J. Non-Newtonian Fluid Mech. 159 (2009) 41–49

Contents lists available at ScienceDirect

Journal of Non-Newtonian Fluid Mechanics

journal homepage: www.elsevier.com/locate/jnnfm

Modelling PTFE paste extrusion: The effect of an objective flow type parameter

Evan Mitsoulis [a,*], Savvas G. Hatzikiriakos [b]

[a] School of Mining Engineering and Metallurgy, National Technical University of Athens, Zografou, 157 80, Athens, Greece
[b] Department of Chemical and Biological Engineering, The University of British Columbia, Vancouver, BC, Canada

ARTICLE INFO

Article history:
Received 15 December 2008
Received in revised form 8 January 2009
Accepted 8 January 2009

Keywords:
Polytetrafluoroethylene
Axisymmetric contraction
Paste extrusion
Slip law
Compressibility
Structural parameter
Flow type parameter
Objectivity

ABSTRACT

In a recent article [P.D. Patil, J.J. Feng, S.G. Hatzikiriakos, Constitutive modelling and flow simulation of polytetrafluoroethylene paste extrusion, J. Non-Newtonian Fluid Mech. 139 (2006), 44–53], a rheological constitutive equation was proposed to model the incompressible flow of poly-tetra-fluoro-ethylene (PTFE) paste and its structural development (fibrillation between the PTFE particles) during flow. The constitutive model was a combination of shear-thinning and shear-thickening viscosity terms, depending on a structural parameter, ξ, which obeyed a convective-transport equation. In the latter, a non-objective flow type parameter, ψ, was employed, which depended on the magnitudes of the rate-of-strain and vorticity tensors. In the present work, an objective flow type parameter is used instead [R.G. Larson, Flows of constant stretch history for polymeric materials with power-law distributions of relaxation times, Rheol. Acta 24 (1985), 443–449]. Despite its complexity and the severe slip at the wall, good convergence has been obtained even for very high apparent shear rates. New results are shown as contours of the ξ-parameter in the flow field as well as contours of the new objective flow type parameter, ψ^*, and compared with previous results. It is shown that the corrected model predicts higher structural levels (more fibrils) in PTFE paste flow through extrusion dies and that the new results are more consistent with experimental observations.

1. Introduction

In a recent article Patil et al. [1] proposed a rheological constitutive equation to model PTFE paste flow and its structural development in tapered axisymmetric dies. The authors performed steady-state flow simulations, which were governed by the conservation equations of mass and momentum under isothermal, incompressible conditions. A linear slip law at the wall was also implemented due to the presence of the lubricant. The constitutive model proposed was a combination of a shear-thinning and a shear-thickening viscosity terms, linearly connected by a structural parameter, ξ, defined as the percentage of fibrillated domains in the paste. This parameter obeyed a convective-transport equation (see below for details of the set of equations). The paste flow was assumed to be incompressible. However, significant amounts of air voids exist in the paste due to low lubricant concentrations. More recent work by Mitsoulis and Hatzikiriakos [2] re-examined the PTFE paste flow problem by taking into account the significant compressibility of the paste and by implementing a slip law based on the consistent normal-to-the-surface unit vector. New results were shown as contours of the ξ-parameter in the flow field in order to study in detail the effect of compressibility on the structure formation and flow kinematics of paste. These results offered a better understanding of the flow behaviour of PTFE in flow through extrusion dies.

A problem with the previous models [1,2] arises from using a non-objective flow type parameter, ψ, that represents the strength of the flow. This measure of flow type was originally introduced by Giesekus [3] and later used by Fuller and Leal for homogeneous flows [4]. It is given by

$$\psi = \frac{|\dot{\gamma}| - |\omega|}{|\dot{\gamma}| + |\omega|}, \tag{1}$$

where $|\dot{\gamma}|$ and $|\omega|$ are the magnitudes of the rate-of-strain tensor $\dot{\gamma} = \nabla \boldsymbol{u} + \nabla \boldsymbol{u}^{\mathrm{T}}$ and the vorticity tensor $\boldsymbol{\omega} = \nabla \boldsymbol{u} - \nabla \boldsymbol{u}^{\mathrm{T}}$, respectively, $\boldsymbol{u}$ is the velocity vector, $\nabla \boldsymbol{u}$ is the velocity gradient tensor, and $\nabla \boldsymbol{u}^{\mathrm{T}}$ is its transpose. As explained in [3,4], $\psi = -1$ in pure rotational flow, $\psi = 0$ in pure shear flow, and $\psi = 1$ in pure elongational flow. In a mixed flow, such as flow through an extrusion die considered here, $-1 \leq \psi \leq 1$. Since values of ψ less than zero correspond to weak flows, and since such flows do not contribute to fibril formation (structure development), Patil et al. [1] were setting all negative values of ψ equal to 0 in their calculations (no structure development).

Astarita [5] pointed out the necessity for a frame-invariant generalization of Eq. (1), and this was also explained in detail by Singh and Leal [6] for the case of flows generated by a co-rotating two-roll

* Corresponding author.
E-mail address: mitsouli@metal.ntua.gr (E. Mitsoulis).

0377-0257/$ – see front matter © 2009 Elsevier B.V. All rights reserved.
doi:10.1016/j.jnnfm.2009.01.001

mill. The new definition of the objective structural parameter, ψ', is given by

$$\psi' = \frac{|\dot{\gamma}| - |\Omega|}{|\dot{\gamma}| + |\Omega|}, \tag{2}$$

where $|\Omega|$ is the magnitude of the relative rate-of-rotation tensor, $\mathbf{\Omega}$, which is the difference between two tensors, the vorticity tensor, $\boldsymbol{\omega}$, and the antisymmetric tensor, $\boldsymbol{W}$, of the rate of rotation of the rate-of-strain tensor, $\dot{\gamma}$. The definition of $\boldsymbol{W}$ is given by [5]:

$$\frac{\mathrm{D}\boldsymbol{e_i}}{\mathrm{D}t} = \boldsymbol{W} \cdot \boldsymbol{e_i}, \tag{3}$$

where D/Dt is the substantial time derivative and $\boldsymbol{e}_i$ are the proper vectors of $\dot{\gamma}$ (i.e., unit vectors along the principal axes of $\dot{\gamma}$). Similarly, ψ' varies between −1 and +1, and in flow simulations the negative values of ψ' should be set arbitrarily equal to zero in order to exclude weak flows that do not contribute to structure development [1,2].

Deriving an objective flow type parameter is clearly a challenge, as it involves the determination of angular velocity of eigenvectors. Some authors [7,8] have worked out such modifications, but it is certainly not a trivial matter. Larson [9] has put forward a frame-invariant flow strength parameter, S_f, defined by

$$S_f = \frac{2(tr\boldsymbol{D}^2)^2}{tr\dot{\mathcal{D}}^2}, \tag{4}$$

where $\dot{\mathcal{D}}$ is the Jaumann time derivative of the rate-of-deformation tensor, $\boldsymbol{D}$, defined as $2\boldsymbol{D} \equiv \dot{\gamma}$, i.e., the time derivative of $\boldsymbol{D}$ with respect to a frame that rotates with the angular velocity of the fluid element (see Appendix). The parameter S_f ranges from 0, for pure rotational flow, to 1, for simple shear flow, to infinite, for pure elongational flow. To make it readily usable in the flow model, S_f can be accordingly normalized to result in a new objective flow type parameter. It is worthwhile to mention that the above flow type or flow strength parameters have been used in the past, particularly in polymeric flows that involve mixing, by Manas-Zloczower and co-workers [10–14]. On the other hand, the non-objective ψ-parameter was primarily used due to its ease of computation, while the much more difficult to calculate S_f-parameter has been used in [13].

It is the purpose of the present work to re-examine the flow of PTFE paste in extrusion dies by using a consistent and objective flow type parameter, based on S_f. The new flow strength parameter and the model are then formulated accordingly. The results are compared with those obtained with the use of the non-objective flow type parameter, ψ, reported in Ref. [2].

2. Governing equations and rheological modelling

We consider the conservation equations of mass and momentum for compressible fluids under isothermal, creeping, steady flow conditions. These are written as [2]:

$$\boldsymbol{u} \cdot \nabla\rho + \rho(\nabla \cdot \boldsymbol{u}) = 0, \tag{5}$$

$$0 = -\nabla p + \nabla \cdot \boldsymbol{\tau}, \tag{6}$$

where ρ is the density, $\boldsymbol{u}$ is the velocity vector, p is the pressure and τ is the extra stress tensor. For a compressible fluid, pressure and density are connected as a first approximation through a simple linear thermodynamic equation of state [2]:

$$\rho = \rho_0(1 + \beta p), \tag{7}$$

where β is the isothermal compressibility with the density being ρ_0 at reference pressure p_0 (=0). The viscous stresses are given for inelastic non-Newtonian compressible fluids by the relation [2]:

$$\boldsymbol{\tau} = \eta(|\dot{\gamma}|)\left(\dot{\gamma} - \frac{2}{3}(\nabla \cdot \boldsymbol{u})\boldsymbol{I}\right), \tag{8}$$

where $\eta(|\dot{\gamma}|)$ is the apparent non-Newtonian viscosity, which is a function of the magnitude $|\dot{\gamma}|$ of the rate-of-strain tensor, and is given by

$$|\dot{\gamma}| = \sqrt{\frac{1}{2}II_{\dot{\gamma}}} = \left(\frac{1}{2}(\dot{\gamma} : \dot{\gamma})\right)^{1/2}, \tag{9}$$

where $II_{\dot{\gamma}}$ is the second invariant of $\bar{\bar{\dot{\gamma}}}$. The tensor $\boldsymbol{I}$ in Eq. (8) is the unit tensor.

In the previous works [1,2], a simple linear combination of shear-thinning and shear-thickening viscosities was assumed for PTFE paste, depending on a structural parameter, ξ:

$$\eta = (1 - \xi)\eta_1 + \xi\eta_2, \tag{10}$$

In the above, η_1 and η_2 are the shear-thinning and shear-thickening viscosities, respectively, that are expressed by a Carreau model [1,2]:

$$\eta_i = \eta_{0,i}[1 + (\lambda_i|\dot{\gamma}|)^2]^{(n_i-1)/2}, \tag{11}$$

where $i = 1$ refers to shear-thinning ($n_1 < 1$) and $i = 2$ refers to shear-thickening ($n_2 > 1$). The model parameters are the zero-shear-rate viscosity, $\eta_{0,i}$, the power-law index, n_i, and the time constant, λ_i. Their values are listed in Table 1.

For the structural parameter, ξ, a convective-transport equation (CTE) was introduced in [1] based on arguments of creation and destruction of fibrils (see [1] for details). The CTE is written as

$$\boldsymbol{u} \cdot \nabla\xi = f - g, \tag{12}$$

where f and g denote the rate of creation and elimination of fibrillated domains in the paste. These functions are given by

$$f(|\dot{\gamma}|, \zeta) = 2|\dot{\gamma}|\sqrt{\zeta(1 - \zeta)}, \tag{13a}$$

$$g(|\dot{\gamma}|, \xi) = \left|\dot{\gamma}\right|\xi, \tag{13b}$$

where the proportionality constants in the above functions proposed by Patil et al. [1] were assumed to be 1 in order to bound ζ between 0 and 1; the parameter ζ is related to S_f, according to Larson [9] as follows. For a planar flow of constant stretch history, there is a frame in which

$$\nabla\boldsymbol{u} = |\dot{\gamma}|\begin{pmatrix} 0 & \zeta \\ 1 - \zeta & 0 \end{pmatrix}, \tag{14}$$

where ζ is related to S_f by

$$S_f = \frac{1}{(1 - 2\zeta)^2}. \tag{15}$$

The largest eigenvalue of the velocity gradient tensor (Eq. (14)) has the form $\left|\dot{\gamma}\right|\sqrt{\zeta(1 - \zeta)}$, and this is why the rate of fibril creation appears as in Eq. (13a). The factor of 2 in Eq. (13a) is added to bound ξ between 0 and 1. According to Larson [9], strong flows are obtained for $\zeta(1 - \zeta) > 0$, and therefore in our calculations all negative values of $\zeta(1 - \zeta)$ are set equal to zero. This corresponds to weak flows for

Table 1
Constants used in the simulations for PTFE paste extrusion at 35 °C.

Parameter	PTFE (35 °C) *Model* (Eq. (11))
$\eta_{0,1}$	0.004 MPa s
n_1	0.5
λ_1	0.3 s
$\eta_{0,2}$	0.0016 MPa s
n_2	1.3
λ_2	1.0 s
β	0.02 MPa^{-1}
ρ_0	1600 kg/m^3
β_{sl}	1.92 m/MPa s