Simulations for Pulsating Breakups of a Nano Taylor Cone

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Abstract. In this paper, a Taylor cone model in the nanoscale is configured using the many-body dissipative particle dynamics method. The sharpening process of the Taylor cone and the breakup process at different electric field intensities and different charge concentrations are systematically investigated. Under a strong electric field, the hemispherical droplet is sharpened over time and evolves into a conic one. Then the conic cusp emits a thin jet. Finally, the cone shrinks into a semi-sphere after jet breaking. These deformation processes occur several times until no charges are emitted from the conic cusp. It is found that the electric field force is responsible for jet emitting, while the Coulombic force causes a jet breakup. With the rising of the intensity of the electric field, the breakup times also increase. However, the breakup times decrease with the rising of the charge concentration. It indicates that a conductive liquid with low electrical conductivities and subjected to a strong electric field is more prone to undergo pulsating breakups.

1. Introduction

Since Taylor [1] studied the electrohydrodynamic (EHD) flows, the Taylor cone, as a fundamental phenomenon in EHD printing, received dramatic attention owing to its wide applications in wearable electronics [2]. A Taylor cone is usually generated by a capillary nozzle when it is imposed by a high potential. If the liquid in the nozzle is electrically charged, the liquid meniscus at the nozzle end suffers several stable or unstable modes due to the electric stress. The liquid meniscus is generated by a flow rate and the electric stress is caused by a high voltage. Early researches [1] found that a steady jet followed the liquid meniscus when the applied voltage exceeds a critical value. The phenomenon is later termed the cone-jet. Through adjusting the flow rates and the voltages, Marginean et al. [3] experimentally exhibited three astable modes including dripping, pulsating breakup, and cone-jet in electrosprays. The three modes are also observed by Lee et al. [4]. Pulsating breakup, as one of the basic instabilities of the Taylor cone, was revealed in several experiments. It is a typically quasi-steady jetting which is related to the steady cone-jet. In most cases, pulsating breakup occurred in the periodic operating conditions, such as a pulse voltage [5]. Under a constant voltage and a constant flow rate, the cone jet may periodically be formed and ruptured at a single frequency [6]. Although the influence of external operating conditions on the occurrence of pulsating breakup has been extensively investigated, the intrinsic mechanisms of pulsating breakup involving the electric stress are still not well-understood. Regarding stress, the leaky-dielectric model (LDM) [7] summarized by Melcher is commonly used to expound the interfacial deformation of the leaky dielectric (LD) liquid. For LD fluids, charge carriers can be migrated from the bulk fluid to its surface due to the electric stress. A tangential stress on the liquid-gas surface drives the fluid. LDM is an effective model to explain the EHD problems. However, the formation of the pulsating breakup is more complicated since the working fluid is usually doped with a few ions [6]. The dynamic behavior
of electrolytes is significantly different to LDs [7] and so the LDM fails to expound the formation of pulsating breakup. It remains unclear how the Taylor cone becomes unstable from the viewpoint of ion motions.

Challenged by the above, the many-body dissipative particle dynamics (MDPD) method is used to examine the formation of pulsating breakup. MDPD, as a mesh-free particle method, has developed to study the nano-droplet dynamics. Plenty of MDPD simulations can be comparable to the results obtained by theoretical analysis [8], [9]. In the work, the behavior of pulsating breakups from a doped-ion droplet is deeply investigated. The formation process of the Taylor cone, the sharpening process of the conic cusp and the breakup process at different electric field intensities and different charge concentrations are simulated. The cone angle, the charge dynamics and the times of pulsating breakup have been systematically investigated.

2. Physical Model and Mathematical Formulation

A Taylor cone model is depicted in Figure 1(a), in which a high electric potential is imposed on the nozzle. The grounded substrate receives the broken droplets. In figure 1(b), a simple but effective model is built, which is similar to the experimental settings of Beroz et. al. [10] Note that the physical parameters in MDPD are dimensionless. In the work, the unit length, time, mass, charge and electric field intensity in MDPD units denote $l_0$, $t_0$, $m_0$, $q_0$ and $E_0$. The real parameters refer to our former work [11]. A semi-spherical droplet with a radius of $R = 20l_0$ wets a solid and rigid wall, of which height ($H$) is $2l_0$ and width ($W$) is $80l_0$. Four typical particles including the wall particle, the water particle, the positive particle and the negative particle compose the model. Here, water, positive and negative particles are liquid particles, but the wall particle is a solid particle. The solid particles are fixed in the grids without any interactions. The working temperature is 300 K and the contact angle is 70°.

The simulation process experiences two stages, in which firstly the wetting process lasts $100t_0$, then the electric field is imposed. LAMMPS [12] is used to implement numerical simulations. Fifteen cases with electric field intensities ranging from $E = E_0$ to $E = 5E_0$ and different charge concentrations including $c = 10^{-3}$, $c = 2 \times 10^{-3}$ and $c = 5 \times 10^{-4}$ are conducted. The charge concentration is defined as the ratio of the number of positive or negative particles to that of total liquid particles.

![Figure 1](image.png)

**Figure 1.** (a) Schematic diagram of a Taylor cone system and (b) a Taylor cone model configured by MDPD.

In MDPD, the motions of fluid particles are governed by Newton’s second law

$$\frac{dr_i}{dt} = \mathbf{v}_i, \quad m_i \frac{dv_i}{dt} = \mathbf{F}_i$$

(1)

where $r_i$, $\mathbf{v}_i$, $m_i$ and $\mathbf{F}_i$ are the position, velocity vector, mass and exerted force, respectively of $i$-th particle. In the simulations, the masses of all particles are $m_i = m_0$. Regarding the four typical particles as shown in figure 1(b), the exerted force $\mathbf{F}_i$ is the sum of the conservation force ($\mathbf{F}_{i}^C$), dissipative force ($\mathbf{F}_{i}^D$), random force ($\mathbf{F}_{i}^R$), electric field force ($\mathbf{F}_{i}^E$) and Coulombic force ($\mathbf{F}_{i}^{\text{Coul}}$):

$$\mathbf{F}_i = \sum_{i\neq j} \left(\mathbf{F}_{ij}^C + \mathbf{F}_{ij}^D + \mathbf{F}_{ij}^R + \mathbf{F}_{ij}^{\text{Coul}}\right) + \mathbf{F}_i^E$$

(2)

The first three terms of the above equation are defined in the original MDPD [13], which are formulated
the negative particles move onto the wall. When the positive particles are accumulated in the cusp, the deformation of the droplet surface. The electric field and the stress at the interface. The tangential electric stress stretches the surface so the round droplet evolves into a conic shape. At about $t = 190t_0$, a standard cone appears due to the electric field and the Coulombic forces balancing the surface tension. After that, the thin jet is emitted from the conic cusp due to the gathered charged particles. As the electric field and the Coulombic forces remarkably drive the charged particles. Most of the positively charged particles finally distribute among the surface. It is known that a droplet tends to form into a smooth sphere due to the surface tension. However, when the conductive droplet is placed in an outer electric field, the Coulombic and the electric field forces exeracting on the surface charged particle give rise to a tangential electric stress at the interface. The tangential electric stress stretches the surface so the round droplet evolves into a conic shape. At about $t = 190t_0$, a standard cone appears due to the electric field and the Coulombic forces balancing the surface tension. After that, the thin jet is emitted from the conic cusp because of the electric field and the Coulombic forces exceed the surface tension. As shown in Figure 2(c), most positive charged particles are dispersed in the conic cusp and the jet. Thus, a large Coulombic force due to the gathered charged particles tears the jet so that the jet breaks up into small drops. In every breakup, the semispherical droplet loses a few charged particles. As the electric field and the Coulombic

\[ F_{ij}^C = A\omega_a(r_{ij})e_{ij} + B(\rho_i + \rho_j)\omega_0(r_{ij})e_{ij}, \]
\[ F_{ij}^R = -\gamma\omega_1(r_{ij})(e_{ij} \cdot v_{ij})e_{ij}, \]
\[ F_{ij}^{\text{Coul}} = \frac{q_i q_j}{r_{ij}} e_{ij} \text{ when } r \leq r_{\text{coul}}, \quad F_{ij}^E = q_i E, \]

where $r_{ij} = |r_i - r_j|$ and $e_{ij} = (r_i - r_j)/|r_i - r_j|$ are the distance between $i$-th and $j$-th particles and their unit direction vector, respectively. $\omega_a(r_{ij})$ and $\omega_0(r_{ij})$ are the weight functions, which are denoted by $\omega_a(r_{ij}) = 1 - r_{ij}/r_c$. The local density $\rho_i$ can be computed as $\rho_i = \sum_{e<j} 15(1 - r_{ij}/r_d)^2/(2\pi r_d^2)$, where $\rho_p$ satisfies the normalized relation $\int_0^\infty 4\pi r^2 \rho_p(r) \, dr = 1$. The dissipative force and the random force satisfy the fluctuation-dissipation theorem, $\sigma^2 = 2\gamma k_B T$ and $\omega^D(r_{ij}) = [\omega^R(r_{ij})]^2 = (1 - r_{ij}/r_c)^2$. $\xi_{ij}$ is symmetric Gaussian random variables with zero mean and unit variance. The Coulombic force and the electric field force of equation (2) only exert on the positive and negative particles. They are respectively given by

\[ F_{ij}^{\text{Coul}} = \frac{q_i q_j}{r_{ij}} e_{ij} \]

where $q_i$ and $q_j$ are the charges on the two charged particles, $C$ denotes the energy-conversion constant, $r_{\text{coul}}$ is the Coulombic cutoff radius and $E$ is the vector of the electric field. The MDPD parameters are listed in Table 1. For the liquid-solid particles, the attractive parameter $A$ is altered to adjust the contact angle to $70^\circ$. For the MDPD fluid, an empirical fitting of the surface tension refers to the work of Arienti et al. [14]. In this study, the surface tension is 4.968 in the MDPD unit.

**Table 1.** MDPD parameters for interaction of liquid-liquid particles and liquid-solid particles.

| MDPD parameters | $A$ | $B$ | $\gamma$ | $r_c$ | $r_d$ |
|-----------------|-----|-----|---------|------|------|
| Liquid-Liquid   | -40 | 25  | 18      | 1    | 0.75 |
| Liquid-Solid    | -25 | 25  | 18      | 1    | 0.75 |

3. Results and Discussions

This section presents the pulsating breakups of a Taylor cone including the sharping process of the cone, the variation of cone angle, and the influences of the electric field intensity and ion concentration on the breakup times.

3.1. Pulsating Breakups

As shown in Figure 2(a), the model firstly experiences the wetting process. Then a conic cusp depicted in Figure 2(b) appears at the bottom of the droplet. Finally, the fine and short jet issuing from the conic apex throws off progeny droplets, as evident in Figure 2(c). In this section, the first breakup of the Taylor cone is primarily investigated. The effect of the electric field force during the formation of the Taylor cone becomes significant and thus the positive particles move towards the bottom of the droplet, while the negative particles move onto the wall. When the positive particles are accumulated in the cusp, the Coulombic force takes effect in the deformation of the droplet surface. The electric field and the Coulombic forces exert on the surface charged particle give rise to a tangential electric stress at the interface. The tangential electric stress stretches the surface so the round droplet evolves into a conic shape. At about $t = 190t_0$, a standard cone appears due to the electric field and the Coulombic forces balancing the surface tension. After that, the thin jet is emitted from the conic cusp because of the electric field and the Coulombic forces exceed the surface tension. As shown in Figure 2(c), most positive charged particles are dispersed in the conic cusp and the jet. Thus, a large Coulombic force due to the gathered charged particles tears the jet so that the jet breaks up into small drops. In every breakup, the semispherical droplet loses a few charged particles. As the electric field and the Coulombic
forces diminish, the large surface tension drives the broken jet. As a result, the conic cusp shrinks under the surface tension so the charged particles move towards the bulk droplet.

Figure 2. Charge motions in pulsating breakups at $E = 4E_0$ and $c = 10^{-3}$. (a) The wetting process, (b) the firstly sharpening process of the Taylor cone and (c) the firstly shrinking process of the conic cusp. The semi-cone angle of the Taylor cone is widely used to characterize the cone deformation [15]. In Figure 3, during the wetting stage, the semi-cone angle keeps a constant value of about 48°. By contrast, in the stage of Taylor cone forming, the angle of the semi-cone decreases until the semi-cone angle reaches the minimum value of about 30°. At this point, the conic cusp rapidly breaks up into small drops. The two stages are demarcated by the black dotted line in Figure 3. Then the semi-cone angle rises to another constant value due to the shrinking of the conic cusp. After shrinking to a maximum value, the semi-cone angle decreases because the Taylor cone becomes sharp again. The conic cusp throws off droplets in the same way as the first breakup. In Figure 3, the Taylor cone will encounter three breakups divided by the blue dotted lines. However, the minimum semi-cone angle in the third breakup only reaches about 38°. Besides, the semi-cone angle finally maintains about 44° and no breakup occurs.

Figure 3. Semi-cone angle of the Taylor cone as a function of time at $E = 4E_0$ and $c = 10^{-3}$.

3.2. Semi-cone Angles

Figure 4. Variation of the semi-cone angle at different electric field intensities and a charge concentration of $10^{-3}$. 
Figure 4 depicts the semi-cone angles varying with different electric field intensities and a constant charge concentration of $10^{-3}$. At low electric field intensities of $E = 1$ or $E = 2$, the semi-cone angles change a little over time. It indicates that the droplet deformation is stable without the Taylor cone. When the electric field intensity increases, the droplet becomes unstable and a thin jet emits from the conic cusp and breaks up into small drops. The times of pulsating breakup increases with the rising of the electric field intensity. For the cases of $E = 1$ and $E = 2$, no breakup occurs during the droplet deformation. For the cases of $E = 3$ and $E = 5$, the Taylor cone encounters 1 and 6 breakups, respectively. Hence, a critical intensity of electric field greater than $2E_0$ and less than $3E_0$ will give rise to a cone-jet.

Figure 5(a) shows the semi-cone angles varying with different charge concentrations and an electric field intensity of $E = 4E_0$. At low charge concentrations ($c = 5 \times 10^{-4}$ and $c = 10^{-3}$), the Taylor cone is prone to undergo pulsating breakup. In the two cases, the Taylor cones rupture 5 times and 3 times, respectively. However, it only ruptures once at a high charge concentration of $c = 2 \times 10^{-3}$. The jet is short at a low charge concentration but is stretched very long at a high charge concentration, as displayed in figure 5(b). Besides, the jet at a low charge concentration is finer than that at a high charge concentration. The electric field force is the cause of jet stretching, while the Coulombic force results in a jet breakup.

### 3.3. Times of Pulsating Breakup

The electric capillary number

$$Ca_E = \varepsilon E^2 R / \Gamma$$

measures the electric stress to surface tension, where $\varepsilon = 78.5$ is the relative electric permittivity of water. The electric stress can reflect the electric field force and the Coulombic force on the atomic scale. As depicted in figure 6, no breakup occurs at a low electric capillary number ($Ca_E < 316$). As the electric capillary number increases, the times of breakup significantly rises for the cases of $c = 5 \times 10^{-4}$ and $c = 10^{-3}$. However, for the case of charge concentration of $c = 2 \times 10^{-3}$, the jet only breaks once and is unaffected by the electric capillary number. At medium electric capillary numbers ($Ca_E = 1264$, $Ca_E = 2844$ and $Ca_E = 5056$), the times of breakup rises with the decreasing of the charge concentration (Marked by the black arrows in figure 6). At an electric capillary of $Ca_E = 7900$ and a charge concentration of $10^{-3}$, the droplet encounters 6 breakups.

### 4. Conclusions

The many-body dissipative particle dynamics simulations demonstrate how a conductive Taylor cone repeatedly forms and breaks up into small droplets. Under a strong electric field, a hemispherical droplet firstly evolves into a conic shape owing to the large electric field force. Then a thin jet is emitted from...
the conic apex. Due to a large Coulombic force, the thin jet subsequently disintegrates into progeny drops. The broken cone finally shrinks into a semi-sphere since the surface tension plays an important role in the cusp deformation. Three forces including the electric field force, Coulombic force and surface tension are responsible for pulsating breakups. The electric field force leads to a thin jet, the Coulombic force causes jet breakup and the surface tension gives rise to cone shrinking. Through analyzing the semi-cone angle of the Taylor cone, it is found that the cone angle is as small as 30° and the Taylor cone even encounters 6 breakups. The times of pulsating breakups increases as the electric field intensity goes up. The result gives an internal mechanism of the deformation of a nano Taylor cone and can improve the droplet dynamics in EHD printing.

Acknowledgements
This work was supported by the National Natural Science Foundation of China (No. 51876071).

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