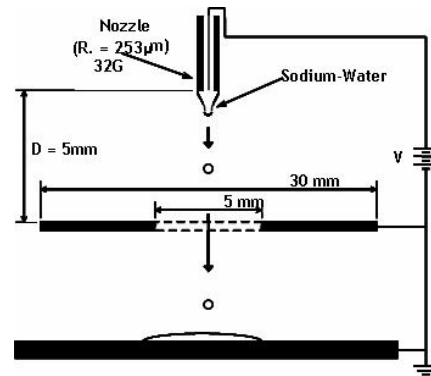
where $K_1 = K_2 = 0.8$ for the ink. If the distilled water is used, K_1 should be changed to 1.28.

3. Experimental setup

The experiment was performed initially to confirm the developed equation for the calculation of the onset voltage and the droplet diameter. Then, the effect of the surfactants was investigated using the same experimental setup. The droplet was formed and ejected form a micro glass capillary tube with an outside diameter of 500 μm including a pole made of Pt wire, an electroplate with a hole of 5 mm at the center, and the distance between the nozzle and electrode plate of 5 mm as shown in Fig. 1. Images of droplet ejection were captured by a high-speed camera (IDT XS-4) at 5000 frames per second and 512 x 512 pixel resolution with a micro-zoom lens (infinity K2), and a 5 W Ar-ion laser. A high-voltage power supply system with maximum voltage of 5.0 kV was used to control the electrostatic field and liquids have been supplied into the glass capillary by a micro-syringe pump. The liquids used in this experiment were distilled water and distilled water with four kinds of surfactants as shown in Table 2. The applied voltage changed from 1.8 kV to 3 kV at the pressure of the syringe pump of 1 kPa.



(a) Overview



(b) Test section

Fig. 1. Experimental setup for electrohydrodynamic jet.

4. Results and discussion

4.1 Equation verification

The first onset voltage is calculated by Eq. (4) clearly, which is a function of the parameters such as the distance between the capillary tip and the electrode plate, the radius of the meniscus, and the surface tension of the liquids. It is difficult to determine the radius of the meniscus at the operating condition. Thus, the curvature radius, which is called the modified radius, based on the initial meniscus of the pendant drop has been determined as follows [8]:

$$r = \begin{cases} K_1(r' - R) + R, r' \leq R \\ K_2(r' - R) + R, r' > R \end{cases} \quad (13)$$

where r' is the radius of the droplet, R is the radius of the nozzle, and K_1 is the correction factor for the radius of the droplet, with K_1 of 0.9 and K_2 of 1.2. However, there is not a general equation to calculate the initial radius of the droplet for this case. Therefore, the ϕ factor [2] can be added in this calculation process. This factor is chosen from experiment depending on the ratio between radius of the nozzle and the capillary length. Then, the onset voltage was calculated assuming that the radius of the droplet is the same as the radius of the nozzle as described in Eq. (4). These calculated results were compared with the experimental results and other published papers.

Table 1. Comparison between experimental and calculated results.

Papers	Onset voltage (kV)		Errors
	Exp.	Eq. (4)	
Present data	3.31	3.374	1.9
	2.5	2.518	0.7
	1.5	1.631	8.8
[10]	8.87	9.012	1.6
[6]	5.04	5.371	6.6
	4.02	4.141	3.0
[9]	4	4.324	8.1
	5.02	4.824	3.9
[11]	4.4	4.651	5.7

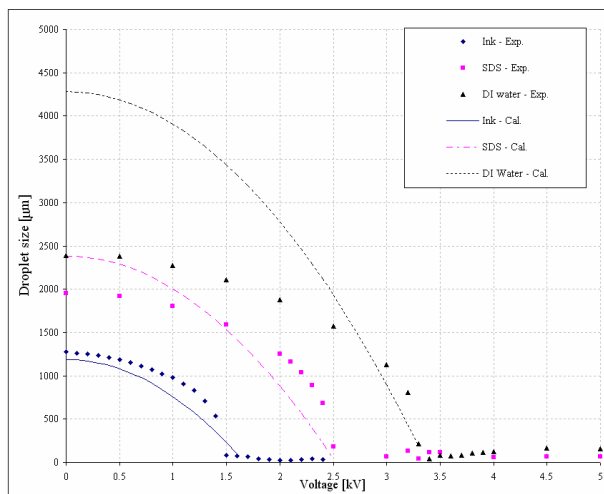


Fig. 2. Droplet diameter with applied voltage.

In the present study, with three solutions and three experimental conditions, the onset voltages were obtained for 1.5 kV, 2.5 kV, 3.3 kV for ink, 1%wt SDS and distilled water respectively. Initially, a uniform droplet size appeared with a regular formation frequency for stable ejection, up to 3.3 kV for the distilled water. The droplet size then decreased rapidly and the ejection became unstable with irregular frequency and various droplet sizes, 10 μm to 200 μm , from 3.3 kV to 5 kV, since the ejection frequency of the droplet became shorter than the relaxation time of the liquid, so as to make the charge for the liquid unstable. Other experimental results were found in Refs. [6, 9, 10, 11]. A comparison of results is shown in Table 1. The calculated for these experimental conditions were similar to the experimental results. The errors were from 0.7 to 8.8%. The confirmation was also performed by comparing with other results and the calculated errors were similar ranging from 1.6% to 8.1%.

As the experimental results, the droplet diameters decreased uniformly until the applied voltage is lower than the first onset voltage. The consequences calculated from Eq. (9) are compared with the experimental results as shown in Fig. 2. The

Table 2. Fixed surface tension for various surfactants.

Surfactant	Surface tension
Cation 0.1%	39.50 (dyne/cm ²)
Nonion 0.007%	38.92 (dyne/cm ²)
Amphoteric ion 0.001%	39.46 (dyne/cm ²)
Anion 0.01%	39.72 (dyne/cm ²)

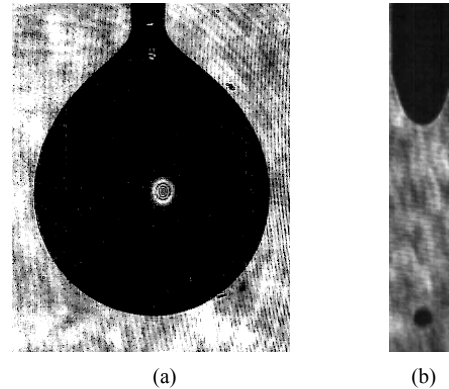


Fig. 3. Droplet ejection mode: (a) Dripping mode; (b) Micro dripping mode.

calculated results show good agreement with the experimental results for the cases of ink and water with 1% SDS. For distilled water the calculated results were higher than the experimental results, which can be explained by the effect of the electric conductivity. Since the normal electric force dominates in the case of the high conductivity, the equation for the electric force just focus on the normal electric force although distilled water has some free charges. In addition, since the surface tension of the distilled water is higher than the others, the calculated results for the diameter of droplets are larger than the experimental results.

4.2 Effect of surfactant

The droplet ejection in this study can be divided into a dripping mode where a droplet size is bigger than a nozzle (Fig. 3(a)) at a relatively lower voltage and a micro dripping mode where a droplet size is smaller than a nozzle (Fig. 3(b)) at a relatively a higher voltage. The onset voltage is a boundary between the dripping and the micro dripping mode.

The onset voltage should be changed by the surfactant even though the surface tension is consistent. Thus, the surfactant was added to the distilled water resulting in similar surface tension as shown in Table 2. The viscosities after adding surfactant were also similar ranging from 1.0 to 1.2 cP. Then, the droplet ejection experiment was performed.

Different onset voltages were measured for different surfactants even though the surface tension was fixed as shown in Fig. 4. The amphoteric ion was not ejected at 1.7 kV and was exhausted from 2.5 kV as a dripping mode. The onset voltage of the nonion was the highest as 3.01 kV and that of the anion

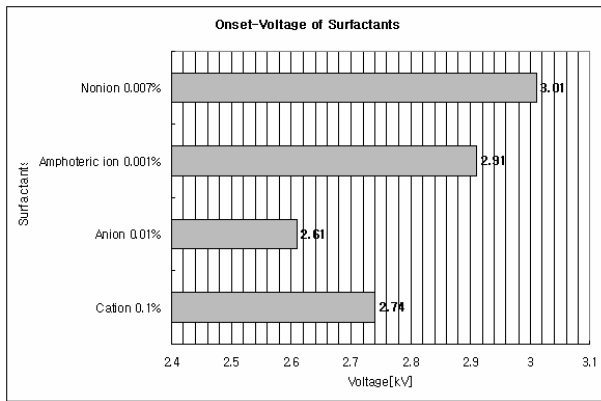


Fig. 4. Onset voltage for various surfactants.

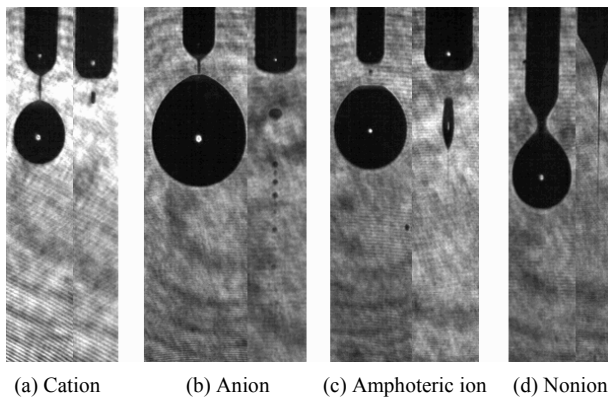


Fig. 5. Droplet ejection pattern.

was the lowest as shown in Fig. 4.

Droplets formed before and after the onset voltage were observed as shown in Fig. 5. The droplet size of the cation was smallest ($370\ \mu\text{m}$) while that of the anion was biggest ($684\ \mu\text{m}$) just before the onset voltage. The nonion surfactant showed similar phenomena as the cation for relatively small droplets before the onset voltage. If the applied voltage increases, the droplet size decreases greatly to produce stable dripping for the cation case while the jetting mode without any droplet was observed for the nonion case. The anion and the amphoteric cases provided relatively big droplets before the onset voltage, and satellite droplets were found after the onset voltage for unstable ejection.

5. Conclusion

A new equation was derived and confirmed with the experimental data in this study to calculate the onset voltage accurately by various parameters. The equation used to estimate the diameters of the droplet below the onset voltage was also developed and confirmed for the dripping mode. The calculated results of the proposed model show good agreement with the experimental results. Nevertheless, more experiments and analyses are required in order to confirm the

developed equation exactly with the factors in physical effects on the ejection of the droplets.

The ejection mechanism was varied by the surfactants, and stable ejection was observed especially for the cation case compared with other cases of the unstable ejection. The conductivity of the liquid, ionization characteristics, internal flow distribution, and charge variation can be the factors for the ejection mechanism.

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Nomenclature

D	: Distance between nozzle tip and electrode plate
d_o	: Droplet diameter
F_g	: Gravitational force
K	: Correction factor
R	: Radius of nozzle
r	: Outer radius of capillary
r'	: Radius of droplet
U	: Applied voltage
U_{onsl}	: Onset voltage.
β	: Shape factor
γ	: Surface tension of liquid
φ	: Harkins correction factor

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