Criterion for Extensional Necking Instability in Polymeric Fluids

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(Received 10 June 2011; published 12 December 2011)

We study the linear instability with respect to necking of a filament of polymeric fluid undergoing uniaxial extension. Contrary to the widely discussed Considère criterion, we find the onset of instability to relate closely to the onset of downward curvature in the time (and so strain) evolution of the zz component of the molecular strain, for extension along the z axis. In establishing this result numerically across five of the most widely used models of polymer rheology, and by analytical calculation, we argue it to apply generically. Particularly emphasized is the importance of polymer chain stretching in partially mitigating necking. We comment finally on the relationship between necking and the shape of the underlying steady state constitutive curve for homogeneous extension.

DOI: 10.1103/PhysRevLett.107.258301 PACS numbers: 83.60.Wc, 83.10.−y, 83.50.Jf, 83.80.−k

Understanding the rheology (flow properties) of polymeric fluids is central to their processing and performance. Many commercially important flows are dominated by extensional components: fiber spinning, film blowing, extrusion, and ink jet printing provide good examples. In fluid dynamical terms, extensional flows cause material elements to separate exponentially quickly and so subject the underlying macromolecules to extreme stretching and reorientation. They are thus highly sensitive to underlying molecular details: linear vs branched polymer chains, for example. Indeed many nonlinear flow features arise only in extension, which thus provides a crucial benchmark for theories of polymer rheology.

In many polymeric fluids, a state of uniform flow becomes unstable when the flow rate exceeds the rate $1/\tau$ on which the underlying molecular structure relaxes [1]. In flows dominated by shear, for example, the phenomenon of shear banding [2] arises widely for shear rates $\dot{\gamma}\tau > O(1)$. In a linear stability analysis, the criterion for onset of a shear banding instability is (usually) that the shear stress is a decreasing function of shear rate.

In extensional rheology [3,4] a common protocol is that of filament stretching [5], in which a cylindrical sample of fluid is drawn out in length. Though the aim is to achieve uniform extensional flow for benchmarking against theory, complications often arise. Particularly serious is the widespread observation of necking instabilities (Fig. 1), in both experiment [6–13] and simulation [14–18]. These lead to heterogeneous deformation and even failure at only modest strains: any small indentations in cross-sectional area become ever more pronounced until the sample breaks. This hinders attempts to characterize these fluids scientifically, and process them commercially.

Although a long-standing problem [19–33], necking remains poorly understood. Crucially lacking is any reliable criterion for its onset, of the same stature as that given above for shear banding. Popularly discussed is the Considère criterion [34], which predicts necking if the tensile force decreases with extensional strain: $dF/d\epsilon < 0$. But taking F to depend only on ϵ in this way assumes the flow purely elastic: it cannot account for the dependence of necking on the strain rate $\dot{\epsilon}$ in these viscoelastic fluids [35], nor predict the rate at which necking sets in.

Here we show that the Considère criterion in fact does not apply in most regimes of polymeric flow. By a linear stability analysis, we demonstrate the onset of necking instead to relate closely to that of downward curvature in the time (or equivalently strain) evolution of the zz component of the molecular conformation tensor, for stretching at constant rate $\dot{\epsilon}$ along the z axis. In most regimes, this further corresponds to downward curvature in the evolution of the tensile stress. In establishing this result numerically across five of the most widely used models of polymer rheology, and by analytical calculation, we argue it to apply generically. The other central contribution of this Letter is to demonstrate the crucial role played by polymeric chain stretching in partly mitigating necking.

We consider a polymeric fluid in inertialess flow, obeying the force balance condition $0 = \boldsymbol{\nabla} \cdot \boldsymbol{T} = \boldsymbol{\nabla} \cdot (\boldsymbol{\Sigma} + 2\eta \boldsymbol{D} - P\boldsymbol{I})$. The total stress $\boldsymbol{T}$ has a viscoelastic contribution $\boldsymbol{\Sigma}$ from the polymer, a Newtonian contribution of viscosity η from the solvent, and an isotropic pressure field P set by mass balance $\boldsymbol{\nabla} \cdot \boldsymbol{v} = 0$ for

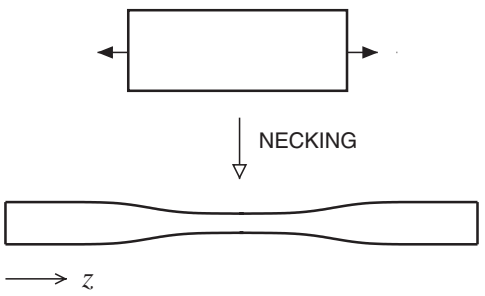

FIG. 1. Cartoon of necking.

incompressible flow. Here $\boldsymbol{v}$ is the velocity field and $\boldsymbol{D}$ the symmetric part of the velocity gradient tensor $\boldsymbol{\kappa}_{\alpha\beta} \equiv \partial_\alpha v_\beta$. We assume the polymeric stress $\boldsymbol{\Sigma} = G_0[k(\boldsymbol{W})\boldsymbol{W} - \boldsymbol{I}]$ with G_0 a modulus and $\boldsymbol{W}$ a dimensionless tensor characterizing the conformation of the chainlike polymer molecules. (In the simplest cartoon these are taken as dumbbells with span $\vec{R}$ and $\boldsymbol{W} \propto \langle \vec{R}\,\vec{R} \rangle$.) This has generalized dynamics

$$(\partial_t + \boldsymbol{v} \cdot \boldsymbol{\nabla})\boldsymbol{W} = \boldsymbol{W} \cdot \boldsymbol{\kappa} + \boldsymbol{\kappa}^T \cdot \boldsymbol{W} - \frac{1}{\tau}\boldsymbol{R}(\boldsymbol{W}), \quad (1)$$

representing a competition between driving out of equilibrium by flow (terms in $\boldsymbol{\kappa}$) and relaxation back to equilibrium $\boldsymbol{W} = \boldsymbol{I}$ on a time scale τ.

We perform calculations for five concrete functional choices for $k(\boldsymbol{W})$ and $\boldsymbol{R}(\boldsymbol{W})$, each corresponding to a particular model used widely in the literature [36]. The Oldroyd B model has $k = 1$, $\boldsymbol{R} = \boldsymbol{W} - \boldsymbol{I}$. The Giesekus model differs from this in having $\boldsymbol{R} = (\boldsymbol{W} - \boldsymbol{I}) + \alpha(\boldsymbol{W} - \boldsymbol{I})^2$ with $0 \le \alpha \le 1$; the Fene-P (in which we assume small δ) in having $\boldsymbol{R} = k(\boldsymbol{W})\boldsymbol{W} - \boldsymbol{I}$ with $k = 1/(1 - \delta T)$ and trace $T = W_{xx} + W_{yy} + W_{zz}$; the Rolie-Poly [37] $\boldsymbol{R} = \boldsymbol{W} - \boldsymbol{I} + 2(1 - \sqrt{3/T}) \times [\boldsymbol{W} + \beta(T/3)^{\delta_{\mathrm{RP}}}(\boldsymbol{W} - \boldsymbol{I})]\tau/[\tau_{\mathrm{R}}(1 - fT/3)]$. Finite τ_{R} in the Rolie-Poly model allows stretching of the underlying polymer chains by the flow field. Indeed for $\dot{\epsilon}\tau_{\mathrm{R}} > 1$ infinite stretch can develop if $f = 0$; $f > 0$ restores finite stretch; $\tau_{\mathrm{R}} \to 0$ disallows it entirely.

We use units in which $\tau = 1$, $G_0 = 1$. This leaves as parameters the Newtonian viscosity η, which we take to zero, and α (Giesekus), δ (Fene-P), β, τ_{R}, δ_{RP}, f (Rolie-Poly) in which we use $\beta = 0.0$, $\delta_{\mathrm{RP}} = -1/2$ following [37].

We consider a long slender [38] tube of fluid subject to uniaxial extension along the z axis. At any time t it has cross sectional area profile $A(z, t)$ and area-averaged fluid velocity $V(z, t)$ in the z direction, with extension rate $\dot{\epsilon}(z, t) = \partial_z V(z, t)$. The mass balance condition is then $\partial_t A(z, t) = -\partial_z(AV)$, and force balance $F(t) = A[k(T)(Z - X) + 3\eta\dot{\epsilon}] \equiv A\sigma_{\mathrm{E}}$ (neglecting surface tension). This defines the tensile stress $\sigma_{\mathrm{E}}(z, t)$ given force $F(t)$, which is uniform along the filament. $Z = W_{zz}$ and $X = \frac{1}{2}W_{xx} + \frac{1}{2}W_{yy}$ (so $Z + 2X = T$) obey

$$\begin{aligned} \partial_t Z(z, t) + V\partial_z Z &= 2\dot{\epsilon}Z - R(Z, T)/\tau, \\ \partial_t X(z, t) + V\partial_z X &= -\dot{\epsilon}X - R(X, T)/\tau, \end{aligned} \quad (2)$$

with $R(Z, T) = Z - 1$ (Oldroyd B); $R(Z, T) = Z - 1 + \alpha(Z - 1)^2$ (Giesekus); $R(Z, T) = k(T)Z - 1$ (Fene-P); $R(Z, T) = Z - 1 + 2(1 - \sqrt{\frac{3}{T}})[Z + \beta(\frac{T}{3})^{\delta_{\mathrm{RP}}}(Z - 1)]/\tau_{\mathrm{R}}(1 - \frac{fT}{3})$ (Rolie-Poly).

Given a time-dependent separation of the sample ends that imposes a global strain $\bar{\epsilon}(t)$ averaged along the filament, we transform to the affinely coextending, cothinning frame by defining $u = ze^{-\bar{\epsilon}(t)}$, $v(u, t) = V(z, t)e^{-\bar{\epsilon}(t)}$ and $a(u, t) = A(z, t)e^{\bar{\epsilon}(t)}$. We then have mass balance, force balance, and viscoelastic dynamics:

$$\partial_t a = -\partial_u[(v - \dot{\bar{\epsilon}}u)a], \quad (3)$$

$$0 = \partial_u[a(k(T)(Z - X) + 3\eta\dot{\epsilon})] = \partial_u[a\sigma_{\mathrm{E}}], \quad (4)$$

$$\begin{aligned} \partial_t Z &= -(v - \dot{\bar{\epsilon}}u)\partial_u Z + 2\dot{\epsilon}Z - R(Z, T)/\tau, \\ \partial_t X &= -(v - \dot{\bar{\epsilon}}u)\partial_u X - \dot{\epsilon}X - R(X, T)/\tau. \end{aligned} \quad (5)$$

For a constant global extension rate $\dot{\bar{\epsilon}}$ commenced at time $t = 0$, a homogeneous "base state" in which uniaxial extension is (artificially) maintained uniformly along the filament, regardless of whether it is in practice unstable to heterogeneous necking, is prescribed by $\dot{\epsilon}(u, t) = \dot{\bar{\epsilon}}$, $v(u, t) = u\dot{\bar{\epsilon}}$, $a(u, t) = a_0 = 1$, and the homogeneous solutions $Z(t)$, $X(t)$ of Eqs. (5) given $Z(0) = X(0) = 1$.

In any regime of finite extensional viscosity this base state attains as $t \to \infty$ a steady state in the cothinning, coextending frame. (In the laboratory frame the sample exponentially extends and thins.) This is described by a homogeneous constitutive curve $\sigma_{\mathrm{E}}(\dot{\epsilon})$ of tensile stress vs strain rate, Fig. 2(a). (For this uniform base state we use symbols $\dot{\bar{\epsilon}}$ and $\dot{\epsilon}$ interchangeably.) The Oldroyd B model has divergent viscosity for $\dot{\epsilon} \to 1/2$. This divergence is avoided (narrowly, for small α, δ) in Giesekus and Fene-P (which reduce to Oldroyd B for $\alpha = 0$, $\delta = 0$). The stretch Rolie-Poly model with $f = 0$ likewise has divergent viscosity for $\dot{\epsilon}\tau_{\mathrm{R}} \to 1$, avoided narrowly for small $f > 0$ and entirely in the nonstretch model $\tau_{\mathrm{R}} = 0$.

The time dependence of the tensile stress σ_{E} in this homogeneous base state as it evolves towards the steady state just described is shown in Fig. 2(b) for the stretch

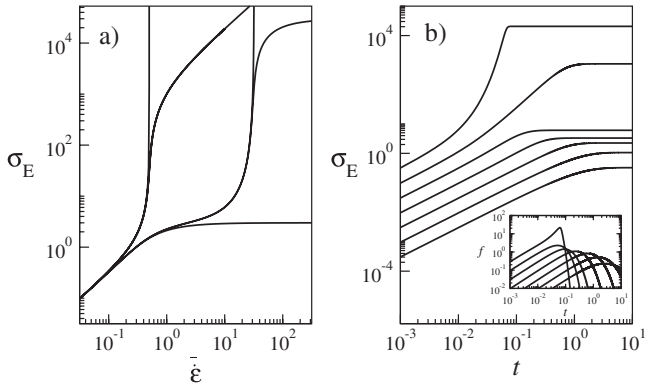

FIG. 2. (a) Homogeneous constitutive curves for steady state uniaxial extension. Clockwise: Oldroyd B; Giesekus ($\alpha = 0.001$); fene-P ($\delta = 0.001$); stretch Rolie-Poly ($\tau_{\mathrm{R}} = 0.0316$) with zero extensibility; the same with finite extensibility ($f = 10^{-4}$); nonstretch Rolie-Poly. (Curves for Giesekus and fene-P indistinguishable.) (b) Tensile stress σ_{E} vs time in startup of uniaxial extension in the stretch Rolie-Poly model with $\tau_{\mathrm{R}} = 0.0316$, $f = 10^{-4}$ for $\dot{\epsilon} = 10^n$, $n = -1, -1/2, 0, 1/2, 1, 3/2, 2$ (curves upwards). Inset: corresponding tensile force.

Rolie-Poly model with finite extensibility. The corresponding force $F(t) = \sigma_E(t)A(t)$ (inset) displays a maximum due to the rise in σ_E after the inception of flow being later overcome by exponentially declining area.

A maximum in force vs time t directly also signifies a maximum vs strain $\epsilon = \dot{\epsilon}t$, given constant $\dot{\epsilon}$. For a nonlinear elastic solid, the Considère criterion decrees such a maximum to herald departure from uniform extension and onset of necking. We now examine whether this criterion also applies to polymeric fluids, as often suggested.

To do so, we analyze the linear stability of the time-dependent homogeneous uniaxially extending base state just described by linearizing in small heterogeneous fluctuations $\sum_q [\delta a_q(t), \delta\dot{\epsilon}_q(t), \delta Z_q(t), \delta X_q(t)] \exp(iqu)$ about it. To first order in the amplitude of these we have

$$\partial_t \begin{pmatrix} \delta a \\ 0 \\ \delta Z \\ \delta X \end{pmatrix}_q = \begin{pmatrix} 0 & -1 & 0 & 0 \\ \sigma_E & 3\eta & \partial_Z \sigma_E & \partial_X \sigma_E \\ 0 & 2Z & 2\bar{\dot{\epsilon}} - \frac{r}{\tau} - \frac{s}{\tau} & -\frac{2s}{\tau} \\ 0 & -X & -\frac{s}{\tau} & -\bar{\dot{\epsilon}} - \frac{r}{\tau} - \frac{2s}{\tau} \end{pmatrix} \begin{pmatrix} \delta a \\ \delta\dot{\epsilon} \\ \delta Z \\ \delta X \end{pmatrix}_q,$$

in which $r = \partial_Z R(Z, T)$, $s = \partial_T R(Z, T)$. The q independence of this matrix suggests all spatial modes will grow (or decay) equally, though we return below to discuss what shape can be expected in practice.

If the time dependence of the base state were disregarded, instability to necking would correspond to any eigenvalue of this matrix having positive real part. However, its entries Z, X, σ_E, T refer to the time-evolving base state, rendering the eigenvalues time dependent: indeed, these can start out all negative before one later goes positive. Numerics and analytics (not shown) reveal this sign change to correspond to excellent approximation to onset of downward curvature $\ddot{Z} < 0$ in the time (and so strain) evolution of the base state $Z(t)$ and also (apart from the small discrepancy seen in Fig. 3) of the tensile stress, $\ddot{\sigma}_E < 0$. This strongly suggests that necking will arise in any regime of $\ddot{Z} < 0$, $\ddot{\sigma}_E < 0$. Some caution is needed, however, because the eigenvectors also evolve. To examine rigorously the onset of necking, therefore, we explicitly integrate the linearized equations numerically. (Nonlinear effects must eventually become important, but when depends on the size of the seeding perturbation, which is not specified.) Resulting contour maps of $\delta a(t)/\delta a(0)$ are shown in Figs. 3 and 4, with the criteria $\ddot{Z} = 0$, $\ddot{\sigma}_E = 0$, $\dot{F} = 0$ for comparison.

In the Oldroyd B model, Fig. 3(a), $\delta a(t)$ grows without bound for $\bar{\dot{\epsilon}} < 1/2$, albeit slowly, predicting that necking should eventually arise in any experiment of long enough duration, even causing filament failure unless mitigated by

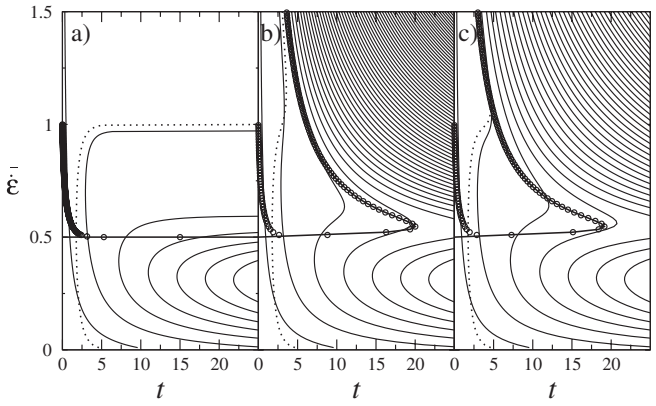

FIG. 3. Contour map for growth of fluctuations in area: $\delta a(t)/\delta a(0) = 2^n$ for $n = 1, 2, 3 \cdots$ shown by thin solid lines from left to right. Dotted lines, open circles, thick solid lines show location, respectively, of $\dot{f} = 0$, $\ddot{\sigma}_E = 0$, $\ddot{Z} = 0$. (a) Oldroyd B, (b) Giesekus ($\alpha = 0.001$), (c) Fene-P ($\delta = 0.001$).

nonlinear effects. For $\bar{\dot{\epsilon}} > 1/2$, in contrast, $\delta a(t)$ grows only weakly before quickly saturating, signifying stability against necking. This is consistent with the divergence in the underlying constitutive curve: for $\bar{\dot{\epsilon}} > 1/2$ the base state $Z(t)$ and $\sigma_E(t)$ diverge in time with ever upward curvature and the eigenvalue remains negative. (For $\bar{\dot{\epsilon}} < 1/2$, $Z(t)$ and $\sigma_E(t)$ curve down towards steady state.).

The Giesekus and fene-P models avoid this viscosity divergence: in the evolution of the base state an early time regime of Oldroyd B-like upward curvature $\ddot{\sigma}_E > 0$, $\ddot{Z} > 0$, inside the nose-shaped region in Figs. 3(b) and 3(c), later gives way to downward curvature toward a high viscosity steady state. Surprisingly δa does grow inside the nose, even though $\ddot{Z} > 0$ and all eigenvalues are negative here. This growth is however weak enough to go unnoticed,

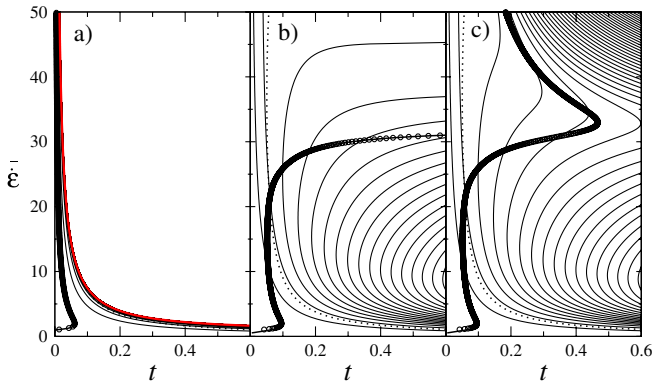

FIG. 4 (color online). Contour maps $\delta a(t)/\delta a(0) = 2^n$ for $n = 1, 2, 3 \ldots$, thin solid lines left to right. Dotted lines, open circles, thick solid lines show location of $\dot{f} = 0$, $\ddot{\sigma}_E = 0$, $\ddot{Z} = 0$. (a) Nonstretch Rolie-Poly, (b) Stretch Rolie-Poly ($\tau_R = 0.0316$, $f = 0.0$), (c) Stretch Rolie-Poly with finite extensibility, $f = 10^{-4}$. Red line in (a) shows analytical prediction for divergence in δa.

assuming the perturbation that initially seeds the instability to be small. Rapid exponential growth and observable necking set in only after the nose, where $\ddot{Z} < 0$, $\ddot{\sigma}_E < 0$ and an eigenvalue is positive.

Can these contour maps be understood analytically? Given an evolution equation $\partial_t \sigma_E = \dot{\epsilon} G(\sigma_E, T) - H(\sigma_E, T)/\tau$ for the base state's tensile stress, one can show (approximately) for Oldroyd B and Giesekus that $\delta a(t)/\delta a(0) = f(x)$ with $f = (1 + bx)^{\bar{\dot{\epsilon}}/b}$ and $x = \sigma_E/\dot{\sigma}_E$. Here $b = H/\tau\sigma_E - \bar{\dot{\epsilon}}$ varies only mildly compared to x in Giesekus, and is constant for Oldroyd B. In any regime of finite viscosity (Oldroyd B for $\bar{\dot{\epsilon}} < 1/2$ and Giesekus for all $\bar{\dot{\epsilon}}$) the argument $x \to \infty$ as $t \to \infty$ due to the downward curvature to steady state $\dot{\sigma}_E \to 0$. Combined with $f(x) \to \infty$ as $x \to \infty$, this gives $\delta a \to \infty$ as $t \to \infty$: necking fluctuations grow without bound, as seen numerically. In contrast the divergent viscosity of Oldroyd B for $\bar{\dot{\epsilon}} > 1/2$ gives $x \to$ const. as $t \to \infty$ and δa saturates.

We now use these findings in the above phenomenological (but widely used) models as pedagogical backdrop to the more realistic Rolie-Poly model, focusing particularly on the dramatic role of polymeric chain stretch.

Without chain stretch, $\tau_R = 0$, the result $\delta a(t) = \delta a(0) f(x)$ with $f = (1 + bx)^{\bar{\dot{\epsilon}}/b}$, $x = \sigma_E/\dot{\sigma}_E$ and $b = 1/\tau - \bar{\dot{\epsilon}}$ holds exactly. Finite extensional viscosity further gives $x \to \infty$ as $t \to \infty$ for all $\bar{\dot{\epsilon}}$, and so unbounded growth of δa. In fact for $\bar{\dot{\epsilon}} > 1/\tau$, f diverges at finite x giving a dramatic finite-time divergence in δa. An analytical prediction for this is shown in Fig. 4(a); the numerical contours indeed accumulate at it. Clearly, suppression of chain stretch confers spectacular necking instability.

For finite τ_R the base state shows divergent extensional viscosity for $\bar{\dot{\epsilon}} \to 1/\tau_R$, associated with the development of infinite chain stretch. This is reminiscent of Oldroyd B for $\bar{\dot{\epsilon}} \to 1/2$. Correspondingly, necking is suppressed altogether for $\bar{\dot{\epsilon}} > 1/\tau_R$ [Fig. 4(b)]. Even for $\bar{\dot{\epsilon}} < 1/\tau_R$ the finite-time divergence of Fig. 4(a) disappears. Chain stretch thus strongly mitigates necking.

Although instructive, infinite chain stretching is clearly unphysical so we now introduce a parameter f to cure it [37], analogous to δ in fene-P. With this, the regime of upward curvature in the base state, $\ddot{Z} > 0$, $\ddot{\sigma}_E > 0$, is again confined to a nose-shaped region [Fig. 4(c)]. As in Giesekus and fene-P, δa grows only slowly inside this, exponential growth sets in only beyond it, once $\ddot{Z} < 0$, $\ddot{\sigma}_E < 0$. The nose's tip at $\bar{\dot{\epsilon}}\tau_R = O(1)$ gives a maximum in onset strain vs $\bar{\dot{\epsilon}}$ that closely resembles experimental data [8,13], as collated onto a master plot in Ref. [4].

Taken together, Figs. 3 and 4 tell us how heterogeneity grows in a necking instability intrinsic to the material's rheology. To specify the process fully also requires knowledge of what initial perturbation seeds the instability. If this is small (thermal noise or minor mechanical imperfection), many contour lines will need to be crossed before necking becomes apparent, well into the regime of exponential growth in $\delta a(t)$, and so of $\ddot{Z} < 0$, $\ddot{\sigma}_E < 0$, which we thus propose as the criterion for onset in this case. This is likely in rheometers that co-thin their end plates as the sample extends. In others, much larger heterogeneous seeding will arise trivially because the sample is constrained to remain thicker at the plates. Only the first few contours might then need to be crossed and, as seen in the maps, the Considère criterion $\dot{f} < 0$ should perform tolerably. In either case we expect such end-plate effects, however small, to select out of the q—independent spectrum of (6) those modes favoring a single neck, midsample.

Finally, we ask if *any* fluid of finite extensional viscosity should neck, via a simplified approach in which (i) the base state has already attained a steady state on its constitutive curve, and (ii) $Z = Z(\dot{\epsilon})$, $X = X(\dot{\epsilon})$ are instantaneously prescribed by $\dot{\epsilon}$. (6) then reduces to $\partial_t \delta a = -\delta\dot{\epsilon}$, $0 = \sigma_E(\bar{\dot{\epsilon}})\delta a + \sigma_E'(\bar{\dot{\epsilon}})\delta\dot{\epsilon}$, and so $\partial_t \delta a = (\sigma_E/\sigma_E')\delta a$. This indeed suggests any regime of $\sigma_E'(\bar{\dot{\epsilon}}) > 0$ is unstable to necking (including any Newtonian fluid, even without surface tension). The physics is clear. Mass balance $\partial_t \delta a = -\delta\dot{\epsilon}$ makes a more strongly stretched section of filament get thinner. This must then develop larger stress σ_E to maintain uniform force along the thread. For positive σ_E' this needs even stronger stretching, giving positive feedback. This also suggests any *negatively* sloping part of a constitutive curve to be *stable* against necking. Whether any such a curve exists, and whether this distilled result still holds given realistic Z, X dynamics, is unclear.

In summary, we have given a new criterion for instability to necking of a filament of polymeric fluid in uniaxial extension of constant rate. Future work will consider the nonlinear dynamics of necking, beyond the linear regime, effects of surface tension, applied force protocols and nonuniform extension rates, and the interplay of "intrinsic" necking instability with "external" (end plate) effects.

The author thanks Gareth McKinley, Mike Cates, Ron Larson, and Helen Wilson for interesting discussions.

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