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Mathematical Properties of the Differential Pom-Pom Model

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Abstract: Recently in order to describe the complex rheological behavior of polymer melts with long side branches like low density polyethylene, new constitutive equations called the pom-pom equations have been derived by McLeish and Larson on the basis of the reptation dynamics with simplified branch structure taken into account. In this study mathematical stability analysis under short and high frequency wave disturbances has been performed for the simplified differential version of these constitutive equations. It is proved that they are globally Hadamard stable except for the case of maximum constant backbone stretch ($\lambda = q$) with arm withdrawal s_c neglected, as long as the orientation tensor remains positive definite or the smooth strain history in the flow is previously given. However this model is dissipative unstable,since the steady shear flow curves exhibit non-monotonic dependence on shear rate. This type of instability corresponds to the nonlinear instability in simple shear flow under finite amplitude disturbances. Additionally in the flow regime of creep shear flow where the applied constant shear stress exceeds the maximum achievable value in the steady flow curves, the constitutive equations will possibly violate the positive definiteness of the orientation tensor and thus become Hadamard unstable.

Introduction

Even though nowadays rheologically complex fluids like particle suspension of polymer melt are very important in theoretical aspect as well as in practical applications, rheological description of some pure polymer melts still remains as a challenging task. Among them, low density polyethylene (LDPE) melt has been considered as the most difficult one probably due to its high degree of long chain branching.[1] Especially so called Melt 1 (IUPAC A or IUPAC X) is the LDPE sample which has been the most extensively characterized in the viewpoint of rheology. It shows even in simple flows highly nonlinear behavior such as high shear thinning and at the same time high extensional strain hardening, which altogether have prevented reasonable rheological description by a single constitutive model with one set of numerical parameters.

Recently McLeish and Larson[2] proposed constitutive equations called the pom-pom model in order to account for these complicated phenomena presumably exhibited by long side branches present in the LDPE melt. They derived the equations based on the reptation dynamics, introducing simplified geometrical molecular structure with long branches named as a pom-pom molecule, for which the schematic illustration is represented in Figure 1. The original pom-pom model is presented as a set of integral/differential equations, and due to their computational inefficiency a simplified differential version is also suggested. Due to its theoretical and also practical importance, these model equations draw many rheologists' attention and quite a few results have already been reported

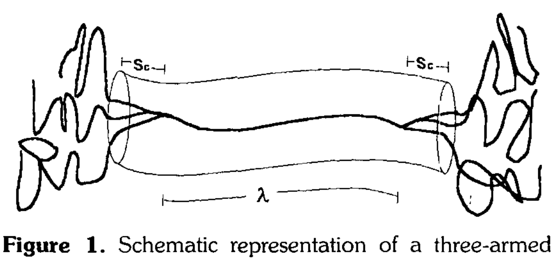

Figure 1. Schematic representation of a three-armed pom-pom molecule (q = 3).

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on their applications.

Regarding LDPE melt rheology, Rubio and Wagner[3] compared the theoretical computation by both versions of the pom-pom model with experimental data, and found some qualitative disagreement. Inkson and coworkers[4] also investigated the rheological behavior of LDPE including Melt 1, and concluded reasonable experimental description by the constitutive equations (the differential version with arm withdrawal length neglected for simplicity) in some transient response of simple shear and extensional flows. However, in their flow modeling they employed about 20 numerical parameters in addition to linear viscoelastic ones, and there is no description of other sets of available experimental data such as strain recovery and so on. On the other hand, in 1995 Simhambhatla and Leonov[5] have already described almost all available experimental data for Melt 1 (also for IUPAC A and IUPAC X) successfully employing the globally stable Leonov constitutive equation, which contains only one nonlinear parameter, and in the above-mentioned works their results have been ignored.

In this study, we investigate the mathematical characteristics of the differential pom-pom constitutive equations in view of stability. Since results obtained by some unstable equations under some simplifying approximations like inertialess approach are not meaningful at all, these mathematical analyses are quite important and should precede extensive applications of the constitutive equations. There are two types of instability in viscoelastic constitutive equations such as Hadamard and dissipative instabilities.[6] In this paper, we mainly examine properties of the pom-pom model in the sense of Hadamard stability that means the stability of differential equations under short and high frequency wave disturbances, and at the end we make a remark on the dissipative instability that arises in simple flows.

The Pom-Pom Model

The pom-pom constitutive equations for polymer molecules with long side branches and more than one branch points are derived on the basis of the reptation dynamics for a melt of identical molecules with a very simplified branching structure, called 'pom-pom' molecules.[2] A typical pom-pom molecule illustrated in Figure 1 consists of two identical q-armed stars connected by a 'backbone' section that pursues hypothetical reptational motion. By McLeish and Larson,[2] this pom-pom molecule is suggested as the simplest analog of the real polymeric molecule with long branching like LDPE (low density polyethylene).

The original pom-pom model is suggested as the following complicated integral/differential constitutive equations that include various variables and parameters in order to take into account molecular geometry such as branch and backbone structures and its time evolution.

The stress representation:

$$\boldsymbol{\sigma} = -p\boldsymbol{\delta} + G\left(\lambda^2 + \frac{1}{\phi_b}\,\frac{2qs_c}{2qs_a+s_b}\right)\boldsymbol{S},\quad G = \frac{15}{4}G_0\phi_b^2\,,$$
$$\phi_b = \frac{s_b}{2qs_a+s_b}\,. \tag{1}$$

Here $\boldsymbol{\sigma}$ is the total stress tensor, $\boldsymbol{\delta}$ the unit tensor, p the isotropic pressure, $\boldsymbol{S}$ the orientation tensor, G_0 the plateau modulus, ϕ_b the fraction of molecular weight contained in the crossbar (backbone), q the number of arms in one of two branches, and s_a and s_b are dimensionless molecular weights of the backbone and arm, respectively, scaled by the entanglement molecular weight. s_c explains the dimensionless length of the arm withdrawn into the backbone tube, λ is the stretch ratio of the backbone under the flow field, and thus both are functions of time.

Backbone orientation:

$$\boldsymbol{S} = \int_{-\infty}^{t}\frac{1}{\tau_b(t_1)}\exp\left(-\int_{t_1}^{t}\frac{dt'}{\tau_b(t')}\right)\boldsymbol{Q}dt_1,\quad \boldsymbol{Q} = \left\langle\frac{\boldsymbol{u}'\boldsymbol{u}'}{(u')^2}\right\rangle_0\,,$$
$$\boldsymbol{u}' = \boldsymbol{E}(t-t_1)\cdot\boldsymbol{u}\,,\quad \frac{d\boldsymbol{E}}{dt} = \nabla\boldsymbol{v}^T\cdot\boldsymbol{E}\,. \tag{2}$$

In these equations, the strain $\boldsymbol{Q}$ is slightly modified to become a universal strain measure of the Doi-Edwards model with independent alignment approximation which is employed by Rubio and Wagner.[3] τ_b is the relaxation time of backbone orientation, $\boldsymbol{u}$ the unit vector, $\boldsymbol{E}$ the deformation gradient tensor, $\boldsymbol{v}$ the velocity, ∇ the gradient operator, $\nabla\boldsymbol{v}^T$ the transpose of the velocity gradient, and the symbol $\langle\ \rangle_0$ is the operation of averaging over the configuration space.

Backbone stretch:

$$\frac{d\lambda}{dt} = \lambda \nabla \boldsymbol{v}^T : \boldsymbol{S} - \frac{1}{\tau_s}(\lambda - 1) \quad \text{for } \lambda < q. \tag{3}$$

Here τ_s is the characteristic time of backbone stretch. The above equation is valid only for $\lambda < q$ and even if equation (3) still expresses the increase of λ after it reaches the value q, λ is fixed at the value of q and the following evolution equation of arm withdrawal starts to act.

Arm withdrawal:

$$\frac{ds_c}{dt} = \left(q\frac{s_b}{2} + s_c\right)\nabla \boldsymbol{v}^T : \boldsymbol{S} - \frac{1}{2\tau_a} \quad \text{for } \lambda = q. \tag{4}$$

Here τ_a is the characteristic time of arm relaxation.

Characteristic time for backbone orientation:

$$\tau_b = \frac{4}{\pi^2} s_b^2 \phi_b \tau_a q\ . \tag{5}$$

Characteristic time for arm relaxation:

$$\tau_a = \tau_a \exp\left[\frac{15}{4} s_a \left\{\frac{(1-x)^2}{2} - (1-\phi_b)\frac{(1-x)^3}{3}\right\}\right],\ x = \frac{s_c}{s_a} \tag{6}$$

Characteristic time for backbone stretch:

$$\tau_s = s_b \tau_a(0) q\ . \tag{7}$$

In the above equations for characteristic times, τ_a and τ_b are dependent upon s_c and thus functions of time, but τ_s is constant.

Due to computational inefficiency as well as complexity of the double integral in the above model equations (2), McLeish and Larson have proposed rather simple differential version of the pom-pom model. Hence in the differential pom-pom constitutive equations, the following evolution equation substitutes equations (2) for backbone orientation:

$$\overset{\nabla}{\boldsymbol{c}} + \frac{1}{\tau_b}(\boldsymbol{c} - \boldsymbol{\delta}) = \boldsymbol{0}, \quad \boldsymbol{S} = \frac{\boldsymbol{c}}{tr\ \boldsymbol{c}} \tag{8}$$

Here $\overset{\nabla}{\boldsymbol{c}} = (d\boldsymbol{c}/dt) - \nabla \boldsymbol{v}^T \cdot \boldsymbol{c} - \boldsymbol{c} \cdot \nabla \boldsymbol{v}$ is the upper-convected time derivative of the configuration tensor $\boldsymbol{c}$, and all the other equations from (1) to (7) except (2) are kept. Note that the evolution equation (8) for the configuration tensor is exactly the same with the evolution equation for the upper-convected Maxwell model in the configuration tensor representation (see for example, the paper by Kwon and Leonov[6]).

In this study, we investigate the mathematical characteristics only of the simplified differential version of the pom-pom model, that is, equations (1) and (3)-(8).

The Hadamard Stability Analysis

The Hadamard stability of differential equations implies the stability of equations under extremely short and high frequency wave disturbances. Hence it accounts for the elastic properties of viscoelastic constitutive equations related to fast responses such as type of differential operator and elastic free energy.[6] Also it is often interpreted as the viscoelastic change of type. In any case, no matter what it means, unstable equations in Hadamard sense should be understood as non-physical formulation of viscoelastic phenomena and discarded from the further application for viscoelastic flow analysis.

Total set of equations for the isothermal incompressible viscoelastic flow is composed of following equations of motion and continuity in addition to the constitutive equations explained in the previous section:

$$\rho\frac{d\boldsymbol{v}}{dt} = \nabla \cdot \boldsymbol{\sigma}, \qquad \nabla \cdot \boldsymbol{v} = 0 \tag{9}$$

Here ρ is the constant density of the fluid and the body force (gravity) is neglected for simplicity of analysis. Upon the above set of equations (1) and (3)-(9), we impose such short and high frequency wave disturbances as

$$\begin{aligned} \{\boldsymbol{\sigma}, \boldsymbol{S}, \boldsymbol{A}, \boldsymbol{v}, \lambda, p, s_c\} &= \{\boldsymbol{\sigma}_0, \boldsymbol{S}_0, \boldsymbol{A}_0, \boldsymbol{v}_0, \lambda_0, p_0, s_{c0}\} \\ &+ \{\delta\boldsymbol{\sigma}, \delta\boldsymbol{S}, \delta\boldsymbol{A}, \delta\boldsymbol{v}, \delta\lambda, \delta p, \delta s_c\}, \\ \{\delta\boldsymbol{\sigma}, \delta\boldsymbol{S}, \delta\boldsymbol{A}, \delta\boldsymbol{v}, \delta\lambda, \delta p, \delta s_c\} \\ &= \varepsilon\{\hat{\boldsymbol{\sigma}}, \hat{\boldsymbol{S}}, \hat{\boldsymbol{A}}, \hat{\boldsymbol{v}}, \hat{\lambda}, \hat{p}, \hat{s}_c\}\exp[i(\boldsymbol{k}\cdot\boldsymbol{x} - \omega t)/\varepsilon^2]\ . \end{aligned} \tag{10}$$

Here $\{\boldsymbol{\sigma}_0, \boldsymbol{S}_0, \boldsymbol{A}_0, \boldsymbol{v}_0, \lambda_0, p_0, s_{c0}\}$ and $\{\delta\boldsymbol{\sigma}, \delta\boldsymbol{S}, \delta\boldsymbol{A}, \delta\boldsymbol{v}, \delta\lambda, \delta p, \delta s_c\}$ are basic solutions and applied disturbances of the corresponding variables, respectively, and from now on we remove the subscript 0 in the basic solutions for convenience of notation. $\{\hat{\sigma},\hat{\mathbf{S}},\hat{\mathbf{A}},\hat{\mathbf{v}},\hat{\lambda},\hat{p},\hat{s}_c\}$ is the amplitude of disturbing wave, $\mathbf{k}$ the wave vector, ω the frequency and ε is the small amplitude parameter that also expresses the short wavelength and high frequency of the disturbing wave. As can be seen from equation (10), if the frequency ω is complex-valued with positive imaginary part, this disturbance implies the exponential growth of perturbation with time, and thus it means the Hadamard instability.

We divide the complete analysis into three parts. First part is the stability analysis in the flow regime of $\lambda < q$, where s_c always vanishes and equation (4) does not play any role. In this case, the relaxation time τ_b is constant and in the perturbed system (10) δs_c is absent. The second and third parts of analysis consist in the region of $\lambda = q$, hence the backbone stretch ratio λ is constant, $\delta\lambda$ vanishes, and equation (4) starts to react in the set of perturbed equations. For the simplest analysis, in the second part we neglect the contribution of the arm withdrawal length s_c, and such a simplified set of the pom-pom constitutive equations has already been employed by Inkson and coworkers[4] to describe the rheological behavior of low density polyethylene melt. However in the third part, we accomplish the complete analysis of stability including the s_c variation for $\lambda = q$.

Actually the second part of the analysis may be regarded as meaningless, since it is composed of equations that do not coincide with the original formulation. However due to its simplicity the set of equations is applied for the viscoelastic flow analysis, and thus for the practical purpose the investigation of its mathematical characteristics deserves its consequence.

Stability Analysis when $\lambda < q$. Since s_c and its perturbation vanish, we disturb equations (1), (3), (8) and (9) according to equation (10). For linear stability analysis we collect only the lowest order terms in ε, and then we obtain following linear set of relations for the amplitudes of disturbances:

$$\rho\Omega\hat{v}_j = -\hat{\sigma}_{jm}k_m\,, \quad k_m\hat{v}_m = 0\,,$$

$$\hat{\sigma}_{ij} = -\hat{p}\delta_{ij} + G(2\lambda\hat{\lambda}S_{ij} + \lambda^2\hat{S}_{ij})\,,$$

$$-\hat{\lambda}\Omega = \lambda S_{mn}\hat{v}_m k_n, \quad \hat{S}_{mn} = \frac{1}{c_{kk}}\left(\hat{c}_{mn} - \frac{c_{mn}}{c_{jj}}\hat{c}_{ii}\right) \tag{11}$$

$$\Omega\hat{c}_{ij} + c_{im}\hat{v}_j k_n + c_{mj}\hat{v}_i k_n = 0$$

Here $\Omega = \omega\text{-}\mathbf{k}\cdot\mathbf{v}$ is the frequency with Doppler's shift on the basic flow field $\mathbf{v}$. We can solve the above linear set and then obtain the equation of stability as follows:

$$\begin{aligned}\rho\Omega^2\hat{v}_j\hat{v}_j &= G\lambda^2\delta_{ij}S_{mn}\hat{v}_i\hat{v}_j k_m k_n \\ \Rightarrow\ \rho\Omega^2 &= G\lambda^2 S_{mn}k_m k_n\,.\end{aligned} \tag{12}$$

Since the frequency ω and thus Ω should be real-valued for stability, the necessary and sufficient condition of the Hadamard stability becomes

$$G\lambda^2 S_{mn}k_m k_n > 0 \tag{13}$$

which exactly requires the positive definiteness of the second rank tensor $\mathbf{S}$. Due to equation (8), the positive definiteness of $\mathbf{S}$ is equivalent to that of $\mathbf{c}$.

Regarding the positive definiteness of the configuration tensor $\mathbf{c}$, Hulsen[7] and Leonov[8] independently proved one theorem in some limited situation. In that theorem, it is stated that for any given piecewise smooth strain history with the initial condition $\mathbf{c} = \boldsymbol{\delta}$ the principal values of tensor $\mathbf{c}$ are positive. Hence we can conclude that *in the flow regime of $\lambda < q$ the pom-pom constitutive equations are Hadamard stable as long as the tensor $\mathbf{c}$ is positive definite, i.e., when the smooth strain history is predefined*. However it is worth mentioning that in some given stress history the constitutive equations with special type of steady flow curves are proved to violate the positive definiteness of the tensor $\mathbf{c}$[9].

Stability Analysis when $\lambda = q$ (s_c neglected). In this case, the backbone stretch λ is fixed at the value of the number of arms q, and thus its disturbance vanishes. Therefore substitution of 0 for $\hat{\lambda}$ in equations (11) and solving the system yield the following dispersion relation:

$$\rho\Omega^2\hat{v}_j\hat{v}_j = Gq^2(\delta_{ij}S_{mn} - 2S_{im}S_{jn})\hat{v}_i\hat{v}_j k_m k_n \tag{14}$$

This specific problem of stability is equivalent to the problem for the constitutive equations in a sort of general Finger form such that

$$\overset{\nabla}{\mathbf{c}}+\frac{1}{\tau_b}(\mathbf{c}-\boldsymbol{\delta})=\mathbf{0},\quad \frac{2\rho}{G}F\equiv U=q^2\ln I_1,\quad I_1=tr\ \mathbf{c}\ ,$$
$$\sigma=G\{\varphi_1\mathbf{c}+\varphi_2(I_1\mathbf{c}-\mathbf{c}^2)+\varphi_3I_3\boldsymbol{\delta}\}=-p\boldsymbol{\delta}+Gq^2\frac{\mathbf{c}}{I_1}\ ,$$
$$\varphi_j=\frac{\partial U}{\partial I_j}\ ,\ I_1=tr\ \mathbf{c},\ I_2=\frac{1}{2}(I_1^2-tr\ \mathbf{c}^2)\ ,\ I_3=det\ \mathbf{c}\ . \tag{15}$$

Here F (or U) is the (dimensionless) elastic free energy, i.e., the Helmholtz free energy corresponding to the pom-pom model for this specific case. According to above equations, for this constitutive model the following holds

$$\varphi_1=\frac{q^2}{I_1},\ \ \varphi_2=\varphi_3=0,\ \ \varphi_{11}=-\frac{q^2}{I_1^2},\ \ \varphi_{12}=\varphi_{21}=\varphi_{22}=0 \tag{16}$$

where $\varphi_{ij}=\dfrac{\partial^2 U}{\partial I_i\partial I_j}$.

After rewriting the constitutive relation as equations (15), we may directly apply the following necessary and sufficient condition for Hadamard stability proved in the paper[6]:

(i) $\beta_i>0$,

(ii) $w_i+2\beta_i\sqrt{c_jc_k}>0\quad (i\neq j\neq k)$,

(iii) $\{(w_i+2\beta_i\sqrt{c_jc_k})^{1/2}+(w_j+2\beta_j\sqrt{c_ic_k})^{1/2}\}^2>w_k-2\beta_k\sqrt{c_ic_j}$
$(i\neq j\neq k)$ (17)

where c_i is the eigenvalue of the tensor $\mathbf{c}$ and

$$\beta_i=\varphi_1+\varphi_2c_i\ ,$$
$$w_i=(I_i-c_i)\beta_i+2(I_i^2-2I_2-c_i^2-2I_3/c_i)\{\varphi_{11}+(\varphi_{12}+\varphi_{21})c_i+\varphi_{22}c_i^2\}\ . \tag{18}$$

Hence for this problem, $\beta_i=\varphi_1$ irrespective of the value of subscript i, and $w_i=(I_1-c_i)\varphi_1+2(I_1^2-2I_2-c_i^2-2I_3/c_i)\varphi_{11}$. In order to demonstrate the instability of these constitutive equations, we here consider the case of simple shear flow, where the principal values and invariants of tensor $\mathbf{c}$ valid at the moment of step strain γ become

$$c_1=c,\ c_2=1/c,\ c_3=1,\ c=(\gamma^2+2+\sqrt{\gamma^4+4\gamma^2})/2\ ,$$
$$I_1=I_2=\gamma^2+3\ . \tag{19}$$

Then the second inequality in (17) for $i=1$ reduces to

$$\sqrt{c}(\gamma^2+3)(\gamma^2+3-c)-2\sqrt{c}(\gamma^4+4\gamma^2+3-c^2-2/c)+2(\gamma^2+3)>0\ , \tag{20}$$

which is always satisfied for all values of γ. However for $i=2$ or 3 it restricts the value of c or γ by

$$(\gamma^2+3)(\gamma^2+3-1/c)-2(\gamma^4+4\gamma^2+3-2c-1/c^2)+2\sqrt{c}(\gamma^2+3)>0\ \text{ and }\ -\gamma^4-\gamma^2+12>0 \tag{21}$$

The latter of inequalities (21) yields the most rigorous constraint such as $\gamma<\sqrt{3}$ for stability. Hence we conclude that *the pom-pom constitutive equations for $\lambda=q$ are Hadamard unstable when the instantaneous shear strain exceeds $\sqrt{3}$ if we neglect the variable s_c*. Also it is highly probable for the results obtained in the paper by Inkson *et al.*[4] to be located in the unstable solution branch when the strain rate is high.

Complete Stability Analysis when $\lambda=q$. In order to study the stability characteristics of the model equations in their full description, now we have to consider the disturbance of s_c, and thus the relaxation times τ_a and τ_b are also perturbed, while λ is constant. The stress relation (1) and the evolution equation for arm withdrawal (4) under the disturbance yield

$$\hat{\sigma}_{ij}=-\hat{p}\delta_{ij}+Gq\left(q+2\frac{s_c}{s_b}\right)\hat{S}_{ij}+\frac{2Gq}{s_b}S_{ij}\hat{s}_c\ ,$$
$$-\Omega\hat{s}_c=\left(\frac{q}{2}s_b+s_c\right)S_{mn}\hat{v}_mk_n\ . \tag{22}$$

Solving the linear system of 1[st], 2[nd], 5[th] and 6[th] of equations (11) with (22) results in

$$\rho\Omega^2\hat{v}_j\hat{v}_j=Gq\left(q+2\frac{s_c}{s_b}\right)(\delta_{ij}S_{mn}-S_{im}S_{jn})\hat{v}_i\hat{v}_jk_mk_n \tag{23}$$

which has to be positive for stability. In the tensor $\mathbf{c}$ representation, due to positivity of G, q, s_c, s_b

and I_1, the inequality imposed on (23) can be rewritten as

$$\left(\delta_{ij}\frac{c_{mn}}{I_1}-\frac{c_{im}}{I_1}\frac{c_{jn}}{I_1}\right)\hat{v}_i\hat{v}_j k_m k_n>0$$
$$\Rightarrow\left(\frac{1}{\sqrt{I_1}}\delta_{im}c_{jn}-\frac{1}{I_1^{3/2}}c_{ij}c_{mn}\right)\hat{v}_i k_j \hat{v}_m k_n>0 \ . \qquad (24)$$

This condition of stability coincides with the case of $\varphi_1=1/\sqrt{I_1}$, $\varphi_{11}=-1/2I_1^{3/2}$ and $\varphi_2=\varphi_{12}=\varphi_{21}=\varphi_{22}=0$ for the following inequality obtained in the reference[10]:

$$\begin{aligned}\boldsymbol{B} &= B_{ijmn}\hat{v}_i k_j \hat{v}_m k_n\\ &= [\varphi_1\delta_{im}c_{jn}+\varphi_2(I_1\delta_{im}c_{jn}-\delta_{im}c_{j\alpha}c_{\alpha n}-c_{im}c_{jn}+c_{ij}c_{mn})\\ &+2\varphi_{11}c_{ij}c_{mn}+2\varphi_{12}c_{ij}(I_1c_{mn}-c_{m\alpha}c_{\alpha n})\\ &+2\varphi_{21}(I_1c_{ij}-c_{i\alpha}c_{\alpha j})c_{mn}\\ &+2\varphi_{22}(I_1c_{ij}-c_{i\alpha}c_{\alpha j})(I_1c_{mn}-c_{m\beta}c_{\beta n})]\hat{v}_i k_j \hat{v}_m k_n>0 \ .\end{aligned} \qquad (25)$$

Then corresponding potential equivalent for the constitutive equations in this particular stability problem becomes

$$U=2\sqrt{I_1} \qquad (26)$$

At this point, we can apply the Renardy's stability condition,[11] which states that for the K-BKZ class of constitutive equations the convexity of the thermodynamic potential U in terms of invariants $\sqrt{I_1}$ and $\sqrt{I_2}$ is the sufficient condition for stability, and it has been verified that Renardy's condition is also sufficient for Hadamard stability when it is applied to differential Maxwell-like models with an upper convected derivative.[10]

Since the equivalent potential (26) clearly satisfies the Renardy's condition, we can finally conclude that *in the flow regime of $\lambda = q$ the pom-pom constitutive equations are Hadamard stable as long as the tensor* **c** *is positive definite.*

Remark on the Properties of Tensor c

According to the preceding results on stability in this work, the pom-pom constitutive equations are globally Hadamard stable (stable in Hadamard sense in any type of flow and in any value of velocity gradient tensor) except for the case of $\lambda = q$ with s_c neglected, as long as the configuration tensor **c** is positive definite. Now we discuss the extreme situation when possibly the positive definiteness can be violated.

In the steady state of simple shear flow, the constitutive equations (1), (3) and (8) reduce to

$$-2\Gamma c_{12}+(c_{11}-1)=0,\ -\Gamma c_{22}+c_{12}=0,\ c_{22}=c_{33}=1,$$
$$c_{13}=c_{23}=0,\ \lambda\Gamma\frac{\Gamma}{3+2\Gamma^2}-\frac{\tau_b}{\tau_s}(\lambda-1)=0\ ,$$
$$\sigma_{12}=G\lambda^2\frac{c_{12}}{c_{11}+2}\ , \qquad (27)$$

where the dimensionless shear rate is defined as $\Gamma=\tau_b\dot{\gamma}$ and τ_b is fixed at the value of $\tau_b(s_c=0)$. Then we finally obtain the backbone stretch ratio and the shear stress as

$$\lambda=\frac{\tau_b}{\tau_s}\left[\frac{\tau_b}{\tau_s}-\frac{\Gamma^2}{3+2\Gamma^2}\right]^{-1},\quad \sigma_{12}/G=\lambda^2\frac{\Gamma}{3+2\Gamma^2} \qquad (28)$$

as long as $\lambda < q$. From the above equations the ratio between the relaxation times τ_b/τ_s should exceed the value of 1/2 to avoid singularity. In Figure 2, the behavior between the dimensionless variables Γ and σ_{12}/G is shown for several values of τ_b/τ_s, and all the curves are valid when $q\geq 2$ (hence they are valid always), since for those values of τ_b/τ_s λ never exceeds 2. All the flow curves show maxima and then decreasing branches of

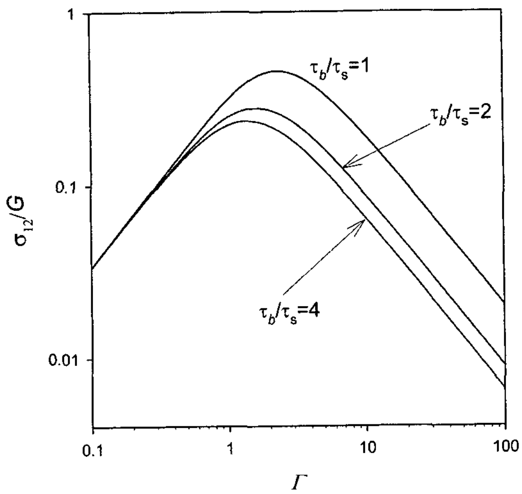

Figure 2. The dimensionless shear stress vs. dimensionless shear rate in steady simple shear flow of the differential pom-pom constitutive equations for various values of the ratio between relaxation times.

solution, and the achievable shear stresses are all bounded below the maximum. Here we can directly apply the theorem derived in the paper[6] that asserts so called 'dissipative instability'. It is evident first that mechanically and thermodynamically, the decreasing branch of the flow curve in Figure 2 is the unstable solution. Consider the system that lies initially at some point of that decreasing branch, and then remove the applied force. Then as the force decreases, the shear rate (Γ) or the flow rate increases in accordance to the flow curve, and finally as the force approaches 0, the flow rate diverges to infinity. Another type of severe blow-up instability exhibited by the dissipative unstable constitutive models has been exposed in reference,[9] where the violation of the positive definiteness of **c** is also demonstrated. Therefore now we can deduce the following: *the pom-pom constitutive equations are dissipative unstable, and in that unstable flow regime, i.e., in the simple shear flow where the constant shear stress greater than the maximum achievable shear stress in the steady flow curves is applied, the positive definiteness of tensor* **c** *is violated and they also become Hadamard unstable.*

Conclusion

Mathematical stability analysis under short and high frequency wave disturbances has been performed for the pom-pom constitutive equations that are recently suggested to describe fluid dynamical behavior of polymer melts with long side branches like low density polyethylene. It is proved that they are globally Hadamard stable except for the case of maximum constant backbone stretch ($\lambda = q$) with arm withdrawal s_c neglected, as long as the orientation tensor $\boldsymbol{S}$ or **c** remains positive definite, that is, the smooth strain history is pre-defined. However in the sense of dissipative stability, this model is unstable, since the steady shear flow curves exhibit non-monotonic dependence on shear rate. Additionally in the flow regime of creep shear flow where the applied constant shear stress exceeds the maximum achievable value in the steady flow curves, the constitutive equations will possibly violate the positive definiteness of the orientation tensor and thus become Hadamard unstable.

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