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Asymptotic decay of radius of a weakly conductive viscous jet in an external electric field

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Motion of a weakly conductive viscous jet accelerated by an external electric field is considered. Nonlinear rheological constitutive equation applicable for polymer fluids (Oswald–deWaele law) is applied. A differential equation for the variation of jet radius with axial coordinate is derived. Asymptotic variation of the jet radius at large distances from the jet origin is analyzed. It is found that the well-known power-law asymptote for Newtonian fluids with the exponent 1/4 holds for more general class of fluids, i.e., pseudoplastic (shear thinning) and dilatant (shear thickening) fluids with the flow index between 0 and 2. Dilatant fluids with the flow index greater than 2 exhibit power-law asymptotes with the exponents depending on the flow index. Results can be applied for the analysis of viscous polymer jets in the electrospinning process. [S0003-6951(98)04447-7]

The flow of liquid jets deformed and accelerated by an external electric field is a coupled electromechanics problem that has attracted considerable interest.[1–4] Excellent reviews on this topic can be found in Refs. 2–4. Electrostatically driven jets are involved in a variety of applications, including electrostatic atomization of liquids.[5] A method of electrostatic drawing of polymer fibers, called electrospinning,[6] is another application. In the latter, a charged jet of a polymer solution or melt is ejected from a capillary tube. The jet is elongated and accelerated by an external electric field, deposited on a substrate, and dried and/or chemically treated to convert it into a thin fiber. Recently, electrospinning was extensively studied experimentally by Reneker *et al.*[6,7] Fibers of over thirty synthetic and natural polymers were spun by this method.

Behavior of electrostatically driven jets at large distances from their origin has not yet been sufficiently studied due to the fact that low-viscosity, low-molecular weight fluids, utilized in most electrostatic jet applications, break up into droplets long before the jet reaches its asymptotic length. This breakup is due to the longitudinal Rayleigh instability, caused by surface capillary waves.[8] The Rayleigh instability is not typically observed in electrospinning of polymer fluids.[6,7] However, a transverse instability or splaying of the jet into two or more smaller jets is sometimes observed. As a rule, the transverse jet splaying occurs further away from the jet origin. Therefore, peculiarities of jet flow at large distances are important for jet splaying analysis. In addition, asymptotic results can be used to evaluate diameters of polymer fibers electrospun in a single-jet flow regime.

A simple model of an electrostatically driven Newtonian jet was developed in Ref. 9. Asymptotic behavior of the model was evaluated under the assumption that the effects of the viscous forces are negligible (ideal liquid approximation). The power-law asymptote with the exponent 1/4 was obtained for the jet radius

$$R \sim z^{-1/4}. \tag{1}$$

The result was experimentally confirmed for several fluids.[9] However, the polymer fluids are highly viscous and the effects of the viscous forces cannot be neglected. In addition, the polymer fluids often exhibit nonlinear rheologic behavior.[10] In the present letter, a broader class of fluids described by the nonlinear power-law rheologic constitutive equation is considered. A model of the jet motion is formulated taking into account inertial, hydrostatic, viscous, electric, and surface tension forces. Asymptotic behavior of the jet at large distances is analyzed.

Consider an infinite viscous jet pulled from the capillary orifice and accelerated by a constant external electric field. Neglecting the magnetic effects, the general three-dimensional linear momentum balance equation for the jet element is

$$\rho(\vec{\boldsymbol{\nu}}\cdot\nabla)\vec{\boldsymbol{\nu}}+\nabla p=\nabla\hat{\tau}^{c}+\nabla\hat{\tau}^{e}, \tag{2}$$

where ρ is fluid density, $\vec{\boldsymbol{\nu}}$ is velocity, p is hydrostatic pressure, $\hat{\tau}^c$ is viscous stress tensor, and $\hat{\tau}^e$ is electric stress tensor. Rheologic behavior of many fluids, including the polymer fluids, can be described by the power-law constitutive equation, known as the Oswald–deWaele law[8,10]

$$\hat{\tau}^{c}=\mu[tr(\hat{\dot{\gamma}}^{2})]^{(m-1)/2}\hat{\dot{\gamma}}. \tag{3}$$

Here, μ is a constant, $\hat{\dot{\gamma}}$ is rate of strain tensor, m is flow index. Viscous Newtownian fluids are described by a special case of Eq. (3) with the flow index $m=1$. Pseudoplastic (shear thinning) fluids are described by the flow indices $0 \leqslant m < 1$.[11] Dilatant (shear thickening) fluids are described by the flow indices $m > 1$.[11] The differential equation of momentum balance [Eq. (2)] is complemented by the equations of mass balance, electric charge balance, and the electrostatic field equation.[11,12]

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Assume that the jet flow is an extensional axisymmetric flow in the direction of the external electric field. Denote z as the axial coordinate. Consider a weakly conductive jet. In such a jet, electrical current due to electronic or ionic conductivity of the fluid is small compared to the current provided by the convective charge transfer with moving jet particles. However, the conductivity is sufficient for the electric charges to migrate the short distance to the jet surface. The bulk electric charge can then be assumed zero, in the asymptotic limit. The surface charge will interact with the external electric field creating the pulling force responsible for jet acceleration. In addition, the surface charge will cause transverse electric repulsion that will lower the hydrostatic pressure. The overall electric potential, ϕ, can be obtained as a sum of the potential of the surface charge, ϕ_s, and the potential of the external field, $\phi_{\mathrm{ext}}=-E_0 z$, where E_0 is the electric field.

Assume that the slope of the jet surface in the direction of the flow is small, $dR/dz \ll 1$, where R is the jet radius. Further, assume that the effect of the surface charge on the axial component of the electric field is negligible. The linear momentum balance [Eq. (2)] can then be averaged over the jet cross-section. Detailed description of the averaging procedure can be found in Ref. 8. The resulting equation in the axial direction is

$$\frac{d}{dz}\left[\frac{\rho}{2}\pi R^2 v^2+\pi R^2 p-\mu\pi R^2\left(\left|\frac{dv}{dz}\right|\right)^{m-1}\frac{dv}{dz}\right]=2\pi R\Omega E_0, \tag{4}$$

where v is average jet velocity in the cross section and Ω is the surface charge density. The average hydrostatic pressure in the cross section is determined by the surface tension and transverse electric repulsion. For a slender jet, it is approximated by

$$p=\frac{\sigma_s}{R}-\frac{\Omega^2}{2\epsilon_0}, \tag{5}$$

where σ_s is the surface tension coefficient and ϵ_0 is the dielectric permeability of vacuum. Equation (5) can be obtained by averaging Eq. (2) in the radial direction.

Averaging the mass balance equation yields $\pi R^2 v=Q$, where Q is the constant volumetric flow rate. Similarly, the electric charge balance equation reduces to $2Q\Omega/R-\pi\sigma R^2 d\phi/dz=I$, where σ is the electrical conductivity of the fluid and I is the constant total electric current. In the weakly conductive jet, the electric current is defined primarily by the convective charge transfer. Therefore in the asymptotic limit, the electric current can be approximated by $I=2Q\Omega/R$ and the ratio of the surface charge density and jet radius is constant $\Omega/R=I/2Q$. The flow rate, Q, and the electric current, I, are considered external parameters of the problem.

Let us introduce the dimensionless jet radius $\tilde R=R/R_0$ and axial coordinate $\tilde z=z/z_0$, where $z_0=\rho Q^3/2\pi^2 R_0^4 E_0 I$. The characteristic jet radius, R_0, is sometimes taken equal to the radius of the capillary orifice.[3,10] Equations (4) and (5) reduce to the following dimensionless equation for the jet radius:

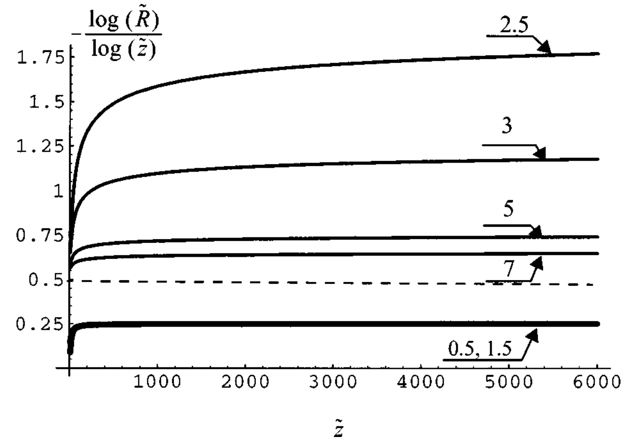

FIG. 1. Results of numerical integration of Eq. (6) for the flow indices $m=0.5$, 1.5, 2.5, 3, 5, 7.

$$\frac{d}{d\tilde z}\left\{\tilde R^{-4}+We\tilde R^{-1}-\Upsilon\tilde R^{2}-\frac{1}{\mathrm{Re}}\left(\frac{1}{2}\frac{d}{d\tilde z}(\tilde R^{-2})\right)^{m}\right\}=1. \tag{6}$$

The dimensionless parameters in Eq. (6) are defined as follows. The Weber number, $We=(2\pi^2R_0^3\sigma_s)/(\rho Q^2)$, describes the ratio of the surface tension forces to the inertia forces. The parameter $\Upsilon=\pi^2 I^2 R_0^6)/(4\epsilon_0\rho Q^4)$ describes the ratio of the electric forces to the inertia forces. The effective Reynolds number for the fluid characterized by the power-law constitutive Eq. (3), $\mathrm{Re}=(Q^2\rho)/(2\pi^2R_0^4\mu)[(4\pi E_0 I R_0^2)/(Q^2\rho)]^{-m}$, describes the ratio of the inertia forces to the viscous forces.

The Bernoulli integral obtained from Eq. (6) is

$$\tilde R^{-4}+We\tilde R^{-1}-\Upsilon R^2-\frac{1}{\mathrm{Re}}\left(\frac{1}{2}\frac{d}{d\tilde z}\tilde R^{-2}\right)^{m}=\tilde z+C, \tag{7}$$

where the integration constant, C, is determined by the boundary condition. A general closed-form solution of Eq. (7) is not available.

By analogy with Ref. 9, let us consider the power-law asymptotic approximation of the jet radius

$$\tilde R\sim\tilde z^{-\alpha}, \tag{8}$$

where the exponent, α, is a positive constant. Substituting Eq. (8) into Eq. (7) gives

$$\tilde z^{4\alpha}+We\tilde z^{\alpha}-\Upsilon\tilde z^{-2\alpha}-\frac{\alpha^m}{\mathrm{Re}}\tilde z^{(2\alpha-1)m}-\tilde z=O(1). \tag{9}$$

The power balance at $\tilde z\rightarrow+\infty$ yields

$$4\alpha=\max[1,(2\alpha-1)m]. \tag{10}$$

For pseudoplastic and dilatant fluids with the flow index m in the range from zero to two, the solution of Eq. (10) is $\alpha=1/4$. That coincides with the asymptotic solution for Newtonian fluids obtained in Ref. 9. For dilatant fluids with the flow index greater than 2, the solution is $\alpha=\frac{1}{2}m/(m-2)$. These two solutions describe two asymptotic regimes of jet motion. In the first regime, the influence of viscosity at large distances is small and the electric field work is fully transformed into the kinetic energy of the jet. In the second regime, the viscous stresses prevail at large distances and asymptotic flow behavior depends on the parameter of the constitutive equation of the fluid.

The analytic solutions presented by Eqs. (8) and (10) are verified by the numerical integration of Eq. (6). The following dimensionless constants are chosen based on typical electrospinning parameters:[7] $\mathrm{Re}=0.10$, $We=10$, $\Upsilon=10$. The boundary condition is $\tilde{R}|_{\tilde{z}=0}=1$. Results of the numerical integration for several flow indices m are shown in Fig. 1. It is seen that at large distances, the ratio $\log(1/\tilde{R})/\log\tilde{z}=-\log\tilde{R}/\log\tilde{z}$ tends to a constant. For the flow indices $m<2$, the asymptote is independent of m and equals $\alpha=1/4$. For the flow indices $m>2$, the asymptote depends on m. As m increases, the asymptotic exponent decreases, approaching the value $\alpha=1/2$. Overall, the results of numerical simulation corroborate with the analytic expressions.

The analysis presented here extends the limit of applicability of the power-law asymptote,[9] Eq. (1), to pseudoplastic and dilatant fluids with the flow index m between 0 and 2. More complicated asymptotic behavior of the dilatant fluids with the flow index $m>2$ is discovered. The obtained results can be used as a basis for stability analysis of viscous polymer jets.

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[1] G. Taylor and M. D. van Dyke, Proc. R. Soc. London, Ser. A **313**, 453 (1969).
[2] M. Cloupeau and B. Prunet-Foch, J. Aerosol Sci. **25**, 1021 (1994).
[3] A. M. Ganan-Calvo, J. Davila, and A. Barrero, J. Aerosol Sci. **28**, 249 (1997).
[4] D. A. Saville, Annu. Rev. Fluid Mech. **29**, 27 (1997).
[5] A. G. Bailey, *Electrostatic Spraying of Liquids* (Wiley, New York, 1988).
[6] J. Doshi and D. H. Reneker, J. Electrost. **35**, 151 (1995).
[7] D. H. Reneker and I. Chun, Nanotechnology **7**, 216 (1996).
[8] A. L. Yarin, *Free Liquid Jets and Films: Hydrodynamics and Rheology* (Longman, New York, 1993).
[9] V. N. Kirichenko, I. V. Petryanov-Sokolov, N. N. Suprun, and A. A. Shutov, Sov. Phys. Dokl. **31**, 611 (1986).
[10] R. Darby, *Viscoelastic Fluids* (Dekker, New York, 1976).
[11] J. R. Melcher and G. I. Taylor, Annu. Rev. Fluid Mech. **1**, 111 (1969).
[12] S. R. de Groot and L. G. Suttorp, *Foundation of Electrodynamics* (North-Holland, Amsterdam, 1972).