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A study of the quadratic molecular stress function constitutive model in simulation

P. Olley*

School of Engineering, Design and Technology/IRC in Polymer Engineering, EDT4, University of Bradford, Chesham Building, Bradford BD7 1DP, UK

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Abstract

Constitutive models that conform to separable KBKZ specification have been shown to fit steady-state strain hardening rheological data in planar and uniaxial elongational flows, but with inaccuracy in the rate of strain hardening. The single parameter molecular stress function model of Wagner [Rheol. Acta 39 (2000) 97–109] has been shown to accurately fit the rise-rate in experimental data for a number of strain hardening and strain-softening materials. We study this models accuracy against the well characterised IUPAC LDPE data, and present a method for full implementation of this model for flow solution which is suitable for incorporating into existing separable KBKZ software. A new method for particle tracking in arbitrarily aligned meshes, which is efficient and robust, is given.

The quadratic molecular stress function (QMSF) model is compared to existing separable KBKZ based models, including one which is capable of giving planar strain hardening; the QMSF is shown to fit experimental rheological and contraction flow data more convincingly. The issue of 'negative correction pressures' notable in some Doi–Edwards based models is addressed. The cause is identified, and leads to a logical method of calculation which does not give these anomalous results.

Keywords: Molecular stress function (MSF); Simulation; Strain hardening; KBKZ; Negative correction pressure

1. Introduction

Forms of the separable KBKZ model [1] have had success (though generally within some limit) in modelling a large number of experimental rheological data. Papanastasiou et al.'s damping function [2] has been very successful in fitting data for steady-state *uniaxial* elongational viscosity, shear viscosity, and first normal stress difference for both strain hardening and strain-softening materials. This has led to considerable success in simulating axisymmetric flows of strain hardening polymer melt [3–9]. The model has also been successful in simulating planar flows of strain-softening melt [6,10–12], but has not been so successful in modelling strain-hardening planar flows as it does not give simultaneous planar strain-hardening and shear softening behaviour [9]. An adaptation of the KBKZ [13] permitted steady-state values for planar elongation viscosity, uniaxial elongational viscosity, and shear viscosity to be fitted simultaneously; the rate of strain hardening was, however, below that seen in experimental results [13]. The model has been applied to several flows, including planar flows of strain hardening polymer melt, with improved success due to capturing significant features of strain hardening behaviour [13–15].

Significant improvement, particularly in capturing the rate of strain hardening, is suggested by use of the molecular stress function (MSF) model [16–20]. The MSF model is based upon considerations of the geometry and stored energy of the polymer segments undergoing deformation; a particular version, the quadratic molecular stress function (QMSF) model [19] has been shown to fit transient planar elongation, uniaxial elongation, biaxial elongation and first and second normal stress difference measurements for strain hardening flows. The accuracy of fit is remarkable particularly as only a

* Tel.: +44 1274 234537.
E-mail address: p.olley@bradford.ac.uk.

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single adjustable parameter is used in the model. convective constraint release (CCR) [21] is incorporated into the model described in reference [20], in a manner that effectively separates elongational and rotational damping functions. The versions preceding the CCR model can be recognized as following the 'separabilty' assumption [20].

There have been many recent developments in simulation of time-integral viscoelastic flows. Two-dimensional time dependent flows have been solved by convecting deformation fields, to achieve a purely Eulerian method [22]; three-dimensional time dependant flows have be modelled using a Lagrangian mesh approach with remeshing, and information transfer, when the mesh becomes distorted [23]. A modified version of the QMSF with CCR has been implemented and applied to membrane inflation [24]. Direct comparison has been made between a time-integral model, its differential approximation and an inelastic model in a complex flow [25].

There have been other recent developments in rheological modelling, with a strong emphasis on molecularly based models. Notable amongst these developments are the Pom Pom model [26,27], and developments in differential models [28,29]. Molecular considerations have led to some models that employ fractional powers of the Finger strain tensor [30,31].

The single parameter QMSF model without constraint release, described in reference [19] has been shown to have many strong points. The model also has an elegant physical derivation in its favour, and provides a natural bridge between earlier KBKZ models, and the MSF with CCR model, described in reference [20]. It is this single parameter QMSF model that this work focuses on.

2. The molecular stress function model

The molecular stress function (MSF) model is based on the extra-stress, $\tau(t)$ being given by:

$$\tau(t) = \int_{-\infty}^{t} m(t-t')\mathbf{S_{MSF}}(t')\,\mathrm{d}t', \tag{1}$$

where $\mathbf{S_{MSF}}$ is the strain measure, and $m(t-t')$ the memory function between time t' in the past, and the current time t. The strain measure of the molecular stress function is given by:

$$\mathbf{S_{MSF}} = 5f^2\left\langle \frac{\mathbf{u}'\mathbf{u}'}{u'^2}\right\rangle, \tag{2}$$

where $\mathbf{u}'$ is a deformed vector given by

$$\mathbf{u}' = \mathbf{F}(\mathbf{t},\mathbf{t}')^{-1}\mathbf{u}_0, \tag{3}$$

where $\mathbf{F}$ is the deformation gradient tensor between time t' in the past, and the current time t, and $\mathbf{u}_0$ a vector of (initially) unit length. The brackets $\langle\,\rangle$, denote the average over the surface of a sphere of unit radius, i.e.:

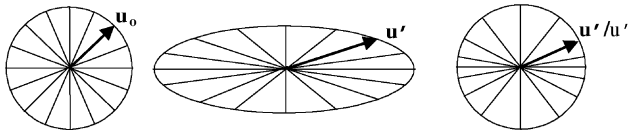

Fig. 1. Schematic illustration of tube deformations: $\mathbf{u}_0$ is the original undeformed vector, $\mathbf{u}'$ the deformed vector given by $\mathbf{u}' = \mathbf{F}^{-1}\mathbf{u}_0$, and $\mathbf{u}'/u'$ the normalised deformed vector.

$$\langle h\rangle = \frac{1}{4\pi}\int_{\Omega} h\,\mathrm{d}\Omega. \tag{4}$$

The strain measure, $\mathbf{S_{MSF}}$ is related to the Doi–Edwards strain function, $\mathbf{S_{DE}}$, the relationship is:

$$\mathbf{S_{MSF}} = f^2\mathbf{S_{DE}}. \tag{5}$$

Fig. 1 suggests the deformations of tube segments, according to the Doi–Edwards model, in an extensional flow. The division by u'^2 in Eq. (2), inherent to the Doi–Edwards strain tensor, normalises the length of the deformed segments, but effectively draws the 'strain ellipse' back to being a 'strain sphere' (see Fig. 1). Thus the Doi–Edwards model assumes a constant cross-section under elongation; the orientation of the segments is accounted for, but the stretching of the tube is not accounted for [19]. This indicates the need for 'f^2' in Eq. (2).

The value f represents the ratio of initial to final tube diameters a_0/a. The ratio f has been defined as a function of $\langle u'\rangle$, or $\langle \ln u'\rangle$. A pair of parameter-free models is derived from first principles: the linear molecular stress function (LMSF) is for modelling linear molecules; the parameter-free model is derived as:

$$f^2 = \mathrm{e}^{\langle \ln u'\rangle}, \tag{6}$$

in reference [18], and the quadratic strain function (QMSF) for modelling long chain branched molecules; is derived in reference [19] in parameter-free form:

$$f^2 = \tfrac{1}{2}\,\mathrm{e}^{2\langle \ln u'\rangle} + \tfrac{1}{2}. \tag{7}$$

We concentrate on a recent single-parameter model for f [19] which includes the notion of a maximum tube stretch. This includes the idea of tube-slip at high deformations, which gives rise to a maximum tube stretch; the tube-slip coefficient is defined in terms of stored energy E, and a maximum value of stored energy, $E_{\max}$. The stored energy is proportional to $f^2 - 1$. These principles are used to derive single parameter models for the LMSF and QMSF:

For the LMSF:

$$f^2 = 1 + (f_{\max}^2 - 1)\left[1 + \exp\left(-\frac{\mathrm{e}^{\langle \ln u'\rangle} - 1}{f_{\max}^2 - 1}\right)\right], \tag{8}$$