

Recent results on the analysis of viscoelastic constitutive equations

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Abstract

Recent results obtained for the pom-pom model and the constitutive equations with time-strain separability are examined. The time-strain separability in viscoelastic systems is not a rule derived from fundamental principles but merely a hypothesis based on experimental phenomena, stress relaxation at long times. The violation of separability in the short-time response just after a step strain is also well understood (Archer, 1999). In constitutive modeling, time-strain separability has been extensively employed because of its theoretical simplicity and practical convenience. Here we present a simple analysis that verifies this hypothesis inevitably incurs mathematical inconsistency in the viewpoint of stability. Employing an asymptotic analysis, we show that both differential and integral constitutive equations based on time-strain separability are either Hadamard-type unstable or dissipative unstable. The conclusion drawn in this study is shown to be applicable to the Doi-Edwards model (with independent alignment approximation). Hence, the Hadamard-type instability of the Doi-Edwards model results from the time-strain separability in its formulation, and its remedy may lie in the transition mechanism from Rouse to reptational relaxation supposed by Doi and Edwards. 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 in the integral/differential form and also in the simplified differential type 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 these constitutive equations. It is proved that the differential model is globally Hadamard stable, and the integral model seems stable, as long as the orientation tensor remains positive definite or the smooth strain history in the flow is previously given. However cautious attention has to be paid when one employs the simplified version of the constitutive equations without arm withdrawal, since neglecting the arm withdrawal immediately yields Hadamard instability. 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 exhibit severe instability that the solution possesses strong discontinuity at the moment of change of chain dynamics mechanisms.

Keywords : time-strain separability, pom-pom model, Hadamard stability, dissipative stability, creep shear

1. Introduction

A rheological constitutive equation expresses the relation between deformation history and stress. Hence in its final form it cannot dispense with thermodynamic as well as continuum mechanical nature however rigorously it may be founded on the molecular physics.

A number of constitutive equations derived from either phenomenological or molecular theories are now available, and it is a tough task to sort among them to formulate an appropriate model equation for a given flow problem. Thus, it is necessary to construct some criteria on which such a choice should be based. Two distinct conditions

have been proposed based on mathematical stability, the Hadamard stability condition and the dissipative stability condition (Kwon and Leonov, 1995). The first of these conditions is related to the rapid elastic response, whereas the latter is associated with the dissipative viscous nature of the constitutive equations. Both conditions express the quality of the relation of rheological equations to the laws of thermodynamics. In this work, we review recent results of stability analysis performed on the constitutive models with time-strain separability and the pom-pom model.

The time-strain separability or factorability has been verified in experiments on the stress relaxation following a step strain, and is found to be valid especially in the long time region. The hypothesis has been widely applied in the formulation of rheological models and the analysis of data. In the case of separable single integral models, the sep-

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arability assumption remarkably simplifies the theoretical procedure to specify the functional term (the so-called “damping function”) introduced in the stress relaxation. In this formulation it is supposed that immediate (but arbitrary) generalization of the damping function to the 3D tensoric form is justifiable. This series of scientific process arrives at a form, which determines the complete viscoelastic constitutive equation merely from the shear stress relaxation experiment. However, the violation of the separability hypothesis in the rapid response has been observed in a number of experiments (Einaga *et al.*, 1971; Archer, 1999), and nowadays it is widely recognized that this hypothesis is invalid in the short time region. On the other hand, time-strain separable constitutive equations are still extensively employed in the modeling and analysis of viscoelastic flows, probably under the assumption that the short-time effect is negligible in the numerical computation and description of flow phenomena.

The theoretical simplicity afforded by the separability hypothesis has led to its frequent use in the formulation of rheological models. This is especially the case for integral constitutive equations, for which only the separable formulation has been employed for the description of viscoelastic flow. While separability can be explicitly expressed for integral equations, its direct implementation for differential models is not obvious. However, the equivalence of the upper-convected Maxwell model to the integral-type Lodge model suggests that the implicit inclusion of separability is possible for some differential models.

Recently McLeish and Larson (1998) proposed constitutive equations called the pom-pom model in order to account for complicated phenomena presumably exhibited by long side branches present in the LDPE molecules. They derived the equations based on reptation dynamics, introducing a simplified geometrical molecular structure named as a pom-pom molecule, for which the schematic illustration is represented in Fig. 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. Owing to its theoretical and also practical importance, these model equations draw many rheologists’ attention and quite a few results have already been reported concerning their appli-

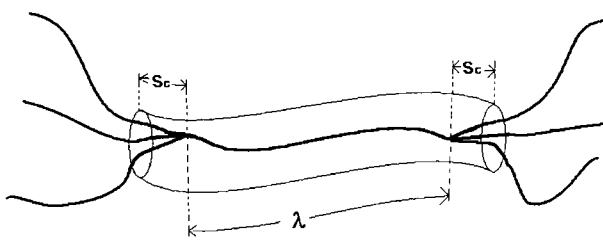


Fig. 1. Schematic representation of a three-armed pom-pom molecule ($q = 3$).

cations.

Regarding LDPE melt rheology, Rubio and Wagner (2000) compared the theoretical computation of the pom-pom model with experimental data, and found some qualitative disagreement. Inkson and coworkers (1999) also investigated the rheological behavior of LDPE including Melt 1, and concluded reasonable descriptive ability of the constitutive equations (the differential version with arm withdrawal length s_c neglected for simplicity) in some transient response of simple shear and extensional flows

2. Stability analysis of the time-strain separable integral models

In this study we consider the time-strain separable form of the Rivlin-Sawyers constitutive equation, because it explicitly expresses the factorability hypothesis:

$$\boldsymbol{\sigma} = -p\boldsymbol{\delta} + \int_{-\infty}^t m(t-t')[\varphi_1(I_1, I_2)\mathbf{C}^{-1} - \varphi_2(I_1, I_2)\mathbf{C}]dt', \quad (1)$$

where $\boldsymbol{\sigma}$ is the total stress tensor, p is the pressure, $\boldsymbol{\delta}$ is the unit tensor, $m(t-t') = \frac{dG(t-t')}{dt'}$, and is the relaxation modulus. \mathbf{C}^{-1} and \mathbf{C} are the total Finger and Cauchy strain tensors, respectively. φ_1 and φ_2 , which should be specified for each constitutive model, are functions of the basic invariants I_n ($n=1, 2, 3$) of \mathbf{C}^{-1} , defined as

$$I_1 = \text{tr}\mathbf{C}^{-1}, \quad I_2 = \text{tr}\mathbf{C}, \quad I_3 = \det\mathbf{C}^{-1} = 1. \quad (2)$$

The second and third identities represent the consequence of fluid incompressibility.

From the class of equations described above we confine our study to the following hyper-viscoelastic type (all the nonhyper-viscoelastic models such as the Wagner and Papanastasiou models are proved Hadamard unstable (Kwon and Leonov, 1995)), which in turn becomes the K-BKZ class of constitutive equations:

$$\varphi_1 = \frac{\partial U}{\partial I_1}, \quad \varphi_2 = \frac{\partial U}{\partial I_2}, \quad (3)$$

$$\varphi_{12} = \varphi_{21} \quad \left(\varphi_{ij} = \frac{\partial^2 U}{\partial I_i \partial I_j} \right), \quad (4)$$

where U is the elastic potential. Denoting \mathbf{C}^{-1} as \mathbf{c} , $\mathbf{C} = \mathbf{c}^{-2} - I_1\mathbf{c} + I_2\boldsymbol{\delta}$ can be substituted into Eq. (1), giving

$$\begin{aligned} \boldsymbol{\sigma} &= -p'\boldsymbol{\delta} + \int_{-\infty}^t m(t-t')[\varphi_1\mathbf{c} + \varphi_2(I_1\mathbf{c} - \mathbf{c}^2) + \varphi_3I_3\boldsymbol{\delta}]dt', \\ &= -p'\boldsymbol{\delta} + \boldsymbol{\tau} \quad (\varphi_3 = 0) \end{aligned} \quad (5)$$

Here $p' = p + \int_{-\infty}^t m(t-t')\varphi_2I_2dt'$, and the term $\varphi_3 \equiv \frac{\partial U}{\partial I_3}$ plays

a role in the compressible fluid flow but vanishes here in the incompressible case. The strain tensor \mathbf{c} satisfies the following evolution equation and the initial condition:

$$\nabla \cdot \frac{d\mathbf{c}}{dt} - (\nabla \mathbf{v})^T \cdot \mathbf{c} - \mathbf{c} \cdot (\nabla \mathbf{v}) = 0 \quad (6)$$

$$c|_{t=t'} = \delta,$$

where ∇ denotes the upper convected time derivative of \mathbf{c} . The constitutive equation (1) or (5) under the condition given in Eq. (6) is combined with the following equation of motion and the continuity equation to solve the flow problem:

$$\rho \frac{d\mathbf{v}}{dt} = \nabla \cdot \boldsymbol{\sigma} \quad (7)$$

$$\nabla \cdot \mathbf{v} = 0. \quad (8)$$

2.1. Hadamard-type stability

To verify the mathematical stability of the above constitutive equations in the Hadamard sense, we first follow the procedure employed by Kwon and Leonov (1995). Hadamard-type stability signifies the stability of equations (originally differential equations) under high frequency short wave disturbances. First, assume that τ_0 , c_0 , v_0 and p_0 are basic solutions of Eqs. (5-8) in an incompressible flow (i.e., $\phi_3=0$ and $I_3=1$), and then upon these variables we impose the following short wave disturbance with high frequency:

$$\{\tau, c, v, p'\} = \{\tau_0, c_0, v_0, p_0\} + \varepsilon \{\tilde{\tau}, \tilde{c}, \tilde{v}, \tilde{p}\} \exp\left[\frac{i(\mathbf{k} \cdot \mathbf{x} - \omega t)}{\varepsilon^2}\right]. \quad (9)$$

Here $\{\tilde{\tau}, \tilde{c}, \tilde{v}, \tilde{p}\}$ are the amplitudes of the corresponding variables, \mathbf{k} is the wave vector, \mathbf{x} is the position vector, ω is the angular velocity, and ε is a small amplitude parameter ($|\varepsilon| \ll 1$) that also accounts for the high frequency and short wavelength of the applied disturbance.

Substitution of Eq. (9) into Eqs. (5)-(8) yields

$$\rho \Omega^2 \tilde{v}^2 = \tilde{v}_i^* k_j \tilde{v}_p k_q \int_{-\infty}^t m(t-t') B_{ijpq} dt' \quad (10)$$

where details of the derivation can be found in the reference (Kwon and Leonov, 1995) and

$$\tilde{v}^2 = \tilde{v}^* \cdot \tilde{v} \quad (\tilde{v}^* : \text{the complex conjugate of } \tilde{v}) \quad (11)$$

$$B_{ijpq} = \varphi_1 \delta_{ip} c_{jq} + \varphi_2 (I_1 \delta_{ip} c_{jq} - \delta_{ip} c_{j\alpha} c_{js} c_{sq} - c_{ip} c_{jq} + c_{ij} c_{pq}) + 2\varphi_{11} c_{ij} c_{pq} + 2\varphi_{12} [(I_1 c_{ij} - c_{is} c_{sj}) c_{pq} + c_{ij} (I_1 c_{pq} - c_{ps} c_{sq})] + 2\varphi_{22} (I_1 c_{ij} - c_{is} c_{sj}) (I_1 c_{pq} - c_{ps} c_{sq}). \quad (12)$$

The necessary and sufficient condition for stability therefore becomes

$$\mathbf{B} = B_{ijpq} \tilde{v}_i^* k_j \tilde{v}_p k_q > 0. \quad (13)$$

If we confine the analysis to the real plane, the inequality (18) can be rewritten as

$$\mathbf{B} = B_{ijpq} \tilde{v}_i k_j \tilde{v}_p k_q > 0. \quad (14)$$

To obtain the simplest necessary condition for stability, we assume the following asymptotic dependence of the potential:

$$U \approx a(\sqrt{I_1})^\alpha (\sqrt{I_2})^\beta \text{ as } I_1 \rightarrow \infty \quad \left(\tilde{\varphi}_i = \frac{\partial U}{\partial \sqrt{I_i}}, \quad \tilde{\varphi}_{ij} = \frac{\partial^2 U}{\partial \sqrt{I_i} \partial \sqrt{I_j}} \right)$$

(and then $I_2 \rightarrow \infty, a > 0$). (15)

In the asymptotic limit shown in Eq. (15), the derivatives of the potential become

$$\begin{aligned} \tilde{\varphi}_1 &\approx a\alpha(\sqrt{I_1})^{\alpha-1}(\sqrt{I_2})^\beta, & \tilde{\varphi}_2 &\approx a\beta(\sqrt{I_1})^\alpha(\sqrt{I_2})^{\beta-1} \\ \tilde{\varphi}_{11} &\approx a\alpha(\alpha-1)(\sqrt{I_1})^{\alpha-2}(\sqrt{I_2})^\beta, & \tilde{\varphi}_{12} &\approx a\alpha\beta(\sqrt{I_1})^{\alpha-1}(\sqrt{I_2})^{\beta-1} \\ \tilde{\varphi}_{22} &\approx a\beta(\beta-1)(\sqrt{I_1})^\alpha(\sqrt{I_2})^{\beta-2}. \end{aligned} \quad (16)$$

Then the inequality $\mathbf{B} > 0$ is in turn written as

$$\begin{aligned} &\alpha(\sqrt{I_1})^{\alpha-4}(\sqrt{I_2})^\beta [c_{vv}c_{kk} - c_{vk}^2 + c_{kk}^2 + c_{kk}c_{nn} + (\alpha-1)c_{vk}^2 \\ &- \beta \left(\frac{\sqrt{I_1}}{\sqrt{I_2}}\right)^2 c_{vk}c_{vk}^{-1}] + \beta(\sqrt{I_1})^\alpha(\sqrt{I_2})^{\beta-4} [(c_{vv}^{-1})^2 + (c_{vv}^{-1})(c_{kk}^{-1}) - (c_{vk}^{-1})^2 \\ &+ (c_{vv}^{-1})(c_{nn}^{-1}) + (\beta-1)(c_{vk}^{-1})^2 - \alpha \left(\frac{\sqrt{I_1}}{\sqrt{I_2}}\right)^2 c_{vk}c_{vk}^{-1}] > 0. \end{aligned} \quad (17)$$

To obtain the simplest necessary condition for the Hadamard-type stability, we hereafter consider only the case of simple shear flow. In this case the Finger tensor \mathbf{c} and its inverse \mathbf{c}^{-1} are expressed in terms of the shear strain γ as follows:

$$[c] = \begin{bmatrix} 1 + \gamma^2 & \gamma & 0 \\ \gamma & 1 & 0 \\ 0 & 0 & 1 \end{bmatrix}, \quad [c^{-1}] = \begin{bmatrix} 1 & -\gamma & 0 \\ -\gamma & 1 + \gamma^2 & 0 \\ 0 & 0 & 1 \end{bmatrix}$$

$$I_1 = I_2 = \gamma^2 + 3 \equiv I. \quad (18)$$

Again, $\mathbf{B} > 0$ becomes

$$\begin{aligned} &\alpha(\sqrt{I})^{\alpha+\beta-4} [1 + \gamma^2 - \gamma^2 + 1 + 1 + (\alpha-1)\gamma^2 + \beta\gamma^2] \\ &+ \beta(\sqrt{I})^{\alpha+\beta-4} [1 + 1 + \gamma^2 - \gamma^2 + 1 + (\beta-1)\gamma^2 + a\gamma^2] > 0. \end{aligned}$$

Eventually we obtain

$$(\alpha + \beta) [3 + (\alpha + \beta - 1)\gamma^2] > 0. \quad (19)$$

Inequality (19) thus yields $\alpha + \beta > 1$ as the simplest necessary condition for a stable constitutive equation.

The analysis can also be performed with respect to the damping function $h(\gamma)$. Consider a step shear experiment in which the shear stress is expressed as

$$\tau_{12}(\gamma, t) = \gamma h(\gamma) G(t). \quad (20)$$

$G(t)$ is the relaxation modulus. In the perfect elastic limit

(i.e., when $t \rightarrow 0+$),

$$\tau_{12}(\gamma) = \gamma h(\gamma) G(0) = \gamma h(\gamma) \sum_i G_i \quad (21)$$

Here G_i is the constant modulus of the i -th Maxwellian spring in multimode representation. For stability, τ_{12} or $\gamma h(\gamma)$ should be monotonically increasing with respect to γ (Simhambhatla, 1994). In conjunction with our analysis, we again assume the following asymptotic dependence:

$$h(\gamma) \approx \kappa \gamma^\mu \quad \text{or} \quad \tau_{12} \approx \kappa \gamma^{\mu+1} \quad \text{as} \quad \gamma \rightarrow \infty$$

leading to the following criterion for stability

$$\frac{d\tau_{12}}{d\gamma} \approx (\mu+1) \kappa \gamma^\mu > 0. \quad (22)$$

Thus μ must be greater than -1 for stability. In the case of the K-BKZ equation (or hyper-viscoelasticity),

$$\varphi_1 + \varphi_2 = h(\gamma) \quad (23)$$

From Eqs. (15) and (16), we have

$$\frac{1}{2\sqrt{I_1}} \tilde{\varphi}_1 + \frac{1}{2\sqrt{I_2}} \tilde{\varphi}_2 = h(\gamma) \xrightarrow{\gamma \rightarrow \infty} \frac{a(\alpha+\beta)}{2\sqrt{\gamma^2+3}} (\sqrt{\gamma^2+3})^{\alpha+\beta-1} \approx \kappa \gamma^\mu \quad (24)$$

Asymptotically, the following relation among α , β and μ can be shown:

$$\frac{a(\alpha+\beta)}{2} \gamma^{\alpha+\beta-2} \approx \kappa \gamma^\mu \Rightarrow \mu = \alpha + \beta - 2 \quad (25)$$

Because $\mu > -1$ for stability, this again results in $\alpha + \beta > 1$, which exactly coincides with the condition in Eq. (19).

2.2. Criterion I of dissipative stability: Integral model

In terms of a thermodynamical description, basic functionals such as the free energy F , extra stress $\boldsymbol{\tau}$ and dissipation D for the hyper-viscoelastic separable integral models are of the form (Kwon and Leonov, 1994):

$$\begin{aligned} F &= \int_{-\infty}^t m(t-t') U(I_1, I_2; t, t') dt' \\ \boldsymbol{\tau} &= \int_{-\infty}^t m(t-t') \mathbf{c} \cdot \frac{\partial U}{\partial \mathbf{c}}(I_1, I_2; t, t') dt' \\ D &= \text{tr}(\boldsymbol{\tau} \cdot \mathbf{e}) - \frac{dF}{dt} = \int_{-\infty}^t \frac{dm(t-t')}{dt'} U(I_1, I_2; t, t') dt' \end{aligned} \quad (26)$$

Here $\mathbf{e} = \frac{1}{2}(\nabla \mathbf{v} + \nabla \mathbf{v}^T)$ is the strain rate tensor. Another stability theorem called the ‘‘dissipative stability criterion I’’ has been proven by Kwon and Leonov (1994). This theorem relates stability to the boundedness of the stress (or other variables such as the free energy and dissipation

functionals) in any regular pre-defined time-dependent flow, and is rewritten below.

Criterion I of dissipative stability (single integral separable models)

In any regular flow, the functionals of free energy and dissipation (26) are bounded if and only if the thermodynamically or Hadamard stable potential function, $U(H_1, H_2, H_3)$ expressed in terms of the principal Hencky strains, H_k , increases more slowly than exponentially.

In the above theorem the principal Hencky strain measure H_i and the Finger strain c_i are related by

$$H_i = \frac{1}{2} \ln c_i. \quad (27)$$

From the result obtained in the previous section, the Hadamard stable constitutive equation is expressed in the asymptotic limit as

$$\begin{aligned} U &\approx a(\sqrt{I_1})^\alpha (\sqrt{I_1})^\beta \quad \text{and} \quad \alpha + \beta > 1 \quad \text{as} \quad I_1 \rightarrow \infty \quad \text{or} \quad I_2 \rightarrow \infty \\ \Rightarrow \hat{U} &\approx a(\sqrt{e^{2H_1} + e^{2H_2} + e^{2H_3}})^\alpha (\sqrt{e^{-2H_1} + e^{-2H_2} + e^{-2H_3}})^\beta \end{aligned} \quad (28)$$

where from incompressibility $H_1 + H_2 + H_3 = 0$. It is clear from expression (28) that the elastic potential U under the stability constraint $\alpha + \beta > 1$ increases at the speed of an exponential function (or faster) in terms of the Hencky strain, as illustrated below.

Without loss of generality, we may assume

$$H_1 \rightarrow \infty \quad \text{as} \quad I_1 \text{ (or } I_2) \rightarrow \infty.$$

Then

$$\begin{aligned} \hat{U} &\approx a e^{\alpha H_1} (\sqrt{e^{2(H_1+H_3)} + e^{2(H_1+H_2)}})^\beta = a e^{(\alpha+\beta)H_1} (\sqrt{e^{2H_2} + e^{2H_3}})^\beta \\ &\geq 2^{\frac{\beta}{2}} a e^{(\alpha+\beta)H_1} e^{-\frac{\beta}{2}H_1} = 2^{\frac{\beta}{2}} a e^{\frac{\alpha}{2}H_1} e^{\frac{\alpha+\beta}{2}H_1}, \quad \text{as} \quad H_1 \rightarrow \infty. \end{aligned} \quad (29)$$

Since the Hadamard-type stability condition states that $\alpha + \beta > 1$, and the Baker-Ericksen inequality (Truesdell and Noll, 1992) requires that $\alpha \geq 0$ and $\beta \geq 0$ for evolutionarity, the Hadamard stable potential U always increases exponentially or faster in terms of the Hencky strain measure. Hence the following conclusion can be drawn:

The time-strain separable single integral constitutive equations for viscoelastic fluids that are Hadamard type stable are always dissipative unstable. In other words, the time-strain separable single integral models are ill-posed either in the Hadamard or in the dissipative sense.

One example of an instability incurred by the hypothesis of time-strain separability is illustrated in Fig. 2, drawn according to the damping function derived by Wagner and Meissner (1980) on the basis of experimental data (Laun, 1978). Because $\gamma h(\gamma)$ is directly proportional to the shear stress, the decreasing branch of the curve after the maximum is Hadamard unstable, proven previously (Simham-

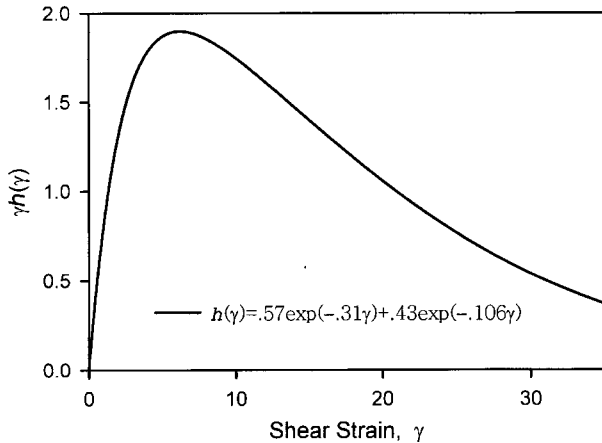


Fig. 2. Behavior of the damping function for the Wagner model [Wagner and Meissner (1980)] in stress relaxation following step shear.

bhatla, 1994; Leonov, 1999). It can therefore be conjectured that the material itself shows this kind of instability right after the large step shear, because the experimental points display the same decreasing response after the maximum. However, this decrease in the experimental points is an artifact induced by the hypothetical time-strain separability, because in the short time region (i.e., immediately after step strain) the experimental behavior definitely deviates (and should deviate if there exists no phase change) from the master curve created under the hypothesis (Einaga *et al.*, 1971). Consequently, all the separable constitutive equations that can appropriately describe the behavior of stress relaxation (except in the short time region) seem to violate the Hadamard stability condition, because they show behavior qualitatively similar to the curve depicted in Fig. 1 (Simhambhatla, 1994).

A similar argument may be applied to the Doi-Edwards constitutive equation. In this section, the analysis has been carried out for the kernel in Eq. (1), i.e., the function (except $m(t-t')$) inside the integral dependent upon the Finger strain c or C^{-1} , but the direct application of the current result to the Doi-Edwards model is not possible since the model equations are represented in terms of the so-called geometric universal tensor Q (Doi and Edwards, 1986). However, Q is related to C^{-1} at least implicitly through some evolution equation imposed on the deformation gradient E and its configurational space averaging; thus the conclusion drawn above regarding the stability of constitutive equations can be applied to the original Doi-Edwards model with the independent alignment approximation. The instability shown in this model, which has been demonstrated previously (Kwon, 1999), possibly results from the time-strain separability present in the model equation. This irrelevance of the Doi-Edwards model in the short time response upon large strain may be

closely associated with the fact that the short time behavior follows the Rouse relaxation mechanism even in the reptation theory (one can find detailed discussion in the paper by Archer (1999)).

3. Remark on differential models with separability in the stress relaxation

The dissipative stability criterion I for the Maxwell-like differential constitutive equations, which is equivalent to the criterion mentioned in the previous section, was proved by Leonov (1992). This criterion is formulated as follows. Criterion I of dissipative stability (Maxwell-like differential models)

Consider the set of upper convected Maxwell-like models with positive dissipation D . Let the free energy F be a non-decreasing smooth function of invariants I_k . If for any positive number, E , the asymptotic inequality

$$D > E \cdot |\sigma_d| \text{ as } |c| \rightarrow \infty \quad (|b| \equiv (\text{tr } b^2)^{1/2})$$

holds, then in any regular flow the configuration tensor c and the stress tensor σ_e are bounded.

In the above theorem, $\sigma_e \equiv Gc$, G is the constant modulus, the invariants I_k are defined as

$$I_1 = \text{tr } c, \quad I_2 = \frac{1}{2}(I_1^2 - \text{tr } c^2),$$

and all other definitions can be found in the reference (Kwon and Leonov, 1995).

Because the differential constitutive equations cannot express the time-strain separability explicitly, in the present analysis we consider only the case of stress relaxation where its explicit representation is possible. In the stress relaxation after some arbitrary step strain or after cessation of any flow field at $t = 0$, the following equations hold

$$v = 0, \text{ and } \nabla v = 0 \text{ at } t \geq 0.$$

Using these equations, the Johnson-Segalman, upper-convected Maxwell and White-Metzner models (the differential models that are time-strain separable at least in stress relaxation) become

$$\dot{\tau} + \frac{1}{\theta}\tau = 0, \quad \tau = G(c - \delta) \tag{30}$$

where θ is the relaxation time. This differential equation can be readily solved, resulting in the solution $\tau = \tau_0 \exp\left(-\frac{t}{\theta}\right)$, where $\tau_0 = \tau(t=0)$ is constant. The dissipation and the magnitude of the stress for these models are

$$D = \frac{G}{2\theta}(I_1 - 3), \quad |\sigma_d| = G|c| = G(I_1^2 - 2I_2)^{1/2}. \tag{31}$$

Hence, the criterion I is violated when $|c| \rightarrow \infty$ (Leonov, 1992). This explains the consequence that the Johnson-

Segalman, upper-convected Maxwell and White-Metzner models all exhibit an unbounded stress response in uniaxial elongational flow when the extensional strain rate exceeds a certain limit (e.g., for the upper-convected Maxwell model the critical strain rate is half of the reciprocal relaxation time). We can also conclude that the dissipative unstable behavior of these constitutive equations occurs due to the time-strain separability (in stress relaxation) inherent in their mathematical form.

The separability $\tau = \tau_0 \exp(-t/\theta)$ in the stress relaxation of the differential equations originates from the linearity of the dissipative term (the term containing the relaxation time in Eq. (30)) with respect to the stress τ . Hence, stable constitutive modeling requires some complicated functional form for the dissipative term rather than a linear function of the stress. It is also worth mentioning that the Johnson-Segalman and White-Metzner models are also Hadamard unstable.

Formulation of integral viscoelastic models that preserves both stability requirements necessitates consideration of a general functional form (Truesdell and Noll, 1992), rather than Eq. (1). Little work has been done in this area of viscoelastic flow analysis, however, probably due to the mathematical complexity or ambiguity of such functional forms. In a practical sense, the differential type with nonlinear dissipative terms surpasses the integral form because previous results show that 3 differential equations are globally stable, whereas no integral models are stable (Kwon and Leonov, 1995). In addition to the models presented above, there also exists a mixed-type formulation of non-separable integral/differential constitutive equations, referred to as the ‘‘pom-pom model’’ (McLeish and Larson, 1998), of which the mathematical stability is discussed in the following.

4. Analysis of 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 under the assumption that a melt consists of identical molecules with a very simplified branching structure, called pom-pom molecules (McLeish and Larson, 1998). A typical pom-pom molecule illustrated in Fig. 1 is composed of two identical q -armed stars connected by a backbone section that pursues hypothetical reptational motion. By McLeish and Larson, the dynamics of this pom-pom molecule is suggested as the simplest analog of the motion of the real polymeric molecule with long branching like LDPE.

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:

$$\boldsymbol{\sigma} = p\boldsymbol{\delta} + G \left(\lambda^2 + \frac{1}{\phi_b} \frac{2qs_c}{2qs_a + s_b} \right) \mathbf{S}, \quad G = \frac{15}{4} G_0 \phi_b^2, \quad \phi_b = \frac{s_b}{2qs_a + s_b} \quad (32)$$

Here $\boldsymbol{\sigma}$ is the total stress tensor, $\boldsymbol{\delta}$ the unit tensor, p the isotropic pressure, \mathbf{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 that also corresponds to the maximum stretch ratio of the backbone, 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 (Fig. 1), and thus both are functions of time.

Backbone orientation:

$$\mathbf{S} = \int_{-\infty}^t \frac{1}{\tau_b(t_1)} \exp\left(-\int_{t_1}^t \frac{dt'}{\tau_b(t')}\right) \mathbf{Q}^{IA} dt_1, \quad \mathbf{Q}^{IA} = \left\langle \frac{\mathbf{u}\mathbf{u}'}{(\mathbf{u}')^2} \right\rangle_0, \\ \mathbf{u}' = \mathbf{E}(t-t_1) \cdot \mathbf{u}, \quad \frac{d\mathbf{E}}{dt} = \nabla \mathbf{v}^T \cdot \mathbf{E} \quad (33)$$

In these equations, the strain measure \mathbf{Q}^{IA} is slightly modified from the original one to a universal strain of the Doi-Edwards model with independent alignment approximation that has been employed by Rubio and Wagner (2000) in their study. is the relaxation time of backbone orientation, \mathbf{u} the unit vector, \mathbf{E} the deformation gradient tensor, \mathbf{v} the velocity, ∇ the gradient operator, $\nabla \mathbf{v}^T$ the transpose of the velocity gradient, and the symbol $\langle \rangle_0$ is the operation of averaging over the configuration space.

Backbone stretch:

$$\frac{d\lambda}{dt} = \lambda \nabla \mathbf{v}^T : \mathbf{S} - \frac{1}{\tau_s} (\lambda - 1) \quad \text{for } \lambda < q. \quad (34)$$

Here τ_s is the characteristic time of backbone stretch. The above equation is valid only for $\lambda < q$ and even if Eq.(34) 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:

$$\frac{ds_c}{dt} = \left(q \frac{s_b}{2} + s_c \right) \nabla \mathbf{v}^T : \mathbf{S} - \frac{1}{2\tau_a} \quad \text{for } \lambda = q. \quad (35)$$

Here τ_a is the characteristic time of arm relaxation.

Characteristic time for backbone orientation:

$$\tau_b = \frac{4}{\pi} s_b^2 \phi_b \tau_a q. \quad (36)$$

Characteristic time for arm relaxation:

$$\tau_a = \tau_0 \exp \left[\frac{15}{4} s_a \left\{ \frac{(1-x)^2}{2} - (1-\phi_b) \frac{(1-x)^3}{3} \right\} \right], \quad x = \frac{s_c}{s_a} \quad (37)$$

Characteristic time for backbone stretch:

$$\tau_s = s_b \tau_a(0) q. \quad (38)$$

In the preceding equations for characteristic times, τ_a and τ_b are dependent upon s_c and thus are 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, 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 Eqs.(33) of backbone orientation:

$$\overset{\vee}{c} + \frac{1}{\tau_b} (c - \delta) = 0, \quad S = \frac{c}{tr \ c} \quad (39)$$

Here $\overset{\vee}{c} = \frac{dc}{dt} - \nabla v^T \cdot c - c \cdot \nabla v$ is the upper-convected time derivative of the configuration tensor c , and all the other equations from (32) to (38) except (33) are kept for the complete set. Note that the evolution equation (39) 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 (1995)).

4.1. Stability analysis of the differential model

The Hadamard stability accounts for the elastic properties of viscoelastic constitutive equations related to fast responses such as type of differential operator and elastic free energy (Kwon and Leonov, 1995). 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. Upon the total set of equations, we impose such short and high frequency wave disturbances as

$$\begin{aligned} \{ \sigma, S, c, v, \lambda, p, s_c \} &= \{ \sigma_0, S_0, c_0, v_0, \lambda_0, p_0, s_{c0} \} \\ &+ \{ \delta \sigma, \delta S, \delta c, \delta v, \delta \lambda, \delta p, \delta s_c \}, \\ \{ \delta \sigma, \delta S, \delta c, \delta v, \delta \lambda, \delta p, \delta s_c \} &= \varepsilon \{ \hat{\sigma}, \hat{S}, \hat{c}, \hat{v}, \hat{\lambda}, \hat{p}, \hat{s}_c \} \times \\ \exp[i(\mathbf{k} \cdot \mathbf{x} - \omega t) / \varepsilon^2]. \end{aligned} \quad (40)$$

Here $\{ \sigma_0, S_0, c_0, v_0, \lambda_0, p_0, s_{c0} \}$ and $\{ \delta \sigma, \delta S, \delta c, \delta v, \delta \lambda, \delta p, \delta s_c \}$ 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. $\{ \hat{\sigma}, \hat{S}, \hat{c}, \hat{v}, \hat{\lambda}, \hat{p}, \hat{s}_c \}$ is the amplitude of disturbing wave, \mathbf{k} the wave vector, the frequency and ε is the small amplitude parameter that also expresses the short wavelength as well as high frequency of the disturbing wave.

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 becomes identically zero and Eq.(35) does not play any role. In this case, the relaxation time τ_b is constant and in the perturbed system (40) 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 Eq.(35) 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 (1999) in order 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 $\lambda = q$.

4.1.1. Hadamard stability analysis when $\lambda = q$

Since s_c and its perturbation vanish, we disturb Eqs.(32), (34), (38) and continuity and momentum equations according to Eq.(40):

$$\begin{aligned} \rho \Omega^2 \hat{v}_j \hat{v}_j &= G \lambda^2 \delta_{ij} S_{mn} \hat{v}_i \hat{v}_j k_m k_n \\ \Rightarrow \rho \Omega^2 &= G \lambda^2 S_{mn} k_m k_n \end{aligned} \quad (41)$$

Here $\Omega = \omega - \mathbf{k} \cdot \mathbf{v}$ is the frequency with Dopplers shift on the basic flow field \mathbf{v} . Since the frequency ω and thus Ω should be real-valued for stability, the necessary and sufficient condition of the Hadamard stability becomes

$$G \lambda^2 S_{mn} k_m k_n > 0, \quad (42)$$

which exactly requires the positive definiteness of the second rank tensor S . Due to the relation (39), the positive definiteness of S is equivalent to that of c .

Regarding the positive definiteness of the configuration tensor c , Hulslen (1990) and Leonov (1992) independently proved one theorem in some limited flow situation. In that theorem, it is stated that for any given piecewise smooth strain history with the initial condition $c = \delta$ the principal values of tensor 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 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 c (Kwon and Leonov, 1992).

4.1.2. Hadamard stability analysis when $\lambda = q$ (s_c neglected)

When the backbone reaches its maximum stretch, λ becomes a constant q , and thus its disturbance vanishes. Therefore substitution of 0 for $\hat{\lambda}$ yield the following dispersion relation and the stability condition:

$$\begin{aligned} \rho \Omega^2 \hat{v}_j \hat{v}_j &= G q^2 (\delta_{ij} S_{mn} - 2 S_{im} S_{jn}) \hat{v}_i \hat{v}_j k_m k_n \\ \Rightarrow G q^2 \left(\frac{1}{I_1} \delta_{im} c_{jn} - \frac{2}{I_1^2} c_{ij} c_{mn} \right) \hat{v}_i \hat{v}_j k_m k_n. \end{aligned} \quad (43)$$

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

$$\begin{aligned} \overset{\nabla}{c} + \frac{1}{\tau_b} (c - \delta) &= 0, \quad \frac{2\rho}{G} F \equiv U = q^2 \ln I_1, \quad I_1 = \text{tr } c, \\ \sigma &= G \{ \varphi_1 c + \varphi_2 (I_1 c - c^2) + \varphi_3 I_3 \delta \} = -p \delta + G q^2 \frac{c}{I_1}, \quad \varphi_j = \frac{\partial U}{\partial I_j}, \\ I_1 &= \text{tr } c, \quad I_2 = \frac{1}{2} (I_1^2 - \text{tr } c^2), \quad I_3 = \det c. \end{aligned} \quad (44)$$

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

$$\varphi_1 = \frac{q^2}{I_1}, \quad \varphi_2 = \varphi_3 = 0, \quad \varphi_{11} = -\frac{q^2}{I_1^2}, \quad \varphi_{12} = \varphi_{21} = \varphi_{22} = 0, \quad (45)$$

where $\varphi_{ij} = \frac{\partial^2 U}{\partial I_i \partial I_j}$. If one substitutes Eqs.(45) into the following inequality (46) obtained in reference (Kwon 1994), one can immediately find that it results in the condition (43):

$$\begin{aligned} B &= B_{ijmn} \hat{v}_i \hat{v}_j \hat{v}_m \hat{v}_n \\ &= [\varphi_1 \delta_{im} c_{jn} + \varphi_2 (I_1 \delta_{im} c_{jn} - \delta_{im} c_{ja} c_{an} - c_{im} c_{jn} + c_{ij} c_{mn}) \\ &\quad + 2\varphi_{11} c_{ij} c_{mn} + 2\varphi_{12} c_{ij} (I_1 c_{mn} - c_{ma} c_{an}) + 2\varphi_{21} (I_1 c_{ij} - c_{ia} c_{aj}) c_{mn} \\ &\quad + 2\varphi_{22} (I_1 c_{ij} - c_{ia} c_{aj}) (I_1 c_{mn} - c_{ma} c_{an})] \hat{v}_i \hat{v}_j \hat{v}_m \hat{v}_n > 0. \end{aligned} \quad (46)$$

After rewriting the constitutive relation as Eqs.(44), we may directly apply the following necessary and sufficient condition for Hadamard stability proved in the paper (Kwon and Leonov, 1995):

- (i) $\beta_i > 0$,
 - (ii) $w_i + 2\beta_i \sqrt{c_j c_k} > 0 \quad (i \neq j \neq k)$,
 - (iii) $\{ (w_i + 2\beta_i \sqrt{c_j c_k})^{1/2} + (w_j + 2\beta_j \sqrt{c_i c_k})^{1/2} \}^2 > w_k - 2\beta_k \sqrt{c_i c_j} \quad (i \neq j \neq k)$,
- (47)

where c_i is the eigenvalue of the tensor c and

$$\begin{aligned} \beta_i &= \varphi_1 + \varphi_2 c_i \\ w_i &= (I_1 - c_i) \beta_i + 2(I_1^2 - 2I_2 - c_i^2 - 2I_3/c_i) \{ \varphi_{11} + (\varphi_{12} + \varphi_{21}) c_i + \varphi_{22} c_i^2 \} \end{aligned} \quad (48)$$

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 c valid at the moment of step strain γ become

$$\begin{aligned} c_1 &= c, \quad c_2 = 1/c, \quad c_3 = 1, \quad c = (\gamma^2 + 2 + \sqrt{\gamma^4 + 4\gamma^2})/2, \\ I_1 &= I_2 = \gamma^2 + 3. \end{aligned} \quad (49)$$

Then the second inequality in (47) for $i=1$ reduces to

$$\sqrt{c}(\gamma^2 + 3)(\gamma^2 + 3 - c) - 2\sqrt{c}(\gamma^4 + 4\gamma^2 + 3 - c^2 - 2/c) + 2(\gamma^2 + 3) > 0, \quad (50)$$

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

$$\begin{aligned} (\gamma^2 + 3)(\gamma^2 + 3 - 1/c) - 2(\gamma^4 + 4\gamma^2 + 3 - 2c - 1/c^2) + 2\sqrt{c}(\gamma^2 + 3) > 0 \\ \text{and } -\gamma^4 - \gamma^2 + 12 > 0. \end{aligned} \quad (51)$$

The latter of inequalities (51) 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.* (1999) to be located in the unstable solution branch when the strain rate is high.

4.1.3. complete Hadamard 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 fixed. The stress relation and the evolution equation for arm withdrawal under the disturbance yield the following final form

$$\rho \Omega^2 \hat{v}_j \hat{v}_j = G q \left(q + 2 \frac{s_c}{s_b} \right) (\delta_{ij} S_{mn} - S_{im} S_{jn}) \hat{v}_i \hat{v}_j k_m k_n, \quad (52)$$

which has to be positive for stability. In the tensor c representation, due to positivity of G , q , s_c , s_b and I_1 , the inequality imposed on (24) can be rewritten as

$$\begin{aligned} \left(\delta_{ij} \frac{c_{mn}}{I_1} - \frac{c_{im} c_{jn}}{I_1 I_1} \right) \hat{v}_i \hat{v}_j k_m k_n > 0 \\ \Rightarrow \left(\frac{1}{\sqrt{I_1}} \delta_{im} c_{jn} - \frac{1}{I_1^{3/2}} c_{ij} c_{mn} \right) \hat{v}_i \hat{v}_j \hat{v}_m \hat{v}_n > 0. \end{aligned} \quad (53)$$

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 preceding inequality (46). Then corresponding potential equivalent for the constitutive equations in this particular stability problem becomes

$$U = 2\sqrt{I_1}. \tag{54}$$

At this point, we can apply the Renardy's stability condition (1985) 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 Renardys condition is also sufficient for Hadamard stability when it is applied to differential Maxwell-like models with an upper convected derivative (Kwon, 1994).

Since the equivalent potential (54) clearly satisfies the Renardys 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.*

4.1.4. dissipative instability in creep shear flow

According to the preceding results on stability in this work, the differential 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 flow situation where possibly the positive definiteness can be violated or another type of instability rather than Hadamard-type can occur.

In the steady state of simple shear flow, the constitutive equations (32), (34) and (39) reduce to

$$c_{11} = 1 + 2\Gamma^2, \quad c_{12} = \Gamma, \quad c_{22} = c_{33} = 1, \quad c_{13} = c_{23} = 0, \\ \lambda = \left[1 - \frac{\tau_s}{\tau_b(0)} \frac{\Gamma^2}{3 + 2\Gamma^2} \right]^{-1}, \quad \hat{\sigma}_{12} \equiv \sigma_{12}/G = \lambda^2 \frac{\Gamma}{3 + 2\Gamma^2} \quad (\lambda < q), \tag{55}$$

where the dimensionless shear rate is defined as $\Gamma = \tau_b(0) \dot{\gamma}$ and $\tau_b(0)$ is the value of τ_b at $x = s_c = 0$. From the above equations the ratio between the relaxation times $\tau_b(0)/\tau_s$ should exceed the value of 1/2 to avoid singularity. In Fig. 3, the behavior between the dimensionless variables Γ and $\hat{\sigma}_{12}$ is shown for several values of $\tau_b(0)/\tau_s$, and all the curves are valid when $q \geq 2$, since for those values of $\tau_b(0)/\tau_s$ λ never exceeds 2. All show maxima and then decreasing branches of solution, and the achievable shear stresses are all bounded below the maximum. Here we can directly apply the theorem derived in the paper (Kwon and Leonov, 1995) that asserts so called dissipative instability. It is evident first that mechanically and thermodynamically, the decreasing branch of the flow curve in Fig.2 is the unstable solution. Another type of severe blow-up insta-

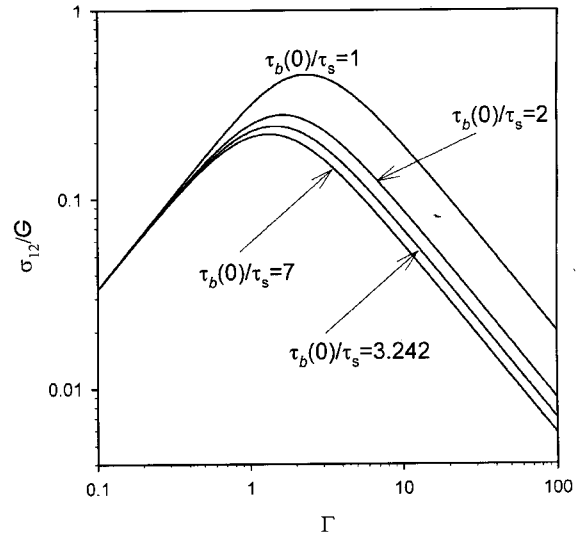


Fig. 3. The dimensionless shear stress σ_{12}/G vs. dimensionless shear rate Γ in steady simple shear flow of the differential pom-pom constitutive equations for various values of the ratio between relaxation times.

bility exhibited by the dissipative unstable constitutive models has been exposed in reference (Kwon and Leonov, 1992), where the violation of the positive definiteness of c is also demonstrated. In this analysis, we consider the flow situation described below in order to demonstrate instability.

In the following simple shear flow after constant step stress $\hat{\sigma}_0$ applied at $t = 0$:

$$\hat{\sigma}_{12} = \hat{\sigma}_0 H(t), \tag{56}$$

initial values of λ and S_{12} become

$$\lambda_0 \equiv \lambda(t=0) = \langle \mathbf{E}_0 \cdot \mathbf{u} \rangle_0, \quad \hat{\sigma}_0 \equiv \hat{\sigma}_{12}(t=0) = \lambda_0^2 S_0, \\ S_0 \equiv S_{12}(t=0) = \frac{(c_0)_{12}}{(c_0)_{11} + 2} \\ \mathbf{E}_0 = \begin{bmatrix} 1 & \gamma_0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{bmatrix}, \quad \mathbf{c}_0 = \mathbf{E}_0 \cdot \mathbf{E}_0^T = \begin{bmatrix} 1 + \gamma_0^2 & \gamma_0 & 0 \\ \gamma_0 & 1 & 0 \\ 0 & 0 & 1 \end{bmatrix}. \tag{57}$$

Here $H(t)$ is the Heaviside function and γ_0 is the initial step strain in this creep flow to be determined from the second of Eqs.(57). Above equations are again valid only in the case of $\lambda < q$, and when λ achieves the value of q , the stress relation for initial values is uncertain whether s_c is zero or nonzero. Due to this ambiguity, we here take into account only the case of initial stretch less than q .

In this type of flow, the evolution and stress equations reduce to

$$\frac{\partial \lambda}{\partial \hat{t}} = \lambda \Gamma S_{12} - \frac{\tau_b(0)}{\tau_s} (\lambda - 1), \quad \hat{\sigma}_{ij} \equiv \sigma_{ij}/G = \lambda^2 S_{ij} \quad \text{when } \lambda < q,$$

$$\frac{\partial S_{ij}}{\partial \hat{t}} = \left(\frac{1}{2} q s_b + s_c \right) \Gamma S_{12} - \frac{\tau_b(0)}{2 \tau_a(x)}, \quad \hat{\sigma}_{ij} = \left(q^2 + 2q \frac{s_c}{s_b} \right) S_{ij},$$

when $\lambda = q$,

$$\frac{\partial c_{11}}{\partial \hat{t}} - 2\Gamma c_{12} + \frac{\tau_b(0)}{\tau_b} (c_{11} - 1) = 0, \quad \frac{\partial c_{12}}{\partial \hat{t}} - \Gamma c_{22} + \frac{\tau_b(0)}{\tau_b} c_{12} = 0,$$

$$c_{22} = c_{33} = 1, \quad c_{13} = c_{23} = 0,$$

$$S_{ij} = \frac{c_{ij}}{c_{11} + 2}, \quad \Gamma = \tau_b(0) \dot{\gamma}, \quad \hat{t} = \frac{t}{\tau_b(0)} \quad (58)$$

In the above stress relation, $\hat{\sigma}_{12}$ is fixed at the value of $\hat{\sigma}_0$ in this creep experiment, and it imposes following constraints:

$$\frac{d\hat{\sigma}_{12}}{d\hat{t}} = \frac{d}{d\hat{t}} (\lambda^2 S_{12}) = 0 \quad \text{when } \lambda < q,$$

$$\frac{d\hat{\sigma}_{12}}{d\hat{t}} = \frac{d}{d\hat{t}} \left[\left(q^2 + 2q \frac{s_c}{s_b} \right) S_{12} \right] = 0 \quad \text{when } \lambda = q. \quad (59)$$

Hence they finally yield

$$\Gamma = \frac{c_{12}}{c_{11} + 2} \left[2 \frac{\tau_b(0)}{\tau_s} \left(1 - \frac{1}{\lambda} \right) (c_{11} + 2) + 3 \right] \quad \text{when } \lambda < q,$$

$$\Gamma = \frac{\tau_a(0)}{\tau_a(x) c_{11} + 2 - c_{12}^2} \left[\frac{\tau_b(0)}{\tau_s} \frac{q s_b}{q s_b + 2 s_c} (c_{11} + 2) + 3 \right] \quad \text{when } \lambda = q. \quad (60)$$

The result of computation according to Eqs.(58) and (60) is illustrated in Fig. 4 for $q = 3$, $s_a = 5$, $s_b = 20$ and $\hat{\sigma}_0 = 2\hat{\sigma}_m$, where $\hat{\sigma}_m$ denotes the maximum value of dimensionless shear stress in steady flow curve. All the solutions except S_{12} manifest rapid increase with respect to time. When the dimensionless time approaches the value of 1.267, the stretch ratio λ becomes close to its maximum admissible value q . When λ attains the value q , the com-

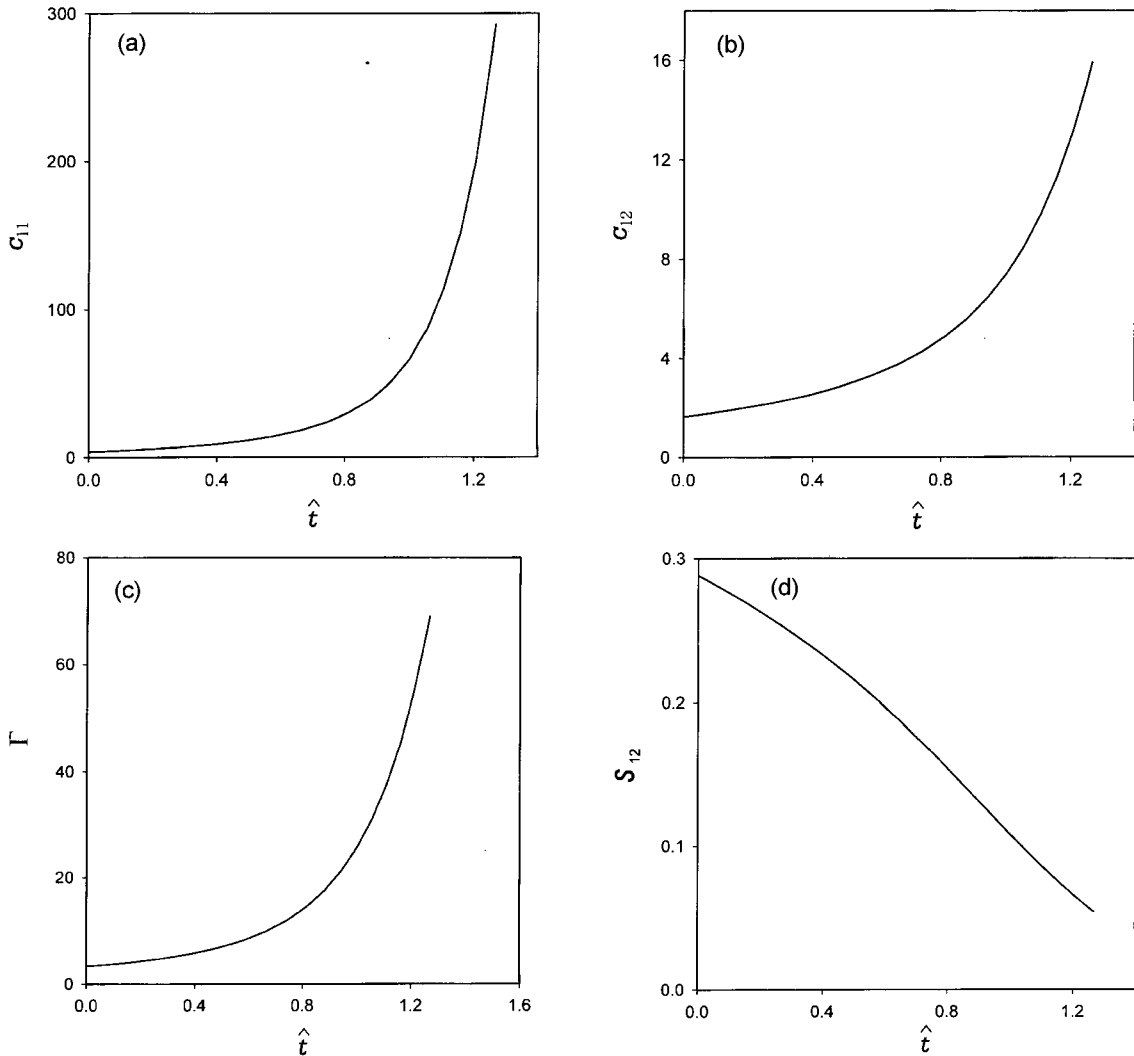


Fig. 4. The behavior of the pom-pom model in creep simple shear flow when applied stress is twice of the maximum achievable stress in steady flow curve (a c_{11} , b c_{12} , c Γ , d S_{12}).

putation regime for shear rate changes from the first to the second of Eqs.(60). However, this model ceases to have physically meaningful solution from this time on, since the shear rate Γ exhibits strong discontinuity from the value 69 to 369. In Eqs.(60), one can easily observe such incompatibility that exists between the first and the second equations, if one assumes continuity of other variables such as c_{11} and c_{12} at $\lambda = q$. Due to this solution jump, we cannot obtain solutions after that instant. This kind of ill-posed behavior occurs for the pom-pom model in creep flow where the stress history is assigned, whereas it may not be observed when strain history is specified.

4.2. Stability analysis of the integral/differential model

In this section, we consider the Hadamard-type stability for the original set of Eqs.(32)-(38). By the same way, we apply the following infinitesimal disturbance on the basic solution:

$$\begin{aligned} \{ \sigma, S, Q^{IA}, E, v, \lambda, p, s_c \} &= \{ \sigma_0, S_0, Q_0^{IA}, E_0, v_0, \lambda_0, p_0, s_{c0} \} \\ &+ \{ \delta\sigma, \delta S, \delta Q^{IA}, \delta E, \delta v, \delta\lambda, \delta p, \delta s_c \}, \\ \{ \delta\sigma, \delta S, \delta Q^{IA}, \delta E, \delta v, \delta\lambda, \delta p, \delta s_c \} &= \varepsilon \{ \hat{\sigma}, \hat{S}, \hat{Q}^{IA}, \hat{E}, \hat{v}, \hat{\lambda}, \hat{p}, \hat{s}_c \} \\ &\times \exp [i(\mathbf{k} \cdot \mathbf{x} - \omega t)/\varepsilon^2]. \end{aligned} \tag{61}$$

Here δQ^{IA} and δE are added instead of δS of Eqs.(40) and from now on we also remove the subscript 0 in the basic solutions.

We carry out the analysis in two folds this time. First is the stability analysis in the flow regime of $\lambda < q$. The second part of analysis consists in the region of $\lambda = q$, and we neglect the contribution of s_c . However for this integral/differential constitutive equations, the complete analysis in the region of $\lambda = q$ with s_c included has not been performed due to their complexity. Hence in the following analyses, the disturbance imposed on the arm withdrawal length always vanishes, that is, $\delta s_c = 0$ in Eqs.(61).

4.2.1. Hadamard stability analysis when $\lambda < q$

The linear perturbation imposed on Eqs.(33) finally yields the resultant inequality for stability in the form of

$$\begin{aligned} \rho \Omega^2 \hat{v}^2 &= G \lambda^2 \left\{ 2 \frac{1}{\tau_b} \int_{-\infty}^t \exp\left(-\frac{t-t_1}{\tau_b}\right) \langle e_i e_j \rangle_0 dt_1 \right. \\ &\cdot \frac{1}{\tau_b} \int_{-\infty}^t \exp\left(-\frac{t-t_2}{\tau_b}\right) \langle e_m e_n \rangle_0 dt_2 + \delta_{im} \frac{1}{\tau_b} \int_{-\infty}^t \exp\left(-\frac{t-t_1}{\tau_b}\right) \langle e_j e_n \rangle_0 dt_1 \\ &\left. - 2 \frac{1}{\tau_b} \int_{-\infty}^t \exp\left(-\frac{t-t_1}{\tau_b}\right) \langle e_i e_j e_m e_n \rangle_0 dt_1 \right\} \hat{v}_i k_j \hat{v}_m k_n > 0. \end{aligned} \tag{62}$$

Here $e_i = \frac{E_{im} u_m}{|\mathbf{E} \cdot \mathbf{u}|}$ and $\hat{v}^2 = \hat{v}_i \hat{v}_j$. Since the complete analysis to yield the necessary and sufficient condition of stability is

not known to authors, we confine the study to following simple situation.

In the case of stress relaxation after some arbitrary step strain, the tensor E and vector e become

$$E = (E_0 - \delta)H(-t_1) + \delta, \quad e_i = \begin{cases} \frac{1}{|\mathbf{E}_0 \cdot \mathbf{u}|} E_{ij}^0 u_j \equiv e_i^0, & t_1 < 0 \\ u_i, & t_1 > 0, \end{cases} \tag{63}$$

where e_i^0 and $E_{ij}^0 = (E_0)_{ij}$ are constant vector and tensor components, respectively. Then the inequality (62) results in

$$\begin{aligned} \rho \Omega^2 \hat{v}^2 &= G \lambda^2 \left\{ \frac{1}{5} \delta_{im} \delta_{jn} + \exp\left(-\frac{t}{\tau_b}\right) \left(\delta_{im} h_{ij} - \frac{1}{5} \delta_{im} \delta_{jn} - 2H_{ijmn} \right) \right. \\ &\left. + 2 \exp\left(-\frac{2t}{\tau_b}\right) h_{ij} h_{mn} \right\} \hat{v}_i k_j \hat{v}_m k_n > 0, \\ h_{jn} &= \langle e_j^0 e_n^0 \rangle_0, \quad H_{ijmn} = \langle e_i^0 e_j^0 e_m^0 e_n^0 \rangle_0. \end{aligned} \tag{64}$$

Right at the moment of strain imposition, i.e., when $t \rightarrow 0+$, this inequality is simplified to

$$\rho \Omega^2 \hat{v}^2 = G \lambda^2 (\delta_{im} h_{ij} - 2H_{ijmn} + 2h_{ij} h_{mn}) \hat{v}_i k_j \hat{v}_m k_n > 0. \tag{65}$$

If we apply the same procedure employed in paper (Kwon, 1999), the following necessary conditions of stability under 1D and 2D disturbances can be derived:

1D disturbance: $h_{ii} - 2H_{iiij} + 2h_{ij}^2 > 0$

2D disturbance:

$$\begin{aligned} &\left(x + \frac{1}{x} \right) \left(x h_{ii} - 2h_{ij} + \frac{1}{x} h_{jj} \right) - 2 \left\{ H_{iiii} + H_{jjjj} + 2 \left(x - \frac{1}{x} \right) (H_{iiij} - H_{ijjj}) \right. \\ &+ \left(x^2 - 4 + \frac{1}{x^2} \right) H_{iiij} \left. \right\} + (h_{ii} - h_{jj})^2 + 2 \left(x - \frac{1}{x} \right) (h_{ii} h_{ij} - h_{ij} h_{jj}) \\ &+ \left(x - \frac{1}{x} \right)^2 h_{ij}^2 > 0 \\ &\text{for } -\infty < x < \infty \text{ (no sum on } i \text{ and } j). \end{aligned} \tag{66}$$

Even though the general analysis of the above simplified stability conditions cannot be achieved, it has been verified that in simple shear flow both inequalities in Eq.(66) are always satisfied.

4.2.2. Hadamard stability analysis when $\lambda = q$ (s_c neglected)

In this case, again $\delta\lambda = \hat{\lambda} = 0$. When the variation of s_c is neglected, the resultant stability condition equivalent to Eq.(62) becomes

$$\begin{aligned} \rho \Omega^2 \hat{v}^2 &= G q^2 \frac{1}{\tau_b} \int_{-\infty}^t \exp\left(-\frac{t-t_1}{\tau_b}\right) \{ \delta_{im} \langle e_j e_n \rangle_0 - 2 \langle e_i e_j e_m e_n \rangle_0 \} dt_1 \times \\ &\hat{v}_i k_j \hat{v}_m k_n > 0. \end{aligned} \tag{67}$$

Now this condition of Hadamard-type stability for the pom-pom model exactly coincides with that for the Doi-Edwards model, i.e., Eqs.(5) and (6) in paper (Kwon, 1999). That is, in stress relaxation after arbitrary step strain, inequality (67) again results in

$$\rho\Omega^2\hat{v}^2 = Gq^2\left\{\frac{1}{5}\delta_{im}\delta_{jn}(1-e^{-t/\tau_b})+(\delta_{im}h_{jn}-2H_{ijmn})e^{-t/\tau_b}\right\} \\ \times \hat{v}_i\hat{k}_j\hat{v}_m\hat{k}_n > 0,$$

and at the moment of strain imposition reduces to the following condition, the upper bound for stability:

$$(\delta_{im}h_{jn}-2H_{ijmn})\hat{v}_i\hat{k}_j\hat{v}_m\hat{k}_n > 0, \quad (68)$$

which precisely corresponds to Eq.(7) with (9) in the reference (Kwon, 1999). Therefore we may draw a same conclusion that the pom-pom constitutive equations are Hadamard-type unstable in the maximum backbone stretch with s_c neglected.

5. Conclusion

In this paper, we examined recent results of mathematical stability for viscoelastic constitutive equations. Employing an asymptotic analysis, we have verified that the time-strain separability implemented in viscoelastic models is the main origin of mathematically ill-posed behavior in these models. In this analysis, the commonly used differential and integral constitutive equations involving separability are proven either Hadamard-type unstable or dissipative unstable. The hypothesis of time-strain separability is shown to cause the Hadamard-type instability of the Wagner, Luo-Tanner, Papanastasiou and K-BKZ models with Larson-Monroe or Mooney potential, and the dissipative instability of the Lodge model. The implicit relation between the Finger strain C^{-1} and the geometric tensor Q (through the evolution equation of a deformation gradient tensor E), allows the same conclusion to be drawn for the Doi-Edwards model. Hence, the Hadamard-type instability of the Doi-Edwards model has its origin in the time-strain separability in its formulation. Regarding the pom-pom constitutive equations, it is proved that the differential version with its full description is globally Hadamard stable, as long as the orientation tensor S or c remains positive definite, in other words when the smooth strain history is pre-defined. For both differential and integral/differential types Hadamard instability occurs in the case of maximum backbone stretch with arm withdrawal s_c neglected. In the sense of dissipative stability, the differential model is also 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 exhibit strong discontinuity of solutions that prohibits further continuation of stable computation along time axis.

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