Critical condition for the transformation from Taylor cone to cone-jet*

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An energy method is proposed to investigate the critical transformation condition from a Taylor cone to a cone-jet. Based on the kinetic theorem, the system power allocation and the electrohydrodynamics stability are discussed. The numerical results indicate that the energy of the liquid cone tip experiences a maximum value during the transformation. With the proposed jetting energy, we give the critical transformation condition under which the derivative of jetting energy with respect to the surface area is greater than or equal to the energy required to form a unit of new liquid surface.

Keywords: Taylor cone, cone-jet, electrohydrodynamics, numerical computation

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1. Introduction

The original work on the evolution behavior of electrified drops was contributed by Rayleigh,[1] in which he proposed the angular frequency expression of a charged drop as

$$\omega^2 = \frac{m(m-1)}{\rho R^3}\left((m+2)\gamma - \frac{Q_R^2}{16\pi^2\varepsilon_0 R^3}\right), \tag{1}$$

where ρ is the liquid density, R is the radius of the spherical drop, γ the surface tension, Q_R the total charge on the drop, and ε_0 is electric permittivity of a vacuum. It is clear that in order to have $\omega^2 > 0$, the minimum value of m is 2. Thus we can obtain the Rayleigh limit $Q_R = 8\pi(\gamma\varepsilon_0 R^3)^{1/2}$, which denotes the theoretical limit of the maximum charge of an isolated charged drop. In 1916, Rayleigh[2] made a further discussion about Eq. (1). Yet, the detailed deduction of this equation was given by Peters[3] in 1980.

In the early twentieth century, researchers started to investigate the dynamic behavior of a charged drop at the capillary tip (CDCT) under an electric field. Then as a base, electrospray, electrospinning, and other relevant technical fields were gradually developed.[4–6] Zeleny[7–9] demonstrated abundant dynamical phenomena of CDCT under an electric field experimentally. Especially in Ref. [8], he first tried to give the critical instability condition of CDCT based on the work of Rayleigh. An unsound assumption was made for the difference in pressure between the inside and the outside of the spherical drop when instability occurred, which results in the confusion of Rayleigh's criterion. In 1964, by supposing that the liquid is a complete conductor and neglecting the part of the jet, Taylor[10] gave the electric potential expression near the CDCT in an electric field, which was later called the basic electrostatic solution.[11] After this, the local convex cone of an electrified liquid was named the Taylor-cone. On this basis, Gañán-Calvo[11] obtained an analytical cone-jet solution. Fernández de la Mora,[12] Gañán-Calvo,[11] and Higuera[13] discussed a series of scale law problems. Collins *et al.*[14] revealed the formation mechanism of a liquid cone for different conductive liquids through numerical simulations, which contains two remarkable results, i.e., the time evolution of the tip curvature and the cross-section variation of axial velocity of the liquid cone.

With the rapid development in chemistry, biology, and other related scientific and technical fields, there is an increasing interest in the Rayleigh limit.[15–20] Especially Duft *et al.*[21] and Achtzehn *et al.*[22] first achieved the imaging of the disintegration of a levitated droplet charged to the Rayleigh limit. The clear pictures provided us with many authentic details. Their analyses showed that the Coulomb emission is an unstable transient catastrophe process. What should be pointed out is that Rayleigh limit is the result of an ideal experiment, which cannot be achieved in reality. First, the spherical symmetry cannot be completely ensured for the charged drop. Also due to the effects of the ion emission and evaporation, the drop often loses some charges and mass before approaching the Rayleigh limit. Therefore, the experimental results are generally lower than the Rayleigh limit. For more information about Coulomb emissions and Taylor-cones of charged fluid, readers can refer to Refs. [23] and [24].

However, the above studies still do not answer a more essential issue: what the critical condition is for the transformation from a Taylor cone to a cone-jet. Both the Rayleigh

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limit and Taylor's result were discussed in terms of the balance between the electric field force and the surface tension, in which the dynamical nature of fluid was ignored. For this issue, it is inadequate that the inertia force and the viscous force are not considered. A more general approach should be from the balance in energy rather than in force. The influences of the kinetic energy, the work of the external forces, the viscous dissipation, and the surface energy should be taken into account comprehensively. Furthermore, for the same liquid, the critical condition for the transformation should be an intrinsic property of the liquid, but not an extrinsic action such as the applied voltage, because different boundary conditions will definitely lead to the change of the so-called applied voltage.

For both the formation of a steady jet at the capillary tip and the transient Coulomb emission of a charged drop, there exists the transformation from a Taylor cone to a cone-jet. The aim of this paper is to answer the question about what the critical condition for the transformation is.

2. Modeling and analysis

Supposing that the electrohydrodynamical process is isothermal and adiabatic, and the fluid is an incompressible consistent Newtonian fluid, we have the following mass conservation equation:

$$\nabla \cdot v = 0, \tag{2}$$

where $\nabla \cdot v$ is the divergence of the fluid velocity vector. The momentum balance equation is

$$\rho \frac{\mathrm{d}v}{\mathrm{d}t} = \rho g + \nabla \cdot (P + T_\mathrm{e}), \tag{3}$$

where $\mathrm{d}/\mathrm{d}t$ is the material derivative, ρ is the fluid density, g is the gravity acceleration vector directing to the negative z axis, T_e is the Maxwell stress tensor, and P is the internal stress tensor. Considering the incompressibility of fluid, we have $P = 2\mu S + (-pI + \nabla \cdot v/3)I = 2\mu S - pI$, where μ is the kinematic viscosity coefficient, $S = [\nabla v + (\nabla v)^\mathrm{T}]/2$ is the strain rate tensor, and p is the liquid pressure. Thus

$$\nabla \cdot P = 2\mu \nabla \cdot S - \nabla p = \mu \nabla^2 v - \nabla p. \tag{4}$$

The Maxwell stress tensor T_e in Eq. (3) is $T_\mathrm{e} = \varepsilon EE - \varepsilon |E|^2 I/2$, where $\varepsilon = \varepsilon_\mathrm{r}\varepsilon_0$, ε_r is the relative dielectric constant, E is the electrical field strength vector, EE is the tensor product, $|E|$ is the Euclid norm of the electric field strength vector, and I is the identical tensor. Hence, we obtain the expression of the electric force density vector as

$$f_\mathrm{e} = \nabla \cdot T_\mathrm{e} = q_\mathrm{b} E - \frac{1}{2}|E|^2 \nabla \varepsilon, \tag{5}$$

where q_b is the body density of charge. Here, we have neglected the interaction between electric dipoles. According to Eqs. (3)–(5), we can obtain the following momentum balance equation:

$$\rho \frac{\mathrm{d}v}{\mathrm{d}t} = \rho g + \mu \nabla^2 v - \nabla p + q_\mathrm{b} E - \frac{1}{2}|E|^2 \nabla \varepsilon. \tag{6}$$

In this paper, we simply consider the case that the characteristic time of an electric phenomenon is far greater than that of a magnetic one. In this case, the magnetic effect can be ignored. So, the corresponding equations for the electrostatic phenomenon are $\nabla \cdot (\varepsilon E) = -\nabla^2(\varepsilon \psi) = q_\mathrm{b}$ and $\nabla \times E = 0$, where ψ is the electric potential. The charge conservation equation is $\partial q_\mathrm{b}/\partial t + \nabla \cdot J = 0$. The current density vector J includes the conductive and the convective items, i.e., $J = \sigma E + q_\mathrm{b} v$, where σ is the conductivity. In some cases, the contribution of ionic migration and charge diffusion can be included in J.[25–28]

Because our initial boundary value problems include the free surface, we now discuss the boundary jump conditions of the hydrodynamic phenomenon and the electric phenomenon. Here we denote n as the outside normal vector of the liquid interface. $\langle Q \rangle = Q_\mathrm{outside} - Q_\mathrm{inside}$ denotes the jump value of Q when the liquid is traveling through the interface. The fluid jump conditions at the interface are $n \cdot \langle v \rangle = 0$ and $\langle [n \cdot (P + T_\mathrm{e})] \cdot t_n \rangle = 0$.

Suppose that the liquid surface is a simple regular one, i.e., a continuously differentiable vector function $r(s_1, s_2)$, $(s_1, s_2) \in D \subset \boldsymbol{R}^2$ with one-to-one correspondence can be used to describe the liquid surface, where $|\partial r/\partial s_1 \times \partial r/\partial s_2| \neq 0, \forall (s_1, s_2) \in D$. The corresponding curvature radii of the two parameters are R_1 and R_2 and the mean curvature is $H = (1/R_1 + 1/R_2)/2$. Thus, the balance equation at the interface is

$$\langle (n \cdot (P + T_\mathrm{e})) \cdot n \rangle = \gamma (1/R_1 + 1/R_2) = 2\gamma H. \tag{7}$$

The term on the right-hand side of Eq. (7) denotes the resultant force of surface tension. t_i $(i = 1, 2)$ are defined as two orthogonal tangent vectors embedded in the interface. The local orthogonal rectangular coordinate system is formed in the order of $t_1 \to t_2 \to n$. Then, the jump conditions of the Maxwell stress tensor are $(T_\mathrm{e}^n \cdot n) \cdot n = \langle \varepsilon (E \cdot n)^2 - \varepsilon (E \cdot t_1)^2 - \varepsilon (E \cdot t_2)^2 \rangle / 2$ and $(T_\mathrm{e}^n \cdot n) \cdot t_i = qE \cdot t_i$. The jump conditions of electric phenomenon are $\langle \psi \rangle = 0$, $\langle E \cdot t_n \rangle = 0$, and $\langle \varepsilon E \cdot n \rangle = q$. The conservative equation of the surface charge is $\partial q_\mathrm{s}/\partial t + v \cdot \nabla_\mathrm{s} q_\mathrm{s} = qn \cdot (n \cdot \nabla) v - n \cdot \langle \sigma E \rangle$, where t_n is the tangent vector orthogonal to n, q_s is the charge at the liquid surface, and $\nabla_\mathrm{s} \equiv \nabla \cdot (I - nn)$ is the surface gradient operator.

Now we employ the general form of the kinetic energy theorem[29], i.e.,

$$\frac{\mathrm{d}U_\mathrm{k}}{\mathrm{d}t} = \frac{\mathrm{d}W_\mathrm{ext}}{\mathrm{d}t} + \frac{\mathrm{d}W_\mathrm{int}}{\mathrm{d}t}, \tag{8}$$

where U_k is the kinetic energy of a given system, W_{ext} is the work done by the external forces, and W_{int} is the cohesion work. We refer to the fluid in domain Ω_t of R^3 at moment t as a system and divide it into two parts (Ω_t^1 and Ω_t^2) along a given surface S_t. Ω_t^1 is the domain of the imminent emitting jet and Ω_t^2 is the domain of the parent drop or the liquid connecting with the capillary tube, which are shown in Fig. 1.

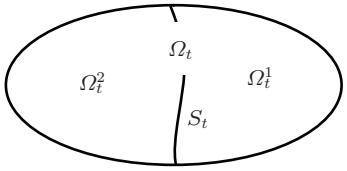


Fig. 1. Partition $\Omega_t^1 \cup \Omega_t^2$ of system Ω_t at time t.

The kinetic energy of subsystem Ω_t^1 is $\mathrm{d}U_k/\mathrm{d}t = \int_{\Omega_t^1} \rho|v|^2/2\,\mathrm{d}\tau$. The work done by the external forces applied in the domain Ω_t^1 is

$$\begin{aligned}\frac{\mathrm{d}W_{ext}}{\mathrm{d}t} &= \int_{\Omega_t^1} v\cdot(\rho g + f_e)\,\mathrm{d}\tau + \int_{\partial\Omega_t^1} v\cdot[n\cdot(P+T_e)]\,\mathrm{d}A \\ &= \int_{\Omega_t^1} v\cdot(\rho g + f_e)\,\mathrm{d}\tau + \int_{S_t} v\cdot[n\cdot(P+T_e)]\,\mathrm{d}A \\ &\quad + \int_{\partial\Omega_t^1\backslash S_t} v\cdot[n\cdot(P+T_e)]\,\mathrm{d}A, \end{aligned} \tag{9}$$

where $\partial\Omega_t^1\backslash S_t$ is a difference set. Considering the jump condition (Eq. (7)), we have

$$\begin{aligned}\frac{\mathrm{d}W_{ext}}{\mathrm{d}t} &= \int_{\Omega_t^1} v\cdot(\rho g + f_e)\,\mathrm{d}\tau + \int_{S_t} v\cdot[n\cdot(P+T_e)]\,\mathrm{d}A \\ &\quad + \int_{\partial\Omega_t^1\backslash S_t} v\cdot(-2n\gamma H)\,\mathrm{d}A. \end{aligned} \tag{10}$$

We define the first two items on the right-hand side of Eq. (10) as the power of the driving forces

$$\frac{\mathrm{d}W_d}{\mathrm{d}t} = \int_{\Omega_t^1} v\cdot(\rho g + f_e)\,\mathrm{d}\tau + \int_{S_t} v\cdot[n\cdot(P+T_e)]\,\mathrm{d}A. \tag{11}$$

Thus, W_d plays the role of driving the liquid Ω_t^1 leaving Ω_t^2. The third item on the right-hand side of Eq. (10) is the negative power of the resultant surface tensor, i.e., the negative time rate of the change of the surface energy when the liquid surface area changes. Hence,

$$\frac{\mathrm{d}\Gamma}{\mathrm{d}t} = -\int_{\partial\Omega_t^1\backslash S_t} v\cdot(2n\gamma H)\,\mathrm{d}A. \tag{12}$$

The cohesion power is

$$\begin{aligned}\frac{\mathrm{d}W_{int}}{\mathrm{d}t} &= \int_{\Omega_t^1} v\cdot(\nabla\cdot P)\,\mathrm{d}\tau - \int_{\partial\Omega_t^1} v\cdot(n\cdot P)\,\mathrm{d}A \\ &= \int_{\Omega_t^1} \nabla\cdot(P\cdot v)\,\mathrm{d}\tau - \int_{\Omega_t^1} P:S\,\mathrm{d}\tau \\ &\quad - \int_{\partial\Omega_t^1} v\cdot(n\cdot P)\,\mathrm{d}A, \end{aligned} \tag{13}$$

where $P:S$ is the scalar multiplication of the tensor P and S. For the incompressibility of the liquid we obtain the relationship $P:S = 2\mu S:S = \phi$. Considering the Stokes formula,

$$\frac{\mathrm{d}W_{int}}{\mathrm{d}t} = -\int_{\Omega_t^1} \phi\,\mathrm{d}\tau = -\frac{\mathrm{d}\Phi}{\mathrm{d}t}, \tag{14}$$

where Φ is the dissipative work, when equations (10)–(12) and (14) are substituted into Eq. (9), the following relationship is obtained:

$$\frac{\mathrm{d}U_k}{\mathrm{d}t} = \frac{\mathrm{d}W_d}{\mathrm{d}t} - \frac{\mathrm{d}\Gamma}{\mathrm{d}t} - \frac{\mathrm{d}\Phi}{\mathrm{d}t}. \tag{15}$$

Equation (15) clearly indicates the allocation of system power. It can be concluded that the changing rate of kinetic energy is equal to the power of the driving forces subtracted by the changing rate of the surface energy and the dissipative work, i.e., the second and the third items on the right-hand side of Eq. (15) constituting the resistance items of the liquid emission.

The time derivative can be expressed as the form $\mathrm{d}/\mathrm{d}t = (\mathrm{d}/\mathrm{d}A)(\mathrm{d}A/\mathrm{d}t)$. After the cancellation of the area changing rate $\mathrm{d}A/\mathrm{d}t$, equation (15) can be rewritten as

$$\frac{\mathrm{d}U_k}{\mathrm{d}A} = \frac{\mathrm{d}W_d}{\mathrm{d}A} - \frac{\mathrm{d}\Phi}{\mathrm{d}A} - \frac{\mathrm{d}\Gamma}{\mathrm{d}A}. \tag{16}$$

Now we define an energy variable, jetting energy, as

$$\Pi = U_k + \Phi - W_d, \tag{17}$$

and we have

$$\frac{\mathrm{d}(\Pi+\Gamma)}{\mathrm{d}A} = 0. \tag{18}$$

Equation (18) indicates that the energy $\Pi+\Gamma$ is stationary in the transformation process from Taylor cone to cone-jet. When there is a small change on the surface area of the Taylor cone, whether the liquid emission occurs depends on the sign on the left-hand side of Eq. (18). The transformation process is considered to be unstable when the energy $\Pi+\Gamma$ is maximum and stable when it is minimum. The sufficient condition for the transformation stability is

$$\frac{\mathrm{d}^2(\Pi+\Gamma)}{\mathrm{d}A^2}\begin{cases} <0, & \text{unstable}, \\ =0, & \text{neutral}, \\ >0, & \text{stable}. \end{cases} \tag{19}$$

3. Numerical computation

Based on the above governing equations and boundary jump conditions, we numerically compute the energy variations during the transformation from a Taylor cone to a cone-jet. Taking the transformation process of the liquid emission at the capillary tip for example, the computational physical model is shown in Fig. 2. Model parameters are $\mu = 0.008\ \mathrm{N\cdot s/m^2}$, $\rho = 859\ \mathrm{kg/m^3}$, $\gamma = 0.0335\ \mathrm{N/m}$, $\sigma = 8.0\times10^{-6}\ \mathrm{S/m}$, $\varepsilon_0 = 8.854\times10^{-12}\ \mathrm{C^2/N\cdot m^2}$, $\varepsilon_r = 12$, $\psi_0 = 10^4\ \mathrm{V}$, and fixed axial velocity $v_0 = 0.018\ \mathrm{m/s}$.

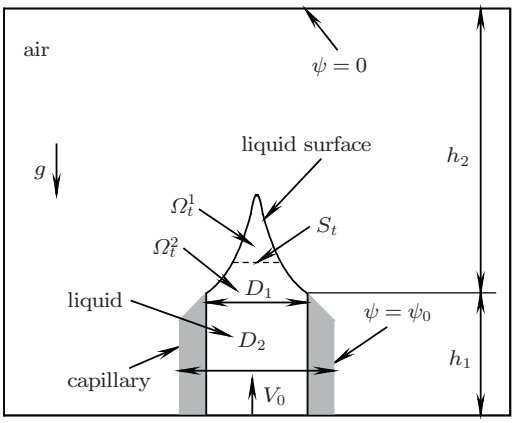


Fig. 2. Configuration of the physical model. The parameters are chosen as $D_1 = 0.35$ mm, $D_1 = 0.45$ mm, $h_1 = 1$ mm, $h_2 = 2.5$ mm, $v_0 = 0.018$ m/s, $\psi_0 = 7000$ V.

Figure 3 shows the variations of surface energy Γ, energies $\Pi + \Gamma$ and Π with the surface area of the domain Ω_t^1 in the transformation process. From Fig. 3, there is a maximum of $\Pi + \Gamma$, which indicates that it is an unstable process when the liquid is changing from a Taylor cone to a cone-jet.

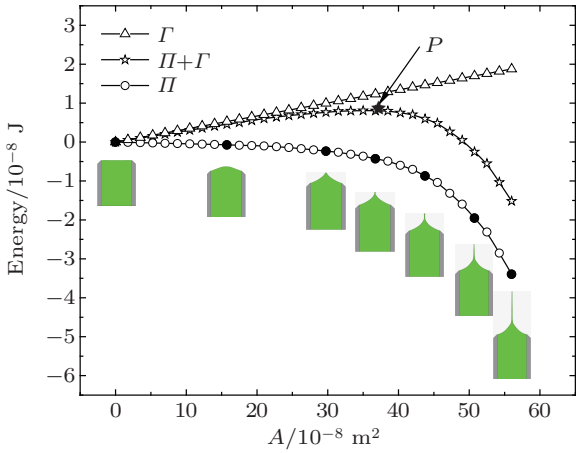


Fig. 3. (color online) Plots of surface energy Γ, energies $\Pi + \Gamma$, and Π versus the surface area of domain Ω_t^1, where P indicates the maximum point of energy $\Pi + \Gamma$.

Now we can define a new parameter $G = -\mathrm{d}\Pi/\mathrm{d}A$ used in the criterion for liquid emission. Considering $\Gamma = \int_{\partial\Omega_t^1/S_t} \gamma \mathrm{d}A$, we can obtain

$$G = -\frac{\mathrm{d}\Pi}{\mathrm{d}A} = \frac{\mathrm{d}\Gamma}{\mathrm{d}A} = \gamma. \tag{20}$$

For the liquid under given experimental conditions, G is an invariant and can be considered as the driving force of liquid emission. The constant γ, in essence, denotes the energy required to form a new unit area of liquid surface. Hence, equation (20) represents the criterion for the transformation from a Taylor cone to a cone-jet: when the derivative of jetting energy G with respect to the surface area is greater than or equal to the energy γ required to form a new unit area of surface, liquid emission will occur. This conclusion is firmly established based on the energy balance.

4. Conclusions

In this paper, the jetting energy is defined to relate the electrohydrodynamics stability to the extremum of energy and consequently the sufficient condition (Eq. (19)) for the stability of the transformation is given. With the jetting energy, the critical condition (Eq. (20)) for the transformation is proposed. We obtain these theoretical results drawn from some ideas of fracture mechanics. It is clear that our approach is not simply restricted to the gas–liquid two-phase case. If we change the surface tension to interface tension and change Eq. (7) to an appropriate form, there is no difficulty in explaining the critical conditions for the coaxial jet.

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