A simple phenomenological non-Newtonian fluid model

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A B S T R A C T

I n this work, a simple phenomenological generalized Newtonian law model has been proposed and tested for different polymer melts by using theological data taken from the open literature. Viscosity is given as a specific function of three principal invariants of the deformation rate tensor, D, and its absolute value defined as %/D • *D.* It has been found that the model predictions are in very good agreement with the strain rate dependent steady shear and uniaxial extensional viscosities for linear and branched polyolefmes. The model behaves correctly in description of steady planar and equibiaxial extensional viscosities and allows their independent strain hardening level control with respect to uniaxial extensional viscosity.

1. Introduction

As early as in 1948 Rivlin observed that for an isotropic generalized Newtonian fluid the viscosity is a function of only three principal invariants of the deformation rate tensor, *r){lD,llD, IIID)* [1,2]. However, for incompressible non-Newtonian liquids (for which the first invariant of the deformation rate tensor is zero) viscosity is usually considered to be a function of only the second and third principal invariants of the deformation rate tensor *r)* (*IIQ, IIID)* [3-10]. The main weakness of generalized Newtonian law model, in which the stress tensor is directly proportional to the deformation rate tensor through *rj* (*IID, IIID),* is its incapability to correctly represent the flow behavior in extensional flows, especially in plane flows, where the third invariant of deformation rate tensor *IIID* is 0. In order to overcome this problem, new definition of the non- Newtonian fluid viscosity is proposed here and tested for three different polymer melts (highly branched LDPE, slightly branched mLLDPE and linear HDPE) whose rheological characteristics were taken from the open literature [11,12].

2. Model development and testing

The simplest non-Newtonian fluid model is generalized Newtonian law in the following form:

$$\tau = 2\eta(I_D, II_D, III_D)D \tag{1}$$

where r means the stress tensor, D represents the deformation rate tensor and *rj* stands for the viscosity which is not constant (unlike in the standard Newtonian law) and is allowed to vary with the first, second and third invariant of the deformation rate tensor, I_D = *tr(D), IID = 2tr* (D^2) and III_D = det(D), respectively. Let us consider viscosity as a variable which depends on three principal invariants of the deformation rate tensor D as well as on its *absolute value \D* i.e.

$$\eta = f\left(I_D, I_{|D|}, II_D, II_{|D|}, III_D, III_{|D|}\right) \tag{2}$$

Here, it is important to mention that |D| is a new tensor defined as |d| = *VD* ■ *D,* which characterizes deformation rate intensity in a particular direction (similarly to D) but without the information about its orientation (positive or negative). From Table 1, where all considered invariants are provided for basic flow situations, it is clearly visible that: firstly, I_D = 0 due to the applied assumption of fluid incompressibility, secondly, /|D| becomes zero and nonzero for pure shear and extensional flow, respectively. Finally, *IID = II\D* and *IIID* = *III*|D| for all considered flow types, except for equibiaxial flow in which *1IID* = *-2s*3 and ///|D| = 2è^3, i.e. J/J|D| = |///d| in this case.

It is worth attention that /|D| characterizes the overall amount of stretching during flow (/jD| ^ 0 in extensional flows) whereas *ID* represents the continuity equation for incompressible fluids *(ID*-0). Based on this, the viscosity definition given by Eq. (2) can be simplified for incompressible fluids as follows:

$$\eta = f\left(I_{|D|}, II_D, III_D, \left|III_D\right|\right) \tag{3}$$

Let us further assume that viscosity dependence on /iD|, *IID, IIID,* I/I/d I can be expressed in the following way:

$$\eta\left(I_{|D|}, II_D, III_D, \left|III_D\right|\right) = \eta(II_D)^{f(I_{|D|}, II_D, III_D, |III_D|)} \tag{4}$$

where *r](IID)* is given by the well known Carreau-Yasuda model presented in Eq. (5) [13,14], and exponent/ (7|D|, *IID, IIID,* |///£»|) is

Table 1
Three principal invariants of the deformation rate tensor *D* and its absolute value |D|.

	Simple shear flow	Uniaxial extensional flow	Equibiaxial flow	Planar flow
D	$\begin{pmatrix} 0 & \dot{\gamma}/2 & 0 \\ \dot{\gamma}/2 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}$	$\begin{pmatrix} \dot{\varepsilon} & 0 & 0 \\ 0 & -\dot{\varepsilon}/2 & 0 \\ 0 & 0 & -\dot{\varepsilon}/2 \end{pmatrix}$	$\begin{pmatrix} \dot{\varepsilon} & 0 & 0 \\ 0 & \dot{\varepsilon} & 0 \\ 0 & 0 & -2\dot{\varepsilon} \end{pmatrix}$	$\begin{pmatrix} \dot{\varepsilon} & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & -\dot{\varepsilon} \end{pmatrix}$
$\|D\|$	$\begin{pmatrix} 0 & \dot{\gamma}/2 & 0 \\ \dot{\gamma}/2 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}$	$\begin{pmatrix} \dot{\varepsilon} & 0 & 0 \\ 0 & \dot{\varepsilon}/2 & 0 \\ 0 & 0 & \dot{\varepsilon}/2 \end{pmatrix}$	$\begin{pmatrix} \dot{\varepsilon} & 0 & 0 \\ 0 & \dot{\varepsilon} & 0 \\ 0 & 0 & 2\dot{\varepsilon} \end{pmatrix}$	$\begin{pmatrix} \dot{\varepsilon} & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & \dot{\varepsilon} \end{pmatrix}$
I_D	0	0	0	0
$I_{\|D\|}$	0	$2\dot{\varepsilon}$	$4\dot{\varepsilon}$	$2\dot{\varepsilon}$
$II_D = II_{\|D\|}$	$\dot{\gamma}^2$	$3\dot{\varepsilon}^2$	$12\dot{\varepsilon}^2$	$4\dot{\varepsilon}^2$
III_D	0	$\frac{1}{4}\dot{\varepsilon}^3$	$-2\dot{\varepsilon}^3$	0
$III_{\|D\|} = \|III_D\|$	0	$\frac{1}{4}\dot{\varepsilon}^3$	$2\dot{\varepsilon}^3$	0

Here, $\dot{\gamma}$ and $\dot{\varepsilon}$ represent shear and extensional strain rates, respectively.

given by Eq. (6), a novel relation proposed here for the first time.

$$\eta(II_D) = \frac{\eta_0 a_T}{\left[1 + \left(\lambda a_T \sqrt{II_D}\right)^a\right]^{(1-n)/a}} \tag{5}$$

$$f\left(I_{|D|}, II_D, III_D, |III_D|\right) = \left\{\tanh\left[\alpha a_T\left(1 + \frac{1}{4\left(\sqrt{3}\right)^3}\right)^{-\psi}\left(\left|1 + \frac{III_D}{II_D^{3/2}}\right|\right)^{\psi}\frac{\sqrt[3]{4\left|III_D\right|} + I_{|D|}}{3} + \beta\right]\frac{1}{\tanh(\beta)}\right\}^{\zeta} \tag{6}$$

Here η_0, λ, a, n, α, ψ, β, and ζ are adjustable parameters and a_T is the temperature shift factor. In simple shear flow, the function $f\left(I_{|D|}, II_D, III_D, \left|III_D\right|\right)$ defined by Eq. (6) is 1 because $\left(\sqrt[3]{4\left|III_D\right|} + I_{|D|}\right)/3 = 0$. On the other hand, in extensional flows, the term $\left(\sqrt[3]{4\left|III_D\right|} + I_{|D|}\right)/3$ becomes nonzero (equal to $\dot{\varepsilon}$ for uniaxial flow, $2\dot{\varepsilon}$ for equibiaxial flow and $2\dot{\varepsilon}/3$ for planar flow) and due to this the function $f\left(I_{|D|}, II_D, III_D, \left|III_D\right|\right)$ starts to deviate from 1. The term $(1 + 1/(4(\sqrt{3})^3))^{-\psi}(\left|1 + III_D/II_D^{3/2}\right|)^{\psi}$ in Eq. (6) allows controlling the significance (amount of difference) of the particular type of flow with respect to uniaxial extensional flow. In more detail, this term is highest for uniaxial extensional flow (equal to 1), lower for planar extensional flow, $(1 - 1/(4(\sqrt{3})^3))^{-\psi}$, and lowest for equibiaxial flow, $(1 + 1/(4(\sqrt{3})^3))^{-\psi}(1 - 1/(12\sqrt{3}))^{\psi}$. The differences between the flow types become more obvious for increased parameter $|\psi|$ in the above mentioned order. In other words, the adjusted value of the term $(1 + 1/(4(\sqrt{3})^3))^{-\psi}(\left|1 + III_D/II_D^{3/2}\right|)^{\psi}$ through ψ parameter allows independent control of the planar/equibiaxial extensional viscosity at the same shear and uniaxial extensional viscosity. It is important to say that in complex flow modeling by using the proposed model, the deformation rate tensor should be evaluated in the local, streamline oriented coordinate system; this helps to distinguish shear and extensional flow components and determination of |D| is less complicated.

To test the proposed model, strain rate dependent steady shear and uniaxial extensional viscosities were taken from the open literature [11,12] for three different polymer melts (highly branched LDPE Escorene LD165 BW1, slightly branched mLLDPE Exact 0201 and linear HDPE Tipelin FS 450-26). These polymers were chosen mainly because they are very well characterized and their Theological properties have already been modeled by advanced differential (XPP model [15], PTT-XPP model [16], modified Leonov model [17]) as well as integral (MSF model [18]) constitutive equations.

The fitting procedure was as follows: In the first step, the shear rate dependent steady shear viscosity was fitted by the Carreau-Yasuda model, Eq. (5), in order to determine 770, *X, a* and n model parameters. In the second step, the full model given by Eqs. (4-6) was used to fit the extensional strain rate dependent steady uniaxial extensional viscosity to determine *a,* and f parameters, whereas the remaining model parameters (*t]0*, A, *a, n)* were fixed. In both steps, non-linear least squares minimization regression method employing the Marquardt-Levenberg algorithm [19,20] was utilized. A comparison between the experimental data and the proposed model predictions for all tested polymer melts is pro-

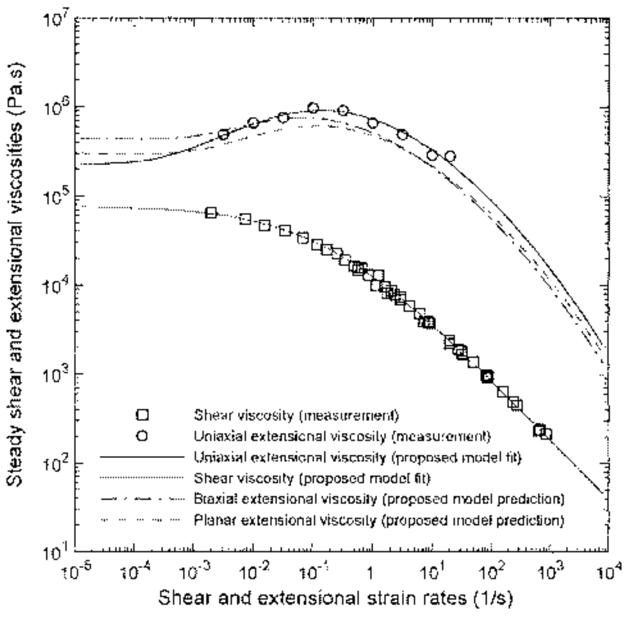

Fig. 1. Comparison between strain rate dependent steady shear and extensional viscosities for highly branched LDPE Escorene LD165 BW1 atT=200°C.

Table 2
Generalized Newtonian model parameters for all tested polymer samples (T^ = 20 for all samples).

	η_0 (Pa s)	λ (s)	a (−)	n (−)	α (s)	β (−)	ζ (−)
LDPE	77,103	6.8553	0.4582	0.3202	1×10^{-5}	3.27×10^{-9}	0.037864
mLLDPE	17,999	0.7078	0.7612	0.4005	192.5×10^{-5}	7.29×10^{-7}	0.011145
HDPE	5,033,405	1000	0.2074	0.3277	1×10^{-5}	3.95×10^{-9}	0.015250

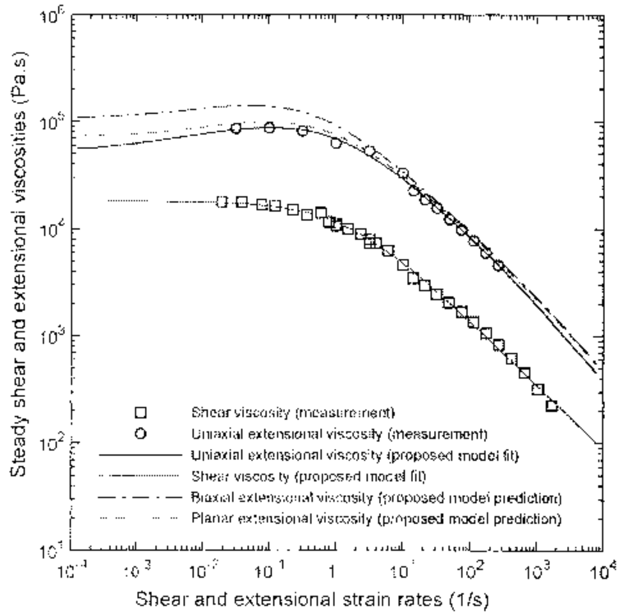

Fig. 2. Comparison between strain rate dependent steady shear and extensional viscosities for slightly branched mLLDPE Exact 0201 at *T*= 180 °C.

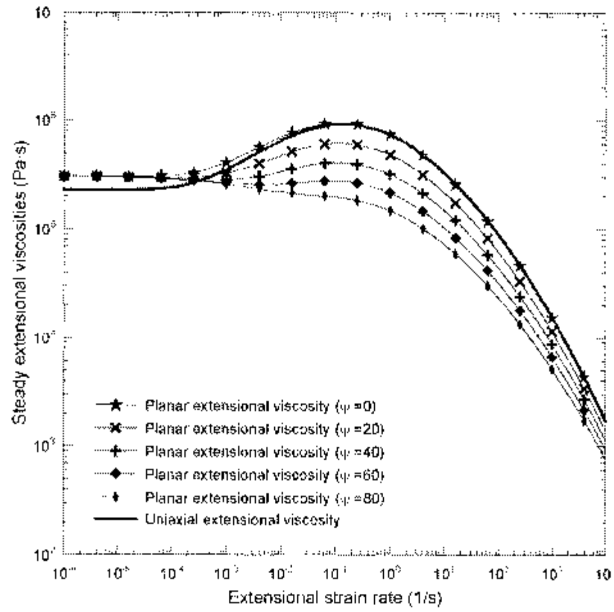

Fig- 4. The effect of parameter *yjr* on the strain rate dependent steady planar extensional viscosity for highly branched LDPE Escorene LD165 BW1 at T=200°C.

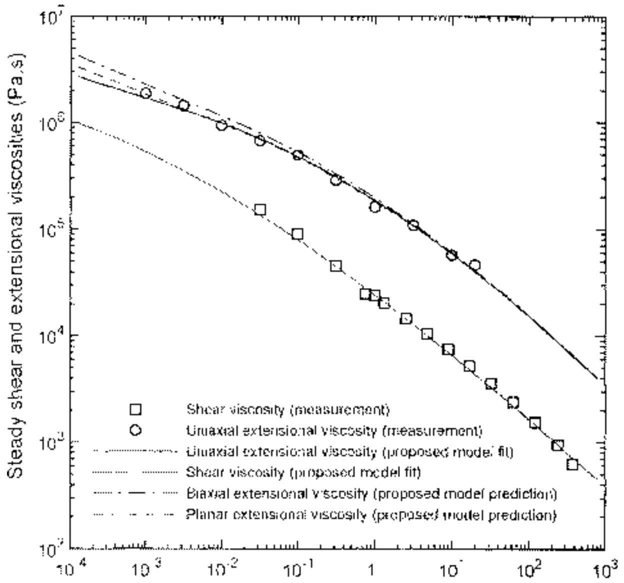

Fig. 3. Comparison between strain rate dependent steady shear and extensional viscosities for linear HDPE Tipelin FS 450-26 at *T*= 180 °C.

vided in Figs. 1-3 and the corresponding model parameters are given in Table 2. As can be seen, the fitting capabilities of the proposed model for steady shear and uniaxial extensional viscosities of the chosen polymer melts are very good.

he steady planar and equibiaxial extensional viscosities predicted by the proposed model for all three samples are also provided in Figs. 1-3, where parameter ^ occurring in Eq. (6) was adjusted *ad hoc* to the value of 20 for all tested samples. As already mentioned, this parameter allows independent control of steady biaxial/planar extensional viscosity. In order to explore this point in more detail, parameter *i/r* was varied from 0 up to 80 for highly branched LDPE Escorene LD165 BW1; the corresponding predictions for steady planar/equibiaxial extensional viscosities are compared with steady uniaxial viscosity in Figs 4 and 5. It can be clearly seen that an increase in ^ parameter leads to a decrease in the strain hardening for both planar and equibiaxial extensional viscosities and the effect is much more pronounced for the latter. This may justify the use of parameter t/^>0 (about 20 in this case) for the studied highly branched LDPE (especially if experimental planar/equibiaxial extensional viscosity data are not available to get the t//" parameter directly by the fitting procedure). In such a case, extensional strain hardening in the uniaxial extensional viscosity is slightly higher than in its planar counterpart, whereas the strain hardening in the equibiaxial extensional viscosity is the lowest, as shown theoretically in Figs. 4-5 and experimentally for branched LDPE polymers in [21-23]. Moreover, if the parameter t/t » 0 (higher than 40 in this case, see Figs. 4-5), strain hardening in equal biaxial and planar extensional viscosities is significantly suppressed while high extension strain hardening remains unchanged in uniaxial extensional viscosity, which is typical for polyisobuty-

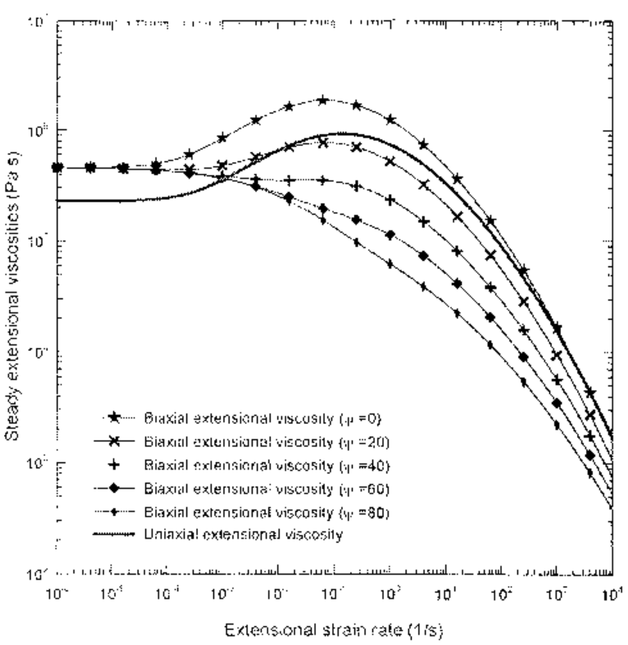

Fig. 5. The effect of parameter \/f on the extensional strain rate dependent steady equibiaxial extensional viscosity for highly branched LDPE Escorene LD165 BW1 at T=200'C.

experimentally in [24]. This suggests that the proposed model may be flexible enough to describe strain rate dependent steady extensional viscosities of different polymer melts.

3. Conclusion

The simple phenomenological generalized Newtonian law model proposed and tested for different polymer melts in this work has proved a good capability to describe the strain rate dependent steady shear and uniaxial extensional viscosities for linear and branched polyolefines. Moreover, it has shown correct predictive behavior for steady planar and equibiaxial extensional viscosities and ability to control their strain hardening level independently of uniaxial extensional viscosity. This supports the claim that the proposed model can be useful in steady shear and uni- axial/planar/equibiaxial extensional viscosity modeling due to its sufficient flexibility and the use of a low number of adjustable parameters which can be identified easily through analytical expressions for steady shear and extensional viscosities. It is also expected that the proposed model can be useful in modeling industrial processes, such as film blowing, fiber spinning, film casting or flat die/profile/annular die flow, where it enables to assess the role of steady shear and extensional viscosities.

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