

Session 42: Electrospray Propulsion

Electrospray thrusters are electrostatic accelerators of charged particles produced from electrified liquid surfaces. There are three types of electrospray thruster technologies:

1. Colloid thrusters, which are accelerators of charged droplets and, under special circumstances, ions and use solvents such as doped glycerol and formamide as propellant.
2. FEEP (field emission electric propulsion) makes use of liquid metals (typically Cs and In) and produce positively charged metallic ions.
3. ILIS (ionic liquid ion sources), that use room-temperature molten salts, also known as ionic liquids and produce salt ion beams, or mixtures of ions and droplets.

The first form of electrospray propulsion (which can be tracked back to the beginning of the 20th century) came in the form of colloid thrusters. They were intensively studied from around 1960 to 1975 as an alternative to normal ion engines. Their appeal at that time rested with the large “molecular mass” of the droplets, which was known to increase the thrust density of an ion engine. This is because the accelerating voltage is $V = \frac{mc^2}{2q}$, where m is the mass of the ion or droplet, and q its charge, and c is the final speed. If c is pre-defined (by the mission), then V can be increased as m/q increases. This, in turn, increases the space charge limited current density (as $V^{3/2}$), and leads to a thrust density, $\frac{F}{A} = \frac{\epsilon_0}{2} \left(\frac{4V}{3d} \right)^2$, (d is the grid spacing), which is larger in proportion to V^2 , and therefore to $(m/q)^2$. In addition to the higher thrust density, the higher voltage also increases efficiency, since any cost-of-ion voltage V_{loss} becomes then less significant, $\eta = \frac{V}{V + V_{loss}}$.

In a sense, this succeeded too well. Values of droplet m/q that could be generated with the technology of the 60’s were so large that they led to voltages from 10 to 100 kV (for typical $I_{sp} \approx 1000$ s). This created very difficult insulation and packaging problems, making the device unattractive, despite its demonstrated good performance. In addition, the droplet generators were usually composed of arrays of a large number of individual liquid-dispensing capillaries, each providing a thrust of the order of 1 μ N. For the missions then anticipated, this required fairly massive arrays, further discouraging implementation.

After lying dormant for many years, there is now a resurgence of interest in electrospray engine technology. This is motivated by:

- (a) The new emphasis on miniaturization of spacecraft. The very small thrust per emitter now becomes a positive feature, allowing designs with both, fine controllability and high performance.
- (b) The advances made by electrospray science in the intervening years. These have been motivated by other applications of electrospraying, especially in recent years, for the extraction of charged biological macromolecules from liquid samples, for very detailed mass spectroscopy. These advances now offer the potential for overcoming previous limitations on the specific charge q/m of droplets, and therefore may allow operation at more comfortable voltages (1 – 5 kV).

- (c) The advances in micro-manufacturing technologies allow for efficient clustering of a large number of emitter tips on a small surface, potentially to the point of competing with plasma thrusters (ion or even Hall) in achievable current density.

With regard to point (a), one essential advantage of electrospray engines for very small thrust levels is the fact that no gas phase ionization is involved. Attempts to miniaturize other thrusters (ion engines, Hall thrusters, arcjets) lead to the need to reduce the ionization mean free path $(\sigma_{ion}n_e)^{-1}$ by increasing n_e , and therefore the heat flux and energetic ion flux to walls. This leads inevitably to life reductions. In the electrospray case, as we will see, the charging mechanisms are variations of “field ionization” on the surface of a liquid; small sizes naturally enhance local electric fields and facilitate this effect.

Basic Physics

Consider first a flat liquid surface subjected to a strong normal electric field, E_0 . If the liquid contains free ions (from a dissolved electrolyte), those of the attracted polarity will concentrate on the surface. Let σ be this charge, per unit area; we can determine it by applying Gauss’ law $\nabla \cdot \vec{E} = \rho/\epsilon_0$ in integral form to a “pill box” control volume,

$$\sigma = \epsilon_0 E_0 \quad (1)$$

A similar effect (change concentration) occurs in a dielectric liquid as well, even though there are no free charges. The appropriate law is then $\nabla \cdot \vec{D} = 0$ where $\vec{D} = \epsilon\epsilon_0\vec{E}$ and ϵ is the relative dielectric constant, which can be fairly large for good solvent fluids ($\epsilon = 80$ for water at 20°C). There is now a non-zero normal field in the liquid (i), and we have,

$$\epsilon_0 E_0 - \epsilon\epsilon_0 E_i = 0 \quad (\text{no free charges}) \quad (2)$$

and, in addition from $\nabla \cdot \vec{E} = \rho_{\text{total}}/\epsilon_0 = 0 + \rho_d/\epsilon_0$,

$$\epsilon_0 E_0 - \epsilon_0 E_i = \sigma_d \quad (\text{dipolar charge}) \quad (3)$$

Eliminating E_i between these expressions,

$$\sigma_d = \left(1 - \frac{1}{\epsilon}\right) \epsilon_0 E_0 \quad (4)$$

which, if $\epsilon \gg 1$ is nearly the same as for a conducting liquid, however, is important to recall that only in a perfect conductor both normal and tangential fields are required to be zero. In a very high ϵ material the normal field,

$$E_i = \frac{1}{\epsilon} E_0 \quad (5)$$

may vanish, but the tangential component, if any, will be continuous across the interface.

Consider a conductive liquid with a conductivity K , normally due to the motion of ions of both polarities. If their concentration is $n^+ = n^- = n$ (m^{-3}), and their mobilities are, μ^+ and μ^- ($\text{m/s}/(\text{V/m})$), then,

$$K = ne(Z^+\mu^+ + Z^-\mu^-) \quad (\text{Si/m}) \quad (6)$$

Suppose there is a normal field E_0 applied suddenly to the gas side of the liquid surface. The liquid surface side is initially un-charged, but the field draws ions to it (positive if E_0 points away from the liquid), so a free charge density σ builds up over time, at a rate,

$$\frac{d\sigma}{dt} = KE_i \quad (7)$$

The charge is related to the two fields E_0 , E_i from the ‘‘pillbox’’ version of $\nabla \cdot \vec{D} = \rho$,

$$\varepsilon_0 E_0 - \varepsilon \varepsilon_0 E_i = \sigma$$

From here we find a differential equation for the free charge density,

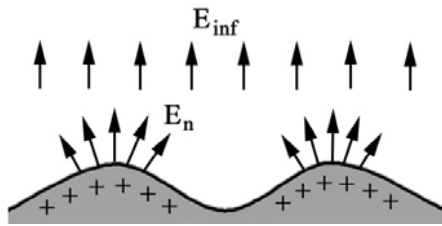
$$\frac{d\sigma}{dt} + \frac{K}{\varepsilon \varepsilon_0} \sigma = \frac{K}{\varepsilon} E_0 \quad (8)$$

The quantity $\tau = \frac{\varepsilon \varepsilon_0}{K}$ is the relaxation time of charges in the liquid. In terms of it, the solution of Eq. (8) that satisfies $\sigma(0) = 0$ (for a constant E_i at $t > 0$) is,

$$\sigma = \varepsilon_0 E_0 (1 - e^{-t/\tau}) \quad (9)$$

Surface Stability

If the liquid surface deforms slightly, the field becomes stronger on the protruding parts, and more charge concentrates there. The traction of the surface field on this charge is $\frac{1}{2}\sigma_s E = \frac{1}{2}\varepsilon_0 E^2$ for a conductor (the $\frac{1}{2}$ accounts for the variation of E from its outside value to 0 inside the liquid). This traction then intensifies on the protruding parts, and the process can become unstable if surface tension γ is not strong enough to counteract the traction. In that case, the protuberance will grow rapidly into some sort of large-scale deformation, the shape of which depends on field shape, container size, etc.



If the surface ripple is assumed sinusoidal, and small (initially at least), then the outside potential, which obeys $\nabla^2 \phi = 0$ with $\phi = 0$ on the surface, can be represented approximately by the superposition of that due to the applied field E_∞ , plus a small perturbation. Using the fact that $Re\{e^{i\alpha z}\}$ is a harmonic function ($z = x + iy$),

$$\phi \approx -E_\infty y + \phi_1 e^{-\alpha y} \cos \alpha x \quad (10)$$

The surface is where $\phi = 0$, and this, when $\alpha y \ll 1$, is approximately given by $0 \approx -E_\infty y + \phi_1 \cos \alpha x$, or,

$$y \approx \frac{\phi_1}{E_\infty} \cos \alpha x \quad (11)$$

The surface has a curvature $\frac{1}{R_c} \approx \left| \frac{d^2 y}{dx^2} \right| = \frac{\phi_1 \alpha^2}{E_\infty} \cos \alpha x$, which is maximum at crests ($\cos \alpha x = 1$),

$$R_c = \frac{E_\infty}{\phi_1 \alpha^2} \quad (12)$$

and gives rise to a surface tension restoring force (perpendicular to the surface) of γ/R_c (cylindrical surface).

The normal field, from Eq. (10), is $E_y = -\frac{\partial \phi}{\partial y} = E_\infty + \alpha \phi_1 e^{-\alpha y} \cos \alpha x$ and at $\alpha y \ll 1$ and on the crests, this is $E_y = E_\infty + \alpha \phi_1$. The perturbation of electric traction (per unit area) is then $\delta \left(\frac{1}{2} \varepsilon_0 E_y^2 \right) = \varepsilon_0 E_\infty \alpha \phi_1$. Instability will occur if this exceeds the restoring surface tension effect,

$$\varepsilon_0 E_\infty \alpha \phi_1 > \gamma \frac{\phi_1 \alpha^2}{E_\infty} \quad \text{or} \quad E_\infty > \sqrt{\frac{\gamma \alpha}{\varepsilon_0}} \quad (13)$$

The quantity α is $2\pi/\lambda$, where λ is the wavelength of the ripple. Thus, if longwave ripples are possible, a small field is sufficient to produce instability. We will later be interested in drawing liquid from small capillaries; if the capillary diameter is D , the largest wavelength will be $2D$, or $\alpha = \pi/D$, which gives the instability condition,

$$E_\infty > \sqrt{\frac{\pi \gamma}{\varepsilon_0 D}} \quad (14)$$

For example, say $D = 0.1$ mm, and $\gamma = 0.05$ N/m (Formamide, CH_3NO). The minimum field to produce an instability is then 1.33×10^7 V/m. This is high, but since the capillary tip is thin (say, about twice its inner diameter, or 0.2 mm), it may take only about 2500 V to generate it. A more nearly correct estimate for this will be given next.

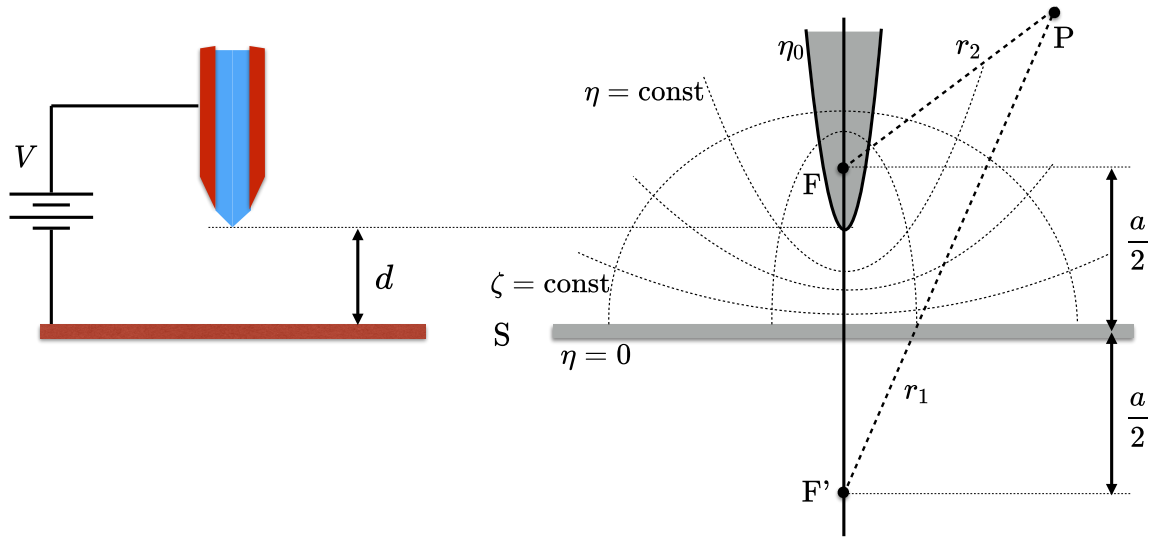
Starting Voltage

The figure shows an orthogonal system of coordinates called ‘‘Prolate Spheroidal Coordinates’’, in which $\eta = \frac{r_1 - r_2}{a}$, $\zeta = \frac{r_1 + r_2}{a}$ and ϕ is an angle about the line FF' .

The cartesian-prolate transformation is,

$$r_1 = \sqrt{x^2 + y^2 + \left(z + \frac{a}{2}\right)^2} \quad \text{and} \quad r_2 = \sqrt{x^2 + y^2 + \left(z - \frac{a}{2}\right)^2}$$

and so, lines of constant η are confocal hyperboloids (foci at F, F') while lines of constant ζ are confocal ellipsoids with the same foci. The surface $\eta = 0$ is the symmetry plane, S , and one of the η -surfaces, $\eta = \eta_0$, can be chosen to represent (at least near its tip) the protruding liquid surface from a capillary.



If the potential ϕ is assumed to be constant (V) on $\eta = \eta_0$, and zero on the plane S , then the entire solution for ϕ will depend on η alone. The η part of Laplace's equation in these coordinates is,

$$\frac{\partial}{\partial \eta} \left[(1 - \eta^2) \frac{\partial \phi}{\partial \eta} \right] = 0 \quad (15)$$

which, with the stated boundary conditions, integrates easily to,

$$\phi = V \frac{\tanh^{-1} \eta}{\tanh^{-1} \eta_0} \quad (16)$$

Let $R^2 = x^2 + y^2$ (cylindrical radius). From $\eta = \frac{r_1 - r_2}{a}$, the (z, R) relationship for a constant η hyperboloid is,

$$a\eta = \sqrt{R^2 + \left(z + \frac{a}{2}\right)^2} - \sqrt{R^2 + \left(z - \frac{a}{2}\right)^2}$$

which, for $z > 0$, can be simplified to,

$$z = \eta \sqrt{\frac{a^2}{4} + \frac{R^2}{1 - \eta^2}}$$

The radius of curvature R_c of this surface is given by,

$$\frac{1}{R_c} = \frac{z_{RR}}{(1 + z_R^2)^{3/2}}$$

which yields,

$$R_c = a \frac{1 - \eta^2}{2\eta} \left[1 + 4 \frac{R^2/a^2}{(1 - \eta^2)^2} \right] \quad (17)$$

Also, the tip-to-plane distance is $d = z(R = 0, \eta = \eta_0) = \frac{a}{2}\eta_0$, so we find the parameters a and η_0 if R_c and d are specified,

$$a = 2d\sqrt{1 + \frac{R_c}{d}} \quad \text{and} \quad \eta_0 = \left(1 + \frac{R_c}{d}\right)^{-1/2} \quad (18)$$

The electric field at the tip is $E_z = -\left.\frac{\partial\phi}{\partial z}\right|_{tip} = -\left.\frac{\partial\phi}{\partial\eta}\frac{\partial\eta}{\partial z}\right|_{tip}$. Now, at the tip we have $R = 0$ and $\eta = \eta_0$, so that $\left.\frac{\partial\eta}{\partial z}\right|_{tip} = \frac{2}{a}$, and using Eq. (16),

$$E_{tip} = -\frac{2V/a}{(1 - \eta_0^2)\tanh^{-1}\eta_0} \quad (19)$$

which can be expressed in terms of R_c and d , when $R_c \ll d$, as,

$$E_{tip} = -\frac{2V/R_c}{\ln(4d/R_c)} \quad (20)$$

Now, in order for the liquid to be electrostatically able to overcome the surface tension forces and start flowing, even with no applied pressure, one needs to have,

$$\frac{1}{2}\varepsilon_0 E_{tip}^2 > \frac{2\gamma}{R_c} \quad (21)$$

($2\gamma/R_c$, because there are two equal curvatures in an axisymmetric tip). Substituting Eq. (20), the ‘‘starting voltage’’ is,

$$V_{start} = \sqrt{\frac{\gamma R_c}{\varepsilon_0}} \ln\left(\frac{4d}{R_c}\right) \quad (22)$$

Returning to the example with $R_c = 0.05$ mm, $\gamma = 0.05$ N/m, and assuming an attractor plane at $d = 5$ mm, the required voltage is 3184 V, whereas if the attractor is brought in to $d = 0.5$ mm, $V_{start} = 1960$ V. These values are to be compared to the previous estimate. They still ignore the effect of space charge in the space between the tip and the plane, which would act to reduce the field at the liquid surface. But we have also ignored the effect of an applied pressure, which can be used to start the flow as well. What an applied pressure cannot do, however, is to trigger the surface instability described before. As Eq. (22) shows, if the radius of curvature at the tip is reduced, so is the required voltage to balance surface tension. One can then expect that, once electrostatics dominates, the liquid surface will rapidly deform from a near-spherical cap to some other shape, with a progressively sharper tip. The limit of this process will be discussed next.

The Taylor Cone

From early experimental observations, it was known that when a strong field is applied to the liquid issuing from the end of a tin tube, the liquid surface adopts a conical shape, with a very thin, fast-moving jet being emitted from its apex. In 1965, G.I. Taylor explained analytically

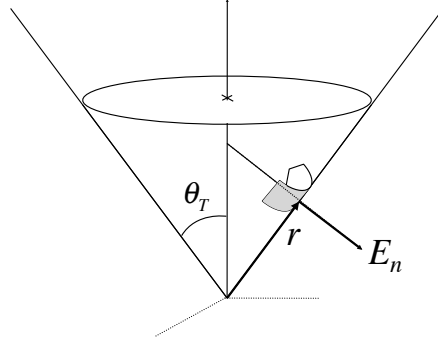
(and verified experimentally) this behavior, and the conical tip often seen in electrospray emitters is now called a “Taylor Cone”. The basic idea is that the surface “traction” $\varepsilon_0 E^2/2$ due to the electric field must be balanced everywhere on the conical surface by the pull of the surface tension. The latter is per unit of area, $\gamma (R_{c1}^{-1} + R_{c2}^{-1})$, where $1/R_{c1}$ and $1/R_{c2}$ are the two principal curvatures of the surface. In a cone, $1/R_c$ is zero along the generator, while the curvature of the normal section is the projection on it of that of the circular section through the same point (Meusnier’s theorem),

$$\frac{1}{R_c} = \frac{\cos \theta}{R} = \frac{\cos \theta}{r \sin \theta} = \frac{\cot \theta}{r} \quad (23)$$

This means that,

$$\frac{1}{2} \varepsilon_0 E_n^2 = \frac{\gamma \cot \theta}{r} \quad \text{or} \quad E_n = \sqrt{\frac{2\gamma \cot \theta}{\varepsilon_0 r}} \quad (24)$$

The question then is to find an external electrostatic field such that the cone is an equipotential, with a normal field varying as in Eq. (24). Notice that the spheroids of the previous section do generate cones in the limit when $r \gg a$ but this type of electrostatic field has $E_n \propto 1/r$, and cannot be the desired equilibrium solution.



If we adopt a spherical system of coordinates, Laplace’s equation admits axisymmetric “product” solutions of the type,

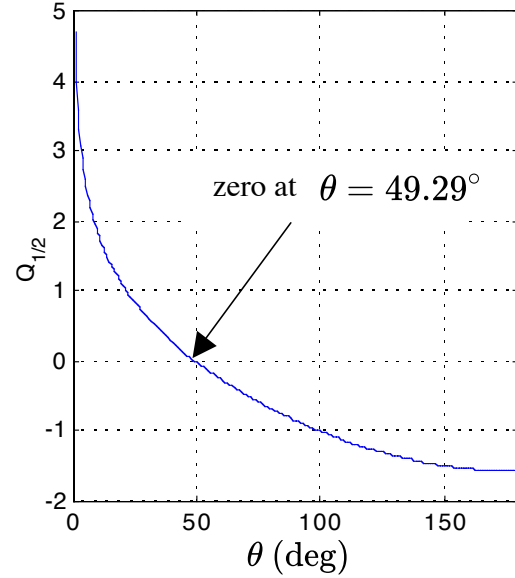
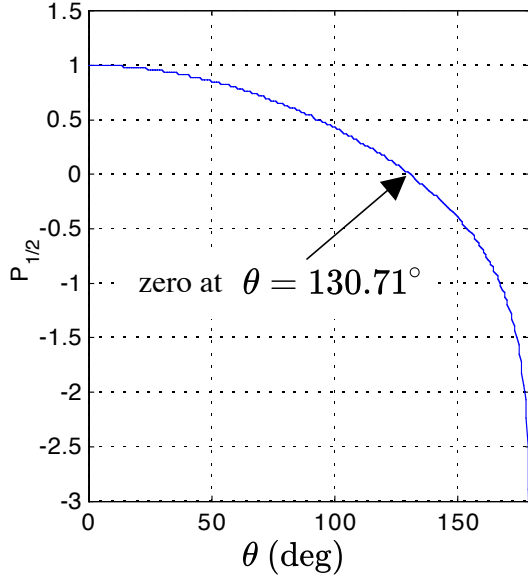
$$\phi \propto Q_\nu(\cos \theta) r^\nu \quad \text{or} \quad \phi \propto P_\nu(\cos \theta) r^\nu \quad (25)$$

where P_ν and Q_ν are Legendre functions of the 1st and 2nd kind, respectively. Of the two, P_ν has a singularity when $\theta = \pi$, and Q_ν has one at $\theta = 0$. The latter is acceptable, because $\theta = 0$ is inside the liquid cone, and we only need the solution outside. The normal field is then,

$$E_n = E_\theta = -\frac{1}{r} \frac{\partial \phi}{\partial \theta} = A \frac{dQ_\nu}{d \cos \theta} \sin \theta r^{\nu-1}$$

where A is a constant. In order to have $E_n \propto 1/\sqrt{r}$, we need $\nu = 1/2$. Thus,

$$\phi = A Q_{1/2}(\cos \theta) r^{1/2} \quad (26)$$



The function $Q_{1/2}(\cos \theta)$ has a single zero at,

$$\theta_T = 49.29^\circ \quad (27)$$

which can therefore be taken as the equipotential liquid surface. Notice that this angle is universal (independent of fluid properties, applied voltage, etc). Taylor (and others) have verified experimentally this value, as long as no strong space charge effects are present, no flow, and as long as the electrode geometry is “reasonably similar” to what is implied by Eq. (26).

The experimental fact that stable Taylor cones do form even when the electrodes applying the voltage are substantially different from the shape given by Eq. (26) apparently indicates that the external potential distribution near the cone is dictated by the equilibrium condition Eq. (24), and that the transition to some other potential distribution capable of matching the real electrode shape takes place far enough from the liquid to be of little consequence. We should expect, however, that the Taylor cone solution will be disturbed to some extent by nonideal conditions, and will eventually disappear. In one respect at least, the Taylor cone cannot be an exact solution: the infinite electric field predicted at the apex will produce various physical absurdities. Something must yield before that point, and that is explored next.

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