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Research Article

Empirical Modelling of Nonmonotonous Behaviour of Shear Viscosity

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Almost all hitherto proposed empirical models used for characterization of shear viscosity of non-Newtonian liquids describe only its monotonous course. However, the onset of new materials is accompanied by more complicated characteristics of their behaviour including nonmonotonous course of shear viscosity. This feature is reflected not only in an existence of one extreme point (maximum or minimum), but also it can appear in both extreme points; that is, this shear viscosity initially exhibits shear thinning; after attaining a local minimum, it converts to shear thickening, and again after reaching a local maximum, it has a shear-thinning character. It is clear that, for an empirical description of this complex behaviour, a hitherto, used number of parameters (four, five) in classical monotonous models (such as Cross or Carreau-Yasuda) are no longer tenable. If more parameters are applied, there should be given an emphasis on a relatively simple algebraic form of the proposed models, unambiguity of the involved parameters, and their sound interpretation in the whole modelling. This contribution provides an overview of the existing empirical nonmonotonous models and proposes a new 10-parameter model including a demonstration of its flexibility using various experimental data.

1. Introduction

Constitutive modelling represents an inevitable step in completing balance equations describing flow behaviour of non-Newtonian fluids. In principle, there are approximately two ways how to cope with this problem. The first one is oriented to a description of one characteristic only (such as for example, shear viscosity) but a sufficiently precise approximation is expected. The second approach consists in a proposal of differential or integral constitutive equation simultaneously describing more rheological characteristics (such as shear and elongational viscosities and first and second normal stress differences). However, accuracy of the individual characteristic (such as for example, shear viscosity) is lower compared to the empirical modelling. It depends on the complexity of the given problem and user's requirements which approach is more suitable to apply.

Empirical modelling of purely viscous and viscoplastic materials started in the 20s with the proposals of 2-parameter constitutive models (power-law, Bingham [1, 2]) followed by the 3-parameter ones: Herschel-Bulkley [2] as a combination of the preceding two models and the Williamson one [3], initiating an algebraic structure appearing in nowadays frequently used models such as the 4-parameter Cross [4] and 5-parameter Careau-Yasuda [5, 6] ones. A detailed analysis with respect to the number of empirical parameters is given for example, in Filip et al. [7]. Recently, the empirical models describing elastoviscoplastic behaviour (Saramito [8], de Souza Mendes and Thompson [9]) have been published.

With the onset of a new larger class of materials, shear viscosity no longer exhibits a monotonous course only. In this case, the classical models fail, and there is a necessity to apply models enabling a description of nonmonotonous course of behaviour of this quantity, that is, to describe simultaneously shear thinning and shear thickening behaviours in their various combinations. Unfortunately, the number of models devoted to this phenomenon is less than scarce. These models can be classified into two groups: either the proposals of algebraically new forms or the modifications of the classical models.

Ranging to the first group, the 6-parameter model proposed in David and Filip [10] enables the description of the

situations when one local extreme (either maximum or minimum) appears between the initial and terminal Newtonian plateaux (with the possibility to include an intermediate one),

$$\eta = \frac{\eta_\infty \exp\left(f\left(\dot{\gamma}; c, p, q\right)\right) + \eta_0 \exp\left(-f\left(\dot{\gamma}; c, p, q\right)\right)}{b + \exp\left(f\left(\dot{\gamma}; c, p, q\right)\right) + \exp\left(-f\left(\dot{\gamma}; c, p, q\right)\right)}, \quad (1)$$

where

$$f\left(\dot{\gamma}; c, p, q\right) = \operatorname{sign}\left(\log\left(c\dot{\gamma}\right)^p\right) \cdot \left|\log\left(c\dot{\gamma}\right)^p\right|^q, \quad (2)$$

the parameters η_0 and η_∞ determine the initial and terminal Newtonian plateaux, respectively. The parameters c, p, and q are supposed to be positive and the parameter $b > -2$.

Nevertheless, due to the low number of parameters, this model exhibits some restrictions summarized in Filip et al. [7], where the presented 8-parameter model is composed of two similar terms simplified from the relation in (1)

$$\eta = \frac{\eta_0 \exp\left(-f_0\right)}{b_0 + \exp\left(f_0\right) + \exp\left(-f_0\right)} + \frac{\eta_\infty \exp\left(f_\infty\right)}{b_\infty + \exp\left(f_\infty\right) + \exp\left(-f_\infty\right)}, \quad (3)$$

where

$$f_0 \equiv f\left(\dot{\gamma}; c_0, p_0\right) = \log\left(c_0\dot{\gamma}\right)^{p_0}, \qquad f_\infty \equiv f\left(\dot{\gamma}; c_\infty, p_\infty\right) = \log\left(c_\infty\dot{\gamma}\right)^{p_\infty} \quad (4)$$

enable the description of consecutively shear thinning, shear thickening, and again shear thinning. In both models, the parameters included participate in a description of the flow curve within the whole range of shear rates.

Ranging to the second group, Zatloukal et al. [12] modified the simplified 4-parameter Carreau-Yasuda model (with $\eta_\infty \equiv 0$) by an additional 3-parameter multiplicative function ensuring nonmonotonous behaviour of viscosity

$$\log\left(\eta\right) = \log\left(\frac{\eta_0}{\left[1 + \left(k\dot{\gamma}\right)^a\right]^{(1-n)/a}}\right) \cdot \left[\frac{\tanh\left(\alpha\dot{\gamma} + \beta\right)}{\tanh\left(\beta\right)}\right]^\xi. \quad (5)$$

Galindo-Rosales et al. [11, 13] have recently published the 11-parameter model describing consecutively shear thinning, shear thickening, and shear thinning behaviour where each monotonous part was described by its corresponding Cross model in such a way that at local extremes, the individual Cross models were coupled.

The increasing number of parameters appearing in these "nonmonotonous" models is not surprising. Even for only shear thinning materials not exhibiting a "symmetry," that is, the drop-off in viscosity from the initial Newtonian plateau is not mirrored by its rapid levelling-off towards the terminal Newtonian plateau, Roberts et al. [14] introduced the 8-parameter model.

Every empirical model describing a nonmonotonous course of shear viscosity should ideally fulfil the following attributes:

(i) it should depend on a limited number of independent model parameters;

(ii) the parameters should participate throughout the whole range of shear rate (stress) as the strict determination of the individual monotonous subintervals can be evaluated only approximately due to both noncontinuous character of experimental data and their inaccuracy;

(iii) the parameters should be readily and unambiguously evaluated from experimental viscosity data;

(iv) the parameters should represent sound physical or mathematical interpretation connected with the measured viscosity function;

(v) a proposed model should exhibit good mathematical structure to enable sufficiently accurate and relatively simple fit to the measured data;

(vi) it should be applicable to a larger class of materials.

The aim of this contribution is to present a 10-parameter empirical model describing sequential changing of shear thinning, thickening, and again thinning regimes. The efficiency of the model is illustrated using data of other authors.

2. Proposal of the 10-Parameter Model

As introduced above, the 6- and 8-parameter nonmonotonous models are not able to evaluate more complicated flow behaviour of shear viscosity. To widen the class of materials for which an evaluation is possible, the following 10-parameter model is proposed:

$$\eta = \frac{c_1\dot{\gamma} + c_2/\dot{\gamma}}{c_3 + \left(\dot{\gamma} + 1/\dot{\gamma}\right)^{c_4}} + \frac{c_5 \cdot \exp\left(f\right)}{c_6 + \left(\exp\left(f\right) + \exp\left(-f\right)\right)^{c_7}}, \quad (6)$$

where

$$f = \operatorname{sign}\left[\log\left(c_8\dot{\gamma}^{c_9}\right)\right] \cdot \left|\log\left(c_8\dot{\gamma}^{c_9}\right)\right|^{c_{10}}. \quad (7)$$

Analogously to the 6-parameter model (relations in (3) and (4)), the proposed 10-parameter model is a sum of two terms. The rescaling of shear rate, that is, the combination of the exponential and logarithmic functions, was motivated by the model presented by Chynoweth and Michopoulos [17].

3. Comparison with the Experimental Data

An applicability of the proposed 10-parameter model is documented by a comparison with the experimental data by Zhang et al. [15] and White et al. [16]. Figure 1 depicts viscosity as a function of shear rate for different solid fractions of fumed silica in ethylene glycol. Figure 2 shows a comparison between the model and the experimental data relating shear rate with viscosity of a suspension of fumed silica in ethylene glycol at 30% (w/w) and carbonyl iron particles for various intensities of magnetic field. The application of the model to the data in White et al. [16] describing shear thickening behaviour of a suspension of cornstarch in water at 55 wt.% is presented in Figure 3. The lists of the values attained by

Table 1: The model parameters corresponding to Figure 1.

Parameter	Solid fraction [%]		
	20	30	40
c_1	2.5381	3.8577	14.512
c_2	−905.5	740.26	1919.2
c_3	−20.17	−4.8718	0.0837
c_4	1.0313	1.0205	1.173
c_5	0.7589	12.264	38.708
c_6	−1.07	−1.234	0.0075
c_7	0.1559	0.7783	1.169
c_8	$5.22 * E - 17$	$1.44 * E - 19$	$9.98 * E - 31$
c_9	5.5967	6.9871	11.424
c_{10}	0.6952	1.0364	0.9263

Table 2: The model parameters corresponding to Figure 2.

Parameter	Magnetic field intensity [kA/m]		
	0	100	200
c_1	0.0002	104.42	257.05
c_2	9.7284	431.74	755.11
c_3	0.04	0.934	−0.5605
c_4	−0.7996	1.572	1.7011
c_5	0.0307	0.0812	62.05
c_6	−4.8503	−10.75	−3.0491
c_7	2.2786	3.4273	2.4126
c_8	$3.9 * E - 04$	$4.4 * E - 04$	$5.4 * E - 05$
c_9	1.0957	1.1149	1.5887
c_{10}	2.0593	4.0544	0.596

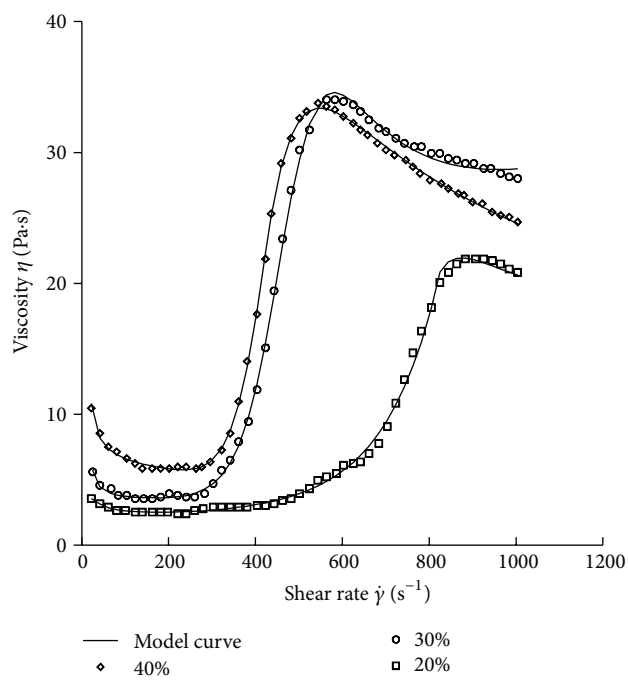

Figure 1: Comparison of the 10-P model with the experimental data presented by Galindo-Rosales et al. [11].

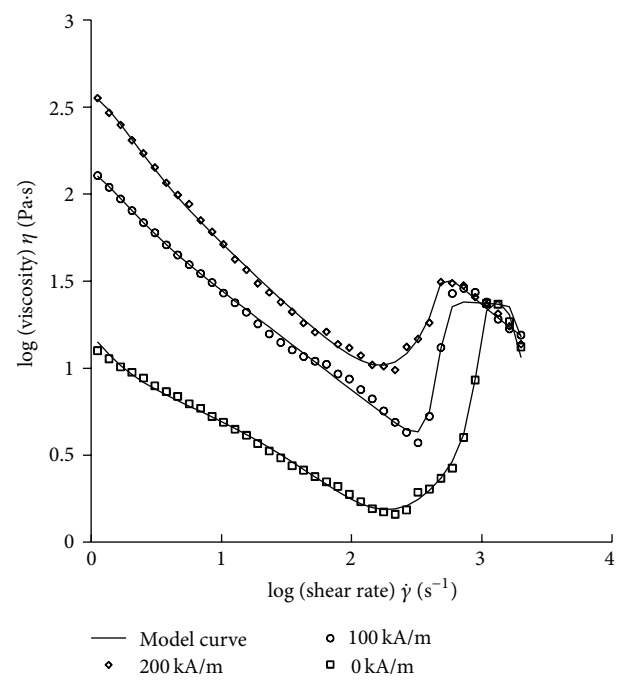

Figure 2: Application of the 10-P model to the experimental data presented by Zhang et al. [15].

the model parameters corresponding to Figures 1–3 are given in Tables 1, 2, and 3. In all cases, the agreement between the empirical fitting and the experimental data is very good.

4. Conclusion

The 10-parameter model was proposed for the description of nonmonotonous behaviour of viscosity in dependence on shear rate. An analogous form can be proposed in dependence on shear stress as for some materials this relation seems to be more corresponding to the physical reality, see Lomellini and Ferri [18]. Seemingly, a large number of parameters are adequate when compared with the other models. The comparisons with the experimental data of other authors showed the very good applicability of the proposed model.

The program in Mathematica software involving the fitting procedure is available at http://www.ih.cas.cz/web_new/cs/index.php?page=Download where the results of this procedure are also documented by the experiments published in the literature. The fitting procedure is organised as a fit-function inside a loop. The fit-function accepts starting values and executes fitting through a standard available routine. In the first run, all starting values equal one, and the fit-function returns a point in the parametric space (i.e., values of 10 parameters) and a corresponding sum of squared errors. Let us denote this point as an acceptable point. For the next cycle of the loop, a starting point is determined according to a multivariate normal distribution with expectation at the

Table 3: The model parameters corresponding to Figure 3.

Parameter	
c_1	0.0706
c_2	0.4169
c_3	−0.4178
c_4	0.3925
c_5	51.762
c_6	−0.3681
c_7	1.9302
c_8	$2.97E-07$
c_9	3.9503
c_{10}	1.5561

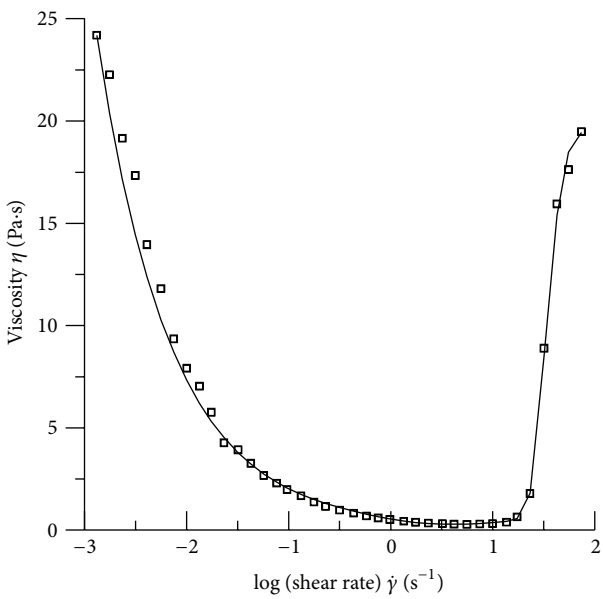

Figure 3: Comparison of the 10-P model with the experimental data presented by White et al. [16].

acceptable point. If the fit-function returns a point with a greater sum of squared errors, then the attempt is considered unsuccessful and the next cycle with the new starting point is run. Otherwise, the attempt is considered successful, and the acceptable status is given to the point found. A criterion to exit the loop can be arbitrary as far as the results are recorded during an execution of the program.

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