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Anisotropy parameter restrictions for the eXtended Pom-Pom model

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ABSTRACT

A significant step forward in modelling polymer melt rheology has been the introduction of the Pom-Pom constitutive model of McLeish and Larson [T.C.B. McLeish, R.G. Larson, Molecular constitutive equations for a class of branched polymers: the Pom-Pom polymer, J. Rheol. 42 (1) (1998) 81–110]. Various modifications of the Pom-Pom model have been published over the years in order to overcome several inconveniences of the original model. Amongst those modified models, the eXtended Pom-Pom (XPP) model of Verbeeten et. al. [W.M.H. Verbeeten, G.W.M. Peters, F.P.T. Baaijens, Differential constitutive equations for polymer melts: the extended Pom-Pom model, J. Rheol. 45 (4) (2001) 823–843] has received quite some attention. However, the XPP model has been criticized for the generation of multiple and unphysical solutions. This paper deals with two issues. First, in the XPP model, anisotropy is implemented in a Giesekus-like manner which is known to result in unphysical solutions for non-linear parameter values $\alpha \geq 0.5$. Hence, we put forward the conjecture that a similar limitation holds for the XPP model. In the present paper, the limits for the anisotropy parameter are elaborated on and result to be most restraining at high deformation rates where the backbone tube is oriented and the backbone tube stretch approaches the number of arms q. By restricting the anisotropy parameter to a maximum critical value the XPP model produces only one solution, which is the correct physical rheology. In the second part we show that, contrary to the results published by Inkson and Phillips [N.J. Inkson, T.N. Phillips, Unphysical phenomena associated with the extended Pom-Pom model in steady flow, J. Non-Newton. Fluid 145 (2–3) (2007) 92–101], for the special case where the anisotropy parameter equals zero, only one physically relevant solution exists in unaxial extensional. In addition to this physically relevant solution, also solutions exist in the physically unattainable part of the conformation space. However, the existence of these physically unattainable solutions is not a unique feature of the XPP model but rather general for non-linear differential type rheological equations.

1. Introduction

The processing of polymer materials has a large influence on the dimensional, mechanical, and optical properties of the end product. The complex rheological behaviour typically encountered in macromolecular fluids is an important reason for that influence. In order to predict the viscoelastic behaviour of polymer melts, simulation tools have been developed, which need constitutive models that can adequately model the polymer dynamics. A significant step forward in modelling polymer melt rheology has been the introduction of the Pom-Pom constitutive model [7]. This model is able to quantitatively predict the correct nonlinear behaviour in both shear and extension simultaneously for branched materials, such as low density polyethylene melts.

Various modifications of the Pom-Pom model have been published over the years in order to overcome several inconveniences of the original model [2,3,8,9,12]. Amongst those modified models, the eXtended Pom-Pom (XPP) model [12] has received quite some attention. This particular model was successfully implemented in a finite element code and was able to satisfactorily predict the behaviour of a commercial LDPE melt in complex flow geometries in a quantitative manner [13,14].

Several authors have criticized the XPP model for both mathematical defects [3] and unphysical solutions [3,5]. Both seemingly alarming issues have each their own specific, yet simple solution. On the one hand, the mathematical defects will mostly be critical in numerical computations in the vicinity of geometric singularities. These can be circumvented by choosing the double-equation version of the XPP model, referred to as DXPP, as mentioned by

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Verbeeten et al. [12] and Clemeur et al. [3]. On the other hand, the unphysical solutions, such as turning points [3] and bifurcation and multiple solutions [3,5], were shown to be related to the anisotropy parameter α. This parameter was introduced to produce a non-vanishing second normal stress difference. By restricting this parameter to a maximum critical value, the correct physical solutions do exist and will be encountered starting from an admissible initial conformation tensor. The restriction still leaves enough freedom to fit the second normal stress difference.

Clemeur et al. [3] propose to use the double-equation version of the eXtended Pom-Pom model and setting the anisotropy parameter $\alpha = 0$. In this way, they avoid the mathematical defects of the single-equation version and by choosing $\alpha = 0$, bifurcation and multiple solutions are also ommited. This unfortunately comes at the cost of losing the second normal stress difference. They suggest an alternative way of introducing a non-vanishing second normal stress coefficient by combining both an upper- and lower-convected time derivative, similar to Johnson and Segalman [6]. However, such a combination of an upper-convected and lower-convected time derivative does not fit within the thermodynamic framework GENERIC [10], i.e. combinations are thermodynamically not allowed and a purely upper-convected or purely lower-convected time derivative is preferred.

Since the anisotropy parameter in the XPP model is Giesekus-like and the anisotropy parameter α in the Giesekus model is restricted [11], it is expected that some restrictions are also present for the α-parameter in the XPP model. The objective of the present paper is to indicate the limits for the α-parameter of the XPP model in order to avoid unphysical and multiple solutions.

2. Modelling

To realistically describe the viscoelastic stresses of polymer fluids over a broad range of deformation rates, a multi-mode approximation of the extra-stress tensor $\boldsymbol{\tau}$ is defined as

$$\boldsymbol{\tau} = \sum_{i=1}^{M} G_i\,(\mathbf{c}_i - \mathbf{I})\,. \tag{1}$$

Here M is the total number of different relaxation times, G_i is the shear modulus of the ith relaxation mode, $\mathbf{c}_i$ is the conformation tensor, and $\mathbf{I}$ is the unit tensor. The conformation tensor of the ith relaxation mode is defined as

$$\mathbf{c}_i = 3\Lambda_i^2 \mathbf{S}_i, \tag{2}$$

with Λ_i the backbone stretch and $\mathbf{S}_i$ the orientation tensor of the backbone tube. In the remainder of this paper we will restrict ourselves to a single mode description of the constitutive behavior and omit the subscript i.

For the eXtended Pom-Pom (XPP) model, time evolution of the conformation tensor follows from

$$\lambda_b \overset{\nabla}{\mathbf{c}} + [f(\mathbf{c}) - 2\alpha]\,\mathbf{c} + \alpha\mathbf{c}^2 + (\alpha - 1)\,\mathbf{I} = 0, \tag{3}$$

in which the function $f(\mathbf{c})$ is given by

$$f(\mathbf{c}) = 2r\,\mathrm{e}^{\nu(\Lambda-1)}\left(1 - \frac{1}{\Lambda}\right) + \frac{1}{\Lambda^2}\left[1 - \alpha - \frac{\alpha}{3}\mathrm{tr}(\mathbf{c}^2 - 2\mathbf{c})\right]. \tag{4}$$

Here λ_b is the relaxation time of the backbone tube orientation, taken from the linear relaxation spectrum. α is the anisotropy parameter that influences the second normal stress difference, $r = \lambda_b/\lambda_s$ with λ_s the relaxation time for the tube stretch, while ν is a parameter determining the influence of the surrounding polymer chains on the backbone tube stretch and is defined as $\nu = 2/q$, where q denotes the amount of arms at the end of a backbone. Since the trace operator acting on the orientation tensor yields 1 by definition, the backbone stretch is defined as $\Lambda = \sqrt{\mathrm{tr}(\mathbf{c})/3}$.

Eqs. (1)–(4) give the same XPP model as given in Clemeur et al. [3] and Inkson and Phillips [5]. However, instead of being written in terms of the orientation tensor $\mathbf{S}$ and backbone tube stretch Λ or extra-stress tensor $\boldsymbol{\tau}$, it is written in terms of the conformation tensor $\mathbf{c}$.

A more appropiate equation for the function $f(\mathbf{c})$, consistent with the thermodynamical framework GENERIC [8,14], reads

$$f(\mathbf{c}) = 2r\,\mathrm{e}^{\nu(\Lambda-1)}\left(1 - \frac{1}{\Lambda^2}\right) + \frac{1}{\Lambda^2}\left[1 - \alpha - \frac{\alpha}{3}\mathrm{tr}(\mathbf{c}^2 - 2\mathbf{c})\right]. \tag{5}$$

Since the introduction of the second normal stress difference is Giesekus-like by means of the anisotropy parameter α, and that parameter in the Giesekus model is restricted [11], the evolution equation of the Giesekus conformation tensor is also given for comparison

$$\lambda \overset{\nabla}{\mathbf{c}} + [1 - 2\alpha]\,\mathbf{c} + \alpha\mathbf{c}^2 + (\alpha - 1)\,\mathbf{I} = 0, \tag{6}$$

with λ the linear relaxation time.

3. Anisotropy parameter restrictions

Our starting point is the limitation on the parameter α in the Giesekus model, $0 \le \alpha \le (1/2)$, as suggested by Bird et al. [1] and later by Schleininger and Weinacht [11], studying the linear stability of Couette flow. The restriction ensures that the Giesekus model does not give solutions with a maximum in the shear or elongational stress, leading to unstable, non-physical solutions. The restriction also ensures that the linear term in Eq. (6) ($[1 - 2\alpha]\mathbf{c}$), is positive. We put forward the conjecture that a similar limitation holds for the XPP model, but we are not able to give a formal prove for this and maybe this is not even possible. However, all numerical experiments that we have performed as well as all relevant data obtained from literature support this conjecture. Also a violation of our proposed limitation on α may lead to the flow condition that no steady state solution exists beyond some finite value of Λ^2 (see Appendix A). With this as a starting point and considering the corresponding term in the XPP model $[f(\mathbf{c}) - 2\alpha]\mathbf{c}$, the restriction we put forward reads $[f(\mathbf{c}) - 2\alpha] \ge 0$. This leads to a positive linear term in the XPP model. In this way, the maximum anisotropy parameter α allowed becomes a function of the other material parameters and the conformation tensor $\mathbf{c}$, and thus depends on the stretch and orientation.

For low shear and elongational rates, stretch and orientation are limited which implies that $f(\mathbf{c}) \approx 1$ and the behavior of the XPP model reduces to the behavior of the Giesekus model. However, at high deformation rates, $f(\mathbf{c})$ changes significantly and, for given ratio r and number of arms q, the critical value of α becomes a function of the applied deformation rate. Computationally, this can be confirmed by computing the values of the function $g(\mathbf{c}) = [f(\mathbf{c}) - 2\alpha]$ for a relevant series of deformation rates and values of α. Contours of $g = 0$, as shown in Fig. 1, reveal the dependence of the maximum value for α as a function of the applied shear or extensional rate. The figure shows that, for fixed ratio of relaxation times r and increasing number of arms q, the minimum allowable value for α appears at high shear or elongational rate. In addition, it shows that the minimum allowable value for α is different in shear and uniaxial extension and appears at different deformation rates.

In order to find an expression for the maximum allowable value of α in general complex flows (denoted by $\alpha_{\max}$) we write the function $g(\mathbf{c})$ in the following way:

$$g(\mathbf{c}) = \frac{1}{\Lambda^2}\left[2r\,\mathrm{e}^{\nu(\Lambda-1)}(\Lambda^2 - \Lambda) + 1 - \alpha(1 + 3\Lambda^4\mathrm{tr}(\mathbf{S}^2))\right]. \tag{7}$$

The possible values for $\mathbf{c}$ (and thus also of Λ and $\mathrm{tr}(\mathbf{S}^2)$) of the XPP model lie on a surface in (c_1, c_2, c_3)-space, where (c_1, c_2, c_3)