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Current/flow-rate characteristic of an electrospray with a small meniscus

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A relation between the electric current and the flow rate of an electrospray operating in the cone–jet mode is proposed for the case of a very small meniscus, a long stationary jet, and a flow with important inertial effects. The finite size of the meniscus then affects the bulk-to-surface current transfer process, though the meniscus is still much larger than the drops of the spray. The result differs from the square-root law which is well-established for large menisci or very viscous flows.

1. Introduction

In the cone–jet regime of electrostatic atomization, a meniscus of the liquid to be sprayed is subjected to an electric field that induces electric charge at the liquid surface. This causes an electric stress that strains the meniscus into a cone with a thin jet emanating from its apex. In a classic analysis, Taylor (1964) showed that a conical meniscus strained by an electric field is an exact equilibrium solution in the absence of any flow, and used a balance of normal electric stress and surface tension to determine the field around the cone as $E = E_T = O(\gamma/\epsilon_0 R)^{1/2}$, where γ is the surface tension of the liquid, R is the distance to the apex of the cone, and ϵ_0 is the permittivity of vacuum. Taylor's analysis should be supplemented with an analysis of the flow and the current transport in the cone-to-jet transition region and in the jet of the electrospray, where the hydrostatic balance is not applicable. A brief summary of these regions follows; further details can be found in Fernández de la Mora & Loscertales (1994), Gañán-Calvo, Dávila & Barrero (1997) and Higuera (2003). Electric charge is carried to the surface of the meniscus and the jet by conduction in the liquid, under the action of the field which strains the meniscus. In combination with the component of the field tangent to the surface, this surface charge generates an electric shear on the liquid that is directed away from the meniscus and favours the observed flow. The surface charge is convected by the flow it helps to generate, causing an additional electric current. The sum of the conduction current in the liquid and the convection current at its surface is a constant. The expressions for these two contributions to the current in the axisymmetric configuration sketched in figure 1 are

$$I_b(x) = 2\pi K \int_0^{r_s} E_x^i r \, \mathrm{d}r \quad \text{and} \quad I_s(x) = 2\pi\sigma v_s r_s,$$

respectively, where K is the electric conductivity of the liquid, E_x^i is the axial (x) component of the electric field in the liquid, $r_s(x)$ is the radius of the cross-section of the surface, and v_s is the velocity of the liquid at the surface. The conduction current dominates in the meniscus, where the speed of the liquid and the density of free-surface charge are small and decrease with increasing distance to the apex. The

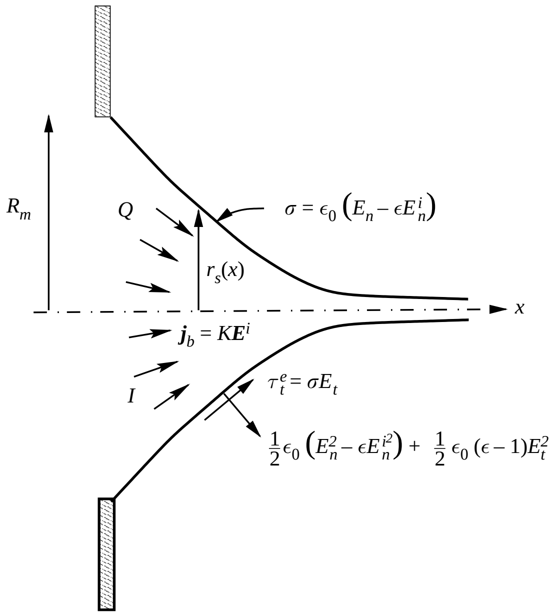

FIGURE 1. Definition sketch.

convection current dominates in the jet far downstream, where the radius r_s and the electric field decrease with streamwise distance. In between, there is a bulk-to-surface current transfer region which determines the total current $I = I_b + I_s$ as a function of the flow rate Q injected through the meniscus.

In many cases, the current transfer region is small compared with the conical meniscus, which makes the structure of this region nearly independent of the configuration of the electrospray system. In these conditions the electric current often follows a universal scaling law, being proportional to the square root of the flow rate and dependent only on physical properties of the liquid (Fernández de la Mora & Loscertales 1994; Gañán-Calvo *et al.* 1997). In other cases, however, the disparity of scales underlying the square-root law is not realized. These include recent applications of electrostatic atomization in which multiple miniature menisci, each in the range of tens of micrometres, are packed in a single device. The electrostatic interaction between elements is then of prime importance and has been studied in some detail (Rulison & Flagan 1993; Almekinders & Jones 1999; Regele *et al.* 2002; Hubacz & Marijnissen 2003; Barrero 2004). The current/flow-rate characteristic of each individual meniscus depends on the electric field prevailing in the current transfer region, and does not necessarily follow a square-root law. Another case in point may be the electrospraying of liquids of low electrical conductivity, which produces thick jets and long transfer regions (Gomez & Tang 1994). The purpose of this paper is to describe the bulk-to-surface current transfer process under a variety of electric field conditions, in order to explore possible current/flow-rate characteristics of a range of electrospray configurations.

2. Governing equations

The axisymmetric flow of the liquid obeys the Navier–Stokes equations. The electric fields in the liquid and outside can be written in terms of electric potentials as $\boldsymbol{E}^i = \nabla\varphi^i$ and $\boldsymbol{E} = \nabla\varphi$, respectively, with $\nabla^2\varphi^i = \nabla^2\varphi = 0$.

The following relations exist between the quantities pertaining to the liquid surface. The density of free-surface charge, σ, determines the jump of the electric field across the surface as (Landau & Lifshitz 1960)

$$\sigma = \epsilon_0 \left(E_n - \epsilon E_n^i\right) \quad \text{and} \quad E_t = E_t^i, \tag{2.1}$$

where E_n^i and E_n are the components normal to the surface of the electric field in the liquid and outside, E_t^i and E_t are the corresponding components of the fields tangent to the surface, and ϵ is the dielectric constant of the liquid. The electric stresses normal and tangent to the surface are (Landau & Lifshitz 1960; Saville 1997)

$$\tau_n^e = \tfrac{1}{2}\epsilon_0\left(E_n^2 - \epsilon E_n^{i^2}\right) + \tfrac{1}{2}\epsilon_0(\epsilon - 1)E_t^2 \quad \text{and} \quad \tau_t^e = \sigma E_t. \tag{2.2}$$

The conservation equation for the surface charge in the axisymmetric configuration of figure 1 is

$$\frac{\mathrm{d}I_s}{\mathrm{d}x} = 2\pi r_s K E_n^i \left(1 + r_s^{\prime 2}\right)^{1/2}, \tag{2.3}$$

where the right-hand side is the rate of bulk-to-surface current transfer.

Much work has been devoted to elucidating the structure of the current transfer region where bulk conduction current becomes surface convection current. Fernández de la Mora & Loscertales (1994) identified the current transfer region with an electric relaxation region around the apex of the cone where conduction fails to carry to the surface the charge that would be needed to screen the liquid from the applied electric field. From this condition they derive the result $I \sim (\gamma K Q/\epsilon)^{1/2}$. Additional estimates have been worked out that take into account the pressure and viscous stresses at the surface necessary to upset the surface tension–normal electric stress balance and deform the surface away from a cone. Work on these lines began with the inertial scaling of Fernández de la Mora *et al.* (1990). When the motion of the liquid is dominated by viscosity, estimates of this kind suggest that the current transfer region is never large compared with the largest of the electric relaxation region and another region around the apex where the viscous stress becomes of the order of the surface-tension stress (Higuera 2003). When the inertia of the liquid dominates, however, most of the current transfer seems to occur in the jet, over a length that increases with the flow rate (Gañán-Calvo *et al.* 1997; Gañán-Calvo 1999; Higuera 2003). The analysis of the current transfer in these conditions has been carried out on the assumption that the field outside the liquid decreases as the inverse of the square root of the streamwise distance, as in Taylor's solution, which is appropriate when the length of the current transfer region is small compared with the characteristic size of the meniscus R_m. This analysis is extended here to the opposite case when the length of the current transfer region is large compared with R_m.

3. Orders of magnitude

The field away from the cone, which will enter the extended analysis, depends on the specific configuration of the system in which the spray is generated. In some of the pioneering experiments of Taylor, the meniscus lies on a horizontal metallic plate and the electric field is due to a voltage V applied between this plate and another parallel plate a distance $L \gg R_m$ apart. The condition of matching of the Taylor field around the apex with the uniform field between the plates is, in orders of magnitude, $E_T(R_m) \sim V/L$, which determines the order of the required voltage as $V = O[(\gamma R_m/\epsilon_0)^{1/2}(L/R_m)]$. A more quantitative analysis turns this estimate into an exact result, and experiments show that the cone–jet regime is realized only when the voltage is in a narrow range around a value of this order.

In a variant of this configuration used extensively, the meniscus is at the end of a long metallic needle of radius R_m held at a voltage V perpendicularly to a grounded plate at a distance L from the needle. The field at a distance R from the end of a needle is of order $[V/\ln(L/R_m)]/R$. Matching this field with the Taylor field E_T for $R = O(R_m)$ requires $V = O[(\gamma R_m/\epsilon_0)^{1/2}\ln(L/R_m)]$, a result often used in the analysis of this configuration.

Assume that current transfer occurs in a region of the jet where the axial field would be $E_{x0}(x)$ in the absence of the jet. (Hereafter x is measured from the apparent apex of the cone.). The velocity of the liquid in the jet is $v \approx Q/\pi r_s^2$ and the pressure variation associated with this velocity is of order $\rho v^2 = O(\rho Q^2/r_s^4)$, where ρ is the density of the liquid. The electric stress normal to the surface is of order $\epsilon_0 E_n^2$, from (2.2). The condition that this electric stress should be of the same order as the pressure variation gives $E_n = O(\rho^{1/2} Q/\epsilon_0^{1/2} r_s^2)$.

On the other hand, seen from a distance large compared with its radius, the jet acts as a line of charge. It induces an axial field that can be approximated by $\ln(r_s/x)\mathrm{d}(E_n r_s)/\mathrm{d}x = O(E_n r_s/x)$ up to logarithmic factors (see, e.g., Ashley & Landahl 1965 for a derivation of this result). The condition that this axial field should be of the order of E_{x0}, so that it can screen the liquid from E_{x0} in the region where conduction dominates and would lead to too large a current if E_{x0} entered the liquid, gives $r_s = O(\rho^{1/2} Q/\epsilon_0^{1/2} E_{x0} x)$.

The axial field needed in the liquid when conduction is responsible for at least a fraction of the total current I is $E_x^i = O(I/K r_s^2) = O(\epsilon_0 I x^2 E_{x0}^2/\rho K Q^2)$. This field increases with x if $x E_{x0}(x)$ increases, and it becomes of the order of the outer field E_{x0} for $x = O(x_t)$, with x_t given by $x_t^2 E_{x0}(x_t) = \rho K Q^2/\epsilon_0 I$. Since E_x^i cannot be large compared with E_{x0}, conduction cannot account for the electric current I when $x \gg x_t$, so that x_t defines the characteristic length of the bulk-to-surface current transfer region.

Insofar as the residence time of the flow in this region ($t_r = x_t/v$) is large compared with the electric relaxation time $t_e = \epsilon_0 \epsilon/K$, the density of free-surface charge is nearly equal to the equilibrium value $\sigma = \epsilon_0 E_n$ that screens the liquid from E_n (i.e. $\epsilon E_n^i \ll E_n$ in (2.1); see Fernández de la Mora & Loscertales 1994; Gañán-Calvo *et al.* 1997; Higuera 2003). This charge leads to a convection current $I_s = 2\pi\sigma v r_s = O(\epsilon_0^{1/2} E_{x0}^{3/2} \rho^{1/2} K^{3/2} Q^2/I^{3/2})$, and the condition that $I_s = O(I)$ in the current transfer region gives finally $I = O(\epsilon_0^{1/5}\rho^{1/5} E_{x0}^{3/5} K^{3/5} Q^{4/5})$, where $E_{x0} = E_{x0}(x_t)$.

Introducing the scaling factors $R_0 = (\epsilon_0^2\gamma/\rho K^2)^{1/3}$, $Q_0 = \epsilon_0\gamma/\rho K$, $I_0 = \epsilon_0^{1/2}\gamma/\rho^{1/2}$ and $E_0 = I_0/K R_0^2$, and taking $E_{x0}(x) = A/x^n$, with $A = O(\gamma^{1/2} R_m^{n-1/2}/\epsilon_0^{1/2})$ to match with the Taylor field around the meniscus, the estimates above can be written as

$$\frac{I}{I_0} \sim \frac{(Q/Q_0)^{\frac{4-5n}{5-4n}}}{(R_m/R_0)^{\frac{3}{2}\frac{1-2n}{5-4n}}}, \tag{3.1a}$$

$$\frac{x_t}{R_0} \sim \left(\frac{R_m}{R_0}\right)^{2\frac{1-2n}{5-4n}} \left(\frac{Q}{Q_0}\right)^{\frac{3}{5-4n}}, \tag{3.1b}$$

$$\frac{r_s}{R_0} \sim \left(\frac{R_m}{R_0}\right)^{\frac{1}{2}\frac{1-2n}{5-4n}} \left(\frac{Q}{Q_0}\right)^{\frac{2-n}{5-4n}}. \tag{3.1c}$$

These results apply when $x_t \gg R_m$, which amounts to $Q/Q_0 \gg R_m/R_0$. On the other hand, the flow rate cannot be arbitrarily large if a conical meniscus has to exist at all. The pressure variations due to the flow in the meniscus are of order $\rho Q^2/R^4$ at a distance R from the apex. These pressure variations become of the order of the surface tension and lead to a deformation of the surface away from a cone when $\rho Q^2/R^4 \sim \gamma/R$, which defines the hydrodynamic region of Fernández de la Mora *et al.* (1990), of characteristic size $R_h = (\rho Q^2/\gamma)^{1/3} = R_0(Q/Q_0)^{2/3}$. This region is small compared with the size R_m of the meniscus only if $Q/Q_0 \ll (R_m/R_0)^{3/2}$.

The case $n = 1/2$ corresponds to a very large meniscus, for which the electric field E_{x_0} is given by Taylor's solution in the current transfer region and beyond ($E_{x_0} = O(A/x^{1/2})$). The estimates (3.1) become independent of R_m/R_0 for this case, as was to be expected. The estimate of the current becomes $I \sim (\gamma K Q)^{1/2}$, which coincides with the result of Fernández de la Mora & Loscertales (1994) up to a factor which is a function of ϵ. The length of the transfer region and the radius of the jet in this region scale as Q/Q_0 and $(Q/Q_0)^{1/2}$, respectively, in agreement with the results of Gañán-Calvo (1999) and Higuera (2003).

The case $n = 0$ corresponds to a meniscus lying on a metallic plate. Then $I \propto Q^{4/5}$ in the range $R_m/R_0 \ll Q/Q_0 \ll (R_m/R_0)^{3/2}$, which is at variance with the usual square-root law. In the jet beyond the current transfer region, the conditions $v \sim Q/r_s^2$, $\sigma v r_s \sim I$ and $\rho v^2 r_s^2 \sim \sigma E_{x_0} r_s x$, expressing the conservation of mass, current (which is almost entirely convection current) and momentum (a balance of the acceleration of the liquid and the electric shear), give

$$\frac{r_s}{R_0} \sim \frac{(Q/Q_0)^{11/20}}{(E_{x_0}/E_0)^{2/5}(x/R_0)^{1/4}}, \quad v \sim \frac{Q}{r_s^2}, \tag{3.2a}$$

$$\frac{\sigma}{\epsilon_0 E_0} \sim \frac{(E_{x_0}/E_0)^{1/5}(Q/Q_0)^{7/20}}{(x/R_0)^{1/4}}. \tag{3.2b}$$

Solutions for other values of n are also of interest. The electric current ceases to be an increasing function of the flow rate when $n > 4/5$, which includes the important case $n = 1$ of a meniscus at the end of a long metallic needle. In this case the current increases as the square root of the flow rate when $x_t \ll R_m$, reaches a maximum of the order of $I_0(R_m/R_0)^{1/2}$ at a flow rate of the order of $Q_0(R_m/R_0)$, and then decreases, if a solution exists at all.

For any n, the asymptotic estimates of this section imply that the surface-tension stress is small compared with the pressure variation and the normal-electric stress in a region of the jet that includes the current transfer region. The stationary solution should become unstable when this region is sufficiently long, and the breakup of the jet could then lead to drops with a charge higher than the Rayleigh limit.

4. Numerical results

The order of magnitude estimates of the previous section have been backed with numerical computations for $n = 0$. The Navier–Stokes equations for the flow in the liquid, the Laplace equations for the electric potentials in the liquid and outside, and the conservation equation (2.3) for the density of free-surface charge have been solved with boundary conditions at the liquid surface expressing the balance of surface tension, pressure, viscous stresses and electric stresses (given by (2.2)), and the electrostatic conditions (2.1). The condition $\varphi = 0$ is imposed at the metallic plate holding the meniscus (see figure 1). The liquid is assumed to flow through a hole of

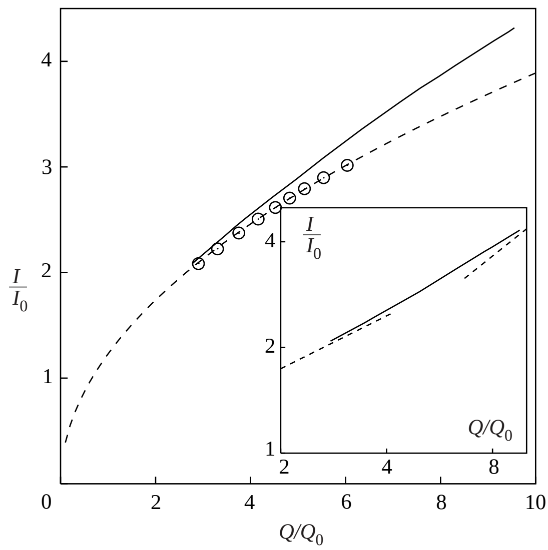

FIGURE 2. The solid curve gives the dimensionless current as a function of the dimensionless flow rate. The dashed curve is $I/I_0 = 1.23\,(Q/Q_0)^{1/2}$. The circles give the current computed for different values of the flow rate in the absence of a uniform far field. Inset: log-log plot. The dashed lines at the left and right in the inset have slopes 1/2 and 4/5, respectively.

radius R_m drilled in this plate, with the meniscus attached at the rim of the hole. Uniform radial velocity and electric field are imposed at the inlet ($r < R_m$). These are only approximate conditions. More accurate conditions should involve the flow and the electric field in the feeding pipe, but these are not expected to affect the current transfer region much because of the large contraction experienced by the flow between the feeding pipe and the current transfer region.

Additional boundary conditions should be specified at the far electrode (extractor) in front of the meniscus. The distance L between the two electrodes may be large compared with the length of the current transfer region, which makes for intensive computations when the whole system is simulated. Here, to decrease the numerical burden and gain some generality, a solution is computed in a reduced domain covering only the meniscus and the current transfer region. For this purpose, the asymptotic results (3.2), along with $\partial\varphi/\partial x \to E_{x0}$ (with E_{x0} a given constant), are used as boundary conditions at the downstream boundary.

The problem can be written in non-dimensional form using the scaling factors introduced above (3.1). The non-dimensional problem contains the four parameters

$$Re = \frac{\rho Q_0}{\mu R_0} = \frac{\rho^{1/3}\epsilon_0^{1/3}\gamma^{2/3}}{\mu K^{1/3}}, \quad \epsilon, \quad \frac{R_m}{R_0}, \quad \frac{E_{x0}}{E_0}, \tag{4.1}$$

where μ is the viscosity of the liquid. (The last two parameters should satisfy the order of magnitude relation $E_{x0}/E_0 = O(R_0/R_m)^{1/2}$ for a cone–jet to exist.) The solution of the problem determines I/I_0 as a function of Q/Q_0 and these four parameters.

The computed dimensionless current is plotted in figure 2 as a function of the dimensionless flow rate for $Re = 0.5$, $\epsilon = 50$, $R_m/R_0 = 20$, and $E_{x0}/E_0 = 0.07$. As can be seen, the current departs from a square-root law (dashed curve) when the flow rate increases, and seems to tend to the predicted 4/5 law for large flow rates. The position of the cross-over point x_c at which $I_s = I_b$, which is an indicator of the current transfer

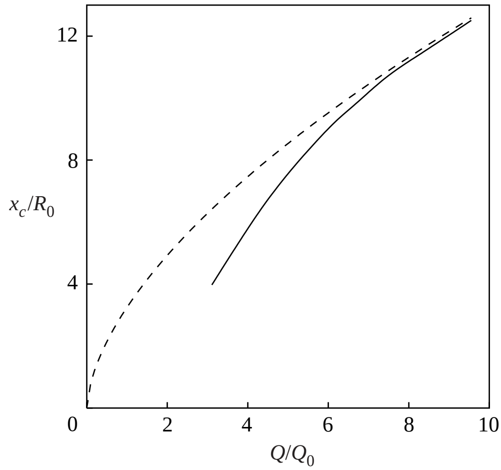

FIGURE 3. Cross-over point at which $I_s = I_b$ as a function of the flow rate (solid). The dashed curve is $x_c/R_0 = 3.25\,(Q/Q_0)^{3/5}$.

progress, is plotted in figure 3 as a function of the flow rate. The cross-over point shifts downstream with increasing flow rate and approaches the 3/5 law predicted by (3.1*b*) $[x_t/R_0 \sim (R_m/R_0)^{2/5}(Q/Q_0)^{3/5}]$ for the largest values of the flow rate. Inspection of the numerical results shows, however, that x_c is never sufficiently far in the jet to assume that most of the current transfer occurs under a constant E_{x_0} in these computations. The flow rate cannot be increased much above the values of figures 2 and 3 because the combined effect of the dynamic pressure variation at the beginning of the jet and the departure of the electric field from the Taylor field around the rim of the injecting hole is already affecting the whole meniscus, which is nowhere a cone. This difficulty is a consequence of the value of the parameter R_m/R_0 used in the computations, which is smaller than in real applications. Increasing R_m/R_0 would increase the range of admissible flow rates at the price of more demanding computations, but the computed results already show a clear departure of the current from a square-root law.

It could be argued that part of this departure might be due to the meniscus never being a Taylor cone. In fact, the coefficient of the dashed curve in figure 2, $I/I_0 = 1.23\,(Q/Q_0)^{1/2}$, is smaller that the coefficient 2.6 proposed by Gañán-Calvo (1999). To assess the importance of this effect, additional computations have been carried out with the far-field condition $\partial\varphi/\partial x \to E_{x_0}$ (a constant) replaced by a far field given by Taylor's solution ($E_T = O(1/R^{1/2})$), and the downstream asymptotic conditions (3.2) changed accordingly (see Higuera 2003 for the modified form of (3.2) when $E_{x_0} \sim A/x^{1/2}$). This far field can be realized in principle using a far electrode of the appropriate shape, though it cannot be a flat plate, for which no cone–jet would exist without a uniform E_{x_0}. The current computed with the modified far field is represented by circles in figure 2. Solutions could not be extended to very large flow rates in the absence of a uniform field but, as can be seen, the current nearly follows a square-root law even though the flow is affected by the finite size of the meniscus.

5. Conclusions

Order of magnitude estimates and numerical computations have been used to analyse the bulk-to-surface current transfer region of an electrospray when the length of this region is not small compared with the size of the meniscus. The electric current is predicted to increase faster/slower than the square root of the flow rate when the electric field in the current transfer region decreases with streamwise distance less/more rapidly than the Taylor field. Estimates of the length of the current transfer region and the radius of the jet in this region have been also worked out. Power-law scalings for large flow rates break down when the electric field decreases sufficiently rapidly with streamwise distance. This result could be hinting at a maximum flow rate, or a maximum current, above which a solution ceases to exist in some conditions of interest.

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