Appendix to "Energy dissipation in electrosprays and the geometric scaling of the transition region of cone-jets".

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Journal of Fluid Mechanics, vol XXX(2010), pp 95-100

The equations of conservation of mass and charge, the Navier-Stokes equations, and the Laplace equation for the electrical potential completely describe the hydrodynamics and geometry of the cone-jet (Melcher & Taylor 1969, Saville 1997, Higuera 2003). The steady-state momentum equation and the resulting equation of mechanical energy for an incompressible fluid can be written as

$$\rho\,\mathbf{v}\cdot\nabla\mathbf{v} = -\nabla p + \nabla\cdot\boldsymbol{\tau}_{\mu} + \nabla\cdot\boldsymbol{\tau}_{M} \qquad \text{(A.1)}$$

$$\rho\nabla\cdot(\mathbf{v}\,\mathrm{v}^2/2) = -\nabla\cdot(p\mathbf{v}) + \nabla\cdot(\boldsymbol{\tau}_{\mu}\cdot\mathbf{v}) - \boldsymbol{\tau}_{\mu}\mathbf{:}\nabla\mathbf{v} + \mathbf{v}\cdot\nabla\cdot\boldsymbol{\tau}_{M} \qquad \text{(A.2)}$$

The electrical forces acting on the fluid are modeled with the Maxwell stress tensor $\boldsymbol{\tau}_M$

$$\boldsymbol{\tau}_M = \varepsilon\varepsilon_0\mathbf{E}\mathbf{E} - \frac{1}{2}\varepsilon\varepsilon_0\left[1 - \frac{\rho}{\varepsilon}\left(\frac{\partial\varepsilon}{\partial\rho}\right)_T\right]\mathbf{E}\cdot\mathbf{E}\,\boldsymbol{\delta} \qquad \text{(A.3)}$$

which yields the volumetric force

$$\nabla\cdot\boldsymbol{\tau}_M = \frac{1}{2}\varepsilon_0\nabla\left[\rho\left(\frac{\partial\varepsilon}{\partial\rho}\right)_T E^2\right] - \frac{1}{2}\varepsilon_0 E^2\nabla\varepsilon + \rho^e\mathbf{E} \qquad \text{(A.4)}$$

This force and the associated terms in the momentum and mechanical energy equations are usually ignored, because to a high degree of accuracy the electrostriction force in the incompressible fluid and the charge density ρ^e can be regarded as negligible, and the dielectric constant can be treated as uniform throughout the fluid. Thus the coupling between electrostatic forces and hydrodynamics is a surface effect, and is enforced by requiring that the free surface of the cone-jet is in mechanical equilibrium. The balance of stresses in the interface Σ_0 (see figure 4(b)) can be written as

$$(\gamma\boldsymbol{\delta}\nabla\cdot\mathbf{n} - p\boldsymbol{\delta} + \boldsymbol{\tau}_\mu + \boldsymbol{\tau}_M^i)\cdot\mathbf{n} = \boldsymbol{\tau}_M^o\cdot\mathbf{n} \qquad \text{(A.5)}$$

where **n** is the outward normal vector and the superscripts *i* and *o* denote the liquid bulk and the surrounding vacuum respectively. In particular, the tangential component of (A.5) in the *rz* plane is

$$\mathbf{t}\cdot\boldsymbol{\tau}_\mu\cdot\mathbf{n} = \mathbf{t}\cdot(\boldsymbol{\tau}_M^o - \boldsymbol{\tau}_M^i)\cdot\mathbf{n} = \varepsilon_0 E_z(E_r^o - \varepsilon E_r^i) = \sigma E_z \qquad \text{(A.6)}$$

$$\mathbf{t}\cdot\boldsymbol{\tau}_\mu\cdot\mathbf{n} = 2\mu\left.\frac{\partial \mathrm{v}_z}{\partial r}\right|_{\Sigma_0} \qquad \text{(A.7)}$$

where σ stands for the surface charge density in the interface.

The macroscopic balance of the mechanical energy in the cone-jet is obtained by integrating (A.2) over the volume Π bounded by the surfaces Σ_0, Σ_1, and Σ_2 (see figure 4(b)). The integration of every term with the exception of $\nabla\cdot(\boldsymbol{\tau}_\mu\cdot\mathbf{v})$ is trivial:

$$\int_\Pi \rho\nabla\cdot(\mathbf{v}\frac{\mathrm{v}^2}{2})\mathrm{d}V = \int_{\Sigma_0,\Sigma_1,\Sigma_2} \rho\frac{\mathrm{v}^2}{2}\mathbf{v}\cdot\mathrm{d}\mathbf{A} = 0 - \rho\mathrm{Q}\frac{\mathrm{v}_1^2}{2} + \rho\mathrm{Q}\frac{\mathrm{v}_2^2}{2} \qquad \text{(A.8)}$$

$$\int_\Pi \nabla\cdot(p\mathbf{v})\mathrm{d}V = \int_{\Sigma_0,\Sigma_1,\Sigma_2} p\mathbf{v}\cdot\mathrm{d}\mathbf{A} = 0 - \mathrm{Q}\,p_1 + \mathrm{Q}\,p_2 \qquad \text{(A.9)}$$

$$\int_\Pi \boldsymbol{\tau}_\mu\mathbf{:}\nabla\mathbf{v}\,\mathrm{d}V = P_\mu \qquad \text{(A.10)}$$

P_μ is the net viscous power dissipated in the cone-jet. To integrate $\nabla\cdot(\boldsymbol{\tau}_\mu\cdot\mathbf{v})$ we make use of the balance of stresses in Σ_0, equation (A.6):

$$\int_\Pi \nabla\cdot(\boldsymbol{\tau}_\mu\cdot\mathbf{v})\mathrm{d}V = \int_{\Sigma_0,\Sigma_1,\Sigma_2}(\boldsymbol{\tau}_\mu\cdot\mathbf{v})\cdot\mathrm{d}\mathbf{A} = \int_{\Sigma_0}\sigma E_z \mathrm{v}\mathrm{d}A - 2\mu\mathrm{Q}\left.\frac{\partial \mathrm{v}_z}{\partial z}\right|_{\Sigma_1} + 2\mu\mathrm{Q}\left.\frac{\partial \mathrm{v}_z}{\partial z}\right|_{\Sigma_2} \qquad \text{(A.11)}$$

where we have assumed flat velocity profiles at Σ_1 and Σ_2. The last two terms in (A.11) are negligible because the viscous stresses rapidly decay both upstream and downstream of the transition region. To advance further we write the differential of area as $\mathrm{d}A = 2\pi R \mathrm{d}s$ (s is the arc length of the generatrix g_0 measured from the intersection with Σ_1)

$$\int_{g_0} 2\pi R \,\mathrm{v}\sigma(E_z \mathrm{d}s) = \int_{g_0} I_s(-\mathrm{d}\phi) \tag{A.12}$$

and notice that charge conservation requires the sum of the surface and bulk conduction currents crossing any $r\theta$ plane to be constant, and equal to the total electrospray current I

$$\int_{g_0} I_s(-\mathrm{d}\phi) = -\int_{g_0} (I - I_c)\mathrm{d}\phi = I(\phi_1 - \phi_2) + \int_{g_0} I_c \mathrm{d}\phi \tag{A.13}$$

We now integrate the last term by parts to obtain

$$\int_{g_0} I_c \mathrm{d}\phi = \int_{g_0} \mathrm{d}(I_c\phi) - \int_{g_0} \phi \,\mathrm{d}I_c = I_c\phi\big|_{\Sigma_2} - I_c\phi\big|_{\Sigma_1} + \int_{\Sigma_0} \phi K \mathbf{E}^i \cdot \mathrm{d}\mathbf{A} \tag{A.14}$$

In this step we have used the equation of conservation of charge in differential form, which requires the variation of the conduction current to be balanced by the charge injected onto the surface, $\mathrm{d}I_c = -K\mathbf{E}^i \cdot \mathrm{d}\mathbf{A}\big|_{\Sigma_0}$. Note that I_c asymptotes to the total current I upstream in the Taylor cone, and is negligible downstream of the transition region. We now use the Gauss theorem to evaluate the last integral in (A.14)

$$\int_{\Sigma_0} \phi K \mathbf{E}^i \cdot \mathrm{d}\mathbf{A} = \int_{\Pi} \nabla \cdot (\phi K \mathbf{E}) \mathrm{d}V - \int_{\Sigma_1, \Sigma_2} \phi K \mathbf{E}^i \cdot \mathrm{d}\mathbf{A} = -\int_{\Pi} \mathbf{J} \cdot \mathbf{E} \,\mathrm{d}V + I_c\phi\big|_{\Sigma_1} - I_c\phi\big|_{\Sigma_2\,1} \tag{A.15}$$

$$\int_{\Pi} \nabla \cdot (\phi K \mathbf{E}) \mathrm{d}V = \int_{\Pi} K\phi \nabla\cdot \mathbf{E} \;\mathrm{d}V + \int_{\Pi} K\mathbf{E}\nabla\phi \;\mathrm{d}V = 0 - \int_{\Pi} \mathbf{J} \cdot \mathbf{E} \,\mathrm{d}V$$

$$\int_{\Sigma_1, \Sigma_2} \phi K \mathbf{E}^i \cdot \mathrm{d}\mathbf{A} = -I_c\phi\big|_{\Sigma_1} + I_c\phi\big|_{\Sigma_2}$$

The last integral in (A.15) is the net ohmic dissipation in the cone-jet, P_Ω. Thus, the sought integral of $\nabla \cdot (\boldsymbol{\tau}_\mu \cdot \mathbf{v})$ over the volume of the cone-jet can be written as

$$\int_{\Pi} \nabla \cdot (\boldsymbol{\tau}_\mu \cdot \mathbf{v}) \mathrm{d}V = I(\phi_1 - \phi_2) - \int_{\Pi} \mathbf{J} \cdot \mathbf{E} \;\mathrm{d}V = I(\phi_1 - \phi_2) - P_\Omega \tag{A.16}$$

and the macroscopic balance of the mechanical energy is finally obtained by combining (A.8), (A.9), (A.10) and (A.16):

$$\rho Q(\frac{v_2^2}{2} + \frac{p_2}{\rho}) - \rho Q(\frac{v_1^2}{2} + \frac{p_1}{\rho}) = I(\phi_1 - \phi_2) - P_\Omega - P_\mu \qquad (A.17)$$

Thus, the total electrical energy transferred to the cone-jet is partially converted into mechanical energy, and partially degraded by viscous and ohmic dissipation.

The macroscopic balance of the internal energy can be derived by using the first law of thermodynamics for an open system. Conservation of energy in the control volume bounded by the surfaces Σ_0, Σ_1 and Σ_2 requires the net flux of energy (internal, kinetic and surface energy) exiting the control volume to be balanced by the net rate of heat and electrical energy addition, combined with the rate of work done by the stresses on Σ_1 and Σ_2 (pressure work and surface tension work, we again neglect the work done by the viscous stresses):

$$Q(\rho u_2 + \rho \frac{v_2^2}{2} + \frac{2\gamma}{R_2}) - Q(\rho u_1 + \rho \frac{v_1^2}{2} + \frac{2\gamma}{R_1}) = I(\phi_1 - \phi_2) + Q(p_1 - \frac{2\gamma}{R_1}) - Q(p_2 - \frac{2\gamma}{R_2}) + \dot{q} \qquad (A.18)$$

u and $\dot{q}$ stand for the internal energy per unit mass and the net rate of heat addition. It is worth noting that in each boundary the flux of surface energy is exactly balanced by the rate of work done by the resultant of the surface tension. The macroscopic balance of mechanical energy can now be subtracted from (A.18) to yield the following macroscopic balance for the fluid's internal energy, and its temperature change

$$u_2 - u_1 = c\,(T_2 - T_1) = \frac{P_\Omega + P_\mu + \dot{q}}{Q\rho} \qquad (A.19)$$

c is the heat capacitance per unit mass. The viscous dissipation of mechanical energy and the ohmic dissipation of electrical energy increase the internal energy of the fluid. Furthermore, the heat fluxes across the boundaries of our cone-jets (radiation through Σ_0, and conduction through Σ_1 and Σ_2) are likely negligible.