# Elastic short wave instability in extrusion flows of viscoelastic liquids

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## Abstract

An analysis of the stability to short waves of the flow of concentric coextruded polymeric liquids modeled by upper convected Maxwell models is presented. The flow can be unstable to short waves under various conditions on the elastic parameter. The growth rates for the short wave instability in the elastic case are finite at leading order. The same short wave instability is known to be two orders of magnitude smaller in the purely viscous case with growth rates proportional to $\alpha^{-2}$ for large $\alpha$. It is argued that this instability could appear in systems with low surface tension, as in polymer-depleted solutions at the walls of a pipe. Some speculative scenarios concerning the appearance of sharkskin after the loss of adhesion, leading to wet slip, are advanced.

## 1. Introduction

Coextrusion is an industrial process frequently used to form multilayered sheets and films having unique optical and mechanical properties. One of the most important problems in coextrusion is to obtain a smooth interface between the individual components in the final product. It has been found that under certain operating conditions or with a certain combination of extruding liquids, the interface between the two extruding components becomes irregular, resulting in interfacial instability that must be avoided in obtaining products of acceptable quality. A better understanding of the interfacial instability and its correlations to the processing conditions, and of the rheological properties of the individual fluids, could be of value in the production of coextruded composites of desired properties and of consistent quality.

It is well known that interfacial instability can occur in plane shearing motions of two liquids of different viscosities (Yih [1967]; Hooper & Boyd [1983, 1987]; etc.). Studies of the stability of plane Couette flow of two viscoelastic liquids to long waves have been carried out by Li [1969] and Waters & Keely [1987]. All of these early investigations concerning viscoelastic liquids conclude that fluid elasticity has no effect on the stability when the viscosities of the two liquids are the same. Recently Y. Renardy [1988] found a new instability in the short wave limit which is solely due to the difference in the elastic properties of two upper convected Maxwell fluids in plane Couette flow. This instability persists even when the viscosities of the two fluids are the same. Chen [1991a] also found this type of instability in the long wave limit for concentric coextrusion flow of two viscoelastic fluids in a circular pipe. In a later publication, Chen [1991b] re-examined the problem of plane Couette flow of two viscoelastic fluids in the long wave limit considered by Li [1969], Waters & Keely [1987], and found an error in their interfacial shear stress condition. The corrected result shows that the instability resulting from the different elastic properties persists in the long wave limit, consistent with Y. Renardy's short wave calculation. It is then apparent that this type of elastic instability will occur for any disturbance wave number. Chen [1991b] showed that for the basic flow, the jump in the first normal stress difference across the unperturbed interface does not need to be balanced. When the interface is perturbed, it is this unbalanced jump in normal stress difference that causes the purely elastic instability found by Y. Renardy [1988] and Chen [1991a, b].

In this paper, we further explore the instability induced by elasticity in extrusion flows. We first study the short wave behavior of this elastic instability for coextrusion flow of two upper convected Maxwell fluids in a circular pipe, an analog of the problem studied by Y. Renardy [1988]. We show that in the short wave limit, elastic effects dominate over viscous effects, while in the long wave limit these effects are of the same order of magnitude (Chen [1991b]). The short wave instability will be stabilized by surface tension and we estimate the cut-off wave length for stabilization by surface tension. We introduce a two-fluid model for the problem of slip at the wall and discuss some speculative ideas about the relationship of the elastic instability to sharkskin.

## 2. Basic flow and formulation of the stability problem

Consider the core-annular flow of two immiscible upper convected Maxwell fluids inside a circular pipe of inner radius $R_2$, driven by a constant pressure gradient. The interface between the two fluids is a perfect cylinder, $r = R_1$. Fluid 1 is located in the core and fluid 2 in the annulus. We are interested in the stability of this core-annular flow.

The continuity and momentum balance equations are

$$\operatorname{div} \mathbf{U} = 0 , \tag{2.1}$$

$$\rho\left[\frac{\partial \mathbf{U}}{\partial t} + (\mathbf{U}\cdot\partial)\mathbf{U}\right] = -\nabla P + \operatorname{div}\mathbf{S} ,$$

where $\rho$ is the fluid density, $\mathbf{U}=\mathbf{e}_r\, u + \mathbf{e}_\theta\, v + \mathbf{e}_x\, w$, and the stress tensor $\mathbf{T}$ is given by

$$\mathbf{T} = -\, P\, \mathbf{I} + \mathbf{S} , \tag{2.2}$$

$\mathbf{S}$ being the extra stress tensor. The upper convected Maxwell constitutive equation is

$$\lambda \overset{\nabla}{\mathbf{S}} + \mathbf{S} = 2\,\eta\,\mathbf{D}[\mathbf{U}],$$

where

$$\overset{\nabla}{\mathbf{S}} = \frac{\partial \mathbf{S}}{\partial t} + (\mathbf{U}\cdot\nabla)\mathbf{S} - \mathbf{LS} - \mathbf{SL}^T,$$
$$\mathbf{L} = \nabla \mathbf{U}\,,\ L_{ij} = \frac{\partial U_i}{\partial x_j}\,, \tag{2.3}$$
$$\mathbf{D}[\mathbf{U}] = \frac{1}{2}(\,\mathbf{L} + \mathbf{L}^T\,)\,,$$

$\lambda$ is the relaxation time and $\eta$ is the constant viscosity (Joseph [1990]). These equations are written in cylindrical coordinates $(r, \theta, x)$ for both fluid 1 and fluid 2.

On the pipe wall, we have the no-slip condition $\mathbf{U} = 0$. The velocity components and the stresses are bounded at the centerline $r = 0$. The equations on the fluids interface $r = R(\theta, x, t)$ are

$$U = \frac{\partial R}{\partial t} + W\frac{\partial R}{\partial x} + \frac{V}{R}\frac{\partial R}{\partial \theta}\,,$$
$$[[\mathbf{U}]] = 0,$$

where $[[f]] = (f)_1-(f)_2$ is the jump in f across the interface, and

$$[[\mathbf{T}]]\,\mathbf{n} + 2\,H\,T\,\mathbf{n} = 0,$$

where $2\,H$ is the sum of the principal curvatures, $T$ is the coefficient of interfacial tension, $\mathbf{n} = \mathbf{n}_{12}$ is the normal to $r-R(\theta, x, t) = 0$ from liquid 1 to 2.

The basic flow with a perfect cylindrical interface $r = R_1$ in the absence of gravity is given by

$$\hat{\mathbf{U}} = \mathbf{e}_x \hat{W}(r)\,, \tag{2.4}$$
$$[[\hat{P}\,]] = \frac{T}{R_1}\,.$$

Let

$$\frac{d\hat{P}_1}{dx} = \frac{d\hat{P}_2}{dx} = -f, \tag{2.5}$$

then the velocity profile and the non-vanishing stress components of the basic flow are

(a) core, $0 \le r \le R_1$,

$$\begin{aligned} \hat{W}_1(r) &= \frac{f}{4\eta_1}(R_1^2 - r^2) + \frac{f}{4\eta_2}(R_2^2 - R_1^2), \\ \hat{S}_{rx} &= -\frac{f}{2}\, r, \\ \hat{S}_{xx} &= 2\frac{\lambda_1}{\eta_1}(\hat{S}_{rx})^2. \end{aligned} \tag{2.6}$$

(b) annulus, $R_1 \le r \le R_2$,

$$\begin{aligned} \hat{W}_2(r) &= \frac{f}{4\eta_2}(R_2^2 - r^2), \\ \hat{S}_{rx} &= -\frac{f}{2}\, r, \\ \hat{S}_{xx} &= 2\frac{\lambda_2}{\eta_2}(\hat{S}_{rx})^2. \end{aligned} \tag{2.7}$$

We choose the centerline velocity $\hat{W}(0)$ as the velocity scale, $R_1$ as the length scale, $\frac{R_1}{\hat{W}(0)}$ as the time scale, and $\eta_l \frac{\hat{W}(0)}{R_1}$ as the stress scale in each region, $l = 1, 2$. The pressure in each region is scaled as a stress. We define the following dimensionless parameters

$$\begin{aligned} &a = \frac{R_2}{R_1}, \\ &(m_1, m_2) = (1, \frac{\eta_2}{\eta_1}), \\ &\boldsymbol{R}_l = \frac{\rho_l \hat{W}(0) R_1}{\eta_l}, \quad l = 1, 2 \text{ (Reynolds numbers)} \\ &W_l = \frac{\lambda_l \hat{W}(0)}{R_1}, \quad l = 1, 2 \text{ (Weissenberg numbers)} \\ &J = \frac{TR_2}{\rho_1 \nu_1^2}, \quad \nu_1 = \frac{\eta_1}{\rho_1}, \\ &Ca = \frac{\lambda_1 T}{\eta_1 R_1} \text{ (capillary number)}. \end{aligned} \tag{2.8}$$

The parameters J and Ca are both independent of flow conditions and the ratio

$$\frac{T/R_1}{\eta_1 \hat{W}(0)/R_1} = \frac{J}{a\boldsymbol{R}_1} = \frac{Ca}{W_1}$$

is a measure of the interfacial tension force relative to the viscous force.

In terms of these controlling parameters, the linearized equations for disturbances, in each region, are given by

$$\text{div}\,\mathbf{u} = 0,$$

$$\boldsymbol{R}\left[\frac{\partial \mathbf{u}}{\partial t} + \hat{W}\frac{\partial \mathbf{u}}{\partial x} + \mathbf{e}_x \hat{W}' u\right] = -\nabla p + \text{div}\boldsymbol{\tau},$$

$$W\left\{\frac{\partial \boldsymbol{\tau}}{\partial t} + \hat{W}\frac{\partial \boldsymbol{\tau}}{\partial x} + (\mathbf{u}\cdot\partial)\hat{\mathbf{S}} - \left[(\partial\mathbf{u})\hat{\mathbf{S}} + \hat{W}'\mathbf{e}_x\mathbf{e}_r\,\boldsymbol{\tau}\right] - \left[(\partial\mathbf{u})\hat{\mathbf{S}} + \hat{W}'\mathbf{e}_x\mathbf{e}_r\,\boldsymbol{\tau}\right]^T\right\} + \boldsymbol{\tau}$$

$$= 2\,\eta\,\mathbf{D}[\mathbf{u}], \tag{2.9}$$

where, $\mathbf{u}$ (= $\mathbf{e}_r$ u + $\mathbf{e}_\theta$ v + $\mathbf{e}_x$ w), p are the perturbation velocity and perturbation pressure respectively, $\boldsymbol{\tau}$ is the perturbation of the extra stress tensor, superscript T stands for transpose, and primes are the derivatives with respect to r.

On the pipe wall, r = a, we have the no-slip condition

$$\mathbf{u} = 0. \tag{2.10}$$

At the centerline of the pipe, $\mathbf{u}$, p and $\boldsymbol{\tau}$ are bounded.

At the unperturbed fluids interface r = 1, we have the

kinematic equation of motion: $u = \frac{\partial \delta}{\partial t} + \hat{W}(1)\frac{\partial \delta}{\partial x}$ ,

the continuity of velocity: [[u]] = 0,

$$[[v]] = 0, \tag{2.11}$$

$$[[w]] + [[\hat{W}'(1)]]\,\delta = 0,$$

and the stress balance: $[[m\,\tau_{rx}]] + [[m\,\hat{S}_{rx}'(1)]]\,\delta - [[m\,\hat{S}_{xx}]]\frac{\partial \delta}{\partial x} = 0,$

$$[[m\,\tau_{r\theta}]] = 0,$$

$$-[[m\,p]] + [[m\,\tau_{rr}]] = \frac{Ca}{W_1}\left(\frac{\partial^2 \delta}{\partial \theta^2} + \frac{\partial^2 \delta}{\partial x^2} + \delta\right),$$

where $\delta(\theta, x, t)$ is the deviation of the interface from r = 1 and the jump

$$[[\,\cdot\,]] = (\,\cdot\,)_1 - (\,\cdot\,)_2$$

is evaluated at r = 1.

The dimensionless basic flow is given by

(1) core: $0 \le r \le 1$,

$$\begin{aligned} \hat{W}_1(r) &= 1 - \frac{m_2 r^2}{a^2 + m_2 - 1}, \\ \hat{S}_{rx} &= -\frac{2 m_2 r}{a^2 + m_2 - 1} = \hat{W}_1', \\ \hat{S}_{xx} &= 2 W_1 (\hat{S}_{rx})^2. \end{aligned} \tag{2.12}$$

(2) annulus: $1 \le r \le a$,

$$\begin{aligned} \hat{W}_2(r) &= \frac{a^2 - r^2}{a^2 + m_2 - 1}, \\ \hat{S}_{rx} &= -\frac{2 r}{a^2 + m_2 - 1} = \hat{W}_2', \\ \hat{S}_{xx} &= 2 W_2 (\hat{S}_{rx})^2. \end{aligned} \tag{2.13}$$

In this paper, only axisymmetric disturbances with $v = \tau_{r\theta} = \tau_{x\theta} = 0$, $\frac{\partial}{\partial\theta} = 0$ are considered. Introducing normal modes

$\{u, w, p, \tau_{rr}, \tau_{rx}, \tau_{\theta\theta}, \tau_{xx}, \delta\} = \{i\,\tilde{u}, \tilde{p}, \tilde{w}, \tilde{\tau}_{rr}, \tilde{\tau}_{rx}, \tilde{\tau}_{\theta\theta}, \tilde{\tau}_{xx}, \tilde{\delta}\}\,\exp[i\,\alpha(x-ct)]$,

and eliminating $\tilde{p}$, $\tilde{w}$ result in, after suppressing ~, the following normal mode equations:

$$\left(\tau_{rx}'' + \frac{\tau_{rx}'}{r} - \frac{\tau_{rx}}{r^2} + \alpha^2 \tau_{rx}\right) - \frac{i\,\alpha}{r}(r\,\tau_{rr})' + i\,\alpha\,\tau_{xx}' + \frac{i\,\alpha}{r}\,\tau_{\theta\theta}$$
$$= -\,i\,\boldsymbol{R}\,(\hat{W}(r) - c)\left(u'' + \frac{u'}{r} - \frac{u}{r^2} - \alpha^2 u\right),$$

$$[i\,\alpha\,W(\hat{W}(r) - c) + 1]\,\tau_{rr} + 2\,\alpha\,W\,\hat{S}_{rx}\,u = 2\,i\,u',$$

$$\alpha\,[i\,\alpha\,W(\hat{W}(r) - c) + 1]\,\tau_{rx} +$$

$$\alpha W [i \hat{S}_{rx}{}' u + \alpha \hat{S}_{xx} u + i \alpha \hat{S}_{rx} \frac{u}{r} - \hat{W}'(r) \tau_{rr}] = -\alpha^2 u - (u'' + \frac{u'}{r} - \frac{u}{r^2}),$$

$$[i \alpha W ( \hat{W}(r) - c) + 1 ] \tau_{\theta\theta} = 2 i \frac{u}{r} , \tag{2.14}$$

$$\alpha [i \alpha W (\hat{W}(r) - c) + 1 ] \tau_{xx} + W \left\{ i\alpha \hat{S}_{xx}{}' u - 2 \left[ - \hat{S}_{rx} \left( u'' + \frac{u'}{r} - \frac{u}{r^2} \right) - i\alpha \hat{S}_{xx} \left( u' + \frac{u}{r} \right) + \alpha \hat{W}'(r) \tau_{rx} \right] \right\} = -2 i \alpha (u' + \frac{u}{r} )$$

$r = 0$: $u(0) = 0$, $u'(0)$ bounded.

$$r = a: u(a) = u'(a) = 0. \tag{2.15}$$

$r = 1$: $u = \alpha ( \hat{W}(1) - c) \delta$,

$$\begin{aligned} &[[u]] = 0, \\ &[[u']] - \alpha [[\hat{W}']] \delta = 0, \\ &[[m \tau_{rx}]] + ([[m \hat{S}_{rx}{}']] - i \alpha [[m \hat{S}_{xx}]]) \delta = 0, \\ &- [[m (\tau_{rx}{}' + \tau_{rx} + i \alpha \tau_{xx} )]] + i \alpha [[m \tau_{rr}]] - \frac{Ca}{W_1} i \alpha (1 - \alpha^2) \delta = 0 , \end{aligned} \tag{2.16}$$

where

$$[[\hat{W}']] = 2 \frac{1-m_2}{m_2 + a^2 - 1} ,$$

$$[[m \hat{S}_{rx}{}']] = 0, \text{ when the densities are matched,} \tag{2.17}$$

$$[[m \hat{S}_{xx} ]] = 8 \frac{m_2 (m_2 W_1 - W_2)}{(m_2 + a^2 - 1)^2} .$$

From the interfacial conditions, it can be seen that interfacial instability can arise from the term containing $\frac{Ca}{W_1}$ in the normal component stress balance equation at the interface. This is the capillary instability, well known since the work of Rayleigh [1879]. Instability can also arise from the jump in the basic flow velocity slope $[[\hat{W}']]$ in (2.16). Since $[[\hat{W}']]$ is proportional to the viscosity stratification $\eta_1-\eta_2$ , this instability is due to viscosity stratification, an instability discovered by Yih [1967]. An instability in vertical flow not considered here has been discussed by Hickox [1971], Smith [1989] and Chen, Bai & Joseph [1990]. This instability is due to the density stratification $\rho_1-\rho_2$ . The term $[[m\,\hat{S}_{xx}]]$ in the shear stress balance equation, identically zero if the fluids under consideration are all Newtonian, can give rise to new instabilities for viscoelastic fluids. This term is the jump in the first normal stress difference of the basic flow across the interface. In the basic flow, the normal stress difference across the interface is not balanced, because it is multiplied by the slope of the interfacial shape which is identically zero for the core-annular flow. The role this term plays in the stability of coextrusion has been explored by Chen [1991a] in the long wave limit $\alpha\to 0$. It is shown that in the long wave limit, the effects of viscosity stratification and of elasticity stratification are of the same order and the growth rate is $O(\alpha^2)$, infinitesimally small as $\alpha\to 0$. However, inspection of (2.11) indicates that the effect of elasticity stratification will dominate in the short wave limit $\alpha\to\infty$, since the jump in normal stress difference $[[m\,\hat{S}_{xx}]]$ is multiplied by the slope of the interface, $\dfrac{\partial\delta}{\partial x}$ . Thus the stability behavior of coextrusion of viscoelastic fluids in the short wave limit is much more interesting than in the long wave limit because of the much stronger effect of the fluids' elasticity.

### 3. Asymptotic solution for short waves

Following Hooper & Boyd [1983], the short wave asymptotic of the problem (2.14)–(2.16) when $\alpha\to\infty$ can be examined by introducing the following transformations:

$$y = \alpha\,(r-1), \quad \eta = \alpha\,\delta\,. \tag{3.1}$$

For simplicity, surface tension is neglected in the following analysis, although there is no difficulty of incorporating such effect. This can be done by setting Ca = 0 or assuming small surface tension (see Chen [1991c]). The variable $\eta$ can be eliminated from the interfacial equations. Since both even and odd derivatives appear in the governing equations, we have to include all the powers of $1/\alpha$ in the asymptotic expansions of u and c in the limit $\alpha\to\infty$ :

$$u(y, \alpha) = u^{(0)}(y) + \frac{u^{(1)}(y)}{\alpha} + \frac{u^{(2)}(y)}{\alpha^2} + \frac{u^{(3)}(y)}{\alpha^3} + O\left(\frac{1}{\alpha^4}\right),$$

$$c = \hat{W}(1) + \frac{c^{(0)}}{\alpha} + \frac{c^{(1)}}{\alpha^2} + \frac{c^{(2)}}{\alpha^3} + O\left(\frac{1}{\alpha^4}\right). \tag{3.2}$$

After substituting (3.1), (3.2) into (2.14)–(2.16), at the lowest order, we obtain

$$\varepsilon \left\{ 1 - i\, W\, c^{(0)} - 2\, i\, W\, s\, y \right\} \tau_{rr}(y) - 4\, W\, s\, u^{(0)}(y) - 2\, i\, u^{(0)\prime}(y) = 0,$$

$$\varepsilon \left\{ 2\, W\, s\, \tau_{rr}(y) + (\, 1 - i\, W\, c^{(0)} - 2\, i\, W\, s\, y\,)\, \tau_{rx}(y) \right\} + \tag{3.3}$$
$$(\, 1 + 8\, W^2 s^2\,)\, u^{(0)}(y) + u^{(0)\prime\prime}(y) = 0,$$

$$\left\{ 1 - i\, W\, c^{(0)} - 2\, i\, W\, s\, y \right\} \tau_{\theta\theta}(y) - 2\, i\, u^{(0)}(y) = 0,$$

$$\varepsilon \left\{ 4\, W\, s\, \tau_{rx}(y) + (\, 1 - i\, W\, c^{(0)} - 2\, i\, W\, s\, y\,)\, \tau_{xx}(y) \right\} +$$
$$(\, 2\, i + 16\, i\, W^2 s^2\,)\, u^{(0)\prime}(y) - 4\, W\, s\, u^{(0)\prime\prime}(y) = 0\,,$$

valid for both the core fluid “1” and annulus fluid “2” and

$$\varepsilon = \frac{1}{\alpha}\,,\ s_1 = \frac{m_2}{a^2 + m_2 - 1}\,,\ s_2 = \frac{1}{a^2 + m_2 - 1}\,. \tag{3.4}$$

At the interface y = 0, we have, to the lowest order,

$$[[u^{(0)}]] = 0,$$

$$[[\hat{W}'(1)]]\, u^{(0)}(0) + c^{(0)}\, [[u^{(0)\prime}]] = 0,$$

$$c^{(0)}\, [[m \left\{ \frac{u^{(0)}}{q} + \frac{8\, W^2 s^2\, u^{(0)}}{q^2} + \frac{8\, W^2 s^2\, u^{(0)}}{q} + \right.$$
$$\left. \frac{4\, i\, W\, s\, u^{(0)\prime}}{q^2} + \frac{u^{(0)\prime\prime}}{q} \right\} ]] - i\, [[m\, \hat{S}_{xx}\,]]\, u^{(0)} = 0, \tag{3.5}$$

$$[[m\ \Big\{[\frac{2\,i\,W\,s}{q^2}-\frac{4\,i\,W\,s}{q}+\frac{16\,i\,W^3\,s^3}{q^2}]\,u^{(0)}+$$
$$[\frac{3}{q}-\frac{8\,W^2\,s^2}{q^2}+\frac{8\,W^2\,s^2}{q}]\,u^{(0)\prime}+$$
$$[-\frac{2\,i\,W\,s}{q^2}+\frac{4\,i\,W\,s}{q}]\,u^{(0)\prime\prime}-\frac{u^{(0)\prime\prime\prime}}{q}\ \Big\}]]=0,$$

where $q=1-i\,W\,c^{(0)}$. When $|\,y\,|\to\infty$, the solution is bounded.

The solution to (3.3)–(3.5) is very similar to that obtained by Y. Renardy [1988] for plane Couette flow. The stress components can be solved in terms of the velocity component $u^{(0)}$. The eigenfunction $u^{(0)}(y)$ is of the form

$$u^{(0)}(y)=\left|\begin{array}{ll}A_1\,(y-G_1)\exp(y)+A_2\exp(\beta_1\,y), & -\infty<y\le 0\ \text{(core)},\\ B_1\,(y-G_2)\exp(-\,y)+B_2\exp(\beta_2\,y), & 0\le y<\infty\ \text{(annulus)},\end{array}\right. \tag{3.6}$$

where

$$G_1=-\frac{c^{(0)}}{2\,s_1}\,,$$
$$G_2=-\frac{c^{(0)}}{2\,s_2}\,, \tag{3.7}$$

$$\beta_1=\sqrt{1+4\,W_1^{\,2}\,s_1^{\,2}}+i\,2\,W_1\,s_1=\gamma_1+i\,2\,W_1\,s_1,$$

$$\beta_2=-\sqrt{1+4\,W_2^{\,2}\,s_2^{\,2}}+i\,2\,W_2\,s_2=-\,\gamma_2+i\,2\,W_2\,s_2\,.$$

From the interfacial conditions (3.5), we obtain the following system of algebraic equations:

$$\mathbf{K}\,\mathbf{x}=0,$$

where $\mathbf{x}=[\,A_1,\ A_2,\ B_1,\ B_2\,]^{Tr}$ and the components of the 4∞4 matrix **K** are given in the appendix.

The eigenvalue $c^{(0)}$ is determined by

$$\mathrm{Det}\{\mathbf{K}\}=0\,,$$

which yields a polynomial of fifth degree in $c^{(0)}$ :

$$D_0 + D_1 c^{(0)} + D_2 c^{(0)^2} + D_3 c^{(0)^3} + D_4 c^{(0)^4} + D_5 c^{(0)^5} = 0. \tag{3.8}$$

The eigenvalues $c^{(0)}$ obtained from solving (3.8) contains the interfacial mode and four bulk modes. The only possible unstable mode is the interfacial mode (see Y. Renardy [1988] for detailed discussions). Equation (3.8) is solved using the method developed by Y. Renardy [1988].

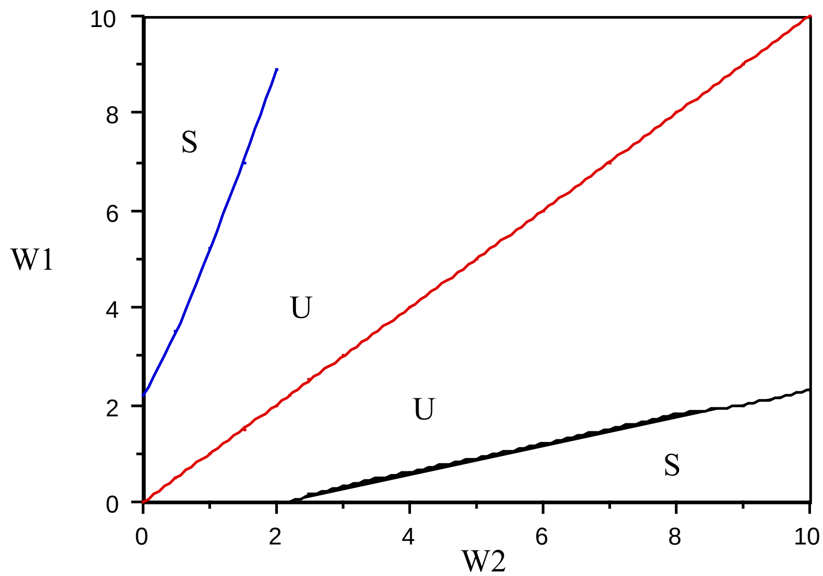


Figure 1. Neutral curves for a=1.05, $m_2$=1.

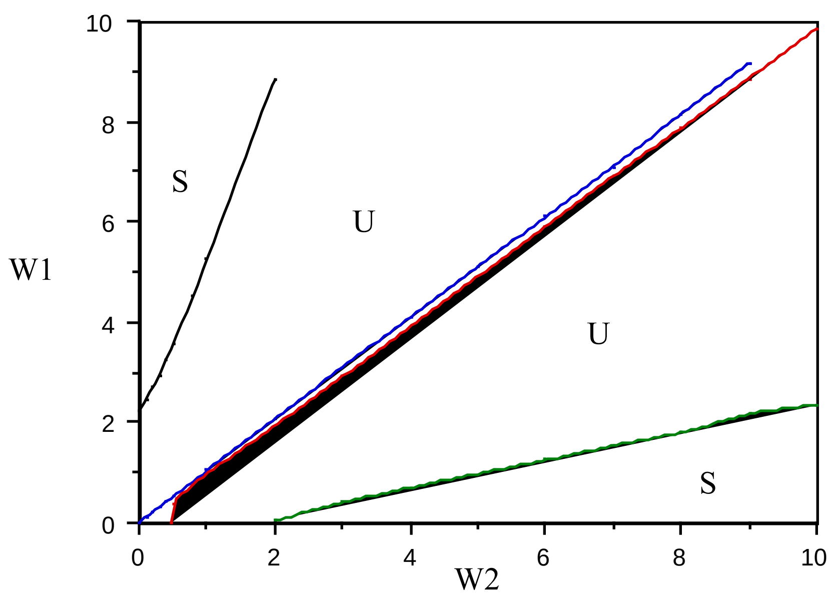


Figure 2. Neutral curves for a=1.05, $m_2$=0.9.

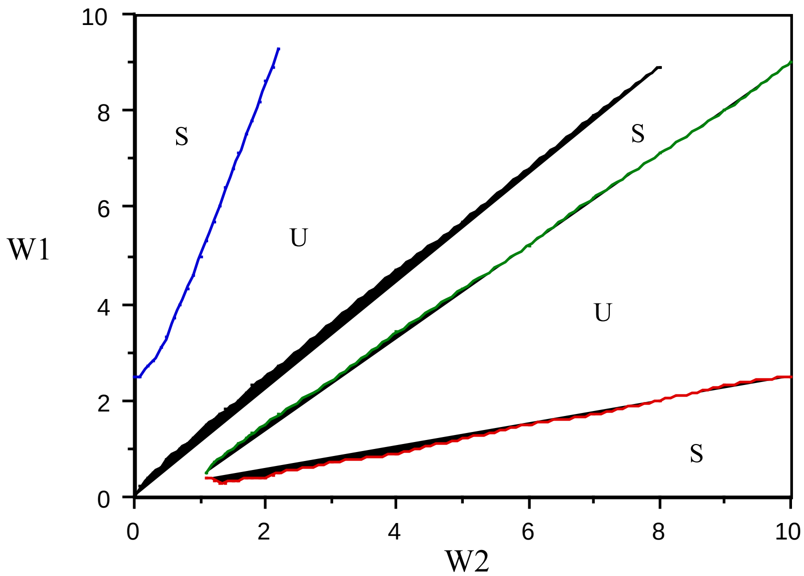


Figure 3. Neutral curves for a=1.05, $m_2$=0.5.

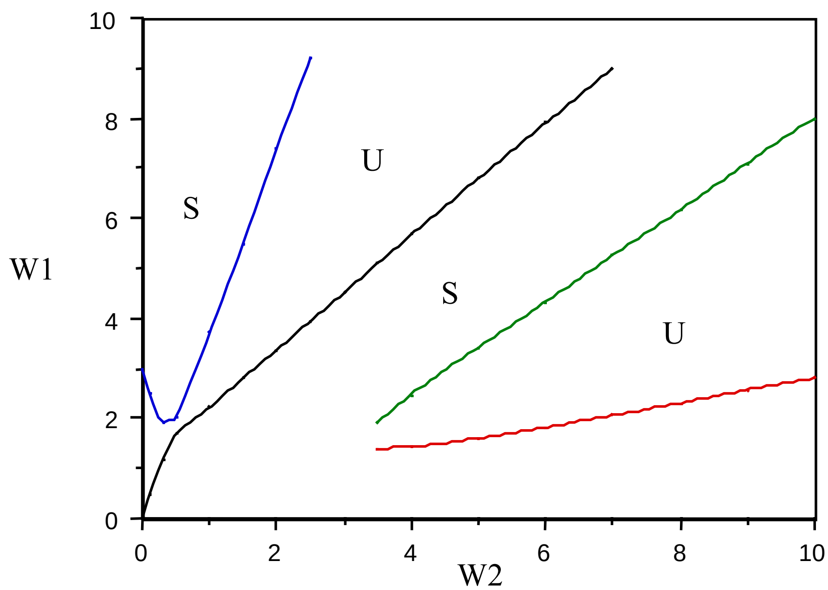


Figure 4. Neutral curves for a=1.05, $m_2$=0.2.

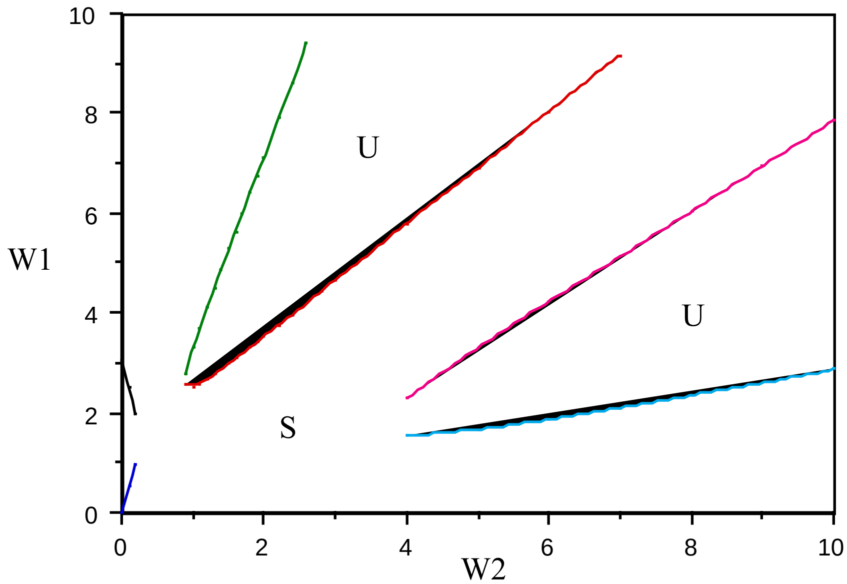


Figure 5. Neutral curves for a=1.05, $m_2$=0.18.

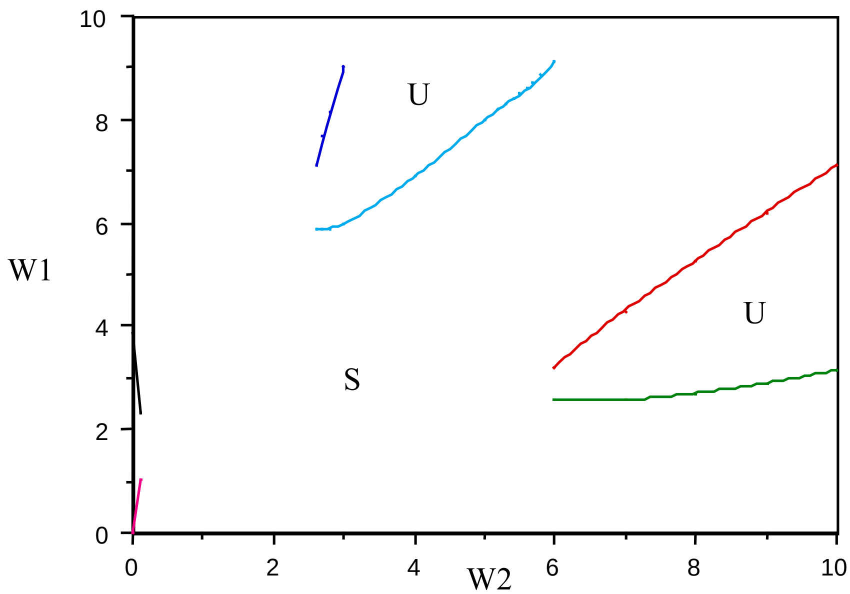


Figure 6. Neutral curves for a=1.05, $m_2$=0.1. Short wave instability of coextrusion with a small lubricating layer. In the case of interest, the core is more elastic, $W_1>W_2$. In this figure we have specified that the fluid in the core is ten times more viscous than the fluid in the slip layer and that the mean thickness a-1 of the slip layer is 0.05. The slip layer can be stable to short waves when the Weissenberg number is small and unstable when it is large.

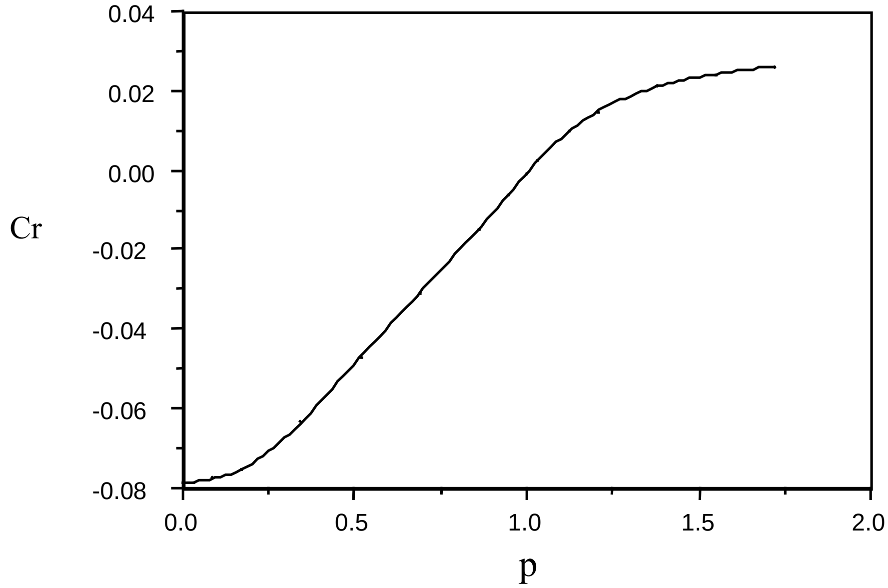


Figure 7. Phase speed $c_r$ vs $p = W_1/W_2$ for $a = 1.05$, $m_2 = 0.1$ and $W_2 = 5.8$. When p is increased past unity, the phase speeds change from negative to positive values.

**4. Short wave instability due to elasticity stratification**

The linear stability of the basic core-annular flow is determined by the sign of the imaginary part of $c^{(0)}$. The flow is unstable if $\mathrm{Im}(c^{(0)})>0$, otherwise it is linearly stable. It is important to note that any instability arising at this order must be caused by elastic effects in the fluids, since instability due to the viscosity difference appears at order $1/\alpha^3$ (see Hooper & Boyd [1983, 1987], Y. Renardy [1988], Chen [1991c]).

Figure 1 shows the neutral stability curves on the $W_2$–$W_1$ plane for the case of matched viscosities, $m_2=1$, and a=1.05. There are three branches separating stable and unstable regions which are marked by "S", "U" respectively. When the elasticities of the two fluids are also matched, $W_1=W_2$, the basic flow is neutrally stable. The neutral curve for the case of matched viscosities $m_2=1$ is symmetric about the line $W_1=W_2$. The flow is unstable when $|W_1 - W_2|>0$ is small but may be stable when $|W_1 - W_2|$ is large. It is unstable when the point $(W_2, W_1)$ falls in one of the two loops on the left or right of the line $W_1=W_2$. When the viscosities are matched, $m_2=1$, the instability is due solely to the difference in the Weissenberg number, a purely elastic effect which was discussed first by Y. Renardy [1988] and first for core-annular flow by Chen [1991a].

Neutral curves for a=1.05 and different $m_2$ are shown in Figures 2 ($m_2=0.9$), 3 ($m_2=0.5$), 4($m_2=0.2$), 5($m_2=0.18$) and 6 ($m_2=0.1$). Figure 2, 3, 4, and m's between are topologically similar. Figures 5 and 6 and for the m's between are topologically similar. In Figure 2, we have plotted neutral curves for $m_2=0.9$ and a=1.05. When the viscosity difference is perturbed away from zero, the symmetry of the neutral curves about the line $W_1=W_2$ is broken. The central branch $W_1=W_2$ for $m_2=1$ splits into two branches, between which is a new stable region. The branch emanating from the origin $(W_2, W_1)=(0, 0)$ is convex relative to the positive $W_1$ axis. There are two branches emanating from the $W_2$ axis. As the viscosity of the fluid in the annulus is further decreased, these two branches move toward each other and eventually coalesce to form a looped branch. This is evident in Figure 3, for $m_2$=0.5, a=1.05. For even smaller viscosities ratios, the loop branch moves away from the origin and the branch emanating from the origin bulges out further towards large $We_1$ (see Figures 4, 5, 6).

We also studied the variation of the stability limits with a for $1<a\leq 1.2$ and $m_2$=0.5 and found that the neutral curves are like those shown in Figure 3. The effect of increasing a is to shift the two side branches away from the origin; the intercept of the leftmost branch with the $W_2$=0 axis moves from about $W_1$=2.5 at a=1.05 to $W_1$=4 at a=1.2. A similar retreat from the origin is exhibited by the nose of the unstable loop on the right as a is increased.

Asymptotic analysis for $(W_1, W_2)\rightarrow(0, 0)$ shows that

$$c^{(0)}\rightarrow i\,\frac{4\,(1-m_2)\,m_2\,s_2^{\,2}}{(1+m_2)^2}\,(m_2\,W_1-W_2)\ . \qquad (4.1)$$

When $|\,m_2\,W_1-W_2\,|$ is small the flow is unstable if

$$0<m_2\,W_1-W_2=\frac{\hat{W}(0)\,\eta_2}{R_1}\,\frac{G_2-G_1}{G_1G_2}\ , \qquad (4.2)$$

where $G=\dfrac{\eta}{\lambda}$ is the shear modulus. Then the flow is unstable against small Weissenberg numbers when the shear modulus of the core fluid is smaller. A more elastic core with $G_1>G_2$, say, with polymer free liquid on the wall, is stable to short waves when the Weissenberg numbers are small.

Another interesting feature of the instability induced by elasticity stratification is that the phase speed changes sign from positive to negative as the value of the core Weissenberg number $W_1$ changes from $W_1>W_2$ to $W_1<W_2$ . An example is shown in Figure 7. Positive wave speed means disturbances propagate along the basic flow direction. Thus, short elastic waves will propagate in the basic extrusion flow direction when the core fluid is more elastic than the annulus fluid and in the direction opposite to the basic flow when the core is less elastic.

**5. Interfacial tension**

Interfacial tension will stabilize the shortest waves. But if the surface tension parameter J is small we may still see a short wave instability. If we include only the leading order effect of surface tension, then the asymptotic expression for c as $\alpha\to\infty$ is given by

$$c = \hat{W}(1) - i\,\frac{1}{2\,(1+m_2)}\,\frac{J}{a\boldsymbol{R}_1} + \frac{c^{(0)}}{\alpha} + O\left(\frac{1}{\alpha^2}\right),$$

where $J = \dfrac{TR_2}{\rho_1 \nu_1{}^2}$, $\dfrac{J}{a\boldsymbol{R}_1} = \dfrac{Ca}{We_1}$ and

$$\mathrm{Im}(c) = -\frac{1}{2\,(1+m_2)}\,\frac{J}{a\boldsymbol{R}_1} + \frac{\mathrm{Im}(c^{(0)})}{\alpha} + O\left(\frac{1}{\alpha^2}\right).$$

A cut-off wave number for neutral stability may be determined by putting Im(c)=0. Thus

$$\alpha_c = \frac{2\,(1+m_2)\,\mathrm{Im}(c^{(0)})}{\dfrac{J}{a\boldsymbol{R}_1}}$$

$$= \frac{2\,a\,(1+m_2)\,\boldsymbol{R}_1\,\mathrm{Im}(c^{(0)})}{J}.$$

In order for $\alpha_c$ to be large (to be consistent with the perturbation scheme $\alpha\to\infty$), we require $J \ll 1$. There are many systems of immiscible polymeric liquids with small interfacial tension. Indeed, if the core fluid is very viscous and T is not large, J will be small.

It is much easier to stabilize the short wave instability in the purely viscous case (Hooper & Boyd [1983, 1987]; Y. Renardy [1988]; Chen [1991c]). In this case the growth rates with interfacial tension neglected are not finite but tend to zero like $\alpha^{-2}$ for large $\alpha$, while the contribution from interfacial tension increases like $\alpha$ J. In other words, to get a short wave instability with a cut-off in the viscous case we must have J so small that $\alpha^3$ J=$O(1)$ for large $\alpha$. In the elastic case $\alpha$J=$O(1)$ suffices.

The results achieved in this and the last section on the stability of coextrusion are very similar to the results for plane Couette flow of two viscoelastic liquids which were obtained by Y. Renardy [1988]. In fact, in the short wave limit, to the lowest order, the curvature effect of the cylindrical geometry does not appear, and the only difference between coextrusion flow and plane Couette flow is the definition of the local shear rate at the interface. This is not true for Newtonian fluids since the short wave instability is caused by viscosity stratification and appears at order $1/\alpha^3$, rather than order $1/\alpha$ as in the present case of elastic instability (Chen [1991c]).

## 6. Wet slip and extrudate sharkskin formation

Melt fracture is a problem that has puzzled the scientific community for many years. It refers to an instability observed in extrusion flow of polymeric liquid. Below a certain critical throughput, the surface of the extrudate is smooth. At a first critical stress, small amplitude short wavelength disturbances called sharkskin appear at the extrudate surface. At a second critical stress, the extrudate surface manifests alternatively relatively smooth and sharkskin regions; this is commonly called stick-slip or spurt flow. As the stress is increased further, the extrudate surface becomes rough and wavy (see Denn [1990]). Melt fracture is a special phenomenon for elastic liquids as it does not occur in Newtonian fluids or in dilute to moderately concentrated solutions. Inertia is not relevant since melt fracture is observed at low Reynolds numbers, even as low as $10^{-15}$(Tordella [1958]). Numerous attempts have been made to explain this instability with very limited success (see Petrie & Denn [1976]; Denn [1990]).

Cohen & Metzner[1985] distinguish between "apparent slip" and “true slip.” Perhaps “true slip” is a dry slip, with one surface sliding along another without benefit of a wet lubricant. Using a slip boundary condition replacing the slip layer simplifies the analysis tremendously. However, we have seen already that the results obtained from this simplification are not satisfactory. In “apparent slip,” the polymer-deficient layer of wet solvent which develops at the wall could be and has been modeled as a lubricating layer of pure solvent. For this problem we could imagine a core fluid in a core-annular flow, and it is natural to ask whether or not such a flow is stable. Since there is perhaps only a polymer-depleted layer, rather then two spatiallysegregated solutions, we are justified in thinking that surface tension effects are negligible. We are therefore led to analysis for short waves of the type constructed in the last two sections. We want to know if after a lubrication layer has formed, can there be a short wave instability induced by the elasticity of the core? Any kind of instability induced by elasticity would be interesting, but the short wave one would be the right place to look for sharkskin.

In fact, it is probable that sharkskin can never develop in the rather mobile solutions studied by Cohen & Metzner [1985]. Sharkskin is a phenomenon which is associated with melts, amorphous polymers, and concentrated solutions in good solvent. In one current line of thought, sharkskin arises in melts as intermittent adhesive failure, sticking and slipping (Denn [1990]). Many persons think that the melt must be prestressed at inlet and in the die and that the slipping and sticking occurs locally at the die exit (see Moynihan, Baird & Ramanathan [1990]).

For the purposes of our discussion, we shall assume that some form of slip occurs in the melt when the stress reaches a certain critical value at the wall. We want to see how far we can go with the argument that we are getting a sort of wet slip, with a segregation or fractionation of molecules on the wall, large molecules inside. This kind of segregation occurs in additive-containing mixtures, say of PVC, where the additives migrate towards the wall forming a lubricating layer between the polymer and the wall. The PVC slips along this lubricating film (Funatsu *et al* [1984], Knappe *et al* [1984]). We should inquire whether such a lubricating layer is stable and what might be the nature of any instability. We are pursuing the thought that the mechanisms for creating slip and the stability of the slip or lubrication layer may be only weakly related or unrelated. For example, the high molecular weight part of a polydisperse polymer could be pulled off the wall at a critical stress, leaving the small molecules behind. This kind of arrangement could then be stable or unstable, depending on conditions.

The idea just discussed can be expanded. First we note that a kind of segregation or fractionation which leaves small molecules on the wall is an old idea which has been advanced by polymer chemists from time to time for thirty years. The idea seems to have been put forward first by Busse [1964] who says that

> The first conclusion, which is at least of some theoretical interest, is that a capillary viscometer should tend to fractionate polymer molecules with respect to molecular weight along the radius of the capillary. Near the wall, molecules of high molecular weight acquire relatively large amounts of free energy of elastic deformation, while very small molecules do not. Hence, there is a thermodynamic force that tends to increase the concentration of very small molecules at the wall, and of the larger molecules nearer the axis.
>
> No measurements of such separations have been reported, to the author's knowledge, but this factor might play a part in the action of die lubricants. It may also cause some of the change in apparent viscosity with the ratio of the capillary length to diameter.

Schreiber & Storey [1965] and Schreiber, Storey & Bagley [1966] have given indirect experimental support for Busse's idea, but more needs to be done. It seems not to be known whether additives can promote fractionation, but Moynihan *et al* [1990] have suggested that 3M's Dynamar additive, which is known to promote slip in LLDPE, forms an LLDPE/fluoroelastomer blend at the surface of the melt.

Additives may enhance or suppress slip. De Smedt & Nam [1987] studied fluoroelastomer additives in the extrusion flow of PE through capillaries of various dimensions. The effect of the additive is to reduce the apparent viscosity of the PE. Spectroscopic analysis indicated that the additive was concentrated at the free surface of the extrudate. This additive suppresses the surface defects which would appear after extrusion in polymer-free PE.

However, these results contradict some of those obtained by Ramamurthy [1986], who used a fluoroelastomer which suppressed slip in a flat die, but not a round one. He says that suppressing slip eliminates surface defects in the melt, by eliminating slip. By using two different fluoroelastomers, both the suppression and enhancement of slip was achieved in the experiments of Hatzikiriankos & Dealy [1991]. They studied the slip of a polydisperse polyethylene in a sliding plate rheometer and they reduced slip by coating the steel plates with “DFL” (dry film lube) and increased the slip by coating it with Dynamar 9613.

It appears not to be known whether or not the various forms of slip are wet or dry. The evidence for wet slip is clear in certain solutions, like those studied by Cohen & Metzner [1985] or in additive-bearing mixtures like the PVC mentioned earlier.

Slip layers which form under adhesive fracture of the high molecular weight molecules from a wall are presently not well understood. We have been looking at the consequences of modeling such layers as a “wet slip” layer with an average thickness, density, viscosity, relaxation time and shear modulus. Using this concept, we must allow that the results of adhesive fracture may lead to a stable or unstable layer, according to conditions. Our analysis was confined to short waves, so that an unstable layer could conceivably lead to sharkskin. Of course, we have only the vaguest idea of how to characterize these layers, but they obviously should be much less viscous and much less elastic than the polymer core.

To see how all this works out for a hypothetical example, consider the case in which the capillary radius is slightly larger than the nominal radius $R_1$ separating high and low molecular weight polymers, say, $R_2/R_1=1.05$. The interfacial tension is zero because our polymers of different molecular weights can mix. The viscosity of the polymer core is, say, ten times greater than the average viscosity of the low molecular weight annulus and since the core is more elastic, $\lambda_2/\lambda_1<1$; say, $10^{-1}<\lambda_2/\lambda_1=W_2/W_1<1$. These assumptions put us in the frame of Figure 6. Inspection of this figure shows that the stability properties of the lubricating layer depend on the slope of $\lambda_2/\lambda_1$, a ray from the origin, and that the layer is stable against short waves when $W_1$ is small, for polymers with low molecular weights. For high molecular weights, $W_1$ is large and we can enter the unstable loop along a ray $10^{-1}<W_2/W_1<1$ at some critical Weissenberg number $W_1$, as shown in Figure 6. We get stability in the lubrication layer when the molecular weight of polymers is low and instability to short waves, sharkskin, when it is high. At least this doesn’t disagree with experiments, but our comparison is much too casual to be anything more than suggestive.

Even in the case of cohesive fracture of the type leading to spurt it is hard to imagine slipping without lubrication of some sort. We can expect a migration of low-viscosity constituents into regions of high shear. These high-shear regions may take form as a wet layer of small thickness or perhaps as a region deficient in high molecular weights defined by large gradients of molecular weight. The slip in such layers is apparent; there is no slip surface, rather there are large gradients across narrow layers which are perceived as slip. In any case, the study of the stability of such layers is of interest.

## Conclusions

We studied the stability of coextrusion of polymers through capillaries modeling the polymers as Maxwell models with single times of relaxation. The analysis of stability to short waves leads to a condition for stability which depends on the Weissenberg numbers, the viscosities, the layer thickness, and interfacial tension. The shortest waves are stabilized by interfacial tension. If the tension is small enough, short wave instabilities with rather large cut-off wave numbers are possible.

We conjectured that adhesive fracture in capillaries and possibly cohesive fractures could give rise to wet slip layers by leaching the polymers from the solution at the wall or by fractionation in amorphous polymers. The properties of such lubricating layers are not well understood, but they may be similar to the better understood lubricating layers in our two-fluid problem. The conditions of existence of such layers and for their stability may be unrelated. We must allow that the results of adhesive fracture may lead to a stable or unstable layer, according to conditions.

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## Appendix

The components of the matrix **K** are:

$$K_{11} = \frac{c^{(0)}}{2\, m_2\, s_2} ,$$

$$K_{12} = 1,$$

$$K_{13} = -\frac{c^{(0)}}{2\, s_2} ,$$

$$K_{14} = -1,$$

$$K_{21} = \frac{c^{(0)}}{m_2} + \frac{c^{(0)2}}{2\, m_2\, s_2} ,$$

$$K_{22} = 2\,(m_2 - 1)\, s_2 + (\,\gamma_1 + 2\, i\, m_2\, W_1\, s_2\,)\, c^{(0)} ,$$

$$K_{23} = -\, c^{(0)} + \frac{c^{(0)2}}{2\, s_2} ,$$

$$K_{24} = (\,\gamma_2 - 2\, i\, W_2\, s_2\,)\, c^{(0)} ,$$

$$K_{31} = 4\, i\, W_2\, s_2 + \frac{c^{(0)}}{m_2\, s_2\, q_1} + \frac{2}{q_1^{\,2}} ,$$

$$K_{32} = \frac{8\, i\, m_2\, s_2^{\,2}(W_2 - m_2\, W_1)}{c^{(0)}} + \frac{2 + 8\, m_2^{\,2}\, W_1^{\,2}\, s_2^{\,2} + 4\, i\, m_2\, \gamma_1\, W_1\, s_2}{q_1} + \frac{4\, i\, m_2\, \gamma_1\, W_1\, s_2}{q_1^{\,2}} ,$$

$$K_{33} = -\,4\, i\, m_2\, W_2\, s_2 - \frac{c^{(0)}\, m_2}{s_2\, q_2} + \frac{2\, m_2}{q_2^{\,2}} ,$$

$$K_{34} = \frac{-2\,m_2 - 8\,m_2\,W_2^{\,2}\,s_2^{\,2} + 4\,i\,m_2\,\gamma_2\,W_2\,s_2}{q_2} + \frac{4\,i\,m_2\,\gamma_2\,W_2\,s_2}{q_2^{\,2}}\ ,$$

$$K_{41} = 4\,i\,m_2\,W_1\,s_2 + \frac{c^{(0)}}{m_2\,s_2\,q_1}$$

$$K_{42} = \frac{2\,\gamma_1}{q_1}\ ,$$

$$K_{43} = 4\,i\,m_2\,W_2\,s_2 + \frac{c^{(0)}\,m_2}{s_2\,q_2}\ ,$$

$$K_{44} = \frac{2\,m_2\,\gamma_2}{q_2}\ .$$

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