J. Fluid Mech. (1982), *vol.* 120, *pp.* 245–266
Printed in Great Britain

Surface-tension-driven breakup of viscoelastic liquid threads

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(Received 25 February 1981 and in revised form 10 August 1981)

A linearized stability analysis is carried out for the breakup of small-diameter liquid filaments of dilute polymer solutions into droplets. Oldroyd's 8-constant model expressed in a corotational reference frame is used as the rheological equation of state. The crucial idea in this theory is the recognition that the liquid may be subject to an unrelaxed axial tension due to its prior history. If the tension is zero, the present analysis predicts that jets of shear-thinning liquids are less stable than comparable jets of Newtonian liquids; this is in agreement with previous analyses. However, when the axial tension is not zero, and provided the stress relaxation time constant is sufficiently large, the new theory predicts that the axial elastic tension can be a significant stabilizing influence. With reasonable values for the tension and stress relaxation time the theory explains the great stability observed for jets of some shear-thinning, dilute polymer solutions. The theory explains why drops produced from jets of such liquids are larger than drops from corresponding Newtonian liquids. The theory also appears capable of explaining the sudden appearance of irregularly spaced bulges on jets after long distances of travel with little amplification of disturbances.

1. Introduction

This paper presents a linearized stability analysis for the breakage of viscoelastic liquid filaments into droplets. Our interest in this problem stems from the fact that the disintegration of accidentally released liquid fuels into fine mists poses a serious fire hazard. Addition of 'anti-misting' polymeric additives to the fuel to suppress disintegration is under active study (Weatherford & Wright 1975; San Miguel 1978). However, the mechanisms by which these additives work are not well understood, so that it is difficult to select polymers which are effective anti-misting agents and whose addition leads to other properties required of the modified fuel. A theoretical and experimental study of the breakup of small-diameter liquid filaments of dilute polymer solutions seems warranted for the following reasons. (i) The formation of small-diameter liquid filaments from larger volumes of accidentally released liquid fuels is an important step in the disintegration process. (Giffen & Muraszew 1953; Hoyt & Taylor 1977*a*, *b*; Altman, Hoyt & Taylor 1979). (ii) Experiments with capillary jets could serve as a rapid and reliable method of screening polymeric additives to select promising candidates for study in more difficult or expensive experiments. (iii) Experiments with capillary jets should lead to better understanding of those physical properties of polymeric additives that are responsible for enhanced stability, and could thereby lead to improved additives. This paper deals only with the theory of jet breakup. An experimental study is underway; the results will be reported at a later date.

Linearized stability analysis of surface-tension-driven breakup of jets of Newtonian liquids is reasonably well understood. In such analyses the jet is presumed to be subjected to small disturbances of the form $\exp(\beta t + i2\pi z'/l)$, where β is the amplification ($\beta > 0$) or damping ($\beta < 0$) factor for growth in time, l is the wavelength of the disturbance, t is time, and z' is the axial distance measured in a co-ordinate system moving with the jet. The linearized equations of motion and boundary conditions constitute an eigenvalue problem which relates β to l and the other relevant variables. These variables include jet radius a, jet velocity U, coaxial gas velocity $\hat{U}$, gas density $\hat{\rho}$, liquid density ρ, surface tension σ, and the rheological properties of the liquid. For a Newtonian liquid only a single rheological parameter is needed, the (constant) shear viscosity η. Most experiments involve spatial growth rather than temporal growth. The eigenvalue problem relating β and l is identical for temporal growth, but for spatial growth $\beta = i\omega$, where the frequency ω of the disturbance is a real number, and $k = 2\pi/l$ is regarded as a complex function of ω and the other variables. The sign of the imaginary part of k determines which frequencies amplify or damp. If the amplification rate is such that the change in amplitude over a distance on the order of the wavelength is small, then numerical results for temporal growth in a moving co-ordinate system are in close correspondence with numerical results for spatial growth. The requirement for this is $\beta l/U \ll 1$, a requirement satisfied for many experimental studies. Once the relationship between β and l is known, the diameter D of the resulting drops and the length L from the orifice to the point of drop formation can be estimated as follows:

$$D = (6la^2)^{\frac{1}{3}}, \quad L = (U/\beta)\ln(a/\epsilon). \tag{1), (2}$$

Here ϵ is the amplitude of the disturbance at the orifice and β is assumed to be constant from the orifice to the point of drop formation.

The first stability analysis of this nature was done by Rayleigh (1879), who examined the breakup of a jet of inviscid liquid having no aerodynamic interaction with the surrounding gas. Rayleigh found that

$$\beta^2 = (\sigma/\rho a^3)(1-\xi^2)\,\xi I_1(\xi)/I_0(\xi), \tag{3}$$

where the wavenumber

$$\xi = 2\pi a/l, \tag{4}$$

and $I_1(\xi)$ and $I_0(\xi)$ are modified Bessel functions of the first kind. For wavenumbers greater than unity, the solutions for β are imaginary, so that short-wavelength disturbances oscillate but do not grow or damp in this inviscid case. For wavenumbers less than unity β has a positive real root, so that disturbances with wavelengths greater than the circumference of the jet are unstable. Rayleigh showed that the wavenumber of greatest growth rate is very nearly $\xi \simeq \sqrt{\frac{1}{2}} = 0{\cdot}707$, and that at this wavenumber β is very nearly given by $\beta \simeq (\sigma/8\rho a^3)^{\frac{1}{2}}$. It is convenient to choose $(8\rho a^3/\sigma)^{\frac{1}{2}}$ as a characteristic time and $2a$ as a characteristic length. In terms of these quantities we define the following dimensionless groups, which are conventional in the study of capillary jets:

$$B = \beta(8\rho a^3/\sigma)^{\frac{1}{2}}, \quad Z = \eta/(2\rho a\sigma)^{\frac{1}{2}}, \tag{5), (6}$$

$$W = 2\rho a U^2/\sigma, \quad \hat{W} = 2\hat{\rho}a(\hat{U}-U)^2/\sigma. \tag{7), (8}$$

B is a dimensionless amplification factor, Z is the Ohnesorge number used to characterize the influence of liquid viscosity, W is the Weber number used to characterize the influence of liquid inertia, and $\hat{W}$ is a Weber number for the gas used to characterize the aerodynamic interaction of the jet with the surrounding gas.

Rayleigh's analysis was extended by Weber (1931) to include the destabilizing influence of aerodynamic interactions of the corrugated liquid jet with the surrounding gas and the stabilizing influence of the liquid viscosity. Sterling & Sleicher (1975) repeated Weber's analysis but without the approximations introduced by Weber which limited his analysis to small values of Z. For $\hat{\rho} \ll \rho$, Sterling & Sleicher's result can be expressed in terms of the above dimensionless groups as:

$$B^2\mathscr{I}(\xi) + 4BZ\xi^2\{4\mathscr{I}(\xi) - 1 + Q(\xi,\xi_1)\} = 4\xi^2\{1-\xi^2+\hat{W}\xi K_0(\xi)/2K_1(\xi)\}, \tag{9}$$

where

$$\xi_1^2 = \xi^2 + B/4Z, \tag{10}$$

$$\mathscr{I}(\xi) = \xi I_0(\xi)/2I_1(\xi), \tag{11}$$

$$Q(\xi,\xi_1) = 4\xi^2\{\mathscr{I}(\xi)-\mathscr{I}(\xi_1)\}/\{\xi_1^2-\xi^2\}, \tag{12}$$

and $K_0(\xi)$ and $K_1(\xi)$ are modified Bessel functions of the second kind. In Weber's approximation $\mathscr{I}(\xi)$ is approximated as unity for $\xi < 1$ and $Q(\xi,\xi_1)$ is approximated as zero for $\xi_1 \gg 1$, i.e. $Z \ll 1$. Computations of $B(\xi, Z, \hat{W})$ from (9) show that, at constant Z as $\hat{W}$ increases the dimensionless amplification factor also increases, indicating shorter jet lengths; that disturbances with wavenumbers greater than unity become unstable; and that the wavenumber for maximum growth shifts to large values, indicating smaller droplets. At constant $\hat{W}$, as Z increases the dimensionless amplification factor decreases, indicating longer jet lengths, and the wavenumber for maximum growth shifts to smaller wavenumbers, indicating larger droplets. For example with $\hat{W} = 0$ and $Z = 0$, the maximum value of B is 0·971, occurring at $\xi = 0{\cdot}697$; with $\hat{W} = 0$ and $Z = 0{\cdot}6$, the maximum value of B is 0·356, occurring at $\xi = 0{\cdot}421$. The reader is referred to the papers of Grant & Middleman (1966) and Sterling & Sleicher (1975) for a comparison of predicted jet lengths with experiment.

Recent research dealing with the breakup of jets of Newtonian fluids has focused on finite-amplitude effects in jets of low viscosity, and the conditions under which satellite drops are formed. Bogy (1979) has given a comprehensive review of this theory, and a very recent article by Chaudhary & Redekopp (1980) also deals with such theory. Chaudhary & Maxworthy (1980*a*, *b*) observed the pattern of jet breakup for disturbances of controlled amplitude and frequency, including harmonics, deliberately imposed on low-viscosity liquid jets. These studies were motivated by the application of ink-jet printing.

Breakage of non-Newtonian liquid filaments, especially viscoelastic liquid filaments, is at best poorly understood. It is important to distinguish between surface-tension-driven breakup, the subject of this paper, and another type of instability known as draw resonance. Draw resonance is the periodic variation in radius of a liquid filament undergoing elongation between an orifice where the thread is formed and a rotating take-up drum. The theory of this phenomenon is outlined in the book by Middleman (1977), and is surveyed in two comprehensive review articles on instabilities in non-Newtonian flows by Pearson (1976) and Petrie & Denn (1976). In this theory, one first derives expressions for the one-dimensional variation of jet radius and jet velocity as functions of the distance from the orifice, the velocity being specified at the orifice and take-up roll. The draw ratio is defined as the ratio of the velocity of the filament at take-up to the velocity of the filament at the orifice. Linearized stability analysis for the flow shows that below some critical draw ratio the steady one-dimensional flow is stable, but that above this critical value the flow is unstable. Surface effects are neglected in both the basic flow and the stability analysis. For Newtonian fluids the critical draw ratio is 20·2. For non-Newtonian fluids the critical draw ratio depends on the rheological equation of state chosen. The interested reader is referred to the two review articles cited above. Apart from the question of stability of these one-dimensional steady flows, there is the question of whether or not the rheological equation of state chosen even permits a one-dimensional steady-state solution at large elongational rates. This aspect of the spinnability of liquids is reviewed in the papers of Pearson (1976) and Petrie & Denn (1976), and an extensive discussion is given by Petrie (1979). Most rheological equations of state do not permit well-behaved one-dimensional steady solutions above some critical elongational rate. The critical elongational rate depends on the rheological equation of state chosen, and is probably related to the fact that the equation of state yields an expression for the elongational viscosity that becomes infinitely large at some finite elongational rate, yielding infinitely large tensile stresses. When the tensile stress produced by the rapid elongation of the filament exceeds the cohesive strength of the material, the filament will rupture. Cohesive fracture has been discussed by Ziabicki & Takserman-Krozer (1964*a*, *b*), and a survey appears in the book by Ziabicki (1976). Cohesive fracture may be coupled strongly with surface-tension-driven breakup because of spatial variation of thread diameter induced by capillarity. The coupling may be particularly important in view of the predictions of Tomotika (1936) and Mikami, Cox & Mason (1975) that threads of Newtonian liquid undergoing continuous extension are much more stable to surface-tension-driven breakup than are threads of constant diameter in plug flow.

We return to the discussion of surface-tension-driven breakage of viscoelastic liquid threads of nearly uniform diameter in nearly plug flow. It is necessary to adapt some rheological equation of state. For the present paper we have used the Oldroyd 8-constant model expressed in a corotational frame of reference; the 'constants' in this model are a zero shear viscosity η_0, a stress relaxation time λ_1, a strain-rate relaxation time λ_2, and five other time constants μ_0, μ_1, μ_2, ν_1 and ν_2 that may be needed to characterize the rheological responses of the liquid. For discussion of the necessity of objective reference frames and of the Oldroyd 8-constant model, the book by Bird, Armstrong & Hassager (1977, p. 336) should be consulted. In the notation of this reference the rheological equation of state used is

$$\boldsymbol{\tau}+\lambda_1\frac{\mathscr{D}\boldsymbol{\tau}}{\mathscr{D}t}+\tfrac{1}{2}\mu_0(\operatorname{tr}\boldsymbol{\tau})\dot{\boldsymbol{\gamma}}-\tfrac{1}{2}\mu_1\{\boldsymbol{\tau}\cdot\dot{\boldsymbol{\gamma}}+\dot{\boldsymbol{\gamma}}\cdot\boldsymbol{\tau}\}+\tfrac{1}{2}\nu_1(\boldsymbol{\tau}:\dot{\boldsymbol{\gamma}})\mathbf{I}$$
$$=-\eta_0\left[\dot{\boldsymbol{\gamma}}+\lambda_2\frac{\mathscr{D}\dot{\boldsymbol{\gamma}}}{\mathscr{D}t}-\mu_2\{\dot{\boldsymbol{\gamma}}\cdot\dot{\boldsymbol{\gamma}}\}+\tfrac{1}{2}\nu_2(\dot{\boldsymbol{\gamma}}:\dot{\boldsymbol{\gamma}})\mathbf{I}\right], \qquad (13)$$

where

$$\dot{\boldsymbol{\gamma}} = \nabla\mathbf{v} + (\nabla\mathbf{v})^{\mathrm{T}}, \tag{14}$$

$$\boldsymbol{\omega} = \nabla\mathbf{v} - (\nabla\mathbf{v})^{\mathrm{T}}, \tag{15}$$

$$\frac{\mathscr{D}\boldsymbol{\tau}}{\mathscr{D}t} = \frac{\partial\boldsymbol{\tau}}{\partial t} + (\mathbf{v}.\nabla)\,\boldsymbol{\tau} + \tfrac{1}{2}\{\boldsymbol{\omega}.\boldsymbol{\tau} - \boldsymbol{\tau}.\boldsymbol{\omega}\}, \tag{16}$$

$$\frac{\mathscr{D}\dot{\boldsymbol{\gamma}}}{\mathscr{D}t} = \frac{\partial\dot{\boldsymbol{\gamma}}}{\partial t} + (\mathbf{v}.\nabla)\,\dot{\boldsymbol{\gamma}} + \tfrac{1}{2}\{\boldsymbol{\omega}.\dot{\boldsymbol{\gamma}} - \dot{\boldsymbol{\gamma}}.\boldsymbol{\omega}\}. \tag{17}$$

Here $\mathbf{v}$ is the velocity vector, $\boldsymbol{\tau}$ is the stress tensor, $\dot{\boldsymbol{\gamma}}$ is the rate-of-strain tensor, $\boldsymbol{\omega}$ is the vorticity tensor, $\mathscr{D}/\mathscr{D}t$ is the Jaumann or corotational derivative, and $\mathbf{I}$ is the unit tensor.

Rheological responses such as the shear viscosity, the first normal-stress coefficient, and the second normal-stress coefficient as functions of rate of strain, the real and imaginary parts of the complex viscosity as functions of frequency, and the elongational viscosity as a function of the rate of elongation, all involving the eight constants of Oldroyd's model, are given explicitly by Bird *et al.* (1977). At present no rheological equation of state can describe accurately every non-Newtonian fluid under every flow situation. However, the Oldroyd 8-constant model is sufficiently flexible that it can be made to match semi-quantitatively rheological responses of many dilute polymer solutions by appropriate choices for the eight constants. In many applications the eight constants are reduced to a smaller number by presuming certain proportionalities among the seven time constants.

All previous linearized stability analyses of surface-tension-driven breakup (with the exception of those that postulate the formation of a gel) have presumed the jet to be in a completely relaxed state, i.e. plug flow and zero residual stresses other than a constant pressure. If small disturbances proportional to $\exp(\beta t + i2\pi z'/l)$ are superimposed on the completely relaxed jet, then to first order in small quantities (13) reduces to

$$\boldsymbol{\tau} = -\eta_0[(1+\beta\lambda_2)/(1+\beta\lambda_1)]\,\dot{\boldsymbol{\gamma}} \tag{18}$$

when expressed in a co-ordinate system moving with the jet. This equation is the same rheological equation of state as for a Newtonian fluid with 'effective' viscosity $\eta(\beta) = \eta_0(1+\beta\lambda_2)/(1+\beta\lambda_1)$. Consequently the theoretical result of Weber (1931) and Sterling & Sleicher (1975) given by (9)–(12) is immediately applicable provided we replace the Ohnesorge number Z by an 'effective' Ohnesorge number $Z(\beta)$:

$$\begin{aligned} Z(\beta) &= [\eta_0/(2\rho a\sigma)^{\frac{1}{2}}]\,(1+\beta\lambda_2)/(1+\beta\lambda_1) \\ &\equiv Z_0(1+\beta\lambda_2)/(1+\beta\lambda_1). \end{aligned} \tag{19}$$

The great majority of dilute polymer solutions are shear-thinning. For shear-thinning fluids $\lambda_2 < \lambda_1$. Under unstable conditions ($\beta > 0$), a shear-thinning fluid has a smaller effective viscosity, hence a smaller effective Ohnesorge number, than does the corresponding Newtonian fluid with constant viscosity equal to η_0. As discussed above, the smaller the Ohnesorge number, the less stable is the jet in the sense that as the Ohnesorge number decreases the dimensionless amplification factor B increases, implying shorter jet lengths to the formation of drops; also, the wavenumber for maximum growth rate shifts to larger values, implying the formation of smaller-diameter drops. This line of reasoning results in the conclusion that the elastic nature of dilute viscoelastic polymer solutions is destabilizing. Such was the theoretical conclusion of Middleman (1965), Paul (1968), Goldin *et al.* (1969), Rubin (1971), Sagiv, Rubin & Takserman-Krozer (1973), Gordon, Yerushalmi & Shinnar (1973) and Lee & Rubin (1975), all of whose chosen rheological equations of state reduced to (18) in their respective analyses.

The conclusion that the elastic nature of dilute viscoelastic polymer solutions is destabilizing is puzzling in view of the great stability observed experimentally for jets of many dilute polymer solutions (Goldin *et al.* 1969; Gordon *et al.* 1973). Indeed, such pronounced stability is essential for the successful operation of wet-spinning processes. Among the proposals offered to resolve this anomaly are the following. (i) There is a change of structure for the liquid associated with large deformation at the nozzle; Goldin, Pfeffer & Shinar (1972) carried out a linear stability analysis for a liquid having a finite yield stress. (ii) The higher amplification rates for truly infinitesimal disturbances are accepted; but it is suggested that even for small disturbances nonlinear terms in the rheological equation of state are important; if a finite-amplitude stability analysis were carried out, such an analysis might show smaller amplification rates (Goldin *et al.* 1969). (iii) Initially higher amplification rates are accepted, but at a later stage thin filaments connect larger bulges along the jet length; the drainage of these filaments into the bulges is an elongational flow which is resisted by a large elongational viscosity (Gordon *et al.* 1973). These proposals may explain the observed enhanced stability of some polymer solutions under some flow conditions. However, some observations for jets of dilute polymer solutions are not explained. In particular, jets of shear-thinning liquids can travel significant distances with very little amplification of disturbances, and then over a relatively short distance the amplitude increases almost explosively. The resulting bulges are often much more irregularly spaced, with larger average spacing than is found for Newtonian fluids. This results in larger drops, but with a wider size distribution, in addition to long breakup lengths.

The crucial idea in the theory to be presented is the recognition that the liquid may be subject to an unrelaxed axial tension. Axial elastic tensions in viscoelastic liquid jets do arise from the large deformation rates within the capillary tube or at the nozzle. Although the elastic tension rapidly decreases within a few jet diameters from the orifice, nevertheless smaller but significant elastic tensions are known to persist for long distances along the jet for some dilute polymer solutions (Gill & Gavis 1956; Gavis & Gill 1956; Gavis & Middleman 1963). Similarly, small-diameter liquid filaments formed from accidentally released fuels containing dissolved antimisting polymer additives will develop axial elastic tensions as a result of the rapid elongation of these threads from larger volumes of liquid. The possibility that an elastic tension could account for the observed great stability was first suggested by Debye & Daen (1956). However, this idea does not seem to have been developed quantitatively in a satisfactory way. As mentioned earlier, all previous theoretical analyses, with the exception of those that postulate the formation of a gel, have presumed the jet to be in a completely relaxed state, i.e. plug flow and *zero* stresses. These previous analyses are not consistent with the observed great stability of many shear-thinning viscoelastic liquids because the analyses actually predict that such jets are less stable than comparable jets of Newtonian liquids. The present analysis also predicts that jets of shear-thinning liquids are less stable than comparable jets of Newtonian liquids when there is no axial elastic tension. However, when the elastic tension T is not zero, and provided the stress relaxation time λ_1 is sufficiently large, the new theory predicts that the elastic tension can be a significant stabilizing influence. We introduce the following two dimensionless groups:

$$Te = -Ta/\sigma, \tag{20}$$

$$D = \lambda_1(\sigma/8\rho a^3)^{\frac{1}{2}}. \tag{21}$$

The dimensionless elastic-tension group includes a minus sign because according to the convention of Bird *et al.* (1977) tensile forces are regarded as negative and compressive forces as positive. D is the Deborah number used to characterize the stress relaxation time. Other factors held constant, the theory predicts that, the larger Te is, then the greater is the stabilizing effect of the tension; the larger D is above some critical value of the order of unity, then the greater is the stabilizing influence of the tension.

2. Theory

Consider an unsteady but axisymmetric jet of viscoelastic liquid moving through an inviscid gas. The local radius of the jet is denoted by $R(z,t)$. In cylindrical coordinates (r,θ,z), the velocity and pressure in the liquid are $(v_r, 0, v_z)$ and p; the corresponding quantities in the gas are $(\hat{v}_r, 0, \hat{v}_z)$ and $\hat{p}$. In the notation of Bird *et al.* (1977, appendix B) the equations of continuity and motion for an incompressible liquid of arbitrary rheology are

$$\frac{\partial v_r}{\partial r}+\frac{v_r}{r}+\frac{\partial v_z}{\partial z} = 0, \tag{22}$$

$$\rho\left\{\frac{\partial v_r}{\partial t}+v_r\frac{\partial v_r}{\partial r}+v_z\frac{\partial v_r}{\partial z}\right\} = -\frac{\partial p}{\partial r}-\left\{\frac{1}{r}\frac{\partial(r\tau_{rr})}{\partial r}+\frac{\partial \tau_{rz}}{\partial z}-\frac{\tau_{\theta\theta}}{r}\right\}, \tag{23}$$

$$\rho\left\{\frac{\partial v_z}{\partial t}+v_r\frac{\partial v_z}{\partial r}+v_z\frac{\partial v_z}{\partial z}\right\} = -\frac{\partial p}{\partial z}-\left\{\frac{1}{r}\frac{\partial(r\tau_{rz})}{\partial r}+\frac{\partial \tau_{zz}}{\partial z}\right\}. \tag{24}$$

The rheological equation of state relating the stress tensor $\boldsymbol{\tau}$ to the velocity field adopted for this study is the Oldroyd 8-constant model expressed in a corotating reference frame; this relationship is given in (13). The gas satisfies a similar trio of equations but, because we assume the gas to be inviscid $\hat{\boldsymbol{\tau}} = 0$,

$$\frac{\partial \hat{v}_r}{\partial r}+\frac{\hat{v}_r}{r}+\frac{\partial \hat{v}_z}{\partial z} = 0, \tag{25}$$

$$\hat{\rho}\,\frac{\partial \hat{v}_r}{\partial t}+\hat{v}_r\frac{\partial \hat{v}_r}{\partial r}+\hat{v}_z\frac{\partial \hat{v}_r}{\partial z} = -\frac{\partial \hat{p}}{\partial r}, \tag{26}$$

$$\hat{\rho}\,\frac{\partial \hat{v}_z}{\partial t}+\hat{v}_r\frac{\partial \hat{v}_z}{\partial r}+\hat{v}_z\frac{\partial \hat{v}_z}{\partial z} = -\frac{\partial \hat{p}}{\partial z}. \tag{27}$$

The liquid and gas velocity fields are coupled by four boundary conditions to be applied at the jet surface $r = R(z,t)$:

$$\hat{v}_r = \partial R/\partial t+\hat{v}_z\partial R/\partial z, \tag{28}$$

$$v_r = \partial R/\partial t+v_z\partial R/\partial z, \tag{29}$$

$$\tau_{rz} + (\tau_{zz} - \tau_{rr})(\partial R/\partial z) - \tau_{rz}(\partial R/\partial z)^2 = 0, \tag{30}$$

$$p + \tau_{rr} - \frac{2\tau_{rz}(\partial R/\partial z)}{1 + (\partial R/\partial z)^2} + \frac{(\tau_{zz} - \tau_{rr})(\partial R/\partial z)^2}{1 + (\partial R/\partial z)^2} = \hat{p} + \sigma\left[\frac{1}{R[1 + (\partial R/\partial z)^2]^{\frac{1}{2}}} - \frac{\partial^2 R/\partial z^2}{[1 + (\partial R/\partial z)^2]^{\frac{3}{2}}}\right]. \tag{31}$$

Equations (28) and (29) express the kinematic condition at the jet surface for the gas and liquid respectively. Equation (30) expresses the vanishing of the tangential forces at the jet surface for an inviscid gas. Equation (31) expresses the balancing of the difference of normal forces at the jet surface by the surface tension and curvature of the surface.

At large radial distances from the jet, the gas moves in plug flow with velocity $\hat{U}$. Thus, for $r \to \infty$

$$\hat{v}_r = 0, \quad \hat{v}_z = \hat{U}. \tag{32}$$

Other boundary conditions are provided by the requirement that the liquid velocity and stress be finite along the jet axis $r = 0$.

It is beyond the scope of the present paper to compute the unperturbed velocity profile and jet radius as functions of axial distance for a viscoelastic liquid jet issuing from an orifice. Our interest is in the region beyond several jet diameters from the orifice where, in the absence of external forces or a take-up drum, the jet approaches a constant radius a with plug-flow velocity U. However, the following analysis shows that in this region, provided $W = 2\rho a U^2/\sigma$ is sufficiently large, variations in jet radius and velocity with axial distance are small even though the axial tension has not completely decayed to zero. Following Bird *et al.* (1977, p. 50), the equations of conservation of mass and momentum may be integrated over a control volume from z to ∞ enclosing the jet. With the neglect of air inertia and air drag, we find

$$\pi R^2 \langle v_z \rangle = \pi a^2 U,$$

$$\pi R^2 \rho \langle v_z^2 \rangle + \pi R^2 \langle p + \tau_{zz} \rangle - 2\pi R\sigma/(1 + R_z^2)^{\frac{1}{2}} - \pi(a^2 - R^2)\hat{p} = \pi a^2 \rho U^2 + \pi a^2 p_\infty - 2\pi a\sigma.$$

Combining (30) and (31) shows that the pressure in the liquid just below the gas–liquid interface is given by

$$p = \hat{p} + \sigma\left\{\frac{1}{R[1 + R_z^2]^{\frac{1}{2}}} - \frac{R_{zz}}{[1 + R_z^2]^{\frac{3}{2}}} - \tau_{rr} - \frac{(\tau_{zz} - \tau_{rr})R_z^2(3 - R_z^2)}{1 - R_z^4}\right\}.$$

In the region of interest $R_z \ll 1$ for the unperturbed flow, and the axial pressure, stresses and velocities can be treated as being constant across the jet's cross-section. Combining the above three equations gives the following dimensionless relationship between the jet radius R/a at position z where the tension is Te, and at infinity where $R/a = 1$ and the tension has decayed to zero:

$$1 - a^2/R^2 + 2(R/a - 1 + Te\, R^2/a^2)/W = 0.$$

Table 1 shows R/a as a function of W for selected values of Te. For example with $Te = 2$ the change in jet radius as the tension relaxes to zero is less than 2 % provided $W \geqslant 100$; the corresponding change in jet velocity is less than 4 %. Larger values of W or smaller values of Te give smaller changes in the unperturbed jet radius and velocity. Since most experimental conditions (beyond a few jet radii from the orifice)

W \ Te	0·1	0·2	0·5	1	2	5	10
10^3	0·9999	0·9998	0·9995	0·9990	0·9980	0·9951	0·9903
10^2	0·9990	0·9980	0·9951	0·9904	0·9814	0·9575	0·9247
10	0·9912	0·9828	0·9603	0·9291	0·8814	0·7935	0·7139
1	0·9559	0·9205	0·8455	0·7659	0·6766	0·5573	0·4742

TABLE 1. Variation of jet radius R/a

correspond to $W > 100$ and $Te < 2$, the results of the stability theory to be presented seem to be generally applicable.

We assume that the filament has 'nearly constant' radius a, moves in 'nearly constant' plug flow with axial velocity U, and is under a 'nearly constant' axial tension T. This state is disturbed by small, axially symmetric perturbations of the form $\epsilon \exp(\beta t + i\pi z/l)$, so that

$$R(z,t) = a + \epsilon \exp(\beta t + i2\pi z/l). \tag{33}$$

Here t is time, z is axial distance measured in a fixed co-ordinate system, l is the wavelength of the disturbance, ϵ is the initial amplitude of the disturbance, and β is the amplification factor for a disturbance of wavelength l. For 'nearly constant' jet velocity and radius, (13) requires that the axial tension decay exponentially with a time constant λ_1:

$$T(z) = T_0 \exp(-z/\lambda_1 U). \tag{34}$$

This represents a stress relaxation at *constant* extension. The results are therefore limited to a distance downstream from the orifice sufficient for the jet radius and velocity to approach their asymptotic values but where the axial tension has not yet decayed to zero (see the discussion above).

The term 'nearly constant' as used above is taken to mean that fractional changes in a, U and T are small over distances comparable to the wavelength, i.e.

$$l/\lambda_1 U \ll 1. \tag{35}$$

In terms of the dimensionless groups defined in § 1, this inequality may be rewritten as

$$\xi D W^{\frac{1}{2}} \gg \pi. \tag{36}$$

To carry out a linearized stability analysis for the basic flow, we superimpose small disturbances on the velocities, pressures, and stress components of the same form as the disturbance superimposed on the jet radius:

$$\mathbf{v} = (0,0,U) + (v_r', 0, v_z')\,\epsilon \exp(\beta t + ikz), \tag{37}$$

$$p = P + p'\epsilon \exp(\beta t + ikz), \tag{38}$$

$$\boldsymbol{\tau} = T\boldsymbol{\delta}_z\boldsymbol{\delta}_z + \boldsymbol{\tau}'\epsilon \exp(\beta t + ikz), \tag{39}$$

$$\hat{\mathbf{v}} = (0,0,\hat{U}) + (\hat{v}_r', 0, \hat{v}_z')\,\epsilon \exp(\beta t + ikz), \tag{40}$$

$$\hat{p} = \hat{P} + \hat{p}'\epsilon \exp(\beta t + ikz). \tag{41}$$

Primed quantities are presumed to be independent of t and z and to depend only on radial position r. Also

$$k = 2\pi/l. \tag{42}$$

When (40) and (41) are substituted into (25)–(28) and (32), and only first-order quantities in ϵ retained, the following set of equations governing the gas are obtained:

$$d\hat{v}'_r/dr+\hat{v}'_r/r+ik\hat{v}'_z = 0, \tag{43}$$

$$\hat{\rho}(\beta+ik\hat{U})\,\hat{v}'_r = -d\hat{p}'/dr, \tag{44}$$

$$\hat{\rho}(\beta+ik\hat{U})\,\hat{v}'_z = -ik\hat{p}', \tag{45}$$

$$\hat{v}'_r(a) = (\beta+ik\hat{U}), \tag{46}$$

$$\hat{v}'_r(\infty) = 0, \quad \hat{v}'_z(\infty) = 0, \quad \hat{p}'(\infty) = 0. \tag{47}$$

The solution to this set of linear equations for the gas pressure is easily shown to be

$$\hat{p}'(r) = \hat{\rho}a(\beta+ik\hat{U})^2\,K_0(kr)/kaK_1(ka). \tag{48}$$

To simplify the analysis for the liquid flow, it is convenient to introduce a stream function $\psi'(r)\exp(\beta t+ikz)$ for the disturbances in the liquid velocities:

$$v'_r = \frac{ik\psi'}{r}, \quad v'_z = -\frac{1}{r}\frac{d\psi'}{dr}. \tag{49}$$

Substituting (37)–(39) and (49) into (23) and (24), retaining only first-order quantities in ϵ, and then eliminating the pressure between the two resulting equations yields the following differential equation for ψ':

$$\rho(\beta+ikU)\,F^2\psi' = G^2(r\tau'_{rz})+ikr\,d(\tau'_{zz}-\tau'_{rr})/dr-ik(\tau'_{rr}-\tau'_{\theta\theta})/r, \tag{50}$$

where

$$F^2 \equiv \frac{d^2}{dr^2}-\frac{1}{r}\frac{d}{dr}-k^2, \tag{51}$$

$$G^2 \equiv \frac{d^2}{dr^2}-\frac{1}{r}\frac{d}{dr}+k^2. \tag{52}$$

Substituting (37)–(39) and (49) into the three boundary conditions expressed in (29)–(31) and retaining only first-order quantities in ϵ gives

$$\psi' = a(\beta+ikU)/ik \quad \text{on} \quad r = a, \tag{53}$$

$$\tau'_{rz} = -ikT \quad \text{on} \quad r = a, \tag{54}$$

$$\begin{aligned}&\rho(\beta+ikU)\,(d\psi'/dr)-d(r\tau'_{rz})/dr-ika(\tau'_{zz}-\tau'_{rr})\\ &\quad = -(ik\sigma/a)\,(1-k^2a^2)+ik\hat{\rho}a^2(\beta+ik\hat{U})^2\,K_1(ka)/kaK_0(ka) \quad \text{on} \quad r = a.\end{aligned} \tag{55}$$

To obtain this last equation from (31), the liquid pressure is first found from the z-momentum equation, and the gas pressure in (48) is used.

It is necessary to relate the perturbation in the stress tensor to the velocity perturbations, and thereby to the stream function for the disturbed flow. Because the basic liquid flow is plug flow, the undisturbed rate-of-strain tensor and the undisturbed vorticity tensor are both zero. Perturbations in the rate-of-strain tensor $\dot{\boldsymbol{\gamma}}'\epsilon\exp(\beta t+ikz)$ and vorticity tensor $\boldsymbol{\omega}'\epsilon\exp(\beta t+ikz)$ are first found as follows:

$$\dot{\gamma}'_{rr} = 2ik\,d(\psi'/r)/dr, \tag{56}$$

$$\dot{\gamma}'_{\theta\theta} = 2ik\psi'/r^2, \tag{57}$$

$$\dot{\gamma}'_{zz} = 2k^2\psi'/r, \tag{58}$$

$$\dot{\gamma}'_{rz} = \dot{\gamma}'_{rz} = -(G^2\psi')/r, \tag{59}$$

$$\omega'_{rz} = -\omega'_{zr} = -(F^2\psi')/r. \tag{60}$$

Components of $\dot{\boldsymbol{\gamma}}'$ and $\boldsymbol{\omega}'$ not shown are zero. The linearization of Oldroyd's 8-constant model (13) results in the expression

$$\begin{aligned}[1+(\beta+ikU)\lambda_1]\boldsymbol{\tau}' = & -\eta_0[1+(\beta+ikU)\lambda_2]\dot{\boldsymbol{\gamma}}' \\ & +\tfrac{1}{2}\mu_0 T\dot{\boldsymbol{\gamma}}'+\tfrac{1}{2}\lambda_1 T\omega'_{rz}\{\boldsymbol{\delta}_r\boldsymbol{\delta}_z+\boldsymbol{\delta}_z\boldsymbol{\delta}_r\} \\ & -\tfrac{1}{2}\mu_1 T\{\dot{\gamma}'_{rz}\boldsymbol{\delta}_r\boldsymbol{\delta}_z+\dot{\gamma}'_{zr}\boldsymbol{\delta}_z\boldsymbol{\delta}_r+2\dot{\gamma}'_{zz}\boldsymbol{\delta}_z\boldsymbol{\delta}_z\}+\tfrac{1}{2}\nu_1 T\dot{\gamma}'_{zz}\mathbf{I}. \end{aligned} \tag{61}$$

In obtaining this result, the term $v'_z\partial T/\partial z$ is neglected, because if the other terms are of order unity then this term is of order $l/\lambda_1 U$. Our assumption that the tension be 'nearly constant' required $l/\lambda_1 U \ll 1$. Inspection of (61) makes it clear that if T is *not* equal to zero then the rheological equation of state does *not* reduce to a form equivalent to that for a Newtonian fluid.

The specific components of $\boldsymbol{\tau}'$ required in (50), (54) and (55) are written explicitly as

$$[1+(\beta+ikU)\lambda_1][r\tau'_{rz}] = \{\eta_0[1+(\beta+ikU)\lambda_2]+\tfrac{1}{2}\mu_1 T-\tfrac{1}{2}\mu_0 T\}G^2\psi'-\tfrac{1}{2}\lambda_1 TF^2\psi', \tag{62}$$

$$\begin{aligned}[1+(\beta+ikU)\lambda_1][\tau'_{zz}-\tau'_{rr}] = & -\left\{\eta_0[1+(\beta+ikU)\lambda_2]-\tfrac{1}{2}\mu_0 T\right\} \\ & \times\left\{\frac{2k^2\psi'}{r}-2ik\frac{d(\psi'/r)}{dr}\right\}-\frac{2\mu_1 Tk^2\psi'}{r}, \end{aligned} \tag{63}$$

$$[1+(\beta+ikU)\lambda_1][\tau'_{rr}-\tau'_{\theta\theta}] = -\left\{\eta_0[1+(\beta+ikU)\lambda_2]-\tfrac{1}{2}\mu_0 T\right\}\left\{2ik\frac{d(\psi'/r)}{dr}-\frac{2ik\psi'}{r^2}\right\}. \tag{64}$$

Substitution of these three expressions into (50) gives the following remarkably simple differential equation for ψ':

$$F^2\{F^2-(k_1^2-k^2)\}\psi' = 0, \tag{65}$$

where

$$k_1^2 = k^2+\frac{\rho(\beta+ikU)[1+(\beta+ikU)\lambda_1]+\lambda_1 Tk^2}{\eta_0[1+(\beta+ikU)\lambda_2]+\tfrac{1}{2}(\mu_1-\lambda_1-\mu_0)T}. \tag{66}$$

Except for the different definition of k_1, this differential equation is identical with that for the breakup of a viscous Newtonian liquid. The solution that remains well-behaved on $r = 0$ is

$$\psi'(r) = c_1 rI_1(kr)+c_2 rI_1(k_1 r), \tag{67}$$

where c_1 and c_2 are constants. Substituting (62) and (67) into (53) and (54) gives a pair of equations by which c_1 and c_2 are determined. Having done this, we substitute (62), (63) and (67) into the third boundary condition at the jet surface, namely (55). This procedure then results in an equation relating β to l and the other variables. After a great deal of manipulation to reduce the relationship to as convenient a form as seems possible, we find

$$\begin{aligned}(\beta+ikU)^2\mathscr{I}(ka)+\frac{(\beta+ikU)\eta_0 k^2[4\mathscr{I}(ka)-1+Q(ka,k_1a)][1+(\beta+ikU)\lambda_2]}{\rho[1+(\beta+ikU)\lambda_1]} \\ = \frac{\sigma k^2}{2\rho a}(1-k^2a^2)-\frac{\hat{\rho}}{\rho}(\beta+ik\hat{U})^2 ka\frac{K_0(ka)}{2K_1(ka)} \\ -\frac{Tk^2}{2\rho}\{(\beta+ikU)[4(\mu_1-\mu_0-\tfrac{1}{2}\lambda_1)\mathscr{I}(ka)+(\mu_1-\mu_0+\lambda_1)Q(ka,k_1a)+\mu_0] \\ -[2\mathscr{I}(ka)+Q(ka,k_1a)]\}/[1+(\beta+ikU)\lambda_1]. \end{aligned} \tag{68}$$

The functions $\mathscr{I}$ and Q are those defined in (11) and (12). Equation (68) can be used to describe spatial growth of disturbances of frequency ω if we set $\beta = i\omega$ and solve for k as a complex function of ω. Here $\beta l/U$ need not be small. When $\beta l/U \ll 1$, spatial growth is equivalent to temporal growth in a co-ordinate system moving with the jet velocity; in such a co-ordinate system the jet velocity appears to be zero and the gas velocity appears to be $\hat{U} - U$. Furthermore, because $\hat{\rho} \ll \rho$ and $\beta \ll lU$, we can neglect the terms involving the gas density except for that multiplied by $(\hat{U} - U)^2$. When we make these substitutions and approximations in (68) and express the result in the dimensionless terms defined in §1, we obtain the following equation for the dimensionless temporal amplification factor:

$$
\begin{aligned}
&B^2\mathscr{I}(\xi) + 4BZ_0\xi^2[4\mathscr{I}(\xi) - 1 + Q(\xi,\xi_1)]\,[1 + (\lambda_2/\lambda_1)\,DB]/[1+DB] \\
&\quad = 4\xi^2[1 - \xi^2 + \hat{W}\xi K_0(\xi)/2K_1(\xi)] \\
&\qquad - 4Te\xi^2\left\{DB\left(4\left[\frac{\mu_1-\mu_0}{\lambda_1} - \frac{1}{2}\right]\mathscr{I}(\xi) + \left[\frac{\mu_1-\mu_0}{\lambda_1} - 1\right]Q(\xi,\xi_1) + \mu_0/\lambda_1\right)\right. \\
&\qquad \left. - [2\mathscr{I}(\xi) + Q(\xi,\xi_1)]\right\}\Big/[1+DB],
\end{aligned}
\tag{69}
$$

where

$$
\xi_1^2 = \xi^2 + \frac{B[1+DB] + 8DTe\xi^2}{4Z_0[1 + (\lambda_2/\lambda_1)\,DB] + 4DTe[(\mu_1-\mu_0)/\lambda_1 - 1]}. \tag{70}
$$

If Te equals zero, then (69) and (70) reduce to the result given by (9) for viscous Newtonian liquid except that the Ohnesorge number Z must be replaced by an effective Ohnesorge number given by $Z_0[1 + (\lambda_2/\lambda_1)\,DB]/[1+DB]$ which is the same as the dimensional form given in (19). For shear-thinning fluids $\lambda_2 < \lambda_1$; if $Te = 0$, shear-thinning fluids will be more unstable than Newtonian fluids of constant viscosity equal to η_0. When $Te > 0$, numerical solutions to (69) show that the axial tension can be a significant stabilizing influence provided D is sufficiently large. Numerical results are given in §3.

3. Results

The relationship between β and the other variables given in (68) does not involve three (ν_1, ν_2 and μ_2) of the seven time constants in Oldroyd's 8-constant model. Furthermore, if we restrict ourselves to sufficiently small growth rates ($\beta l/U \ll 1$), then the disturbances can be treated as growing in time in a co-ordinate system moving with the jet. In this approximation, the dimensionless amplification factor B does not depend on the Weber number W for the liquid. Equation (69) can be written symbolically as

$$
B = B(\xi, Z_0, \hat{W}, Te, D, \lambda_2/\lambda_1, \mu_1/\lambda_1, \mu_0/\lambda_1). \tag{71}
$$

If $Te = 0$, B also becomes independent of μ_1/λ_1 and μ_0/λ_1, and (71) reduces to the result for a viscous Newtonian liquid except that Z is replaced by

$$
Z_0[1 + (\lambda_2/\lambda_1)\,DB]/[1+DB].
$$

Jets of shear-thinning liquids with $Te = 0$ are less stable than corresponding Newtonian jets.

In view of the many variables on which B depends, it is desirable to adopt some restrictions. Most dilute polymer solutions show very small second normal-stress coefficients. Oldroyd's 8-constant model requires $\mu_1/\lambda_1 = 1$ for fluids with zero second

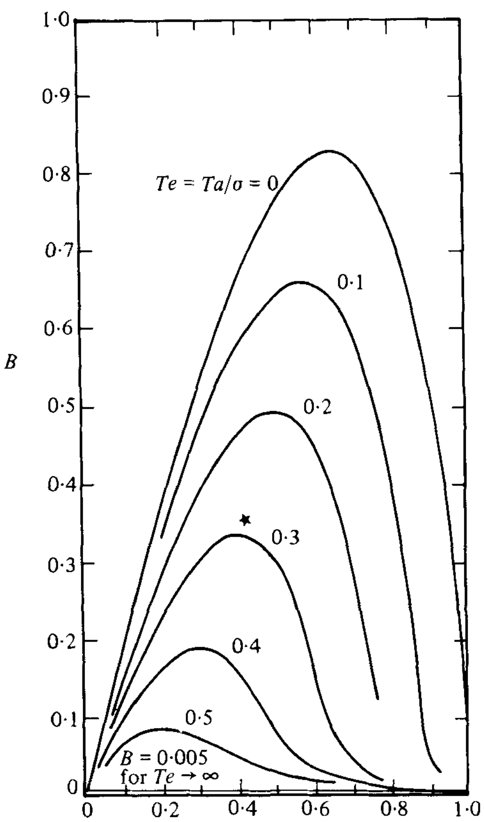

FIGURE 1. Amplification factor $B = \beta(8\rho a^3/\sigma)^{\frac{1}{2}}$ as a function of wavenumber $\xi = 2\pi a/l$ for a viscoelastic liquid jet sustaining various elastic tensions. The star marks the wavenumber and amplification factor for a Newtonian fluid whose Ohnesorge number Z is 0·6. $Z_0 = 0{\cdot}6$, $D = 200$, $\lambda_2/\lambda_1 = 0{\cdot}1$, $\mu_1/\lambda_1 = 1$, $\mu_0/\lambda_1 = 0$, $\hat{W} = 0$.

normal-stress coefficient. In all of the calculations reported here μ_1/λ_1 has been set equal to unity.

Most, but not all, flow calculations based on Oldroyd's 8-constant model take $\mu_0 = 0$. Experimental justification for this is not clear to the present authors. However, $\mu_0 = 0$ is inherent in the following simplifications of Oldroyd's 8-constant model: Oldroyd's fluid B, Williams' 3-constant Oldroyd model, corotational Jeffreys model, Denn's modified convected model, second-order fluid, and the convected Maxwell model (Bird *et al.* 1977). Most of the calculations reported here also set $\mu_0 = 0$.

In order to simplify the calculations and their interpretation further, we have set $\hat{W} = 0$ for all calculations reported here. The presumption is that we can make the density of the surrounding gas sufficiently small that, even for large relative velocities between the gas and jet, $\hat{W} \ll 1$. Depending on the other variables it may be necessary to require $W \gg 1$ in order to treat the disturbances as growing in time, but this inequality is not inconsistent with $\hat{W} \ll 1$ when $\hat{\rho} \ll \rho$.

Figure 1 is a plot of the dimensionless amplification factor B as a function of the wavenumber ξ for several dimensionless elastic tensions Te. The parameters held

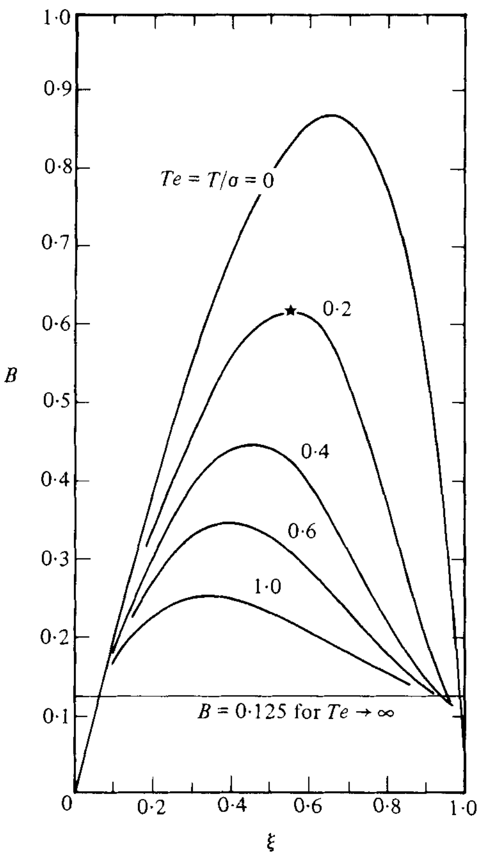

FIGURE 2. Amplification factor B as a function of wavenumber ξ for a viscoelastic liquid jet sustaining various elastic tensions. $Z_0 = 0{\cdot}2$, $D = 8$, $\lambda_2/\lambda_1 = 0{\cdot}1$, $\mu_1/\lambda_1 = 1$, $\mu_0/\lambda_1 = 0$, $\hat{W} = 0$.

constant for this plot are $Z_0 = 0{\cdot}6$, $D = 200$, $\lambda_2/\lambda_1 = 0{\cdot}1$, $\mu_1/\lambda_1 = 1$, $\mu_0/\lambda_1 = 0$ and $\hat{W} = 0$. The values selected for the first three groups are order-of-magnitude estimates for a 100 μm diameter thread formed from a 0·74 % solution of 'AM-1' in kerosene; 'AM-1' is a commercial antimisting additive (manufactured by CONOCO) investigated by Peng & Landel (1980). As a point of reference, the star marks the wavenumber and amplification factor for maximum amplification rate for a Newtonian fluid whose Ohnesorge number Z is 0·6; for this fluid the respective quantities are $B = 0{\cdot}356$ at $\xi = 0{\cdot}421$. For the viscoelastic liquid at zero elastic tension the maximum amplification factor is 0·825 at a wavenumber of 0·650. We see clearly that this shear-thinning liquid is less stable than the corresponding Newtonian liquid at $Te = 0$. As Te increases, B decreases, indicating longer distances to the point of drop formation. As Te increases, the wavenumber for maximum amplification shifts to smaller values, indicating longer wavelengths and therefore larger droplets upon breakage. For the parameters selected, at $\xi \simeq 0{\cdot}4$, it is necessary for Te to exceed about 0·3 before the stabilizing influence of the tension counterbalances the destabilizing effect associated with shear thinning. Further increases in Te result in greater stabilization.

Although it is not clear from figure 1, as Te increases, some disturbances with wavenumbers greater than unity also become unstable, and the maximum in ampli-

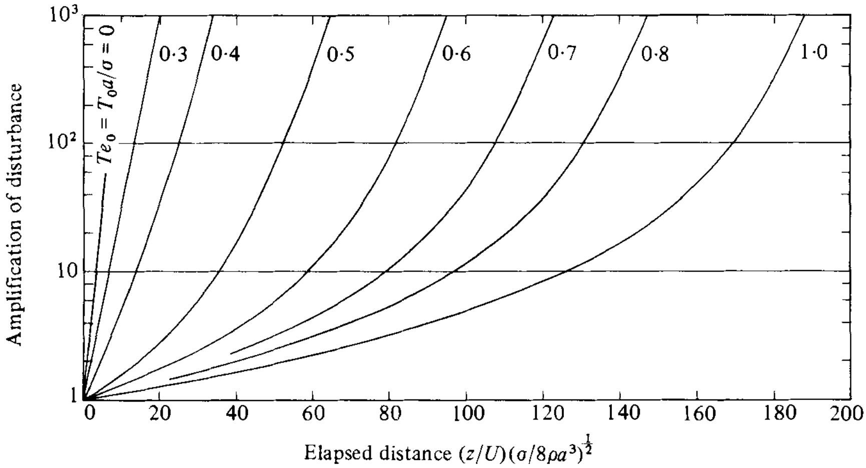

FIGURE 3. Amplification of disturbances as a function of elapsed dimensionless distance (or time) for viscoelastic liquid jets of various initial elastic tensions. The tension decays exponentially with a time constant λ_1 equal to the stress relaxation time. $Z_0 = 0{\cdot}6$, $D = 200$, $\lambda_2/\lambda_1 = 0{\cdot}1$, $\mu_1/\lambda_1 = 1$, $\mu_0/\lambda_1 = 0$, $\hat{W} = 0$, $\xi = 0{\cdot}421$.

fication rate becomes less pronounced. These trends are more clearly evident at smaller Deborah numbers. For example, for figure 2, $Z_0 = 0{\cdot}2$, $D = 8$, $\lambda_2/\lambda_1 = 0{\cdot}1$, $\mu_1/\lambda_1 = 1$, $\mu_0/\lambda_1 = 0$ and $\hat{W} = 0$; these values are order-of-magnitude estimates for a 1 mm diameter thread formed from a 0·74 % solution of 'AM-1' in kerosene. Again, as a point of reference the star marks the wavenumber ($\xi = 0{\cdot}55$) for the maximum amplification rate ($B = 0{\cdot}62$) for a Newtonian fluid of the same Ohnesorge number. When the maximum in the amplification factor becomes less pronounced, it is likely that the distribution in the resulting drop sizes will broaden because there is no disturbance that amplifies much more rapidly than the many other unstable waves. For $\mu_1/\lambda_1 = 1$ and $\mu_0/\lambda_1 = 0$, (69) requires that, as $Te \to \infty$, $B \to 1/D$ or $\beta \to 1/\lambda_1$. Thus at very high tensions the fluid can respond no faster than the stress relaxation.

The stress relaxation time constant λ_1 is important in another way. For a jet with sufficiently large Te the stabilizing influence of the tension exceeds the destabilizing effect of shear thinning, and the amplification rate is small. However, as the jet moves away from the orifice the elastic tension decays. The rate of amplification will increase with distance from the orifice. When the tension decays sufficiently, the destabilizing influence of shear thinning exceeds the stabilizing influence of the tension; here the rate of amplification is large. Liquids with large stress relaxation times, i.e. large D, will travel long distances from the orifice with little relaxation of the tension and consequently smaller amplification of disturbances than will comparable jets with smaller D. These trends are illustrated in figure 3. Here we plot the cumulative amplification of a disturbance as a function of a dimensionless distance (L/U) $(\sigma/8\rho a^3)^{\frac{1}{2}}$ from the orifice. The parameter is a dimensionless tension number $Te_0 = -T_0 a/\sigma$ based on the tension T_0 at the orifice. The parameters held constant for this figure are $\xi = 0{\cdot}421$, $Z_0 = 0{\cdot}6$, $D = 200$, $\lambda_2/\lambda_1 = 0{\cdot}1$, $\mu_1/\lambda_1 = 1$, $\mu_0/\lambda_1 = 0$ and $\hat{W} = 0$. To obtain these curves we have numerically solved the equation $U\,d\epsilon/dz = \beta(z)\,\epsilon$, where $\beta(z)$ is the local amplification, which depends on the local tension $T(z)$ as given by our theory, and $T(z) = T_0 \exp(-z/\lambda_1 U)$.

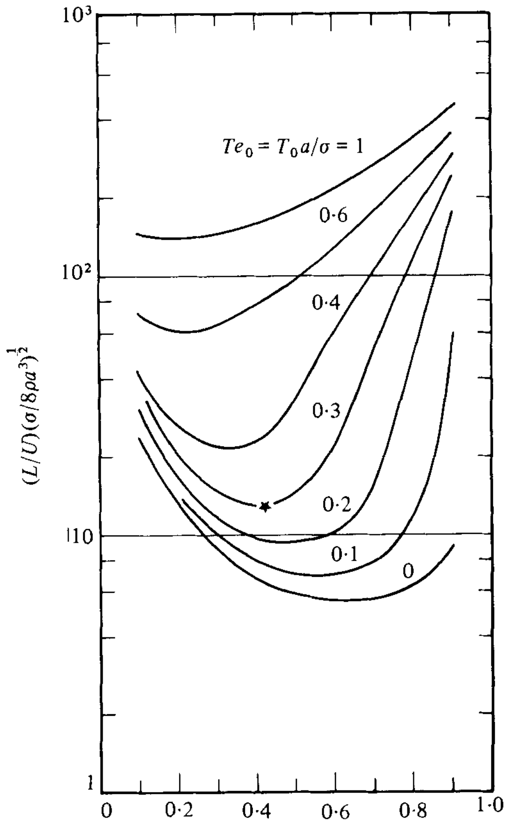

FIGURE 4. Dimensionless length $(L/U)(\sigma/8\rho a^3)^{\frac{1}{2}}$ required for disturbances to amplify 100-fold as a function of wavenumber for various initial elastic tensions. The tension decays exponentially with a time constant λ_1 equal to the stress relaxation time. The star marks the minimum length for 100-fold amplification of a Newtonian liquid whose Ohnesorge number is 0·6. $Z_0 = 0{\cdot}6$, $D = 200$, $\lambda_2/\lambda_1 = 0{\cdot}1$, $\mu_1/\lambda_1 = 1$, $\mu_0/\lambda_1 = 0$, $\hat{W} = 0$.

In carrying out the integrations to produce figure 3 and subsequent figures, spatial variations in $T(z)$ only and not in a and U have been considered. As discussed above, this is justified when W is greater than about 100. Also, to apply the results of a stability calculation for a locally 'frozen' flow to one with a spatially varying axial tension, it is necessary that $l/\lambda_1 U = 1/\xi DW$ be much smaller than unity. The value depends on the sensitivity of β to T. By comparing $\beta[T(z+l)]$ with $\beta[T(z)]$ for small $l/\lambda_1 U$, we arrive at the criterion

$$D \gg -\frac{Te}{W\xi B}\frac{dB}{dTe}.$$

For example from figure 2 at $Te = 0{\cdot}4$ and $\xi = 0{\cdot}5$ we estimate

$$B = 0{\cdot}45 \quad \text{and} \quad dB/dTe = (0{\cdot}60-0{\cdot}33)/(0{\cdot}2-0{\cdot}6) = -0{\cdot}68;$$

consequently we require $D \gg 1{\cdot}2/W$. Since W is expected to be of the order of 100, the choice of $D = 8$ for this calculation certainly seems adequately large. The longer the elapsed time or distance, the smaller are Te and dB/dTe and the larger is B, so that the approximation of a 'frozen' flow becomes even better far from the orifice.

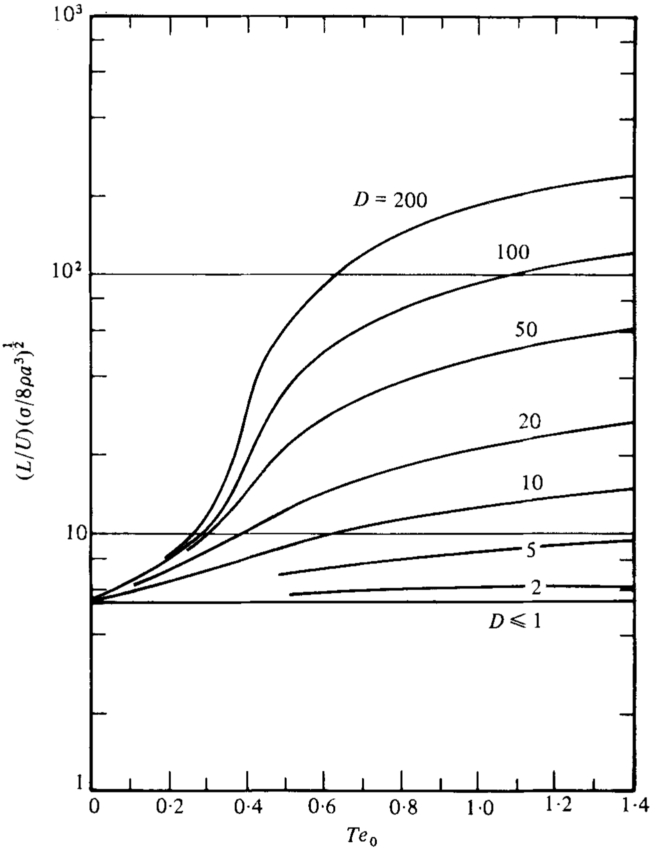

FIGURE 5. Parametric study of the dimensionless length $(L/U)(\sigma/8\rho a^3)^{\frac{1}{2}}$ required for 100-fold amplification of disturbances as a function of the dimensionless initial elastic tension $Te_0 = T_0 a/\sigma$ and dimensionless stress relaxation time D. For this graph the Ohnesorge number $Z_0 = 0{\cdot}001$ is sufficiently small that the shear-thinning property of the liquid is irrelevant. $\xi = 0{\cdot}5$, $\lambda_2/\lambda_1 = 0{\cdot}1$, $\mu_1/\lambda_1 = 1$, $\mu_0/\lambda_1 = 0$, $\hat{W} = 0$.

Figure 3 provides an explanation of the observation that some jets of dilute polymer solutions can travel significant distances with relatively small amplification of disturbances, and that then, suddenly, very large amplification rates develop. For example, with $Te_0 = 1$ and the other conditions of figure 3, the dimensionless distance required for the disturbance to first increase tenfold is 125; a second tenfold increase in the disturbance amplitude will require an additional dimensionless distance of $168 - 125 = 43$; a third tenfold increase in the disturbance amplitude will require an additional dimensionless distance of only $187 - 168 = 19$. The sudden development of large-amplitude waves after long distances of small amplification are associated with larger values of Te_0 and D and smaller values of λ_2/λ_1.

Calculations of this type can be used to relate the length of jet from orifice to point of drop formation to the other variables if the amplitude at the orifice is known. For example suppose the amplitude of the disturbance at the orifice is 0·01 times the jet radius. The disturbance must amplify 100-fold before breakage occurs. For $Te_0 = 1$ and the conditions of figure 3, breakage occurs at a dimensionless length of 168. Figure 4 shows the dimensionless length $(L/U)(\sigma/8\rho a^3)^{\frac{1}{2}}$ required for breakage as a

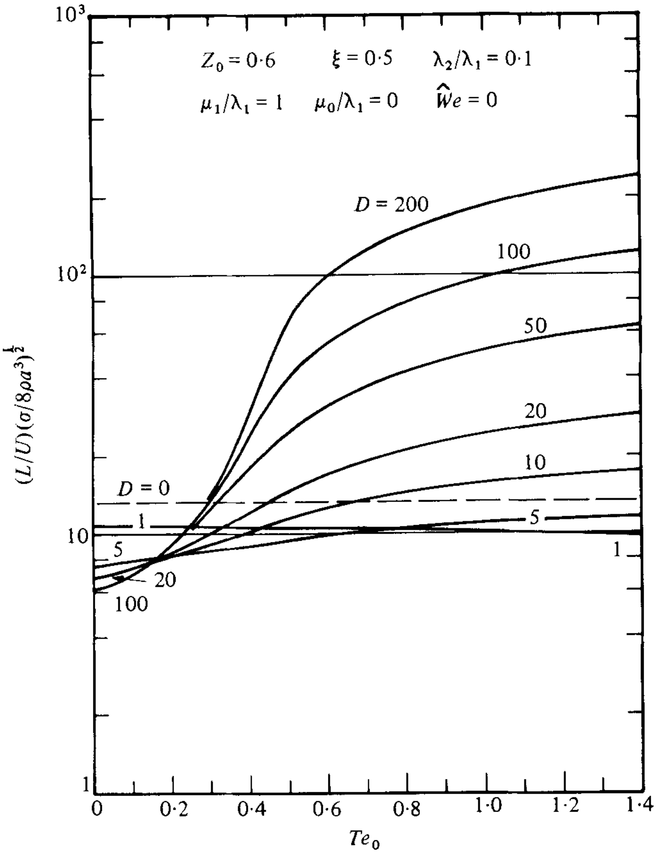

FIGURE 6. Parametric study of the dimensionless length $(L/U)(\sigma/8\rho a^3)^{\frac{1}{2}}$ required for 100-fold amplification of disturbances as a function of the dimensionless initial elastic tension Te_0 and dimensionless stress relaxation time D. For this graph the Ohnesorge number $Z_0 = 0{\cdot}6$ sufficiently large that the competition between the stabilizing influence of the tension and the destabilizing influence of shear thinning is evident. $\xi = 0{\cdot}5$, $\lambda_2/\lambda_1 = 0{\cdot}1$, $\mu_1/\lambda_1 = 1$, $\mu_0/\lambda_1 = 0$, $\hat{W} = 0$.

function of the wavenumber when the initial amplitude is 0·01 times the jet radius. The parameter is the dimensionless tension Te_0 based on the tension at the orifice. Other parameters held constant for this figure are $Z_0 = 0{\cdot}6$, $D = 200$, $\lambda_2/\lambda_1 = 0{\cdot}1$, $\mu_1/\lambda_1 = 1$, $\mu_0/\lambda_1 = 0$, and $\hat{W} = 0$; these are the same conditions as for figure 1. The star marks the minimum length to breakup for a Newtonian liquid jet with $Z = 0{\cdot}6$. We see, in analogy with figure 1, that, the larger Te_0 is, the greater is the length to breakage. Also, as Te_0 increases, the wavenumber for minimum jet length shifts to smaller wavenumbers, indicating longer wavelengths and larger resulting drops. The cumulative effect of small amplification rates at larger Te with larger amplification rates at smaller Te results in a jet length to breakage having a much more shallow minimum than might have been guessed from figure 1 without these calculations. The minimum in jet length is especially shallow for larger Te_0 and D. These calculations may explain the sudden appearance of irregular bulges on jets not subjected to controlled disturbances; the irregularity is traceable to the nearly constant jet length for a spread of wavelengths, so that the wavelength observed more nearly reflects the

Z_0 \ λ_2/λ_1	0·001	0·1	0·4	1·0
0·001	34·0	34·0	34·0	34·0
0·1	34·4	35·0	36·4	39·1
0·6	36·1	38·9	46·0	56·9
1·0	37·4	41·9	52·4	67·9

TABLE 2. Dimensionless breakage length

randomness of the input disturbances than would be the case for a pronounced minimum in breakup length as a function of wavenumber.

Figure 5 is a parametric study of the breakage length as a function of Te_0 and D for disturbances whose initial amplitude is 0·01 times the jet radius. For this figure $\xi = 0{\cdot}5$, $Z_0 = 0{\cdot}001$, $\lambda_2/\lambda_1 = 0{\cdot}1$, $\mu_1/\lambda_1 = 1$, $\mu_0/\lambda_1 = 0$ and $\hat{W} = 0$. At this low value of Z_0, viscous effects are negligible, so that the shear-thinning property of the liquid is irrelevant. The figure was prepared to show the order-of-magnitude increases in jet length brought about by the tension. For $D < 1$ there is a negligibly small effect of Te_0 on the dimensionless breakage length, which is nearly constant at the inviscid value of 5·41. For $D > 1$, as either D or Te_0 increase, the dimensionless breakage length monotonically increases. Tenfold and more increases in jet length are achieved with reasonable values of D and Te_0.

Figure 6 also is a parametric study of the breakage length as a function of Te_0 and D. For this figure $Z_0 = 0{\cdot}6$; the other variables are the same as for figure 5. $Z_0 = 0{\cdot}6$ is sufficiently large that viscous effects are important and the shear-thinning nature of the liquid results in reduced stability. This is most evident at small values of Te_0; increasing D decreases the dimensionless breakage length from the Newtonian value of 13·3 by perhaps a factor of two. At larger Te_0, increasing D from zero first results in a reduction of jet length until D is in the range 1–2 where the reduction is about 30 %. With further increases in D, the jet length increases monotonically. Increases up to tenfold are achievable with reasonable values of D and Te_0.

Comparison of figures 5 and 6 suggests that the dimensionless breakage length is relatively independent of Z_0 for $Z_0 < 0{\cdot}6$ when De is large and Te_0 is not too small. Table 2 was prepared to evaluate the dependence of breakage length on the two parameters Z_0 and λ_2/λ_1 that influence the 'viscosity'. Other conditions for this table are an initial amplitude of 0·01 times the jet radius, $\xi = 0{\cdot}5$, $Te_0 = 0{\cdot}5$, $D = 100$, $\mu_1/\lambda_1 = 1$, $\mu_0/\lambda_1 = 0$ and $\hat{W} = 0$. The entries in table 2 suggest that, if Z_0 or λ_2/λ_1 is sufficiently small, the breakage length is relatively insensitive to changes in these groups. Consequently, under such conditions it is not necessary to know the zero shear viscosity η_0 and the retardation time constant λ_2 in order to estimate the breakage length (unless these parameters are important in predicting T_0 from the prior history of the fluid).

Table 3 has been prepared to give some indication of the sensitivity of the breakage length to μ_0/λ_1. Other conditions for this table are initial amplitude of 0·01 times the jet radius, $\xi = 0{\cdot}5$, $Te_0 = 0{\cdot}5$, $D = 100$, $\lambda_2/\lambda_1 = 0{\cdot}1$, $\mu_1/\lambda_1 = 1$, and $\hat{W} = 0$. An examination of the expressions for the rheological responses of an Oldroyd 8-constant fluid does not appear to restrict μ_0 to positive values only. In fact the kinetic theory of 'dumb-bells' predicts that $\mu_0/\lambda_1 = -\frac{1}{7}$.

Z_0 \ μ_0/λ_1	−0·2	−0·1	0	0·1	0·2	0·5
0·001	57·2	46·2	34·0	22·7	13·9	6·65
0·1	58·0	46·9	35·0	23·5	14·5	6·80
0·6	61·7	50·7	38·9	27·6	17·4	7·64
1·0	64·6	53·6	41·9	31·5	20·0	8·39

TABLE 3. Dimensionless breakage length

The calculated values of the dimensionless breakage length show that positive values of μ_0 are destabilizing and negative values of μ_0 are stabilizing. The lengths appear to be more sensitive to μ_0/λ_1 than to λ_2/λ_1 and Z_0. Consequently, jet-stability measurements may provide some useful information on the elusive physical property μ_0.

It appears from our calculations that for dilute polymer solutions which show zero second normal-stress coefficient (so that $\mu_1 = \lambda_1$), the most-important parameter in the rheological equation of state with regard to jet stability is the stress relaxation time constant λ_1. Larger values of λ_1 give larger values of D and therefore slower growth rates as long as Te is sufficiently large. Larger values of λ_1 give slower relaxation of the axial elastic tension, so that the growth rate remains smaller for a longer elapsed time or distance. Also, it is likely that, the larger λ_1 is, the larger will be the initial axial tension for given kinematics of the prior history.

It is perhaps worth noting the relationship between λ_1 and the elongational viscosity $\bar{\eta}$ at a given rate of strain $\dot{\epsilon}$. One of the simplest cases is a Maxwell fluid which is embedded within the Oldroyd 8-constant model with the following choices for the constants: $\mu_1 = \lambda_1$ and $\mu_2 = \lambda_2 = \nu_1 = \mu_0 = 0$. For this fluid the elongational viscosity is given by $\bar{\eta} = 3\eta_0/(1-\lambda_1\dot{\epsilon}-2\lambda_1^2\dot{\epsilon}^2)$. Clearly, for a given elongational rate, the larger the stress relaxation time is, the larger is the elongational viscosity. It is therefore possible to associate the enhanced stability of liquids with large λ_1 to large $\bar{\eta}$. We think it is preferable to interpret the results in terms of the time constants as these are presumably independent of flow conditions, whereas the elongational viscosity is generally dependent on flow conditions.

4. Conclusion

A linearized stability analysis has been developed for the breakup of viscoelastic liquid threads that may be under elastic tension. The theory appears to account, at least qualitatively, for most of the observed breakage patterns for jets of dilute polymer solutions. For a quantitative comparison of the theory with experimentally measured breakage patterns one would need independent measurement of the rheological responses of the fluid from which the time constants could be inferred. One would also need an independent measure of the initial elastic tension T_0. An experimental study of jet breakup for dilute polymer solutions is currently underway. Our results will be reported in a future paper.

This research was supported by NASA under NASA–Ames University Consortium Number NCA2-OR050-001.

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