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RAPID COMMUNICATION

The *W*-criterion for the onset of shear banding in complex fluids

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Abstract A stability analysis of planar shear flow shear of a homogeneous, complex fluid predicts that shear banding instabilities can grow in fluids with a shear thinning strength above $W_c = 1$ and dampen out in fluids with $W < 1$. The shear thinning strength, $W(\dot{\gamma}) = -\partial \log\eta / \partial \log\dot{\gamma}$, arises naturally as the lead material function for the stability analysis of shear thinning fluids. The onset of shear banding is modeled as shear-thinning instability, which is attributed to anomalously strong shear thinning. Not considered here are inertial or elastic instabilities. In lack of suitable viscosity data from experiments, a Carreau powerlaw fluid and a Carreau-Yasuda powerlaw fluid serve as testbeds for the *W*-criterion. The analysis shows that the limiting high shear viscosity, η_∞, plays an important role in shear banding and that the ratio of the limiting high shear viscosity and zero shear viscosity, η_∞/η_0, has to be sufficiently small for shear banding to occur. The main purpose of this brief communication is to share this new stability criterion. Extensive testing is still needed and is planned for future study.

Keywords Shear instability · Shear banding · Shear thinning strength · Carreau-Yasuda fluid · Limiting high shear viscosity

Introduction

Complex materials have been reported to become unstable when sheared beyond some critical condition. This includes shear banding, which divides an otherwise homogeneous shear flow into macroscopic regions ("shear bands") parallel the shear planes. In spite of being at the same shear stress (no curvature considered here), the shearing in the bands occurs at different shear rates. For homogeneous fluids, shear bands consist of the same material, but exhibit different viscosities (!). The shear banding phenomenon is accepted to be real. It has been observed for a rich variety of complex fluids such as foams, surfactant solutions, gels, soft glassy materials, and concentrated particle suspensions as reported in extensive reviews and research publications (Divoux et al. 2016, Ovarlez et al. 2013, Fielding et al. 2009, Ovarlez et al. 2009, Callaghan 2008, Dhont and Briels 2008, Manneville 2008, Olmsted 2008). Some materials develop shear bands under transient conditions only (Moorcroft and Fielding 2013) or never reach steady shear conditions at all (Vasisht et al. 2016). Shear banding can be viewed as an instability phenomenon in which, under some conditions, fluctuations grow into bands while, under slightly different conditions, such fluctuations dampen out. Shear banding originates for reasons which strongly change from material to material and are not obvious in many cases.

This communication concerns the type of shear banding, which is caused by exceptionally strong shear thinning. For that purpose, we assume that steady shear conditions exist for such material and define a shear thinning strength

$$W(\dot{\gamma}) = -\frac{\partial \log\eta}{\partial \log\dot{\gamma}}. \quad (1)$$

where W is the magnitude of the gradient in the conventional log–log plot of the steady shear viscosity over the shear rate, $\eta(\dot{\gamma})$. The format of W arises naturally in the stability analysis as will be shown. To my knowledge, this is the first time that W is used as a meaningful material function.

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Since not all shear thinning liquids are able to form shear bands, at least not under all shearing conditions, a criterion should be made available that lets us distinguish fluids that shear band from those that are not, and under which shearing conditions.

Derivation of the *W*-criterion

For the stability analysis, we use a Gedankenexperiment, in which a homogeneous fluid with known $W(\dot{\gamma})$ is sheared between two planar, parallel surfaces at an average shear rate of $\dot{\gamma}_0 = U/H$ as shown in Fig. 1. The question arises about the conditions at which the shear flow might get unstable and possibly split into bands as indicated in the figure.

When splitting into a low-shear and a high-shear band of sizes *a* and (*H*-*a*), respectively, the actual band sizes depend on the relative shear rates (assuming a constant overall velocity *U*)

$$\frac{a}{H} = \frac{\dot{\gamma}_2 - U/H}{\dot{\gamma}_2 - \dot{\gamma}_1}. \quad (2)$$

Near onset conditions, the shear rates in the two bands will barely deviate from the average. Fluctuations $\Delta\dot{\gamma}$ are considered to be small. The viscosity adjusts accordingly

$$\eta(\dot{\gamma}) = \eta(\dot{\gamma}_0) + \Delta\dot{\gamma}\left(\frac{\partial\eta}{\partial\dot{\gamma}}\right)_{\gamma_0} + O^2 \quad (3)$$

with $\Delta\dot{\gamma} = (\Delta\dot{\gamma})_1$ or $\Delta\dot{\gamma} = (\Delta\dot{\gamma})_2$.

$\Delta\dot{\gamma}$ is negative in the low-shear band and positive in the high-shear band. $\Delta\dot{\gamma}$ also differs in magnitude for the two bands, but this difference is of minor importance for the following derivation.

The shear stress is uniform throughout the gap, with or without shear bands. Before splitting into separate shear bands, the stress is

$$\sigma_0 = \dot{\gamma}_0\eta(\dot{\gamma}_0); \quad \dot{\gamma}_0 = U/H. \quad (4)$$

After splitting into shear bands, the stress becomes

$$\sigma_{sb} = \dot{\gamma}_1\eta(\dot{\gamma}_1) = \dot{\gamma}_2\eta(\dot{\gamma}_2) \quad (5)$$

or in general

$$\sigma_{sb} = (\dot{\gamma}_0 + \Delta\dot{\gamma})\left[\eta(\dot{\gamma}_0) + \Delta\dot{\gamma}\left(\frac{\partial\eta}{\partial\dot{\gamma}}\right)_{\dot{\gamma}_0}\right]. \quad (6)$$

For the prescribed condition of at constant *U*/*H* (see Fig. 1), the total rate of energy dissipation goes down when the shear stress decays. This is the case with shear banding. Shear bands will grow if their formation reduces the stress in both bands: $(\sigma_{sb})_1 < \sigma_0$ and $(\sigma_{sb})_2 < \sigma_0$. Vice versa, shear fluctuations dampen out if shear bands would cause the shear stress to increase. The decision about instability is with the shear stress criteria

$$\sigma_{sb}\begin{cases} < \sigma_0 & \text{shear banding} \\ = \sigma_0 & \text{onset} \\ > \sigma_0 & \text{stable shear flow} \end{cases}. \quad (7)$$

$\sigma_{sb} = \sigma_0$ is the dividing condition for banding or not banding. We explore this further by equating Eqs. 4 and 6

$$(\dot{\gamma}_0 + \Delta\dot{\gamma})\left[\eta(\dot{\gamma}_0) + \Delta\dot{\gamma}\left(\frac{\partial\eta}{\partial\dot{\gamma}}\right)_{\dot{\gamma}_0}\right] = \dot{\gamma}_0\eta(\dot{\gamma}_0) \text{ for } \sigma_{sb} = \sigma_0. \quad (8)$$

The $\dot{\gamma}_0\eta(\dot{\gamma}_0)$ term cancels and the equation consolidates to

$$\dot{\gamma}_0\Delta\dot{\gamma}\left(\frac{\partial\eta}{\partial\dot{\gamma}}\right)_{\dot{\gamma}_0} + \Delta\dot{\gamma}\;\eta(\dot{\gamma}_0) + (\Delta\dot{\gamma})^2\left(\frac{\partial\eta}{\partial\dot{\gamma}}\right)_{\dot{\gamma}_0} = 0. \quad (9)$$

The fluctuations are assumed to be small with $(\Delta\dot{\gamma})^2 << \Delta\dot{\gamma}$ and the onset criterion $\sigma_{sb} = \sigma_0$ appears as

$$\left(\frac{\partial\ \log\eta}{\partial\ \log\dot{\gamma}}\right)_{\dot{\gamma}_0} = -1 \quad \text{or} \quad W(\dot{\gamma}_c) = +1 \text{ with } \dot{\gamma}_0 \equiv \dot{\gamma}_c. \quad (10)$$

The stability analysis in conjunction with the stress criteria of Eq. 7 shows that

- Complex fluids with $W < 1$ are always stable and without shear banding. Their steady shear viscosity can be measured and stability can be confirmed experimentally
- Complex fluids with $W(\dot{\gamma})$ above $W_c = 1$ may form shear bands as sketched in Fig. 1, where a band of low shear rate coexists with a band of high shear rate at the same overall shear stress σ_{sb}. The shear stress- shear rate function is double valued

$$\dot{\gamma}_1 = \sigma_{sb}/\eta_1 \text{ and } \dot{\gamma}_2 = \sigma_{sb}/\eta_2, \quad (11)$$

For the splitting into $\dot{\gamma}_1$ and $\dot{\gamma}_2$, see also Fig. 2.

Powerlaw viscosity fluid with *W* > 1

The shear instability interferes with viscosity measurements in high *W* fluids with the consequence that viscosity data are unavailable (a suggestion to overcome this problem in case of steady banding will be made further below). In view of this lack of experimental viscosity data, an analytical viscosity

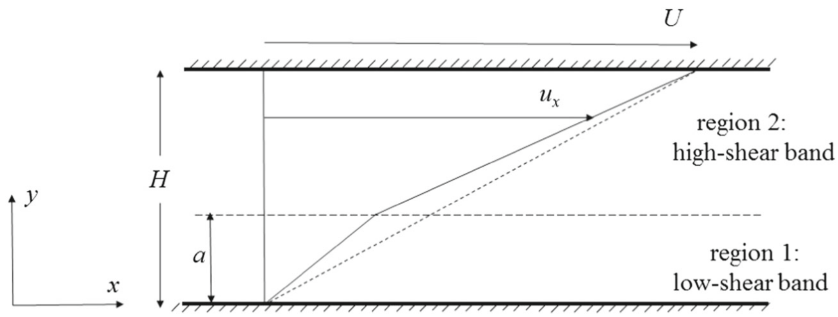

Fig. 1 Schematic velocity $u_x(y)$ of planar shear flow with an average shear rate $\dot{\gamma}_0 = U/H$. The following analysis will assume that it is possible to keep the velocity of the top surface, U, unchanged for flow with or without bands. During shear banding, the flow presumably splits (forming "bands") into two regions of slightly different shear rate, $\dot{\gamma}_1 < U/H$ and $\dot{\gamma}_2 > U/H$. The interface is assumed to be clearly defined and planar (neglecting the fluctuations which have been reported about the interface). The choice of two bands, instead of three or more, and the specific choice of band location do not cause any loss in generality for the derivation below

function needs to be chosen for testing the W-criterion. The Carreau (1972) steady shear viscosity model

$$\sigma(\dot{\gamma}) = \eta(\dot{\gamma})\ \dot{\gamma}; \quad \frac{\eta(\dot{\gamma})}{\eta_0} = \left(1 + (\tau\dot{\gamma})^2\right)^{-n/2} \tag{12}$$

might serve the purpose. Its shear thinning strength calculates as

$$W(\dot{\gamma}) = \frac{n\tau^2\dot{\gamma}^2}{1+\tau^2\dot{\gamma}^2}. \tag{13}$$

Equation 12 is a widely used analytical function for describing shear thinning in quantitative ways. The function includes both the constant zero shear viscosity η_0 and shear thinning at steady shear rates above $1/\tau$, where τ is a characteristic material time for the transition from linear to non-linear shear behavior.

According to the W-criterion, a Carreau fluid is always stable for $n < 1$and has the potential for shear banding for $n > 1$. The following analysis will focus on high-n Carreau fluids (n>1) with the objective of gaining insight into shear banding. To begin with, homogeneous shear flow is assumed (no banding) and the steady shear viscosity $\eta(\dot{\gamma})$ and steady shear stress $\sigma(\dot{\gamma})$ are predicted using Eqs. 12 and 13; see Fig. 2 with solid lines for $W < 1$ and dashed lines for $W > 1$. For these calculations, the viscosity is assumed to be the same throughout a flow region.

$W = W_c = 1$ provides an important reference state for the high-n Carreau fluid. Using Eq. 13, the critical shear rate at $W = 1$ calculates as

$$\tau\dot{\gamma}_c = (n-1)^{-1/2} \quad \text{for} \quad n > 1. \tag{14}$$

Strong shear thinning with $W > 1$ sets in when shearing at a rate above this critical value, $\dot{\gamma}_c$; see Fig. 2. The corresponding stress has the upper limiting value

$$\frac{\tau\sigma(\dot{\gamma}_c)}{\eta_0} = n^{-n/2}(n-1)^{(n-1)/2}, \tag{15}$$

which cannot be exceeded by the high-n Carreau fluid under any steady shearing condition. Shear banding potentially can set in at any macroscopic shear rate U/H with one band shearing at high rate $\dot{\gamma}_2 > \dot{\gamma}_c$ and the other one shearing at low rate $\dot{\gamma}_1 < \dot{\gamma}_c$; see Fig. 2, right side. The shear rate ratio $\dot{\gamma}_2/\dot{\gamma}_1$

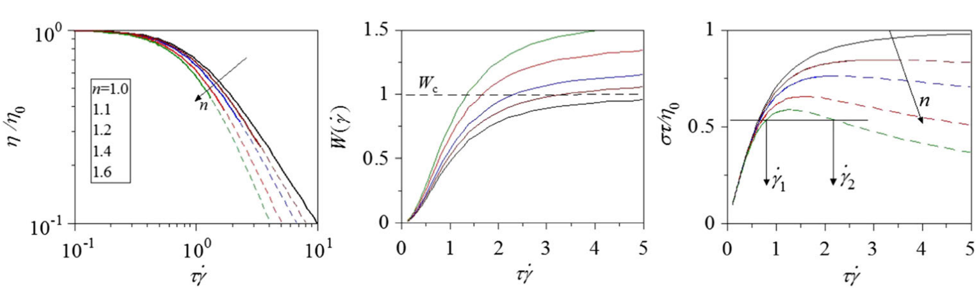

Fig. 2 Steady shear viscosity $\eta(\dot{\gamma})$, shear thinning strength W, and the corresponding shear stress $\sigma(\dot{\gamma})$ of Carreau fluids with $W = 1$ or higher. The shear stress curve becomes double valued in shear rate, $\dot{\gamma}_1$ and $\dot{\gamma}_2$, as indicated in the *right* figure for $n = 1.6$ (and some arbitrary stress); see Eq. 11. The steady shear stress cannot exceed an upper limit which is defined by $W = 1$ (when evaluated for the corresponding n value). The zero shear viscosity η_0 and the characteristic relaxation time τ of the Carreau fluid do not need to be specified in this dimensionless representation. Anomalously strong shear thinning is modeled here by choosing $n > 1$

decreases with $\sigma_{sb} \to \sigma_c$. There is no analytical solution for the shear rate ratio of the Carreau fluid, except for small stress values $\sigma_{sb} << \sigma_c$, at which the shear rate ratio approximately decays with

$$\dot{\gamma}_2/\dot{\gamma}_1 \approx (\tau\sigma/\eta_0)^{\,n/(1-n)}. \tag{16}$$

For shear banding at very low stress, this ratio of shear rates becomes very large. It looks almost as if the high shearing band lubricates the low shearing band. The Carreau fluid is unrealistic in this context since it predicts a decaying viscosity approaching zero at high shear rates in the high-shear band.

A more realistic picture arises when including the limiting high shear rate viscosity, η_∞. Such lower limit was found for many fluids (Ferry 1980), for which shear thinning is not effective any more at very high shear rates. The Carreau-Yasuda fluid (Yasuda et al. 1981) with its steady shear viscosity

$$\frac{\eta(\dot{\gamma})}{\eta_0} = \frac{\eta_\infty}{\eta_0} + \left(1 - \frac{\eta_\infty}{\eta_0}\right)\left(1 + (\tau\dot{\gamma})^a\right)^{-n/a}, \tag{17}$$

can capture this phenomenon; see Fig. 3 with specific values for a and η_∞/η_0. Its shear thinning strength

$$W(\dot{\gamma}) = \left[\frac{1 - \frac{\eta_\infty}{\eta_0}}{1 - \frac{\eta_\infty}{\eta_0}\left(1 - (1 + \tau^a\dot{\gamma}^a)^{n/a}\right)}\right]\frac{n\tau^a\dot{\gamma}^a}{1 + \tau^a\dot{\gamma}^a} \tag{18}$$

passes through a maximum at intermediate shear rates with $W_{\max}$ exceeding $W_c = 1$ as shown in Fig. 3. Interestingly, the Carreau-Yasuda fluid with n = 1.1 barely reaches the critical value of $W_c = 1$ because of the stabilizing effect of η_∞ (see also Fig. 4 about this effect). The corresponding steady shear stress, $\sigma(\dot{\gamma}) = \eta(\dot{\gamma})\ \dot{\gamma}$, rises at first and then decays as shear rates exceed a critical value $\tau\dot{\gamma}_c \approx (n-1)^{-1/a}$, very much like in the high-n Carreau fluids shown above. However, the stress rises again at the very high shear rates when the viscosity approaches its lower limiting value. In this way, the stress assumes a sigmoidal shape with maximum and minimum at $W = W_c = 1$. The shear rate becomes triple-valued (unstable) at intermediate stress, but is single-valued (stable) at low and at high stress as indicated in Fig. 3 at the right.

The origin of the three shear rates becomes obvious when considering the three asymptotes as demonstrated in Fig. 4, using n = 1.6 as example. The three asymptotes are

$$\frac{\tau\sigma(\dot{\gamma})}{\eta_0} = \begin{cases} \tau\dot{\gamma} & 1\text{ - low shear rate}, \quad W < 1 \\ (\tau\dot{\gamma})^{1-n} & 2\text{-shear thinning region}, \quad W > 1 \\ \tau\dot{\gamma}\eta_\infty/\eta_0 & 3\text{ - high shear rate}, \quad W < 1 \end{cases} \tag{19}$$

Only the second shear rate, $\dot{\gamma}_2$, falls into a shear rate region with $W > 1$. This suggests that the shear thinning strength has to exceed the critical value of unity at some intermediate shear rate for shear banding to occur even if the shear bands themselves are regions of $W < 1$.

The mere existence of a limiting high shear viscosity, η_∞, stabilizes the flow. For shear banding to occur, η_∞ has to be much below the zero shear viscosity, $\eta_\infty << \eta_0$. Figure 5 demonstrates this phenomenon for high-n Carreau-Yasuda fluids. They remain stable up to high n values ($W < 1$) when the two viscosity values are close together. Even a decade of difference between η_∞ and η_0 does not seem to be enough for shear banding to set in.

Using the Carreau-Yasuda equation, Eq. 17, together with the kinematic constraint, Eq. 2, and the equations for the stress, Eq. 5, one could calculate the shear rate in the bands and the band thickness by minimizing the rate of energy dissipation. This, however, would exceed the immediate objective of this brief communication.

Discussion

The W-criterion confirms (as it should) the well-known stability of polymer melt and solutions. Their typical shear thinning may reach W-values of 0.4 to 0.7, but never gets close to $W_c = 1$.

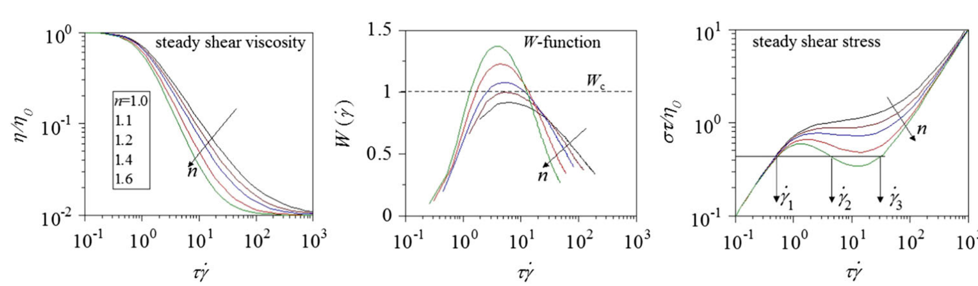

Fig. 3 High-n Carreau-Yasuda fluids with viscosity ratio $\eta_\infty/\eta_0 = 0.01$ and exponent a = 2. The shear rate becomes triple valued at intermediate shear stress as indicated in the *right* figure for n = 1.6. Parameters η_0 and τ of Eq. 17 do not have to be specified in the dimensionless representation. Anomalously strong shear thinning is modeled here by choosing $n > 1$

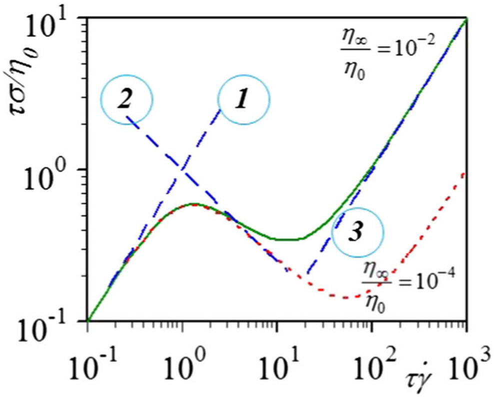

Fig. 4 Three asymptotes for sections of the shear stress function demonstrate the origin of three shear rates at intermediate stress. Parameter n = 1.6. The asymptotes are number-marked according to Eq. 19. The shear thinning region, using parameters of Fig. 3, is too small for the stress to get close to asymptote no. 2. However, when increasing the difference between the two leading viscosities, η_∞/η_0, shear thinning increases and the stress approaches asymptote no. 2 more clearly (*dashed line* with deep minimum)

The *W*-criterion predicts the leveling off of the shear stress at increasing shear rates, which already had been reported by Berret (2005) and many others since. At least qualitatively, there seems to be agreement of the experimental data with the stress evolution in Fig. 3.

The W-criterion can be expressed in terms of the steady shear stress σ by rearranging

$$W(\dot{\gamma}) = -\frac{\partial \ \log\eta}{\partial \ \log\dot{\gamma}} = -\frac{\partial \ \log\sigma}{\partial \ \log\dot{\gamma}} + 1. \tag{20}$$

Integration results in

$$\sigma = C\dot{\gamma}^{(1-W)}. \tag{21}$$

In case of $W > 1$, the shear stress decays with increasing shear rate. This decaying shear stress function, also shown in Figs. 2 and 3, is a commonly used criterion for the occurrence of steady shear banding in homogeneous fluids (Moorcroft and Fielding 2013). W = 1 describes the onset condition, σ = constant. It should be mentioned, however, that a sigmoidal flow curve, having a decaying shear stress at intermediate shear rates, has also been reported to cause elastic instabilities in flows such as melt fracture (Yerushalmi et al. 1970, Petrie and Denn 1976). Such elastic instabilities, where the entire flow alternates between two or more states, might be related to shear banding in some complicated way.

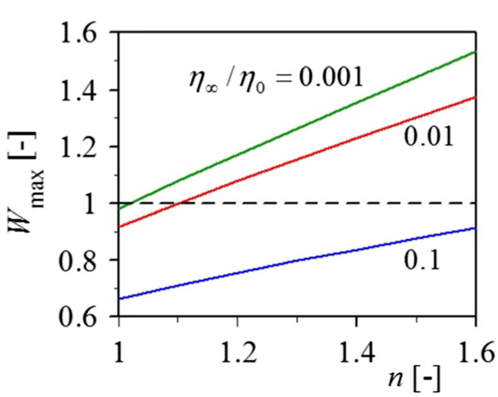

Fig. 5 $W_{\max}$ for Carreau-Yasuda fluids for $n > 1$ and various η_∞/η_0. According to the *W*-criterion, $\eta_\infty/\eta_0 = 0.1$ stabilizes the flow up to high n values while complex fluids with $\eta_\infty/\eta_0 = 0.01$ and 0.001 might destabilize and form bands under suitable conditions

Generally, the condition for band development, exceeding $W_c = 1$, amounts to a severe drop in shear viscosity. A shear-induced structural breakdown may be able to achieve such viscosity drop in complex fluids of fragile structure (surfactants, colloidal gels, coacervates). It will not matter whether the structural breakdown is reversible or irreversible. The banding might generate new structural states in sheared samples. Very few viscosity data are available. More experiments are needed for determining *W*-values for such highly sensitive (to shear) materials.

The above analysis postulates the existence of a continuous viscosity function up to high shear rates. Not all shear banding fluids might provide such behavior, especially those with yield stress (Besseling et al. 2010, Ovarlez et al. 2009, 2013). This needs to be looked into. It will require a more extensive study to analyze published shear banding data in terms of the *W*-criterion with the aim of confirming or dismissing it for certain material groups.

As mentioned above, the steady shear viscosity cannot be measured any more when shear banding takes over. However, there might be a possibility to overcome this problem. In case of two stable bands, for instance, it might be possible to measure the shear rates in the two bands, the relative band sizes, and the shear stress and use these measurements to recreate the $\eta(\dot{\gamma})$ and $\sigma(\dot{\gamma})$ plots along the lines of Fig. 3. In this way, the viscosity function might be extracted in spite of shear banding. However, a more complete model for stable bands will be needed for that purpose. This brief communication only concerns the onset conditions for shear bands.

The above analysis is based on the steady shear viscosity and misses out on the transient character of the band formation in time-dependent flows and the fluctuations of the interface between bands (flat interface simplification). The dynamics of shear band formation cannot be predicted without expanding the analysis to a full viscoelastic model. Such expansion is not considered in this brief communication.

In a multi-component material, shear banding might also be the result of flow-induced composition gradients. Inhomogeneity adds a whole new level of complexity which is beyond consideration here.

Conclusion

$W > 1$ means that there are shearing conditions in which shear thinning is so strong that the stress decays at increasing shear rates. Instability occurs only if a sample is actually able to access the shearing at $W > 1$. The W-criterion also predicts that low-W materials, defined by a $W(\dot{\gamma})$ below $W_c = 1$ at all shear rates, will always be stable. The existence of a limiting high shear rate viscosity, η_∞, potentially stabilizes shear flows, which otherwise would develop shear bands. The range of validity of the W-criterion needs to be tested further and is planned for the future, but it would exceed this brief communication by far.

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