



On the modelling of highly elastic flows of amorphous thermoplastics

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ABSTRACT

Two approaches to the kinematic structuring of constitutive models for highly elastic flows of polymer melts have been examined systematically, assuming either: (1) additivity of elastic and viscous velocity gradients or (2) multiplicability of elastic and viscous deformation gradients. A series of constitutive models were compared, with differing kinematic structure but the same linear responses in elastic and viscous limits. They were solved numerically and their predictions compared, and they were also compared to those of the Giesekus model. Several variants, previously proposed as separate models, are shown to be equivalent and qualitatively in agreement with experiment, and therefore a sound basis for construction of models. But the assignment of viscous spin is critical: if it is assumed equal to the total spin with approach (1), or equal to zero with approach (2), then unphysical viscoelastic behaviour is predicted.

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1. Introduction

Industrial forming processes for thermoplastic polymers frequently involve large deformations in a time/temperature range where flow is highly elastic. Physically, this arises from the great lengths of the molecules. All molecules with molar mass M larger than a monomer-specific critical value M_e are topologically constrained by their neighbours, linking them into a continuous molecular network even in the molten state—that is, when they are amorphous and above the glass transition temperature T_g . Connectivity is provided by molecular entanglements. Such a network has an elastic, rubber-like, constitutive response when unrelaxed. It can relax fully, but only by the tortuous process of molecular disengagement that has come to be known as “reptation”, with an associated relaxation time τ_d . Industrial polymers usually have $M \gg M_e$, and $\tau_d \propto (M/M_e)^\beta$ where $\beta \sim 3.4$ [30], so relaxation times are exceptionally long compared to other viscoelastic liquids. Moreover, economic necessity requires industrial forming processes for polymers to be as rapid as possible. Consequently polymers are frequently melt-processed on time scales not far from τ_d .

In such flows, elastic stretch of the entanglement network is only partially relaxed. This is especially true of processes such as stretch-blow moulding and thermoforming of sheets where substantial elasticity of the melt is advantageous to stability of the process. Network stretch, and hence mutual alignment of the molecules, is

also an essential requirement for stress-induced crystallisation (e.g. during blow moulding of polyester bottles or spinning of polyamide fibres). Moreover, the complexity of non-linear viscoelastic material behaviour is often combined with large-scale geometrical non-linearity. An example of this is the finite rotation encountered when the flow has a large shear component, for example around rigid particles in modelling of the forming of particulate-reinforced polymers.

This paper is motivated by the engineering need to model highly elastic polymer flows, in a manner suitable for optimisation in the context of numerical simulation of polymer processes. Clearly, numerical modelling of such processes requires a constitutive model that is robust under arbitrarily large deformations and in the presence of a high degree of elasticity. The question of how best to achieve this remains a matter of dispute. A particular difficulty is that solutions have been proposed in two different branches of the literature—solid mechanics and fluid mechanics—and hence it has been unclear how they are all related. The present note aims to clarify the issues, and assist the development of suitable constitutive models, by comparing systematically the kinematic assumptions embedded in various approaches, and highlighting how they are related.

Previous authors attempting to capture accurately, but empirically, highly elastic flows of amorphous polymers have adopted a range of strategies. Some have ignored the problem altogether and have approximated the polymer response as wholly viscous [15,6] or wholly elastic but with rate and temperature-dependent parameters [31,23]. While it is possible to fit experimental data for a given monotonic strain sequence in this way, it is clearly impossible to capture an arbitrary deformation history with such approaches. Other authors proposing finite deformation viscoelastic models, in view of the lack of experimental evidence for how the antisymmetric part

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of the velocity gradient (i.e. the spin) should be apportioned between viscous flow and elastic deformation, have cautiously declined to speculate on this point and their proposed models are incomplete in this respect [34,1,9,22].

Those authors that have proposed complete three-dimensional models have adopted one of three approaches. One group have employed models based on additive split of the rate of deformation tensor [25,26] together with the assumption of zero viscous spin [21]. Another group assumed multiplicative decomposition of the deformation gradient [16,20], together with a particular, convected, interpretation of the viscous velocity gradient and the assumption of zero viscous spin [4,5,11]. Finally, several authors have employed models expressed in terms of convected derivatives of stress, such as the upper convected Maxwell model (UCM), see for example [28], or the more robust Giesekus modified UCM or a finite extensibility adaptation of it [10]. In addition, the literature provides a number of physically based models with similar structure, which quite successfully capture polymer melt viscoelasticity under a wide range of conditions. An attractive feature of these is that they embody awareness of molecular architecture. Examples are the Pom-Pom model [24] for branched molecules and the Rolie-Poly model [17] for linear molecules. However, these do not yet capture accurately the highly elastic extensional flows of interest here, without empirical extensions. For authoritative reviews of polymer melt constitutive models to date, and their links to molecular structure, the reader is referred to [2,19,8].

The present paper considers classes of constitutive models that may be conveniently fitted to experimental data under relevant conditions. These models are kinematically structured a priori to capture naturally the geometrical non-linearity, before insertion of a description of the physical response, that may be either empirical or physically inspired. Two approaches to their kinematic structure are compared in this work: (1) approach I is based on additive decomposition of the velocity gradient tensor, while (2) approach II is based on multiplicative decomposition of the deformation gradient tensor. In order to highlight the consequences of purely *geometric* non-linearity arising from different kinematic assumptions made, models considered here based on each approach are *linear* in both the elastic and viscous limits. Thus elastic response is taken to be neo-Hookean, while viscous response is taken to be Newtonian.

The Giesekus model provides a convenient benchmark, as its variable parameter α ($0 \leq \alpha \leq 1$) allows several models to be recovered from a single equation.

2. Approaches

Consistent with the aim of modelling highly elastic flows where stresses may be sufficient for detectable volume change to occur, in the models considered here there is a reversible volumetric contribution to the deformation gradient. Thus we begin with a multiplicative decomposition of the deformation gradient \mathbf{F} into its volumetric (\mathbf{F}_{vol}) and isochoric ($\hat{\mathbf{F}}$) parts [12]

$$\mathbf{F} = \mathbf{F}_{\text{vol}} \hat{\mathbf{F}} \quad \text{where } \hat{\mathbf{F}} = J^{-1/3} \mathbf{F} \text{ and } J = \det \mathbf{F}. \quad (1)$$

J is the volume ratio. The viscoelastic response is then all contained within the isochoric part of the velocity gradient $\hat{\mathbf{F}}$. Its corresponding left Cauchy–Green tensor and velocity gradient, and the latter's symmetric (deformation rate) and skew-symmetric (spin) parts are defined by

$$\hat{\mathbf{B}} = \hat{\mathbf{F}} \hat{\mathbf{F}}^T, \quad \hat{\mathbf{L}} = \dot{\hat{\mathbf{F}}} \hat{\mathbf{F}}^{-1} \quad \text{and} \quad \hat{\mathbf{D}} = \text{sym}[\hat{\mathbf{L}}] \quad \text{and} \quad \hat{\mathbf{W}} = \text{skew}[\hat{\mathbf{L}}]. \quad (2)$$

In order to avoid unnecessary complexity, we now restrict attention to only those materials whose response to volume change is purely elastic (a good approximation in the case of elasto-viscous polymer

melts). In the two limits of fast (elastic) deformation, or slow (viscous) deformation, respectively, the deviatoric Cauchy stress may then be written as

$$\hat{\boldsymbol{\sigma}} = \hat{\boldsymbol{\sigma}}(\hat{\mathbf{B}}, J) \quad \text{or} \quad \hat{\boldsymbol{\sigma}} = \hat{\boldsymbol{\sigma}}(\hat{\mathbf{D}}, J), \quad (3)$$

respectively, where the two functions must be determined by experiment. In the general case of an intermediate rate (viscoelastic) deformation, we assume that “elastic” and “viscous” deformation gradients exist, $\hat{\mathbf{F}}_E$ and $\hat{\mathbf{F}}_V$, such that the instantaneous Cauchy stress is given in terms of the left Cauchy–Green tensor and the rate of deformation derived from them, respectively, thus

$$\hat{\boldsymbol{\sigma}} = \hat{\boldsymbol{\sigma}}(\hat{\mathbf{B}}_E, J) \quad \text{and} \quad \hat{\boldsymbol{\sigma}} = \hat{\boldsymbol{\sigma}}(\hat{\mathbf{D}}_V, J). \quad (4)$$

Completion of a three-dimensional constitutive model requires knowledge of how $\hat{\mathbf{B}}_E$ and $\hat{\mathbf{D}}_V$ are related to $\hat{\mathbf{F}}$.

2.1. Approach based on additive decomposition of the velocity gradient (Approach I)

In this approach, additive decomposition of the isochoric velocity gradient $\hat{\mathbf{L}}$ into elastic $\hat{\mathbf{L}}_E^I$ and viscous $\hat{\mathbf{L}}_V^I$ parts is assumed *a priori*

$$\hat{\mathbf{L}} = \hat{\mathbf{L}}_E^I + \hat{\mathbf{L}}_V^I \quad \text{hence} \quad \hat{\mathbf{D}} = \hat{\mathbf{D}}_E^I + \hat{\mathbf{D}}_V^I \quad \text{and} \quad \hat{\mathbf{W}} = \hat{\mathbf{W}}_E^I + \hat{\mathbf{W}}_V^I, \quad (5)$$

where superscript I is introduced to distinguish elastic and viscous quantities from their counterparts in approach II presented in the next section. The second of Eq. (5) expresses the additivity of rates of deformation proposed by Nemat-Nasser and others, where $\hat{\mathbf{D}}_V^I$ is defined by a flow rule (see Section 3).

There is no attempt to attribute precise physical meaning to $\hat{\mathbf{F}}_E^I$ and $\hat{\mathbf{F}}_V^I$ in terms of macroscopic response. In terms of polymer physics, $\hat{\mathbf{F}}_E^I$ relates to molecular chain configurations, whose perturbation from equilibrium at any instant gives rise to the entropic stress dependent on $\hat{\mathbf{B}}_E^I$. To exploit Eq. (5), we make use of the following kinematic identity for the time derivative of $\hat{\mathbf{B}}_E^I$ with respect to a fixed reference frame

$$\dot{\hat{\mathbf{B}}}_E^I = (\hat{\mathbf{F}}_E^I (\hat{\mathbf{F}}_E^I)^T)^\circ = \hat{\mathbf{L}}_E^I \hat{\mathbf{B}}_E^I + \hat{\mathbf{B}}_E^I (\hat{\mathbf{L}}_E^I)^T, \quad (6)$$

where superscript T denotes the usual transpose, and the definition of the co-rotational (Jaumann) derivative of $\hat{\mathbf{B}}_E^I$

$$\hat{\mathbf{B}}_E^I \overset{\circ}{=} \dot{\hat{\mathbf{B}}}_E^I - \hat{\mathbf{W}} \hat{\mathbf{B}}_E^I + \hat{\mathbf{B}}_E^I \hat{\mathbf{W}}. \quad (7)$$

There is no means of completing the model rigorously. An assumption must be made concerning apportionment of spin $\hat{\mathbf{W}}$ between elastic and viscous parts of the velocity gradient, in the third of Eq. (5). Here we consider two possible cases:

- (i) Case 1: We follow Giesekus' plausible physical argument for polymers that $\hat{\mathbf{L}}_V^I$ must be an inner variable of the configurational state, as expressed by symmetric tensor $\hat{\mathbf{B}}_E^I$, and hence must itself be symmetric, giving $\hat{\mathbf{W}}_V^I = \mathbf{0}$ [13]. Hence, it follows from Eq. (5) that $\hat{\mathbf{W}}_E^I = \hat{\mathbf{W}}$. Then, combining Eqs. (6) and (7) with the *a priori* assumption Eq. (5) gives

$$(\hat{\mathbf{B}}_E^I)^\circ = \hat{\mathbf{D}}_E^I \hat{\mathbf{B}}_E^I + \hat{\mathbf{B}}_E^I \hat{\mathbf{D}}_E^I = (\hat{\mathbf{D}} - \hat{\mathbf{D}}_V^I) \hat{\mathbf{B}}_E^I + \hat{\mathbf{B}}_E^I (\hat{\mathbf{D}} - \hat{\mathbf{D}}_V^I). \quad (8)$$

We shall refer to Eq. (8) as the Leonov equation after its original proposer [21]. When constitutive representations are provided for $\hat{\mathbf{D}}_V^I$ and $\hat{\mathbf{B}}_E^I$ in terms of $\hat{\boldsymbol{\sigma}}$, Eq. (8) may be integrated to obtain the evolution of stress for a given deformation history. This approach has been used by Tervoort et al. [32] in modelling elastic-viscoplastic deformation of glassy polymers.

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