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On the Initiation of Melt Fracture

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Synopsis

The importance of $(\tau_{11} - \tau_{22})/\tau_{12}$ (sometimes called the Weissenberg number) in determining the onset of melt fracture is examined using classical linearized hydrodynamic stability analysis. The constitutive relation used is that proposed by Bird and Carreau. It is shown that simple shearing flow of a viscoelastic fluid becomes unstable at a critical value of the Weissenberg number. Implications with regard to polymer processing are discussed.

INTRODUCTION

The presence of a hydrodynamic instability in the extrusion of polymer melts is a problem in the processing of viscoelastic materials. The cause of this instability is still not completely understood despite the extensive work of Tordella,[1,2] Bagley et al.,[3-5] Pearson,[6-8] Bogue and White,[9,10] Han,[11,12] and many others.[13] The recent paper by Ballenger et al.[14] summarizes many possible approaches taken in the past and calls attention to the critical role played by the ratio of the first normal stress difference divided by the shear stress, $N_1 = (\tau_{11} - \tau_{22})/\tau_{12}$ (see Fig. 1). Below, a simple classical linearized hydrodynamic stability problem is posed as a model for the initiation of instabilities in the extrusion process. It is shown that at a critical value of N_1 a hydrodynamic instability is developed. Implications with respect to melt fracture are then discussed.

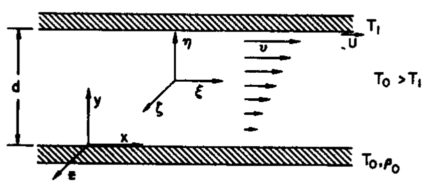

Fig. 1. The physical situation.

CONSTITUTIVE RELATION

The constitutive relation used in this work is that proposed by Bird and Carreau[15,16]

$$\boldsymbol{\tau} = \mathbf{S} + p\mathbf{I} = \sum_p \int_0^\infty m_p[(t - t'),\mathrm{II}(t')] \left[\left(1 + \frac{\delta}{2}\right)\mathbf{B} + \frac{\delta}{2}\mathbf{C}\right] dt' \quad (1)$$

where

$$m_p = \frac{\eta_p \exp[-(t - t')/\lambda_{2p}]}{\lambda_{2p}^2[1 + 2\mathrm{II}(t')\lambda_{2p}^2]}$$

$$C_{ij} = \frac{\partial x_\alpha'}{\partial x_i}\frac{\partial x_\alpha'}{\partial x_j} - \delta_{ij} \tag{2}$$

$$B_{ij} = \frac{\partial x_i}{\partial x_\alpha'}\frac{\partial x_j}{\partial x_\alpha'} - \delta_{ij}$$

$\delta = \dfrac{\tau_{22} - \tau_{33}}{\tau_{11} - \tau_{22}}$ = ratio of normal stress differences in simple shearing flow

δ_{ij} = Kroneker delta

$\eta_p, \lambda_{1p}, \lambda_{2p}$ = material constants

and

$\mathrm{II}(t')$ = second invariant of the rate of strain tensor.

This equation is of the same general form as many recently proposed modern nonlinear viscoelastic constitutive relations. The exact form of the relation is not crucial to the argument below, but the model must at least allow for finite normal stress differences.

DEVELOPMENT OF THE HYDRODYNAMIC STABILITY PROBLEM

Consider the model fluid given above flowing in plane Couette flow (simple shearing flow) with a superposed temperature gradient (see Fig. 1). A solution to the conservation equations, neglecting viscous dissipation and assuming a Fourier heat conduction vector is given by[17]

$$v = i\dot{\gamma}y$$
$$T = T_0 - \beta y \tag{3}$$

where $\beta = (T_0 - T_1)/d$ and $\dot{\gamma}$ = magnitude of the velocity gradient.

Following the usual linearized hydrodynamic stability theory (see Chandrasekhar[18]), the steady-state solution is perturbed by a small disturbance. It is assumed that the altered motion satisfies the conservation equations. All products of the disturbances and their derivatives are neglected (linearization assumption). The steady-state flow is then subtracted, and the result is a set of linear partial differential equations for the disturbances. The pressure disturbance is normally eliminated by taking the curl of the linear momentum conservation equation. In the present problem three additional assumptions are made:

1. The Boussinesq assumption, that is, all the material fluid properties are unaffected by the temperature fields except the density in the gravita-

tional force form. The variation of density is assumed to be a linear function of temperature in this term:

$$\rho = \rho(1 + \alpha\beta y)$$

where $\alpha = \frac{1}{V}\left(\frac{\partial V}{\partial T}\right)$ = volume coefficient of expansion and V = specific volume.

2. The disturbances depend only on two coordinates—the flow direction and the vertical direction.

3. The normal mode analysis is used.

The equations are non-dimensionalized using the following variables:

$$\frac{v}{U} = (u_\xi, u_\eta, u_\zeta)$$

$$\xi = \frac{x}{d}$$

$$\eta = \left(\frac{y}{d} - \frac{1}{2}\right)$$

$$\zeta = \frac{z}{d}$$

$$\frac{\partial}{\partial t} = \frac{d}{U}\left(\frac{\partial}{\partial t}\right)$$

$$U = \dot{\gamma} d$$

$$\psi = \frac{T'}{T_0 - T_1} = \frac{T}{\Delta T}$$

$$\theta = \frac{\alpha g \Delta T d^2 \rho_0 \nu_x{}^2 \psi}{\beta_0 U}$$

$$u_\xi = \frac{i}{\nu_x}\frac{dv(\eta)}{d\eta}\exp(i\,\nu_x\xi + \sigma t)$$

$$u_\eta = v(\eta)\exp(i\nu_x\xi + \sigma t)$$

$$u_\zeta = \frac{-i}{\nu_x}\frac{dv(\eta)}{d\eta}\exp(i\nu_x\xi + \sigma t)$$

$$\theta = \theta(\eta)\exp(i\nu_x\xi + \sigma t)$$

where ν_x = disturbance wave number (real), σ = disturbance growth rate (complex), and $i = \sqrt{-1}$. The reduced linearized momentum and energy conservation equations for the disturbance functions $v(\eta)$ and $\theta(\eta)$ are given by

Momentum:

$$Re(1 + \sigma\lambda)^2(\sigma D^2 v + i\nu_x\eta D^2 v) = (1 + \sigma\lambda)D^4 v + (2 + \sigma\lambda)(i\nu_x A - i\nu_x B)D^3 v + \frac{2Cg(\sigma\lambda)\nu_x{}^2}{(1 + \sigma\lambda)}(D^2 v + \nu_x{}^2 v) + \frac{2Ei\nu_x{}^3 h(\sigma\lambda)}{(1 + \sigma\lambda)^2}Dv - (1 + \sigma\lambda)^2\theta \quad (4)$$

Energy:

$$Pe(1 + \sigma\lambda)^2(\sigma\theta + i\nu_x\eta\theta) = (1 + \sigma\lambda)^2[Ra\nu_x{}^2v + D^2\theta] \quad (5)$$

where

$$Ra = \text{Rayleigh number} = \frac{\alpha g \Delta T d^3 \rho^2 C_v}{k\beta_0}$$

$$Re = \text{Reynolds number} = \frac{\rho_0 d^2 \dot{\gamma}}{\beta_0}$$

$$Pe = \text{Peclet number} = \frac{\rho_0 \hat{c}_v \dot{\gamma} d^2}{k}$$

$$g(\sigma\lambda) = 2[3 + 3(\sigma\lambda) + (\sigma\lambda)^2]$$

$$h(\sigma\lambda) = 6[4 + 6\sigma\lambda + 4(\sigma\lambda)^2 + (\sigma\lambda)^3]$$

$$A = \left[\sum_{p=1}^{\infty} m_p\lambda_{2p}{}^3 \Big/ \sum_{p=1}^{\infty} m_p\lambda_{2p}{}^2\right]\dot{\gamma}\left[1 + \frac{\delta}{2}\right] \quad (6)$$

$$B = \left[\sum_{p=1}^{\infty} m_p\lambda_{2p}{}^3 \Big/ \sum_{p=1}^{\infty} m_p\lambda_{2p}{}^2\right]\dot{\gamma}\,\frac{\delta}{2}$$

$$C = \left[\sum_{p=1}^{\infty} m_p\lambda_{2p}{}^4 \Big/ \sum_{p=1}^{\infty} m_p\lambda_{2p}{}^2\right]\left(1 + \frac{\delta}{2}\right)\dot{\gamma}^2$$

$$E = \left[\sum_{p=1}^{\infty} m_p\lambda_{2p}{}^5 \Big/ \sum_{p=1}^{\infty} m_p\lambda_{2p}{}^2\right]\left(1 + \frac{\delta}{2}\right)\dot{\gamma}^3$$

$$\beta_0 = \sum_{p=1}^{\infty} m_p\lambda_{2p}{}^2 - \text{non-Newtonian viscosity}$$

$$D = \frac{d}{d\eta} \qquad \lambda = \sum_{p=1}^{\infty} \lambda_{2p}$$

It should be noted here that: $N_1 = A - B = (\tau_{11} - \tau_{22})/\tau_{12}$ = reduced first normal stress difference and $N_2 = B = (\tau_{22} - \tau_{33})/\tau_{12}$ = reduced second normal stress difference.

Equations (4) and (5) are simplified somewhat by assuming that near the neutral stability curve, $\sigma \ll 1$ and all terms of order σ^2 are neglected. Also, as experimental information is not available, it is assumed $C = E = 0$.

The resulting equations to be solved below are given by

Momentum:

$$\sigma[Re\, D^2v - \lambda[D^4v - 2Re\, i\nu_x\eta D^2v + i[\nu_x(A - B)D^3v - 2\theta]]] = D^4v - Re\, i\nu_x\eta D^2v + 2i\nu_x(A - B)D^3v - \theta \quad (7)$$

Energy:

$$\sigma[Pe\,\theta - \lambda[2Ra\nu_x{}^2v - 2Pe\, i\nu_x\eta\theta + 2D^2\theta]] = Ra\nu_x{}^2v - Pe\, i\nu_x\eta\theta + D^2\theta \quad (8)$$

with boundary conditions

$$\left.\begin{aligned} v = Dv = 0 \\ \theta = 0 \end{aligned}\right\} \text{at } \eta = \pm \tfrac{1}{2}. \tag{9}$$

SOLUTION OF THE PERTURBATION EQUATIONS

The method of solution employed is a modified Galerkin procedure due mainly to Finlayson[19] and used successfully by others in similar hydrodynamic stability problems.[17,20] The functions $v(\eta)$ and $\theta(\eta)$ are approximated by a complete set of functions which satisfy the problem boundary conditions. The coefficient of each approximation function is determined by making the equation residuals orthogonal to the set of approximation functions in the domain of interest. This procedure gives a set of $2N$ algebraic equations for the $2N$ unknown coefficients. Finlayson[19] modified this technique by allowing the coefficients to be time dependent. The left-hand side of eqs. (7) and (8) are then regarded as being differentiated with respect to time rather than multiplied by σ:

$$v \cong \sum_{i=1}^{N} a_i^1(t)\Phi_i \qquad \theta \cong \sum_{i=1}^{N} a_i^2(t)\phi_i \tag{10}$$

where $\Phi_i = \eta^{i-1}\left(\eta^2 - \dfrac{1}{4}\right)^2$ and $\phi_i = \eta^{i-1}\left(\eta^2 - \dfrac{1}{4}\right)$.

Putting these approximations into eqs. (7) and (8), forming the equation residuals, and orthogonalizing with respect to the Φ_j and ϕ_j over $-\frac{1}{2} \leq \eta \leq \frac{1}{2}$ results in the following set of equations, represented symbolically by

$$\frac{dA}{dt} = \mathbf{H}A \tag{11}$$

where A is a column matrix such that $A^T = (a_1^1 \cdots a_N^1, a_1^2 \cdots a_N^2)$ and $\mathbf{H}$ is a $2N$-by-$2N$ complex valued matrix whose coefficients are functions of the dimensionless groups of the problem and the disturbance wave number. The condition of stability is that all of the eigenvalues of $\mathbf{H}$ have negative real parts. The dimensionless groups and wave number can be varied to determine the point of neutral stability—the point where one $\sigma_r = 0$. Numerically, it was found that taking three terms in each expansion ($N = 3$) produced agreement to four places with the tabulated values of critical Rayleigh number for the Newtonian problem given in Chandrasekhar.[18] This value of N was used for all the reported numerical work.

RESULTS AND DISCUSSION

It is of interest to note that the only non-Newtonian parameter appearing in eqs. (7) and (8) is $B - A = -N_1$ (recalling the assumption $C = E = 0$). This is similar to the work of McIntire and Schowalter[17] who examined the same problem for a second-order fluid and small values of N_1

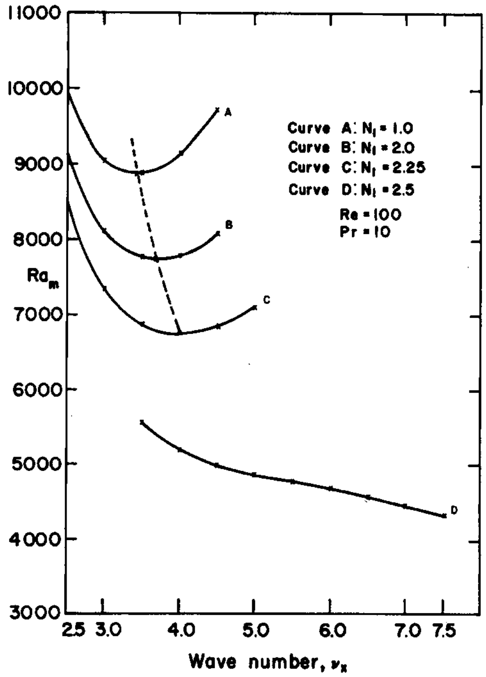

Fig. 2. Marginal Rayleigh numbers as a function of disturbance wave number. The critical Rayleigh number is indicated by the dotted line.

($N_1 < 0.5$). The results for the critical Rayleigh number are extremely interesting. As shown in Figures 2 and 3, when N_1 is increased, for given values of Reynolds number and Prandtl number, the critical Rayleigh number (dimensionless temperature gradient) first increases, then decreases rapidly; and at a value of $2.25 < N_1 < 2.5$, a flow with zero Rayleigh number is unstable. This indicates a purely rheological hydrodynamic instability with no buoyancy effects, as isothermal Newtonian plane Couette flow is known to be stable at all Reynolds numbers to linearized disturbances. The critical wave number of the instability is quite large. The implications with regard to melt fracture are interesting. This analysis indicates that simple shearing flow of a viscoelastic material becomes unstable at a critical value of $(\tau_{11} - \tau_{22})/\tau_{12} = N_1$. This is in agreement with the collected experimental results of Ballenger et al.[14] and others. Curves of N_1 as a function of shear rate are given in the work of Ballenger et al.[14] and are shown qualitatively in Figure 4. In the range of shear rates where data are available, two types of behavior are shown: (a) N_1 monotone increasing with shear rate, and (b) N_1 passing through a local maximum and local minimum with increasing shear rate.

Behavior of type (b) is interesting in that there are sometimes two regions of stable operation of extrusion; a low shear rate region and a high shear rate region. If a critical value of N_1 is the determining factor of the melt fracture instability, then the high shear rate region of the local minimum of N_1 may be responsible for the second region of stable operation. This is admittedly a speculation, but from a quantitative stability result. Investigation is proceeding into the effects of allowing viscous

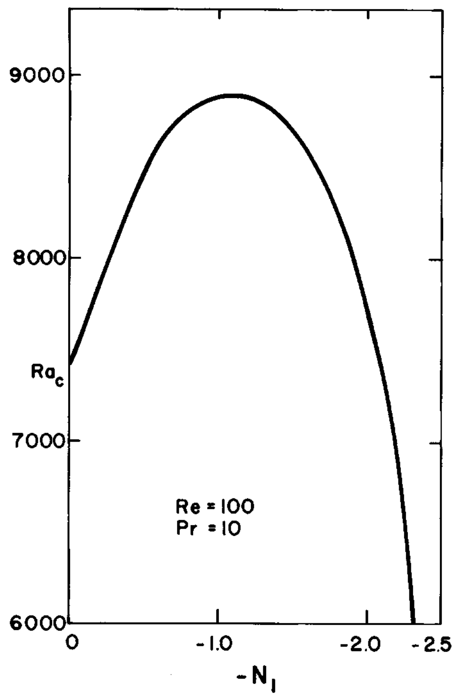

Fig. 3. Critical Rayleigh number as a function of the rheological parameter N_1 (Weissenberg number).

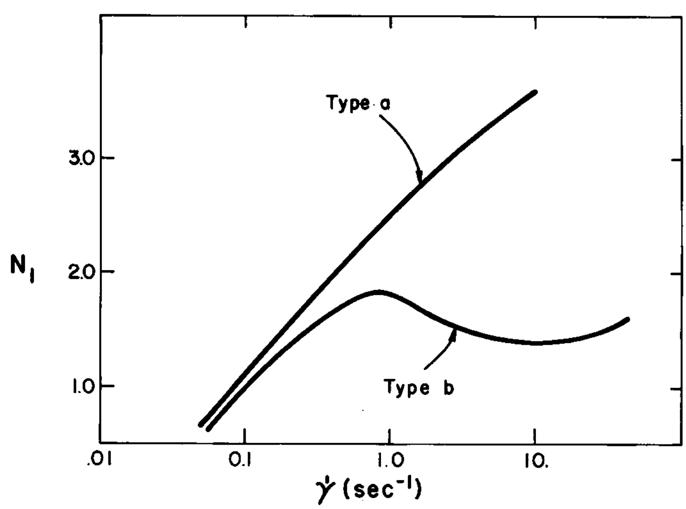

Fig. 4. Qualitative behavior of N_1 as a function of shear rate for polymer melts.

dissipation, non-zero rheological parameters C and E, and of changes in Reynolds and Prandtl numbers.

CONCLUSIONS

A quantitative explanation, using classical linearized hydrodynamic stability theory, of the possible importance of the parameter $N_1 = (\tau_{11} - \tau_{22})/\tau_{12}$ in the problem of melt fracture has been given. The results indicate that at a critical value of N_1, simple shearing flow of a viscoelastic material exhibits a hydrodynamic instability. This is in dramatic contrast to a Newtonian fluid where hydrodynamic instability in isothermal simple shearing flow is not found. An explanation of the high and low shear rate range of stable operation in extrusion processes is also postulated.

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